DIAGENESIS AND INCIPIENT METAMORPHISM OF THE CARBONIFEROUS ROCKS IN THE SOUTH WALES COALFIELD

By:

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ABSTRACT

A petrographic study of both carbonate rocks forming the Carboniferous Limestone Series and the terrigenous rocks forming the Millstone Grit and Coal Measures Series has been carried out. The carbonate rocks are represented by five main lithotypes, namely: micrites, biomicrites, biosparites, oolitic limestones and dolomites; each lithotype has been further classified into severel sublithotypes. The terrigenous rocks are represented mainly by three lithotypes of argillaceous rocks (mudstone, shale and claystone); two siltstone lithotypes, and three sandstone lithotypes (quartzarenite, lithic arenite and quartzwacke).

The diagenetic processes which affected both the carbonate and the terrigenous rocks have been discussed, and the minerals and textures formed by these processes described.

Four indicators have been used for the determination of the incipient metamorphic grades of the rocks studied, namely, fabric modifications as revealed from thin section study, modifications in the clay mineral composition and in the structure of illite, reflectivity values of the vitrinite grains, and the degree of carbonization of the spores and pollen grains.

The results of the various techniques employed have been discussed in relation to the fixed carbon ratio of the coal seams.

Finally, the relationship between the degree of incipient metamorphism of the source rocks and the occurrence of hydrocarbon accumulation has been discussed.

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CHAPTER I

INTRODUCTION

INTRODUCTION

LOCATION AND GEOGRAPHY

The South Wales Coalfield is one of the principal coalfields of Great Britain. It produces about 1/10th of the total British coal output. It occupies the south-eastern part of South Wales (Fig. 1.1).

The Coalfield has a total area of about 750 sq. miles and is an oval-shaped region extending as a single entity from Pontypool in Monmouthshire for 55 miles westwards to the Gwendraeth Valley in Carmarthenshire. Over much of this length the width is about 16 miles from north to south. The greater part of the coalfield lies in the County of Glamorgan; marginally it extends into the County of Brecknock. The South Wales Coalfield is separated from the Pembrokeshire Coalfield, which may be regarded as a westward extension, by an unproven gap of 14 miles under Carmarthen Bay. To the north of the Coalfield lies the high moorland tract of the Black Mountains, Forest Fawr and the Brecon Beacon. To the south and south-east of the Coalfield is the undulating coastal belt including the agricultural vale of Glamorgan and the seaports of Barry, Cardiff and Newport.

In the South Wales Coalfield a distinctive feature is the outcropping of massive Pennant Sandstone giving rise to

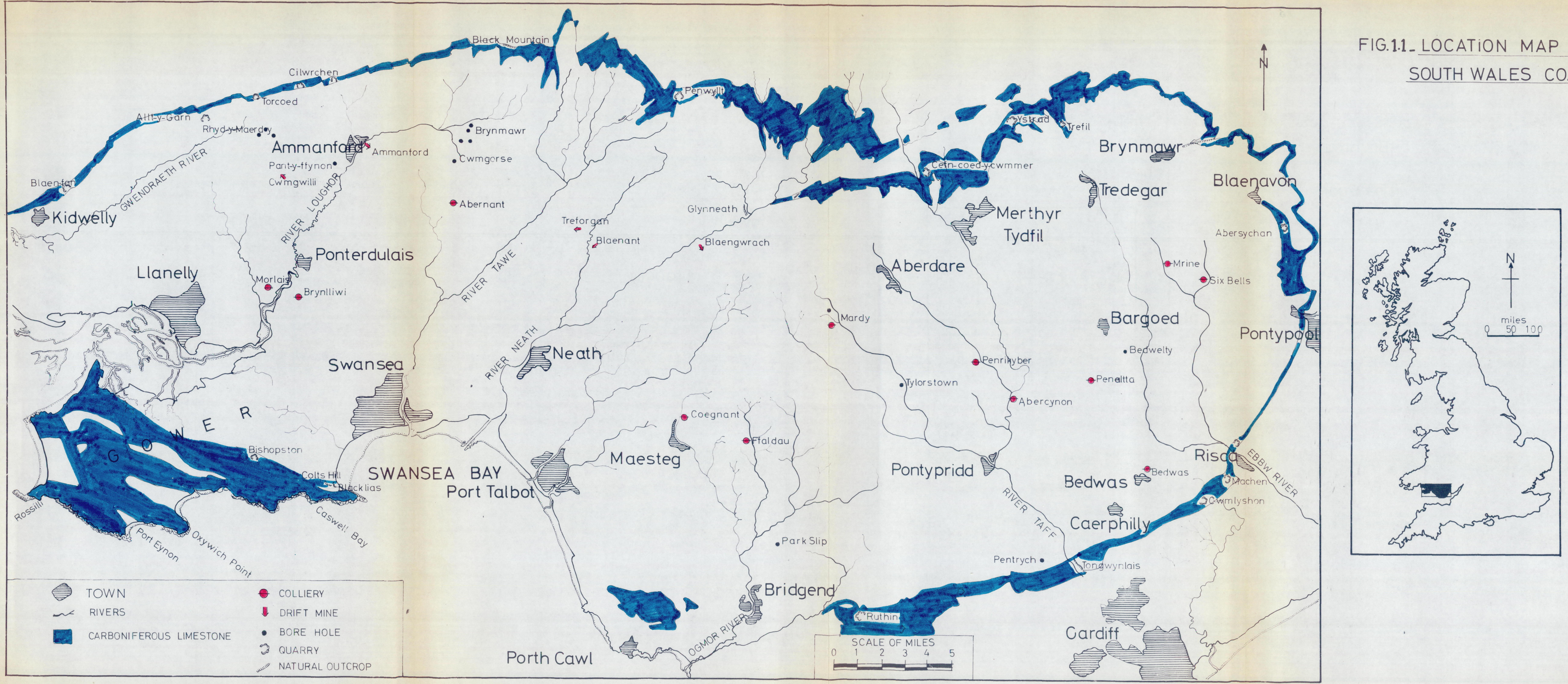


FIG.1.1_ LOCATION MAP OF SOUTH WALES COALFIELD

the high plateau of the region. This plateau is intersected by numerous deep valleys that render the principal coal seams more accessible. Most of the collieries, the mining towns and the main lines of communication lie along the valley floors, especially in the central and eastern parts of the Coalfield.

The Coalfield is sharply delineated by the differential erosion of hard and soft beds. The grits and conglomerates of the upper Old Red Sandstones, tough and resistant, form the summit of Pen-t-fan at 2,906 ft. in the Brecon Beacons, the highest in South Wales. Dipping gently southwards, they form a bold and almost unbroken escarpment which runs for 30 miles from Banau Sir Gaer eastwards to the Black Mountains and then at lesser heights continues southwards by Abergavenny and Pontypool to the Ebbw and Taff valleys. Within the major escarpment are parallel ridges rising to 2,000 ft. in several summits, formed by the Carboniferous Limestone and the Millstone Grit, which dip under low ground along the outcrop of softer shales of the Lower and Middle Coal Measures, an outcrop drained by a number of subsequent strike streams. In their turn, the shales dip beneath the thick Pennant Sandstone, which rises in a steep and forbidding scarp to heights of nearly 2,000 ft. in Craig-y-Llyn, and which forms within the Coalfield an undulating plateau, unbroken except by narrow deeply incised valleys. The rivers of the coalfield, cutting through the escarpment, follow courses that bear little relationship to the geological outcrops and have all the appearance of being superimposed. Only the Neath and the Tawe flow for a considerable distance along structurally determined lines.

The upland regions of the northern part of the South Wales Coalfield present a general similarity of aspect because they have been affected in similar ways by the erosive events of a post-Carboniferous geological history. They form a geomorphic unit that is sharply demarcated from the low country of the Vale of Glamorgan, of Gower and of the coastal regions of Carmarthenshire and Pembrokeshire.

The coastal tracts have been submerged beneath the sea intermittently since early Mesozoic times and are the eroded relics of low-level platforms benched by marine denudation. The lithological varieties of the rocks forming the coast area have controlled the coastal geomorphology of the area. In the Gower Peninsula, massive Lower Carboniferous Limestone tends to be the dominant element of the headlands and the steep cliffs. Carmarthen Bay is eroded partly in upper Carboniferous Shales, partly in Red marls of the Old Red Sandstone. The alternations of bay and headland in Gower coincide with Millstone Grit Shales and Lower Carboniferous Limestone outcrops.

REGIONAL GEOLOGICAL SETTING

Regional Stratigraphy:

The Carboniferous rocks of the South Wales Coalfield are represented by the following general succession and almost entirely exposed as shown in Figure 1.2:-

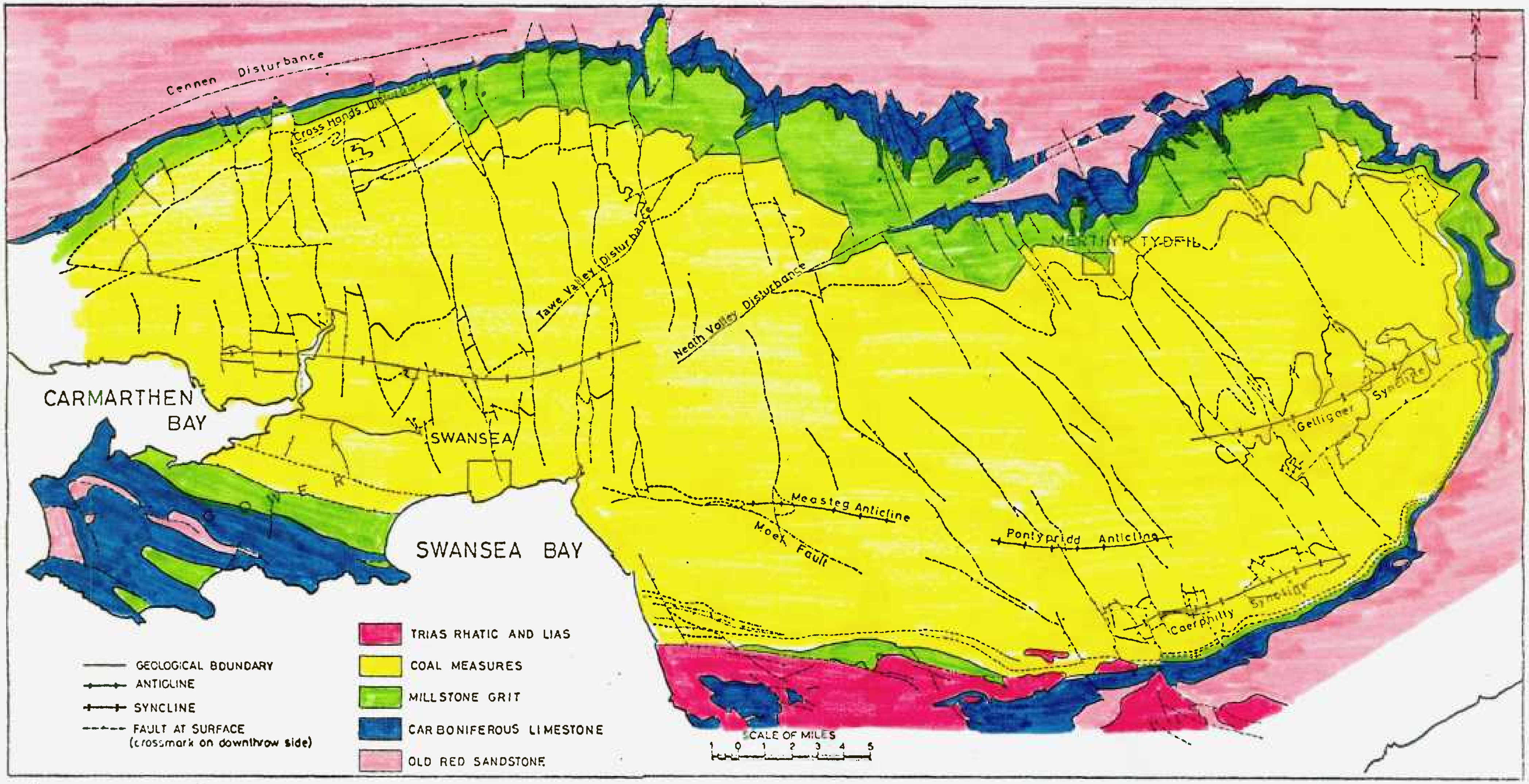
Upper Carboniferous	Coal Measures	Upper Coal Measures
		Middle Coal Measures
		Lower Coal Measures
	Millstone Grit	Farewell Rock
		Middle Shales
		Basal Grits
Lower Carboniferous	Carboniferous Limestone	Upper Limestone Shales
		Main Limestone
		Lower Limestone Shales

(A) Carboniferous Limestone Series

The outcrop of the Carboniferous Limestone Series marks the outer limit of the South Wales Carboniferous Basin. It presents a relatively thin outcrop rimming the coalfield in a narrow belt along the north and east borders. In the southern border of the coalfield and the Gower Peninsula it is represented by relatively broad outcrops.

The lithological sequence is broadly divisible into the three groups of the Lower Limestone Shales, the Main

FIG.1.2 – GEOLOGICAL MAP OF SOUTH WALES COALFIELD.



Limestone and the Upper Limestone Shales. The Carboniferous Limestone Series is most completely developed along the southern outcrops, where the broad lithological groups can be recognised. There is a consistent north and north-eastwards thinning which is accompanied by marked changes in lithology. The pure limestones of the south are replaced by more sandy and terrigenous deposits in the north, particularly in the upper part (S_2 and D zones). In the Forest of Dean the S and D zones are formed mainly of pure terrigenous sandstone (Drybrook Sandstone).

Dixon and Vaughan (1911) showed that the strata are more precisely subdivided on the basis of the fossils found in different types of limestone into the following zones:-

6.	Dipunophyllum zone	(D)
5.	Seminula zone	(S_2)
4.	Upper Caninia zone	(C_2S_1)
3.	Lower Caninia zone	(C_1)
2.	Zaphrentis zone	(Z)
1.	Cleistopora zone	(K)

Application of zonal subdivision has demonstrated unconformity between the lower parts (C_1 and C_2) in Gower, but of greater significance is the fact that much of the zonal succession of the Carboniferous Limestone is not represented in the north-western portion of the coalfield owing to the greater development of the unconformity referred to.

1. Cleistopora Zone (K):

This zone is approximately conterminous with the Lower Limestone Shale rocks, consisting of alternations of calcareous shales and thin limestones. The whole zone bears evidence of accumulation in shallow water into which much terrigenous detritus was being brought. Although the Lower Limestone Shales show little lateral change in lithology they vary greatly in thickness. In Gower they are about 500 ft. thick, but eastwards they decrease to 350 ft. near Bridgend, to 250 ft. in the Taff valley, to 120 ft. in the Ebbw valley, and to 100 ft. on the north-east crop near Abergavenny.

2. Zaphrentis Zone (Z):

Rocks belonging to this zone consist mainly of highly fossiliferous crinoidal limestones which mark the beginning of the "Zaphrentid-phase" deposits that continue into the overlying Lower Caninia zone and are not easily distinguishable from the higher beds. In the Vale of Glamorgan the beds become dolomitized as they are traced eastwards and between the Taff and Ebbw valleys they consist wholly of crystalline dolomites. Rocks of the Zaphrentis zone progressively decrease in thickness from over 500 ft, in south-western Gower to 300 ft. in eastern Gower and the Bridgend district, and to about 150 ft. in the Ebbw valley. Oolites interrupt the sequence of crinoidal limestones at Pendine, Gower and the Vale of Glamorgan. From Blorenge westwards along the north crop they become dominant in the zone, the beds being called the oolite group and only occasional layers are richly fossiliferous.

3. Lower Caninia Zone (C_1):

In the Gower and Bridgend districts the rocks of this zone fall into two lithological formations, namely, Caninia oolites and Laminosa dolomites. Laminosa dolomites are dark grey or black, finely crystalline dolomites. The lower part of the laminosa dolomites consists of about equal proportions of dolomites and of unaltered limestones; the limestones are rich in corals and also contain some crinoid-cups. The upper part is normal in its dolomitization, but includes occasional chert nodules and beekitized fossils. The Caninia Oolite as a rule is a remarkably pure limestone, distinguished from neighbouring horizons by its light-grey, often white colour and thick bedding.

In the eastern outcrop along the south crop between the Bridgend district and the Ebbw valley, Dixey and Sibley

(1918) showed that the fabrics of the laminosa dolomites encroach upon the overlying and underlying strata, so that near Cardiff and Newport the whole of the C_1 zone is an unbroken group of secondary dolomitized limestones. At the same time the zone is much reduced in thickness from about 500 ft. in Gower and Bridgend to about 350 ft. in the Ebbw valley.

4. Upper Caninia Zone (C_2S_1):

This zone consists of highly fossiliferous limestones, many crinoidal but some oolitic or porcellaneous. The upper beds are locally more oolitic than the lower and may merge upwards without marked break into the thick oolites of the Seminula Oolites. In most of South Wales there is an abrupt but otherwise normal junction with the Caninia Oölite beneath, but in some localities as in eastern Gower and in the Miskin district of the Vale of Glamorgan, the upper surface of the Caninia oolite is uneven and slightly eroded and is followed by a thin group of shales and fine-grained compact porcellaneous limestines, the Calcite-Mudstone group, that locally is the first formed member of the upper Caninia zone.

At maximum thickness the zone exceeds 600 ft. in Gower and in the west of the Vale of Glamorgan. Traced eastwards from the Bridgend district along the south crop to the Taff and Ebbw valleys the zone is reduced to some 200 ft.

near Risca. At the same time there is a great change in lithology, the crinoidal beds of the west being progressively dolomitized.

5. Seminula Zone (S_2):

The most widespread of all the Lower Carboniferous Limestone zones, the Seminula zone, is dominated by massive coarse oolites. Pisolites, algal limestones and calcite mudstones are intercalated in the upper part and are particularly strongly developed along the north crop. The zone is about 800-1,000 ft. thick in Gower and the Vale of Glamorgan. Along the north crop, however, its thickness does not exceed 400 ft. The thinning is not accompanied by any marked lithological or faunal changes, except that sandy beds appear both at the base and in the upper beds beneath the Dibunophyllum zone.

6. Dibunophyllum Zone (D):

The beds of the Dibunophyllum zone characteristically consist of neritic crinoid, coral and brachiopod limestones, many richly foraminiferal, some with oolites. In Gower, Dixon and Vaughan (1911) divided this zone into three lithological units, namely, subzone D_1, subzone D_{1-2} and subzone D_{2-3}. The D_1 subzone or the lower Dibunophyllum zone is 400-450 ft.

thick and consists chiefly of light grey, thick-bedded limestones, many partly dolomitized. The limestones are predominantly of the type characterised by the remarkable "pseudo-brecciated" structure, which is in fact a partly dolomitized foraminiferal limestone. Subordinate to the pseudobreccias are limestones of ordinary types. In places a coarse-grained oolite marks the base of the zone. This subzone is further distinguished by the presence of thin coal seams.

D_{1-2} subzone, of about 120 ft. thick, consisted mainly of thickly bedded, fine-grained limestones including oolite. D_{2-3} subzone is equivalent to the Upper Limestone Shales Group. It consists of limestones and shales with cherts, the whole known locally as "black lias". The limestones are thin-bedded, almost invariably dark-grey or black. At Oystermouth they reach 175 ft. thick.

The whole zone reaches a thickness of about 800 ft. in Gower. Along parts of the main north crop, the zone is progressively overstepped by the Millstone Grit. Between Llandybei and the Tawe valley and at Kidwelly all of the three subzones can be recognised; they show great attenuation, reaching a maximum thickness of less than 300 ft. The thinning of the zone along the north crop is accompanied by notable lithological changes. The Lower Dibunophyllum subzone consists mainly of a massive cross-bedded oolitic limestone, resting on a calcareous sandstone, the honeycombed sandstone.

(B) Millstone Grit

The Officers of the Geological Survey divided the Millstone Grit of the north crop of the South Wales Coalfield into three groups, namely, Basal Grit, Shale Group and Farewell Rock.

In the northern outcrops the fullest Namurian development is found between the Laughor and Tawe valleys, where the broad sequence of stages is complete with a thickness of about 800-900 ft. Westwards the thickness is reduced to about 600 ft. near Kidwelly. Eastwards the changes in the Namurian sequence are more rapid and more radical. The thickness is reduced to about 450 ft. in the Neath valley, to about 300 ft. in the Taff valley, to 150 ft. near Brynmawr and to less than 100 ft. to the east. Along the east crop southwards to Risca the series remains very thin. Further south, few details are known as the rocks swing into the south crop, but Squirrel and Downing (1964) have recorded a thickness of 100 ft. in the Ebbw valley, of 140 ft. near Rudry and of 175 ft. in the Taff valley. There is a great expansion to 900 ft. towards Bridgend. The sequence in Gower, described by Dix (1931) is exceptionally thick, nearly 2,300 ft.

(C) Coal Measures

The Coal Measures of South Wales consist almost wholly of terrigenous detritus derived from nearby sources and carried into a shallow subsidiary trough of sedimentation by rivers from a land-mass lying, as in Avonian and Namurian times, mainly to the north. The deposits appear to have been laid down under estuarine or fresh water conditions as alluvial muds and sands, swamp clays and other delta fans and aprons; the coarser layers are often cross-bedded in lenticular units; and the channelled washouts are common. Marine beds are few and thin, and are restricted to the lower part of the sequence. The recurrent coal seams were formed as thick beds of waterlogged peaty humus in swamps and marshes supporting a luxuriant vegetation, through which tributaries and distributaries of large rivers meandered.

Sedimentation of the Coal Measures, as of the Millstone Grit, was in rythmic cycles. In each typical cycle (cyclothe~~r~~m) a coalseam is succeeded by fine-grained shale, with goniatites, and bivalves, brachiopods as a sign of sharp subsidence and a marine incursion. There follow shales of increasingly shallow-water and non-marine character, with a fauna of mussels, which in turn gradually pass upwards into sandy shales, sandstones and sometimes grits and conglomerates.

The close of the cycle is marked by very shallow-water muds, now converted to underclays and rootlet beds by the growth of forests on them, which are overlain by a coal seam initiating the next cyclothem. Usually the full cyclothem is not represented, one or more elements being omitted.

Lower and Middle Coal Measures

Over a great part of the coalfield the Coal Measures as a major lithological suite are dominated by shales in the lower part and by sandstones in the upper part. The shales of the lower part of the Coal Measures crop out in major development at the foot of the "Pennant" escarpment around the coalfield. They also outcrop in the floors of some of the deeply entrenched valleys of the Rhondda. They consist for the most part of a monotonous series of grey, blue and black pyriteous and micaceous shales often imperfectly laminated. Usually there is only a minor development of sandstones, but over much of the coalfield a hard siliceous sandstone or grit forms a characteristic bed between the Red and the Stwrin coals or their equivalents. The measures contain the greatest number, the thickest and the most important coal seams in South Wales.

The Lower Coal Measures are characterised by the abundance of iron ore occurring as a penecontemporaneous segregation of argillaceous chalybite (clay ironstone) in nodules or bands arranged parallel with the stratification.

At maximum the thickness of the Lower and Middle Coal Measures reaches some 3,000 ft. in the Swansea district, but it diminishes as the group is traced towards the north-east and east. In the Maesteg district it is about 2,100 ft., near Merthyr Tydfil and in the south-east near Cardiff 1,400 ft. In the Pontypool district and the outcrops about Abertillery they are slightly less than 800 ft.

Upper Coal Measures

Over most of the coalfield there is a clear line of division, both lithological and topographical, between soft shales of the Middle Coal Measures and the massive scarped sandstones of the Pennant Measures. Typically, the Pennant Measures include thick massive sandstones (Pennant sandstone) much used for building. Work by Bluck and Kelling (1963) on the abundant washouts and channels in the rocks suggests an expected derivation of the detritus mostly from the north and east in the Lower and Middle Coal Measures, but contrary to this inferred palaeogeography and thickness variations, a wholly unexpected derivation mainly from the south occurs in the Pennant Measures, implying a land-mass nearby to the south where the Bristol Channel now lies.

Using some of the more promiment and persistent coal seams as convenient markers, Woodland et al. (1957) have recently reorganised and systematised the classification of the Upper Coal Measures, hitherto chaotic, into two major groups and six subgroups:

2. <u>Upper Pennant Measures</u>: the measures upwards from the Hughes Seam, including the beds, the highest preserved in South Wales, formerly called the Upper Coal Series.

    c) Grovesend Beds, the Mynyddislwyn Seam at the base.
    b) Swansea Beds, the Swansea Seam at the base.
    a) Hughes Beds, the Hughes or Wenalt Seam at the base.

1. <u>Lower Pennant Measures</u>: the measures containing the main beds of typical massive Pennant sandstones.

    c) Brithdir Beds, the Brithdir Seam at the base.
    b) Rhondda Beds, the No. 2 Rhondda Seam at the base.
    a) The Llynfi Beds,their base at the top of the Upper Cwmgorse Marine Band.

The Upper Coal Measures exceed 5,000, perhaps 6,000 ft. in thickness in the western part of the coalfield about Swansea. They are reduced to about 3,500 ft. in the Dyffryn trough near Neath, and to about 2,000 ft. in the Llantwit syncline. At the same time there is a great reduction westwards in the development of massive sandstones.

Regional Structural Geology:

The majority of the structures in the South Wales area were the products of the Variscan orogeny. This includes earth movements which took place during Devonian and Carboniferous times, but which culminated in important mountain building late in Carboniferous times after the deposition of the Coal Measures. In Britain and in Ireland the intensity of deformation is greater in the South than the North.

Four main Variscan orogenic phases have taken place in South Wales during Carboniferous times; these are:-

(a) Nassanic phase (pre-Upper Caninia Zone)
(b) Sudetic phase (pre-Namurian)
(c) Malvernian phase (pre-Morganian)
(d) Asturic phase (pre-Stephanian)

Many of the coalfields of Britain appear to have been parts of a wider spread of Upper Carboniferous deposits.

Their separation today was the result of Variscan flexuring and subsequent erosion of the upfolded areas. Owen (1971) suggested that the South Wales Coalfield achieved an identity during Westphalian times (~~Austric~~ Asturic phase).

Structural Elements of the South Wales Coalfield

The South Wales Coalfield consists essentially of a synclinical depression which extends westwards from Monmouthshire, through Glamorgan and into Carmarthenshire, where, after passing under Carmarthen Bay, it continues to form the Pembrokeshire Coalfield to the coast at St. Bride's Bay. The structure is that of a rather complex downfold containing a number of flexures which tend to diminish when traced for any distance and which are replaced by other structures, folds and faults arranged en echelon in a general east-west direction. Numerous powerful faults cross the basin cutting through the structures. The basin is characteristically asymmetric, with a gently dipping northern rim (10 degrees) and a more steeply inclined southern border with a dip of 45 degrees and over.

1. Folds:

Folding is most prominent in the eastern region of the coalfield. The most remarkable feature of this region is the Pontyprid anticline which, running east-west, swings northwards and dies out towards the east crops. It separates two basins containing upper groups of the Upper Coal Measures; to the north the Gelligaer syncline and to the south the Llantrisant-Caerphilly syncline. The Pontyprid anticline brings Middle Coal Measures to the surface over a restricted portion of its length. Westwards this anticline is less evident and appears to be replaced in part by a number of powerful east-west trending strike faults.

Maeros

The ~~Meiro'~~s anticline, a much-faulted structure, lies south west of the Pontypridd field and is separated from it by the Ton-yrefail syncline.

The Maesteg anticline brings Lower Coal Measures to the surface from which the measures dip northwards into the broad Glynn Gorrwg syncline.

Generally, all of these folds are parallel to each other and to the Variscan trend.

2. Faults:

The faulting in the South Wales Coalfield is of three main types, namely, east-west faults, north west-south east faults, and north east-south west faults.

East-west faults:

This is the least common type of fault and they are present in the eastern and western regions of the coalfield. They are usually associated with the folds. Many are overthrust faults with considerable throw; normal faults may also follow this general direction, of which the Moel Gilan fault has a southerly downthrow of 600 ft.

North west-south east faults:

The predominant fault trends are in a general north-west and south-east direction at fairly regularly spaced intervals across the coalfield and tend to occur in pairs, often producing trough-faulted structures. Within the coalfield such faults may reach considerable dimensions, but their effect on the outcrop of the margin of the basin is often relatively small. In the western region of the coalfield, many of these faults can be traced in the upper and into the lower coal measures on the north crop, the larger showing vertical displacements of between 1,000 and 2,400 ft. A characteristic of all the faults is the variation in throw along their length.

North east-south west faults:

These are the most powerful faults, usually associated with the compression belts of the western region of the coalfield. Three main disturbance belts of this trend have affected the coalfield, namely, Neath disturbance, Tawe disturbance, and Cennen disturbance. These three narrow belts of folding and faulting trend NE-SW from the eastern side of Wales into the main coalfield (in the case of the Neath and Tawe belts) or to skirt its north-western rim (in the case of Cennen). Folds along the belts are narrow and often impersistent, but the faults are usually very persistent. Those along the Neath and Tawe belts appear to be major wrench faults of sinistral displacement, but that along the Cennen line is of more complex history with appreciable thrusting along its more western length. The trend of these disturbance belts suggests that they are inherited from Caledonian fractures in the pre-Carboniferous rocks.

COAL OF SOUTH WALES

The South Wales Coalfield differs from other British Coalfields in its wide range of coals, from bituminous through semibituminous to anthracite. It is particularly in the widespread occurrence of high rank sub-bituminous and anthracite coals that the range differs.

Many of the physical and chemical properties of coal are interrelated and some of the more important can be used as indices of the general properties of the coal. These properties are to some extent related to the degree of metamorphism of the coal, i.e. the coal rank. Of these properties are the percentage of the volatile matter, the fixed carbon ratio and the carbon-hydrogen ratio.

Regional Variation of Coal Rank:

A striking feature of the South Wales Coalfield lies in the extent and regularity with which the limits of the previously discussed rank vary across the coalfield from one place to another. Fig. 1.3 is the isovol map of the nine-feet seam as produced by the National Coal Board and Fig. 1.4 is the isocarb map of the same seam produced by Mohafez (1966). These two maps illustrate the general and regular pattern of the regional variations of the coal rank in the South Wales Coalfield. The isovols and isocarbs run approximately parallel to the southern and eastern borders of the coalfield. The coal seams progressively increase in rank towards the West. It is clear that the rates of change vary in different parts of the coalfield. The anthracite region to the north-west provides a large area in which the total variation in the volatile constituents is only about 5 percent. A similar pattern of regional variation is exhibited by seams higher in the sequence but with progressive shift to the west and north.

FIG.1.3– COAL RANK AND ISOVOL MAP OF SOUTH WALES COALFIELD
(based on Nine feet seam.)

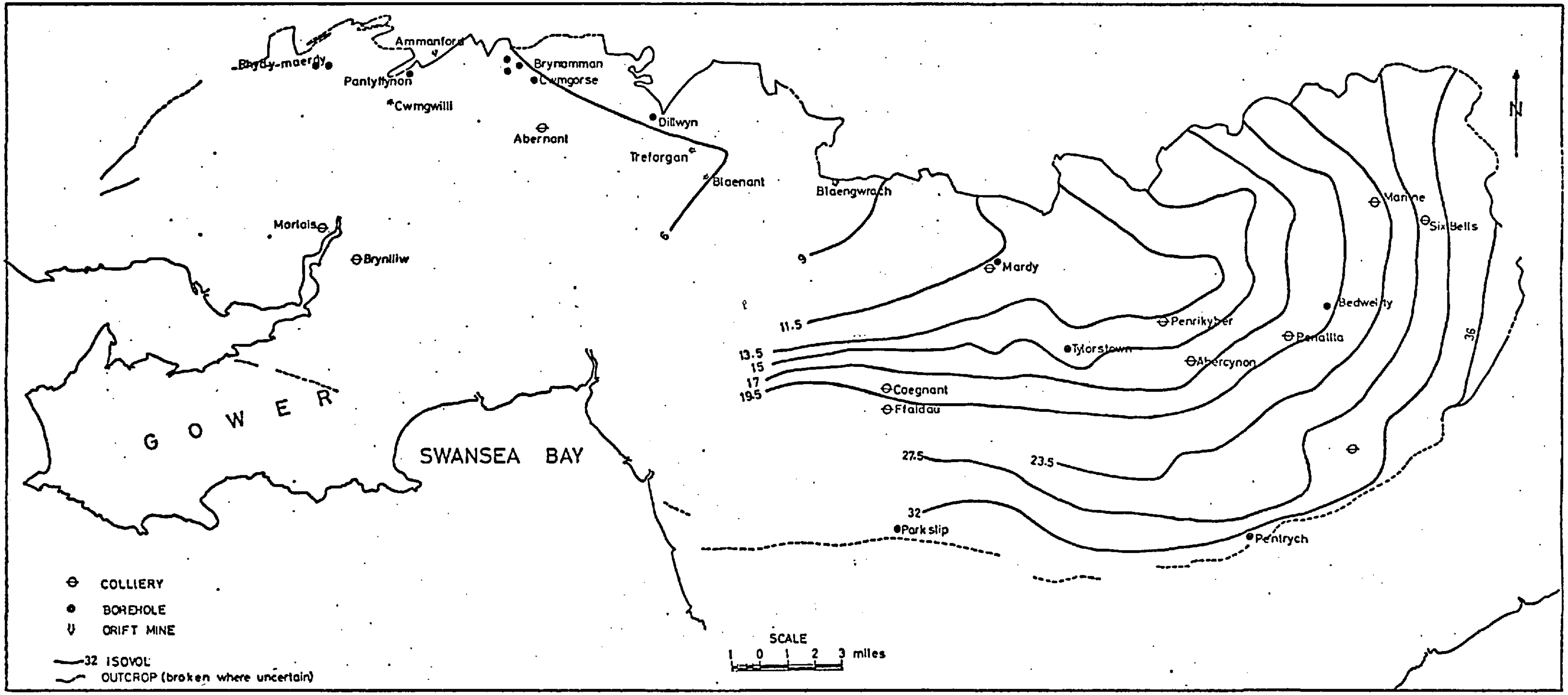
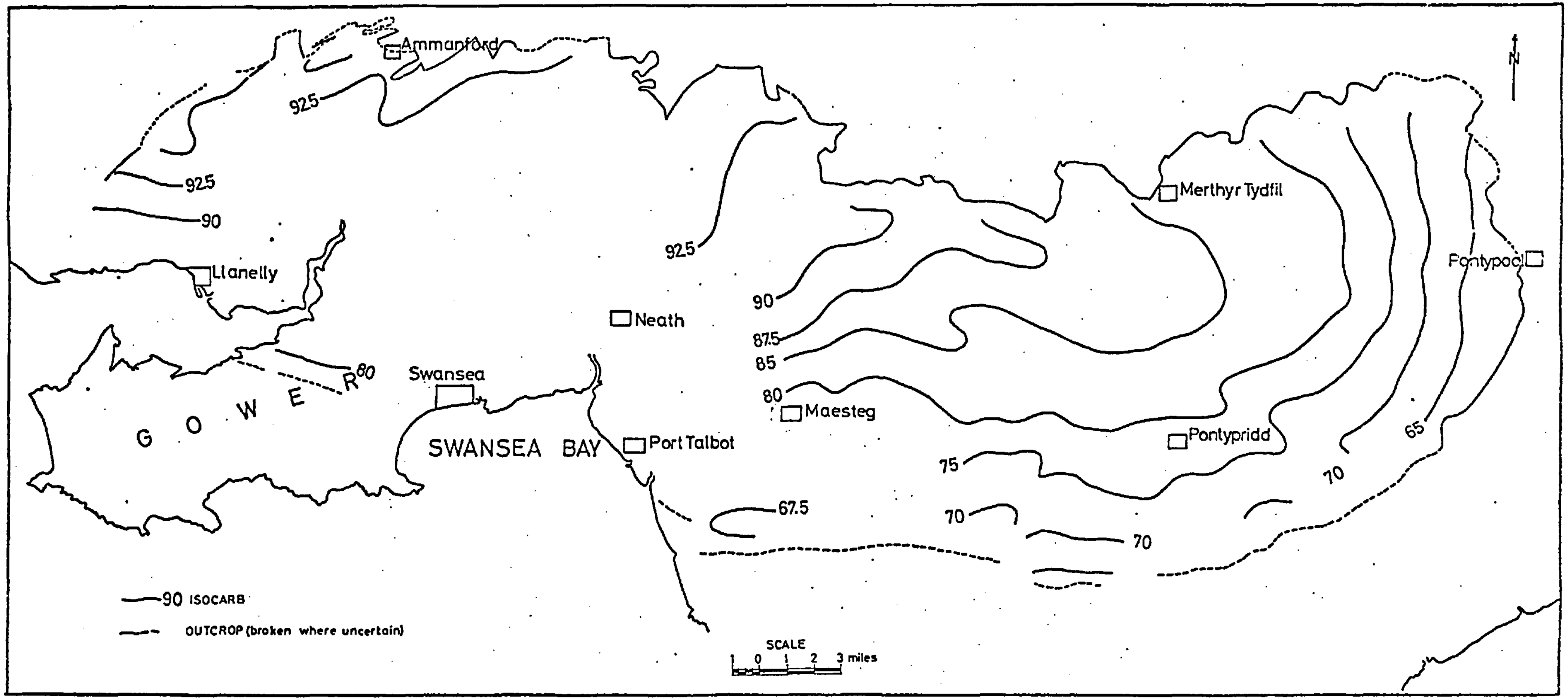


FIG. 1.4- ISOCARB MAP OF SOUTH WALES COALFIELD. (After Mohafez, 1966)
(based on Nine feet seam - dry & ash free)



Vertical Variation of Coal Rank:

The dominant variation in coal rank in the major seams is regional, but significant differences may occur between the upper and lower seams of the main group in any one locality. When seams in the main productive portion of the Lower Coal Measures are considered at a given colliery, the changes are small, frequently not more than 2-3 percent. The changes are more noticeable between the coal seams in the Pennant sandstones and those in the Lower Coal Measures, especially in the western part of the coalfield. The vertical changes are less significant in the eastern part of the coalfield.

Objects of the Present Study:

The objectives of the present investigation were twofold. The first involved a study of the diagenesis of the various lithotypes of the Carboniferous rocks of the South Wales Coalfield. The second objective was concerned with the incipient metamorphism of these rocks and its relation with the carbonization of the associated coal seams.

For this purpose the following studies were carried out:-

1. Detailed petrographical study of the various rock units forming the Carboniferous sequence in order to

establish the following:

(a) Identification of the various lithotypes and sub-lithotypes, and the description of their petrographic characteristics.

(b) To know the various diagenetic processes that affected the original sediments, and therefore their diagenetic history.

(c) The recognition of incipient metamorphic fabrics and their regional distribution in the various lithotypes.

2. Qualitative and quantitative X-ray diffraction analysis of clay minerals separated from the various lithotypes. The main purpose of this study was to establish a relationship between the degree of incipient metamorphism and the variations in the clay minerals.

3. Detailed petrographical study of the polished surfaces of the organic matter dispersed in the various lithotypes. This study involved the reflectivity measurements of the vitrinite grains.

4. Measurements of the degree of carbonization of spores and pollen grains that separated from the various lithotypes.

The South Wales Coalfield has been chosen as a suitable area for studying incipient metamorphism in the sedimentary rocks for the following reasons:-

1) As indicated from the isocarb and the isovol maps of South Wales Coal, the area has been gradually and progressively metamorphosed and this is demonstrated by the wide range of the fixed carbon content of the coal seams (from about 65% in the east and the south to about 95% in the north-west).

2) The Carboniferous sequence in the area contains various types of sedimentary rock, and this was necessary to show the response of the different rock types to the various grades of incipient metamorphism.

Sampling:

For the purposes of the present investigation, about one thousand samples have been collected from the Carboniferous rocks of South Wales. Some samples were collected also from the Carboniferous rocks of the Forest of Dean for the purpose of correlation. Sampling was carried out in such a way as to collect representative samples from the various lithotypes, and from different localities in order to demonstrate regional and vertical variations.

Sampling of the Carboniferous Limestone Series was facilitated by the frequent number of quarries scattered in the limestone terrains in South Wales, Fig. 1.1. Samples were also collected from accessible valley escarpments and road cuttings. Cores from two bore holes were provided by the Ystrad quarry authorities. Fig. 1.5 (enclosures) is the lithological logs of the Carboniferous Limestone Series in the localities studied; sample numbers are located on these logs.

Samples of the Millstone Grit Series were collected from the natural outcrops mainly.

Middle and Lower Coal Measures rocks were mainly sampled from coal mines. Seventeen collieries in the South Wales Coalfield have been visited and samples were collected from rock outcropping in the underground tunnels. These are: Abernant, Abercynon, Ammanford, Bedwas, Bleanant, Bleangwrach, Brynlliw, Coegnant, Cwmgwilli, Ffaldau, Mardy, Marine, Morlais, Penallta, Penrikyber, Sixbells and Treforgan. In the Forest of Dean Coalfield five collieries have been visited; these are: Cinderford, Colford, Hinton, Mableford and Yorkly.

Upper Coal Measures rocks were mainly sampled from cores provided by the National Coal Board. The following are the localities of these bore holes: Abercynon, Bedwality, Brynamman, Cwmgorse, Dillwyn, Mardy, Pantyffnon, Parkslip, Pentrych, Rhydymeardy and Tylorstown, Fig. 1.1.

Fig. 1.6 (enclosures) is the lithological logs of the Coal Measures Series in the localities studied; samples numbers are located on these logs.

CHAPTER II

PETROGRAPHY OF THE CARBONIFEROUS LIMESTONE SERIES

PETROGRAPHY OF THE CARBONIFEROUS LIMESTONE SERIES

INTRODUCTION

Very little work has been published on the petrography of the Carboniferous Limestone rocks of the South Wales coalfield. Dixon and Vaughan (1911), Dixey and Sibley (1918) briefly discussed the petrographic characteristics of the various lithotypes in their general geological survey of part of the South Wales coalfield. Recently, Whitecombe (1970) studied the diagenesis of the Carbonate rocks of the Lower Limestone Shale Group, and George (1972) proposed a classification of the Avonian limestones in the Bristol area and in South Wales. MacQuown and Bloxam (1972) gave petrographic descriptions of certain parts of the Middle Visean Section that crop out on the Gower Peninsula, in their study of the depositional history of the Middle Visean limestones in the Bristol area and parts of South Wales.

In this Chapter the author attempts to describe the regional petrographical characteristics of the various lithologies that comprise the Carboniferous Limestone Series in South Wales and the Forest of Dean.

CLASSIFICATION

The classification followed in this work is that of Folk (1959). The following lithotypes have been identified in the Carboniferous Limestone Series: (a) terrigenous rocks; (b) micrites, (c) biomicrites; (d) biosparites; (e) oolitic limestones; and (f) dolomite. Each lithotype is classified into various sublithotypes.

PETROGRAPHY OF LITHOTYPE-A: TERRIGENOUS ROCKS

(Plate 1)

Terrigenous rocks are predominant in the Lower Limestone Shale Group. They occur also as thin beds in the Seminula zone of the Main Limestone Group in the South Wales Coalfield. In the Forest of Dean Coalfield they are known as the Lower Limestone Shale Group and the Drybrook Sandstone.

The terrigenous rocks of the Carboniferous Limestone Series may be divided into four sublithotypes, namely, shale, seatearth, siltstone and sandstone.

Sublithotype A-1; Shale

The Lower Limestone Shale of South Wales and the Forest of Dean are represented mainly by this sublithotype, which is dominant also in the Upper Limestone Shale Group of the former area. Usually it contains intercalations of crinoidal biosparites.

Macroscopically, the shales comprising the Lower Limestone Shale Group are fissile and of dark grey colour.

In the Upper Limestone Shale Group the bedding becomes relatively thicker and the rock is almost black in colour, with bands very rich in brachiopod shells.

Microscopically, the rock is made up mainly of clay minerals with up to 10% detrital grains. The clays consist of very fine shreds of illite and mixed layer minerals. The average length of the shreds is usually about 7µ, and they usually lie parallel to the bedding of the rock. Silt-sized quartz and muscovite grains represent the detrital constituents of the rock. The quartz grains are moderately sorted and subangular to subrounded, and the muscovite flakes are oriented parallel to the bedding. The latter commonly show partial kaolinitization which seems preferentially to attack the cleavage planes of the flakes. Some muscovite flakes are completely kaolinitized.

Orthochemical minerals are represented mainly by ferroan dolomite which is abundant in the shales of the southern parts of the South Wales Coalfield, and comprises up to 15 percent of the rock. Ferroan dolomite occurs as single euhedral rhombs of about 20µ in average diameter, but reaching 90µ in size in some samples.

The shales from the Upper Limestone Group of South Wales (Gower) are more calcareous, very rich in organic matter and diagenetic pyrite. The fossiliferous bands are rich in brachiopod shells and crinoid debris. Organic matter is finely disseminated in the clayey ground mass.

X-ray analysis of the $< 2\mu$ fraction has revealed that illite and irregular mixed layer illite-montmorillonite are the main clay minerals present, with small amounts of kaolinite.

Sublithotype A-2; seatearth

Seatearth occurs as thin bands of about one to two feet in thickness. It is present as thin intercalations in the Seminula zone of the Gower Peninsula, "Ilston Quarry", and at the contact between the S_2 and D_1 zones at "Colts Hill Quarry", where it overlies partially dolomitized biomicrite (pseudobreccia) and is overlain by a thin coal seam about 6 in. thick. Seatearth is present also in the northern outcrops at the contact between the S_2 and D_1 zones.

Macroscopically, it is a light grey, friable rock with abundant black rootlets. Due to its friability it is very difficult to make a thin section of this rock. However, X-ray analysis of the $< 2\mu$ fraction showed the rock to consist mainly of illite.

Sublithotype A-3; siltstone

Two types of siltstone have been identified, namely, a clayey siltstone and a calcareous variety.

Clayey siltstone occurs in the upper part of the Lower Limestone Shale Group of the Forest of Dean. It is thin-bedded, microcross bedded, hard and pale brown in colour, with frequent burrows.

Microscopically, it consists mainly of well-sorted, subangular to subrounded quartz grains, muscovite flakes and plagioclase, bound together by a clay matrix. Dolomite is present as euhedral rhombs about 0.15 mm. across. It is sporadically distributed, comprising up to 10 percent of the rock mass.

Calcareous siltstone occurs as thin bands intercalated in the limestones and the calcareous sandstones of the S_1 zone in the northern outcrops of the South Wales area.

Microscopically, it consists of subsngular to subrounded quartz grains of about 0.05 mm. average diameter. The quartz particles are cemented by a fine-grained sparry calcite. Silt-sized faecal pellets and shell debris are commonly present (Plate 1, Fig. 1).

Sublithotype A-4; sandstones

Two types of sandstone are found in the Carboniferous Limestone Series. These are the Drybrook Quartzarenite and the Calcareous Sandstones.

A. Drybrook Quartzarenite: (Plate 1, Figs. 2, 3 & 4)

Quartzarenite occurs only in the Forest of Dean Coalfield and is represented by the Drybrook Sandstone. It reaches about 700 ft. in thickness and is divided by about 200 ft. of thick-bedded oolitic limestone intervals into an upper unit and a lower unit. The two units are lithologically different. The quartzarenite of the lower unit is about 100 ft. in thickness, thin-bedded and varies in colour from various shades of grey to white and cream. It contains bands of shale and mudstone, and is frequently burrowed.

The upper unit is thickly stratified with large scale cross-bedding; it is friable and varies in colour from red, pink to indian red.

Microscopically, the lower unit is an ill-sorted, fine-grained quartzarenite. Angular to subangular monocrystalline quartz grains commonly form about 88 percent of the sand. Rock fragments, mainly chert, average about 4 percent. Micas are represented by rare muscovite flakes. The quartz grains usually have silica overgrowths, and a ferruginous clay matrix binds the framework together.

The upper unit is a moderately to well-sorted, medium-grained quartzarenite, consisting of 76-89 percent mono- and polycrystalline quartz grains. Rock fragments are rare, and amount to about 1 percent, mainly chert. Quartz overgrowths on the detrital quartz grains are the main cementing agent of the framework. The interstitial pore spaces generally contain well-crystallized vermicular kaolinite aggregates, which form up to 16 percent of the sandstone. Table 2.1 shows the modal analysis of the two units of the Drybrook quartzarenite.

TABLE 2.1 MODAL ANALYSIS OF THE DRYBROOK SANDSTONES

	Upper Unit			Lower Unit		
	1023	1024	1027	1030	1031	1032
Quartz:						
Monocrystalline	85.5	74.2	74.0	84.0	88.0	85.5
Polycrystalline	3.6	1.6	4.0	-	2.0	1.0
Total:	89.1	75.8	78.0	84.0	90.0	86.5
Chert	1.4	1.0	1.5	4.4	4.0	3.5
Mica	-	-	-	1.0	1.5	1.0
Clays	-	-	-	-	2.5	1.0
Organic Matter	-	-	-	-	2.0	2.0
Silica Overgrowths	6.5	7.0	6.0	8.8	-	5.0
Authigenic Kaolinite	3.0	16.2	14.5	1.0	-	1.0
Average Diameter	0.21 mm.	0.21 mm.	0.25 mm.	0.1 mm.	0.15 mm.	0.15 mm.

In general the fabric of the Drybrook quartzarenite is that of a diagenetically immature quartzarenite. The rock is supported structurally by the quartz-chert framework. Authigenic minerals, represented by quartz overgrowths, iron oxide and kaolinite form the cement. Visual estimation of the grain size distribution shows that the rock is moderately well to poorly sorted, fine to medium grained sand. The lower unit is texturally immature, as indicated by its degree of sorting and roundness, while the upper unit is texturally mature.

Mineralogy:

1. Terrigenous detritus

a - Quartz: Two varieties of quartz have been recognised, viz., mono- and polycrystalline grains. Monocrystalline quartz is the most frequent type, ranging from 76 to 90 percent. Polycrystalline quartz is rare, and varies from about 1 to 4 percent in the rock. Polycrystalline quartz grains usually consist of three or four quartz crystals of equant shape. Generally, both types of the quartz grains are unstrained. Authigenic quartz overgrowths are very common and range in frequency from 5 to 9 percent.

b - Chert is present in small amounts; it makes up from about 1 to 4 percent of the rock mass and is common in the lower unit. It is very fine-grained and pale brown in colour.

c - Mica is almost always muscovite. It occurs only in the sandstones of the lower unit, where it amounts to about only 1 percent of the rock mass. The flakes are slightly bent between the quartz grains.

d - Detrital clay minerals are present in the sandstones of the lower unit. They are represented by minute shreds of illite. X-ray analysis of the $< 2\mu$ fraction suggests that the clay minerals present are illite, illite-montmorillonite mixed layer, kaolinite and chlorite. This analysis includes authigenic kaolinite, as it is impossible to separate detrital clays from authigenic clays.

e - Heavy minerals are present in trace quantities and are represented mainly by tourmaline and zircon. The tourmaline is of the green variety and occurs as well-rounded grains. The zircon grains are also well-rounded and some are spherical.

2. Chemical Constituents

a - Authigenic silica is represented by two types; (i) secondary overgrowths deposited in optical continuity on the detrital quartz grains, and (ii) microcrystalline quartz. The secondary overgrowth quartz is clearly identifiable as the detrital grains are usually coated with an iron oxide film. Inclusions of hematite spicules are frequent in the quartz overgrowth rim. The overgrowth quartz rim is the main cementing agent in the sandstones of the upper unit. In some places former pore spaces are completely occupied by the overgrowth quartz, but in most of the rock, overgrowths have only partially filled the voids, and the rest of the space has a filling of hematite and authigenic kaolinite. That is why the rock can be easily disaggregated. Microcrystalline quartz also acts as cementing agent, particularly in the sandstones of the lower unit.

b - Authigenic kaolinite is present as well crystalline vermicular aggregates filling the pore spaces. Authigenic kaolinite is common in the sandstones of the upper unit where it comprises about 16 percent of the rock mass. The occurrence of kaolinite has been confirmed by X-ray analysis of the $< 2\mu$ fraction of the sandstones of the upper unit. Usually the authigenic kaolinite has attacked the quartz grains. In some cases kaolinite vermicules are enclosed in the quartz overgrowth rim.

c - Iron oxide is represented by the hematite spicules and is very common in the sandstones of the upper unit. It is responsible for the red colour of the rock, and occurs as minute spicules, about 10μ in average diameter. Three modes of occurrence of the hematite spicules have been noticed: (i) as very thin rims coating the detrital quartz grains; (ii) floating in the quartz overgrowth rim; (iii) filling interstitial spaces between the kaolinite vermicules.

B. Calcareous Sandstones: (Plate 1, Figs. 5 & 6)

These rocks constitute thin beds, about 3 to 4 ft. thick, within the calcarenites of the S_1 zone at Ystrad Quarry, and the D_1 zone in the Llandybei area in the northern outcrop of the South Wales Coalfield.

Macroscopically, the rock is very hard and of light grey colour. Brachiopodal shell debris is usually abundant and forms thin bands.

Microscopically, it consists mainly of well sorted quartz grains with subordinate amounts of brachiopod shell debris and oolites. The framework grains are bound together by a micrite matrix, and sparite cement, and/or dolomite. There are mono- and polycrystalline quartz grains. Monocrystalline quartz grains are more frequent. The micrite matrix is usually recrystallised, while the sparry calcite cement consists of equant calcite crystals forming a mosaic texture. Some rocks are cemented by dolomite.

Both calcite and dolomite are frequently seen to have corroded quartz grains. In some rocks the corrosion is so severe that it makes the identification of the texture of the detrital quartz grains very difficult. Poikilotopic texture developed by the enclosure of more than one quartz grain inside single calcite or dolomite crystals is frequent.

PETROGRAPHY OF LITHOTYPE-B: MICRITE

(Plate 2)

This lithotype is divided into two main sublithotypes, namely, a massive dismicrite and a thin-bedded micrite.

Sublithotype B-1; Massive Dismicrite

Massive dismicrite occurs as a thin bed of about 3 ft. thick in the D_2 zone in Gower Peninsula and Porthcawl. It was called calcite-mudstone by Dixon & Vaughan (1911). Similar beds are exposed in the northern outcrops, but there it was found that they consist of an algal biomicrite.

Macroscopically, the rock is very hard, light grey to creamy-grey in colour. It has a porcellaneous appearance, and breaks with concoidal fractures. Stylolitic seams are commonly seen.

Microscopically, the rock consists wholly of a homogenous mass of micrite with very little silt-size algal and other skeletal debris. Patchy recrystallization of the micrite is not uncommon. The recrystallized calcite crystals are equidimensional with irregular boundaries, and average about 10-15μ in size. The rock is highly vugy. The vugs are of irregular shape and range in size from fractions of millimetres to a few millimetres. The following vug types are recognised:

(i) Vugs filled with sparry calcite only. Bladed, fine-grained, scalenohedral calcite crystals fringe the wall of each vug. The rest of the cavity is filled with a coarse-grained mosaic of sparry calcite.

(ii) Vugs occupied by micrite and sparry calcite (Plate 2, Fig. 3). Each vug has a lining of drusy calcite, and micrite then accumulated in the bottom half of the vug, sometimes filling about three-quarters of the cavity. The upper surface of the micrite mass is usually flat and is covered by a thin layer of drusy calcite crystals. The upper half of the vug is filled with a clear mosaic of calcite crystals. In some, the upper half of the vug is occupied by a large single calcite crystal.

(iii) Vugs filled with vermicular kaolinite and sparry calcite. The vug is lined with a thin layer of drusy calcite crystals. The lower half has a filling of coarsely crystalline aggregates of vermicular kaolinite or dickite covered by a thin layer of fine-grained calcite. The upper half of the vug is filled with a mosaic of sparry calcite crystals or a large single calcite crystal.

(iv) Vugs occupied completely by vermicular kaolinite aggregates. These vugs are lined on the inside by drusy calcite (Plate 2, Fig. 1).

The sparry calcite infilling the cavities is highly strained and this is demonstrated by the development of lamellar twinning. The detrital micrite filling the vugs is partially recrystallised and frequently contains authigenic pyrite. Kaolinite has been observed only as coarsely crystalline vermicular aggregates reaching up to 20μ in size. The boundaries between kaolinite and sparry calcite are usually irregular and kaolinite seems to have corroded the sparry calcite lining of the vugs (Plate 2, Fig. 4). The micrite ground mass contains some authigenic idiomorphic quartz crystals which range in size from 8 to 70μ.

Sublithotype B-2; Thin-bedded Micrite

This rock occurs as a thin-bedded, laminated, fine-grained, grey to dark grey limestone in the C_2 zone of the Ruthin area in the southern outcrop of the South Wales Coalfield.

Microscopically, it consists wholly of a thinnly-bedded homogenous micrite. The bedding is caused by concentrations of silt-size skeletal debris in very thin laminae. These laminae also contain appreciable amounts of silt-size angular quartz grains. This rock is slightly dolomitized. Dolomite occurs as small rhombohedra of about 30μ in average size (Plate 2, Fig. 5).

Stylolites are common in the micrite in the Gower Peninsula. Organic matter has been concentrated along the stylolite seams and thin calcite veinlets often cut across the stylolites (Plate 2, Fig. 6).

PETROGRAPHY OF LITHOTYPE-C: BIOMICRITE

This is the most common lithotype in the Carboniferous Limestone Series of the South Wales Coalfield. On the basis of the relative frequency of micrite matrix, this lithotype is divided into two sublithotypes, these are; sparse biomicrite, in which the micrite matrix constitutes more than 50% of the rock, and packed biomicrite, in which the micrite matrix accounts for less than 50% of the rock.

Sublithotype C-1; Sparse Biomicrite (Plate 3)

This sublithotype is dominant in the Main Limestone Group; especially in the S_2 and D zones. It is present as thickly bedded layers usually interstratified with oolitic bands.

Macroscopically, the rock is very hard, massive, light grey in colour. A porphyrotopic texture of coarse-grained skeletal debris floating in a fine-grained micrite groundmass is very prominent. In the western part of the northern outcrops and in the Gower Peninsula, the rock is dissected by calcite veins which range in width from a few millimetres to several centimetres.

Microscopically, this rock is made up of various types of skeletal debris, intraclasts, pellets and some oolites. The allochemes are usually poorly sorted and float in the micrite matrix. In many rock samples of this sublithotype, the micrite matrix has been recrystallised partially or completely into pseudosparite. At the first glance, the rock gives the impression of being a biosparite, but the rock is in fact recrystallised biomicrite.

Many rock varieties have been recognised in this sublithotype. In the following paragraphs their gross petrography will be described collectively and then the main characteristics of each type will be briefly discussed.

Allochems:

The following allochems have been identified in the biomicrite lithotype.

Echinoderms:

Echinoderm fragments varying in size from 0.1 to 4.0 mm. occur. A more precise differentiation of the echinoderm remains into crinoid and echinoid debris is possible in many instances. Stems dominate the crinoid debris. Abundant echinoid spine fragments together with echinoid plates have been observed. The single crystal habit has almost always been preserved, even in cases of dolomitization or heavy micritization.

The reticulate texture, caused by the original porosity of the echinoderm skeletons, generally remains recognisable in spite of many changes. Pores filled with calcite, syntaxial with the skeletal monocrystal, remain visible as relicts because of the impurities which originally lined them. In many cases the pores are filled with micrite grains which are randomly oriented relative to the skeletal monocrystal. These micrite infillings are usually concentrated in the outer third of the echinoid fragment. In some fragments the pores are filled with black organic matter, which clearly brings out the reticulate structure.

Syntaxial overgrowths of calcite on the echinoid fragments are not frequent. If present, they occur as thin rims that usually replace the micrite matrix. Micritization of echinoidal remains is very frequent. The micrite envelope usually occurs as a thin rim, but in some fragments it replaces the outer third of the fragments. Almost complete micritization of smaller fragments is not uncommon.

Brachiopods:

Both complete shells and individual valves of brachiopods are present, but detrital shell fragments are the most frequent. The shell structure is characterised by a thin outer lamellar layer and a thicker inner layer that comprises long slender calcite prisms oriented obliquely to the shell surface. Shells with punctate structure have been observed.

Micritization of brachiopod shell debris is very frequent. The micrite envelope varies in thickness from a thin rim to a very thick rim that replaces about half of the shell. The interface between the thick micrite envelope and the original shell is very irregular. Algal encrustation is very common on the surface of shell debris. The algal micrite crust reaches up to 1.0 mm. in thickness. Borings are very common in the algal micrite crust, bores being filled with sparry calcite.

Molluscs:

Molluscan remains are represented mainly by pelecypod shell debris. The original aragonitic shells have been dissolved and the moulds are filled with drusy calcite. Severe micritization has affected the shell fragments and algal encrustation and borings have been observed.

Algae:

Many varieties of calcareous algae have been recognised in the sparse biomicrite sublithotype. MacQuown and Bloxam (1972) listed the algae preserved in the Middle Visean limestones of the Bristol area and the Gower Peninsula and the present writer has nothing to add to their list. Micritization, algal encrustation and boring have frequently affected the skeletal structures of the various types of the calcareous algae.

Algal grains, similar to those described by Wolf (1965), Monty (1967) and De Meijer (1971) have also been observed. The grains are rounded and are very extensively micritized.

Bryozoans:

Fenestrate types of bryozoa are the most abundant in this sublithotype. They possess circular, elliptical or polygonal zoecial chambers. The fronds of the fenestrate forms have a fine fibrous structure in which the fibres lie more or less parallel to the Zoecia.

Ostracods:

Both complete shells and single valves of ostracods are present. In the former case, the upper side consists of a stack of valves. The shells have a lamellar structure with very fine calcite fibres oriented obliquely to the surface.

Calcispheres:

These are minute spherical bodies, each consisting of a dark thin wall surrounding a cavity filled with calcite cement. Spheres reach an average diameter of about 0.08 mm.

Foraminifera:

Most of the foraminiferal tests are those of benthonic to semipelagic forms. They have microcrystalline calcite walls. In some tests, the original texture has been completely obliterated by diagenetic changes.

Pellets:

The pellets in the biomicrite lithotype consist of microcrystalline calcite (micrite) and are devoid of internal structure. Two types of pellets have been recognised in this lithotype. The first variety consists of well sorted, fine grained, spherical pellets. They are very similar to faecal pellets of Recent sediments described by Kornicker and Purdy (1957). The second type is less uniform in size and usually has a very small core of skeletal fragment. This could be produced by the complete micritization of fine grained skeletal debris (Wolf, 1965). These pellets have been called algal pellets, by Monty (1967) and by De Meijer (1971). in the Upper Carboniferous Limestone of Spain.

Oolites:

The oolites in this sublithotype are made up of a coarse-grained core comprising a bioclast or quartz grain surrounded by oolite shells. They are thought to represent the combined result of algal coating and regular rolling.

Micrite Matrix:

In the sparse biomicrite rocks the micrite matrix forms more than 50 percent of the rock. It is represented by a very fine grained mosaic of calcite crystals about 5μ in average diameter and is usually very rich in organic matter. The presence of organic matter gives a light grey colour to the micrite. Patchy to complete recrystallisation of the micrite matrix into neomorphic pseudospar has been observed.

Cavity-filling calcite:

Clear sparry calcite fills foraminiferal chambers and Calcispheres. It also occurs as cavity filling of mollusc shell moulds. Two generations of calcite infilling have been recognised. The first generation is represented by minute, bladed prismatic crystals that line the cavity. The second generation is a coarse-grained mosaic of sparry calcite filling the rest of the cavity.

The sparse biomicrite sublithotype has been divided into six varieties on the basis of the dominant type of skeletal debris. The following is a brief description of each variety.

Crinoidal biomicrite: (Plate 3, Fig. 1)

This occurs generally in the lower part of the Z zone, and sometimes as thin intercalations in the shales of the Lower Limestone Shale Group of the South Wales and Forest of Dean Coalfields.

Macroscopically, it is very hard and greyish to black in colour. The crinoidal fragments are coarse enough to be visible to the naked eye. In some specimens, these fragments reach up to 4 mm. in diameter. On the weathered surface of some samples, lengths of crinoid stems in articulating positions have been observed. The rocks in the Gower Peninsula and in the western part of the northern outcrops are highly fractured. Fractures are filled with sparry calcite crystals, ranging in width from a few millimetres to several centimetres.

Microscopically, the rock consists of crinoidal fragments with subordinate amounts of brachiopod shell debris, ostracod shells, and calcareous algal fragments, all floating in a micrite matrix. The crinoidal fragments are comprised mainly of ossicles. Partial silification is found to have preferentially attacked the central parts of the ossicles.

The micrite matrix in samples from the northern outcrop and the Gower Peninsula of the South Wales Coalfield is usually recrystallized into pseudosparry calcite crystals. Neomorphic pseudo-spar is present as mosaics of equidimensional and strongly strained crystals. Individual pseudo-spar crystals reach 60μ in average diameter.

Foraminiferal biomicrite: (Plate 3, Fig. 4)

These rocks are frequently present in the Main Limestone Group, especially in the S_2 and the D zones. They occur as massive dark grey limestones, usually interbedded with the oolitic layers.

Macroscopically, the rocks are very hard, black to greyish in colour, with aphanetic texture.

Microscopically, the rock consists mainly of foraminiferal tests with subordinate amounts of calcareous algal debris, ostracod shells, crinoidal fragments and brachiopod shell debris floating in a micrite matrix. The micrite matrix of samples from the northern outcrops is recrystallized into pseudo-spar. Recrystallization of micrite is patchy or complete. Stylolitization is frequent, and predates the calcite veinlets. In some rocks, authigenic idiomorphic quartz crystals and vermicular kaolinite-filled cavities are frequent.

Algal biomicrite: (Plate 3, Fig. 3)

This occurs as thin bands, about 2 to 3 ft. thick, in the upper parts of the C zone.

Macroscopically, it is very hard, varying in colour from creamy-white to black. It is usually of aphanitic texture and has a porcellanous appearance.

Microscopically, it consists mainly of calcareous algal debris with frequent amounts of ostracod shells and foraminiferal tests, in a micrite matrix. Also present are vugs and moulds of mollusc shells filled partially by geopetal micrite and partially by sparry calcite. Oncolite algal biomicrite usually contains considerable amounts of glauconite. The glauconite pellets ressemble ostracod shell moulds. Some glauconite seems to be pseudomorphous after dolomite rhombs.

The glauconite is often partially or completely oxidised to haematite. Glauconite nodules are usually concentrated along stylolite seams. Authigenic idiomorphic fluorapatite crystals are frequent in the oncolite bodies.

The micrite matrix in the algal biomicrite rocks seems to be derived from disintegration of the algal bodies. In this respect it is similar to the algal micrite described by Wolf (1965) and De Meijer (1971). Recrystallization of the algal micrite is very rare and only a few recrystallised patches of microcrystalline calcite (15μ in average diameter) have been observed. Stylolitization is very frequent, with concentrations of organic matter and other insoluble residues in the stylolite seams.

Brachiopod biomicrite: (Plate 3, Fig. 2)

The rock consists of brachiopod shells with subordinate amounts of pelecypod shells, echinoid debris, and tests of foraminifera floating in a micrite matrix. Considerable amounts of pellets have been noticed in this rock type.

Brachiopodal shell debris is represented by separated valves and fragments. Brachiopod cavities are often partially filled with geopetal micrite and sparry calcite cement. The aragonite of the pelecypod shell debris has been dissolved and the moulds filled with drusy calcite. Partial dolomitization of the brachiopodal shell debris has occurred. Dolomite is present as curved subhedral dolomite crystals.

In samples from the northern outcrops, idiomorphic authigenic quartz crystals are developed and vermicular kaolinite cavity-filling is frequent. The micrite of these samples is usually recrystallized to pseudo-spar with crystals of about 35µ in average diameter. It was found that the micrite forming the envelopes resists recrystallization.

Ostracodal biomicrite: (Plate 3, Fig. 3)

This occurs as about 1 ft. thick intercalations in the shales of the Lower Limestone Shale Group of the Gower Peninsula.

Macroscopically, the rock is very hard and reddish in colour. Ostracodal shells can be seen easily by the naked eye.

Microscopically, it consists mainly of articulated ostracod valves with subordinate amounts of echinoid debris and foraminiferal tests. Ostracod shells are usually filled with glauconite showing various degrees of oxidation. This can be seen by the gradual change of colour from fresh green through brownish green, to brown and black. The micrite is completely recrystallized into coarse-grained pseudosparite.

Calcisphere biomicrite: (Plate 3, Fig. 6)

This facies occurs as 1 ft. thick bands in the C_2 zone of the Gower Peninsula. The rock is very hard and is dark grey in colour. Microscopically, it consists mainly of calcispheres floating in a micrite matrix.

Sublithotype C-2; Packed biomicrite (Plate 4)

The rocks belonging to this sublithotype occur as thin beds interbedded with other limestone types at various horizons in the Carboniferous Limestone Series.

Macroscopically, the rocks are hard, of phaneritic texture, usually black in colour and very rich in organic matter.

Microscopically, they consist of crinoid ossicles, brachiopod shells, ostracod shells, algal grains, coral fragments and oolites with less than 15% of micrite matrix. Quartz grains are also present in considerable amounts. According to ~~to~~ the dominant allochem type, the following varieties have been identified; packed crinoidal biomicrite, packed brachiopodal biomicrite, packed oobiomicrite, packed coral biomicrite.

In these rocks the allochems are closely packed with sutured grain contacts and organic matter is concentrated along the contacts. Although compressed, bivalves shells are articulated. In some cases, the concave-convex valves are squashed, and this is evidence for very early compaction which pre-dates lithification of the rock. In the case of oolitic rocks, the oolites are elliptical in shape with their long diameter parallel to the bedding.

Generally, the skeletal debris is micritized. Recrystallization or grain diminution has taken place in the rocks from the northern outcrops of the South Wales Coalfield. Authigenic idiomorphic quartz is frequent in the algal grains. Patchy recrystallization of the micrite matrix to microcrystalline pseudosparite crystals, up to 30μ in average diameter, is not uncommon.

In the packed coral biomicrite from Gower Peninsula, the micrite matrix is strongly strained and recrystallized to pseudo-sparite crystals up to 50μ in average diameter. The coral chambers are filled with two phases of cementation. The first phase is represented by bladed fine-grained crystals lining each chamber, and the second phase is an equidimensional mosaic of sparry calcite that fills the rest of the chamber. Dolomitization is frequent in this rock type. The dolomite formed as coarse crystals, most of which are now dedolomitized. The strained lattice of dolomite is inherited by the replacing calcite.

Quartz grains trapped in the micrite matrix are severely cracked, and the cracks are filled with calcite. Vermicular aggregates of authigenic coarsely-crystalline kaolinite fill cavities and corrode the calcite matrix.

PETROGRAPHY OF LITHOTYPE-D: BIOSPARITE

Rocks of this lithotype consist of various types of allochems cemented together by sparry calcite. On the basis of the dominant type of bioclasts, this lithotype is divided into three main rock groups, namely, crinoidal biosparite and biosparudite, brachiopod biosparite, and algal biosparite.

Crinoidal biosparite and biosparudite: (Plate 5, Figs. 1 & 2)

These rocks occur as thick beds interstratified with the shales in the Lower Limestone Shale Group of South Wales and the Forest of Dean Coalfields. They are present also in the main Limestone Group.

The rocks are very hard, light grey in colour and phaneritic in texture. The crinoid ossicles are usually large, reaching up to 7 mm. in the sparudite.

Microscopically, they consist mainly of crinoidal fragments with subordinate amounts of algal debris, brachiopod shell debris and oolites. The framework fragments are bound together with a clear sparry calcite cement.

In the crinoidal biosparite, grain size distribution is that of a well sorted, medium-sand, skeletal debris of about 0.3 mm. in average diameter, being equidimensional in shape and well rounded. In the case of the crinoidal biosparudite, bioclasts are of moderately sorted granules, of about 3 mm. in average diameter. Generally, there is no preferred orientation of skeletal debris, except in a sample from a thin band in the Lower Limestone Shale Group of the Forest of Dean. In this rock the crinoidal ossicles lie parallel to bedding.

Crinoid fragments comprise more than 70 percent of the framework grains, and are represented mainly by ossicles. Echinoid fragments are much less common. Brachiopod shell debris is represented by well-rounded shell fragments. Oölites are usually spherical, well sorted and of about 0.3 mm. average diameter. They often contain cores of coarse-grained crinoidal debris. Brown phosphatic fragments (collophane) are common in the crinoidal biosparudite of the Lower Limestone Shale Group.

Skeletal fragments are usually surrounded by micrite envelopes, but micritization and algal boring are more common in the brachiopod shell fragments. Algal grains produced by the micritization of skeletal debris have been observed. Aragonite of molluscan shells has been dissolved and the shell cavities filled with drusy calcite. The crinoidal biosparudite rocks are commonly poorly washed and contain appreciable amounts of micrite. By contrast, the well sorted crinoidal biosparite rocks are usually very clean.

Syntaxial calcite overgrowth on the crinoidal fragments is the main cementing agent. It constitutes up to 70 percent of the cement in most crinoidal biosparite and biosparudite rocks. In some crinoidal biosparudite rocks, syntaxial calcite overgrowth is the only cement observed.

The texture of the syntaxial overgrowth calcite is sometimes poikilotopic and encloses more than one allochem. In some specimens, the syntaxial overgrowth calcite partially replaces other skeletal debris.

Authigenic minerals are also frequent. Authigenic quartz is present as overgrowths on the detrital quartz grains and authigenic illite, pseudomorphing vermicular kaolinite, is common.

Brachiopodal biosparite:

This occurs only in the Main Limestone Group of South Wales. Microscopically, these rocks consist mainly of brachiopod shells and shell debris and crinoidal fragments, with considerable amounts of pellets and subordinate amounts of algal debris and oolites. Appreciable amounts of micrite have been found in these rocks. Because of this, the rock is regarded as a poorly-washed biosparite.

The skeletal debris is often severely micritized. Two types of pelletal grains have been identified, namely, well sorted faecal pellets, and poorly sorted coarse algal pellets.

The framework grains are cemented mainly by a granular mosaic calcite.

Algal biosparite: (Plate 5, Fig. 3)

These facies occur only in the Main Limestone Group of South Wales. The rock consists mainly of algal fragments, crinoidal and brachiopod debris, with subordinate amounts of foraminiferal tests and oolites. The algal element is represented by various types of calcareous algae and algal lumps. Generally, the skeletal debris is severely micritized. The rock contains appreciable amounts of micrite. Cementation is similar to that of the brachiopod biosparite.

PETROGRAPHY OF LITHOTYPE-E: OOLITIC LIMESTONE

Rocks belonging to this lithotype consist mainly of oolites and subordinate amounts of skeletal debris cemented by sparry calcite. On the basis of the relative frequency of the skeletal debris in the rocks, this lithotype is further subdivided into two sublithotypes, namely, oosparite and bio-oosparite.

Sublithotype E-1; Oosparite: (Plate 5, Figs. 4, 5 and 6)

These are the main rocks comprising the Caninia Oolite zone (C) in South Wales and the Forest of Dean Coalfields. They are present as beds of about 6 ft. thick in the Lower Limestone Shale Group in some parts of the northern outcrops of South Wales, and are the dominant lithology in the Drybrook Limestone of the Forest of Dean (S_2 zone).

Macroscopically, they are very hard, with a clear oolitic texture, ranging in colour from white to dark grey.

Microscopically, they consist almost wholly of oolites. Skeletal debris comprises less than 7 percent of the rock grains. The oolites are cemented by a clear sparry calcite. Textureally, the oolites are well sorted, and are of medium-sand size.

The oolites show an exceptionally well-developed internal texture. Those in the Caninia oolitic limestone contain nuclei about 30μ in average diameter. Occasional coarse-grained nuclei in Caninia oosparite are also present. Oolites of the Lower Limestone Shale Group of South Wales and of the Drybrook Limestone of the Forest of Dean contain coarse-grained nuclei up to 0.2 mm. in diameter. The oolite nuclei are usually made up of crinoid and brachiopod shell fragments and occasional quartz grains. In the Drybrook Limestone almost all the oolites contain nuclei of coarse-grained angular quartz (Plate 5, Fig. 6). The thickness of oolite shells in the Drybrook oosparite ranges from 40 to 50μ with an average of about 60μ.

The oolite shells are now composed of radially arranged, elongated calcite crystals which reach across several original layers. It is probable that the original mineral comprising the oolitic shells was aragonite, as is the case in Recent marine carbonate oolites.

Skeletal debris is usually well rounded and is covered by a thin micrite envelope. Molluscan shell remains are usually filled by cavity-filling sparry calcite. Crinoidal and echinoidal fragments are often surrounded by syntaxial calcite overgrowths.

The cementation history of this rock is simple. Two crystal fabrics in the sparry calcite cement have been recognised: a finely crystalline, prismatic calcite fringe around the oolites, followed by an equidimensional mosaic calcite occupying the rest of the pore spaces. Both are of non-ferroan calcite. The syntaxial overgrowths on echinoderm debris are fringed by mosaic calcite. When oolites are surrounded by syntaxial calcite overgrowths, the fringe calcite cement is absent.

Authigenic quartz is frequent and is represented by euhedral crystals. Most of them are the product of overgrowths on the quartz grains forming the nuclei of the oolites. The original shape of the quartz grains is preserved by a dirt coating. Ghosts of oolite shells replaced by authigenic quartz overgrowths have been noticed. In the course of their growth some of the quartz crystals have replaced the oolite shells and parts of the fringe and the mosaic cement.

In some oolites the outer shell has been spalled off. A fringe of finely crystalline calcite covers the outer and the inner surface of spalled off shells. This is an indication of very early deformation. In some rocks, partial to complete recrystallization of the oolites has been observed. Microstylolitic contact between oolites has been noticed.

Sublithotype E-2: Bio-Oosparite

Rocks belonging to this sublithotype comprise the bulk of the Seminula and Dipnophyllum zones of South Wales. They are thick bedded to massive, hard rocks, varying in colour from white to dark grey.

Microscopically, they consist of oolites and skeletal debris cemented by sparry calcite. They usually contain considerable amounts of micrite and may thus be regarded as poorly washed calcarenites. Texturally, they consist of well to moderately sorted medium sand size material.

Oolites form about 50 percent of the allochems. The oolites are of two types: (a) those with well developed internal structure; and (b) those devoid of any internal structure. The term pseudo-oolite will be used here for the latter type. The oolites have coarse-grained nuclei of crinoid and brachiopod debris. Quartz nuclei are not uncommon. The skeletal debris forming the nuclei of many oolites is severely micritized. The oolite shells may range in thickness from 40 to 200 microns, with an average of 120 microns. Pseudo-oolites are very frequent, forming about 20 percent of the allochems.

Skeletal elements consist of crinoid and brachiopod shell debris and algal remains. They form about 50 percent of the allochems. Crinoidal debris is the most frequent bioclast. The skeletal debris is usually micritized. This applies particularly to the brachiopodal shell fragments.

The allochems are cemented by sparry calcite. Syntaxial calcite overgrowths on the crinoidal debris form more than 40 percent of the cement. Poikilotopic texture, of syntaxial calcite overgrowth enclosing more than three allochem grains, is frequent. Mosaic calcite forms 60 percent of the cement. It fills the intergranular pores between grains other than crinoidal debris. Mosaic calcite is represented by two crystal fabrics: prismatic fine calcite crystals fringing the oolites and allochems except crinoidal ossicles; and granular mosaic calcite filling the rest of the pore spaces. Usually the cement is non-ferroan calcite.

Oolites with off-centre nuclei and sparry calcite interiors occur in several bio-oosparite beds of the Main Limestone Group. In these cases, the oolite shells are partially or completely dissolved and the relatively insoluble nuclei have come to rest in the fluid-filled voids. Some oolites have completely dissolved. It was found that almost all the insoluble nuclei have settled in the bottom of the voids. The voids are filled with clear sparry ferroan calcite with a mosaic texture.

Recrystallization of the geopetal micrite in samples from the northern outcrops has taken place. The micrite is recrystallized to pseudospar crystals, about 50μ in average size. Ghosts of the original micrite material are clearly preserved in the pseudospar crystals.

PETROGRAPHY OF LITHOTYPE-F: DOLOMITE

(Plate 6 and 7)

Rocks belonging to this lithotype are very common in the Carboniferous Limestone sequences in both the South Wales and the Forest of Dean Coalfields. In the South Wales Coalfield, dolomite rocks are confined generally to the eastern and southern outcrops. Here, dolomite forms the main bulk of the rocks of the K, Z, C and S zones. It occurs also in the D zone in the Gower and Ruthin areas. In the Forest of Dean Coalfield, almost all the Carboniferous Limestone rocks are composed of dolomite, with the exception of some thin beds in certain localities along the eastern edge of the Coalfield.

On the basis of their texture, the dolomite rocks are classified into two main sublithotypes, namely, coarsely crystalline dolomites, and finely crystalline dolomicrite.

Sublithotype F-1; Coarsely crystalline dolomite

This is the most frequent type of dolomite. Macroscopically, the rock is very hard, of buff to cream in colour.

It has a porous saccaroidal texture, and is clearly of diagenetic origin. It is formed by dolomite replacement of the original limestones, the original texture of which has been well preserved.

Three grades of dolomitization are distinguished, these are: (i) incipiently dolomitized limestones, in which the dolomite represents less than 10 percent by volume of the original rock; (ii) partially dolomitized limestones, in which the dolomite represents from 10 to 80 percent of the original rock; and (iii) dolomite, in which the entire limestone rock has been altered.

Incipiently dolomitized limestone: (Plate 6, Fig. 1)

This grade of dolomitization is frequent in most of the biosparite and oosparite rocks. In thin sections, dolomite occurs as idiomorphic rhombs, sporadically distributed in the rock. The rhombs range from 0.06 to 0.2 mm. in size, and are usually of the non-ferroan variety. They are cloudy and rich in inclusions. The rhombs preferentially occur in oolites and in the micrite envelopes around the bioclasts. In some cases, the sparry calcite cement has been partially replaced by dolomite.

Partially dolomitized limestone: (Plate 6, Figs. 2 & 3)

Partial dolomitization is very common in the biomicrite rocks. Two prominent textures have been observed.

(1) In the crinoidal biomicrites the micrite matrix is partially or completely dolomitized, while the crinoidal fragments resist dolomitization. Some fine dolomite rhombs have also been developed in the micrite envelopes of crinoidal ossicles. The dolomite occurs as finely crystalline mosaic crystals, about 60μ in average size.

(2) Patchily dolomitized foraminiferal biomicrite of the D_1 zone of the Gower Peninsula. This was described by Dixon and Vaughan (1911) as 'Pseudobreccia'. Macroscopically, this rock consists of angular dark limestone patches of various sizes, up to several inches across, in a ground mass of coarsely crystalline, saccaroidal, light-coloured dolomite.

Microscopically, the rock is identified as a partially dolomitized foraminiferal biomicrite. The dolomitized groundmass consists of euhedral to subhedral dolomite crystals of about 0.8 mm. in average size. The general texture of the dolomite groundmass is equidimensional hypidiomorphic. The contacts between the dolomite crystals are usually of irregular shape. Dolomite crystals are very rich in inclusions. Many contain ghosts of foraminiferal shells (Plate 5, Fig. 4). The rock is fractured and the fractures are filled with a mosaic of calcite crystals. The fractures are post-dolomitization as they cross the dolomite crystals. Where the fractures cut dolomite crystals they are partially lined with a thin rim of clear ferroan dolomite, which has grown in optical continuity with the dolomite groundmass. The rest of the fracture is filled with mosaic calcite.

Completely dolomitized limestone (dolomites):

In most of these rocks the original limestone textures are very well preserved. Their original limestone textures have a very great influence on the present dolomite textures. Texturally, two main types of dolomite are recognised, namely, porphyrotopic dolomites and granulotopic dolomites.*

(i) Porphyrotopic dolomite: (Plate 7, Figs. 1 & 2)

These rocks consist of coarse dolomite crystals, ranging from 0.2 to 2 mm. in size with an average of about 0.8 mm., floating in a medium to finely crystalline dolomite groundmass. It was found that the coarsely crystalline phenocrysts are dolomitized crinoidal debris and other coarse-grained skeletal fragments. The original shape of the

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* This terminology is after Friedman (1965).

crinoidal fragments and their syntaxial calcite overgrowth are very well preserved. The part of the dolomite crystal originally occupied by the crinoidal fragment is usually cloudy and the micrite envelopes are preserved, while the part formerly occupied by the syntaxial calcite overgrowth is very clear. In some samples the incipient dolomitization of the micrite envelope of the crinoidal ossicles can be recognised. It is represented by finely crystalline idiomorphic dolomite rhombs concentrated in the micrite envelope and in the micrite filling cavities in some skeletal fragments. These dolomite crystals are now enclosed in a single large dolomite crystal, which was produced by dolomitization of the rest of the rock. Because of this, these crystals show poikilotopic texture.

(ii) Granulotopic dolomite: (Plate 7, Figs. 3, 4 & 5)

This type of dolomite is generally produced by complete dolomitization of oosparite, biomicrite and biosparite. It usually consists of dolomite crystals almost uniform in size. Actually, two types can be recognised, based on size variation. These are: medium-crystalline dolomites which are built up of dolomite crystals of about 0.15 mm. in average size, and coarsely crystalline dolomites with crystals of about 1 mm. across. There appears to be no relation between size variation of the dolomite crystals and the original texture of the limestones.

Ghosts of ooliths are very well displayed. In the medium crystalline dolomites each oolith is replaced by more than one dolomite crystal (Plate 6, Fig. 5), while in the case of the coarsely crystalline dolomites, one or more oolites are enclosed in a single dolomite crystal (Plate 6, Fig. 6).

According to the fabric of the dolomite crystals, three varieties of the granulotopic dolomites have been recognised, namely: idiotopic, hypidiotopic and xenotopic dolomites. No relation was found between these and the original texture of the limestones. Xenotopic dolomites are usually dense with very few voids. By contrast, the idiotopic and hypidiotopic dolomites are very porous.

Two types of pores have been recognised in the dolomites:

(i) Intergranular pores:

These are pores which exist between 3 or 4 idiomorphic or hypidiomorphic dolomite crystals of random orientations. Usually the pores are tiny and filled with insoluble organic matter. Dolomite replaced calcite, while the organic matter, that now fills pore spaces, was formerly present within the skeletal structures, or was present as finely disseminated grains in the micrite matrix. This organic matter could not be incorporated into the dolomite lattice due to the large size of its molecules. Thus during the growth of the dolomite, the organic matter was continually pushed away, and finally became trapped in the intergranular pores.

(ii) Shrinkage pores:

These are relatively large pores that range in size from about 80μ to 3 mm., and are bounded by idiomorphic dolomite crystals. Although filled in some cases, the pores are mostly empty. In the former case, they are occupied by a composite cement. This consists of an outer rim of clear ferroan dolomite, developed in optical continuity with the dolomite crystals enclosing the pores, and an inner zone of ferroan calcite. The ferroan calcite cement has a coarsely crystalline texture.

Sublithotype F-2; Finely crystalline dolomite (Dolomicrite)

This rock occurs as thin beds in the Lower Limestone Shale Group and at various horizons in the Main Limestone Group in both the South Wales and the Forest of Dean Coalfields.

In the field, the rock is thin-bedded, very hard, usually of greyish to pale brownish or buff colour. It has an aphanetic texture.

Microscopically, it has a finely crystalline granulotopic texture. It consists of very fine idiomorphic dolomite rhombs, ranging in size from 20 to 60µ. The rock lacks shrinkage porosity (coarse pores) but intergranular pores are frequent. The intergranular pores are extremely small and filled with displaced insoluble organic matter (Plate 7, Fig. 6).

Stylolites are abundant. Seismograph type stylolites have been seen. Organic matter is concentrated in the stylolite seams, some of which are up to 3 mm. thick.

Fractures have developed in the organic matter of the stylolite seams. These fractures are filled with fibrous calcite.

PLATE 1

Fig. 1 Calcareous siltstone, general texture. Sample BY54.

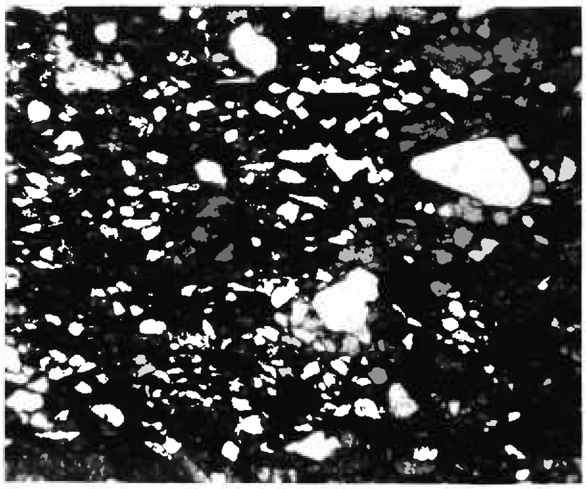
Fig. 2 Quartzarenite of the upper unit of the Drybrook Sandstone, general texture. Sample 1024. Crossed polars.

Fig. 3 Quartzarenite of the lower unit of the Drybrook Sandstone, general texture. Sample 1031. Crossed polars.

Fig. 4 Quartzarenite of the upper unit of the Drybrook Sandstone, with hematite cement filling most of the intergranular pores. Sample 1023.

Fig. 5 Calcareous sandstone cemented by dolomite (D). Sample BY58.

Fig. 6 Calcareous sandstone, consists of quartz grains cemented by calcite. Sample BY55.



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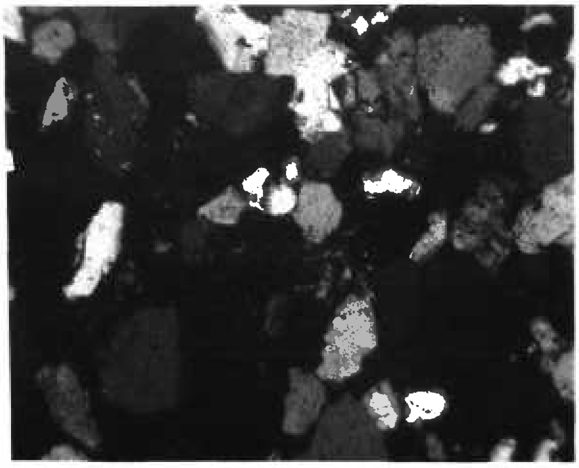

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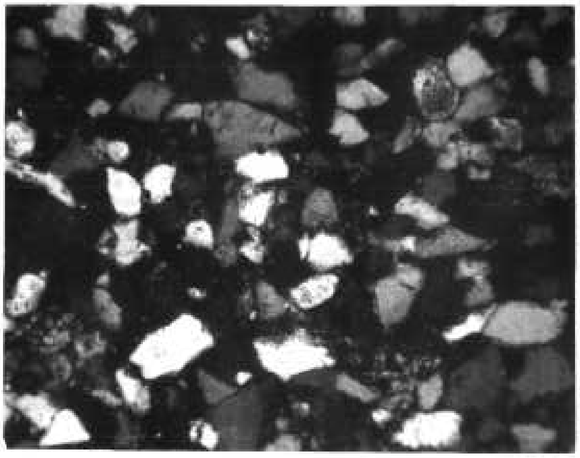

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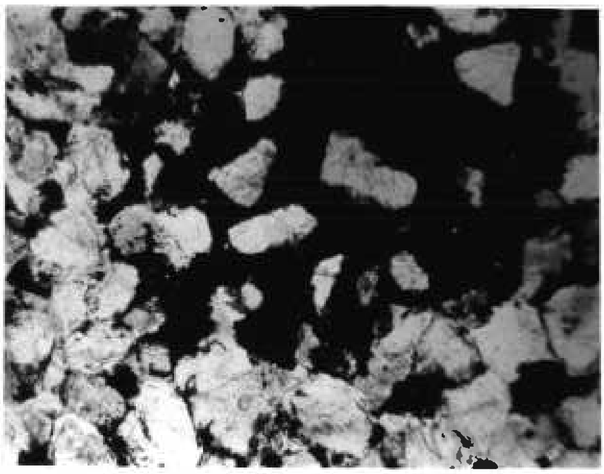

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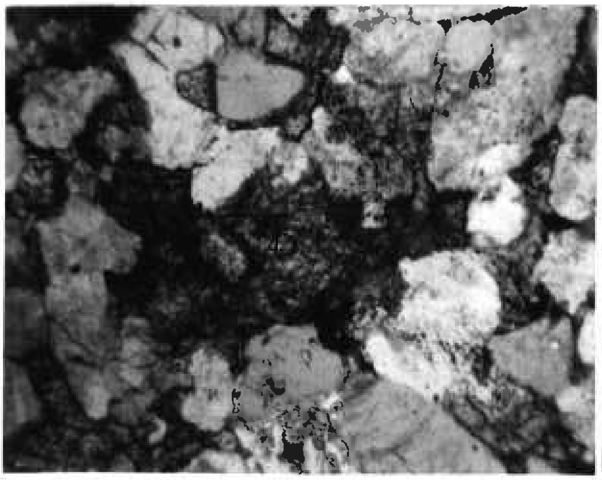

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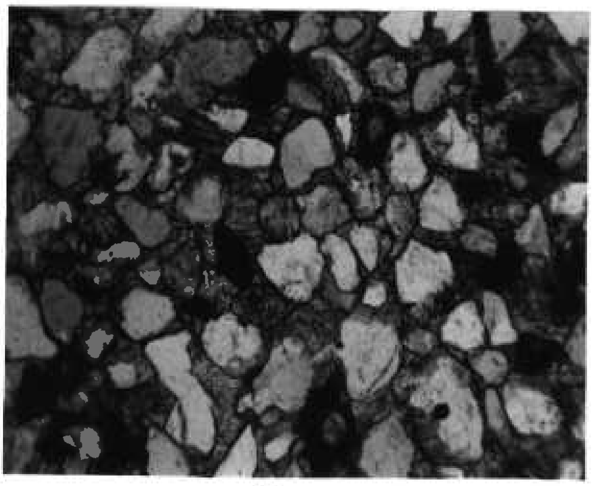

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PLATE 2

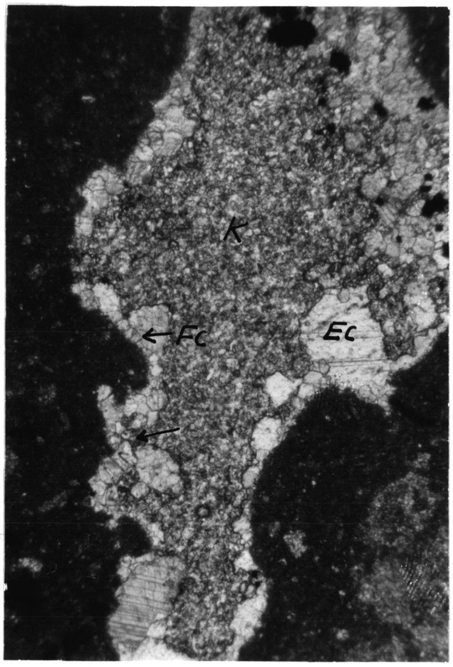
Figs. 1 & 2 Dismicrite, a vug filled with:- a rim of finely crystalline fibrous calcite (FC), equicrystalline mosaic calcite (EC) and vermicular kaolinite (K). Note corrosion of calcite cement by kaolinite (arrow). Sample 601.

Fig. 3 Dismicrite, a vug partially filled with recrystallized micrite (Rm), and a coarsely crystalline calcite crystal (Ec). The vug is lined with finely crystalline drusy calcite (FC). Sample 601.

Fig. 4 Corrosion of sparry calcite by kaolinite within a calcite-filled vug in dismicrite. Sample 601. Crossed polars.

Fig. 5 Thin-bedded micrite, partially dolomitized. Dolomite (D). Sample 605.

Fig. 6 Calcite veinlets cutting across a stylolite seam in Micrite. Sample 617.



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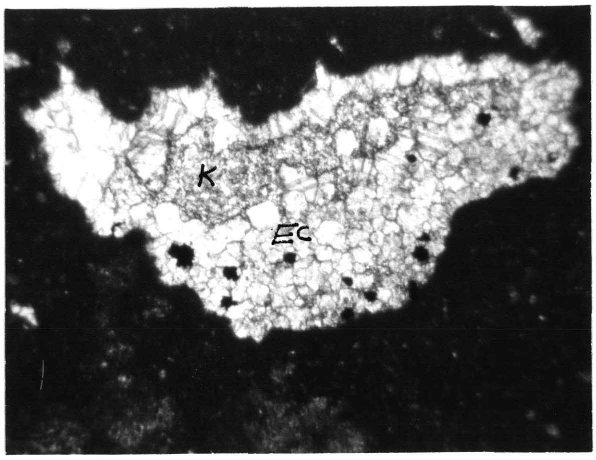

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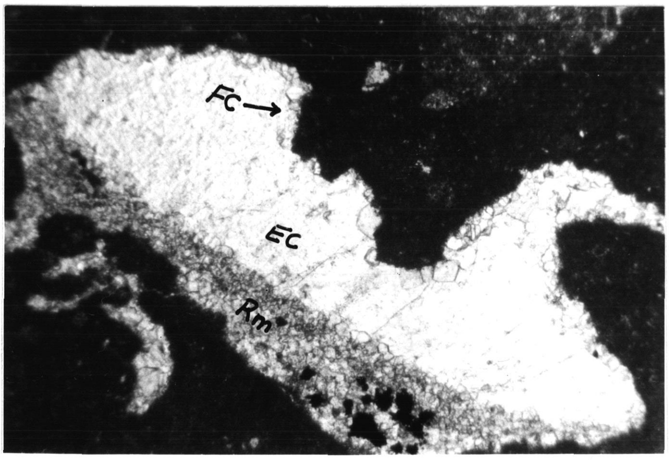

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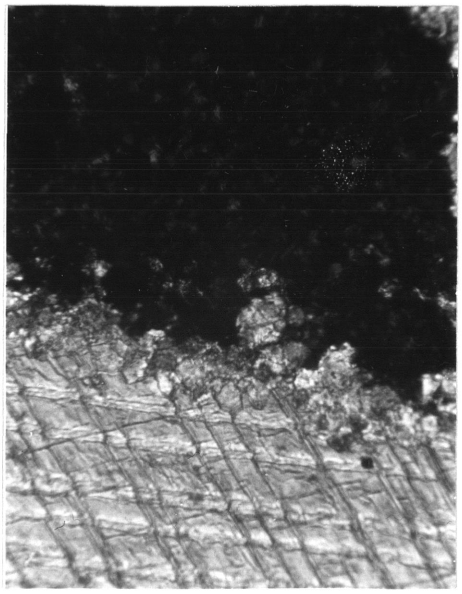

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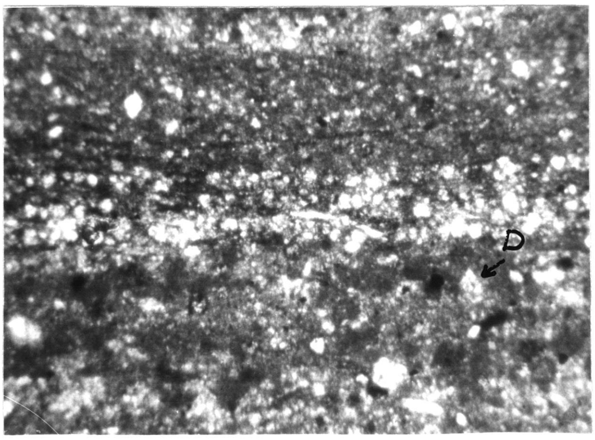

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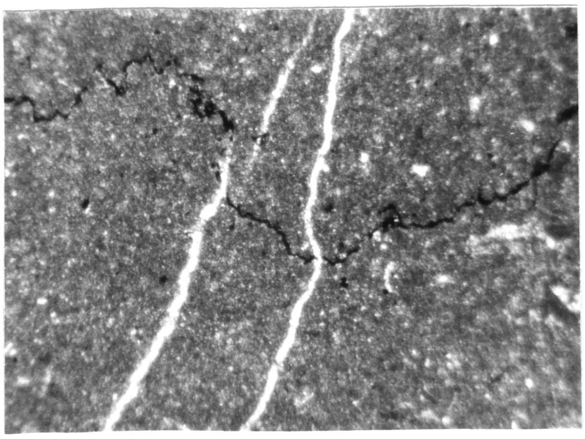

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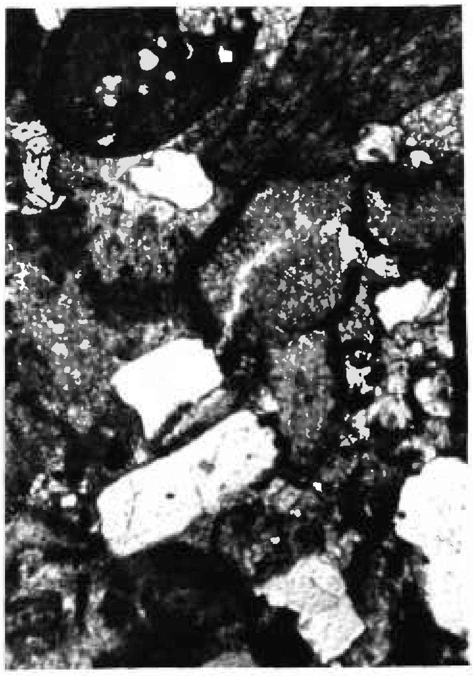
PLATE 3

Fig. 1	Crinoidal biomicrite. Micrite matrix is partially recrystallized. Sample 107.
Fig. 2	Brachiopod biomicrite. Micrite matrix is partially recrystallized. Sample 655.
Fig. 3	Algal biomicrite. Consists mainly of algal pellets (Gp) and faecal pellets (Fp), micrite is recrystallized. Sample 628.
Fig. 4	Foraminiferal biomicrite, micrite is slightly recrystallized. Sample 642.
Fig. 5	Ostracod biomicrite, ostracod body cavities filled with glauconite (G). In some shells, it is altered to limonite (L). Sample 764.
Fig. 6	Calcisphere biomicrite, calcisphere cavities filled with sparry calcite. Sample 606.



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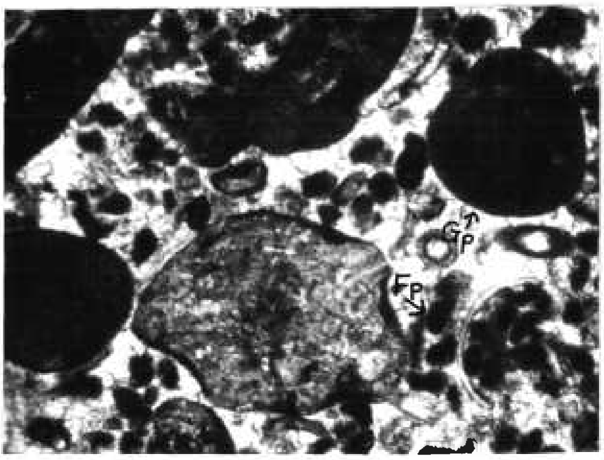

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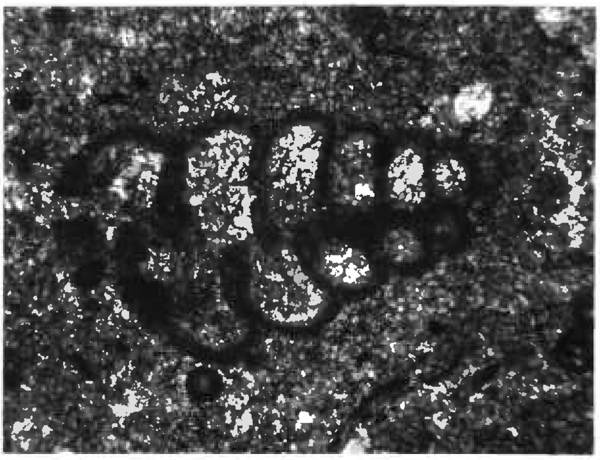

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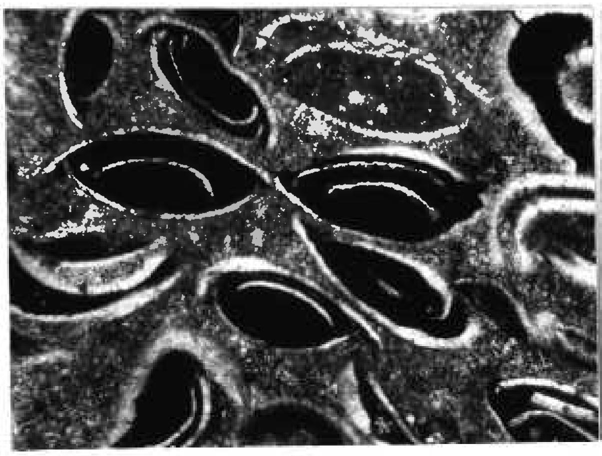

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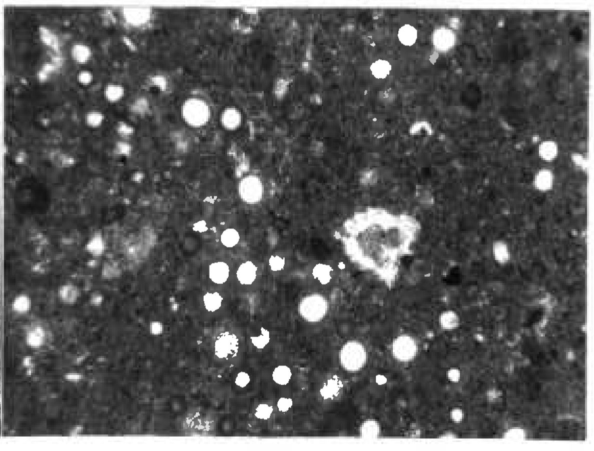

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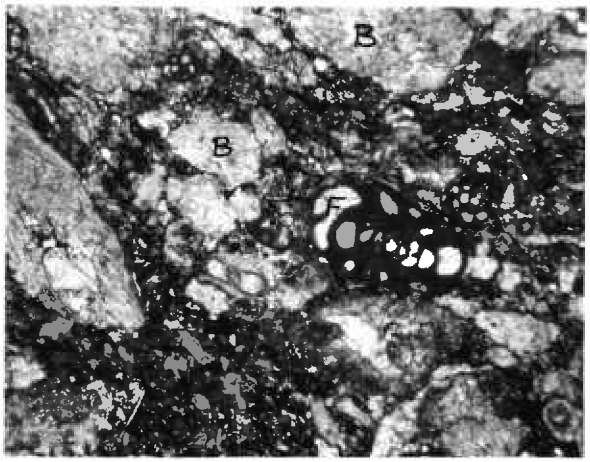
PLATE 4

Fig. 1	Packed brachiopod biomicrite, consisting mainly of brachiopod shell fragments (B) and foraminiferal tests (F). Sample 622.
Fig. 2	Packed crinoidal biomicrite. Note the stylolitic boundaries between the crinoidal fragments. Sample BY29.
Fig. 3	Packed fine-grained crinoidal biomicrite, partially dolomitized. Dolomite (D) is present as small rhombs scattered in the rock. Sample 624.
Fig. 4	Packed bio-oomicrite. Note deformed oolites (O) and broken ostracod shells (OS). Sample BY48.
Fig. 5	Packed brachiopod biomicrite. Sample BY53.
Fig. 6	Packed algal biomicrite. Note concentration of the organic matter on the stylolitic contacts. Sample 105.



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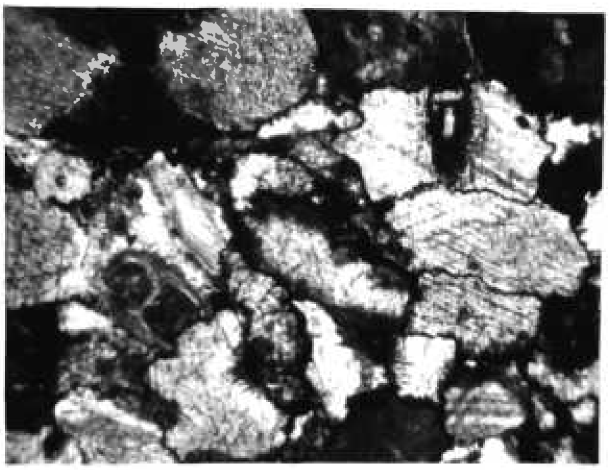

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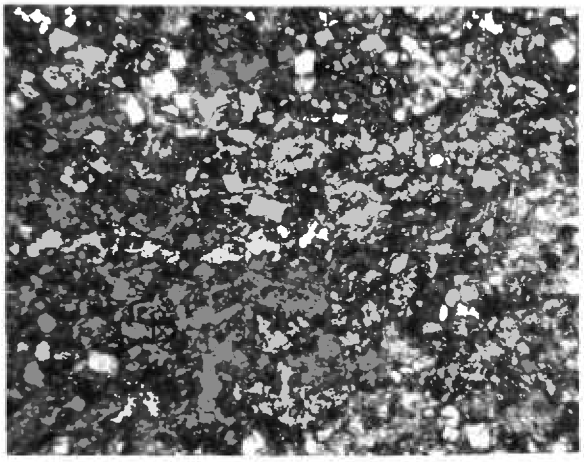

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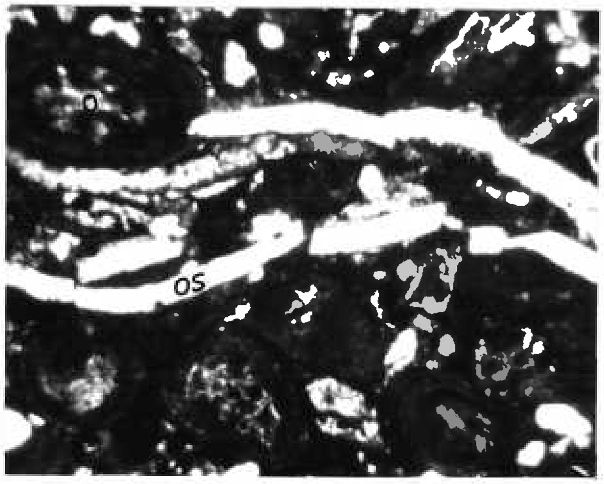

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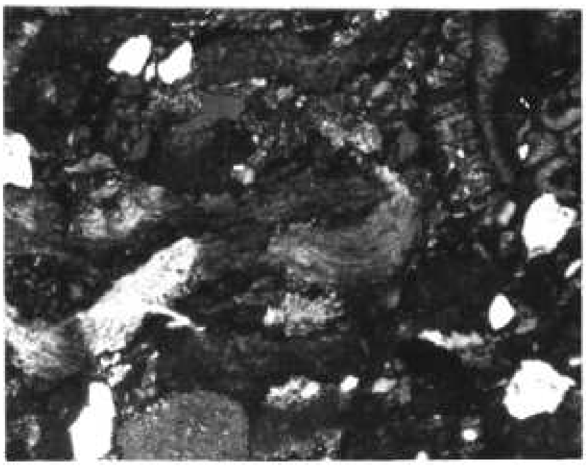

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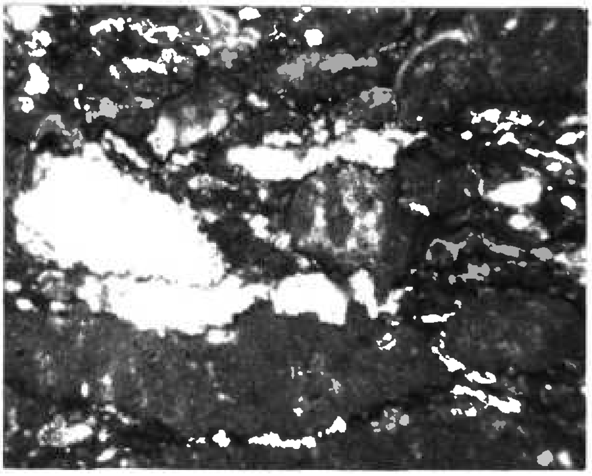

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PLATE 5

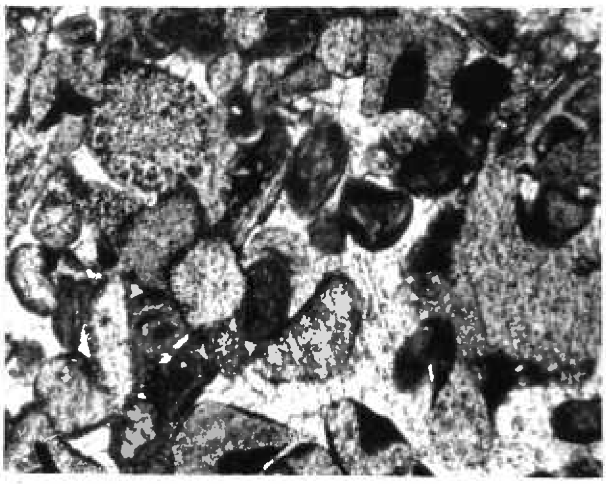
Fig. 1 Crinoidal biosparite. Sample 626'.

Fig. 2 Crinoidal biosparite. Note poikilotopic texture of syntaxial calcite overgrowth contains numerous allochem grains. Sample 673.

Fig. 3 Algal biosparite. Sample BY41.

Fig. 4 Oosparite, consists of well-sorted oolites cemented by sparry calcite. Sample BY31.

Fig. 5 Quartz-rich oolitic limestone. Oolites consist of coarse-grained quartz nuclei surrounded by a thin shell of calcite. Sample 1026.



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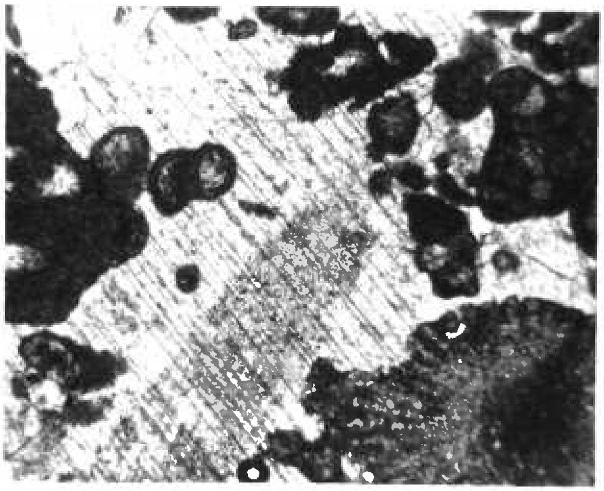

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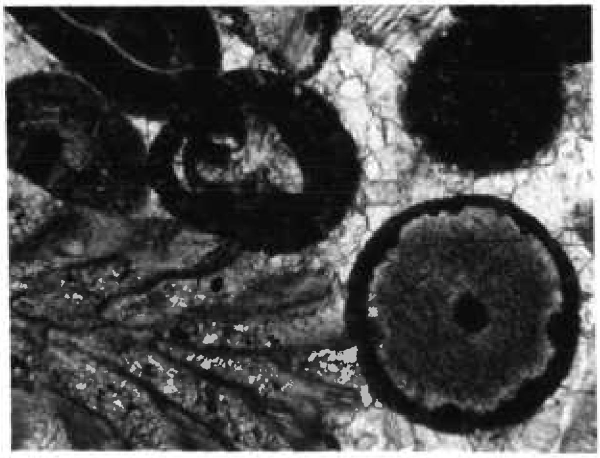

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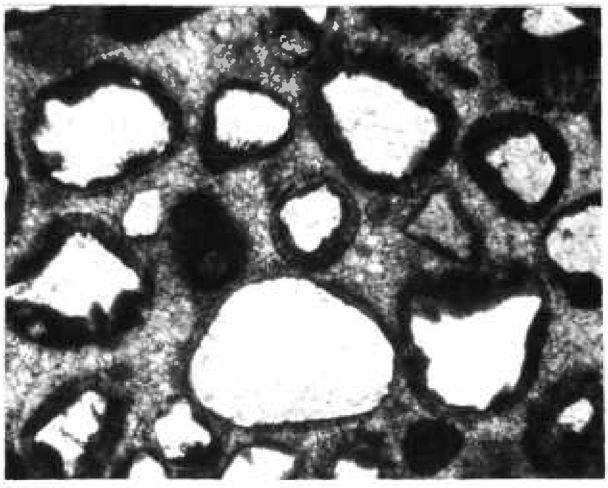

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PLATE 6

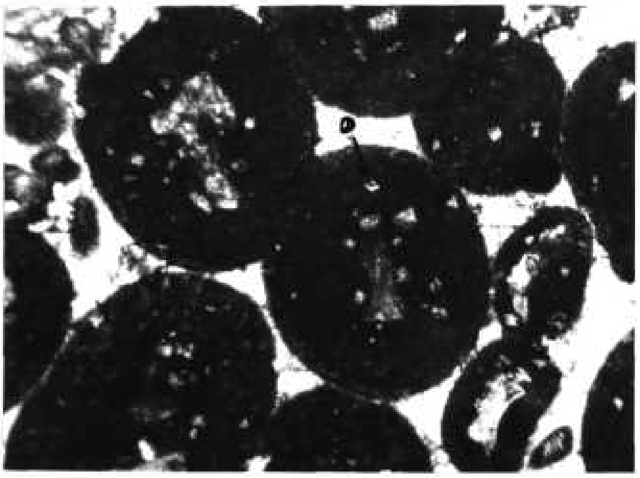
Fig. 1 Incipient dolomitization of oosparite. Finely crystalline dolomite rhombohedra (D) are preferentially replacing the micritic calcite of the oolites. Sample M2.

Fig. 2 Partially dolomitized oosparite. Dolomite (D) is replacing both oolites and calcite cement. Ghosts of oolites are displayed in some coarsely crystalline dolomite rhombs (arrow). Sample 607.

Figs. 3 & 4 Partially dolomitized biomicrite. Note irregular boundaries between dolomite crystals and ghost of foraminiferal test displayed in a single dolomite crystal. Sample 603.

Fig. 5 Completely dolomitized calcarenite. Ghosts of rounded allochems show that each grain has been replaced by a mosaic of dolomite crystals. Sample 1012B.

Fig. 6 Coarsely crystalline dolomite. Ghosts of an oolite displayed in a single dolomite crystal (arrow). Sample BY20.



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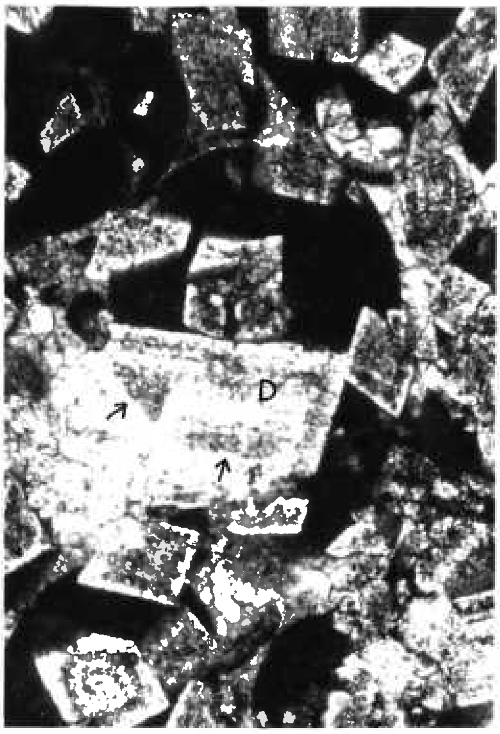

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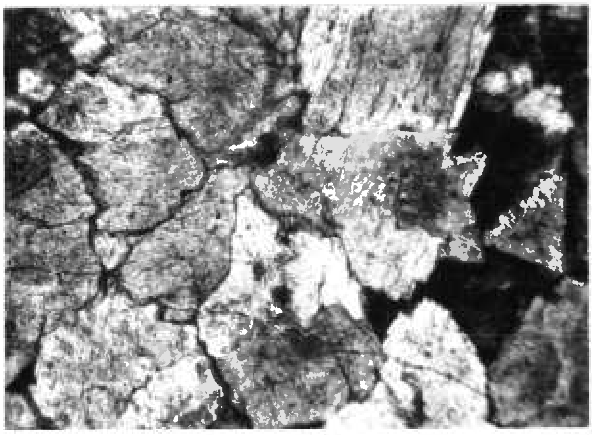

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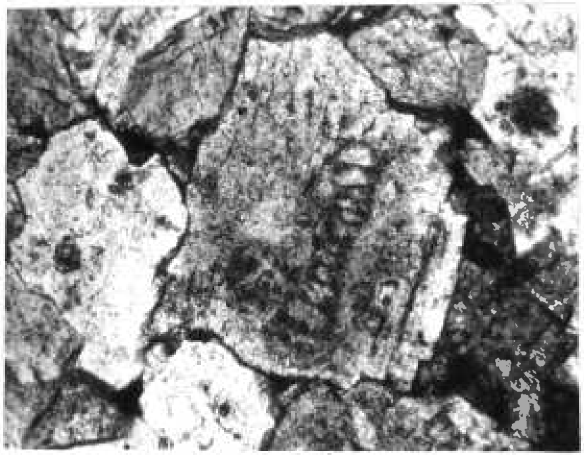

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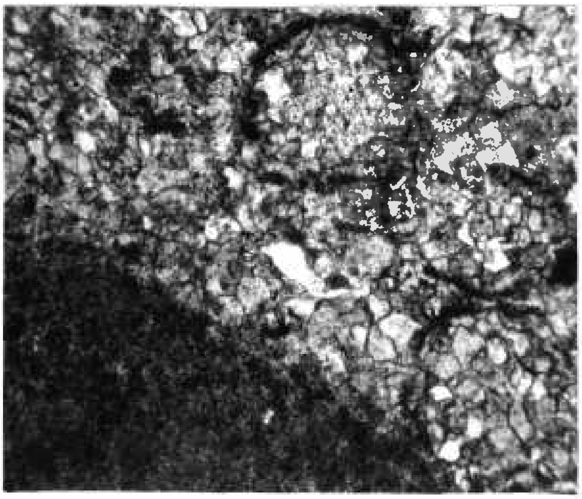

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PLATE 7

Figs. 1 & 2 Porphyrotopic hypidiomorphic dolomite consists of coarsely crystalline subhedral dolomite crystals embedded in a groundmass of moderately crystalline subhedral dolomite crystals. In Fig. 2, the dolomite phenocryst is most probably replacing a crinoid fragment (area with high content of dusty inclusions) and its syntaxial overgrowth (clear area). Sample BY23.

Fig. 3 Coarsely crystalline, granulotopic xenomorphic dolomite. Intergranular pores filled with organic matter. Sample BY25.

Fig. 4 Coarsely crystalline, granulotopic, hypidiomorphic dolomite. Sample BY20.

Fig. 5 Moderately crystalline, granulotopic xenomorphic dolomite. Sample SA17.

Fig. 6 Dolomicrite. Sample BY30.


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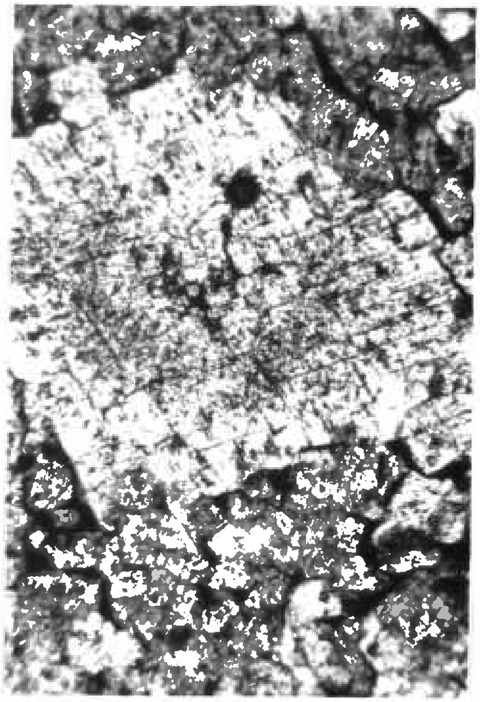
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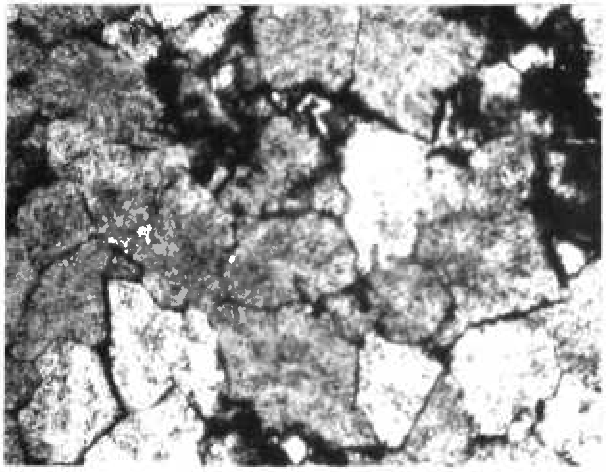
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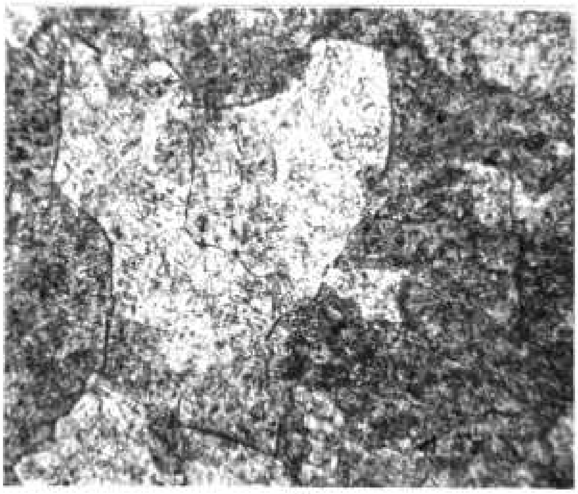
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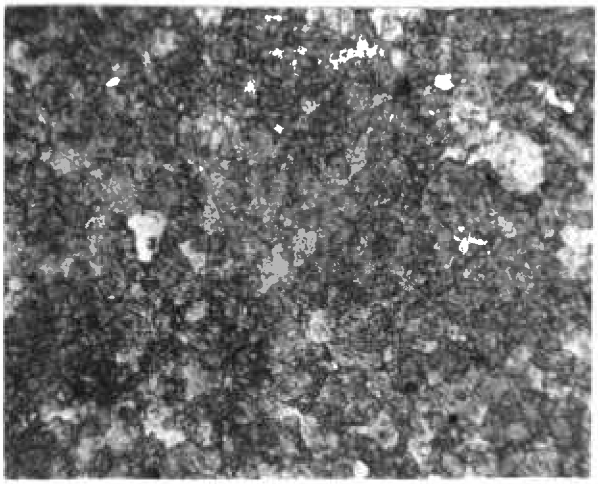
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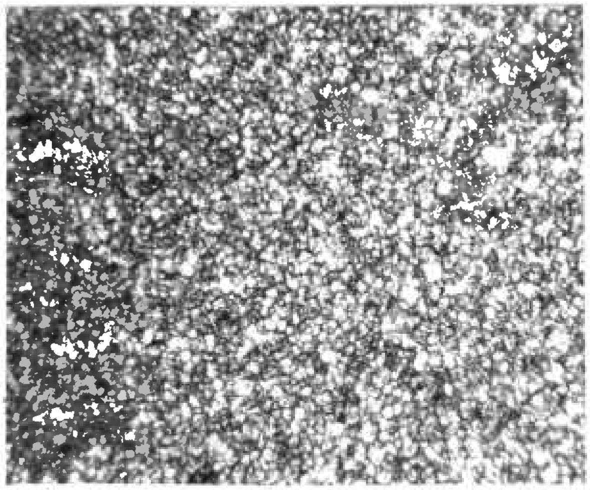
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CHAPTER III

DIAGENESIS OF THE CARBONIFEROUS LIMESTONE ROCKS

DIAGENESIS OF THE CARBONIFEROUS LIMESTONE ROCKS

INTRODUCTION

In this chapter the diagenetic changes and their sequence, in the various rock types of the Carboniferous Limestone Series are described. The chapter consists of two parts. The first deals with the diagenesis of the carbonate rocks (Lithotypes B, C, D, E and F). The second part deals with the diagenesis of the non-carbonate rocks (Lithotype A).

DIAGENESIS OF THE CARBONATE ROCKS

Because most of the main diagenetic changes encountered in the various lithotypes of the carbonate rocks are found to be similar, so they will be described collectively.

DIAGENETIC PROCESSES

For the purposes of description the diagenetic changes are classified as biological, physical, physiochenical and chemical.

A. BIOLOGICAL PROCESSES

These processes have taken place during the initial stage of syndiagenesis and began to be effective at the moment the allochems touched the bottom of the sedimentary basin. The main biological processes affected the bioclasts and displayed in the studied rocks are: micritization, algal boring and algal coating.

A.1 MICRITIZATION:

Two forms of micritization have been distinguished, namely, algal micritization and micritization by infilling of the skeletal pores.

(i) Algal micritization is the most common form. The mechanism of this process has been studied in detail by Bathurst (1966) and summarised in the following points: (a) Algae bore into the shell wall, (b) the algal filaments die and decay, (c) micrite aragonite fills the tubes.

(ii) Micritization by micrite infilling of skeletal pores of the echinoderm fragments. Sometimes it is difficult to recognise this form of micritization without high magnification (Plate 8, Figs. 1 and 2).

Micritization of skeletal debris is very prominent in the biomicrite and biosparite rocks. Centripetal micritization of most of the molluscan shell debris which has resulted in the formation of micrite envelope was responsible for the preservation of the shells as calcite casts. Micritization varies in intensity from slight micritization represented by the developments of thin micrite envelopes, to almost complete micritization of the bioclasts. An advanced stage of micritization generally ends in complete loss of the skeletal microstructure and consequently the bioclasts are identified as algal grains or algal pellets, which are frequent in most biomicrite and biosparite rocks. This phenomenon has been described by Monty (1967) in recent sediments, and by Wolf (1965) and De Meijer (1971) in Devonian and Carboniferous rocks respectively.

It was found that the various types of bioclasts vary in their susceptibilities to micritization. The following are the main bioclasts arranged in order of decreasing susceptibility to micritization: (a) Calcareous algae, (b) brachiopods, (c) molluscs, (d) echinoderms, (e) foraminifera, and (f) ostracods.

A.2 ALGAL BORING:

Algal boring in bioclasts has been noticed frequently in the biomicrite and biosparite rocks. Two types of algal boring have been distinguished, namely, fine boring and coarse boring. Fine borings are of about 10µ in average diameter and are responsible for the formation of the micrite envelopes. Coarse boring, ranging from 60 to 150 microns in diameter, are filled with micrite and fine-grained skeletal debris (Plate 9, Fig. 1), or by sparry calcite cement (Plate 9, Figs. 2 and 3).

Coarse boring was responsible for the biofragmentation of some bioclasts. (See Swinchatt, 1965.)

A.3 ALGAL COATING:

Algal coatings have been developed on many skeletal fragments in biomicrites and biosparites. The algal coating generally consists of dark algal micrite with calcareous algal tubes or moulds of algal tubes. Algal tubes are usually filled with clear microcrystalline sparry calcite. The algal coating and the micritization processes are generally found together in the same bioclasts. Generally, the volume of the algal coating is small compared to the volume of the allochems, but there are cases in which the algal coating has grown out of all proportion (Plate 9, Fig. 2).

These biological processes were found to be responsible for the following textural changes in the allochems of the original sediments:

(i) Disturbance in the original texture of the allochems by biofragmentation of some bioclasts on the one hand and increase in size of others by algal coating.

(ii) Replacement of the original microstructure of the bioclasts by the cryptocrystalline texture of the micrite, and the formation of algal grains and pellets.

(iii) The most important result is the formation of the micrite envelopes, which are responsible for the preservation of the skeletal moulds and therefore the original texture of the sediments.

B. PHYSICAL PROCESSES

These are classified into: (a) compaction, (b) mechanical deformation, and (c) pressure solution.

B.1 COMPACTION:

This process affected the sediments just after deposition and before complete cementation. The load created by overburden was the main responsible factor. It was found that the response of the allochems to the compaction depended on their nature, especially shape and fabric. Oolites, algal grains and pellets were found to be very responsive to the compaction, while the coarsely crystalline calcite fragments such as brachiopods and echinoderm debris were usually resistant to the compaction.

Compaction of the calcarenites is responsible for the alteration of the original texture of the sediments, e.g. the shape of the grains.

Compaction is manifest in the micrite rocks by the presence of vugs, which could be moulds of air or water bubbles. In these rocks compaction was responsible for the movements of the connate water which may form tubes, channels and bubbles (Cloud, 1962), which when filled with calcite cement produce the so-called birds-eye structure. This structure is prominent in the dismicrite rocks.

The following is evidence for early diagenetic compaction:

(i) Oolites are commonly flattened with thin long diameters parallel to the bedding.

(ii) The concave-convex bivalvia shells (brachiopods and ostracods) are usually squashed.

(iii) The presence of vugs in the dismicrite rocks, and which indicate migration of gas bubbles before lithification of sediments.

B.2 MECHANICAL DEFORMATION:

Two stages of mechanical deformation have been recognised, namely, early diagenetic and late diagenetic.

(a) Early diagenetic deformation:

This process commonly took place just after the dissolution of aragonite and was continuous even after the precipitation of the fibrous drusy calcite cement. Early diagenetic deformation was a result of the instability conditions created in the sediments during and after the dissolution of the aragonitic skeletal debris under overburden load. Evidence for this process is recorded by the following features:

(i) Spalling-off of the outer shells of the oolites (Plate 10, Fig. 2). In some oolites the outer shell is completely spalled-off and has fallen in the intergranular spaces, but in the majority it is still attached to the oolite.

(ii) The presence of crumbly fractures in oolites and bioclasts (Plate 10, Fig. 1). These fractures are on a microscopic scale, they are not uniform in width, and are usually limited to the allochems.

(iii) Partial collapse of the micrite envelopes of some shell debris.

(b) Late diagenetic deformation:

This process took place after the complete lithification of the rocks. This type of deformation is represented mainly by the intensive fracturing of the rocks. Fractures formed by this process are mainly of sharp-edged type and reach up to several centimeters in thickness. They are related to tectonism.

B.3 PRESSURE SOLUTION:

Two stages of pressure solution have been identified, namely, early diagenetic and late diagenetic.

(a) Early diagenetic pressure solution:

This process was active before the complete cementation of the sediments. It is very well displayed in the packed biomicrite rocks. A result of this process was the formation of microstylolitic grain boundaries. Pressure solution usually results in the dissolution of the carbonates at grain contacts and reprecipitation of the dissolved calcium carbonate in pores and cavities. In the early diagenetic pressure solution, the dissolution fabric is very well preserved (microstylolites), while the reprecipitation fabric is absent. This is because the dissolved carbonates could migrate out of the sediments via the connate water.

(b) Late diagenetic pressure solution:

This process was responsible for the late diagenetic compaction of the carbonate rocks and led to their bulk volume reduction. The mechanism was by stylolitization through pressure solution of carbonate mineral matter.

Pettijohn (1957, p. 213) defined stylolites as: "as surface marked by interlocking of mutual interpenetration of the two sides. The teeth-like projection of the one side fits into sockets of like dimensions on the other". The apex of each of the cone-like projections is characterized by the presence of a grain or groups of grains less susceptible to pressure solution than the grains on the opposite side of the stylolitic seams.

Sometimes, the pressure solubility of the grains on both sides of the pressure solution contact was equal, and the only visible remnant of this type of solution is a thin seam of insoluble residue that may easily be mistaken for a primary sedimentological feature. The factors that govern the absolute and relative pressure-solubility of carbonate grains are not completely understood at present. Trunit (1968) has shown that, among other things, shape of grains, size of grains, surface tension, temperature and pore fluid composition are important.

Stylolites are very common in most of the lithotypes studied (Plate 10, Figs. 3 and 4). Although most of them are horizontal, many are vertical or inclined. They vary in shape from simple undulatory to complicated ceratitic types. The cone-like apices vary from smooth slightly flat to very sharp elongated shapes. The stylolite amplitude varies from 1 to 15 mm . Concentrations of insoluble residue along stylolitic seams are common. They range in thickness from 0.1 to 2.00 mm.

In the light-coloured limestones, stylolitic seams consist of clay minerals and silt-size quartz grains, while in the dark-coloured rocks, they are formed of organic matter.

C. PHYSIO-CHEMICAL PROCESSES

Replacement of aragonite by calcite is the only process noticed in the studied rocks.

ARAGONITE-CALCITE REPLACEMENT:

By comparing the carbonate mineralogy of the rocks under study with their analogues in Recent carbonate sediments, it is obvious that one of the main differences is the complete absence of aragonite in the ancient rocks. It is suggested that the original aragonite was completely replaced by calcite during the early stages of diagenesis.

Stehli and Hawer (1961) reported that of Recent high-Mg calcite, aragonite and low-Mg calcite, the first is very unstable whereas the other two may persist for a long time under natural conditions. They suggested that the increase of volume during change from aragonite to low-Mg calcite may affect the porosity, cementation and dolomitization of the sediments.

It is suggested that aragonite could have been present in three forms, in the original sediments, These are:

(a) as an aragonite 'micrite' matrix in biomicrites and poorly washed biosparites. This might have consisted of fine silt size grains formed by the mechanical breakdown of aragonite skeletal debris and by direct precipitation from the sea water; (b) as coarse-grained skeletal debris; and (c) as oolites and pellets.

The problem of the relative stability of calcite and aragonite has long been of interest to both sedimentologists and geochemists. Fyfe and Bischoff (1965) and Bathurst (1971) have recently reviewed the problem. Bathurst (1964) suggested two processes for the replacement of aragonite by calcite, namely: (i) in situ inversion; and (ii) solution-deposition conversion. Friedman (1964) has also recognised these two basic processes, which he termed a paramorphic replacement and solution-infilling.

Fyfe and Bischoff (1965) separated the problem into dry transformation and wet transformation of aragonite to calcite. The most compelling argument against inversion of aragonite skeletal material in dry system seems to be the lack of evidence for a volume change corresponding to the density differences between calcite and aragonite. The inversion processes should involve an 8 percent increase in volume. Such a volume change should leave obvious petrographical evidence, but none has been observed in this study or reported in the literature.

Two processes of replacement of aragonite by calcite appear to have taken place in the studied rocks.

(a) Leaching of aragonite during the early stages of diagenesis was found to be the most common process. By this process aragonite of bioclasts was dissolved and the resulting cavities then filled with drusy calcite cement. The micrite envelopes of the skeletal fragments played an important role in their preservation. After the aragonite core had been dissolved away, it left the micrite envelope as a mould.

Although micrite envelopes in Recent sediments are known to be made of aragonite and high-Mg calcite (Purdy, 1968 and Winland, 1968), it has been found that it survives after the dissolution of the aragonite cores of the skeletal fragments. Shearman and Skipwith (1965) and Kendall et al (1966) suggested that where the grain was dissolved during diagenesis, it was the organic matter of the envelope that remained as the mould. Winland (1968) suggested that the stability of the micrite envelope is due to the fact that high-Mg calcite forming the micrite envelope would not dissolve but would simply lose its Mg^{2+}, as in Land's stage III. The present petrographic studies have revealed that micrite envelopes collapsed and broke down during compaction in the early diagenetic stage. These phenomena are not in favour of the ideas of Kendall et al., because the micrite envelopes were brittle at the time of fracture and not simply elastic organic matter.

(b) In situ inversion of aragonite to calcite has been rarely observed in the present study. The process is evidenced by the presence of some molluscan shells that are now formed of calcite, but in which the fine lamellar fabric of the former aragonite shell is preserved as ghosts by organic matter preserved in the calcite. This fabric could only be formed by in situ replacement of aragonite or high-Mg calcite by low-Mg calcite.

The mechanism of the replacement of aragonite by calcite in oolites has been described by Shearman et al. (1970). They suggested that the tangentially arranged aragonite crystals were dissolved leaving cavities between the concentric layers of algal mucilage, which were then filled with prismatic calcite crystals radially arranged on the algal mucilage layers. It is most likely that the same mechanism was responsible for the replacement of aragonite by calcite in the oolites of the rocks under review.

D. CHEMICAL PROCESSES

These include cementation, dolomitization, dedolomitization and silicifation.

D.1 CEMENTATION:

The cementation in the various carbonate lithotypes was studied in detail. The distinction between chemically precipitated sparry calcite cement and pseudo-sparite formed by the recrystallization of micrite is based on the sixteen fabric criteria that have been listed by Bathurst (1971, p. 417).

Regarding cementation, four aspects have been studied: (a) the sources of the carbonate cement (b) the fabric of the cement; (c) the mineralogy of the cement; and (d) the genesis of the cement.

a. The sources of the carbonate cement:

It is suggested that the carbonate material forming the cement could have been derived from the following sources:

(i) Calcium carbonate in solution in the original sea water could have been important in the very early stage of cementation. Calcium carbonate could have been precipitated in the pore spaces of loose sediments while they were still submerged under the sea water. Direct precipitation of low-Mg calcite in the pore spaces of Recent and Sub-recent carbonate sediments from sea water has been reported by MacIntyre et al., (1968) and Milliman (1966).

(ii) Excess calcium carbonate produced by the change from aragonite to calcite: "inversion" of aragonite to calcite should produce an excess of 8.48% calcite by volume to act as an interparticle cement (Oldershaw and Scoffin, 1967). In the case of biomicrites, the change of aragonite matrix to calcite matrix would produce an amount of calcium carbonate sufficient for the formation of the present calcite cement.

Comparing the volume of the oolites to that of the present cement in the oosparite rocks, excess calcium carbonate resulting from the alteration of the original aragonitic oolites to the calcite could be responsible for the precipitation of at least one third to one half of the overall cement of the rock. In the biosparites, it seems to be that this source was less important.

(iii) Calcium carbonates released by pressure solution: two stages of pressure solution have been identified in the rocks studied, namely: early diagenetic and late diagenetic. Early diagenetic pressure solution along grain contacts (microstylolites) would release calcium carbonate which could then reprecipitate as intergranular cement. Calcium carbonate in solution produced by the late diagenetic stage of pressure solution may contribute to the cementation of the cavities, vugs and fractures.

b. Cement fabrics:

It was found that two main factors controlled the fabrics of the cement. These were: (i) the nature of the place in which the cement precipitated (e.g. intergranular pores, vugs, moulds, caves, fractures, etc); (ii) the nature of the substrata to which the crystal of cement was attached (e.g. polycrystalline grains, monocrystalline grains etc.).

In the description of the textures and fabrics of the cement, the terms suggested by Folk (1965), Friedman (1965) and Bathurst (1959), have been used.

On the basis of fabric, the following types of calcite cement have been recognised.

(i) Fibrous calcite cement:

In this type, the calcite cement consists of finely crystalline, bladed crystals arranged radially around the grains. This fabric is common in almost all of the samples studied. It commonly lines the intergranular pore spaces, moulds and vugs. On the basis of Folk's classification of cement fabrics, this texture is, a "precipitative finely crystalline bladed crust ($P.B_3.C.$)", (Plate 11, Fig. 1).

(ii) Radiaxial fibrous mosaic cement:

Calcite crystals forming this type of cement have their long axes normal to the surface of growth. They have curved cleavage traces, and generally show wavey extinction that extends through essentially the entire thickness of the crust. This type of cement occasionally lines the brachiopod body cavities (Plate 11, Fig. 3). The thickness of the radiaxial fibrous crust is approximately constant in a single specimen, but varies in different specimens from 0.2 to 0.8 mm.

All radiaxial fibrous calcite crystals are distinctly "dirty" or "cloudy" in thin section, as a result of the presence of numerous extremely fine inclusions. The texture of this cement is: "precipitative, - moderately to coarsely crystalline fibrous crust $(P.F_{4,5}.C.)$".

In some specimens it was found that radiaxial fibrous calcite crystals are developed as overgrowths on the prismatic calcite crystals of some brachiopod shells (Plate 11, Figs. 5 and 6). This fabric has been observed to form a discontinuous crust. It is usually developed when the prismatic calcite crystals forming the shells are perpendicular to the shell surface and where the surface of the shell is very clean.

(iii) Equicrystalline mosaic cement:

This cement fabric is divided into two types according to crystal size, namely: medium equicrystalline mosaic cement, and coarsely crystalline equicrystalline mosaic cement.

1. Medium equicrystalline cement:

The sparry calcite forming this type of cement consists of equidimensional crystals, uniform in size, and with axial ratios ranging from 1:1 to 2:1. They vary in diameter from 80 to 200μ. The crystals have straight boundaries and usually become coarse towards the cavity centre. This fabric is frequently developed in intergranular pore spaces, moulds, small cavities, and in some narrow fractures where fibrous calcite cement was not sufficient to completely fill these spaces. The texture of this cement is a "precipitative medium equicrystalline, randomly oriented calcite crystals (P.E_4)".

2. Coarsely to very coarsely equicrystalline mosaic cement:

Calcite crystals forming this type of cement are well to moderately sorted, equant, with an axial ratio of about 1:1. They range in diameter from 0.3 to more than 3 mm. They are characterised by straight boundaries and with enfacial junctions (see Bathurst, 1971, p. 423). This cement usually fills body cavities, coarse fractures and dolomite cavities (Plate 13, Fig. 2). The texture is "precipitative, coarsely to very coarsely crystalline, randomly oriented calcite crystals (P.$E_{6,7}$)".

(iv) Rim cementation:

This is the syntaxial overgrowth of calcite on echinoderm fragments. This type has been described in detail by Evamy and Shearman, (1965) and (1969). It is the most frequent type of cementation in the biosparites and bio-oosparites. In the crinoidal biosparites, rim cementation comprising from 70 to 100 percent of the cement. Serrated compromise boundaries are common between adjacent crystals (Plate 12, Fig. 3). Poikilotopic textures of more than one allochem enclosed in monocrystalline calcite overgrowths are frequent (Plate 5 , Fig. 2).

Two types of rim cementation have neen noticed, namely: precipitative and neomorphic. The precipitative rim cement was formed by the precipitation of calcium carbonate from solution in optical continuity on the echinoderm fragments ($P.E_{5,7}.O_M$). The neomorphic rim cement was developed during the "inversion" of the aragonitic micrite that formerly surrounded the echinoderm fragments, to calcite. The newly formed calcite crystals were rearranged in such a way as to be in optical continuity with the echinoderm fragments ($N.E_{43}.O_M$), (Plate 12, Fig. 4).

Precipitative rim cement is commonly of clear calcite and occurs in the biosparites, while the neomorphic type usually is present in biomicrites and is rich in inclusions.

c. Mineralogy of the cement:

By applying staining techniques it was found that the cement consists of non-ferroan calcite, ferroan calcite and dolomite.

(i) Non-ferroan calcite cement:

It is the most frequent cementing mineral. It was found that the following cement types are formed mainly of non-ferroan calcite: the fibrous cements; radiaxial fibrous mosaic cement; most of the medium equicrystalline mosaic cement, and part of the rim cement.

Non-ferroan calcite cement can be precipitated subaerially or submarine, but in oxidising conditions (Evamy, 1969). It could have been formed while the sediments were still submerged, or after the emergence of the sediments in a subaerial setting, but above the water table.

(ii) Ferroan calcite cement:

This mineral usually forms the coarsely equicrystalline mosaic cement and sometimes the medium equicrystalline mosaic cement and occasionally the outer zones of rim cement. It commonly occurs filling brachiopod body cavities, some of the fractures, and is occasionally present in intergranular pore spaces. In dolomites, it is the most frequent mineral cement, filling most of the pore spaces and the fractures.

Zoned calcite, in which zones of ferroan and non-ferroan calcite are developed in a single crystal, are very frequent in the fracture-filling cement of some of the dolomites

(Plate 13, Fig. 1). In these fractures, calcite cement starts to develop as ferroan calcite, followed by zones of non-ferroan calcite precipitated in optical continuity on the earlier formed ferroan calcite. Zoned calcite has also been recognised in the syntaxial calcite overgrowths on echinoderm fragments in the crinoidal biosparites (Plate 12, Fig. 5).

The precipitation of the ferroan calcite is controlled by two main factors, namely: (1) the availability of ferrous ions; and (2) physiochemical conditions of the diagenetic environment. In their study on the Wenlock limestones of North Wales, Oldershaw & Scoffin (1967) suggested that clay minerals were the source of iron in the ferroan calcite cement. Evamy (1969) suggested that ferroan calcite is precipitated in reducing environments where the ferrous form of iron is usually stable. He suggested also that this condition is commonly developed in sediments rich in organic matter and buried below the water table.

(iii) Dolomite cement:

Dolomite cement has been noticed to occur only in the dolomite rocks. It is present as idiomorphic equicrystalline crystals filling fractures and intergranular pore spaces. Although ferroan dolomite is the most frequent form of dolomite cement, non-ferroan dolomite occasionally is present as a thin clear zone within the ferroan dolomite filling fractures in some of the rocks (Plate 14, Fig. 1). As in the case of ferroan calcite, ferroan dolomite is characteristic of reducing diagenetic environments which are commonly present below the water table. Oscillation in the Eh value of the diagenetic environment could have been responsible for the formation of alternate zones of ferroan and non-ferroan dolomite cement.

In some dolomite rocks, barite and galena have been occasionally seen to occur as fracture-filling cement with calcite and dolomite (Plate 14, Fig. 1).

d. Genesis of Carbonate Cement:

As a result of the detailed petrographical study of the cement of the carbonate rocks, four stages of cementation have been identified.

(i) Stage 1: In this stage cement has been developed shortly after the deposition of sediments, in submarine or subaerial conditions. The fibrous cement and partial rim cementation were developed in this stage. Cement of this type is usually formed of non-ferroan calcite.

(ii) Stage 2: The medium equicrystalline mosaic is commonly developed in this stage, also precipitation of rim cement continued to develop. Cementation of most of the moulds, the chambers of the microorganisms and the intergranular pores was completed in this stage. Cement developed in this stage is generally of non-ferroan calcite, but in a few rocks ferroan calcite has been precipitated.

(iii) Stage 3: Coarsely equicrystalline mosaic cement was precipitated as ferroan calcite at this stage.

(iv) Stage 4: This was a late diagenetic process that took place after dolomitization and tectonic fracturing of the rocks. Non-ferroan and ferroan calcite and dolomite cement filling fractures and intergranular pore spaces of dolomites were also precipitated in this stage.

D.2 DOLOMITIZATION:

Dolomitization was the most important and the most frequent diagenetic process in the Carboniferous Limestone Series of the Forest of Dean and South Wales. It varies in extent from slight dolomitization represented by a few rhombohedra scattered in the limestones, to complete dolomitization of all of the rock. Dixon and Vaughan (1911, p. 510) described the partially dolomitised biomicrite bed, known as pseudobreccia, of the D zone of Gower, and they suggested that the dolomitization was mainly due to the effect of Avonian sea water.

Dixey and Sibley (1918) suggested two origins of the dolomites of the south eastern margin of South Wales, namely:

Penecontemporaneous dolomitization and subsequent dolomitization (vein-dolomite). They mentioned also that dolomite could be of double origin - "Moreover, it is probable that many cases occur in which limestone partly altered by penecontemporaneous dolomitization was further affected by vein dolomitization at a later date".

Dixon* in his study of some parts of the South Wales Coalfield mentioned that dolomitization was contemporaneous rather than the result of subsequent alteration along veins or faults. The following paragraph is quoted from the Newport memoir, 2nd edition*, pages 19-20: "The rocks were at first calcite as shown by their containing crinoidal remains and the expression contemporaneous is introduced as implying dolomitization while they were still under the influence of the Carboniferous Limestone sea". Trotter (1942) distinguished two types of dolomite in the Forest of Dean, namely, dolomite and dolomite mudstone. He suggested a contemporaneous origin to the dolomites, which were produced by the alteration of limestones in the Lower Carboniferous Sea during or shortly after deposition. The dolomite mudstones on the other hand were the result of the deposition of dolomite mud.

Parsons (1918) distinguished two main genetic types of dolomite of Leicestershire, primary and secondary dolomite. Primary dolomites are of two types, those deposited as clastic

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* Strahan, M.A., 1909: Newport Mem. British Regional Geology. London.

rocks derived from pre-existing dolomite and those chemically precipitated as dolomite with or without the agency of organisms. Secondary dolomites are also of two types, contemporaneous dolomites are those deposited as ordinary limestones which were altered soon after deposition by the influence of magnesium salts in the sea in which the rocks were originally deposited. Subsequent dolomites are those deposited as ordinary limestones, but which were altered by the influence of waters belonging to some later period.

George (1972) proposed a penecontemporaneous origin of the dolomites in South Wales and the Forest of Dean, based on the occurrence of dolomite pebbles found to occur in the Upper Avonian basal conglomerates in Breconshire. He mentioned also that there are no recognisably primary dolomites amongst the Avonian limestones.

a. Fabric types of dolomites:

According to the fabric study of dolomites and dolomitized limestones, two types have been revealed, namely: finely crystalline dolomites or dolomicrite and coarsely to very coarsely crystalline dolomite. The fabric and the mineralogy of each type have been described in detail in Chapter II.

Finely crystalline dolomite or dolomicrite is similar to the dolomite-mudstone of Trotter (1942) and primary chemical precipitated dolomite of Parsons(1918). Coarsely to very coarsely crystalline dolomite is equivalent to the contemporaneous and subsequent dolomite of Parsons(1918) Dixey and Sibly (1918), Dixon (1911) and Trotter (1942).

b. The mechanism of dolomitization:

The discussion below is based on the unifying model for dolomitization of Friedman and Sanders (1967, p. 334), who concluded that all "dolostones" whether syndiagenetic, anadiagenetic or epidiagenetic are the result of the action or reaction of hypersaline brines. The mechanism by which the finely crystalline and the coarsely crystalline dolomites of South Wales and the Forest of Dean were formed was that of reaction between hypersaline brines and the pre-deposited desiments. The main variations between the different types of dolomite are related to time of dolomitization and the diagenetic environments.

b.1 Finely crystalline dolomites (syndiagenetic)

Based on the study of Recent carbonate sediments, the finely crystalline dolomites could be of primary or penecontemporaneous replacement origin. Recent primary dolomites have been reported by Alderman and Skinner (1957) from shallow isolated lakes of Coorong Lagoon, South Australia, and by Jones (1961) from the Deep Spring Lake Playa, California, U.S.A. In all of the previous examples the primary dolomites were believed to be precipitated directly from solution, where the water was very saline, the pH was high and plants abundant. Plants extract carbon dioxide from water during photosynthesis, this raises the pH and promotes precipitation of dolomite.

Penecontemporaneous replacement as a process for formation of Recent dolomite has been described and reported by many authors, (see Friedman and Sanders, 1967). Two processes have been suggested for the formation of this type of dolomite.

(i) Capillary movement of sea water, initially of normal or near normal salinity through sediments on marginal supratidal flats and leads to concentration by evaporation.

(ii) Downward flow of dense brine may occur either in the main water mass or in the interstitial fluids of sea or lake-marginal sediments. This is the seepage refluxion process of Adams and Rhodes (1960).

Usually the finely crystalline dolomites of both South Wales and the Forest of Dean are thin-bedded with traces of boring and burrows, so they were most probably deposited in subtidal or lagoonal environments. Concerning the origin of these dolomites, the direct precipitation from sea water is unlikely, because the conditions under which this type of dolomite could be formed should lead to precipitation of evaporite minerals. The latter were not observed or reported in the area of study. Because of this, it is suggested that the most likely process for the formation of the finely crystalline dolomites was penecontemporaneous replacement of previously deposited micrite (most probably as aragonite) by the downward flow of dense brine in the main water mass.

b.2 Coarsely and very coarsely crystalline dolomites:

Diagenetic coarsely and very coarsely crystalline dolomites have been developed by partial or complete dolomitization of limestones. Two stages of dolomitization have been recognised. In the first stage, dolomite, represented by finely to medium crystalline rhombohedra, preferentially replaced micrite bodies such as micrite matrix, micrite envelopes, oolites and pellets. In the second stage, dolomitization affected the rock as a whole and dolomite is present as coarsely crystalline rhombohedra. The following are the main lines of petrographic evidence:

(i) In the partially dolomitized limestones, dolomite is present as finely crystalline rhombohedra preferentially replacing micrite bodies.

(ii) In the completely dolomitized rocks, finely crystalline dolomite rhombohedra that were concentrated in pre-existing micrite envelopes, are now enclosed in one large dolomite crystal. This is termed poikilotopic texture, (Plate 15, Fig. 1).

It is suggested that the first stage of dolomitization could have occurred just after deposition by the action of dense brine seepage downward through the water mass. Thus it was actually penecontemporaneous dolomitization.

The second stage of dolomitization is believed to have started after partial cementation, by the action of hypersaline connate water. It is difficult to deduce the age of dolomitization, but in some places in Gower it is evident that dolomitization started in Avonian times. The evidence for this is the occurrence of unconformity surface above the partially dolomitized biomicrite of D zone (pseudobreccia). This means that dolomitization pre-dated the unconformity surface. At this surface dedolomitization has been identified.

In the present study it was difficult to differentiate between the diagenetic and epigenetic dolomites petrographically with certainty. However, in the field, epigenetic dolomites are usually associated with sulphides and ore deposits.

c. Selective dolomitization:

In partially dolomitized rocks, certain depositional carbonate constituents were more readily replaced by dolomite than others. From microscopic observations, an order of decreasing susceptibility of the depositional constituents to dolomitization can be established:

(i) The carbonate content of shale and shaly carbonates was selectively dolomitized in those rocks that were incipiently dolomitized.

(ii) In the biosparites and biomicrites it was found that the micrite envelopes that covered most of the calcitic and aragonitic bioclasts are selectively dolomitized with respect to both the micrite matrix and the allochems.

(iii) Where micrite matrix and allochems occur together, the micrite matrix is selectively dolomitized. In many samples of crinoidal biomicrites, the micrite matrix is completely dolomitized, whereas the allochems are only lightly dolomitized or unaffected.

(iv) Pellets, oolites and original aragonitic bioclasts were selectively dolomitized, while original calcitic bioclasts were unaltered.

(v) It was found that the original calcitic bioclasts (crinoidal and brachiopod debris) were least susceptible to dolomitization.

d. Sources of magnesium needed for dolomitization:

d.1 Sea water;

Magnesium can be concentrated in hypersaline solutions by evaporation of sea water in supratidal environments. Hypersaline solutions can also be developed by evaporation of connate water trapped in pores of incipiently cemented sediments. These are thought to be the main sources of magnesium responsible for the formation of most of the penecontemporaneous dolomites.

d.2 High-Mg calcitic skeletal grains:

Magnesium could be derived from dissolution of high-Mg calcitic skeletal grains by fresh water in the vadose zone. Precipitation of calcium carbonate from interstitial solution in the form of calcite, as a first phase of cementation, results in the concentration of Mg ions in the rest of the solution. This source could contribute to the early stage of diagenetic dolomitization.

d.3 Clay minerals:

Clay minerals may contain appreciable quantities of magnesium ions either in interlattice sites, or in exchange positions resulting from broken edges or unbalanced charges. Magnesium is especially common in some montmorillonite, chlorite and vermiculite which may contain 25 percent or more MgO (see Deer, Howie and Zussman, 1962, vol. 3).

There are two main mechanisms by which clay minerals could contribute to the addition of magnesium:

d.3.1 Ion exchange:

Ion exchange mechanism might be activated by dissolution of carbonate rocks and release of appreciable quantities of calcium ions to the pore waters. According to Chave (1960) the addition of such ions to interstitial waters could increase the amount of exchangeable magnesium ions that could be removed from clay minerals and replaced by calcium. The clays that acted as potential sources of magnesium in the Carboniferous Limestone Series of South Wales and the Forest of Dean could have been one of the following:

(i) Clays of shale beds in the Lower Limestone Shale Group.

(ii) Disseminated clay particles in the micritic parts of the limestone rocks.

The addition of magnesium ions to the interstitial water by ion exchange could have occurred in two stages.

Firstly, in the early diagenetic stage during wet replacement of aragonite by calcite. Ion exchange could have been activated between the calcium carbonate solution and the suspended clay particles that were disseminated in the aragonitic micrites. Secondly, ion exchange could have taken place in the late diagenetic stage between the insoluble clays, concentrated on the stylolitic surfaces, and the calcium carbonate solution.

d.3.2 Reaction between clay minerals and carbonates:

Possible ways that clay minerals within sediments may take part in reactions which involve the formation of dolomite have been investigated by several workers, especially by Zen (1959). From Zen's diagrams and equations possible reactions may be illustrated as follows:

Calcite + chlorite + carbon dioxide = dolomite + kaolinite
+ quartz + water

Montmorillonite + calcite + carbon dioxide = dolomite+kaolinite
+ quartz + water

Limited petrographical and mineralogic work supports the essential validity of these reactions although numerous problems remain concerning the chemistry involved and the quantitative importance of dolomites which may have been derived by such reactions.

D.3 DEDOLOMITIZATION:

Replacement of dolomite by calcite becomes one of the interesting processes in the diagenesis of the carbonate rocks. The process has been termed dedolomitization by most authors. Some other authors used the term calcitization, among them Swett (1965) and De Meijer (1971). The term dedolomitization was first used by Von Morlot (1848) who suggested the following reaction:

$$CaMg(CO_3)_2 + CaSO_4.2H_2O = 2CaCO_3 + MgSO_4 + 2H_2O$$

Sulphate ions were originally suggested as being derived from the dissolution of gypsum or anhydrite, but they could also be formed from pyrite oxidation (Evamy, 1963). According to Evamy (1967) and De Groot (1967) dedolomitization is caused by solution with a high Ca/Mg ratio reacting with dolomite to form calcite. Experimental work by Yanateva (1955) and De Groot (1967) indicates that dedolomitization can only take place at or near the earth's surface, where carbon dioxide pressure and temperature are relatively low.

a. Occurrences of dedolomitization:

Evidence of dedolomitization is frequent in the study area, but it is more common in the Forest of Dean rather than in South Wales. On the basis of field relations two types of dedolomite have been distinguished, namely: recent dedolomite and fossil dedolomite (Fig. 3.1).

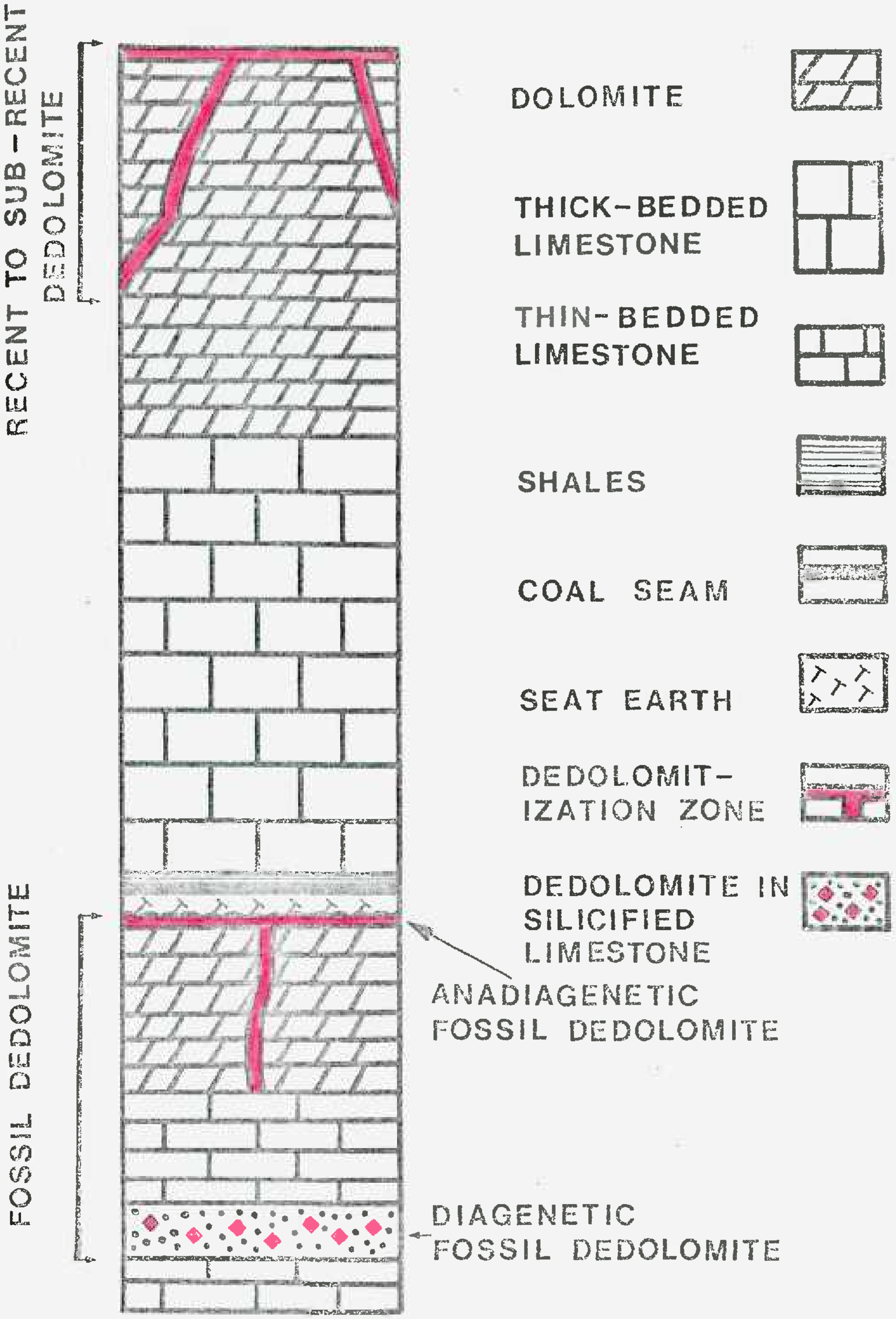


Fig. 3-1 Schematic diagram shows the mode of occurrance of Dedolomite in the Carboniferous Limestones of South Wales.

(i) Fossil dedolomite:

The term fossil dedolomite refers to dedolomite formed during subaerial exposure of dolomite in Carboniferous times. This type of dedolomite is frequently found in the top part of the partially dolomitized biomicrite beds of the D zone in Gower (pseudobreccia). There, dedolomitization occurred just at the contact between this bed and the overlying clayey seatearth. The occurrence of the seatearth and thin coal seam is absolute evidence for subaerial exposure of the dolomitized bed in pre-seatearth time. In this locality it was found that dedolomitization took place on the surface and by the action of meteoric water which penetrated only about 8 cm. into the rock. This indicates that the rock at that time had some porosity. Deeper penetration took place by way of fractures. On both sides of the fractures the meteoric water again penetrated zones of only 1 to 2 cm.

Another form of fossil dedolomite has been identified in the silicified rocks. These rocks are silicified partially-dolomitized biomicrites, and they contain considerable amounts of finely crystalline dedolomite. Dedolomitization in these rocks took place concurrently with silicification. Silicification of calcite liberates calcium ions. Solutions near such silicified calcite crystals will consequently have a high Ca/Mg ratio. Reaction of these solutions with dolomite crystals could result in dedolomitization. In the completely silicified rocks, dolomite rhombohedra are preserved due to the fact that dolomite was less susceptible to silicification than calcite. After dedolomitization of the dolomite rhombohedra, silicification started to corrode the calcite pseudomorph after dolomite. Some of the rhombohedra were completely silicified and some only partially silicified. This is evidence that the dedolomitization was concurrent with silicification (Plate 16, Fig. 3).

Two genetic types have been recognised amongst the fossil dedolomites. Firstly, anadiagenetic fossil dedolomite. This usually occurs near unconformities. Secondly, diagenetic dolomite, which is that which was generated as a by-product of silicification of dolomitized limestones.

(ii) Recent and sub-Recent dedolomite:

This is the most frequent form of dedolomite. The surface of most of the present day exposures are severely dedolomitized and alteration also occurs along fractures which sometimes reach up to 30 ft. in depth.

The dedolomitised zones are usually reddish and yellowish-brown in colour, which gives the rock a rusty appearance. This phenomenon is very well displayed by the dolomites of the Forest of Dean. The rusty appearance is usually more pronounced in those parts of the rock which are heavily fractured or jointed. In many cases the "rust" forms selvages to joints and bedding planes. This phenomenon has been reported by Al-Hashimi et al.(1973) in the dolomitic Carboniferous rocks of the Middle Limestone Group of the north east coast of Northumberland. He suggested that sea or fresh water circulating along permeable zones exposed at or near the present surface is responsible for, the oxidation and hydration of the ferrous iron in the metastable ferroan dolomites, as well as the dedolomitization of these dolomites. It is most likely that the same mechanism might be responsible for the formation of the rusty crust and dedolomitization in the study area.

b. Dedolomitization fabrics and textures:

The following textures and fabrics were seen in the dedolomites, amongst the rocks studied in this project.

(i) Composite calcite rhombohedra:

This is similar to the type (a) of Shearman et al., (1961). Former dolomite rhombohedra with micrite inclusions are now seen as rhombohedra composed of equicrystalline mosaics of calcite. The replacement of the dolomite crystals by calcite may be centripetal or centrifugal, and is usually not complete. This type of replacement is frequent in the partially dolomitized crinoidal biomicrites of the Lower Limestone Shale Group of the Gower Peninsula (Plate 16, Fig. 2).

(ii) Abundant massive limonite and limonite aggregates occur as pseudomorphs after dolomite rhombohedra. They frequently contain inclusions of dolomite or calcite. Usually the dolomite or calcite remnants occupy the centre of the rhombohedra.

(iii) Monocrystalline calcite rhombohedra:

This is the most abundant of the dedolomitization fabrics. Dolomite rhombohedra with zones of ferric oxide are now seen as single rhombohedral crystals of calcite that contain the original ferric oxide zones. This fabric has been noticed in the dedolomitized coarsely crystalline dolomites and in the dedolomite rhombohedra preserved in the silicified limestones. In this type of replacement original features of some of the former dolomite, such as the curved cleavage and brush extinction, are retained in the newly formed calcite.

(iv) Partially dedolomitized rhombohedra:

This is a very frequent fabric, especially in the eastern area of South Wales and the Forest of Dean. In these rocks the rhombohedra comprise alternating zones of dolomite and dedolomite with the same optical orientation.

The following are the fabric types that have been recognised in this rock variety:

(a) The rhombohedron consists of dedolomite with an outer thin zone of clear dolomite (Plate 15, Fig. 5).

(b) The rhombohedron consists of a dolomite core followed outwards by a zone of dedolomite and then another zone of clear dolomite (Plate 15, Fig. 6).

(c) The rhombohedron consists of dedolomite with irregular patches of relict dolomite.

In the unaltered equivalent rock, it was found that the original dolomite rhombohedra consist of alternating zones of cloudy ferroan and clear non-ferroan dolomite. Because ferroan dolomite is usually unstable under the sub-aerial conditions, it is readily altered to calcite, while the clear zones of non-ferroan dolomite, being less susceptible, stay unaltered. It is obvious that the present dedolomitization fabrics are controlled by the original mineralogy of the dolomite.

Many authors have reported that dedolomitization could lead to regeneration of the pre-dolomitization texture of the original limestone; Shearman et al (1961), Evamy (1967) and De Meijer (1971). There is no evidence of this in the rocks under discussion.

D.4 SILICIFICATION:

By contrast to dolomitization, silicification of the Carboniferous Limestone rocks is rare in South Wales and almost absent in the Forest of Dean. Silicified carbonate rocks are present as thin bands of about 5 cm. in thickness and as elongated nodules parallel to the bedding planes. They occur in the Z zone at Gower and in the S zone of the northern outcrop in South Wales.

Beekitized skeletal debris is frequently present in the crinoidal limestones of the Lower Limestone Shale Group.

a. Fabrics of silica

The following fabric types of silica were recognised:

(i) Idiotopic quartz:

Idiotopic quartz crystals were found preferentially replacing micritic bodies, such as oolites, pellets and algal micrite. Silicification was responsible for the development of quartz overgrowths on the detrital quartz grains forming cores of some oolites (Plate 17, Figs. 3 and 4). The overgrowth rims vary in width from very thin of about 10 microns replacing one or two oolite shells, to almost complete replacement of an oolite and part of the sparry calcite cement. Ghosts of oolite shells are displayed in some idiotopic quartz crystals occupying the cores of some oolites. In the oosparites, idiotopic quartz crystals were found to range in size from 50 to 200 microns. In the algal lumps, the quartz crystals are prismatic and finely crystalline, being about 20 microns in length (Plate 17, Fig. 5). Clusters of idiotopic and hypidiotopic quartz crystals were observed to partially replace some brachiopod shells (Plate 17, Fig. 6). The original textures of the shells are preserved as ghosts in the quartz aggregates.

(ii) Microcrystalline granular quartz:

This is the most frequent fabric in the completely silicified micrites and biomicrites of the Z zone in Gower. These rocks consist completely of microcrystalline granular quartz. The quartz crystals are xenotopic and are aphanocrystalline to finely crystalline; they are of about 10 microns in average size. The interparticle relations are too small to be determined optically. This type of quartz fabric is the main fabric recognised in the partially silicified crinoidal limestone (Plate 17, Figs. 1 and 2). It was found also that the completely silicified calcarenites of the S zone of the northern outcrop of South Wales contain appreciable amounts of this quartz type, and it usually occupies the central part of the allochems.

(iii) Chalcedonic overlay:

This fabric was described by Wilson (1966). Under crossed polars, the chalcedonic overlays appear fibrous, each fibre having an extinction approximately parallel to that of the overlay. It commonly rims silicified allochems and lines voids. Successive layers of chalcedonic overlays are recorded by brown colour bands (Plate 16, Figs. 5 and 6).

(iv) Spherulitic chalcedony:

This fabric is characterised by radiating fibrous extinction under crossed polars. It is usually pale olive in colour. This variety occurs in incipiently silicified brachiopod shell fragments.

b. Sources of silica

Silicification usually occurs in the biomicrite and biosparite beds intercalated with shales in the Lower Limestone Shale Group, and in various lithotypes in the Main Limestone Group.

There are two possible sources of the silica:

1) Silica could have been leached from the clay minerals of the shaley beds by the action of interstitial water. On compaction, these interstitial silica-rich waters could

have migrated to the relatively more porous biomicrites and biosparites present within the shale sequence. Leaching of silica, from clays, by connate water, is accelerated in reducing conditions. The connate water in the shales becomes isolated, all its sulphate ions have been removed by reduction to sulphide (pyrites are abundant in the associated shale beds), and all its calcium, magnesium and carbonate ions are precipitated as dolomite. This results in a water with an extremely high pH (see Bass, Becking et al., 1960, p. 276), which facilitates the dissolution of the silica.

2) The slight silicification of the oosparites and biosparites in the Main Limestone Group is explained by the ability of the living algae to hold considerable concentrations of silica in their skeletons. The presence of algae in the original sediments is demonstrated by the presence of algal mucilage in the oolites and in the algal micrite that envelopes some of the bioclasts. It seems quite probable that in the case of the oosparites and biosparites high pH was created by algal photosynthesis and this allowed high concentrations of silica in the connate water. The silica was subsequently precipitated when the pH fell.

c. Genesis of silica

Genetically, two types of silica have been distinguished, namely, replacive and precipitative. The replacive type of silica is the most frequent. Replacement of carbonate by silica occurred in various stages of the diagenetic history of the carbonate rocks studied.

In the completely silicified calcarenites it was found that silica started to replace the rock just after the first phase of cementation because the silica replaces the allochems and the fringes of calcite cement. The main evidence for replacement is the well-preserved original texture of the rock. The inter-allochem pore spaces are filled with precipitative silica of chalcedonic overlay fabric. Precipitative and replaceive silica could be synchronised in these rocks.

In the completely silicified micrites and biomicrites it was found that the original texture of the rock is well preserved. A replacive origin of silica in these rocks is suggested. The occurrence of former dolomite rhombohedra which is now dedolomite indicates that replacement by silica took place after incipient dolomitisation.

In some crinoidal limestones of the Lower Limestone Shale Group it was found that silicification has partially affected both dolomotized and undolomitized rocks. In the dolomitized rocks, two stages of dolomitization were recognised, namely, early stage represented by finely crystalline penecontemporaneous dolomites, and a late stage represented by coarsely crystalline diagenetic dolomite. The finely crystalline dolomite rhombohedra were found enclosed in the silicified areas of the bioclasts, while the coarsely crystalline dolomite was found to be present in the unsilicified areas. Therefore, it is concluded that silicification post-dated the penecontemporaneous dolomitization and pre-dated the diagenetic dolomitization.

DIAGENETIC HISTORY OF THE CARBONATE ROCKS

The following is a chronological review of the main diagenetic events that took place during the history of each lithotype.

1. MICRITE

a. Thin-bedded micrite:

i) Compaction and penecontemporaneous deformation resulted in formation of cracks and crumbly fractures.

ii) Replacement of aragonite by calcite and slight penecontemporaneous dolomitization.

iii) Stylolitization took place after complete lithification of the rock.

iv) Tectonic deformation then took place resulting in the formation of late diagenetic fractures.

v) Precipitation of the sparry calcite of stage (4) was responsible for cementation of the tectonic fractures.

b. Dismicrite:

i) Early diagenetic compaction of micritic sediments was the main cause of expulsion of intergranular water and gas to form relatively large bubbles by coalescence.

ii) Incipient lithification of the micrite sediment was responsible for the preservation of the gas and water bubbles as vugs.

iii) Replacement of aragonite by calcite, and enlargement of vugs by slight dissolution then took place.

iv) Precipitation of fibrous drusy calcite as a crust that lines most of the vugs (stage 1 of cementation).

v) Deposition of the internal sediment from material in suspension in the water that filled the vugs.

vi) The rest of the vugs left after deposition of internal sediment were subsequently filled by equicrystalline mosaic calcite (stage 2 of cementation).

vii) After complete lithification of the rock, late diagenetic pressure-solution was responsible for the formation of stylolites.

2. BIOMICRITES

i) Biological processes affected the bioclasts just after their deposition. These included: micritization, algal boring and algal coating.

ii) Dissolution of aragonite then took place. The aragonite cores of the partially micritized skeletal fragments were then dissolved producing cavities supported by micrite envelopes.

iii) Precipitation of fibrous drusy calcite cement which lined most of the skeletal moulds as well as some shell body cavities. Also, neomorphic rim cement developed as overgrowths on echinoderm fragments.

iv) Incipient penecontemporaneous dolomitization was responsible for the development of finely crystalline dolomite rhombohedra scattered in the micrite matrix and the micrite envelopes.

v) Partial silicification of brachiopod shell fragments, micrite envelopes and algal grains by idiotopic quartz crystals, and spherulitic chalcedony then took place. Some rocks were completely silicified.

vi) The dolomite rhombohedra that were preserved in the completely silicified rocks were then dedolomitized.

vii) Precipitation of equicrystalline mosaic non-ferroan calcite was then precipitated as that filled most of the skeletal moulds and shell body cavities (stage 2 of cementation).

viii) Precipitation of equicrystalline mosaic ferroan calcite that filled the rest of the brachiopod shell body cavities (stage 3 of cementation).

ix) Late diagenetic pressure-solution was responsible for stylolitization. Concentration of idiotopic quartz crystals and finely crystalline dolomite along the stylolite planes is evidence for the fact that stylolitization post-dated incipient penecontemporaneous dolomitization and silicification.

x) Diagenetic dolomitization was responsible for the partial dolomitization of some rocks and complete dolomitization of others. Coarsely crystalline dolomite rhombs cut across stylolite seams in a manner that suggests that stylolitization pre-dated diagenetic dolomitization.

xi) Ancient anadiagenetic dedolomitization of diagenetic dolomites took place in Carboniferous time.

xii) Tectonic fracturing affected most of the rocks. It is very prominent in the western outcrops of the South Wales Coalfield.

xiii) Precipitation of the equicrystalline mosaic cement, built up of ferroan dolomite and non-ferroan and ferroan calcite as fillings of tectonic fractures and intergranular pores in dolomites (stage 4 - cementation).

xiv) Recent anadiagenetic dedolomitization. This was the latest process in the diagenetic history of the rock. It affected the rocks after they have been subaerially exposed in Recent or Sub-Recent times.

In the case of the packed biomicrites, early diagenetic compaction and pressure-solution were important. They were responsible for squashing of some shells and compression of oolites and algal pellets causing them to change their shape from spherical to elliptical. They are also responsible for the development of the microstylolitic boundaries between the allochems.

3. BIOSPARITES

i) Biological processes represented by micritization, algal encrustation and algal boring affected the skeletal debris just after deposition.

ii) Dissolution of the aragonite cores of the partially micritized skeletal fragments.

iii) Precipitation of fibrous non-ferroan calcite cement as a crust that lined moulds of skeletal fragments and intergranular pores. Syntaxial overgrowths of calcite on echinoderm fragments took place at the same time (stage 1 of cementation).

iv) Early diagenetic deformation was responsible for fracture of some brachiopod shell fragments and echinoderm debris, and partial collapse of some micrite envelopes. Crumbly fractures were originated at that time.

v) Incipient dolomitization represented by finely crystalline dolomite rhombohedra preferentially occurred in the micrite envelopes and in the micrite-filled cavities in some of the echinoderm debris.

vi) Silicification represented by microcrystalline quartz partially replaced some crinoid fragments. Complete silicification of crinoidal biosparites was rare.

vii) Precipitation of the equicrystalline mosaic calcite cement in the rest of the intergranular pore spaces and the rest of the skeletal moulds (stage 2 of cementation). In the case of the crinoidal biosparites, the development of the syntaxial calcite overgrowths was completed in this stage.

viii) Late diagenetic pressure-solution led to formation of stylolites.

ix) Diagenetic dolomitization, represented by coarsely crystalline dolomite rhombs replaced skeletal fragments as well as the cement.

x) Tectonic fracturing.

xi) Cementation of the tectonic fractures by coarsely equicrystalline ferroan and non-ferroan calcite cement (stage 4 of cementation).

4. OOLITIC LIMESTONES

i) Micritization of the skeletal fragments.

ii) Replacement of the aragonite of the oolites by calcite. The aragonitic cores of the skeletal fragments were also dissolved, leaving empty moulds supported by micrite envelopes.

iii) Early diagenetic deformation was responsible for the collapse of some micrite envelopes and spalling-off of the outer shells of some oolites. Crumbly fractures were formed at the same time. The presence of fibrous drusy calcite cement on the outer and inner surfaces of the spalled-off shell is clear evidence that early diagenetic deformation pre-dated the first stage of cementation.

iv) Development of the fibrous drusy calcite cement fringing the inner surfaces of the moulds and the intergranular pore spaces. Syntaxial calcite overgrowths were also developed on the echinoderm fragments (stage 1 of cementation). The rate of growth of the syntaxial calcite was faster than that of the fibrous drusy calcite. This is manifested by the local absence of fibrous drusy calcite crystals where syntaxial calcite overgrowths reach the surface of an oolite. Fibrous drusy calcite crystals completely fringe the intergranular pore surfaces in places where there are no echinoderm fragments.

v) Incipient dolomitization represented by finely crystalline dolomite rhombs selectively replaced the micrite in oolite shells and micrite envelopes.

vi) Partial silicification represented by the development of secondary quartz overgrowths on detrital quartz grains present as nuclei in some oolites.

vii) Precipitation of the equicrystalline calcite cement in the rest of the intergranular pore spaces and the skeletal moulds. In most cases this type of cement is non-ferroan calcite, but in a few rocks it is ferroan calcite.

viii) Diagenetic dolomitization represented by the occurrence of coarsely crystalline dolomite.

ix) Tectonic fracturing.

x) Cementation of the tectonic fractures by equicrystalline mosaic calcite crystals.

5. DOLOMITES

a. Finely crystalline dolomite (dolomicrite):

i) Penecontemporaneous complete dolomitization of formerly deposited aragonitic micrite. This resulted in the formation of the dolomicrite.

ii) Lithification of dolomicrite was completed by the interlocking of the finely crystalline dolomite rhombohedra during their growth.

iii) Late diagenetic pressure solution affected the rock and resulted in the formation of stylolites.

iv) Tectonic fracturing.

v) Cementation of the tectonic fractures by the development of equicrystalline mosaic calcite cement (stage 4 of cementation.

b. Coarsely crystalline or diagenetic dolomite:

Because these are secondary rocks formed by the complete diagenetic dolomitisation of former limestones, their diagenetic histories were influenced by the variations in the lithology of the original rocks.

Petrographic studies of dolomites have revealed that coarse-grained diagenetic dolomites were formed by complete dolomitization of biomicrites, biosparites and oosparites. The general texture of the dolomites as well as the allochem ghosts displayed in them can be used for the identification of the original rock types.

The pre-dolomitization history of a dolomite formed from a biosparite rock will have consisted of the first eight stages in the diagenetic history of the normal biosparites. The post-dolomitization history consists generally of four further stages:

i) Cementation of the intercrystalline pore spaces in the dolomite by equicrystalline mosaic cement that consists of ferroan dolomite, non-ferroan and ferroan calcite.

ii) Tectonic fracturing.

iii) Cementation of the tectonic fractures by equicrystalline mosaic cement consists of ferroan and non-ferroan dolomite and ferroan and non-ferroan calcite.

iv) Dedolomitization of the subaerially exposed surfaces of the dolomite.

DIAGENESIS OF THE NON-CARBONATE ROCKS

The non-carbonate rocks of the Carboniferous Limestone Series are represented mainly by three rock types, namely: shale in the Lower Limestone Shale Group; calcareous sandstone in the Main Limestone Group of the northern outcrops of the South Wales Coalfield; and the Drybrook quartzarenite of the Forest of Dean. In the following paragraphs, the diagenesis of the two main types of sandstones will be discussed. The diagenesis of each rock type will be dealt with separately and the diagenetic processes will be arranged in chronological order.

DIAGENESIS OF THE CALCAREOUS SANDSTONES

1. Precipitation of Carbonate Cement:

Calcareous sandstones consist mainly of detrital quartz grains and a little skeletal debris cemented by carbonate minerals. Two types of calcareous sandstones have been recognised, namely; dolomite-cemented sandstones and calcite-cemented sandstones. In the calcite-cemented sandstones the cement may be finely crystalline or coarsely crystalline. In the rocks cemented by coarsely crystalline calcite, it was found that large calcite crystals fill one or more pore spaces. Poikilotopic texture is developed where two or three quartz grains are enclosed within a single calcite crystal.

In the dolomite-cemented sandstones, each pore space is filled with one or two coarse dolomite crystals. Poikilotopic texture formed by enclosing more than two quartz grains in a single dolomite crystal is frequent. Dolomite is usually present as ferroan dolomite.

2. Replacement of detrital quartz grains by carbonate cement:

This process involves partial replacement of detrital quartz grains by the carbonate cement. In the calcite-cemented sandstones, partial replacement of quartz grains by calcite is very common. The detrital components display open packing, but on close inspection the outlines of formerly existing grains can be recognised as ghosts within the calcite mosaic. In the dolomite-cemented sandstones, replacement of the detrital quartz grains by ferroan dolomite is commonly displayed.

3. Grain fracturing:

Fracturing of the quartz grains in the calcareous sandstones is a common phenomenon. Fractures are usually closed and of irregular shape. It is suggested that this type of fracturing may be due to pressure exerted on the quartz grains by the force of crystallization of the carbonate cement.

4. Pressure-solution:

This process is usually manifest by the presence of the stylolites. These were rarely observed in the calcareous sandstones.

5. Tectonic fracturing:

Tectonic fractures are commonly present especially in the western outcrops of the South Wales Coalfield. They are cemented by coarsely crystalline calcite cement with coarse mosaic texture.

DIAGENESIS OF THE DRYBROOK QUARTZARENITES

The diagenetic modifications found in this sandstone could be attributed mainly to chemical processes. The only chemical process which has played a major role was the chemical precipitation of pore-filling minerals.

This involved the precipitation of minerals either as partial or complete filling of pore spaces, without modification of the adjoining framework minerals. Three minerals have participated in this process: quartz, kaolinite and iron oxide.

1. Developments of authigenic quartz:

Authigenic quartz was precipitated in two forms: as secondary overgrowths on detrital quartz grains, and as microcrystalline quartz. Secondary quartz overgrowths on the detrital quartz grains are the most common form of authigenic quartz in the upper unit of the Drybrook quartzarenite. They occur as thin rims of clear quartz in lattice continuity with the host quartz grains. Occasionally, they completely fill the pore spaces. The recognition of quartz overgrowths is greatly facilitated by the presence of discontinuous rims of iron oxide along the original detrital grain boundaries.

Microcrystalline quartz was found to be the most common form of authigenic quartz in the lower unit of the Drybrook quartzarenite. It is composed of equant microcrystals of quartz and is present as pore fillings.

2. Developments of authigenic kaolinite:

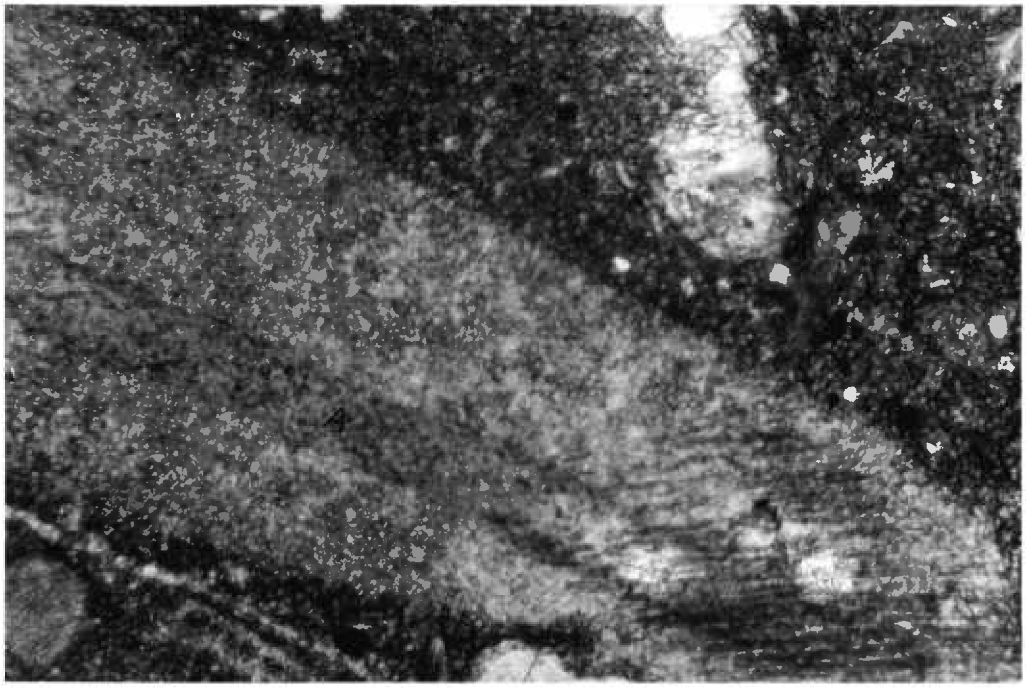
One of the prominent characteristics of the Drybrook sandstones is the abundance of kaolinite as pore-filling cement. Pores are usually lined with a thin rim of authigenic quartz and the rest of the pore is filled with kaolinite. Kaolinite is present as coarsely crystalline vermicular aggregates. The absence of any sort of minerals which could alter to kaolinite (e.g. clay minerals or mica) and the presence of undisturbed vermicules, suggested authigenic origin of kaolinite. It is most probable that kaolinite was chemically precipitated from intergranular pore solutions rich in silica and aluminium in an acidic environment.

3. Precipitation of iron oxide:

The frequent amount of iron oxide present in the Drybrook sandstones, especially in the upper unit, is responsible for their red colouration. Iron oxide, which is most probably haematite, is present as minute spicules, usually coating the detrital quartz grains and partially or completely filling some of the pore spaces. Sometimes it is mixed with the vermicular kaolinite aggregates in the pore spaces.

PLATE 8

Figs. 1 & 2 Biomicrite. Note micrite infilling skeletal pores of an echinoderm fragment, (A) unmicritized core of the echinoderm fragment. Sample BY37.



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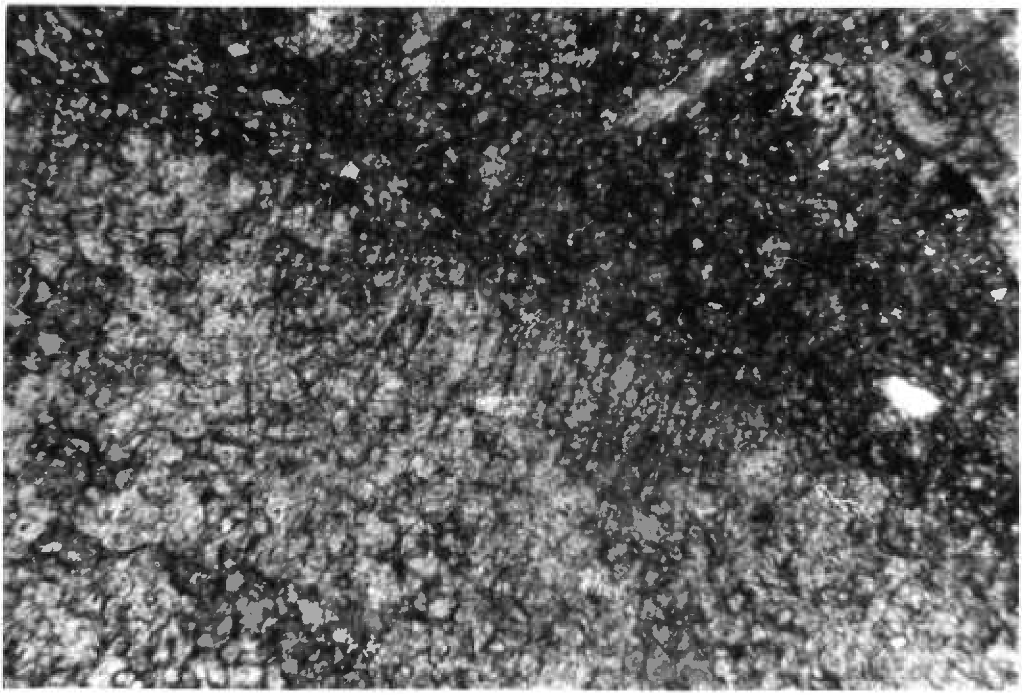

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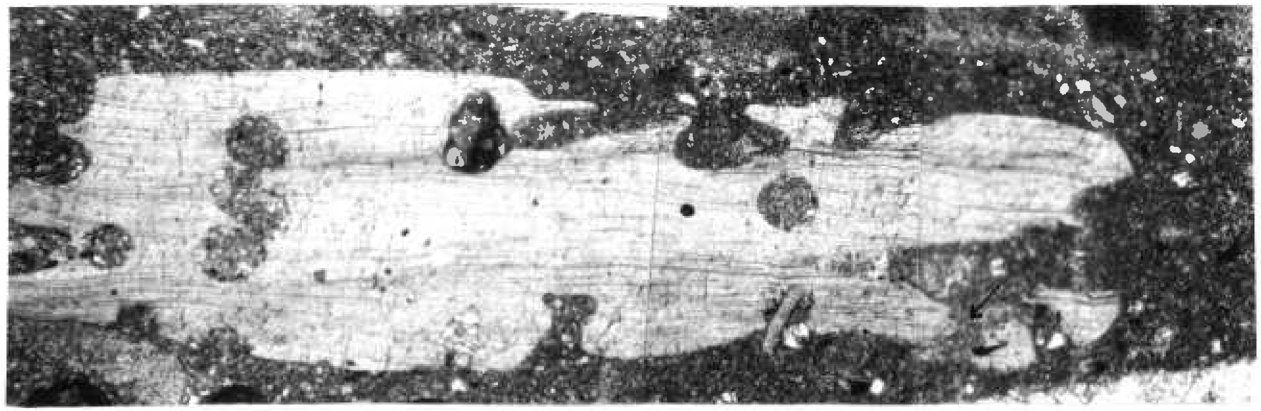
PLATE 9

Fig. 1 Algal boring in a brachiopod shell fragment. Borings filled with micrite and fine-grained skeletal debris. Note biofragmentation (arrow). Brachiopod biomicrite, Sample BY37.

Figs. 2 & 3 A brachiopod shell fragment encrusted by algal micrite exhibiting algal borings. Algal micrite crust was affected by algal borings which are filled with sparry calcite (arrow). Algal biosparite, Sample 671.



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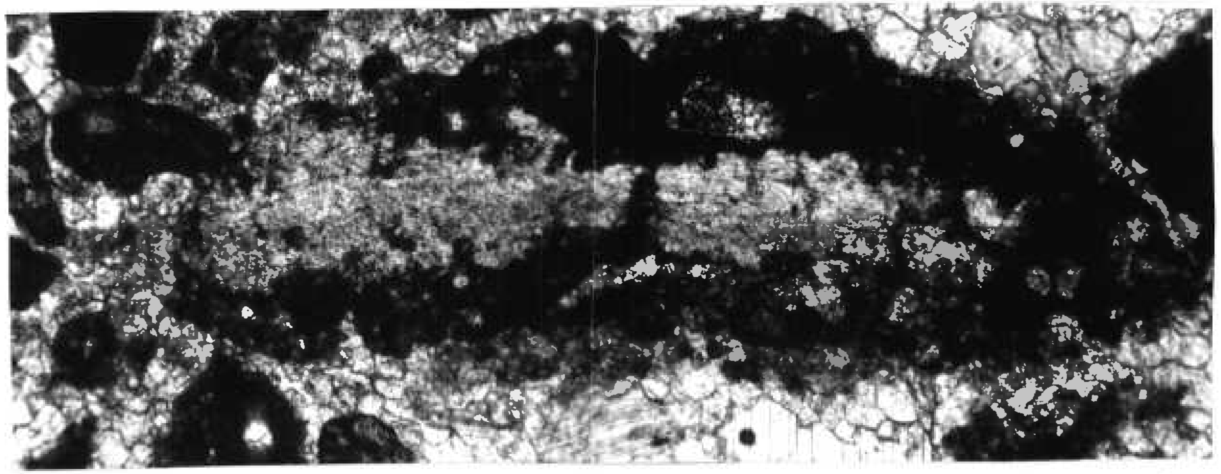

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PLATE 10

Fig. 1 Irregular fractures cutting across an echinoderm fragment. The sparry calcite filling the fractures is in optical continuity with that of the echinoderm fragment and its syntaxial overgrowth. Biomicrite, Sample C05.

Fig. 2 Spalled-off shell of an oolite, with fibrous drusy calcite cement (FC) covering both the outer and the inner surfaces of the spalled-off shell (arrows). Oosparite, Sample RU3.

Fig. 3 Stylolite in a biosparite. Note concentration of organic matter and finely crystalline dolomite rhombs (D) along the stylolite seam. Sample BY45.

Fig. 4 Concentration of organic matter in a stylolite seam. Dolomicrite, Sample BY65.

Fig. 5 Stylolitic contacts between oolites. Oosparite. Sample BY31.

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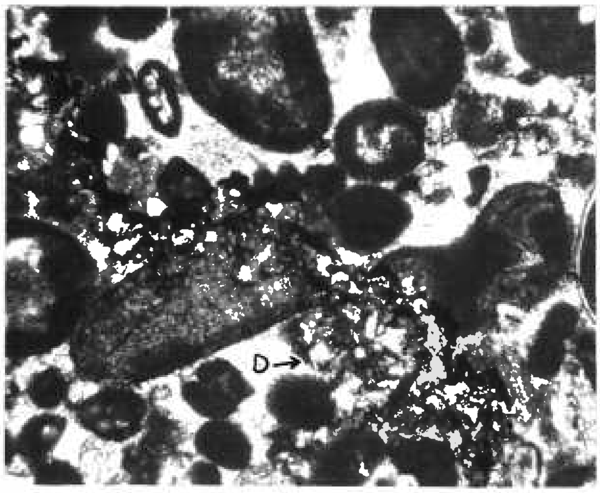

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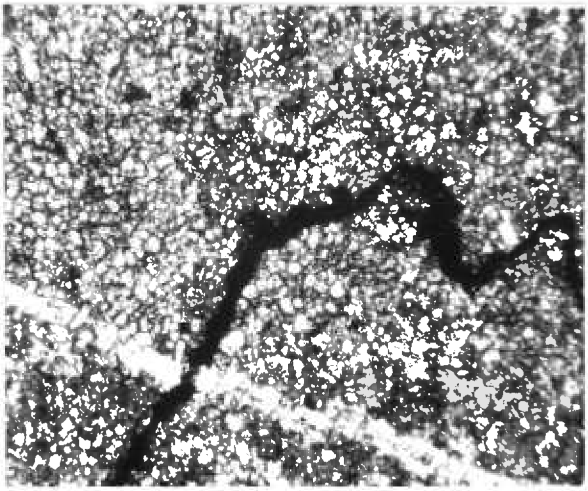

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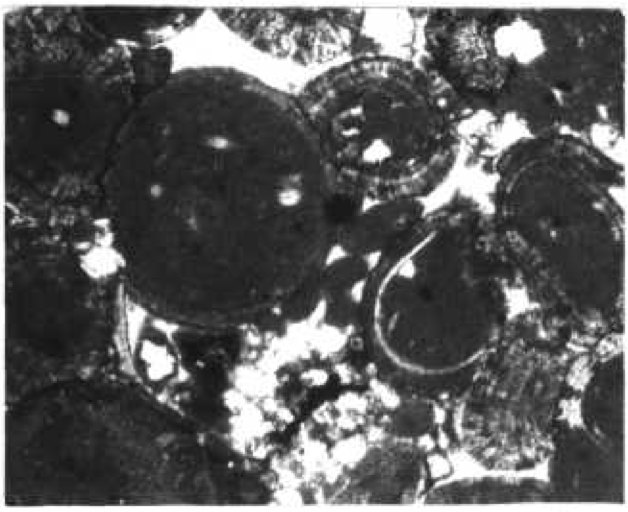

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PLATE 11

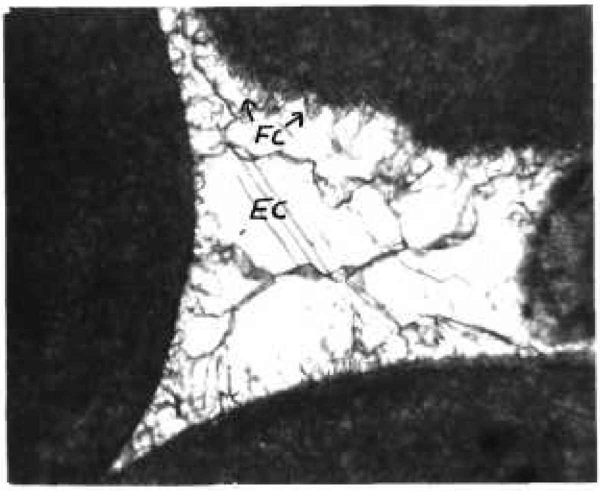
Fig. 1 Two phases of calcite cement fill in a pore space in an oosparite. Fibrous drusy calcite lines the pore space (FC), equicrystalline calcite mosaic (EC) fills the rest of the pore space. Sample 604.

Fig. 2 Off-centred oolites. The shells of the oolites were completely dissolved and their nuclei (crinoidal fragments) being unsupported settled down to the base of the cavities. The cavities were then filled with equicrystalline sparry calcite (EC). Oolitic limestone, Sample BY62.

Fig. 3 Radiaxial fibrous mosaic calcite cement (RC) developed on a brachiopod shell fragment. Idiotopic quartz (Iq) partially replaces both the shell fragment and the calcite cement. Sample 655.

Fig. 4 Coarsely equicrystalline ferroan calcite cement (FEC) lining cavity in dolomite. Sample 1043, stained with a mixture of alizarin red S and potassium ferricyanide.

Figs. 5 & 6 Radiaxial fibrous mosaic calcite cement (RC) developed on a brachiopod shell fragment. The calcite cement has grown in optical continuity with the prismatic calcite crystals forming the shell fragment. Fig.6 crossed polars. Sample 655.



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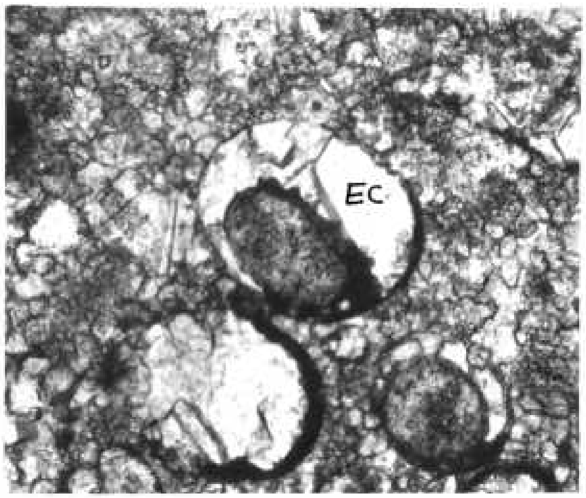

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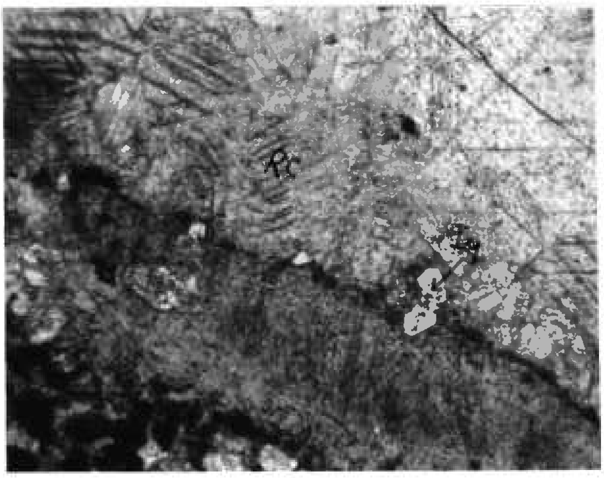

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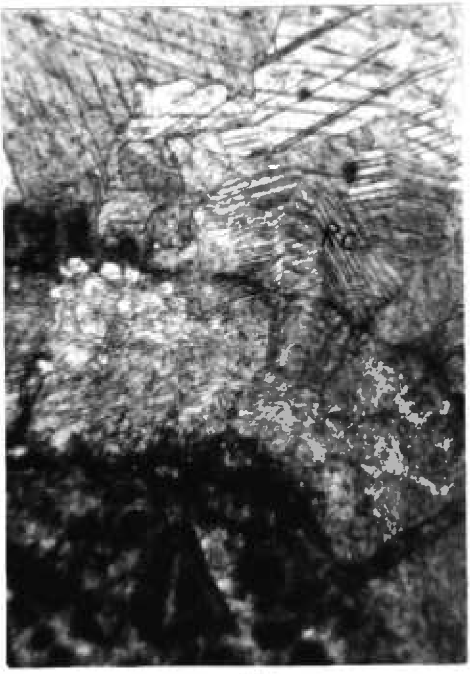

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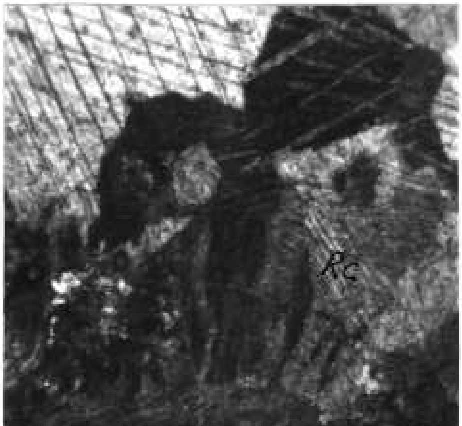
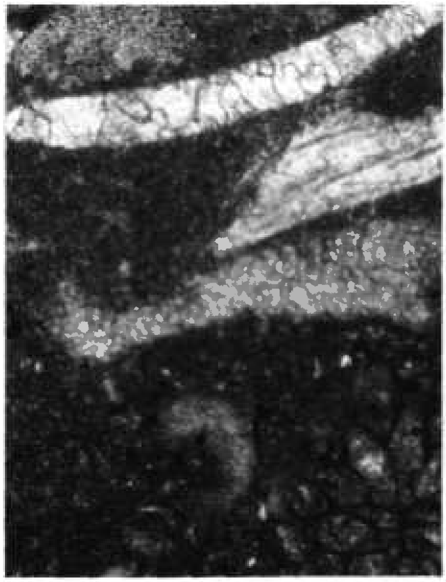


6 260 μ

PLATE 12

Fig. 1	Shell moulds, in biomicrite, filled with equicrystalline calcite mosaic. Sample BY37.
Fig. 2	Oolites partially cemented by finely crystalline fibrous calcite (FC) and by the syntaxial calcite overgrowth of an echinoderm fragment (Oc). Oosparite, Sample 604.
Fig. 3	Serrated compromise boundaries between adjacent syntaxial calcite overgrowths. Crinoidal biosparite, Sample 1015.
Fig. 4	Neomorphic syntaxial calcite overgrowth developed on an echinoderm fragment. Biomicrite, Sample BY44.
Fig. 5	Zoned syntaxial calcite overgrowth, with an inner zone of non-ferroan calcite and an outer zone of ferroan calcite. Sample 765, stained with a mixture of alizarin red S and potassium ferricyanide.



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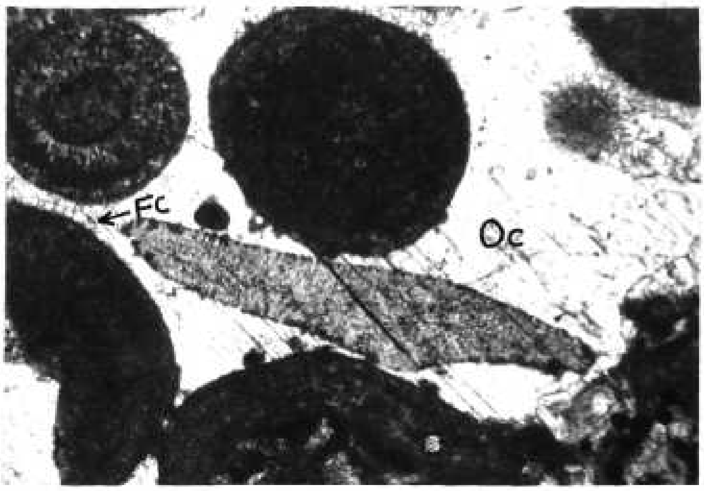

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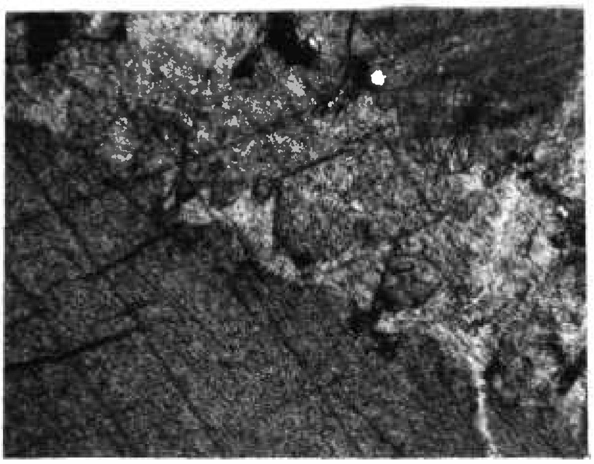

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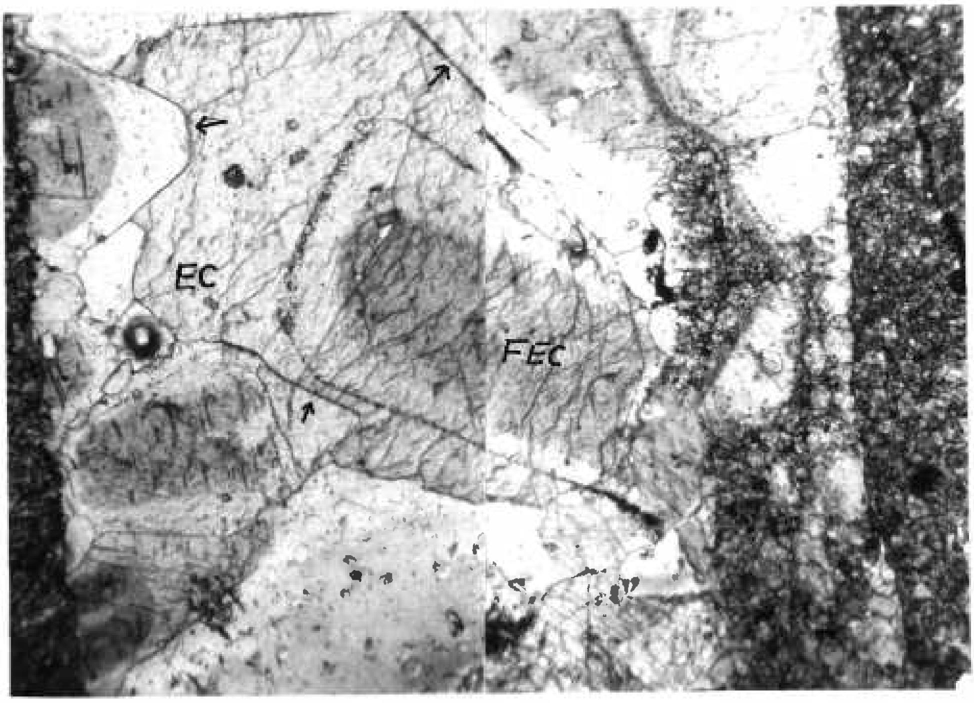
PLATE 13

Fig. 13 Zoned prismatic calcite cement filling a fracture in dolomite. The calcite prisms consist of a ferroan core (FEC) overlain by a non-ferroan zone (EC). Note the straight compromise boundaries between the adjacent crystals (arrows). Sample MC4, stained with a mixture of alizarin red S and potassium ferricyanide.

Fig. 2 A brachiopod body cavity filled with equicrystalline calcite mosaic. First phase shows a relatively finely equicrystalline non-ferroan calcite cement (EC) followed by a coarsely equicrystalline ferroan calcite cement (FEC). The latter phase shows plane intercrystalline boundaries and enfacial junctions (arrows). Sample 1042, stained with a mixture of alizarin red S and potassium ferricyanide.



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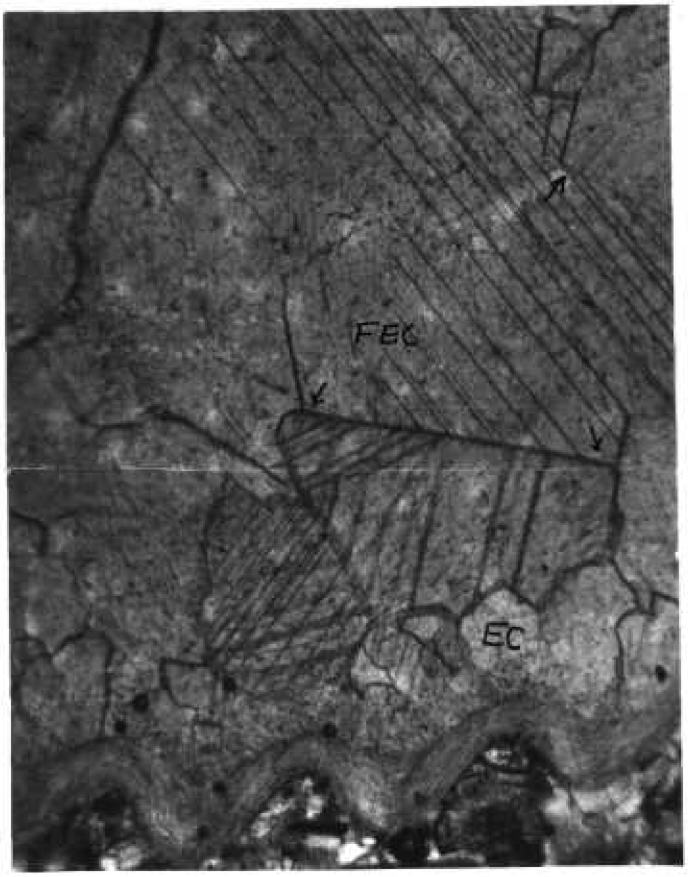

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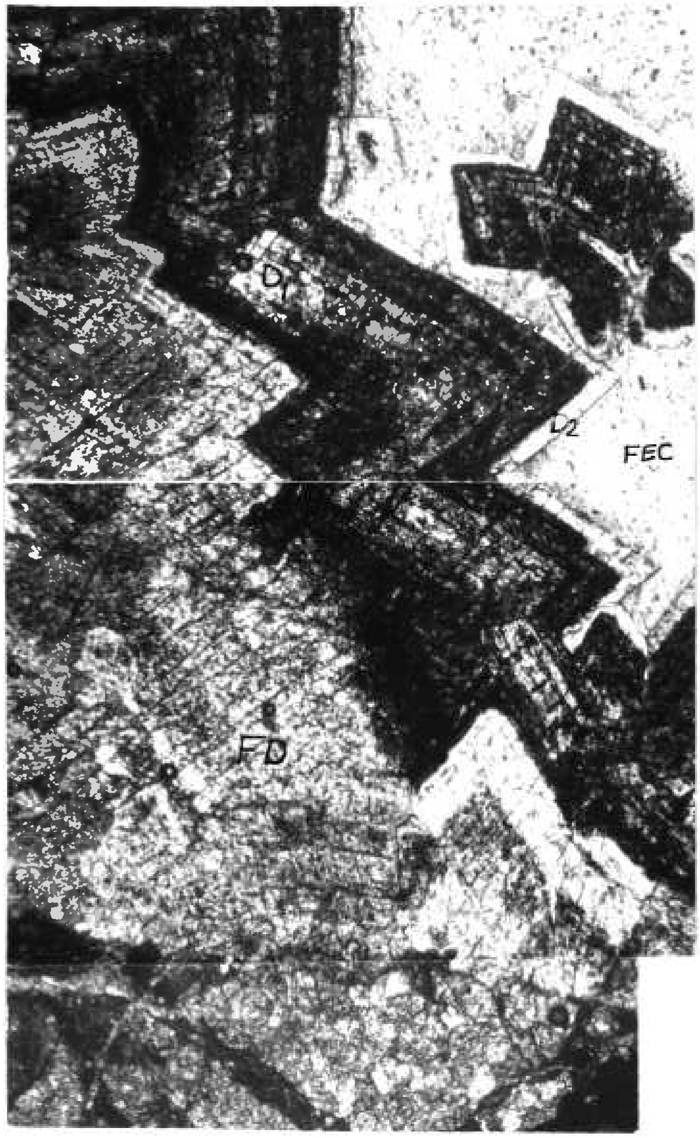
PLATE 14

Figs. 1 & 2 Fracture in dolomite filled with a composite cement of dolomite, calcite and barite. Dolomite cement is coarsely crystalline and zoned; it consists of an early phase of ferroan dolomite (FD) followed by zones of non-ferroan dolomite rich in iron oxide inclusions (D_1), and finally by a narrow zone of clear non-ferroan dolomite (D_2). The rest of the fracture is filled with clear coarsely equicrystalline ferroan calcite (FEC). Barite (B) is floating in the calcite cement. Sample MC4, stained with a mixture of alizarin red S and potassium ferricyanide.

Figs. 3 & 4 Coarsely crystalline ferroan calcite (FEC) filling cavities in dolomite. Note the development of clear non-ferroan dolomite (D_1) as an overgrowth on the ferroan dolomite crystals (D_2) that form the wall of the cavities. Samples 1038 and CWM1, stained with a mixture of alizarin red S and potassium ferricyanide.



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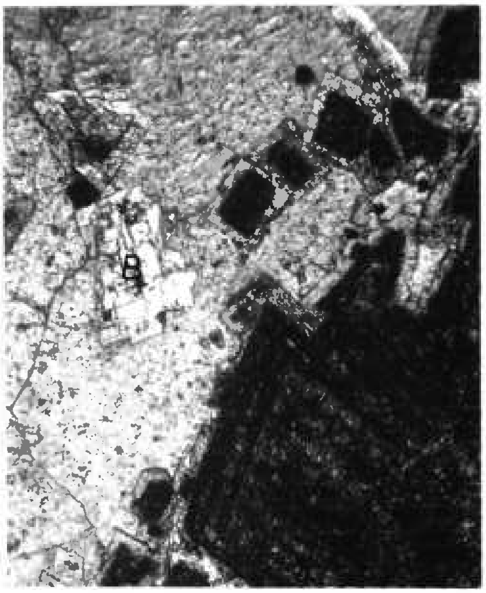

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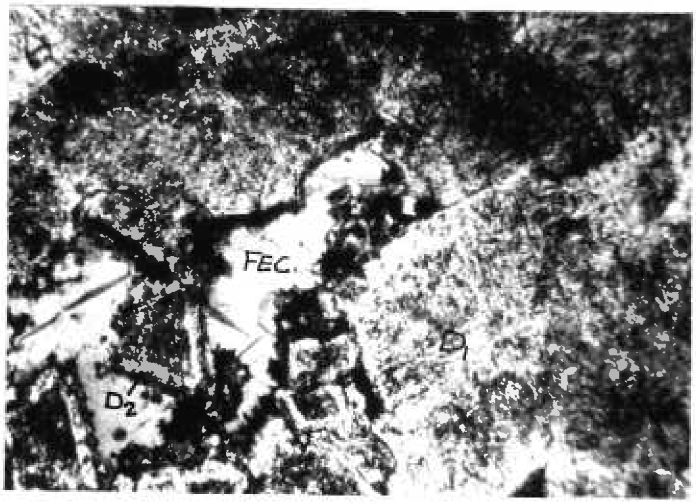

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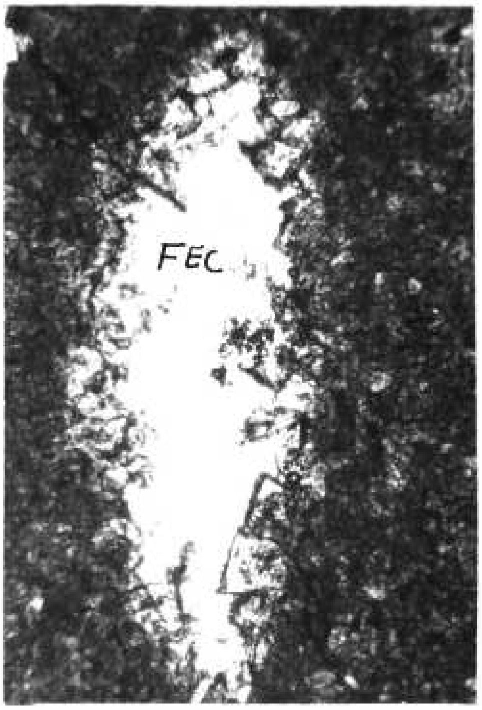

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PLATE 15

Fig. 1 Completely dolomitized crinoidal limestone. It shows a porphyrotopic texture with finely crystalline dolomite rhombs (d) floating in a large single dolomite crystal (D). Sample A5.

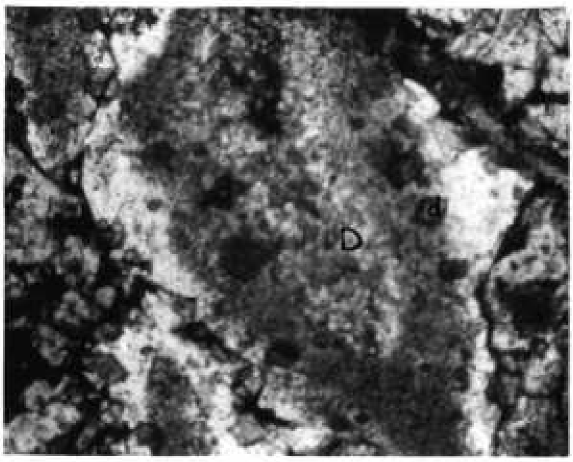
Fig. 2 Organic matter trapped in the intercrystalline pores. Granulotopic hypidiomorphic dolomite. Sample MC1.

Fig. 3 Completely dolomitized calcarenite. Sample A16.

Fig. 4 Partially dolomitized foraminiferal biomicrite. Sample 616.

Fig. 5 Partially dedolomitized zoned dolomite. The ferroan dolomite zones are replaced by calcite (C), while the non-ferroan dolomite zones are unaltered (D).

Fig. 6 Preferential dedolomitization of the ferroan dolomite zones in zoned dolomite. Dedolomitization did not affect the original dolomite fabrics. Dedolomite (C), non-ferroan dolomite (D). Sample SY5, stained with a mixture of alizarin red S and potassium ferricyanide.



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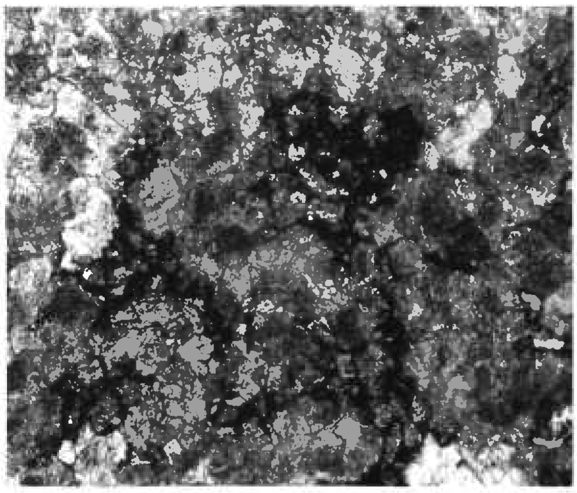

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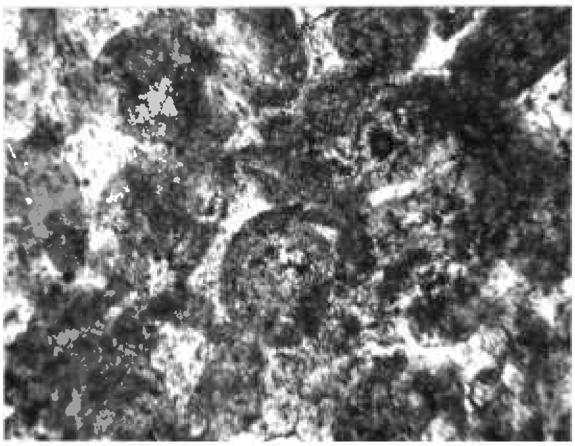

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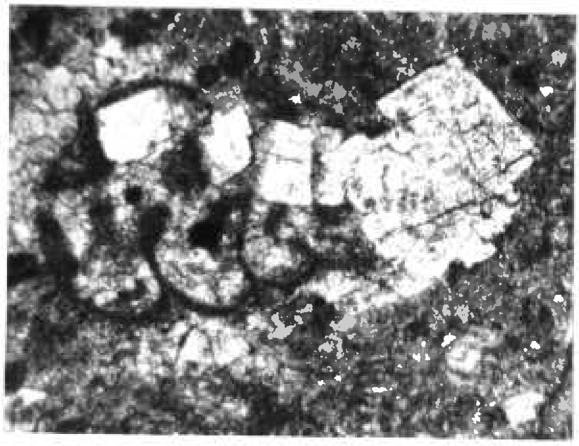

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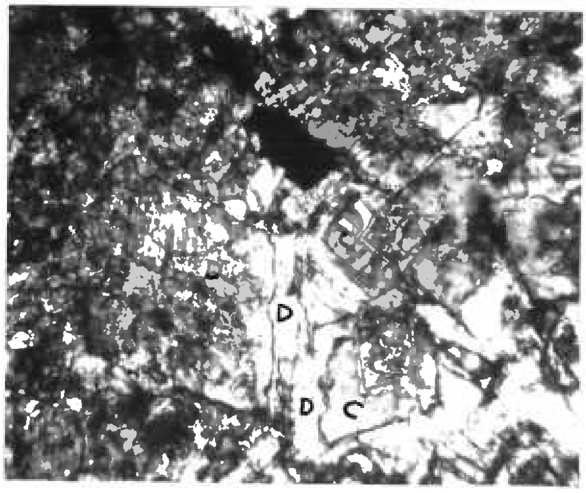

6 75 μ

PLATE 16

Fig. 1 Completely dedolomitized zoned dolomite rhombs. Sample 686. Stained with a mixture of alizarin red S and potassium ferricyanide.

Fig. 2 Dedolomite rhomb. The former dolomite rhomb has been replaced by calcite mosaic. Sample 1021B.

Figs. 3 & 4 Silicified, partially dolomitized micrite. The micrite is replaced by microcrystalline quartz (Mq), dolomite is replaced by calcite (C). Sample 648, stained with alizarin red S. Fig. 4 crossed polars.

Figs. 5 & 6 Completely silicified calcarenite. Sample 647.

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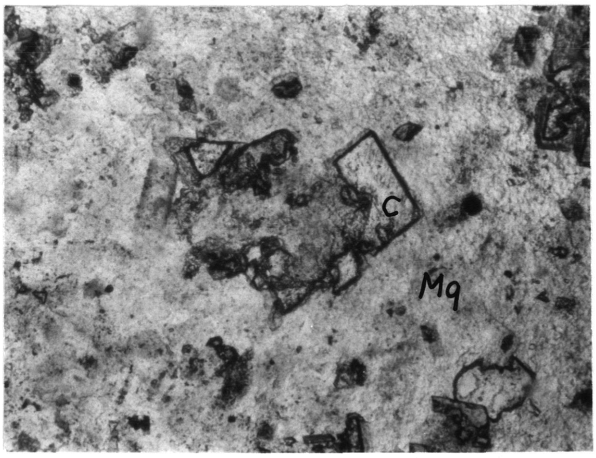

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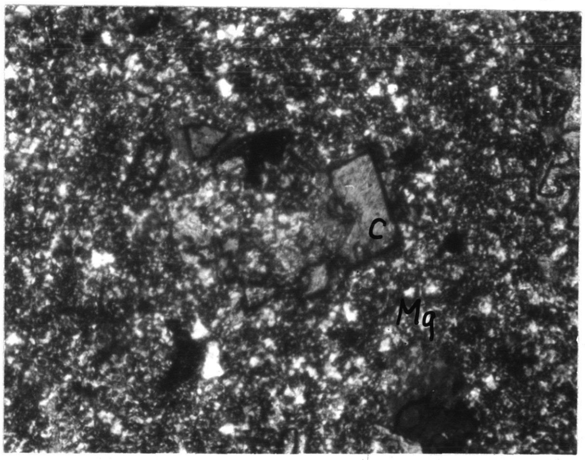

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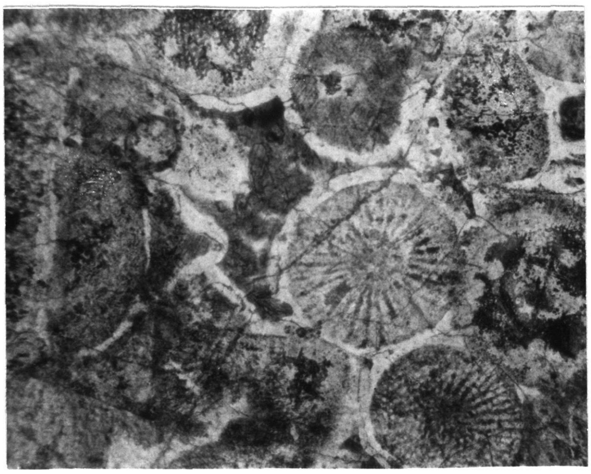

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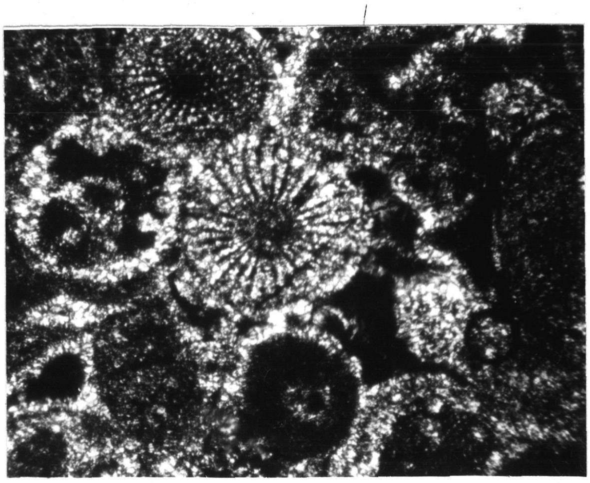

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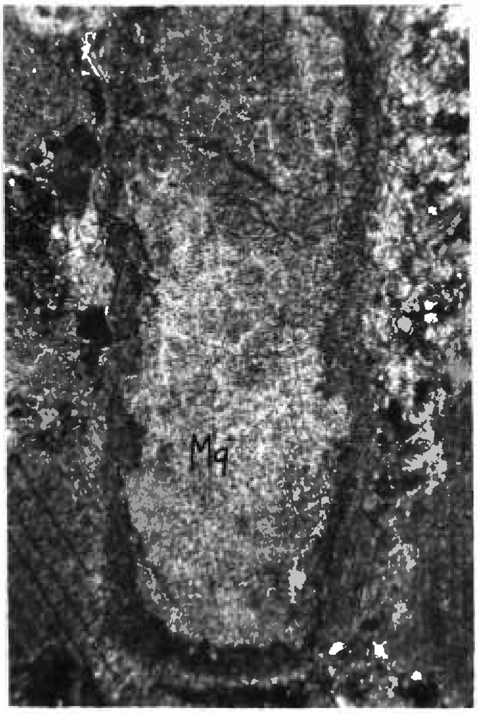
PLATE 17

Figs. 1 & 2 Silicified crinoid ossicle, the original calcite is replaced by microcrystalline quartz (Mq). Sample 1013.

Figs. 3 & 4 Authigenic idiomorphic quartz replacing oolite shells and sparry calcite cement. Note the ghost of the original detrital quartz nucleus (dq).

Fig. 5 Authigenic idiomorphic quartz crystals preferentially replace micrite in a thick algal crust. Sample Rul.

Fig. 6 Idiomorphic quartz crystal replacing calcite in a severely micritized brachiopod shell fragment. Sample Rul.



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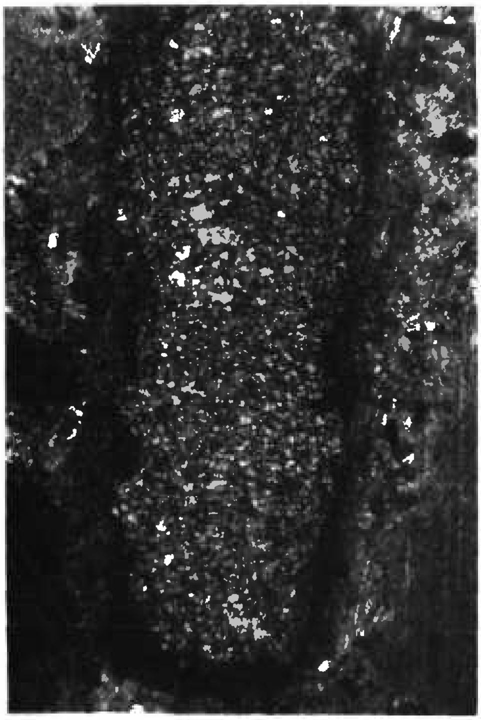

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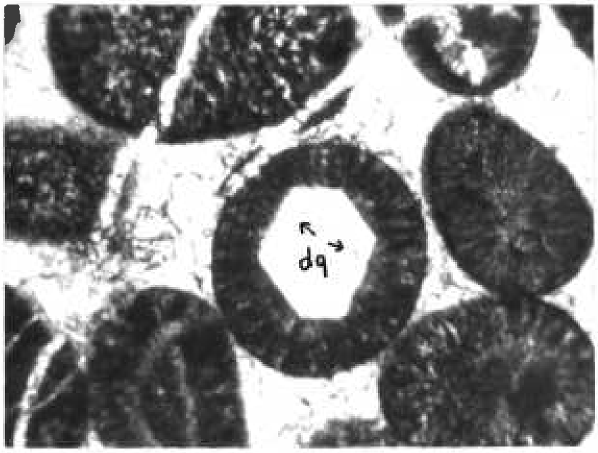

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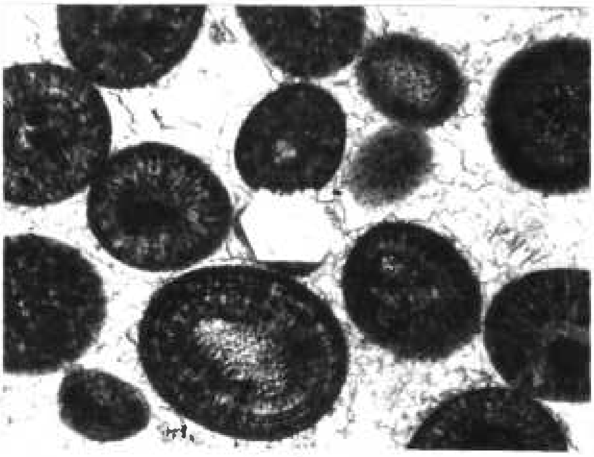

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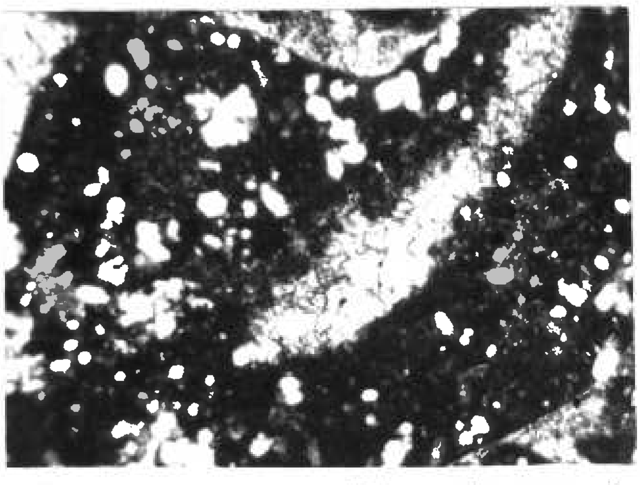

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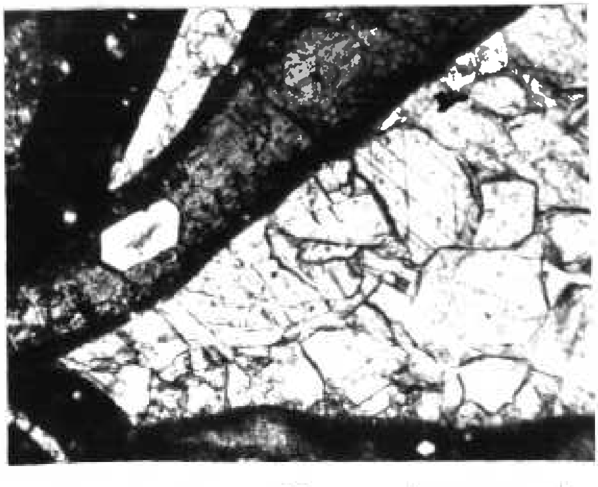

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CHAPTER IV

PETROGRAPHY OF THE MILLSTONE GRIT AND THE COAL MEASURES SERIES

PETROGRAPHY OF THE MILLSTONE GRIT AND THE COAL MEASURES SERIES

INTRODUCTION

Although much geological research has been done on the Coal Measures of South Wales, little work has been published on the petrography of these rocks. Heard (1922) studied the petrology of the Pennant Series, east of the River Taff in South Wales, with special emphasis on the heavy mineral constituents. Wilson (1965) studied the origin and the mineralogy of South Wales underclays. Jones (1972) studied in detail the petrology of the Trenchard sandstones in the Forest of Dean coalfield. He recognised two lithotypes, namely; quartzarenite and litharenite.

This Chapter deals with the petrography of the terrigenous rocks of the Millstone Grit and the Coal Measures of South Wales. The main objective of this study is to identify and describe the various lithotypes forming these rocks.

CLASSIFICATION

On the basis of the framework grain size, the following terrigenous rock types have been recognised: conglomerates, sandstones, siltstones and argillaceous rocks. Conglomerates are present mainly in the Basal Grit

Group of the Millstone Grit and are represented by the orthoquartzitic type.

The classification of the sandstones in the present work follows that used by Pettijohn, Potter and Siever (1972). This classification basically uses only framework grains of quartz, feldspars and rock fragments of sand size. As a secondary criterion the classification distinguishes between the "clean sands" or arenites (sandstones with less than 15 percent matrix), and the "dirty sands" or wackes (sandstones with more than 15 percent matrix).

Arenites are divided into three main classes, namely: quartzarenites, lithic arenites, and arkosic arenites. Quartzarenites are arenites with no more than five percent of either feldspars or rock fragments. Lithic arenites (litharenites) are those arenites with 25 or more percent of rock fragments and a minimal matrix content. It was found that lithic arenites in the rocks studied are mainly of the phyllarenite type. These are lithic arenites which consist mainly of pelitic rock particles, (Folk, 1968). Arkosic arenites are those arenites with 25 or more percent feldspar and lesser amounts of rock fragments. The term feldspathic arenite is used for any sandstone with five or more percent feldspars. As thus defined feldspathic sandstones include subarkoses,

arkoses, some litharenites and many graywackes. The transitional class sublitharenite has a lesser rock particles content, likewise the term subarkose is used for a transitional class of arenites with less feldspar than an arkose and with few or no rock fragments.

Wackes are divided into three main classes, namely: quartzwackes, feldspathic graywackes and lithic graywackes, depending on whether detrital quartz, detrital feldspars, or rock particles dominate the rock framework. Quartzwacke was found to be the dominant graywacke type in the rocks studied. 30 microns was used as the upper limit of the grain size of matrix particles. The origin of the matrix in graywackes and arkoses has been studied by Dickinson (1970). He designated the following types of interstitial material: (a) exotic cements like calcite or zeolites; (b) homogeneous, monomineralic phyllosilicate cement displaying textures indicative of pore filling; (c) clayey detrital lutum called protomatrix; (d) recrystallized lutum or protomatrix called orthomatrix with relicts detrital texture; (e) polymineralic, diagenetic pore-filling called epimatrix, whose growth is accompanied by alteration of framework grains; and (f) deformed and recrystallized lithic fragments called pseudomatrix. He described the criteria for the recognition of each type.

Siltstones are divided into two types, namely; quartzitic siltstones and clayey siltstones, in the present study. Quartzitic siltstones are those with less than 10 percent matrix, while clayey siltstones are those with more than 10 percent clayey matrix.

Classification of the argillaceous rocks in the present study is based mainly on their megascopic appearance. Three main types of argillaceous rocks have been recognised, namely; claystones, mudstones, and shales. Claystones are usually indurated and massive. They are commonly associated with coal beds and occur directly beneath the coal seams. They are generally slickensided and contain traces of plant roots. In the geological literature the words "fireclay", "seatearth" and "underclay" are used for naming this rock type. The term "underclay" is preferred since the term "fireclay" is related to the refractory nature of the clays and the term "seatearth" is not frequently used in geological literature. The majority of the underclays studied are claystones, but some contain considerable amounts of silt-size particles, and so they are silty claystones. The term "mudstone" is used for thin-bedded, indurated argillaceous rocks, while the term "shale" is applied to those with bedding cleavage or fissility approximately parallel to the bedding.

PETROGRAPHY OF THE MILLSTONE GRIT

The Millstone Grit Series in South Wales is divided into three lithological units, namely; Farewell Rock (Sandstones), Middle Shales (dark grey and black shales with sandstone bands) and Basal Grits (sandstones and conglomerates). The following is the detailed petrographic descriptions of the various lithotypes forming these units.

A. ORTHOQUARTZITIC CONGLOMERATES

These rocks generally form the lowest part of the Basal Grit Group and mark the unconformity between the Millstone Grit Series and the underlying formations.

Macroscopically, they are thick-bedded and occasionally show graded bedding. They are light in colour, very hard, and consist of well rounded quartzitic pebbles and cobbles cemented by siliceous and calcareous cement. Texturally, the framework grains are usually moderately sorted and range in size from granule to cobble; they have an intact framework.

Microscopically, they consist mainly of polycrystalline quartz pebbles with subordinate amounts of quartzitic rock pebbles and chert debris. The intergranular pore spaces are filled partially with a matrix of ill-sorted sands and clays, and by siliceous and calcareous cement.

Mineralogy:

Terrigenous detritus:

a. Quartz: Quartz forms about 85 percent of the rock, occurring as framework debris and as sandy and silty matrix. Quartz in the framework debris is represented by only polycrystalline variety. Each pebble consists generally of about 10 quartz crystals. The quartz crystals within the polycrystalline quartz pebbles are highly strained. Quartz grains in the matrix are separable into polycrystalline and monocrystalline. Generally quartz grains of fine-sand and silt-size are monocrystalline, while those of coarse-sand size are polycrystalline.

b. Chert: Chert is present in small amounts in most samples. Usually it occurs in the sandy matrix, but a few chert pebbles are present in the framework of the rock. It is pale brownish in colour and of coarsely crystalline type.

c. Rock fragments: Rock fragments represented by metaquartzite are present in small amounts. They occur as framework pebbles, and occasionally as sand grains in the matrix.

d. Detrital Clays: detrital clays are present in small amounts in the matrix, and they are of the illitic type.

Chemical Constituents:

a. Calcite: calcite is present as clear, coarsely equicrystalline crystals that partially fill the intergranular spaces. In the lower beds which covered the underlying Carboniferous limestones, calcite is very abundant and fills most of the intergranular pore spaces. Commonly it partially replaces the detrital quartz grains, especially those of the matrix.

b. Authigenic quartz is present as secondary overgrowths precipitated in optical continuity on the detrital quartz grains. It occurs in small amounts and more frequently is developed on the detrital quartz grains of the matrix rather than on those of the framework.

c. Authigenic Kaolinite presents as coarsely crystalline vermicular aggregate filling some of the pore spaces. Usually it is partially replacing the carbonate cement as well as the matrix material.

B. SANDSTONES

Two main sandstone lithotypes have been recognised, namely quartzarenite and lithic arenite. Table 4.1 summarizes their modal analysis. Fig. 4.1 shows the framework mineral composition and classification of these sandstones.

TABLE 4.1 MODAL ANALYSIS OF THE MILLSTONE GRIT SANDSTONES

	Lithiarenite			Quartzarenite								
	90	91	652	BY8	BY10	BY14	82	99	683	631	660	676
Quartz:-												
Monocrystalline	6.0	8.0	5.4	5.0	2.0	5.0	7.0	3.0	2.0	3.0	2.0	2.5
Polycrystalline	64.8	62.5	57.5	82.0	86.0	85.0	80.0	85.0	90.5	85.0	89.0	90.0
Total	70.8	70.5	62.9	87.0	88.0	90.0	87.0	88.0	92.5	88.0	91.0	92.5
Argillaceous R.F.	20.8	21.0	22.6	-	-	-	-	-	-	-	-	-
Chert	1.0	2.0	3.5	2.0	2.0	3.0	5.0	3.0	-	2.0	2.0	-
Feldspars	1.0	2.0	-	-	-	-	-	-	-	-	-	-
Muscovite	3.4	3.0	3.0	-	T	T	-	1.0	-	-	-	-
Biotite	2.0	-	3.0	-	-	-	-	-	-	-	-	-
Silica Overgrowths	1.0	1.0	-	3.0	5.0	7.0	8.0	8.0	7.5	7.0	6.0	7.5
Clays	-	-	5.0	-	-	-	-	-	-	-	-	-
Authigenic Kaolinite	-	-	-	8.0	5.0	-	-	-	-	2.0	1.0	-
Av. diameter in mm.	0.3	0.3	0.3	0.20	0.50	0.50	0.40	0.20	0.30	0.20	0.30	0.40

T = Traces

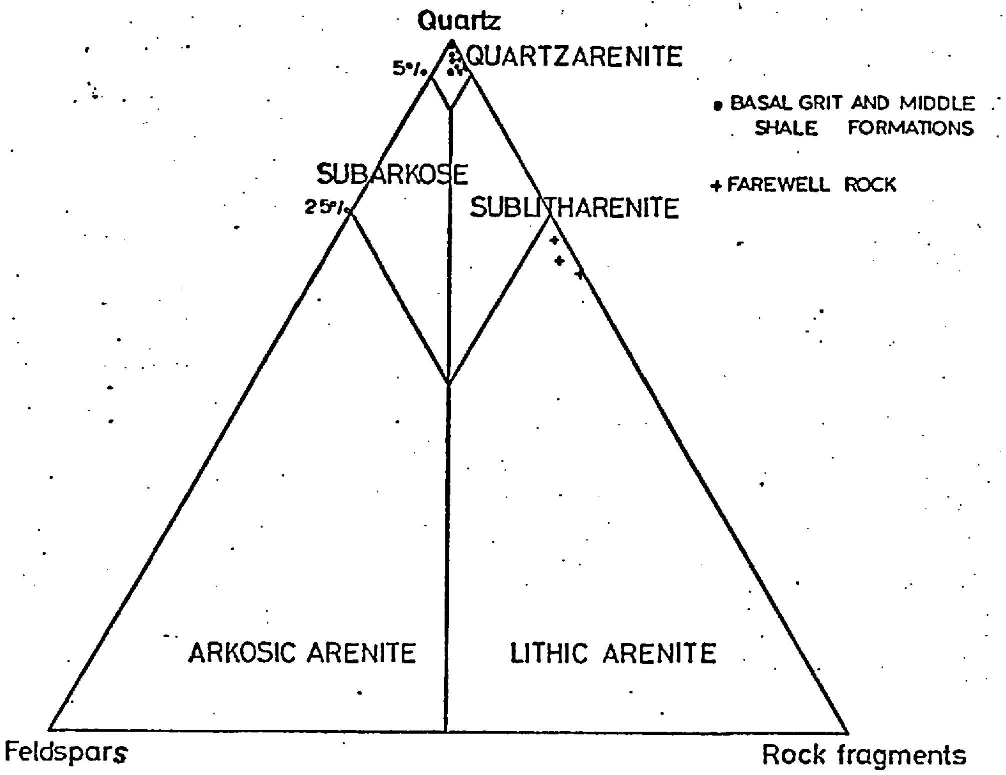


FIG.4.1 FRAMEWORK MINERAL COMPOSITION AND CLASSIFICATION OF THE MILLSTONE GRIT SANDSTONES.

B.1 QUARTZ ARENITES (Plate 18 , Fig. 1)

Most of the sandstones of the Basal Grit Group are of the quartzarenitic type. Quartzarenites are present also as thin beds in the Middle Shale Group.

In the field, they are thick-bedded and show cross-stratification. They are very hard and creamy and white in colour. Sparkling facets of the euhedral quartz overgrowth hide detrital outlines.

Microscopically, these rocks consist almost wholly of mono- and polycrystalline quartz grains cemented by authigenic quartz overgrowths. Small amounts of chert grains and authigenic kaolinite complete the rock constituents. Detrital quartz grains with their secondary quartz overgrowths commonly form about 90 percent of the rock. Quartz grains are commonly spherical and well rounded. They are usually of medium sand size, about 0.3 mm. in average diameter, and are well sorted. This lithotype is mineralogically and texturally mature.

Mineralogy:

Terrigenous Detritus:

a. Quartz is represented mainly by the monocrystalline type. Polycrystalline type is present in small amounts.

Quartz grains commonly form about 90 percent of the rock. Strained grains are frequent in the rocks from the western area of the Coalfield.

b. Chert is present in small amounts, about 2 percent of the rock. It is pale brownish in colour and of microcrystalline type.

Chemical Constituents:

(a) Authigenic quartz is present as secondary overgrowths deposited in optical continuity on the detrital quartz grains. Compromise boundaries between adjacent quartz overgrowths are common. Authigenic quartz crystals occur also as fracture fillings. They have been developed in such a way that their C-axes are normal to the fracture wall and they are usually in optical continuity with the detrital quartz grains forming the fracture wall.

(b) Authigenic kaolinite is present as coarsely crystalline vermicular aggregates filling some of the intergranular pore spaces. Commonly, authigenic kaolinite fills pore spaces which are partially filled with authigenic quartz overgrowths. In some samples it was found that this vermicular kaolinite is further cemented by authigenic quartz overgrowths.

B.2 LITHIC ARENITE (Plate 18 , Fig. 2)

The Farewell Rock unit is formed mainly of this sandstone lithotype. It is light grey in colour (salt and pepper appearance), and consists of well sorted, medium sand-size grains, with shaley and carbonaceous partings.

Microscopically, the rock consists mainly of quartz grains and rock fragments with subordinate amounts of feldspar grains and mica flakes. Squashed argillaceous rock fragments and mica flakes are bounding the other framework grains.

Because most grains tend to be equant, and the mica flakes and the argillaceous rock fragments are strongly deformed and squashed between the competent grains, a preferred shape orientation of the framework grains is not obvious. Grain size distribution is that of a moderately well sorted, medium grained sand (about 0.3 mm. in average diameter). The roundness and sphericity of the framework grains are dependent on their mineral composition. Usually, the quartz grains are ellipsoidal to spherical and rounded to sub-rounded. The original shape of the argillaceous rock fragments is often demolished due to their deformation but some of them still preserve their original shape, which is lensoidal with subrounded edges.

This lithotype is texturally mature, as indicated by sorting and roundness of the framework grains and by the absence of detrital clay matrix.

Mineralogy:

Terrigenous Detritus:

(a) Quartz is separable into mono- and polycrystalline grains. The majority of the quartz grains are monocrystalline, but polycrystalline quartz grains are present in small amounts. Several varieties of the monocrystalline quartz have been identified. These include strained monocrystalline quartz, unstrained monocrystalline quartz with inclusions, and unstrained clear monocrystalline quartz. Generally the monocrystalline quartz grains possess a thin rim of quartz overgrowths. Two varieties of polycrystalline quartz have been distinguished, namely; grains comprising two or three crystals, and grains containing numerous crystals of both equant and elongated shape. Commonly polycrystalline quartz is present as coarse grains.

(b) Feldspars: three varieties of feldspars have been recognised, namely; untwinned potash feldspars, perthite and plagioclase. The latter variety was found to be the most frequent, while the other types are represented by very few grains. Some feldspar grains show very slight alteration to kaolinite. There is no evidence that kaolinitization has taken place in situ. Coarse-grained feldspars are commonly fractured.

(c) Rock fragments: the following varieties of the rock fragments have been identified; slates, grey claystone, green claystone, chert and siltstone. Slates and claystone rock fragments are the most frequent. Slate fragments consist mainly of chlorite. The grey claystone fragments generally consist of illite shreds mixed with kaolinite. The green claystone fragments consist mainly of microcrystalline chlorite flakes.

(d) Mica is represented by three varieties, namely; muscovite, biotite and chlorite. They occur as shreds and flakes, usually bent, broken and squashed between the quartz grains. Muscovite shows alteration to kaolinite. Kaolinitization of the muscovite flakes usually takes place at the feathery edges and is intensive on the cleavage planes. Some muscovite flakes have been completely altered to kaolinite. Slight alteration of biotite and chlorite to ferrigenous material was observed.

Chemical Constituents:

(a) Authigenic quartz is present as secondary overgrowths on the detrital quartz grains as well as microcrystalline quartz. Secondary quartz overgrowths are commonly present as thin rims around some of the monocrystalline quartz grains. Microcrystalline quartz is less common and is partially filling some pore spaces.

(b) Authigenic chlorite is present as greenish minute flakes (about 10 microns in size) lining some of the pore spaces. It occurs also as vermicular aggregates filling some pore spaces.

(c) Authigenic kaolinite occurs as coarsely crystalline vermicular aggregates filling some pore spaces. The occurrence of muscovite remnants within the vermicular aggregates of kaolinite suggests that this kaolinite could have been produced by almost complete kaolinitization of former squashed muscovite flakes.

C. SILTSTONES

Siltstone is one of the most frequent lithotypes in the Millstone Grit Series. Generally it occurs as thin beds intercalated with the sandstone of the Basal Grit Group and with the shales of the Middle Shale Group. Two siltstone lithotypes have been identified, namely; quartzitic siltstone and clayey siltstone.

C.1 QUARTZITIC SILTSTONE

This type of siltstones was found to occur in the Middle Shale Group. These rocks are very hard and usually grey in colour. They commonly display various sedimentary structures, which can be classified into mechanical primary structures and organic structures. Mechanical primary structures are represented by horizontal stratification, microscale cross-stratification, ripple marks and convolute bedding. Organic structures are very frequent in these rocks and two types have been recognised, namely; thin bedding and borings. Organic thin bedding is displayed by alternation of thin siltstone laminae and thin carbonaceous bands which could have been laminae of algal mats. Borings are very common and are represented by a wide range of varieties. They are usually cut across several silt laminae and lined by carbonaceous matter.

Microscopically, the rock consists mainly of quartz grains, which commonly form about 60 percent of the rock, mica flakes with subordinate amounts of feldspars and argillaceous rock fragments.

Mica flakes are usually concentrated on the bedding planes, with organic matter and authigenic siderite. Mica flakes, other than those present on the bedding planes, are bent and squashed between the quartz grains. Most of the framework grains are angular to subangular and equant in shape. They are of about 40 microns in average size and are generally well sorted.

Mineralogy:

Terrigenous Detritus:

(a) Quartz is represented only by the monocrystalline type. The majority of the grains show undulose extinction.

(b) Mica is represented by muscovite, chlorite and biotite. Squashed muscovite flakes between the quartz grains are usually kaolinitized. In the case of partially kaolinitized muscovite flakes, kaolinitization is preferential to the cleavage planes and the shreddy edges of the flakes. The partially kaolinitized mica flakes appear as stripped flakes of alternating bands of high bireferengent mica and low bireferengent kaolinite. Partial sideritization of the muscovite flakes has been recognised. It is commonly preferential to the cleavage flakes.

(c) Feldspar is represented by a few plagioclase grains. They are commonly fresh.

(d) Rock fragments are represented only by a few grains of claystones. They are commonly deformed and squashed between the quartz grains and acted as pseudomatrix.

(e) Clay minerals are represented by micaceous clays filling some of the intergranular pore spaces. X-ray analysis of the $< 2\mu$ fraction revealed that the clay minerals present are: illite, kaolinite, chlorite and illite-montmorillonite mixed layer.

(f) Heavy minerals are represented mainly by zircon and tourmaline. They are usually concentrated in bands parallel to the bedding, and are also sporadically distributed in the rock. Zircon occurs as rounded and euhedral grains. Tourmaline is of the green variety.

Chemical Constituents:

(a) Authigenic quartz is represented mainly by microcrystalline quartz filling the intergranular pores. It is commonly found replacing the clay minerals.

(b) Authigenic siderite is present in two forms, namely; microcrystalline siderite forming micronodules concentrated on the bedding planes and associated with the organic matter, and finely crystalline rhombohedra of about 15 microns in size. The latter type usually occurred in the pore spaces partially replacing the clay matrix and the microcrystalline quartz cement.

C.2 CLAYEY SILTSTONE

This type of siltstone generally occurs in the Basal Grit Group, interbedded with quartzarenites and argillaceous rocks.

Clayey siltstones are present as thin beds of seatearth, and as thick beds intercalated with the argillaceous rocks. The clayey siltstone seatearth is usually very hard, dark grey in colour and with abundant randomly oriented plant rootlets. They do not show any type of sedimentary structures.

Microscopically, the rock is wacke siltstone and consists of monocrystalline quartz grains which commonly form about 50 percent of the rock, and mica flakes floating in organic matter-rich clay matrix. Organic matter is present as fine-grained debris and rootlet fragments.

Quartz grains are moderately to poorly sorted, subangular to subrounded, and of about 40 microns in average size.

Mineralogy:

Terrigenous Detritus:

(a) Quartz is represented only by the monocrystalline type, some grains have displayed slight undoluse extinction. Few chert grains have been identified.

(b) Mica is present as fine-grained muscovite flakes. They are usually undeformed. The majority of the muscovite flakes are partially or completely kaolinitized, and are commonly bulged.

(c) Clay minerals consist mainly of micaceous clays. They comprise about 35 percent of the rock. The original orientation of the micaceous clay flakes seems to be disturbed by the plant rootlets. They could have been pushed away from the horizontal position to an inclined position to give way to the growing rootlets. X-ray analysis of the $< 2\mu$ fraction of the rock revealed that the following clay minerals are present; illite-montmorillonite mixed layer, illite, chlorite and kaolinite.

(d) Heavy minerals are represented by a few grains of zircon and tourmaline. They are usually sporadically distributed in the clayey matrix. Zircon is present as rounded grains and tourmaline as green, well rounded grains.

Chemical Constituents:

These are represented mainly by authigenic siderite which usually occurs as microcrystalline siderite micronodules associated with the organic debris.

D. ARGILLACEOUS ROCKS

Argillaceous rocks form the main lithology of the Middle Shale Group and are frequent in the Basal Grit Group. Two main lithotypes have been recognised, namely, shales and claystones.

D.1 SHALES (Plate 18, Fig. 3)

These are the most frequent type of the argillaceous rocks in the Millstone Grit. They are black, fissile and fairly hard. Two types of shale are present, namely; non-marine shale and marine shale. Non-marine shale forms the main sequence of the Middle Shale Group and is usually lighter in colour than marine shale.

Microscopically, they consist wholly of micaceous clay minerals. Clays are usually dark brownish in colour due to their enrichment in organic matter. Organic matter is frequently occurring as fine-grained debris sporadically scattered throughout the rock and as thin fossilized algal mats lying parallel to the bedding. Marine shales are rich in framboidal pyrite, and brachiopod and ostracod shell fragments. Skeletal debris are intensively silicified. Silica replacing the calcitic shell debris is present as microcrystalline quartz and as spherulitic chalcedony.

Shell body cavities are commonly filled with vermicular kaolinite aggregates. Authigenic siderite is frequent and occurs as microcrystalline micronodules of ellipsoidal shape lying parallel to the bedding. They are usually associated with the organic matter debris.

Non-marine shales usually contain appreciable amounts of silt-size quartz grains and muscovite flakes. Muscovite flakes are commonly kaolinitized, and accordion-shaped flakes are produced. The non-marine shales of the Middle Shale Group of Gower area are usually found to contain appreciable amounts of detrital chlorite. X-ray analysis of the $< 2\mu$ fraction revealed that the clay minerals of these shales are; illite, illite-montmorillonite mixed layer, kaolinite and chlorite. Kaolinite was found to be more frequent in the non-marine shales than in marine shales.

D.2 CLAYSTONES

A thick sequence of about 70 ft. of claystones occurs at the bottom of the Basal Grit Group in the northern outcrops near Kidwelly (ALT-Y-GARN silica quarry). They are overlying the D zone of the Carboniferous Limestone Series and are overlain by the basal conglomerates of the Basal Grit Group.

These claystones are plastic, massive, some are thin bedded, and white to brown in colour. X-ray analysis of the $<2\mu$ fraction revealed that it consists mainly of illite and illite-montmorillonite mixed layer with small amounts of kaolinite.

PETROGRAPHY OF THE COAL MEASURES SERIES

The Coal Measures of South Wales consist almost wholly of terrigenous detritus derived from nearby sources and carried into a shallow subsiding trough by rivers from a landmass lying, as in Avonian and Namurian times, mainly to the north (see George, 1970). Over a great part of the coalfield the coal measures as a major lithological suite are dominated by shales in the lower part and by sandstones in the upper. The following are the generalised lithological characteristics of the three stratigraphical units of the coal measures.

3. Upper Coal Measures or Pennant Measures consist mainly of thick-bedded sandstones with subsequent intercalations of shales.

2. Middle Coal Measures consist mainly of argillaceous rocks with relatively few intercalations of sandstones.

1. Lower Coal Measures consist mainly of argillaceous rocks with thin intercalations of sandstone.

The detailed petrographic description of the various rock types forming the Coal Measures will be given below.

A. SANDSTONES

Three main sandstone lithotypes have been recognised, namely, lithic arenites, quartzarenites and quartz wackestones. Fig. 4.2 shows the framework mineral composition and classification of the Coal Measures arenites.

A.1 LITHIC ARENITES (Plate 20, Figs. 2, 3 & 4)

This rock is the main rock forming the Upper Coal Measures. It is known in the literature as the "Pennant Sandstone". These lithic arenites are thick-bedded and cross-stratified. The overall sedimentary structures of these rocks reveal that they are closely comparable to those laid down in modern alluvial plain environments.

In the field, they are grey in colour generally (salt and pepper appearance) in the fresh surface, and reddish-brown in the weathered surface. They are very hard but usually easy to break on the bedding planes. Thin streaks of carbonaceous and argillaceous matter are frequently found parallel to the bedding. Occasionally, they contain conglomerate bands of about 1 ft. thick, which sometimes reach up to 1 m. in thickness.

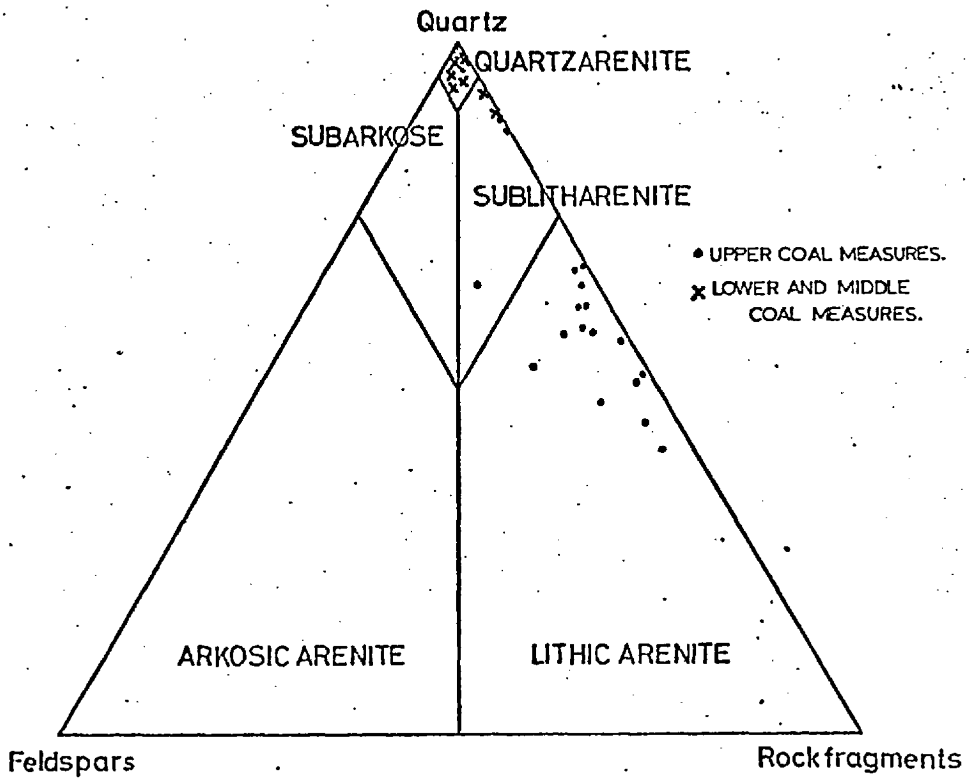


FIG.4.2 FRAMEWORK MINERAL COMPOSITION AND CLASSIFICATION OF THE COAL MEASURES SANDSTONES.

Microscopic description:

The rock consists mainly of quartz, rock fragments, feldspars and mica, and small amounts of authigenic minerals. Mono- and polycrystalline quartz are common, ranging in frequency from 41 to 75 percent of the rock. Rock fragments are represented by various rock types forming from 8 to 50 percent of the rock. Feldspars are usually frequent in the coarse-grained rocks and reach up to 15 percent of the rock. Mica reaches up to 8.8 percent of the rock. Table 4.2 summarises the modal analysis of the Coal Measures lithic arenites.

Textures:

Most of the rock is supported structurally by the quartz-feldspars-mica-rock fragment framework. Argillaceous rock fragments have been squeezed between the competent grains and intrude into the pore spaces to form a pseudomatrix. This pseudomatrix is responsible for binding the framework grains of the rock. The recognition of the pseudomatrix has been based mainly on criteria mentioned by Dickinson (1970). These are: (a) flame-like wisps of crushed lithic fragments extended into narrowing orifices between undeformed rigid grains; (b) the pseudofluidal internal fabric of lithic fragments deformed by pseudoplastic

TABLE 4.2 MODAL ANALYSIS OF THE LITHARENITES OF THE COAL MEASURES SERIES

	Bryn Amman						Morlias		Treforgan			Mardy		Tylorstown		Bedwelty		
	829'	832	834	866	778	821	747'	745	723	729	728	310	306	304	304C	560	585	574
Quartz																		
Monocrystalline	47.8	39.6	60.9	63.0	60.0	48.0	55.4	20.9	29.0	25.8	28.4	48.0	32.4	28.5	26.5	48.2	60.6	58.1
Polycrystalline	14.2	18.0	2.6	12.3	5.2	7.2	5.9	20.0	18.6	26.6	28.4	4.8	23.3	24.0	23.5	12.3	5.7	7.7
Total	62.0	57.6	63.5	75.3	65.2	55.2	61.3	40.9	47.6	52.4	56.8	52.8	55.7	52.5	50.1	60.5	66.3	65.8
Rock fragments																		
Argillite	19.7	19.0	21.9	16.2	7.8	31.0	19.8	22.8	17.6	18.2	20.4	18.4	16.4	25.2	23.6	13.8	-	17.4
Schists	6.1	4.0	3.6	1.3	0.3	2.0	3.6	7.8	6.7	4.0	4.7	7.2	4.5	1.6	3.0	12.65	-	-
Quartzite	5.7	11.1	3.6	-	-	10.6	7.7	20.9	16.8	2.7	3.8	1.6	17.3	19.0	17.6	3.4	4.0	-
Chert	1.7	1.2	-	-	-	1.1	-	2.6	2.5	7.5	4.2	8.0	2.7	1.0	1.2	5.0	4.4	7.7
Feldspars	3.8	4.8	T	T	-	-	1.6	3.4	7.8	14.2	7.3	3.2	1.0	0.7	1.7	2.3	13.3	T
Mica	1.0	2.0	3.1	4.5	T	T	4.4	-	T	1.0	T	8.8	2.4	T	1.0	1.3	-	-
Clays	T	-	T	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Dolomite	-	-	-	2.6	27.6	-	1.6	1.6	1.0	-	2.8	-	-	-	1.8	-	1.0	7.8
Siderite	-	-	4.1	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Av. Diameter in mms.	0.40	0.28	0.14	0.10	0.10	0.30	0.25	0.40	0.60	0.80	0.50	0.25	0.60	0.60	0.60	0.30	0.40	0.40

flow commonly conforms to the margins of confining rigid grains as concentric drape lines; (c) large "matrix" filled "gaps" in the framework suggest pseudomatrix, and the suggestion is strengthened where each "gap" filling is semi-homogeneous but texturally distinct from other "gaps". Muscovite flakes usually are squashed between the competent framework grains. They are also bent and cracked. Authigenic carbonate minerals are represented by ferroan dolomite and siderite and occupy some pore spaces and they are commonly replacing the argillaceous rock fragments.

According to the grain size distribution, two types of lithic arenites can be recognised, namely; (a) coarse-grained lithic arenite ranging in average size from 0.5 to 0.8 mm., usually rich in feldspars, and (b) medium-grained lithic arenite ranging in average size from 0.2 to 0.4 mm. Both types are usually moderately sorted to well sorted.

On the basis of the shape of the grains, the framework grains can be classified into three categories. These are: (a) equant grains (quartz, chert, quartzitic rock fragments and feldspar); (b) elongated bladed or lensoidal grains (argillaceous rock fragments and schist rock fragments); (c) flakey mica grains. Almost all of the grains are subrounded to subangular.

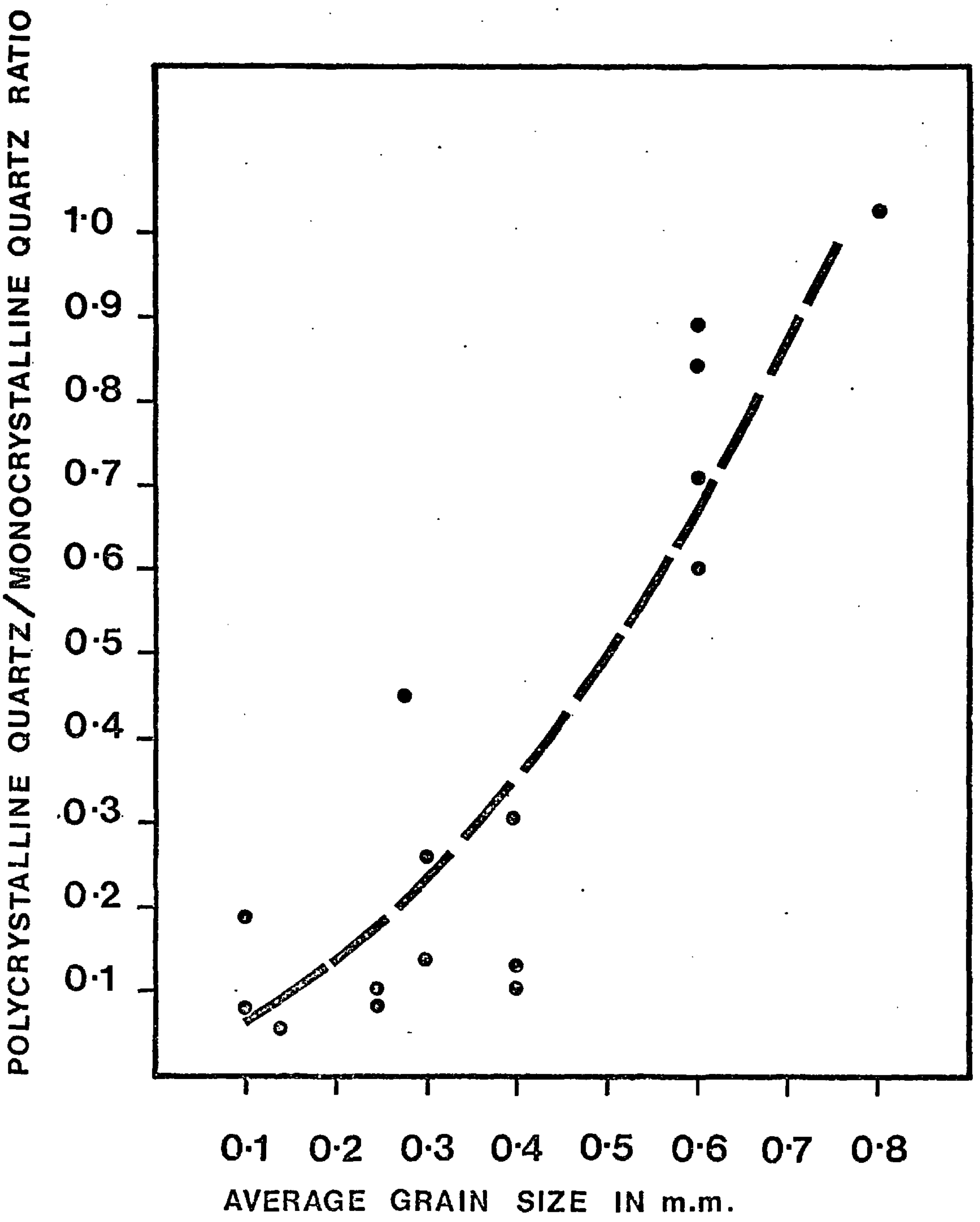
Mineralogy:

Terrigenous detritus:

(a) Quartz:

Quartz is separable into mono- and polycrystalline. Monocrystalline quartz grains are usually the most common. The following monocrystalline quartz varieties have been identified: (i) clear unstrained grains, (ii) inclusions rich unstrained quartz grains, (iii) inclusions rich strained quartz grains. The latter variety is the most frequent among the monocrystalline quartz grains. Polycrystalline quartz grains are represented by the following varieties: (i) grains consisting of equigranular equant small crystals, (ii) grains consisting of elongated and equant small crystals of various sizes, some grains showing biomodality of size, (iii) grains consisting of elongated stretched quartz crystals of metamorphic textures. The number of quartz crystals within a single polycrystalline quartz grains varies from three to about ten crystals. This depends on the size of the grain and the size of the crystals forming these grains. The polycrystalline quartz/monocrystalline quartz ratio was found to be generally related to the average diameter of the framework grains of the rock, Fig. 4.3. The ratio of polycrystalline to monocrystalline quartz grains is increased by increasing the average diameter of the framework grains of the rock.

Fig 4·3 The Relationship between Polycrystalline Quartz / Monocrystalline Quartz ratio and the average grain size in the Coal Measures Lithic arenite



(b) Rock fragments:

Four main types of rock fragments have been distinguished, namely; argillaceous, schists, quartzitic and chert.

(i) Argillaceous rock fragments

These are the most common type of rock fragment. They are generally grey and green in colour and they are the pepper in the pepper and salt mixture appearance of the rock. These rock fragments are composed of two main rock types, namely; mudstones and slates. Slate fragments are usually of greenish colour and consist mainly of micaceous clay minerals and finely crystalline chlorite flakes. They are thin laminated due to the presence of alternating bands of clay and chlorite flakes, and microcrystalline light-coloured quartz bands. The long diameter of the lensoidal grains is usually parallel to the laminations. They are slightly deformed and bent between the rigid quartz grains, but still preserve their original form and texture.

Mudstone fragments are the most abundant and they are grey and greenish in colour. They are commonly deformed and squashed between the competent framework grains. Some of these grains act as pseudomatrix. Some of the deformed mudstone fragments still preserve their original texture.

(ii) Schist fragments

They range in frequency from 0.3 to 12.6 percent of the rock. The following types have been recognised; muscovite schist, chlorite schist, quartz schist and kyanite schist. The latter type is represented only by a few grains. The most frequent type is the muscovite schists.

(iii) Quartzitic rock fragments

Two types of quartzitic rock fragment have been identified, namely; metamorphic quartzite and fine-grained quartz arenite. The metamorphic quartzite rock fragments consist of strongly deformed elongated quartz crystals of mosaic texture. The quartzarenite rock fragments are composed of rounded to subrounded fine-grained quartz grains cemented by secondary quartz overgrowths. Quartzarenite rock fragments are the most abundant.

(iv) Chert

This is generally the least frequent type of rock fragment. It reaches up to 8 percent of the rock in some specimens. It is separable into finely crystalline and coarsely crystalline chert. The latter type is the most frequent.

(c) Mica:

Mica is represented mainly by muscovite, and small amounts of biotite and chlorite. They are usually bent and broken, many are deformed between the quartz grains. The majority of the muscovite flakes are clear, but some contain fine grained reddish inclusions which are most probably rutile crystals. Some flakes are partially kaolinitized on their cleavage planes.

(d) Feldspars:

Three types have been identified; these are twinned plagioclase, untwinned feldspars and perthite. Twinned plagioclase grains are the most frequent among the other feldspars. Feldspar grains commonly show signs of alteration. Kaolinitization is the most frequent form of alteration, especially in the plagioclase grains. Kaolinitization is usually intensive along the cracks and the cleavage planes. Sericitization has also been recognised in some of the untwinned feldspar grains. Sericite usually present as fine grains displays the characteristic bireferengence. They are usually concentrated along cleavage planes. Feldspar grains are commonly mechanically deformed. Deformation is displayed by cracking, twisting and bending of the grains. Dolomite replacing feldspars along cracks and cleavage planes is common.

(e) Clay minerals:

Clay minerals as proto or orthomatrix (Dickinson, 1970) are almost absent. They are only present as the main mineral composition of the argillaceous rock fragments. In some clayey sublithic arenites, detrital clay matrix is present. It was difficult to determine their mineral composition because it is impossible to separate them from that forming argillaceous rock fragments, for the purposes of X-ray analysis.

(f) Heavy minerals:

Heavy minerals are present in small amounts. They are represented mainly by zircon, tourmaline and opaque minerals. Zircon occurs as both rounded or re-cycled grains as well as with euhedral crystals. They are usually rich in fibrous inclusions. Tourmaline is commonly represented by worn down green grains. In a polished section prepared for reflectivity measurements it was easy to identify the opaque mineral grains. They are represented by pyrite, chalcopyrite and magnetite. Magnetite grains are usually containing exsolved ilminite lamellae, and they are usually martitized along their octahedral cleavage planes.

Chemical Constituents:

(a) Iron carbonates, namely, siderite and ferroan-dolomite, are commonly present in small amounts. In some samples where the rock has been severely dolomitized, the ferroan-dolomite comprises up to 27.6 percent of the rock.

Ferroan-dolomite usually occurs as fine-grained rhombohedra partially filling some pore spaces. It is commonly found preferentially replacing argillaceous rock fragments and the other framework grains. The following are the framework grains arranged in descending order according to their susceptibility to ferroan dolomite replacement; argillaceous rock fragments, feldspars, mica and quartz grains.

Authigenic siderite is generally present in three forms; these are:-

i) as finely crystalline rhombohedra scattered in the pore spaces, and also replacing some argillaceous rock fragments.

ii) as microcrystalline micronodules concentrated on the bedding planes and which are usually associated with the plant debris.

iii) as finely crystalline mosaics filling cellular cavities of some plant debris.

In severely sideritized rocks, poikilotopic texture of remnant quartz grains floating in a single siderite crystal has been observed.

(b) Authigenic quartz is represented by overgrowths precipitated in optical continuity on some detrital quartz grains. Authigenic microcrystalline quartz is also present, filling some intergranular pore spaces.

(c) Authigenic kaolinite is present in small amounts. It usually occurs as vermicular aggregates filling fractures and some pore spaces.

A.2 QUARTZ ARENITES (Plate 19, Figs. 1, 2 & 3)

This lithotype is present mainly in the Middle and Lower Coal Measures and in the lower part of the Upper Coal Measures, usually as thin beds interbedded with the argillaceous rocks. In the field it varies in colour from grey to very light grey, and is extremely hard and massive.

Microscopic description:

It consists mainly of mono- and polycrystalline quartz commonly more than 75 percent of the rock. Mica and feldspars are present in small amounts. Authigenic quartz overgrowths on the detrital quartz grains is the main cementing agent of the rock. Authigenic kaolinite is commonly present as pore filling. Detrital clays are present, but generally less than 15 percent of the rock. Table 4.3 summarises the modal analysis of some of the studied quartz arenite samples.

TABLE 4.3 MODAL ANALYSIS OF THE QUARTZARENITES OF THE COAL MEASURES SERIES

-	121	141	149	179	181	187	207	214	224-2	244	295-5	354	355	569	760	778	810	827	858	870	911
Quartz																					
Monocrystalline	81.8	77.1	73.2	74.0	69.3	75.9	73.0	72.7	59.6	78.7	76.5	81.3	76.3	80.6	80.0	71.0	74.8	45.0	65.7	43.0	77.0
Polycrystalline	7.7	7.8	-	11.2	3.6	13.2	1.8	3.6	3.2	10.2	6.0	5.7	5.4	10.2	2.0	-	3.4	-	6.0	-	2.3
Total	89.5	84.9	73.2	85.2	72.9	89.1	74.8	76.3	62.8	88.9	82.5	87.0	81.7	90.8	82.0	71.0	78.2	45.0	71.7	43.0	79.3
Rock fragments																					
Argillite	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Schists	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Quartzite	-	-	-	-	1.1	-	-	0.7	-	-	-	-	-	-	-	-	-	-	-	-	-
Chert	-	-	-	3.7	1.4	-	1.3	1.0	-	-	-	4.0	5.5	-	3.0	-	-	-	-	-	-
Feldspars	-	-	-	-	-	-	-	-	-	-	-	0.8	-	-	-	-	-	-	-	-	-
Mica	-	0.6	-	-	T	-	1.0	T	-	-	1.0	0.5	0.5	-	2.0	0.7	2.1	-	-	-	1.1
Clays	-	1.3	8.4	-	14.6	-	14.6	10.9	5.8	-	5.0	-	-	-	4.0	-	-	-	11.6	-	-
Dolomite	-	-	19.4	T	1.8	T	2.9	2.9	31.4	1.0	7.0	1.4	4.0	4.0	-	11.2	1.7	55.0	-	57.0	5.8
Siderite	-	1.2	-	-	3.6	T	T	4.3	-	1.3	1.0	T	T	-	-	-	0.8	-	-	-	2.3
Authigenic Quartz	3.5	3.6	-	7.4	3.6	6.6	5.4	3.6	-	6.9	3.5	1.7	4.0	5.2	7.0	-	-	-	-	-	7.0
Kaolinite	7.0	8.4	-	3.7	1.0	4.3	T	-	-	1.9	-	4.6	4.3	-	2.0	17.1	17.2	-	5.7	-	4.5
Av. Diameter in mms.	0.30	0.15	0.12	0.20	0.08	0.20	0.08	0.08	0.14	0.20	0.14	0.17	0.15	0.30	0.15	0.15	0.16	0.07	0.10	0.07	0.25

T = Traces

Textures:

The fabric is typical of the mosaic quartz arenite. The rock is supported structurally by a quartz framework cemented by authigenic quartz overgrowths. It was difficult in most cases to recognise the original shape of the detrital grains. Mica flakes are usually bent, buckled, and squashed between the quartz grains. The majority of the samples are well sorted and of fine to very fine sand-size.

Mineralogy:

Terrigenous detritus:

(a) Quartz: It is separable into mono- and polycrystalline quartz. Monocrystalline quartz is the most frequent type, while the polycrystalline quartz is less frequent and ranges in frequency from 0 to 13 percent of the rock. Monocrystalline quartz grains are commonly strained and rich in inclusions. Polycrystalline quartz grains comprise two or three equant quartz crystals.

(b) Mica: is represented by muscovite, biotite and chlorite and ranges in frequency from 0 to about 5 percent of the rock. Muscovite is relatively the most frequent while the other two varieties are rare. Mica flakes are usually bent, cracked and squashed between the quartz grains, and partially altered to kaolinite. Kaolinitization is preferentially found to occur on the cleavage planes and the frayed ends of the muscovite flakes. Some of the severely squashed mica flakes, present as aggregates of minute shreds blocking some intergranular pore spaces, are almost completely kaolinitized. In the dolomitized rocks, it was found that muscovite flakes are partially replaced by ferroan dolomite, and replacement took place on the mica cleavage planes. The force of crystallization of the ferroan-dolomite on the cleavage planes of the mica was responsible for the disruption of some muscovite flakes.

(c) Feldspars: they are present as traces in some samples, represented mainly by plagioclase. It is commonly fractures and partially replaced by dolomite.

(d) Clay minerals are present in small amounts. It was difficult to identify the composition of the detrital clays because of the occurrence of authigenic clays. X-ray analysis of the $< 2\mu$ fraction of both detrital and authigenic clays revealed that they consist of illite, illite-montmorillonite mixed layers, kaolinite and chlorite.

(e) Heavy minerals are rare, and are represented by zircon and tourmaline. Zircon is present as well rounded grains. Tourmaline is represented by rounded green grains.

Chemical constituents:

(a) Authigenic quartz: it is present in three forms, namely; secondary overgrowths on the detrital quartz grains, microcrystalline quartz, and spherulitic fibrous chalcedony. Authigenic quartz overgrowths is the most frequent type and is the main cement mineral. Straight compromise boundaries between the authigenic quartz overgrowths have been noticed. Occasionally, authigenic quartz overgrowths contain kaolinite vermicules. Microcrystalline authigenic quartz is present in small amounts filling partially or completely some pore spaces. Usually, it replaces clay matrix. Spherulitic chalcedony is rare and occurs as pore-filling cement.

(b) Authigenic clays are represented mainly by kaolinite. It is present as coarsely crystalline vermicular aggregates filling pore spaces which were lined by authigenic quartz overgrowths. In many cases, the kaolinite vermicules are cemented by authigenic quartz and so they appear as if they are floating in the silica cement. It is also present filling some fractures. Authigenic vermicular chlorite was observed lining some pore spaces.

(c) Iron carbonates are siderite and ferroan dolomite. Both types are generally present in small amounts, except in some quartzarenite thin beds, where the rock is severely replaced by carbonates.

Ferroan dolomite occurs as finely crystalline rhombohedra starting to develop in the pore spaces. It replaces detrital clays, authigenic clays and authigenic quartz. With increased dolomitization, ferroan dolomite starts to replace the quartz grains. Quartz grains appear to be freely floating in the ferroan dolomite cement, developing poikilotopic texture. Ferroan dolomite rhombohedra commonly occur as rims surrounding the detrital quartz grains and floating in the authigenic quartz overgrowths.

Siderite is present as single rhombohedra and as clusters of microcrystalline siderite. Commonly it is associated with the organic debris. In the severely sideritized rocks the same fabric textures have been developed as in the dolomitized rocks.

(d) Authigenic pyrite has been recognised in quartzarenites of the anthracite area. It occurs as well euhedral cubes commonly corroding the quartz grains. It is also present in the form of thin veinlets filling narrow fractures.

A.3 QUARTZ WACKESTONE (Plate 20, Fig. 1)

This is the dominant sandstone type in the Lower Coal Measures. It occurs as thin beds intercalated with the argillaceous rocks. The rock is greyish in colour, commonly hard, massive with frequent amounts of organic debris.

Microscopic description:

The rock consists mainly of mono- and polycrystalline quartz and subordinate amounts of silica and feldspars. The framework grains are bounded by clay matrix which forms more than 15 percent of the rock. Ill-sorted carbonaceous debris is frequent. Authigenic minerals are represented mainly by siderite, dolomite and kaolinite. Table 4.4 shows the modal analysis of some quartzwacke samples.

Texture:

The rock is supported structurally by quartz grains bounded by clay matrix. The majority of the samples have intact framework, but a few have open framework structures. Commonly, the framework grains are angular to subangular and of fine to very fine sand-size.

Mineralogy:

Terrigenous detritus:

(a) Quartz is separable into mono- and polycrystalline quartz. Monocrystalline quartz grains vary in frequency from 47.8 to 77.8 percent of the rock, while the polycrystalline grains range in frequency from 0 to 15.6 percent of the rock. Monocrystalline quartz is present as strained and unstrained quartz grains. Polycrystalline quartz grains comprise two or three equant quartz crystals.

TABLE 4.4 MODAL ANALYSIS OF THE QUARTZ WACKES OF THE COAL MEASURES SERIES

	166	170	180	184	242	258	262	388-2	388-3	568'	719	740	748	758	760B	761	818	819	821'
Quartz																			
Monocrystalline	60.7	47.8	51.7	60.7	71.3	50.2	76.9	56.9	54.1	77.8	73.3	53.3	69.5	65.0	66.6	75.4	59.0	55.5	58.8
Polycrystalline	3.5	15.6	12.0	9.2	2.2	5.9	1.5	13.0	4.8	-	-	1.8	3.3	3.2	4.1	5.0	3.8	5.2	3.8
Total	64.2	73.4	63.7	69.9	73.5	56.1	78.4	69.9	58.9	77.8	73.3	55.1	72.8	68.2	70.7	80.4	62.8	60.7	62.6
Rock fragments																			
Argillite	-	-	-	-	-	-	-	7.3	5.5	-	-	1.3	2.4	4.8	-	-	7.2	8.3	7.1
Schist	-	-	-	-	-	-	-	-	-	-	-	-	2.0	-	-	-	-	-	-
Quartzite	-	-	-	-	-	-	-	1.6	3.0	-	-	-	2.0	-	-	-	-	-	-
Chert	2.2	1.7	1.3	-	-	-	-	-	-	-	-	-	1.0	-	-	-	-	-	-
Feldspars	-	-	-	-	-	T	-	-	-	T	-	T	-	-	-	-	-	-	-
Mica	T	T	T	1.4	-	-	-	1.6	3.0	-	-	1.3	T	T	T	1.0	5.5	15.5	5.4
Clays	23.7	21.8	22.8	18.8	25.0	25.4	15.4	14.6	14.2	22.2	20.0	26.7	14.8	20.5	25.0	14.7	16.9	15.5	13.9
Dolomite	7.8	T	3.4	2.8	1.5	2.4	6.2	1.0	1.8	-	-	4.5	T	-	-	-	4.2	-	8.0
Siderite	T	T	4.3	-	-	11.7	-	4.0	10.1	T	6.7	4.4	-	T	-	-	3.4	-	3.0
Authigenic Quartz	2.1	8.7	4.5	7.1	-	3.4	-	-	8.5	T	-	6.7	5.7	6.5	4.3	3.9	-	-	-
Kaolinite	-	4.4	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Av. Diameter in mms.	0.17	0.30	0.12	0.10	0.12	0.12	0.10	0.14	0.14	0.10	0.08	0.08	0.15	0.14	0.12	0.15	0.11	0.10	0.12

T = Traces

(b) Mica is represented mainly by muscovite with subsequent amounts of biotite and chlorite. Mica flakes are slightly deformed, bent and strained. Most of the muscovite grains are severely kaolinitized, numerous muscovite flakes are completely kaolinitized. Kaolinitized muscovite flakes are commonly of two shapes; namely, barrel-shaped flakes and fan-shaped flakes. These two forms are produced by intensive kaolinitization along the cleavage planes in the centre of the flake or on the frayed extremeties of the flakes, respectively.

(c) Feldspars are represented by albite and perthite and are present as traces in some samples. Most of the grains are severely altered to kaolinite or sericite.

(d) Rock fragments are identified in some samples and they reach up to 10 percent of the sand. They are mainly represented by grey and green mudstone fragments. In some rocks, cherts and quartzitic rock fragments have been identified.

(e) Heavy minerals are present in traces and are represented mainly by zircon and tourmaline. Zircon occurs as well rounded grains and tourmaline as rounded green grains.

(f) Clay minerals; they reach up to 26.7 percent of the rock. X-ray analysis of the $< 2\mu$ fraction revealed that clay minerals are represented by the following species; illite, illite-montmorillonite, kaolinite and chlorite mainly.

Chemical constituents:

(a) Authigenic quartz is represented mainly by microcrystalline quartz which is present as pore-filling cement. They commonly replace the clay matrix.

(b) Authigenic kaolinite has been identified in some samples. It is present as well crystalline vermicular aggregates filling some pore spaces.

(c) Authigenic carbonate minerals represented by ferroan dolomite and siderite have been recognised. Siderite is the most frequent carbonate mineral in the rock. Both minerals occur as fine rhombs sporadically scattered in the clay matrix. Siderite commonly is present as microcrystalline aggregates concentrated near and around the carbonaceous fragments.

B. SILTSTONES

Siltstones are abundant in the Lower and Middle Coal Measures. They are hard and vary in colour from white to dark grey. The following sedimentary structures have been identified:

1. Horizontal stratification: This is very common and is displayed by alternation of very thin laminae of carbonaceous clay rich in iron carbonates and relatively thick, light-coloured siltstone laminae.

2. Small cross-stratification: This is of macro- and micro-scale. It consists of alternating sets of microcross laminae composed of carbonaceous clay rich in brown siderite micronodules and light-coloured siltstone laminae.

3. Current lineations: This is represented mainly by ripple marks on the mudstone surfaces which are now preserved as casts on the underside of the siltstone laminae. These markings are usually of about one to 3 mm. in depth and a few centimetres in length.

4. Biogenic structures are very common in the siltstone, especially in the thin laminae intercalated with the mudstone. It is represented mainly by boring and burrows. Borings are very frequent and are present in various shapes. They range in diameter from 1 mm. to about 5 mm. Usually they are lined with carbonaceous matter.

On the basis of the clay content of the rock, two types of siltstone have been recognised, namely, quartzitic siltstone and clayey siltstone.

B.1 QUARTZITIC SILTSTONE (Plate 21, Fig. 1)

Quartzitic siltstones are the most frequent type of siltstone especially in the silt laminae intercalated with the mudstone and in the relatively thick, pure siltstone beds.

It is usually cross-bedded, very light in colour and very hard. Some samples are relatively very rich in mica flakes which usually lie parallel to the bedding planes and it gives a shiny lustre to the rock especially with surface parallel to the bedding planes.

Microscopic description:

The rock consists mainly of monocrystalline quartz grains and mica flakes with subordinate amounts of feldspars and cherts. Intergranular pore spaces are filled with clay matrix. The rock contains considerable amounts of siderite and carbonaceous debris.

Textures:

The rock is supported structurally by quartz and muscovite framework, bound by detrital clay matrix and authigenic quartz. Because most grains tend to be equant, a preferred shape orientation is not obvious but orientation of some of the detrital mica flakes is excellent and parallel to the bedding. Mica flakes mixed with the quartz grains are commonly bent, cracked and squashed. Grain size distribution is that of a moderately well sorted and of coarse to medium silt-size. Most of the quartz grains are subrounded to subangular.

Mineralogy:

Terrigenous detritus:

(a) Quartz: monocrystalline quartz is the most dominant type; polycrystalline quartz is represented by very few grains. Unstrained and strained quartz has been recognised.

(b) Mica is the second most frequent detrital grain. It is represented by muscovite, biotite and chlorite. Biotite is the least frequent, while muscovite is the most frequent. Some muscovite flakes are replaced by siderite which initially crystallized out along cleavage planes. Other muscovite flakes are altered to kaolinite along their cleavage planes. Biotite flakes are usually altered to brownish matter. Chlorite is present as the main mica mineral in some samples.

(c) Feldspars are present in small amounts. Two varieties were identified, namely, plagioclase and perthite. Most of the grains display alteration to illitic clay along cleavage planes.

(d) Rock fragments are rare and are represented mainly by mudstone fragments which consist almost entirely of illitic clay.

(e) Heavy minerals have been recognised in most of the studied samples. They are present as scattered grains or concentrated in thin bands parallel to the bedding. The following heavy minerals have been identified; tourmaline, zircon, sphene and iron ore minerals. Zircon is the most frequent and is present as rounded grains and euhedral crystals. The former type is most common. Tourmaline is present as rounded blue and green grains.

(f) Clay minerals: The majority of the clay minerals present are of the micaceous type. X-ray analysis of the $<2\mu$ fraction revealed that clay matrix consists of the following minerals; illite, illite-montmorillonite mixed layer, kaolinite and chlorite. Generally the first two minerals are the most important while the other two are present in subordinate amounts.

Chemical constituents:

(a) Authigenic quartz is present in two forms, namely; overgrowths around detrital quartz grains and microcrystalline quartz aggregates. Volumetrically the quartz overgrowths are the most important pore-filling cement. Microcrystalline quartz is present as pore-filling cement and commonly replaces the clay matrix.

(b) Authigenic siderite is the most frequent orthochemical mineral in most of the siltstones. It is generally associated with the carbonaceous debris. Siderite is present as very finely crystalline rhombohedra floating in the clay matrix. Authigenic ferroan dolomite is present in considerable amounts in some siltstones. It commonly occurs as small rhombohedra filling pore spaces and replacing both clay matrix and microcrystalline quartz cement. Some samples are intensively dolomitized, in which dolomite replaces considerable amounts of the framework grains. Ferroan dolomite is frequent also as fracture-filling cement.

In some samples authigenic siderite is present in the form of spherulitic siderite filling some pore spaces. They are usually of about 30 microns in average size, and of brown amber colour. They consist of fibrous crystals arranged radially around fine-grained core of carbonaceous debris. They are commonly altered to reddish or brownish ferrogenous material.

(c) Authigenic kaolinite is present as vermicular aggregates filling the fractures in some rocks.

B.2 CLAYEY SILTSTONES (Plate 21, Fig. 2)

Clayey siltstones are generally present as seatearth underlying some coal seams or interbedded with the argillaceous rocks of the Lower and Middle Coal Measures rocks. They are usually of dark grey or greenish grey in colour, and very hard with abundant rootlet traces.

Microscopic structure:

The rock consists mainly of ill-sorted quartz grains with subordinate amounts of mica floating in a clayey groundmass. The clayey groundmass is usually of brownish colour and it consists of organic matter-rich clays. Frequent carbonaceous matter is present as ill-sorted silt-size debris and as carbonaceous rootlets. Authigenic siderite is commonly present.

Textures:

The rock consists of open framework of quartz grains and mica flakes floating in clayey ground mass. Clays consist mainly of micaceous clay minerals which show an original preferred orientation parallel to the bedding, but it has been disturbed by the rootlets' penetration, so most of them are pointed towards the rootlets growth distribution. Quartz grains are of a wide range of silt size, ranging from very coarse silt to very fine silt size. They are commonly subrounded to subangular.

Mineralogy:

Terrigenous detritus:

(a) Quartz is represented by monocrystalline quartz grains, the majority are unstrained. It represents more than 70 percent of the framework grains.

(b) Mica is represented mainly by muscovite and chlorite. They are usually undeformed. Chlorite is present as isotropic and low bireferengent types. Mostly mica flakes are altered to kaolinite.

(c) Clay minerals represent more than 20 percent of the rock. Clay minerals occur as micaceous clay minerals which are of low bireferengence and these are illite and illite-montmorillonite mixed layer. Granular grains with first order grey interference colour have been recognised and identified as kaolinite. X-ray analysis of the $<2\mu$ fraction confirmed the presence of these minerals plus small amounts of chlorite.

(d) Heavy minerals are rare and are represented by zircon and green tourmaline.

Chemical constituents:

(a) Authigenic quartz is present mainly as microcrystalline quartz aggregates filling some intergranular pore spaces.

(b) Siderite is the most frequent orthochemical mineral present in the rock. It was found to be present in two forms, namely; rhombohedral siderite and spherulitic siderite. Rhombohedral siderite occurs as brownish rhombs of about 20 microns in average size, scattered in the clayey groundmass. Many rhombohedra have nuclei of dark brown material which could be organic debris. Spherulitic siderite is the most common form of siderite. Spherulites are usually scattered between the framework grains and grown on the expanse of clay minerals of the matrix. They are of about 60-80 microns in size, spherical, ellipsoidal or irregular in shape. They usually have a rosette-shape outline and the inner inclusion zones follow the same shape. Spherulites consist of fibrous siderite crystals arranged radially around organic debris nuclei.

(c) Francolite is present as finely crystalline idiotopic crystals of about 30 microns in average size. It occurs as prismatic rectangular crystals, colourless or very pale yellowish in colour. It is characterised by its low bireferengence and strong relief. It is present as single scattered crystals or as crystal aggregates of two or three crystals in the clayey matrix. It is very rare and has been noticed only in siltstones of the anthracite area.

C. ARGILLACEOUS ROCKS

Argillaceous rocks are the most frequent rocks in the Lower and Middle Coal Measures. On the basis of megascopic and microscopic characteristics, three types of argillaceous rocks have been recognised, namely; shale, mudstone, claystone.

C.1 SHALE

Commonly it occurs as thin beds forming the roof of the coal seams. They usually contain thin streaks of coal and frequent amounts of siderite nodules of varying sizes and shapes. It is black in colour and highly fissile.

Microscopic description:

It consists mainly of micaceous clay minerals with subordinate amounts of quartz. Frequent amounts of carbonaceous matter and pyrite are present. Micaceous clay mineral flakes commonly lie parallel to the bedding.

Mineralogy:

Terrigenous detritus:

(a) Clay minerals; they form more than 70 percent of the rock. They are mainly present as micaceous flakes reaching up to 20 microns in length. Micaceous flakes consist mainly of illite, which is identified by its characteristic bireferengence. Kaolinite is present as equigranular grains in small amounts. Generally the clay mineral groundmass is of brownish colour. This is due to the occurrence of organic matter. Organic matter as extremely fine-grain particles occurs in the intergranular spaces between the illite flakes. X-ray analysis of the $< 2\mu$ fraction proved that illite and illite-montmorillonite mixed layer are the main clay minerals present. Kaolinite and chlorite are commonly present in small amounts.

(b) Quartz is commonly present in small amounts, but in some rocks it reaches up to 30 percent. It occurs as monocrystalline grains of about 10-15 microns in size. It is usually sporadically distributed in the shale but sometimes it concentrates on the bedding or fills some lenticular cavities.

Chemical constituents:

(a) Siderite is the most common orthochemical mineral in the shales. It is present as minute rhombs randomly distributed in the rock, and as microcrystalline siderite aggregates commonly frequent along bedding and associated with the organic debris.

(b) Francolite is present only in the shales of the anthracite area. It is present as minute idiotopic prismatic crystals of very low birefirengence. They usually contain very fine inclusions but reject the organic matter during their crystallization. This is evident by the presence of organic matter surrounding most of the crystals, as if it has been pushed outward during the replacement of the francolite to the clay minerals.

C.2 MUDSTONE (Plate 21, Fig. 3)

This is the most frequent type among the argillaceous rocks. It is commonly of dark grey to black in colour, very hard and characterised by thin laminations which are due to alternating black or dark grey laminae and light grey laminae. The following are the main sedimentary structures which have been recognised:

a. Horizontal stratification. This is the most dominant structure throughout most of the mudstone surfaces. Horizontal stratification is displayed by the alternating laminae of dark grey or black argillaceous laminae rich in organic matter and light grey laminae rich in quartz grains of fine silt size.

b. Borings and burrows are common and have been recognised in most of the mudstones.

Microscopic description:

The rock consists of alternating laminae of light and dark colour. The dark coloured laminae consist mainly of clayey matter with abundant amounts of organic matter and siderite. The light laminae consist of fine-grained mica flakes and quartz grains of silt-size floating in clayey matrix.

Mineralogy:

Terrigenous detritus:

(a) Quartz is represented by monocrystalline quartz grains of about 30 microns in average size. It is usually concentrated in the light coloured laminae.

(b) Clay minerals; these form the main mineral constituent of the rock. Micaceous clay minerals are the most frequent and are represented by illite and illite-montmorillonite mixed layer minerals. X-ray analysis of the $< 2\mu$ fraction has confirmed the occurrence of both minerals, plus small amounts of kaolinite and chlorite.

(c) Mica is present as silt-size flakes of muscovite and chlorite. It usually lies parallel to the bedding. Most of the muscovite flakes show partial or complete alteration to kaolinite.

(d) Organic matter is very frequent and is present as silt-size debris randomly distributed in the rock and as thin sheaths of carbonaceous matter lying parallel to the bedding.

Chemical constituents:

(a) Authigenic siderite is the most important orthochemical mineral. It is commonly present as microcrystalline aggregates concentrated on the bedding and associated with the organic debris. Siderite rhombs of very fine silt-size have been noticed.

C.3 CLAYSTONE (Plate 21, Figs. 4, 5 & 6)

Claystones are present throughout the Coal Measures sequence, but they are most frequent in the Lower and Middle Coal Measures. They are present usually beneath the coal seams as seatearth or underclay rock. It varies in thickness from a few inches to several feet.

Varieties of Claystones:

On the basis of megascopic description the following varieties of claystones have been identified:

1. Underclays: underclays or claystone seatearth are those which commonly underlay the coal seams. They are the most frequent type of claystones. They are usually of greyish colour and range from friable to very hard. Generally they are massive and rich in randomly oriented rootlet imprints and plant fossils. On the basis of the frequency of the silt-size quartz grains two types of underclays have been recognised, namely; clayey underclays and silty clayey underclays. Usually the silty clayey underclays are lighter in colour and less frequent.

2. Thin-bedded claystones; these are massive laminated claystones, friable to very hard. They usually underlie the underclay and are interbedded with the mudstones. They also occur as intercalations with the sandstones. On the basis of their colour, three varieties have been recognised, namely, grey, green and red claystones. The two latter varieties are rare and commonly present in the Upper Coal Measures, while the greyish claystone is the most frequent type, especially in the Lower and Middle Coal Measures.

Sedimentary structures:

The main sedimentary structure displayed by the claystones is the horizontal stratification. Horizontal stratification is very common among the thin-bedded claystones. This structure is displayed by alternating very thin laminae of varying colours. It was found that lamination was mainly due to alternating laminae of organic matter-rich clays and clays poor in organic matter.

Tectonic structure:

The main and most prominent tectonic structure among the claystones is the slickensides; this is common in the underclays. This structure forms polished, very finely striated surfaces which cut the bedding at a low angle.

Microscopic description:

Claystones consist mainly or wholly of clay minerals, silty types contain about 10 percent of quartz grains. Mica flakes and authigenic siderite are common as well as the carbonaceous matter.

Textures:

In the case of the thin-bedded claystones, micaceous clay mineral flakes as well as the small amounts of detrital mica flakes are generally preferably oriented parallel to the bedding. While in the underclays the original fabric of the micaceous flakes is disturbed by the penetration of the plant rootlets just after the sedimentation of the clayey sediments.

Mineralogy:

Terrigenous detritus:

(a) Clay minerals:

Clay minerals forming most of the claystone varieties are of two types, namely, micaceous clay minerals and granular clay minerals. On the basis of their optical properties it was found that micaceous clay minerals are mainly of illite and chlorite, while the granular clay minerals are mainly of kaolinite. Illitic clays are the most frequent type among the other clay minerals. X-ray analysis of the $< 2\mu$ fraction revealed that illite and illite-montmorillonite mixed layer are the most dominant clay minerals in most of the claystone varieties, while kaolinite and chlorite are present in subordinate amounts. In the reddish claystones of the Upper Coal Measures abundant amounts of iron oxides have been detected microscopically and by the X-ray analysis.

(b) Quartz:

Detrital quartz grains of siltsize are present in considerable amounts in the silty claystones. They are usually of about 30 microns in average size. Quartz is present as monocrystalline unstrained quartz.

(c) Mica:

Mica is present in considerable amounts in most of the claystones. Three types have been recognised, namely, muscovite, biotite and chlorite. Muscovite is the most frequent mica mineral in most of the claystones. In green thin-bedded claystones of the Upper Coal Measures chlorite was found to be the most frequent mica mineral. Muscovite flakes are usually partially or completely altered to kaolinite. Kaolinitization is preferential to the cleavage planes of the muscovite flakes. Kaolinitized muscovite flakes are commonly bulged in a direction perpendicular to the cleavage plane. Chlorite is represented commonly by pale green, low bireferengent type. Biotite is present in some claystones and it is the least frequent mica mineral. Biotite of underclays is usually altered to brownish ferrogenous material.

(d) Organic matter:

Organic matter is very abundant in most claystone types and especially in the underclays. The following are the different types of organic matter which have been identified in most of the claystones:

i) Extremely fine-grained particles of organic matter debris filling pore spaces between the clay particles. These are responsible for the colouration of the clay mineral matter by grey or greyish-brown colour in this section.

ii) Organic matter debris ranging in size from coarse silt to coarse clay size. Most of these debris are carbonaceous plant remains.

iii) Plant remains of well defined structure, e.g. spores and cuticle. They are usually of brownish colour.

iv) In some thin-bedded claystones, algal mat structure has been recognised. This occurs as very thin sheets of carbonaceous matter, usually continuous, or sometimes discontinuous (Plate 21, Fig. 4).

Chemical constituents:

(a) Siderite:

Siderite is the most common orthochemical mineral in most of the claystones. It commonly occurs in spherulitic form and to a lesser extent as microcrystalline particles or of rhombohedral form. Spherulites vary in size from 80 microns to about 2 mm. They are composed of radially arranged laths. The outer boundaries of the laths are often irregular and usually exhibit iron staining. On occasions quartz grains and clay matter occur as inclusions within the laths. Many spherulites exhibit thin zones of organic matter. These zones are usually present parallel to the surface of the spherulites. The occurrence of the organic matter zones within the spherulites suggests that the sideritic spherules could have started to grow inside a globular organic body, such as a spore or any plant cell, and by growing up of the spherulites the organic membrane stretched to a certain extent, after which siderite laths continued their growth outside the organic membrane in optical continuity with the laths inside the stretched membrane. Siderite spherulites usually occur filling rootlet cavities or surrounding them.

(b) Kaolinite:

Authigenic kaolinite represented by vermicular aggregates has been recognised in some claystones. It usually fills cavities and thin fractures in the rock.

(c) Francolite:

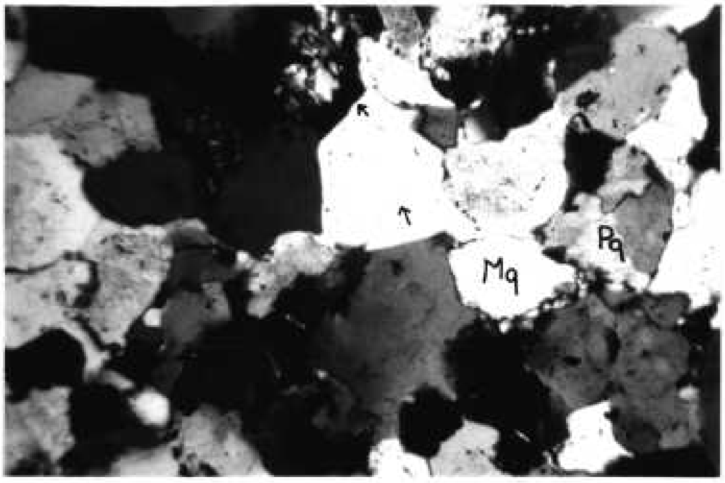
Francolite is a common authigenic mineral in the claystones of the anthracite area.

PLATE 18

Fig. 1 Quartzarenite of the Basal Grit Group of the Millstone Grit. The rock consists mainly of monocrystalline quartz (Mq) and polycrystalline quartz grains (Pq). Note the secondary overgrowths (arrows). Sample 82. Crossed polars.

Fig. 2 Lithic arenite of the Farewell Rock Group of the Millstone Grit. Lithic fragments (LF), quartz (Q). Sample 652. Crossed polars.

Fig. 3 Shale of the Middle Shale Group of the Millstone Grit. Sample 614.



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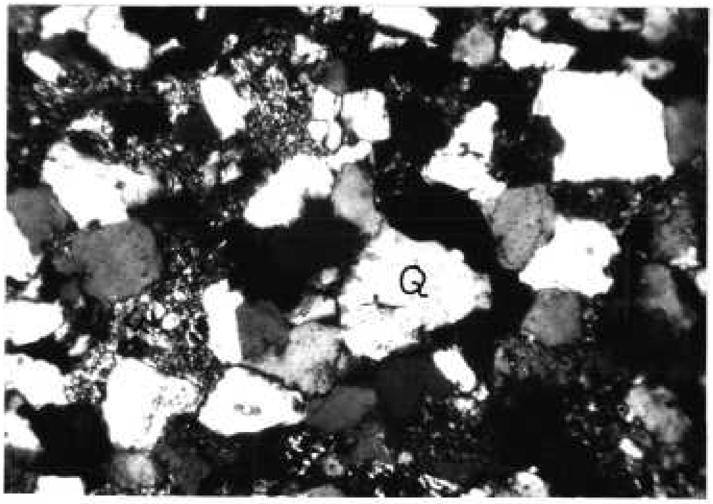

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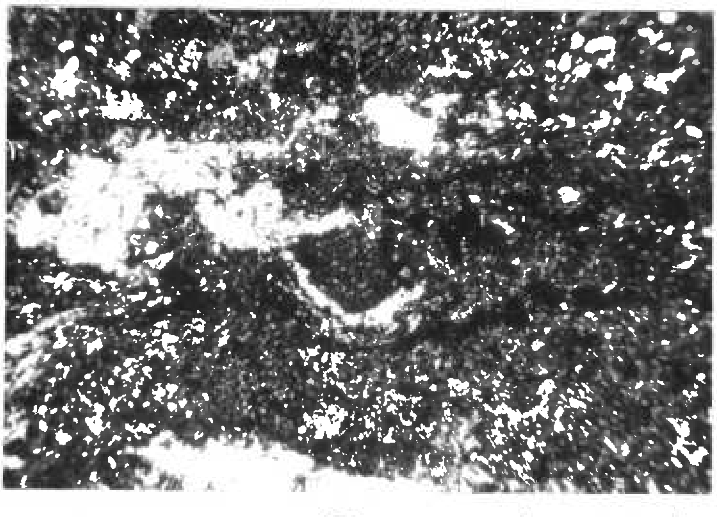

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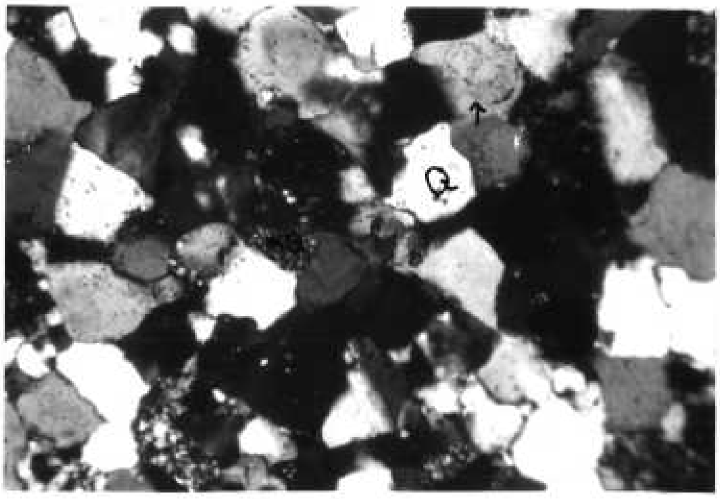
PLATE 19

Fig. 1 Quartzarenite of the Lower Coal Measures, consists mainly of monocrystalline quartz (Q), cemented by quartz overgrowths (arrow) and microcrystalline quartz (mQ). Sample 141. Crossed polars.

Fig. 2 Poorly sorted quartzarenite from the Lower Coal Measures. Sample 154. Crossed polars.

Fig. 3 Quartzarenite of the Middle Coal Measures, from the anthracite area. Sample 911. Crossed polars.



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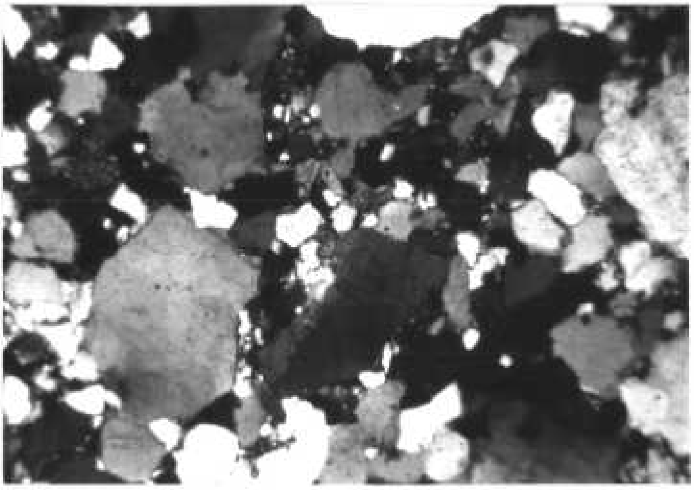

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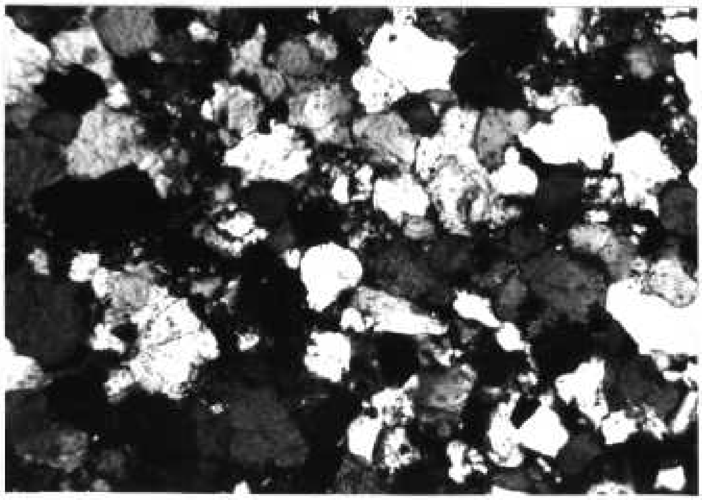

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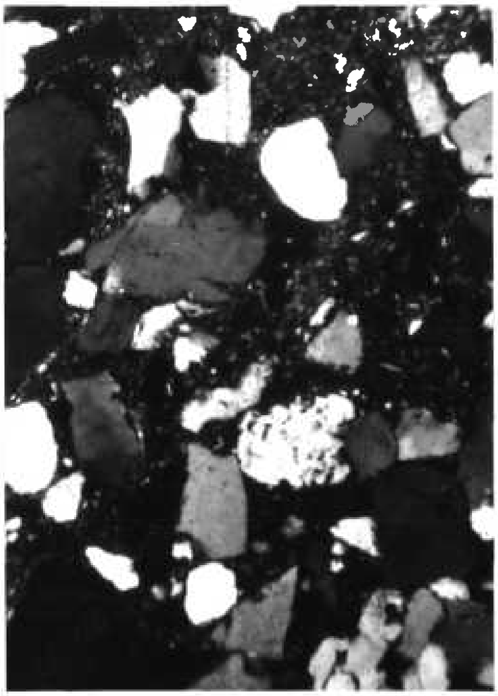
PLATE 20

Fig. 1 Quartzwacke, Coal Measures. Sample 170. Crossed polars.

Fig. 2 Lithic arenite of the Upper Coal Measures. (AF) argillaceous rock fragment. Sample 722. Crossed polars.

Fig. 3 Lithic arenite of the Upper Coal Measures. (AF) argillaceous rock fragment, (PF) phyllite fragment. Sample 745. Crossed polars.

Fig. 4 Lithic arenite of the Upper Coal Measures. (QF) quartzitic rock fragment. Sample 832. Crossed polars.



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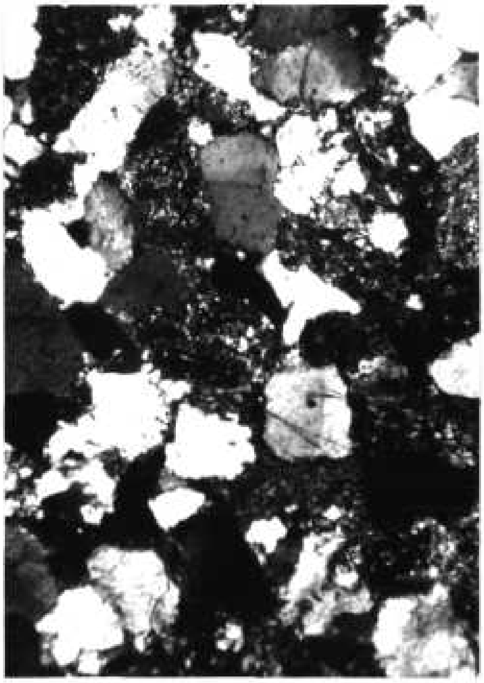

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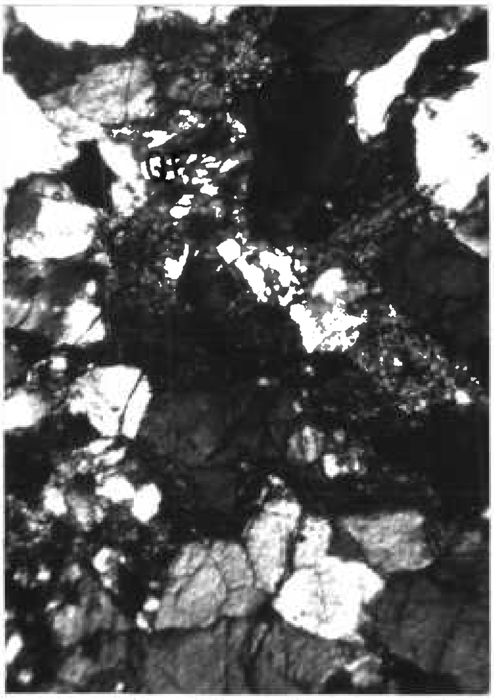

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PLATE 21

Fig. 1 Micaceous siltstone, Coal Measures. Sample 231.

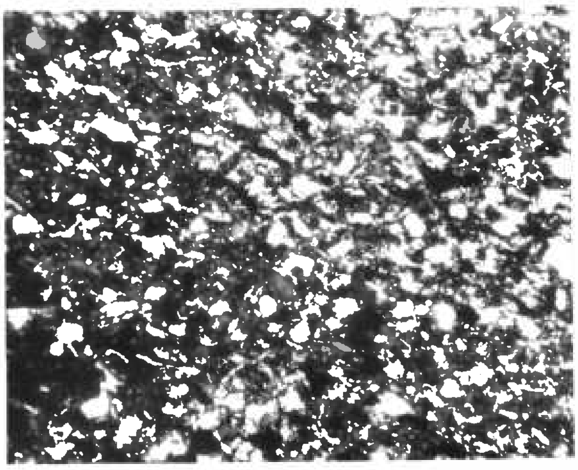
Fig. 2 Clayey siltstone, Coal Measures. Sample 802.

Fig. 3 Mudstone, Coal Measures. Sample 813.

Fig. 4 Algal mat (?) in claystone, Coal Measures. Sample 183.

Fig. 5 Underclay, Coal Measures. Note the carbonaceous plant rootlet (arrow). Sample 914.

Fig. 6 Underclay, Coal Measures. Sample 2493-10. Crossed polars.



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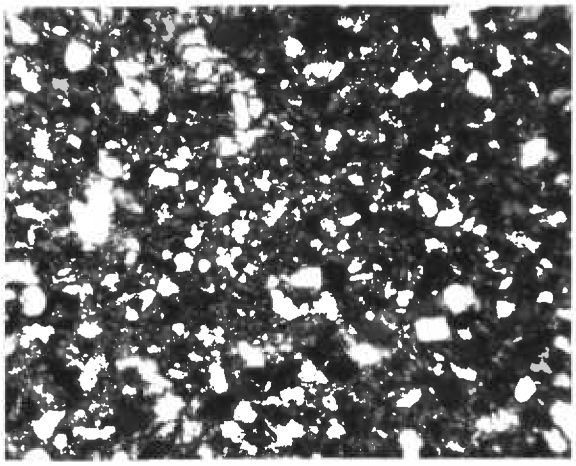

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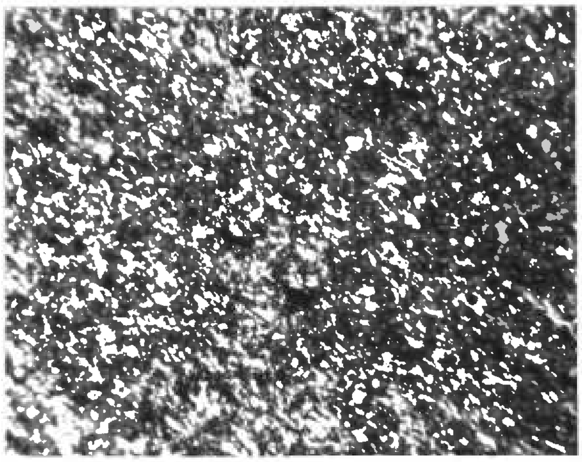

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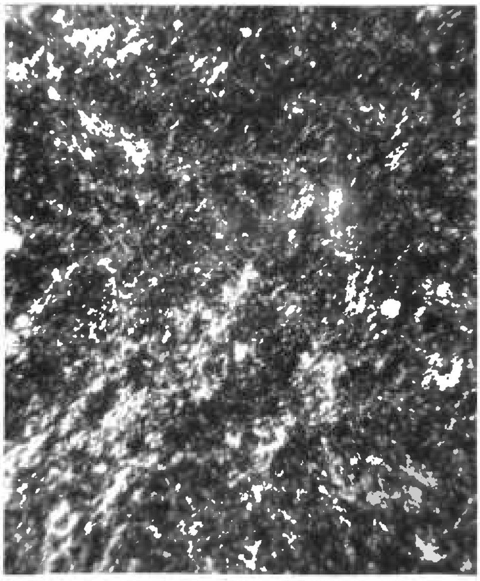

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CHAPTER V

DIAGENESIS OF THE TERRIGENOUS ROCKS OF THE MILLSTONE GRIT AND THE COAL MEASURES SERIES

DIAGENESIS OF THE TERRIGENOUS ROCKS OF THE MILLSTONE GRIT AND THE COAL MEASURES SERIES

INTRODUCTION

As no published information is available on the diagenesis of the terrigenous rocks of the Millstone Grit and the Coal Measures Series of the South Wales Coalfield, the attempt of the present study is to discuss in detail the diagenetic processes which have modified these rocks. However, references to cementation and diagenesis do exist for sandstones of Millstone Grit and Lower Coal Measures observed in other areas; among the most significant contributions are those of Sorby (1859), Gilligan (1920), Dunham (1953), Greensmith (1957) and Hawkins (1972).

This chapter consists of two parts. The first part will deal with the diagenetic modifications of the studied rocks. The second part deals with the origin and the conditions of formation of the authigenic minerals.

DIAGENETIC MODIFICATIONS

Introduction

In this part, the diagenetic modification which affected the various lithotypes of the terrigenous rocks of the Millstone Grit and the Coal Measures Series of South Wales will be discussed in detail. To avoid repetition, the diagenetic modifications displayed in all of the studied lithotypes will be described collectively. For the purpose of description, the diagenetic processes are grouped as physical, physiochemical and chemical.

A. PHYSICAL PROCESSES:

Generally, these processes have affected the rocks through their early stages of diagenesis and have been continued until the very late stages. These processes can be grouped as : (1) arrangement of the micaceous flakes; (2) grain fracturing and crushing; (3) grain bending; and (4) grain squeezing.

(1) Arrangement of the micaceous flakes:

This process is most evident in the argillaceous rocks, especially the shale. Compaction usually affects the orientation of the clay flakes forming the bulk of these rocks. O'Brien (1970) studied the fabric of the various types of shales and claystones of various ages from different localities in the United States and Japan with the aid of the electron microscope. He suggested that the fabric of shales may result either from the deposition of dispersed clay or by the collapse of clay globules after deposition.

White (1961) and O'Brien (1964) have found that in very fissile grey shales clay platelets are oriented with their C-axes perpendicular to the fissility plane. Heling (1970) in his study of the microfabrics of tertiary shales from the Rhein Graben and their rearrangement by compaction, found that the compaction of these shales below a depth of 100 m. can be differentiated in two stages: (a) shallow-burial stage, and (b) deep burial stage. From his detailed study on the mineralogy and porosity and surface area measurements, he concluded that only in the shallow burial stage the geometric model of increasing parallel orientation with increasing compaction pressure agrees with the data evaluated from pore size and specific surface area measurement. In the deep buried shales the porosity of the clay mineral fabric is no longer controlled by mechanical rearrangement but by the formation of illite aggregates which are coarser and more perfectly crystallized as overburden grows.

In the studied argillaceous rocks of the Millstone Grit and the Coal Measures, it was found that the fabric of micaceous clay flakes is dependent mainly on the depositional environment. Therefore in the shales the clay flakes are very well oriented parallel to the bedding, while in the underclays the original fabric of the clay flakes has been disturbed by the action of the plant bioturbation.

(2) Grain fracturing and crushing:

This process is most evident in the litharenites of the Millstone Grit and the Coal Measures Series. Coarse-grained quartz and feldspar grains are found to be the most susceptible mineral grains to fracturing. Fractured quartz grains are not uncommon, and occurred where they are surrounded by other competent framework grains. Fractures in quartz grains are commonly filled with the flowing pseudomatrix of argillaceous rock fragments. Occasionally intensive fracturing of the quartz grains is accompanied by spalled-off slivers of quartz, some of which may display rotation.

Feldspar grains are found to be the most sensitive to fracturing. Fractures in these grains are usually irregular, some are parallel to the cleavage. Fracturing is commonly resulting in the displacement of twin planes, some grains are crushed and the fragments have been rotated. Rotation of the fragments in the crushed feldspar grains is usually produced by the force of crystallization of the dolomite precipitated along the fractures (Plate 22 , Fig. 1).

(3) Grain bending:

This process manifests itself in most of the sandstones and siltstone lithotypes where mica flakes and lensoidal argillaceous rock fragments are bent so as to conform to the configuration of the adjacent framework grains. Occasionally, muscovite flakes are bent in a way so as to envelope about three-quarters of a quartz grain perimeter and some are folded. Bent mica flakes usually display strained extinction (Plate 22 , Figs. 3 and 4).

Commonly argillaceous rock fragments are smoothly bent between the rigid framework grains.

(4) Grain squeezing:

This process is demonstrated in most of the sandstones and siltstone lithotypes. Squeezing of plastic and flexible grains to fill the pore spaces between the rigid framework grains is the main result of this process. Argillaceous rock fragments and mica flakes are the only grains that have been affected by this process.

In the quartzarenites and quartzitic siltstones, muscovite and biotite flakes are commonly squeezed between the quartz grains. In the case where the mica flakes have been squeezed at their central part, their extremities usually develop a fan-shape of mica shreds filling the adjacent intergranular pore space. In some cases mica flakes have been squeezed between the quartz grains to completely fill the pore spaces between these grains. In the latter case the flakes are usually squashed to form a lump consisting of randomly oriented minute mica shreds (Plate 22 , Fig. 5).

In the litharenites, where argillaceous rock fragments are abundant, grain squeezing process has played a great role in the diagenetic modification of these rocks. In these rocks claystone fragments have been squeezed to fill partially or completely the intergranular pore spaces. In the early stages of grain squeezing the original fabric of the claystone rock fragments can easily be recognised, while in the advanced stages it was difficult to recognise. The developments of the pseudomatrix as a result of the deformation of the argillaceous rock fragments are the most important diagenetic change that has taken place as a result of the action of the grain squeezing process in the litharenites (Plate 22, Fig. 2).

B. PHYSIO-CHEMICAL PROCESSES:

The role pressure solution:

This process is displayed by most of the sandstone and siltstone lithotypes. The role of pressure solution process has been evident from the nature of the framework grain boundaries. It was found that the pressure solution process is effective in rocks where the orthochemical cementation is insignificant. Therefore the process has played an important role in most of the litharenites and wackestones, while it was insignificant in quartzarenites.

The following types of grain boundary have been recognised; tangential, straight, concavo-convex, and stylolitic. It was found that the type of the grain contact is a function of the degree of compaction and the nature of the framework grains. Straight contacts are usually developed between quartz grains and mica flakes and also between rock fragments and mica flakes. Concavo-convex contacts are commonly found between two quartz grains, and between quartz grains and argillaceous rock fragments. Two types of concavo-convex contact can be recognised. These are, contacts developed by dissolution of one grain faster than the other; and contacts developed by pressing of a rigid grain, such as quartz, into a flexible argillaceous rock fragment. The shape of the latter type of concavo-convex contact depends on the original shape of the quartz grains, i.e. a smooth concavo-convex contact usually develops between rounded quartz grains and an argillaceous rock fragment, while an angular concavo-convex contact commonly exists between an angular quartz grain and an argillaceous rock fragment.

Stylolitic grain contact usually develops between two adjacent quartz grains and between quartz grains and argillaceous rock fragments. On occasions a very thin film of illitic clay is present on the stylolitic fragments.

C. CHEMICAL PROCESSES:

(1) Precipitation of pore-filling minerals:

This process involves the precipitation of minerals within empty pore spaces, resulting in either partial or complete filling of pores without modification to the adjoining framework minerals. Precipitation of pore-filling minerals has played a great role in the lithification of most of the sandstone and siltstone lithotypes. Four minerals have precipitated in this process, namely, quartz, dolomite, siderite and kaolinite, with quartz playing the most active role.

(a) Development of quartz cement:

Authigenic quartz has played an important role in the cementation of the sandstones and siltstones of the Millstone Grit and the Coal Measures Series. It has been developed as pore-filling as well as fracture-filling cement. According to the fabric, four types of quartz cement have been recognised, namely, quartz overgrowths, microcrystalline quartz, spherulitic chalcedony and idiotopic quartz crystals.

(i) Quartz overgrowths:

Quartz overgrowths have been recognised in most of the sandstones and siltstone lithotypes. Quartz overgrowths fill almost all of the pore spaces in the quartzarenites and quartzitic siltstones, while it is less important in the quartz wackestones and clayey siltstones.

Quartz overgrowths have been developed by the precipitation of clear quartz rims in lattice continuity with the host quartz grains. It was found that grain growth takes place preferentially in the direction of the C-axis. Commonly it was found that the lattice properties of the detrital quartz grains are inherited by their overgrowths. Therefore strained lattice structure of some host quartz grains has been transmitted to their overgrowths.

The recognition of quartz overgrowths has been greatly facilitated by the presence of vacuoles and inclusions in the host quartz grains; while the overgrowths are usually free from impurities (Plate 23 , Fig. 1). The presence of minute dolomite or siderite rhombs forming a rim around the detrital quartz has helped in the recognition of the quartz overgrowths.

In the quartzarenites, pore spaces are usually completely filled with the quartz overgrowths. Where pore spaces were large enough, quartz overgrowths usually develop with euhedral terminations. Overgrowths mutually interfere with one another along compromise boundaries to produce a mosaic of interlocking crystals. Compromise boundaries may be straight or irregular, this being dependent on the orientation of the C-crystallographic axes of the adjacent quartz grains. Commonly apophyses of illite clay have been seen trapped along compromise boundaries of the overgrowths, suggesting that clay once existed in pore spaces but was subsequently squeezed out during the crystallization of the overgrowths.

In the litharenites, quartz overgrowths have been seen to develop mainly on the monocrystalline quartz grains as a thin rim. The degree of development of the quartz overgrowths depends on the nature of framework grains adjacent to the quartz grains.

In the quartz wackestones, the development of the quartz overgrowths is very limited, especially where it is adjacent to the clay matrix. The quartz overgrowths usually grow by pushing aside the clay matrix to a certain limit at which the clayey matrix attains its maximum compaction. Commonly the boundaries between the overgrowths and the adjacent clay matrix are very irregular.

(ii) Microcrystalline quartz:

Microcrystalline quartz is the most abundant form of quartz cement in the quartz wackestones and clayey siltstones. It has been recognised also in the other lithotypes. It is present as patchy aggregates consisting of equant quartz microcrystals, filling partially or completely the intergranular pore spaces (Plate 23, Fig. 2). Occasionally microcrystalline quartz occurs infilling plant rootlets in silty seatearth. It was also recognised filling some of the shell cavities in the marine shales.

In the sandstone and siltstone lithotypes, microcrystalline quartz is generally found to be filling the rest of the pore spaces left after the precipitation of the quartz overgrowths. Where microcrystalline quartz occurs adjacent to quartz overgrowths, the microcrystalline quartz passes from fine to coarser grained crystals, with the latter appearing to merge into the overgrowths. Microcrystalline quartz occurs infrequently filling microfractures in quartzitic siltstone.

(iii) Spherulitic chalcedony:

Spherulitic chalcedony is infrequently present as pore filling cement. It is relatively most common in the quartzarenites and quartzitic siltstones of the Coal Measures Series. Where spherulitic chalcedony occurs, it is present as a spherule consisting of fibrous radiating quartz crystals, (Plate 23, Fig. 3).

(iv) Idiotopic quartz cement:

Idiotopic quartz crystals have been recognised only in the fracture-filling cement of some quartzarenites. They are commonly present as single euhedral crystals floating in dolomite or kaolinite fracture-filling cement. Near the border of the fracture, idiotopic quartz crystals seem to be developed as an overgrowth on the detrital quartz grains forming the wall of the fracture. In the latter case, the idiotopic quartz crystals have been developed in such a way that their C-crystallographic axes are normal or slightly inclined to the fracture wall.

(b) Development of dolomite cement:

Dolomite has been recognised as pore-filling and fracture-filling cement in the sandstone lithotypes of the Coal Measures. It is rarely observed in the Millstone Grit rocks. Staining by potassium ferricyanide has proved that dolomite is present mainly as ferroan dolomite.

In the quartzarenites, dolomite is present in two forms, namely, as finely crystalline rhombs floating in the intergranular cement (Plate 23, Fig. 2), and as coarsely crystalline cement filling completely some of the pore spaces. In the latter form, dolomite is present as a single xenotopic crystal filling one pore space, sometimes it fills more than one pore space enclosing two or three quartz grains forming a poikilotopic texture (Plate 24, Fig. 3). Therefore two stages of dolomite cementation are evident in these rocks. An early stage pre-dates the quartz overgrowth cementation, and is represented by the finely crystalline dolomite rhombs; and a late stage post-dating the quartz overgrowths cementation and represented by the coarsely crystalline xenotopic dolomite crystals.

In the litharenites and quartz wackestones, dolomite has been developed partially filling the pore spaces. It is usually present as idiotopic and hypidiotopic crystals with sharp rhombohedral edges, usually filling the pore space.

Dolomite is frequently observed as fracture-filling cement. It is commonly present as coarsely crystalline , mosaic cement (Plate 23, Fig. 4). Dolomite cement fills thin fractures and commonly consists of discrete elongated crystals.

(c) Development of siderite:

Siderite is present in most of the lithotypes of the Millstone Grit and the Coal Measures Series.

In the litharenites and the quartz wackestones of the Coal Measures, siderite is present mainly in two forms, namely, as minute rhombs usually surrounding the detrital quartz grains, and as medium-crystalline rhombs lined some of the pore spaces. In some litharenites siderite occurs as infilling in the cellular cavities of some fossilized plant tissues (Plate 24, Figs. 5 and 6). Cellular cavity-filling siderite is present in one of the following forms; (a) as a single crystal filling one cell cavity; (b) as a coarse single crystal filling more than one cell cavity; and (c) as finely crystalline mosaic filling cell cavities.

In the siltstones of the Millstone Grit and the Coal Measures, siderite is present mainly as microcrystalline aggregates filling partially or completely some of the pore spaces. Microcrystalline aggregates are commonly concentrated along the bedding and associated with the carbonaceous matter debris.

In the argillaceous rocks, especially in the shale of the Millstone Grit and the Coal Measures, siderite occurs in two forms, namely, as minute rhombs sporadically distributed in the rock; and as microcrystalline aggregates preferentially occurring along the bedding associated with the organic matter debris.

(d) Development of kaolinite:

Authigenic kaolinite is frequently present as pore-filling cement in the quartzarenites of the Millstone Grit and the Coal Measures. In this lithotype, kaolinite is present as coarsely crystalline vermicular aggregates filling the pore spaces left after the development of the quartz overgrowths (Plate 26, Fig. 1). The absence of any traces of illitic clays or muscovite shreds within the pore-filling kaolinite cement adds more evidence to the precipitation origin of such kaolinite.

Authigenic vermicular kaolinite has been recognised also as pore-filling cement in the quartzitic siltstones of the Coal Measure Series. In the sideritic nodules which are frequently present in the argillaceous rocks of the Coal Measures and the Millstone Grit, authigenic kaolinite has been recognised as the main fracture-filling cement in these nodules.

(2) Mineral replacement:

This process involves partial or complete replacement of either framework components or newly formed authigenic minerals. The important minerals in this process are; siderite, dolomite, microcrystalline quartz, kaolinite and pyrite.

(a) The role of siderite as a replacement mineral:

Siderite as a replacement mineral has played a great role in the diagenetic modification of most of the Millstone Grit and Coal Measures lithotypes, with the exception of the quartzarenites. In the Coal Measures litharenites, the following fabrics of siderite replacement have been identified:

i) Microcrystalline siderite present as aggregates filling some of the pore spaces are found to be progressively replacing the adjacent framework grains. Occasionally, undigested remnants of quartz grains are found to be enclosed within the microcrystalline siderite aggregates.

ii) Finely crystalline siderite which has been developed as cement is found to be partially replacing the framework grains resulting in the development of a false open-textured framework (Plate 24, Fig. 4).

iii) Medium-crystalline siderite which had been developed as scattered rhombs in some of the argillaceous rock fragments has completely replaced the latter. The general shape of these rock fragments is preserved as a ghost formed of the undigested particles.

iv) The various stages of muscovite replacement by siderite can be seen in these rocks. Incipient sideritization of the muscovite is displayed by the presence of fine siderite rhombs replacing muscovite flakes along thin cleavage planes. Progressive sideritization results in almost complete replacement of the muscovite flakes by siderite. Sometimes thin shreds of the muscovite flakes are present as remnants within the siderite pseudomorph.

In the argillaceous rocks, siderite as a replacement mineral has played an important role in the diagenetic modification of the rocks. In the shales of the Millstone Grit and the Coal Measures, siderite is present mainly as nodules ranging in length from a few millimetres to up to 50 cms. and ranging in height from a few millimetres up to 20 cms. These nodules had originally developed as minute microcrystalline pellets precipitated around the carbonaceous matter debris concentrated along the bedding. Progressively, these microcrystalline-siderite aggregates have grown replacing the other rock constituents. The replacive nature of the nodules is evident from the presence of undigested inclusions of illitic clays and quartz grains.

Replacive siderite nodules commonly consist of a mosaic of microcrystalline siderite and they usually contain pyrite. Pyrite is present as hypidiotopic crystals concentrated in the core of the siderite nodules and sometimes concentrated in a zone near the periphery of the nodule. The presence of the pyrite within the siderite nodules could be due to the fluctuation of the sulphate content of the connate water in the shale.

Micaceous clay flakes surrounding the siderite nodules usually show a flow texture. It is suggested that replacing siderite nodules have been developed in early stages before the rock has reached its maximum compaction. During the compaction process the siderite nodules resist the compaction, while the host clays have been liable to the compaction and as a result flow texture has been developed.

In the claystones of the Coal Measures, especially in the underclays, replacive siderite is frequently present in the form of spherulites. Siderite spherulites are ranging in size from 0.1 to 1.5 mm. They are usually randomly distributed in the clayey groundmass. Concentrations of spherosiderite nodules around the fossilised plant rootlets have been recognised (Plate 25, Fig. 3). Siderite spherulites commonly consist of a core of microcrystalline siderite surrounded by a relatively thick envelope consisting of fibrous siderite crystals radially arranged around the core (Plate 25, Fig. 1). Some fine-grained spherules consist of a mosaic of radially arranged fibrous siderite crystals. Inclusions of clays and quartz grains frequently occur in the spherulites.

On occasions replacive spherosiderite is present as discontinuous thin bands in the siltstones of the Coal Measures. These bands consist wholly of a mosaic of siderite spherulites (Plate 25, Fig. 2). Remnants of the original rock constituents commonly occur between the spherulites. The spherulites in these rocks are commonly uniform in size and spherical in shape. They usually consist of a mosaic of three or four equicrystalline siderite crystals. Some of the spherulites are formed of a single siderite crystal.

(b) The role of dolomite as a replacement mineral:

The replacement role of dolomite can be demonstrated in many of the sandstone and siltstone lithotypes of the Millstone Grit and the Coal Measures, but it is more significant in the quartzarenites and the quartzitic siltstone.

In the quartzarenites it was found that dolomite which had been developed as pore-filling cement has progressively replaced the quartz overgrowths and in advanced stages it has replaced the detrital quartz grains. In these rocks the framework grains display open packing, but on close inspection the outlines of the formerly existing grains can be recognised (Plate 24, Fig. 2).

Replacive dolomite veinlets are commonly present in the quartzarenites. These veinlets are characterised by irregular boundaries which are in fact the interface between the detrital quartz grains and the replacive dolomite. Dolomite forming these veinlets is usually coarsely crystalline with brush extinction. They frequently show a poikilotopic texture formed of more than one quartz grain enclosed within a single dolomite crystal.

In the quartz wackestones, replacive dolomite is present as idiotopic rhombs commonly replacing partially or completely the clayey material and the microcrystalline quartz filling the pore spaces.

In litharenites and feldspathic litharenites, dolomite has replaced feldspar grains (Plate 24, Fig. 1). Three forms of the fabric of the dolomite replacing feldspars have been distinguished. These are: (i) replacive dolomite present as minute rhombs randomly scattered in the feldspar grains; (ii) replacive dolomite present as fracture-filling cement and replacing the feldspars on both sides of the fracture; (iii) centripetal replacement of the feldspar grains by dolomite. Occasionally muscovite flakes and argillaceous rock fragments have been partially replaced by dolomite. In the case of the muscovite flakes, replacive dolomite is preferentially present along the cleavage planes.

In some of the Coal Measure siltstones dolomite replacement has played an important role in the diagenetic changes of the rock. In these rocks the framework grains display an open packing due to the replacement of the matrix and cement minerals by the dolomite. Quartz grains display irregular contacts with the dolomite, where solution of quartz has taken place accompanied by the crystallization of dolomite producing a corroded quartz boundary.

(c) The role of microcrystalline quartz as a replacement mineral:

The role of the microcrystalline quartz as a replacement mineral can be demonstrated in the quartz wackestone and rarely in the quartzarenites. In these rocks microcrystalline quartz commonly replaces the pore-filling clayey matrix (Plate 26, Fig. 2). Isolated patches of clays enclosed in microcrystalline quartz filling pore spaces have been recognised. The boundaries between the microcrystalline quartz cement and the clayey matrix are commonly irregular.

(d) The role of kaolinite as a replacement mineral:

The role of the authigenic kaolinite as a replacement mineral is very limited. In the quartzarenites, kaolinite has been noticed replacing the quartz overgrowths and occasionally replacing the detrital quartz grains (Plate 26, Fig. 3).

(e) The role of pyrite as a replacement mineral:

The role of pyrite as a replacement mineral is restricted and has been recognised in some Coal Measure quartzarenites. In these rocks, pyrite is present as idiotopic crystals replacing the quartz grains and the quartz overgrowths (Plate 25, Fig. 5). Also pyrite has partially replaced some of the muscovite flakes.

(f) The role of francolite as a replacement mineral:

Francolite has been recognised in the argillaceous rocks of the Coal Measures in the anthracite area of the South Wales Coalfield. Francolite is present as small minute idiotopic to hypidiotopic crystals. It is also present as aggregates of three or four crystals. It is suggested that francolite in its late stages of development has replaced the clay minerals forming the argillaceous rocks (Plate 25, Figs. 2 and 4).

(3) Crystallization involving expansion and rearrangement of grains:

This process commonly took place where authigenic minerals have been developed on the cleavage planes and the cracks of the detrital mineral grains. The minerals most actively involved in this process are dolomite and siderite and very rarely quartz.

In feldspathic litharenites, the force of crystallization of dolomite arrests itself along the fractures of some coarse-grained plagioclase grains resulting in the displacement of the fragments (Plate 22, Fig. 1). It has been observed also that extensive shredding and disruption to cleavage planes in some muscovite flakes is the result of the force of crystallization of siderite and dolomite (Plate 26, Fig. 4).

Occasionally the displacement of the siderite envelope around a coaly fragment by the force of crystallization of authigenic quartz has been observed (Plate 26, Fig.5).

Here the siderite has been precipitated as an envelope consisting of a mosaic of minute fibrous siderite crystals arranged radially around the coaly fragments. Authigenic quartz has been developed in the space between the coaly fragment and the siderite envelope pushing aside the latter.

In the partially dolomitized and partially sideritized quartzarenite and quartzitic siltstones, the force of crystallization of dolomite or siderite on the fabric of the framework grains can be demonstrated where open packing occurs. Here many quartz grains are either floating or have tangential contacts in areas pervaded by dolomite or siderite. Although some corrosion of quartz occurs in these areas, there is no evidence to suggest that these large areas are the result directly of grain replacement. Therefore this open packing fabric could be due to the movements of the quartz grains caused by the force of crystallization of the dolomite or the siderite.

(4) Chemical alterations:

Chemical alterations process has played an important role in the modifications of the original mineralogy and the fabric of the framework grains. This process is demonstrated in most of the Millstone Grit and the Coal Measures lithotypes, but it is insignificant in the quartzarenites. This process is manifested mainly by the transformation of the mica and feldspars to clay minerals.

(a) Alteration of feldspars:

Partial alteration of feldspars to clay minerals is demonstrated in the feldspathic litharenites of the Coal Measures. This process is represented by the formation of illite flakes and kaolinite on the expanse of the plagioclase. Sometimes it is concentrated along the cleavage planes and on the grain boundaries as embayments filled with illitic clay.

Occasionally feldspar grains have been partially altered to kaolinite which is present as tiny flakes commonly concentrated along the cleavage plane.

(b) Alteration of muscovite:

This process has played a great role in the diagenetic modifications especially in the siltstones and the argillaceous rocks. It was found that muscovite is commonly altered to kaolinite and illite.

(i) Kaolinitization of muscovite:

Transformation of muscovite to kaolinite is found to be preferential to the areas which can be reached by the aqueous solition, such as fractures and cleavage planes.

Kaolinite can be distinguished in the muscovite flakes by its characteristic low bireferengence. Kaolinitization was found to be responsible for the thickening of the muscovite flakes. Accordion appearance of the partially kaolinitized muscovite flakes is due to the preferential kaolinitization along the muscovite cleavage planes. Here the muscovite flake is formed of alternating laminae of low birefringent kaolinite and relatively high bireferengent muscovite (Plate 27, Fig. 2). The term "accordion" is used after Millot (1970).

The kaolinitization front usually moves in a direction normal to the cleavage plane of the muscovite. The various stages of kaolinitization of the muscovite, from fresh muscovite to completely kaolinitized muscovite, can be demonstrated in the same sample. In the case where a muscovite flake has been crushed into a number of minute sheaves and squashed in a pore space, complete kaolinitization will alter these sheaves to form an aggregate of vermicular kaolinite very similar to the authigenic kaolinite (Plate 27, Fig. 3).

(ii) Illitization of muscovite:

Illite, which is characterized by a bireferengence higher than that of kaolinite and lower than that of muscovite, is commonly developed along the cleavage planes of the muscovite flakes. Illite is usually present as very fine aggregates. It was found that illite is generally present in the kaolinitized flakes. This suggested that illite has been developed on the expanse of the kaolinite formed by the alteration of the muscovite.

(c) Alteration of biotite:

Biotite flakes are commonly altered to brownish ferrugenous matter. Alteration of biotite is usually preferential to the fractures and the cleavage planes. Complete alteration of some fine biotite flakes has been recognised. The early stage of alteration can be demonstrated in some flakes where ferrugenous globules have been seen to occur along the cleavage planes.

(d) Alteration of siderite:

This process is very well displayed in the weathered litharenite. In these rocks siderite was found to be partially altered to a brownish ferrugenous matter. It was found that coarse crystalline rhombohedral siderite crystals are more susceptible to alteration than the microcrystalline siderite forming the siderite nodules.

ORIGIN AND CONDITIONS OF FORMATION OF MINERALS

This part deals with the study of the origin and the conditions of formation of the authigenic minerals present in the terrigenous rocks of the Millstone Grit and the Coal Measures. The following authigenic minerals have been identified; quartz, siderite, dolomite, kaolinite, illite, pyrite and francolite.

QUARTZ:

The study of the dissolution and precipitation of silica have been a cause of intensive studies carried out by many authors, amongst whom are Roy (1945), Kennedy (1950), Alexander et al. (1954), Krauskopf (1956, 1959), Siever (1957, 1959), Dapples (1959) and Thomson (1959).

It was evident that the dissolution and precipitation of silica is governed by three main factors, namely, temperature, pH and EH values.

Siever (1959) concluded from the results of a study of silica cementation in some Pennsylvanian sandstones that temperature was an important factor in the quartz-calcite relationship. With an increase in depth of burial, carbonates are less soluble and commence to precipitate, replacing silica which is more soluble at higher temperature, provided the CO_2 pressure remains constant.

Laboratory experiments undertaken by Maxwell (1964) shows that the solubility of quartz in water increases with an increase in temperature and pressure. Silica was found to be dissolved in water and forms a true solution (H_4SiO_4) in concentrations up to 120 to 140 ppm. at room temperature (Degens, 1965). Correns (1941) in his geochemical studies of silica showed that the solubility of quartz increased with an increase in pH. He assumed that with an increase in pH of the migrating fluids, quartz would go into solution.

Alexander et al. (1954), Krauskopf (1956) and Okamoto et al. (1957) have studied the effect of pH on the solubility of mica. It was found that in some approximation the pH between 0 and 9 has no effect on the solubility, but the solubility increases sharply as the pH rises above 9 because of the ionisation of H_4SiO_4. From the previous review it is concluded that silica is precipitated in acidic environment at low temperatures.

In the case of the studied rocks, it is most likely that authigenic quartz, especially the quartz overgrowths, have been precipitated in the very early stage of diagenesis from acidic water. This is because in most of the quartz-arenites of the Millstone Grit and the Coal Measures, authigenic quartz has been precipitated while the detrital quartz grains were still in tangential contact, i.e. before any pressure solution took place. It was found also that the original texture of the sediments has played an active role in the quartz diagenesis. So in the well sorted clean quartzitic sediments quartz precipitation was found to be facilitated by the high porosity which helps in the circulation of the silica-rich solution, and by the occurrence of clean quartz grains which have acted as seeds for the precipitation of the quartz overgrowths.

Origin of silica:

Many authors, including Waldschmidt (1941), Heald (1956), Thomson (1959) and Füchtbauer (1967) considered that silica was derived by pressure solutions, whilst others such as Goldstein (1948) invoked the solution of fine quartz grains as a possible source. However, Siever (1959) was of the opinion that ground water contributed to much of the silica found in sandstones. Von Engelhardt (1967) considered that silica dissolved at depth could subsequently be transported upwards either by fluid movement or diffusion to be precipitated as quartz near the surface where lower temperature existed. Vatan (1962) suggested that silica could be derived from the transformation of hydromica to kaolinite; he calculated that 20% of silica held in hydromica could be released during this process.

In the sandstones and siltstones of the Coal Measures and the Millstone Grit, two stages of silica cementation have been recognised, namely, an early stage of cementation represented by the quartz overgrowths, and a late stage of cementation manifested by the microcrystalline quartz. It is suggested that the early stage of silica precipitation has taken place by the precipitation of silica from the surface water or very shallow underground water which was rich in silica. The silica in this water could be derived from the silica released during the weathering of the detrital micas.

Silica precipitated in the late stage of cementation could be derived from two sources; (a) from the silica released during the transformation of the muscovite flakes present in the rock to clay minerals. This process most probably took place while the rock was porous and before complete cementation of the rock. It is suggested that this autochtonous silica has been precipitated in the nearby pore spaces as microcrystalline quartz; (b) During the dolomitization and sideritization of the sandstones and siltstones considerable amounts of silica were released and migrated to precipitate in places attaining conditions of low pH and low temperature and pressure.

SIDERITE:

Siderite in the various lithotypes of the Millstone Grit and the Coal Measures is generally present as chemically precipitated filling pore spaces, and as diagenetic replacive mineral. The latter is the most frequent type of siderite. The genesis of the siderite as well as the other iron minerals in the Coal Measures of the Pennine Coal Fields has been a subject of detailed study by many authors, while that of the South Wales Coalfield has not received such interest.

According to Curtis and Spears (1968) five factors control the formation of siderite, namely; Eh, pH, activity of dissolved HS^-, activity of dissolved HCO_3^-, and activity of dissolved Fe^{2+}. It was found that from the stability field diagrams the optimum chemical environment for the siderite appears to be one with zero sulphide activity, low Eh (-0.25 to -0.35 volts) and severely restricted circulation, and pH ranging from 7 to 8. They considered that siderite can only attain equilibrium with sediment pure waters, so it is only stable therefore within sediment masses. However, it is suggested that siderite has been initially precipitated in the pore spaces in the sediments below the water/sediment interface where the conditions suitable for the formation of siderite prevailed.

Dunham (1960) has discussed the possible sources of iron necessary for the formation of siderite. It has been supposed by a majority of writers that the iron is derived from terrestrial weathering products and transported by rivers. This suggestion is chemically feasible, for average river water, as determined by Clarke (1924), has a salinity of 146 ppm. of which $Fe_2O_3 + Al_2O_3$ amounts to 4.02 ppm. Against this view a very different concept has been developed by Bachet (1952) by which the iron is derived from the sediments on the sea floor in regions of stagnant bottom conditions and transported by current action.

Curtis and Spears (1968) have suggested that only ferric compounds can precipitate from normal depositional water. They stated that where, for any reason, sulphide activity in the diagenetic environment is very low, a biotic reaction between organic matter and precipitated ferric compounds would be likely to create conditions favourable for siderite formation:

$$4Fe(OH)_3 + 3C_{org} \longrightarrow 3CO_2 + 6H_2O + 4Fe^{2+} - 4e$$

Since chemically precipitated siderite in the studied rock is usually found to be associated with the organic matter debris, and is rarely present in organic matter free rocks, therefore the mechanism suggested by

Curtis and Spears (1968) is most likely to be responsible for the development of this siderite.

The abundance of siderite in the argillaceous rocks (shales and claystones) more than in the coarser-grained rocks, is due to the fact that the physio-chemical conditions within the argillaceous sediment bodies could be more firmly separated from the water body above the water/sediment interface. Where in the case of the coarse-grained sediments, because they are usually very porous, so the separation between the pore water in the sediments and the water body above the sediment/water interface is impossible. This is assuming that the physio-chemical conditions of the water body were not suitable for the formation of the siderite.

The second stage of siderite formation is the development of the diagenetic siderite. This stage has taken place just after a shallow depth of burial. Two mechanisms have been suggested for the formation of this siderite, namely, by displacement and by replacement.

After shallow burial and before the completion of the compaction of the argillaceous sediments, the iron-rich solutions were segregated and siderite nodules have been formed. These nodules progressively grow pushing the clay matrix aside forming a texture similar to the agglomeroporphyritic texture of the volcanic rocks, and usually the micaceous clay flakes have a flow texture around these nodules. In late diagenetic stages and after the sediments have reached their maximum compaction, segregated siderite was unable to displace the clayey mass, therefore siderite started to replace the rock constituents.

The geometry of the replacive siderite bodies is governed by the original rock texture and composition. In the case of sandstones and siltstones, siderite usually starts to replace the clayey matrix or the quartz cement and then progressively replaces the framework grains. In these rocks replacive siderite is commonly present as discontinuous bands.

In the shales and where the micaceous clay flakes are oriented parallel to the bedding, it was found that the rate of the replacement of the clay flakes by siderite in the direction parallel to the bedding is faster than in the direction normal to the bedding. This resulted in the development of elongated nodules with their long dimension parallel to the bedding.

In the underclays in which the clay flakes are usually randomly oriented, spherulitic siderite is the frequent form of the replacive siderite. This is because the rate of growth of the siderite crystals was equal in all directions. Deans (1934) has suggested that the siderite spherulites have started to develop as aggregates of fine needle-shaped crystals. As is to be expected in siderite, the elongation of these crystals occurs in the direction of the optic axis. At the centre of crystallization the minute crystals will at first be directed in all directions, but as growth proceeds only those crystals whose optic axes are directed radially outwards can continue to grow, and so the radial structure develops around a cryptocrystalline nucleus. This mechanism could be responsible for the development of some of the siderite spherulites in the underclays.

Taylor and Spears (1967) suggested that ankerite and siderite-forming carbonate bands in the Coal Measures of the East Pennine coalfields have been formed diagenetically by the alteration of originally precipitated calcite. Such phenomenon has not been recognised in the diagenetic siderite in the studied rocks.

DOLOMITE:

Ferroan dolomite is very frequent as a replacement mineral and is rarely present as chemically precipitated pore filling. Chilinger, Bissell and Wolf (1967) suggested that diagenetic dolomitization occurs in strongly reducing to weakly reducing environment having high alkinity. Chilinger and Bissel (1963) assigned a pH value of 8 to the environment necessary, in their evaluation, for the formation of primary sedimentary dolomites.

In the sandstones and the siltstones of the Coal Measures, ferroan dolomite has started to precipitate as fine dolomite rhombs before and after the development of the secondary quartz overgrowths. Ferroan dolomite has been starting to develop replacing firstly the clay matrix and then the detrital quartz grains. The precipitation of dolomite as ferroan dolomite indicates that the mineral has developed in a reducing environment (Evamy, 1969).

Magnesium and iron necessary for the formation of the ferroan dolomite are most probably derived from the clay minerals during their weathering.

PYRITE:

Pyrite is frequently present in the marine shales and in some quartzarenites of the Coal Measures Series. It is present as idiotopic crystals and as framboidal aggregates.

Berner (1963 and 1964.a, b, c, d) from his studies on pyrite development in Recent sediments, concluded that within a sediment profile, the concentration of sulphide species increases with depth from some point below the sediment/water interface at the expense of SO_4^{2-}. The sulphide species are generated by sulphate reducing bacteria. The rate of generation depends on the availability of utilizable organic matter and on the rate of diffusion of SO_4^{2-} down from the overlying waters.

Love (1967) in his study on the iron sulphide in Recent sediments of the Wash, England found that pyrite as an early diagenetic mineral is present as single isolated grains and framboidal spherules formed by the conversion of ferric grains to mono-sulphide and then to pyrite.

Curtis (1967) in his study on some British Coal Measures Shales found that pyrite is predominant in marine shales whereas siderite is frequent in the non-marine shales.

In the studied rocks it was found that pyrite is commonly frequent in the marine shales which are rich in organic matter. It is more likely that the pyrite has been formed in the same way by which the Recent pyrite of the Wash has been formed (Love, 1967). Intercalation of pyrite rich shales and siderite rich shales is thought to be due to the fall-off in available sulphate which is itself a reflection of change in the salinity of the environment of deposition (see Taylor, 1971).

KAOLINITE:

Besides its detrital origins, kaolinite has been recognised to occur as authigenic vermicular aggregates present frequently as pore filling in the quartzarenites, and as alteration product of the mica minerals. Degradation of mica minerals to kaolinite and illite is well displayed in most of the Coal Measures lithotypes. This feature has been described by Millot (1970).

In the rocks under investigation, the development of both kaolinite and illite from mica is thought to be a result of leaching process. However, Millot (1970) maintained that the illite fans found in mica have resulted from the fixation of K^+ during a later stage of diagenesis and refers to this process as Illitization of Kaolinite.

It is considered that soon after burial, hydrolysis becomes active due to the passage of fresh water through the porous and permeable rocks, resulting in kaolinitization of the mica flakes. In the argillaceous rocks the organic matter could have played an important role in the kaolinitization of micas. Release of CO_2 during the decaying of the organic matter decreases the pH value of the interstitial water, which facilitates the leaching process of mica and therefore their kaolinitization.

In the case of vermicular kaolinite filling the pore spaces in the quartzarenites, there is no evidence to suggest that this kaolinite is formed by kaolinitization of mica. Its occurrence as fracture-filling cement and its fabric in the pore spaces suggested that this kaolinite was formed by direct precipitation from supersaturated solutions.

Bucke and Mankin (1971) have reported the occurrence of similar kaolinite in the sandstone beds of the Desmoinesian deltaic strata in Oklahoma, and they suggested that this kaolinite has been precipitated from the pore solution.

Shelton (1964) suggested three possible origins of the authigenic kaolinite in sandstones, namely, by crystallization from solution, by alteration in place of some parent minerals, and by recrystallization of fine-grained detrital kaolinites. Carrigy and Mellon (1964) have described the authigenic kaolinite in the Cretaceous and Tertiary sandstones of Alberta and they suggested that kaolinite has been precipitated from alminia-rich solutions expelled from adjacent shaley strata.

Wilson et al. (1972) have reported the presence of authigenic vermicular kaolinite in the sandstones of the Coal Measures of Scotland and suggested that this kaolinite could be formed by recrystallization of clay particles during diagenesis.

FRANCOLITE:

Francolite is present in small amounts in the shales and occasionally in the siltstones. Francolite has been recognised in ironstones in the Coal Measures rocks near Leeds by Deans (1938).

Contrary views relating to the mode of formation of francolite are apparent in the literature. Ames (1959) summarised the different viewpoints with respect to the mineralogical and geochemical aspects of francolite. These views can be grouped in three theories. The first theory is a chemical precipitation formed upon supersaturation of natural waters (Kazakov, 1937, 1950 and McKelvey et al., 1953). The second theory considers francolite as a biochemical product formed by the action of microorganisms (Cayeux, 1936). The third theory suggested that francolite is a simple replacement product caused by the introduction of PO_4 units into pre-existing calcareous materials (Bushinsky, 1935, 1964 and Ames, 1959).

At first sight, a detrital origin could be postulated for the francolite in the studied rocks, but the fact that the mineral is present as idiotopic and hypidiotopic crystals besides its presence as fracture-filling cement in some siderite nodules is not really consistent with such genesis.

The balance of evidence is therefore in favour of the francolites having been formed by direct precipitation from supersaturated pore solution. The phosphorus necessary for the formation of francolite might be derived from the plant tissue and the fish bone remains.

PLATE 22

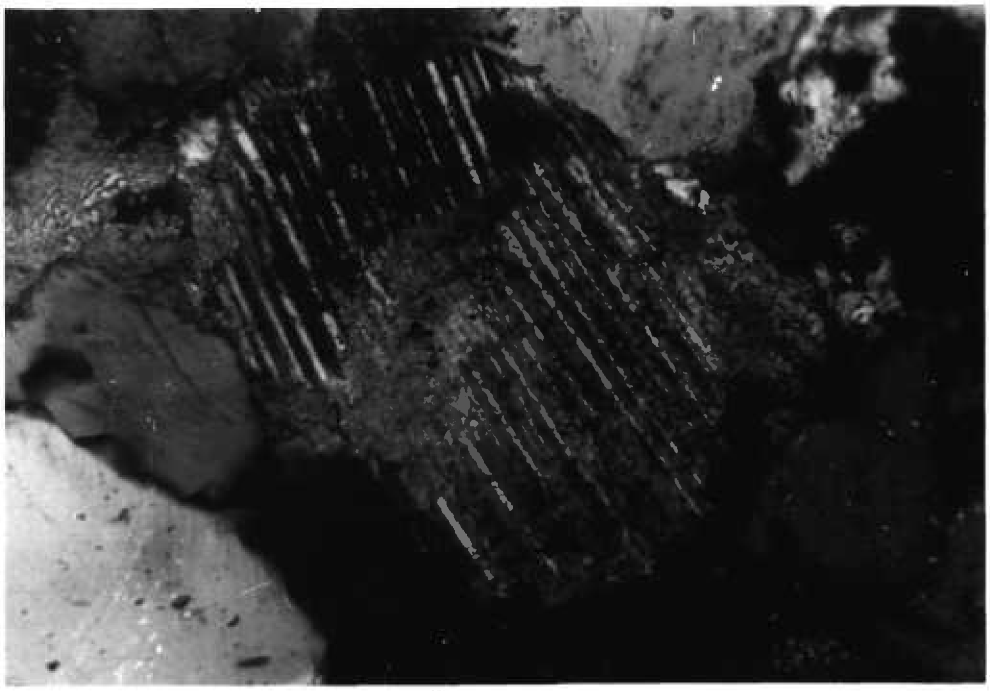
Fig. 1 Fractured plagioclase grain showing displacement of twin planes. Feldspathic sandstone. Sample 574, crossed polars.

Fig. 2 Argillaceous rock fragment (AF) has flowed between quartz (Q) and feldspar grains (F). Note the stylolitic contact between a feldspar and a quartz grain (arrow). Sample 729.

Fig. 3 Bent muscovite flake (M) in lithic arenite of the Upper Coal Measures. Note the wavey extinction of the flake. Sample 316. Crossed polars.

Fig. 4 Bent and fractured muscovite flake (arrow), trapped between quartz grains (Q) in coarse-grained area within siltstone. Sample 276. Crossed polars.

Fig. 5 Squashed muscovite flake in quartzarenite. Sample 760A. Crossed polars.



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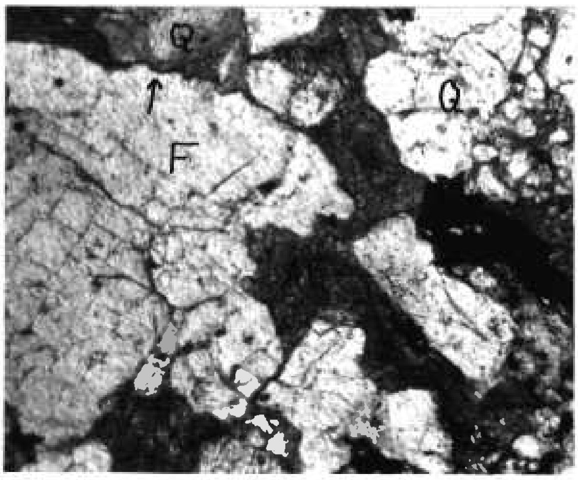

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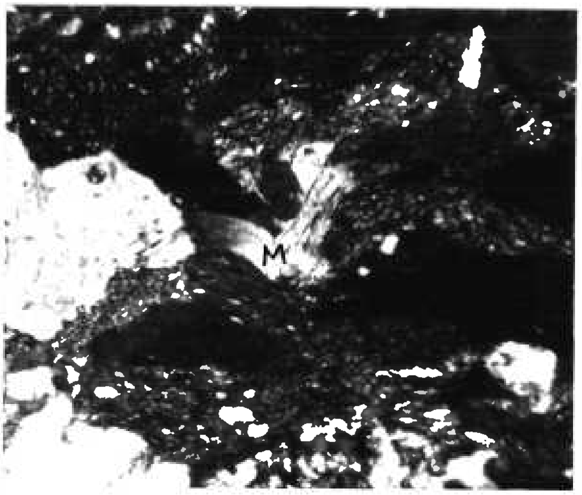

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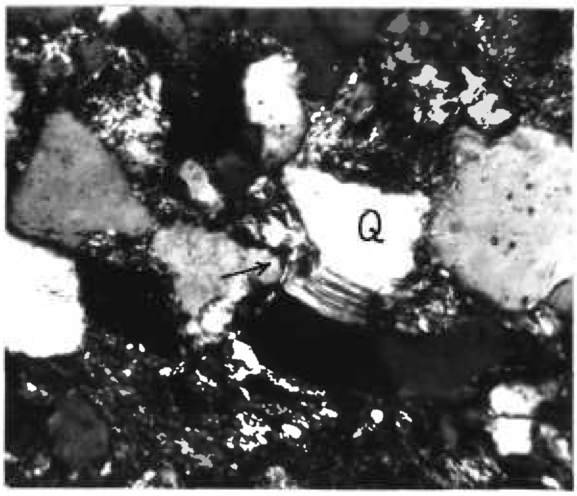

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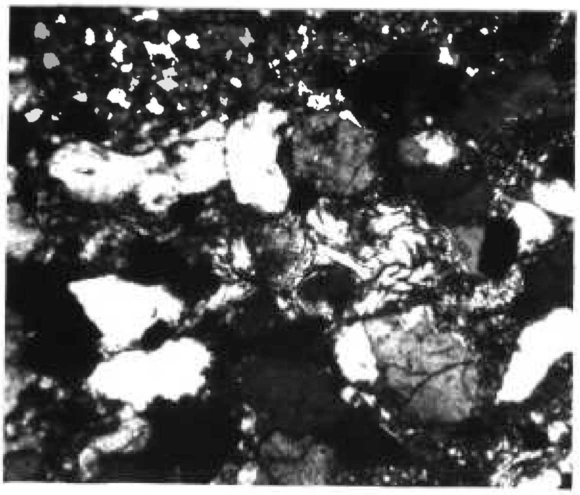

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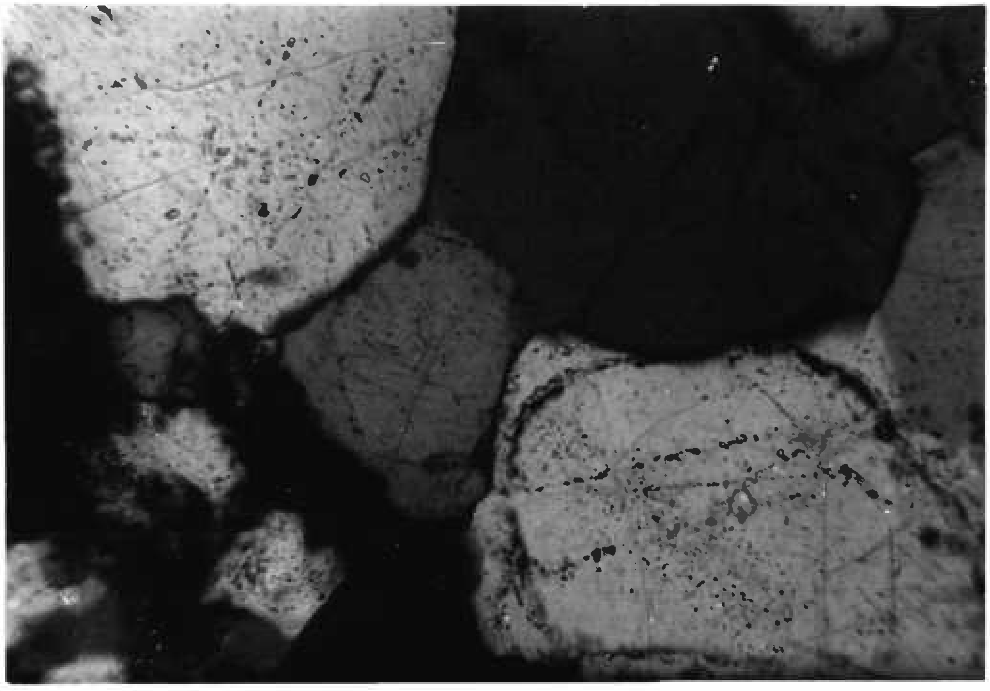
PLATE 23

Fig. 1 Quartz overgrowths. Quartzarenite of the Lower Coal Measures. Sample 187. Crossed polars.

Fig. 2 Microcrystalline quartz (MQ), kaolinite (K) and idiomorphic dolomite (D) filling a pore space in fine-grained quartzarenite. Sample 335. Crossed polars.

Fig. 3 Spherulitic chalcedony filling a pore space in quartzarenite. Sample 911. Crossed polars.

Fig. 4 Dolomite veinlet in fine-grained quartzarenite. Note the irregular boundaries of the veinlet. Sample 149.



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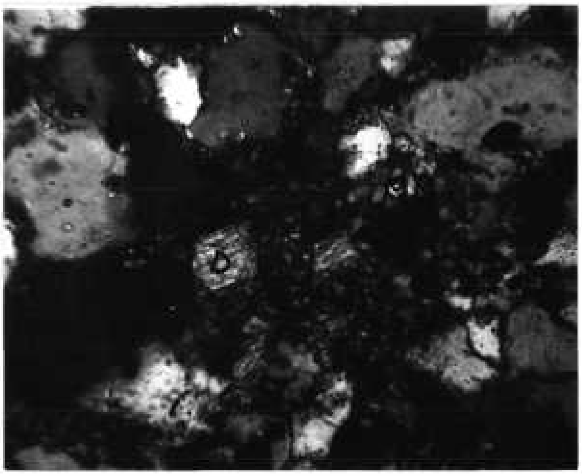

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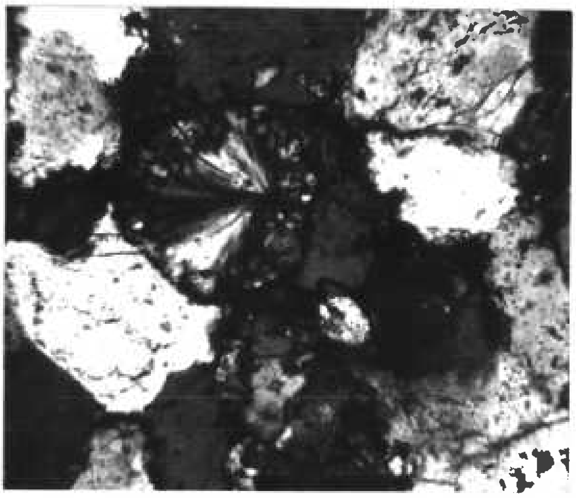

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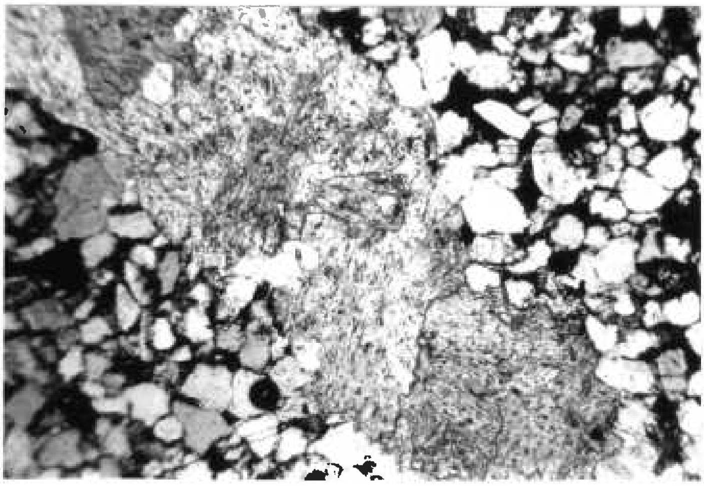

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PLATE 24

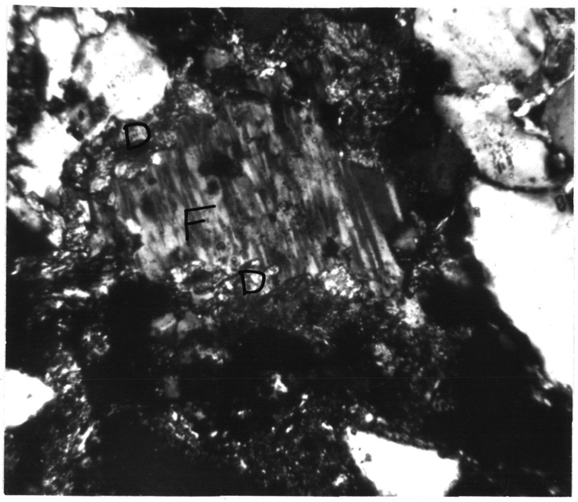
Fig. 1 Corrosion and replacement of feldspar grain (F) by dolomite (D). Sample 307. Crossed polars.

Fig. 2 Open framework texture produced by partial replacement of the quartz grains by dolomite. Note the poikilotopic fabric. Sample 870. Crossed polars.

Fig. 3 Replacement of quartz grains and cement by coarsely crystalline dolomite. Note remnants of quartz grains floating in the dolomite crystals forming poikilotopic texture (arrow). Sample 355. Crossed polars.

Fig. 4 Open framework texture in lithic arenite produced by partial replacement of the framework grains by microcrystalline siderite (S). Sample 733.

Figs. 5 & 6 Siderite filling the cellular cavities in plant fragments. Sample 175. Fig. 6 between crossed polars.



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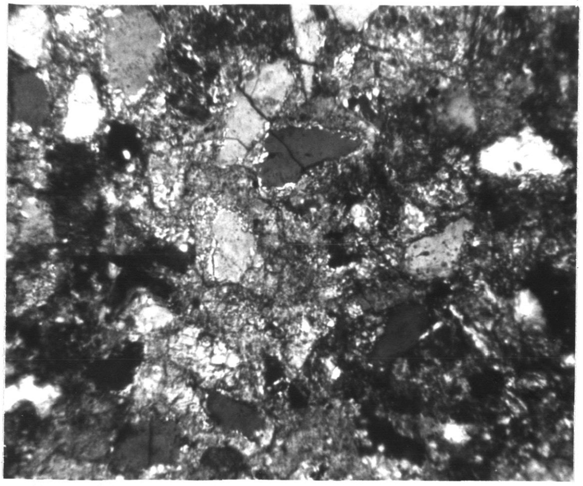

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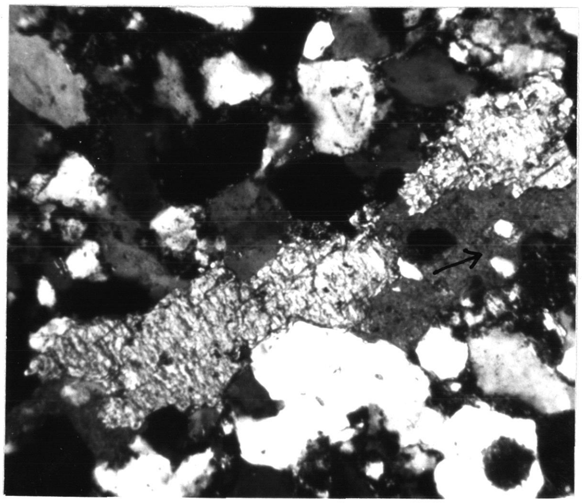

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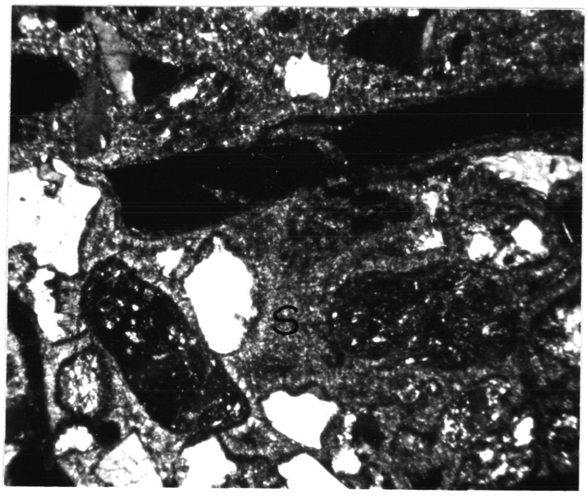

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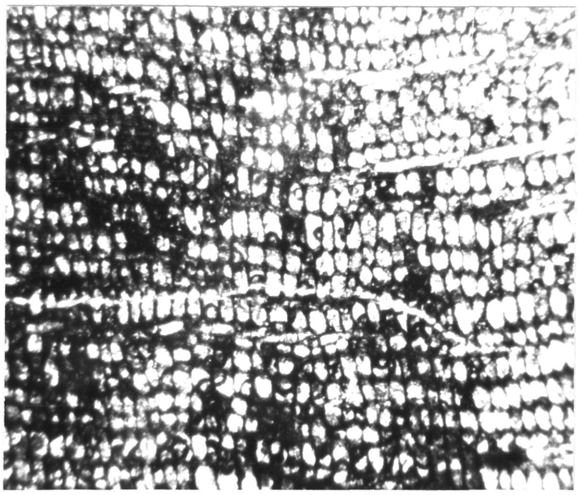

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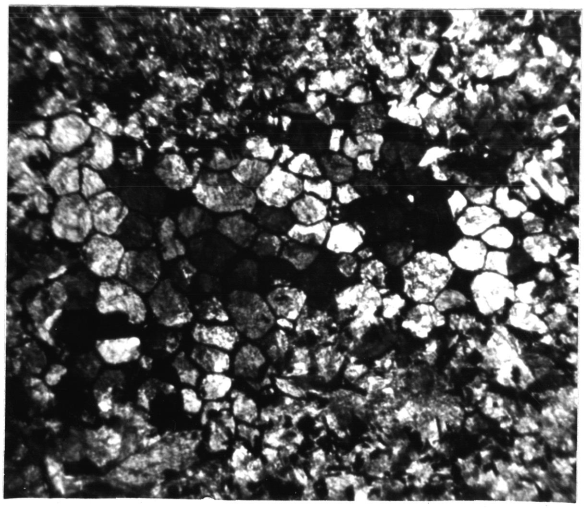

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PLATE 25

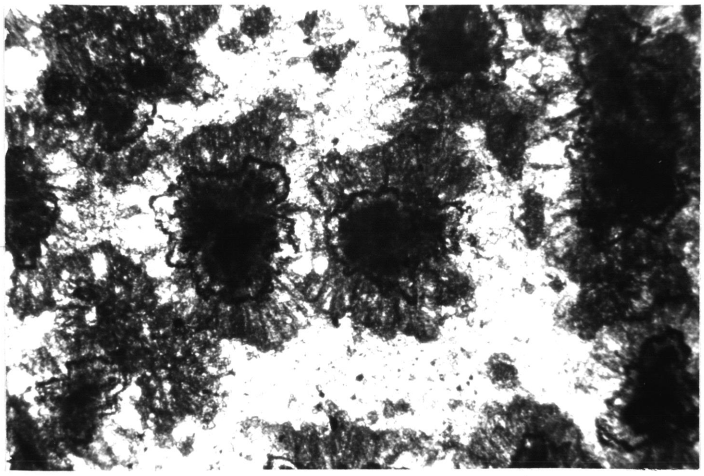
Fig. 1 Spherulitic siderite replacing the clayey groundmass of an underclay rock. Note the organic matter envelope enclosed in the prismatic siderite crystals. Sample 420-5.

Fig. 2 Spherosiderite (S) replacing a clayey siltstone rock. A fracture filled with micricrystalline quartz (mQ) and prismatic crystals of francolite (Fr). Sample 791. Crossed polars.

Fig. 3 Sphaerosiderite nodules encrust a plant rootlet in an underclay rock. Sample 863.

Fig. 4 Subhedral crystal of francolite (Fr) in shale. Sample 805. Crossed polars.

Fig. 5 Euhedral pyrite (P) replacing quartz in a quartzarenite. Note squeezing of a mica flake between the quartz grains (arrow). Sample 760A.



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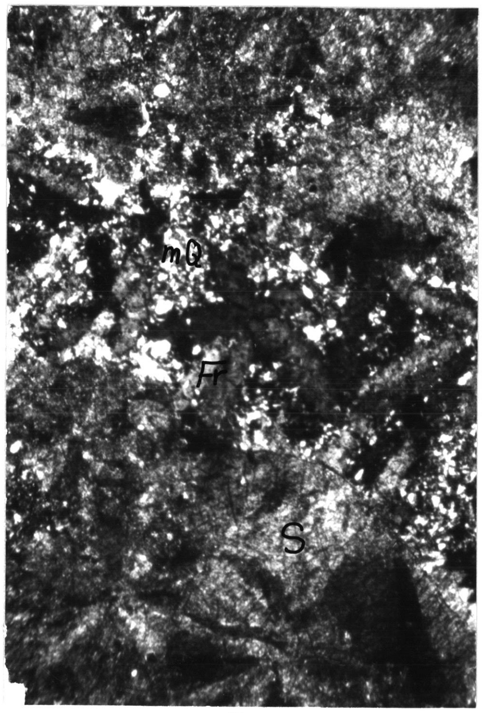

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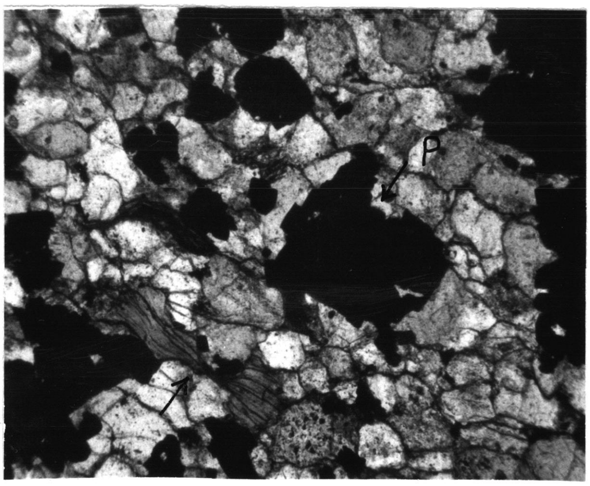

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PLATE 26

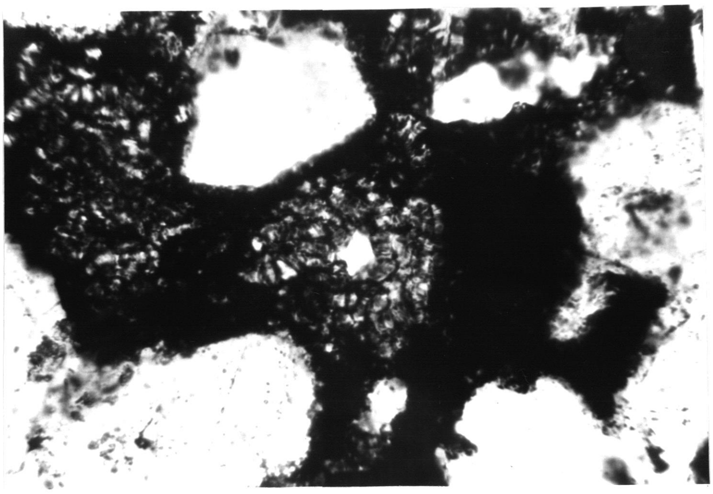
Fig. 1 Coarsely crystalline vermicular kaolinite filling the pore spaces in a quartzarenite rich in organic matter. Sample 179.

Fig. 2 Replacement of clay matrix (C) by microcrystalline quartz (mQ) in a quartz wacke stone. Sample 170. Crossed polars.

Fig. 3 Vermicular kaolinite partially replacing quartz grains and quartz overgrowths (arrows). Sample 94. Crossed polars.

Fig. 4 Shredded muscovite flakes due to the force of crystallization of siderite. Lithic arenite. Sample 733. Crossed polars.

Fig. 5 Displacement of fibrous siderite rimming a coaly fragment by the force of crystallization of quartz (arrow). Sample 715. Crossed polars.



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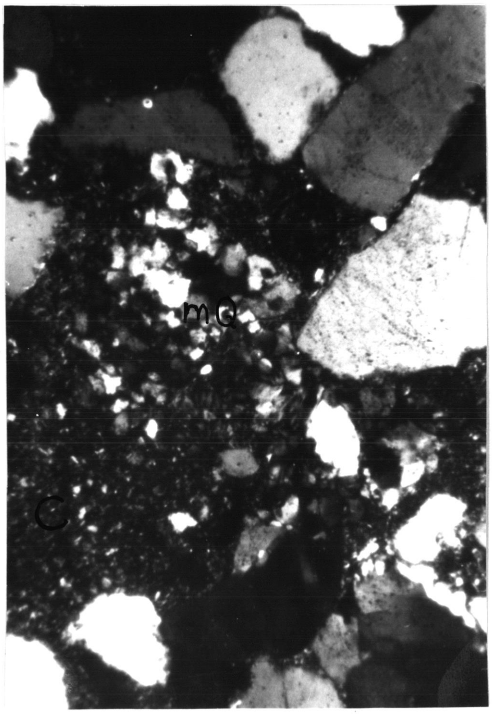

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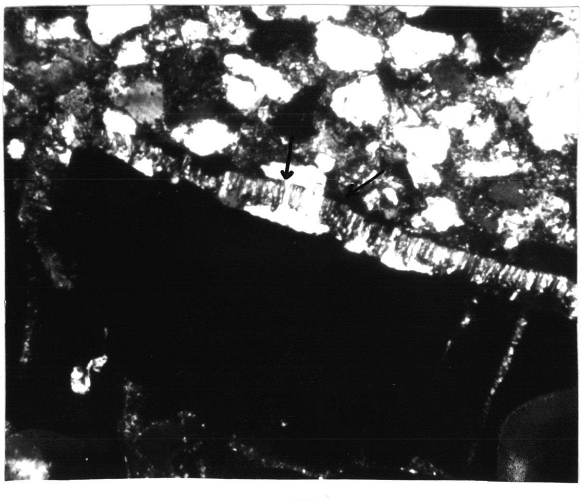

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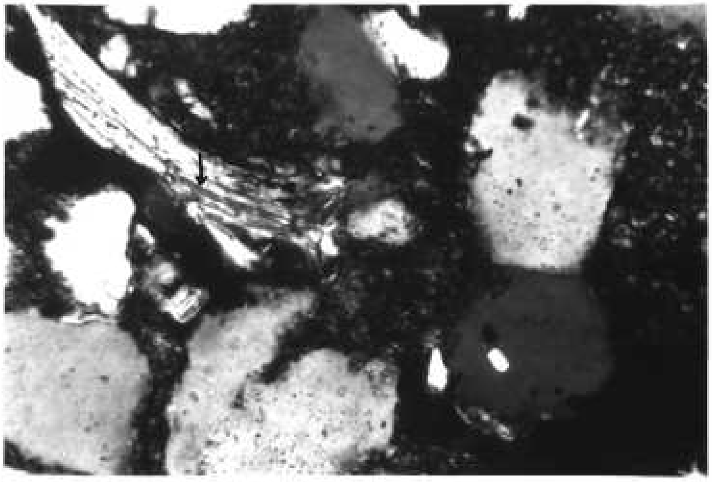
PLATE 27

Fig. 1 Kaolinitization of a muscovite flake in a quartz wackestone. Note the preferential kaolinitization of the muscovite flake on the cleavage plane (arrow), the frayed edge of the flake is highly kaolinitized. Sample 170. Crossed polars.

Fig. 2 Kaolinitized squashed muscovite flake. Sample 276. Crossed polars.

Fig. 3 A severely squashed muscovite flake, almost completely kaolinitized and filling a pore space in a quartzarenite. Sample 141. Crossed polars.



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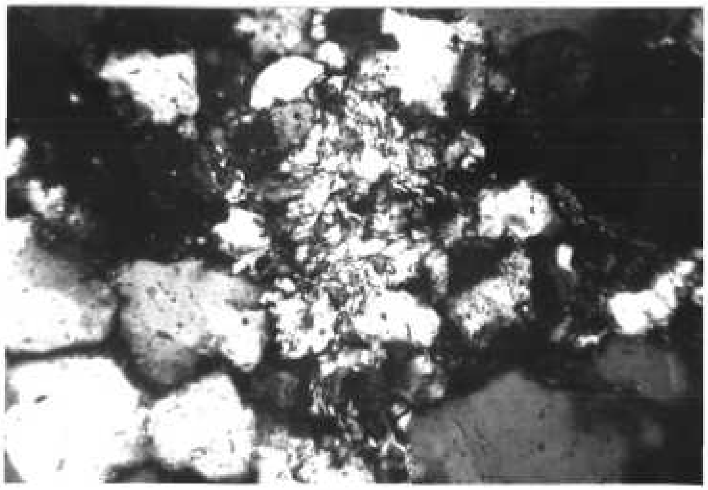

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CHAPTER VI

INCIPIENT METAMORPHIC FABRICS OF THE CARBONATE AND THE TERRIGENOUS ROCKS

INCIPIENT METAMORPHIC FABRICS OF THE CARBONATE AND THE TERRIGENOUS ROCKS

INTRODUCTION:

This chapter deals with the study of the fabrics and textures of the incipiently metamorphosed carboniferous rocks of the South Wales Coalfield. Incipiently metamorphosed rocks have been recognised in the western part of the Coalfield, i.e. in the anthracite area. This chapter consists of two parts, the first part deals with the description of the fabrics and textures characteristic of the carbonate rocks, and the second part deals with the fabrics and the textures characteristic of the non-carbonate rocks.

INCIPIENT METAMORPHIC FABRICS OF THE CARBONATE ROCKS

As yet, little has been published on the fabric changes that can occur in carbonate rocks within the incipient metamorphic stage. Wardlaw (1962) described fabric alterations in some Irish carbonates that had been deformed. He emphasised recrystallization or crystal diminution that resulted in grain. Brown (1972) described fabric alteration in incipiently metamorphosed mud-supported carbonate rocks in parts of the Central Appalachians. He found that the fine-grained matrix is progressively getting coarser by recrystallization.

The following are the most important fabrics characterising the incipiently metamorphosed carbonate rocks of the South Wales Coalfield: (A) recrystallization fabrics; (B) fabrics due to deformation.

A. RECRYSTALLIZATION:

Definition:

The definition of the recrystallization process was the subject of controversy between many carbonate petrologists. Folk (1965) has reviewed most of these ideas. The term "neomorphism" has been introduced by Folk (1965) to include two processes, namely, inversion and recrystallization. The first embodies all of the polymorphic transformation, such as transformation of aragonite to low-Mg calcite, and high-Mg calcite to low-Mg calcite. He defined the recrystallization as such process by which formerly-found calcite is replaced by calcite of different grain size, morphology and orientation.

Bathurst (1958) attempts to explain some of the coarser recrystallization fabrics found in true stone by analogy with process taking place during the annealing of work-hardened metals. But later in 1964 he shows that the analogy was in error as it dealt with rocks showing no metamorphism. Folk (1965) pointed out that the process occurring in metals should have little in common with those taking place in porous, fluid-saturated carbonates, which are initially composed of metastable minerals such as aragonite and high-Mg calcite. The latter idea of Folk is applicable only in the early stages of diagenesis, but eventually carbonate rocks may lose their effective porosity and most of their interstitial fluids, and being formed essentially of low-Mg calcite, so they are mainly monomineralic and non-porous.

Wardlaw (1962) has defined recrystallization as the replacement of physically unstable grains by grains of the same mineral which are less strained or more stable.

Voll (1960) suggested that the free energy of deformation is the main deriving force of recrystallization. Griggas et al. (1960) in their detailed experimental work on annealing recrystallization in calcite crystals and aggregates have concluded that single crystals and aggregates of calcite recrystallized when annealed at high temperature. They suggested that the driving force for recrystallization is the internal strain energy of crystals and/or the grain boundary energy. There is a critical temperature below which recrystallization does not occur, this critical temperature is thought to be dependent on the difference in free energy between the initial and recrystallized state.

They also concluded that an increase in strain prior to annealing decreases the critical temperature. A decrease in grain size of unstrained aggregates also decreases the critical temperature for annealing recrystallization. They found that Solenhofen Limestones recrystallized at 800°C without prior strain.

In the present study, recrystallization is defined as such process which takes place in rock, commonly after complete lithification, due to the effect of temperature and pressure and resulted in the transformation of the original rock fabrics to a newly-formed fabric stable under the new conditions of temperature and pressure.

Recognition of the recrystallized calcite:

One of the most difficult problems in the identification of the incipiently metamorphosed carbonate rocks is the differentiation between the diagenetic sparry cement and the recrystallized pseudosparry calcite. The recognition of the recrystallized calcite was based mainly on Stauffer's Criteria (1962), summarized in the following points:

- Irregular grain size of crystal mosaic with no systematic variation.
- Truncation by crystal mosaic of previously existing structures, such as laminae of oolites

and fossil structures.

- Ghosts of allochems in crystal mosaic.
- Embayments by calcite crystals which may have plane sides, into microcrystalline calcite or allochems.
- Curved serrated or interlocking intergranular boundaries between crystals.

Besides the above-mentioned criteris, there are a number of others that would suggest that a crystal mosaic is recrystallized calcite:

- Loosely packed allochems.
- Poorly sorted allochems.
- Angular allochems.
- Gradational boundaries between calcite mosaic and microcrystalline calcite.
- Calcite crystal size never smaller than 4μ and generally larger than 7μ.
- Irregular patches of crystal mosaic in microcrystalline calcite.
- Residual, irregular, floating patches of microcrystalline calcite in the middle of a coarser calcite mosaic.

Recrystallization fabrics:

In the recrystallized carbonate rocks of the Carboniferous Limestone Series of the South Wales Coalfield two types of recrystallization fabric have been recognised, namely, aggrading recrystallization fabrics and degrading recrystallization fabrics.

1. Aggrading recrystallization:

This is the process by which fine crystal mosaics are transformed to coarser crystal mosaic of the same mineral. This process is described in detail by Folk (1965). Bathurst (1958) has used the term "grain growth" for the same process. The term "pseudospar" has been introduced by Folk (1965) to cover the sparry calcite formed by neomorphism which is inversion or recrystallization in solid state in the presence of hybrid films. In the present study the term pseudospar will be used to indicate sparry calcite formed by recrystallization only.

In the carbonate rocks of the Carboniferous Limestone Series in the anthracite area of South Wales, aggrading recrystallization has been recognised affecting the micrite matrix, the allochems, and the calcite cement filling crumbly fractures.

a. Aggrading recrystallization of the micrite matrix:

Recrystallization of the micrite has been frequently recognised in the micrite rocks and the other micrite lithotypes in the anthracite area of South Wales. It was found that the dismicrite is the least susceptible rock to recrystallization. In these rocks, recrystallization is confined to its effect on the geopetal micrite which partially fills the vugs. This micrite has been recrystallized to micropseudospar. Occasionally, irregular patches of micropseudospar mosaic is present within the micrite groundmass. They usually have aggradational boundaries with the groundmass micrite.

Biomicrites, especially the foraminiferal biomicrites, were found to be the most susceptible rocks for recrystallization. These rocks are generally formed of fine-grained allochems floating in a groundmass consisting of pseudospar mosaic. Remnant micrite patches are frequently present in the pseudosparite groundmass. Pseudosparry calcite crystals are commonly ill-sorted and have irregular boundaries. They range in size from 10μ to 150μ. It was found that increase in the grade of incipient metamorphism is manifested in the micrite matrix by increasing the crystal size and the sorting of the pseudosparry calcite crystals (Plate 28). Micrite in the micrite envelopes and the algal grains is found to be very resistant to recrystallization.

In the poorly-washed oolitic biosparites, it was found that micrite patches have been completely recrystallized to pseudospar mosaic. Pseudosparry calcite crystaly commonly display lamellar twinning.

b. Aggrading recrystallization of the allochems:

In some biomicrites and poorly washed oosparites, partial and complete recrystallization of some of the allochems has been recognised. In foraminiferal biomicrites where the micrite matrix is almost completely recrystallized, complete recrystallization of foraminiferal tests has been recognised. The original shape of the recrystallized foraminiferal tests are preserved as ghosts floating in coarsely crystalline pseudosparite mosaic (Plate 29, Fig. 3).

In the poorly washed oolitic biosparites, partial and complete recrystallization of the oolites have been observed. In these rocks the micrite or microsparite forming the oolites have been partially or completely recrystallized to pseudosparry calcite mosaic, in which the crystals range in size from 10μ to 85μ. In the case where the oolite consists of a coarse grained crinoidal debris covered by thin oolite shells of about 100μ thick, the micrite forming the oolite shell has been recrystallized to a mosaic of bladed calcite crystals (Plate 29, Fig. 4). In the case of the nucleus free oolites, the whole oolite has been recrystallized to a mosaic of equigranular pseudosparite crystals (Plate 29, Fig. 5). The original shape of the completely recrystallized oolites is commonly preserved as ghosts.

c. Aggrading recrystallization of fracture-filling cement:

In some of the micritic rocks, the fracture-filling cement which usually consists of a mosaic of equicrystalline sparry crystals, has been recrystallized to a mosaic of pseudosparry calcite crystals (Plate 31, Fig. 1). The recrystallized sparry calcite crystals are characterised by irregular crystal boundaries. They commonly have a strained fabric, displayed by the brush extinction and the abundance of lamellar twinning. The fractures have an irregular boundary which could be due to the outward growth of the pseudosparite replacing the groundmass micrite. This type of fracture-filling cement has been called "recrystallization veinlets" by Misik (1968, p. 132).

2. Degrading recrystallization:

This is the process by which coarse-grained calcite crystal is replaced by a number of unstrained fine-grained calcite crystals. In the case of a recrystallization of the coarse-grained calcite crystals, nuclei of strain-free grains originate at several points, producing a polycrystalline texture instead of the original monocrystalline texture.

Attention was drawn to textures of this type by Voll (1960) who recorded that in low temperatures recrystallization of limestone even after light straining, numerous nuclei have been formed within the coarse-grained calcite crystals. Wardlaw (1962) has described this process in the Irish Carboniferous limestones. He suggests that this could be due to minor chemical differences between fine-grained aggregates and the first rock and that the replacement has the nature of a chemical reaction, for which the energies of reaction are the driving force for the recrystallization. Orme and Brown (1963) introduced the term "grain diminution" to describe the in situ replacement of a deformed crystal by crypto-crystalline unstrained crystals of the same mineral.

a. Degrading recrystallization of the crinoidal fragments;

Degrading recrystallization is a common feature among the crinoidal biomicrites and crinoidal biosparite of the anthracite area of the South Wales Coalfield. In these rocks degrading recrystallization mainly affects the crinoidal fragments and their syntaxial calcite overgrowths.

The following fabrics have been recognised:

i) Partial recrystallization of the monocrystalline crinoidal debris, represented by the occurrence of minute calcite crystals floating in the central area of a coarse-grained calcite crystal built up of a crinoidal ossicle and its syntaxial overgrowth (Plate 30, Fig. 3).

ii) In some other rocks recrystallization is confined to the crinoidal ossicles without affecting their syntaxial overgrowths. In this case, the monocrystalline crinoidal ossicle has been transformed to a mosaic of pseudosparry calcite crystals which is surrounded by the unaffected monocrystalline syntaxial overgrowths. Pseudosparry crystals are equicrystalline with the characteristic irregular boundaries and are of about 55μ in average size. Cleavage traces of the original monocrystalline calcite are preserved within the pseudosparite mosaic (Plate 30, Fig. 1).

iii) In some biosparite, recrystallization was found to be confined to the syntaxial overgrowths on the crinoidal debris. Here, the crinoidal debris still preserve their original fabric, while the monocrystalline calcite overgrowths have been partially replaced by a mosaic of pseudosparry calcite crystals (Plate 30, Figs. 3 and 4).

b. Degrading recrystallization of the fracture-filling cement:

Calcite veins are frequently present in the North Western outcrops of the Carboniferous limestone of the South Wales Coalfield. These calcite veins are found to reach up to 80 cms. in width. They usually consist of a mosaic of very coarse calcite crystals, which reach up to 20 cms. in size. In thin sections it was found that these calcite crystals have been degradingly recrystallized. Newly-formed unstrained crystals are present as aggregates of equicrystalline crystals of about 130μ in average size, which are usually developed adjacent to the twin lamellae of the host crystal. While twin lamellae are frequently present in the strained host crystal, the newly formed crystals are almost untwinned, which is an indication of their strain freedom (Plate 30, Fig. 5).

Twinning in the strained host crystals is represented by the presence of well developed, thick twin lamellae. Incipient recrystallization of these twin lamellae is demonstrated by the development of newly formed fine crystals within the twin lamellae. This gives the twin lamellae the appearance of having been corroded by the calcite groundmass (Plate 32, Fig.1).

These fabrics have been described by Griggars et al. (1960) in their experimental work. They found that these fabrics have developed in a single calcite crystal strained at low temperature and annealed at 800°C.

B. DEFORMATION:

Deformational fabrics have been frequently observed in the carbonate rocks outcropping in the anthracite area of the South Wales Coalfield. The following deformational fabrics have been recognised: (a) brush extinction; (b) distortion of twin lamellae; and (c) deformed calcite veinlets.

(1) Brush extinction:

Wavey extinction in calcite and dolomite crystals has been commonly seen in the incipiently metamorphosed carboniferous limestone rocks. Brush extinction of the deformed crystals is a product of the dislocation of the lattice boundaries, Voll (1960). Lattice dislocation displayed by brush extinction has been noticed in the strained unrecrystallized crystals as well as in the pseudo-sparry calcite crystals.

In the porphyrotopic dolomites outcropping in the western area of the South Wales Coalfield, coarse-grained dolomite crystals commonly exhibit brush extinction (Plate 32, Figs. 3 and 4). These crystals are characterised by curved cleavage traces. In some partially dolomitized biomicrites curved dolomite crystals have been dedolomitized and the deformed lattice has been inherited by the newly formed calcite (Plate 32, Fig. 6). This phenomenon added evidence that the dedolomitization of these rocks has taken place very late and after the incipient metamorphism of the rock.

In some incipiently metamorphosed crinoidal biomicrites, the microcrystalline crinoidal ossicles show signs of lattice dislocations. This is displayed by the presence of adjacent disoriented bands (Plate 31, Fig. 5). These bands can be clearly seen between crossed nicols and present at alternating sectors radially arranged. This fabric is commonly seen in the horizontal sections of the crinoidal ossicles.

(2) Bending of the twin lamellae and the cleavage traces:

Bending of the twin lamellae is commonly seen in the deformed calcite crystals forming the crinoidal debris as well as in the intergranular calcite cement. This phenomenon is usually displayed by the coarse grained crystals. It was found that twin lamellae have been elastically bent and occasionally they are mechanically displaced.

In the recrystallized biomicrites it is often found that the cleavage traces of some pseudosparry calcite crystals are bent (Plate 31, Fig. 2). The same feature has also been seen in the deformed crinoidsl ossicles and their overgrowths.

(3) Deformed calcite veinlets:

An indication of the incipient metamorphism is the presence of deformed calcite veinlets within the Carboniferous limestone rocks. Calcite veinlets have a fabric similar to that of the fibrous gypsum veins filling the hydraulic fractures in the evaporite rocks described by Shearman et al. (1972), have been noticed. The following types of deformed calcite veinlets have been recognised: (i) deformed calcite veinlets in coaly fragments present in packed biomicrite; (ii) deformed calcite veinlets in completely chertified micrite; and (iii) deformed calcite veinlets developed in the stylolite seams in dolomicrite.

(a) Deformed calcite veinlets in packed biomicrite:

This type of calcite veinlet has been recognised occurring in the incipiently metamorphosed packed biomicrites. Calcite veinlets are present filling fractures in coaly fragments trapped and deformed between the skeletal debris forming the packed biomicrites. Each calcite veinlet consists of bundles of calcite fibres which extend from wall to wall transversely. The fibres are commonly slightly bent and of sigmoidal shape. Fine pieces of the coal material detached from the fracture wall are often seen floating in the fibrous calcite mass. Some of these pieces are included by more than one fibre. In thin sections parallel to the fibre axes, the fibrous structure is more commonly expressed by optical extinction shadows than by physical grain boundaries (Plate 33, Figs. 1 and 2).

Shearman et al. (1972) suggested that the fibrous gypsum veins have been developed in spaces formed by hydraulic fracturing of rocks. The gypsum of the veins has grown in the water-filled fractures, while the overburden was supported on the water. When the residual water escaped from the fractures, the overburden load was transferred to the gypsum of the vein, producing the fibrous structure.

Shearman's theory is applicable to a great extent to the origin of the deformed fibrous calcite veinlets in the studied rocks. It is suggested that during the early diagenetic pressure solution in the packed biomicrite, considerable amounts of $CaCO_3$ have been dissolved along the fissility planes of the coaly streaks which have been jacked open by the hydraulic pressure of the injected water. Coarse-grained calcite crystals grew up in the water-filled fractures, while the overburden was supported on the water. Due to the release of the residual water for any reason, the overburden load was transferred to the calcite of the vein producing the present fibrous structure. By increasing the overburden pressure, the calcite fibres tended to be bent and curved.

(b) Deformed calcite veinlets in silicified limestones:

Deformed fibrous calcite veinlets have been identified in some samples from the chert nodules in the Z zone of the Gower Peninsula. These nodules commonly consisted of completely silicified micrite or biomicrite which contain dolomite rhombs. Calcite veinlets of about 2 mm. thick are usually horizontal to sub-horizontal, and are characterised by sharp boundaries. These calcite veinlets consist of sigmoidal calcite fibres extending from wall to wall transversely. These fibres are uniform in shape but vary in width. Optical study has revealed that groups of adjacent fibres are basically in lattice continuity with one another (Plate 33, Figs. 3 and 4).

The mechanism of the formation of these veinlets is similar to that found in the packed biomicrites. The source of the calcite forming these veinlets is most probably from the $CaCO_3$ released during the replacement of the original carbonate rock by silica.

(c) Deformed calcite veinlets in dolomite:

In the dolomicrites of the S zone outcropping in the northern part of the South Wales Coalfield, very thin fibrous calcite veinlets have developed within the organic matter concentrated on the stylolites. These stylolite seams reach up to 3 mm. in thickness and are commonly fractured. Fractures are filled with fibrous calcite veinlets, similar to those previously described (Plate 33, Fig. 5).

INCIPIENT METAMORPHIC FABRICS OF THE TERRIGENOUS ROCKS

The fabrics and textures of incipiently metamorphosed terrigenous rocks have been described by many Russian geologists. Kossovskaya and Shutov (1970) summarised some of the Russian work on the alteration of terrigenous rocks during deep burial. They particularly pointed out the economic importance of such studies. In petroleum geology, for example, the destruction of porosity and permeability and the generation, migration and ultimate destruction of hydrocarbons is clearly related to the temperature and pressure that have affected the rock.

Kossovskaya and Shutov (1958) in their study on the terrigenous rocks of the Veikloyansk geosyncline, Mesozoic deposits in the eastern part of the Siberian platform, and of Paleozoic strata of the south eastern part of the Russian platform, have distinguished four incipient metamorphic zones. These are: (i) zone of unaltered clay cement in which the rocks are fully preserved in their original textural and mineralogical properties; (ii) zone of altered clay cement which is characterised by recrystallization of clays, hydromicatization of kaolinite, and complete disappearance of montmorillonite; (iii) zone of quartzite-like structure and hydromica-chlorite-cement; and (iv) zone of spine-like structure characterised by the perpendicular orientation of chlorite and muscovite lamellae penetrating into the composite regenerated grains of quartz and feldspar.

In the following paragraph, the characteristic fabrics of the incipiently metamorphosed Coal Measure rocks of the South Wales Coalfield will be discussed.

(A) REACTION OF CLAY MINERALS WITH THE FRAMEWORK GRAINS

In the sandstones and siltstones of the anthracite area, reaction between the clay matrix minerals and the quartz grains has been noticed. Clay minerals in these rocks are formed mainly of well crystalline illite and chlorite. They are usually found to be arranged perpendicular to the quartz grain boundaries, forming a texture similar to the spine-like structure of Kossovskaya and Shutov (1958). Recrystallized micaceous clay minerals seem to be corroding the quartz grains which are characterised by hazy outlines (Plate 34, Figs. 4 and 5).

(B) DEFORMED ROCK FRAGMENTS

The difference in the fabric of the argillaceous rock fragments in the litharenites of the eastern part and the western part of the South Wales Coalfield is very clear. In the anthracite area the argillaceous rock fragments in the litharenites are completely deformed, losing their original shape and internal fabric. They have been transformed to a clayey matrix, "pseudomatrix". Commonly in these rocks a reaction rim on the contact between argillaceous rock fragments and quartz grains has been developed. These reaction rims consist of newly formed micaceous clay minerals which are arranged normal to the quartz grain surfaces.

(C) DEFORMED FIBROUS QUARTZ VEINLETS

Fibrous quartz veinlets have been occasionally noticed in the quartzarenites of the anthracite area. These veinlets are very similar from the textural point of view to those described in the carbonate rocks. Fibrous quartz veinlets are usually developed along horizontal weak planes, such as contacts between coal streaks and the quartzitic groundmass. These veinlets consist of quartz fibres extending from wall to wall transversely. Some of the fibres are bent and curved. Between crossed nicols, each group of bundles basically has an optical continuity, but the lattice of each individual fibre has slightly dislocated from the lattice of the adjacent fibre (Plate 34, Figs. 1, 2 and 3).

The mechanism by which the fibrous quartz veinlets have been formed is similar to that responsible for the formation of the formerly described fibrous calcite veinlets.

DISTRIBUTION OF THE INCIPIENT METAMORPHIC FABRICS IN THE SOUTH WALES COALFIELD

The development of the previously described incipient metamorphic fabrics in both the Carbonate and terrigenous rocks were found to be related to their grade of incipient metamorphism as revealed from the fixed carbon % of the associated coal seams.

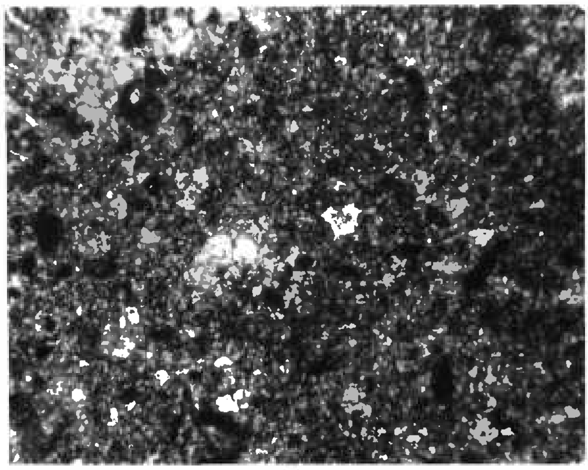
In the Carbonate rocks, the degree of recrystallization of the micrite was found to be increased westwards, i.e. in the direction of increasing the fixed carbon %. Therefore it was difficult to observe even slight recrystallization in the limestones of the southern and eastern outcrops. In the central part of the northern outcrop (from Brynmawr in the east to Penwyllt in the west) recrystallization starts to affect the matrix of the biomicrites. In the western outcrops (anthracite area) recrystallization was found to affect most of the rocks, the matrix and the allochems. Deformation fabrics in both calcite and dolomite crystals have been observed in the rocks of the western outcrops only.

In the Coal Measures litharenites the degree of deformation of the argillaceous rock fragments is clearly increased towards the anthracite area. It was very difficult to recognise the original fabric of the argillaceous rock fragments in lithicarenites from the western outcrops, while in the eastern part of the coalfield they are slightly deformed. At the same time, the clayey matrix of the greywackes is progressively getting coarser in size and develops a micaceous fabric westwards. In the anthracite area, the recrystallized clays are commonly reacting with the framework grains. Also, fibrous quartz veinlets were observed only in the anthracite area.

PLATE 28

Aggrading recrystallization of micrite. The figures are arranged in order of increasing grade of recrystallization.

Fig. 1	Sample 655
Fig. 2	Sample 93.
Figs. 3 & 4	Sample 102.
Fig. 5	Sample 101.
Fig. 6	Sample 615.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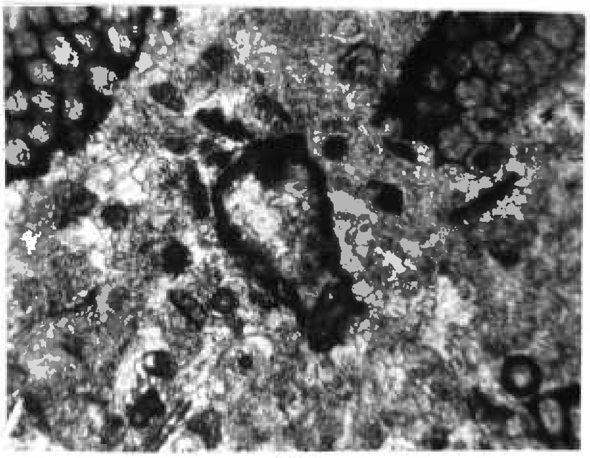
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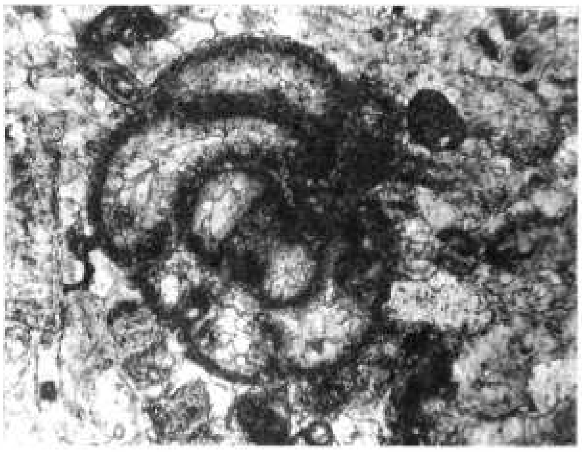
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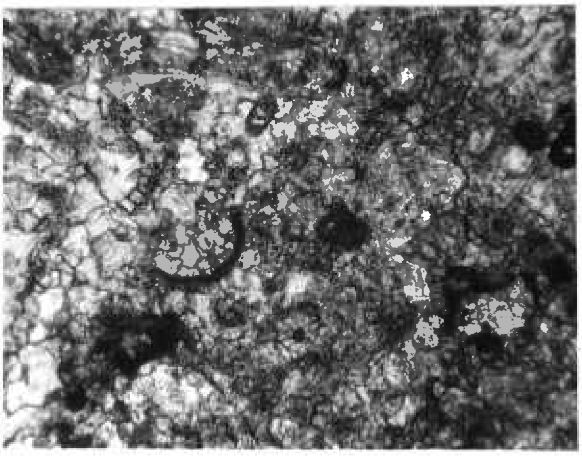
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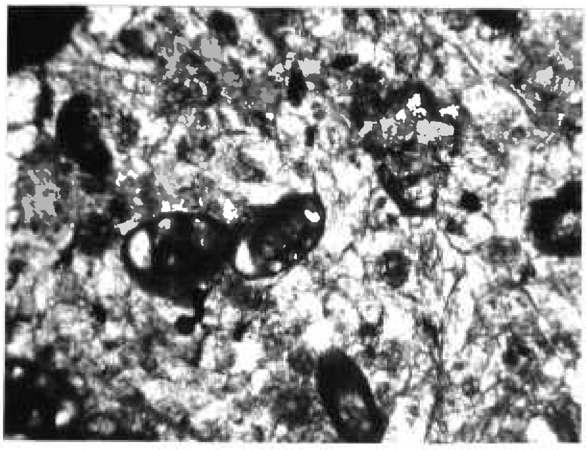
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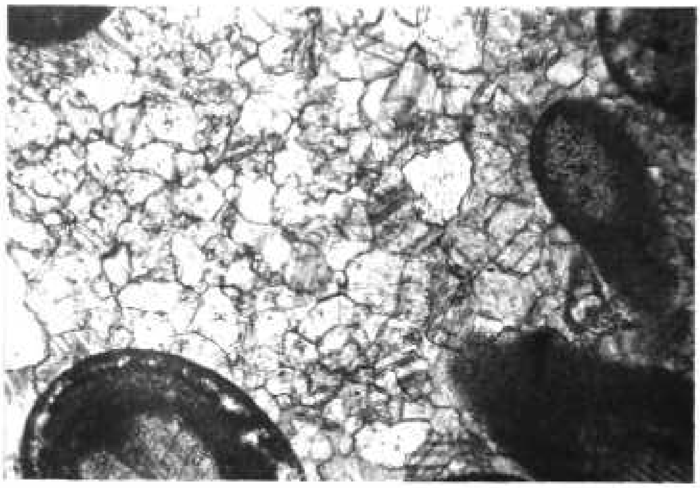
PLATE 29

Fig. 1 Completely recrystallized matrix in a biomicrite rock. Note the curved boundaries between the adjacent pseudospar crystals. Sample BY62.

Fig. 2 Ghosts of allochems in an almost completely recrystallized matrix in a biomicrite (arrow). Sample 629.

Fig. 3 Ghost of a foraminiferal test (?) in a completely recrystallized matrix in a biomicrite. Sample 673.

Figs. 4 & 5 Partially recrystallized oolite in an oolitic limestone. Note ghost of the original oolite displayed in the pseudospar (arrow). Sample BY43.



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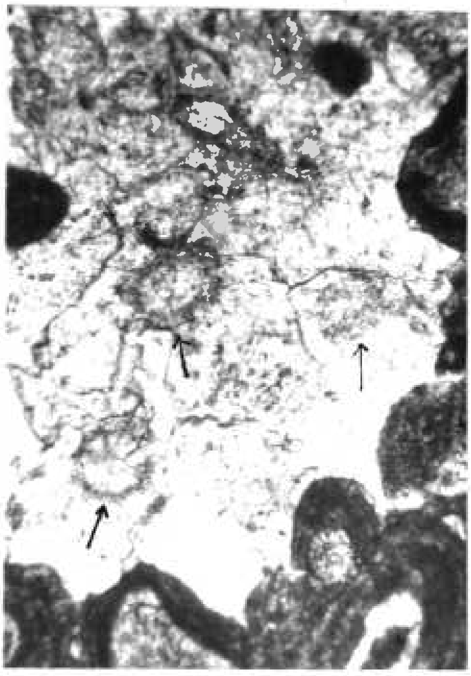

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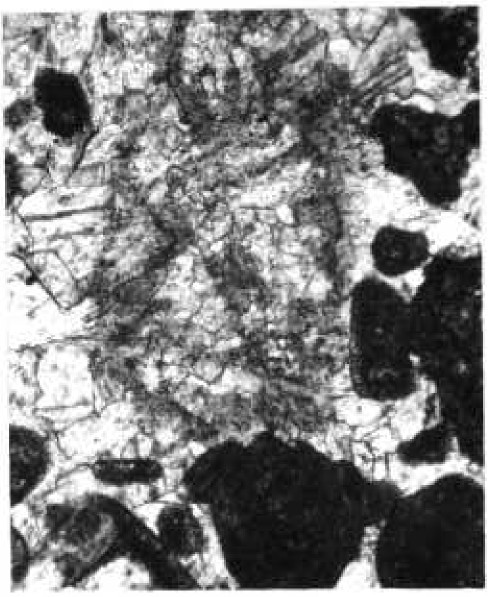

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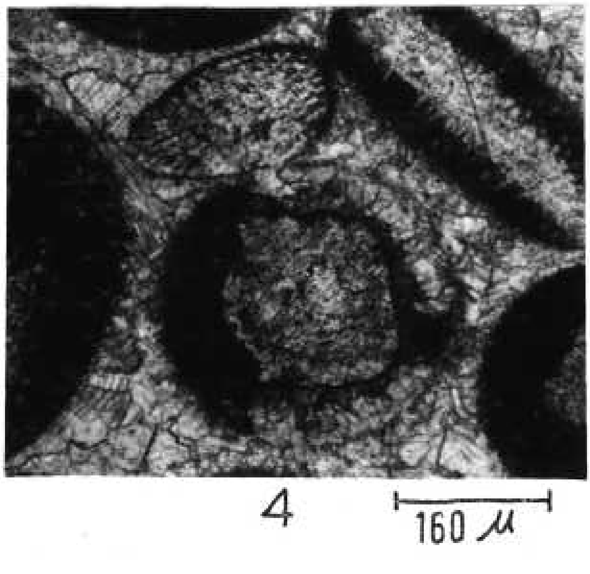

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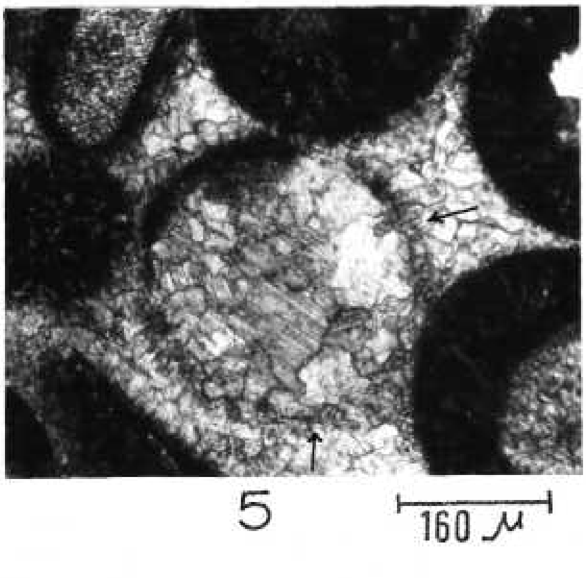

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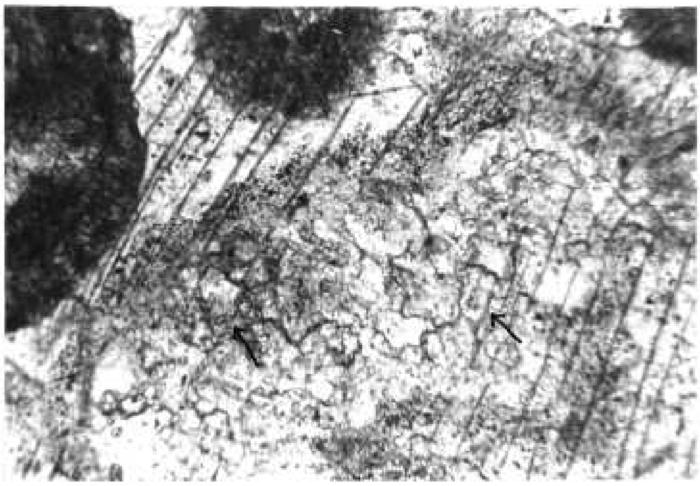
PLATE 30

Fig. 1 Degrading recrystallization of a microcrystalline crinoid fragment. Note, recrystallization is confined to the crinoid fragment and it does not affect the calcite overgrowth. The cleavage of the original coarsely crystalline calcite crystal is still preserved in the newly formed pseudospar (arrow). Sample 101.

Fig. 2 Incipient stage of degrading recrystallization of an echinoderm fragment. Sample 671.

Figs. 3 & 4 Degrading recrystallization of syntaxial calcite cement (arrow). Samples BY41 and 101.

Fig. 5 Degrading recrystallization of a coarsely crystalline calcite infilling fractures. Note the occurrence of broad twin lamellae in the original crystal while the newly formed calcite crystals are strain-free. Sample 1000. Crossed polars.



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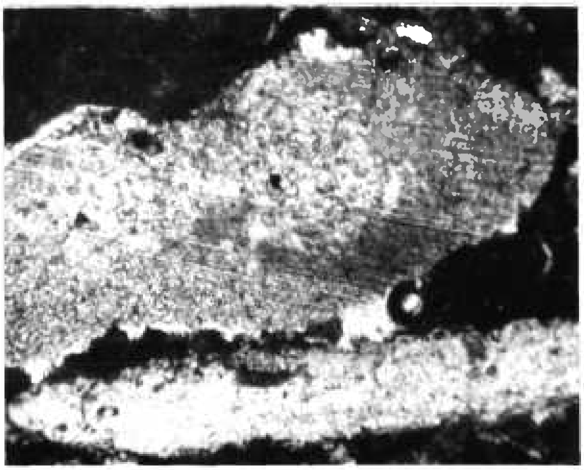

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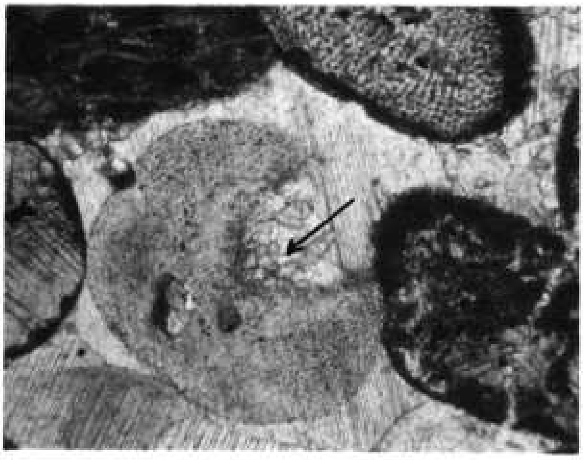

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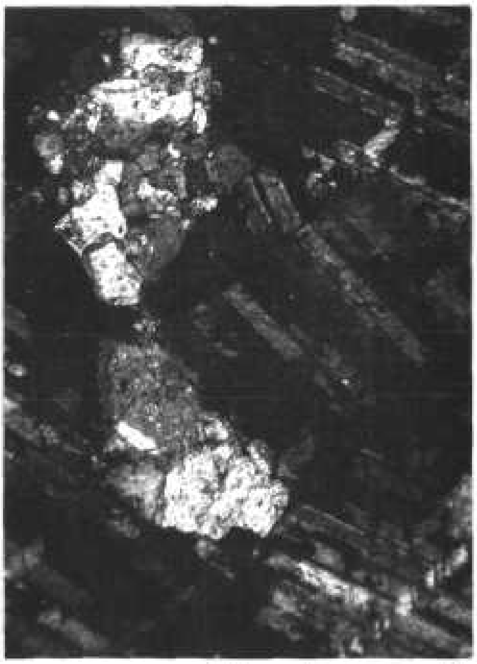

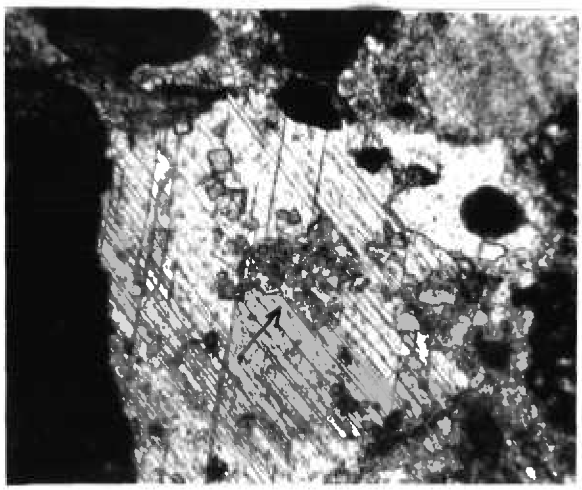

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PLATE 31

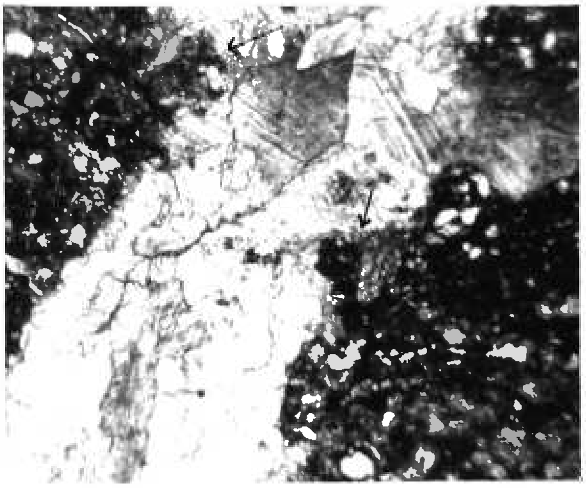
Fig. 1 Recrystallized sparry calcite veinlet. Note the irregular boundaries of the veinlet due to partial recrystallization of the rock groundmass near the veinlet (arrow). Sample 618.

Fig. 2 Deformed pseudospar crystal (arrow). Note ghosts of the original micrite. Sample 655.

Fig. 3 Deformed coarsely crystalline pseudospar in a micritic rock. Note ghost of allochem (arrow) and curved cleavage plane. Sample 621.

Fig. 4 Deformed calcite crystals in a biomicrite. Note curved twin lamellae. Sample 93.

Fig. 5 Deformed crinoid ossicle. Sample 765. Crossed polars.



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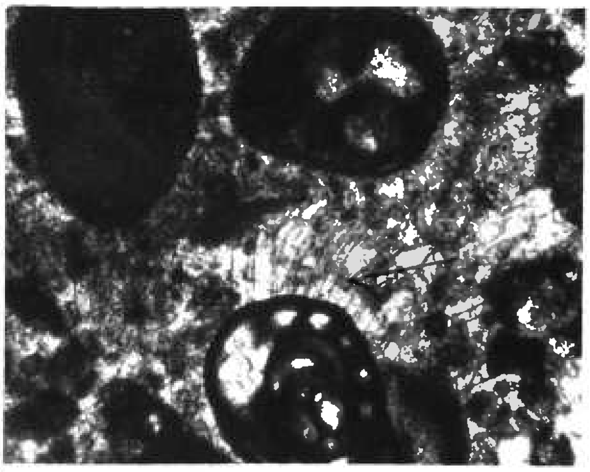

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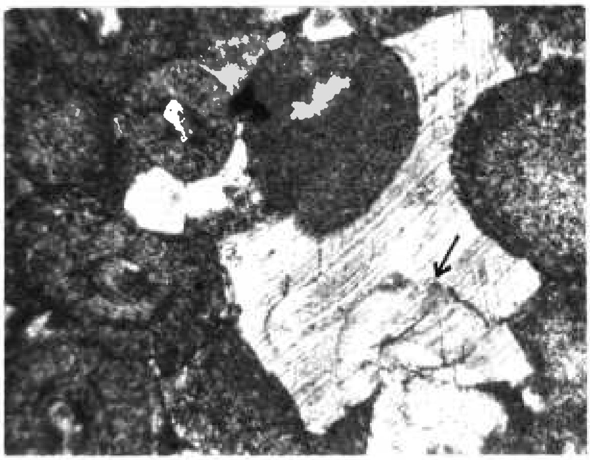

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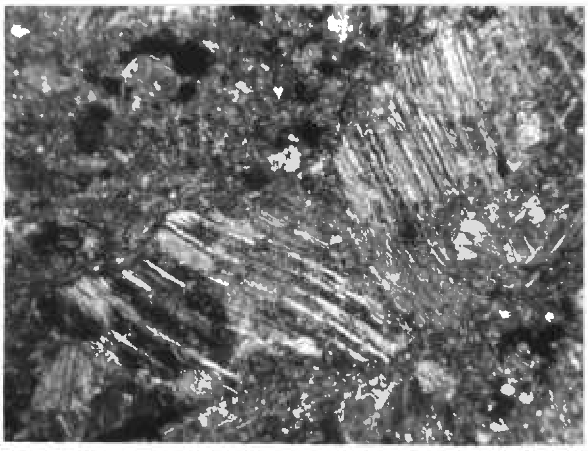

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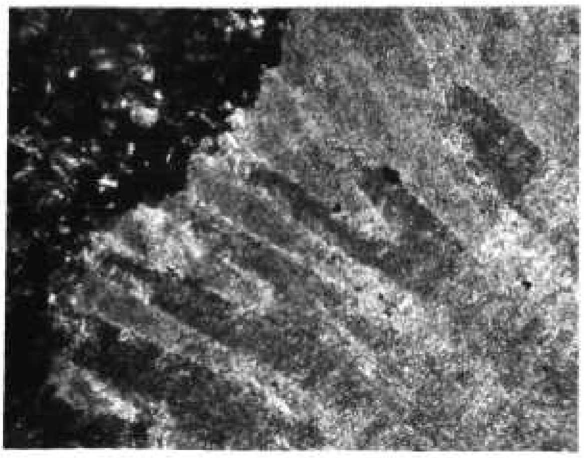

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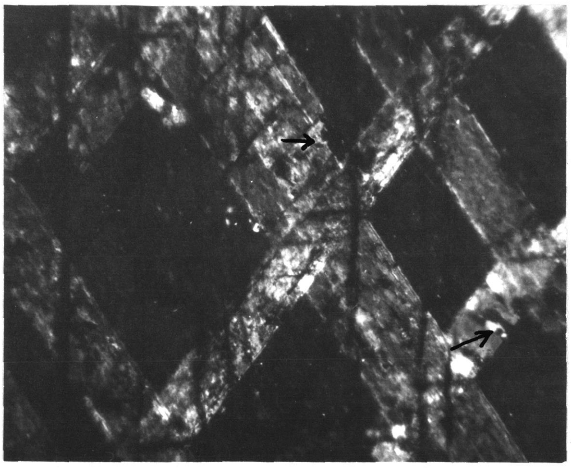
PLATE 32

Fig. 1 Broad twin lamellae in a deformed coarsely crystalline calcite crystal. Note the corrosion of the twin lamellae by the calcite in the groundmass (arrow). Sample 1000. Crossed polars.

Fig. 2 Curved boundaries between two adjacent pseudospar crystals (arrow). Sample 1000. Crossed polars.

Figs. 3, 4 & 5 Deformed dolomite crystals. Note curved cleavage planes and brush extinction. Sample 621.

Fig. 6 Deformation fabric represented by brush extinction displayed by calcitized dolomitr crystal. Sample 615. Crossed polars.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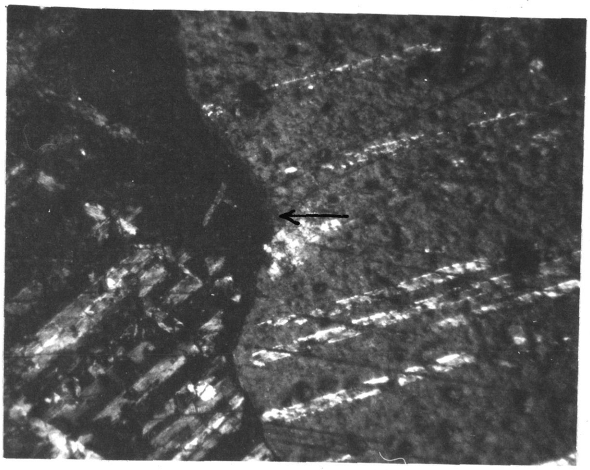
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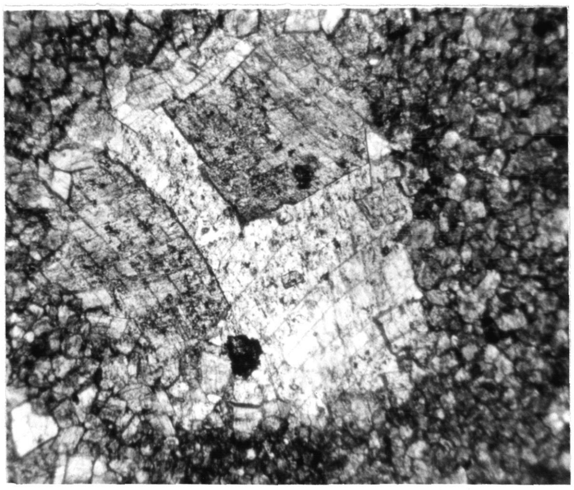
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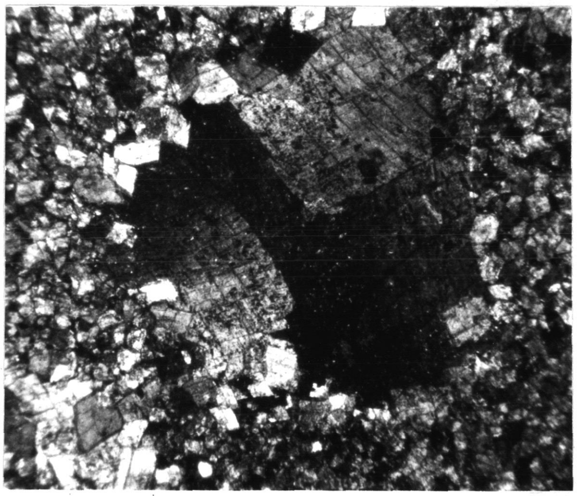
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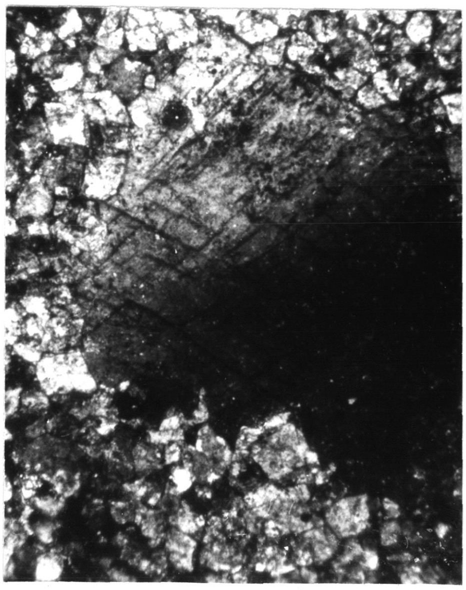
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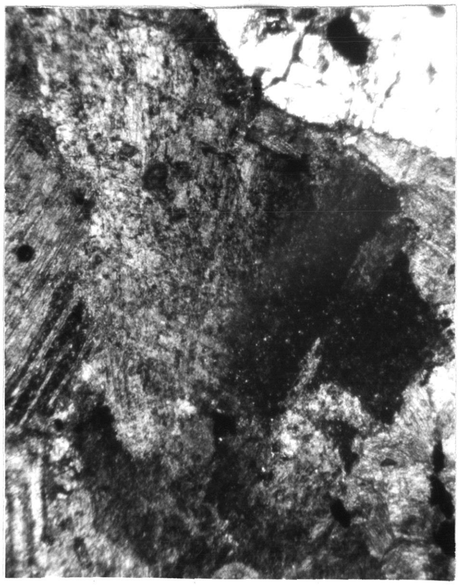
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PLATE 33

Figs. 1 & 2 Fibrous calcite veinlet developed in hydraulic fractures in a coal streak. Note signoidal shape of the calcite fibres (arrow). Sample 615. Crossed polars.

Figs. 3 & 4 Fibrous calcite veinlet in a silicified biomicrite. Sample 624.

Fig. 5 Fibrous calcite veinlet developed in an organic rich stulolitic seam. Sample BY65.


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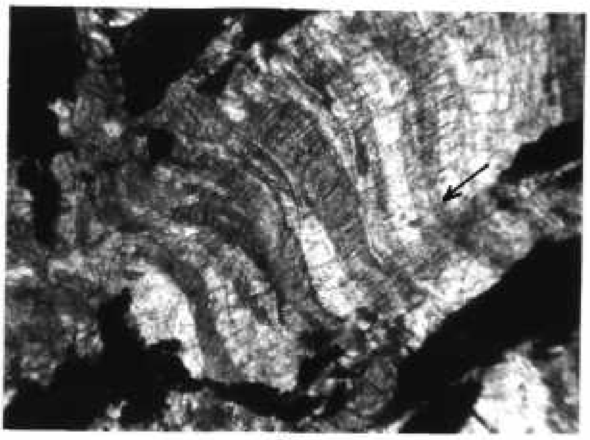

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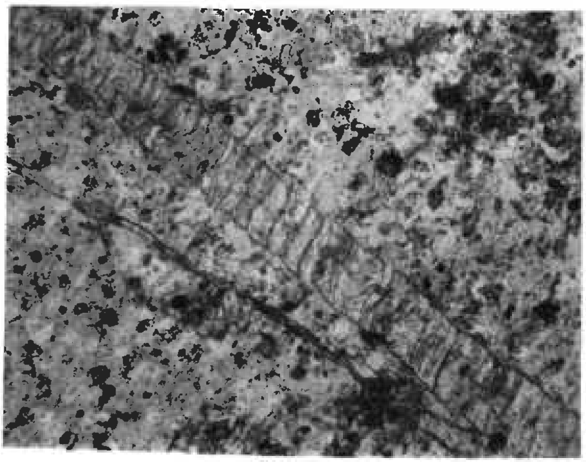

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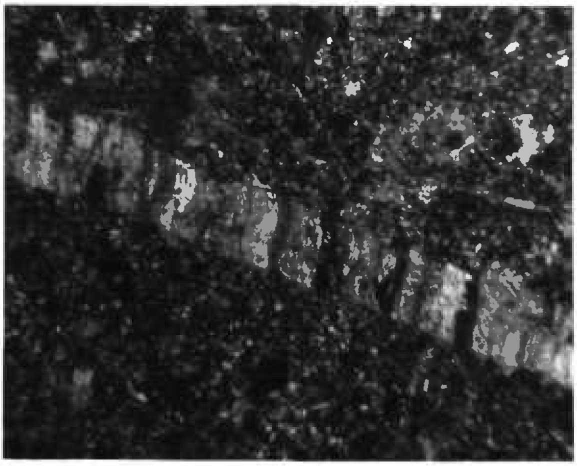

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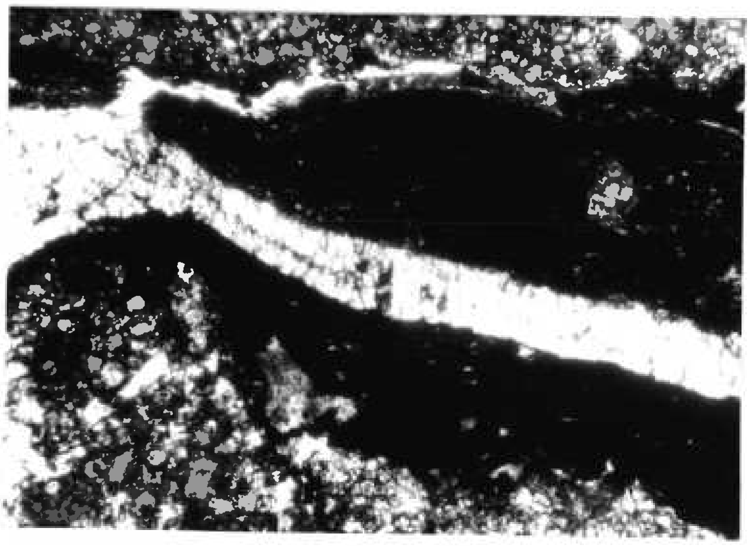

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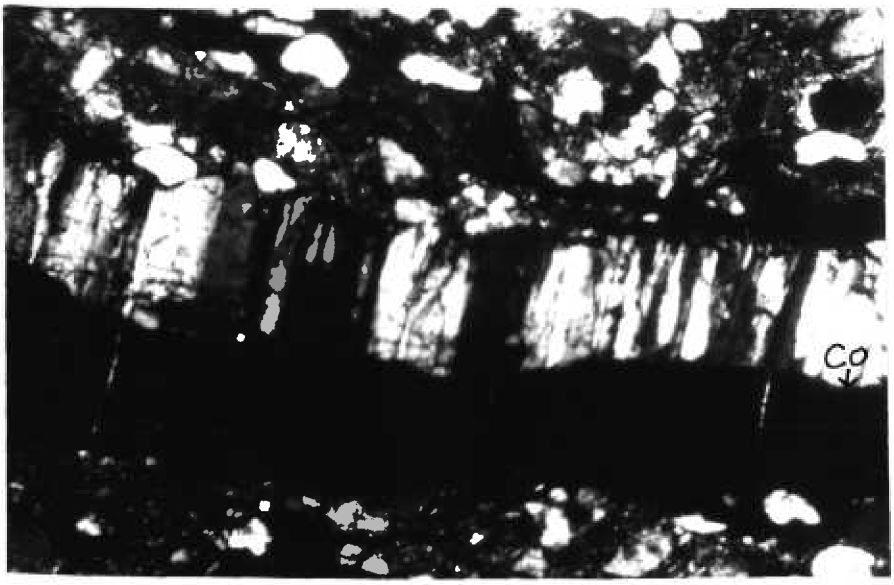
PLATE 34

Figs. 1 & 2	Fibrous quartz veinlet. (Co) coaly fragment. Sample 760A.
Figs. 3 & 4	Incipient metamotphism of sandstone. Note the development of micaceous minerals and the reaction between the matrix and the quartz grains (arrow). Sample 908, 851.



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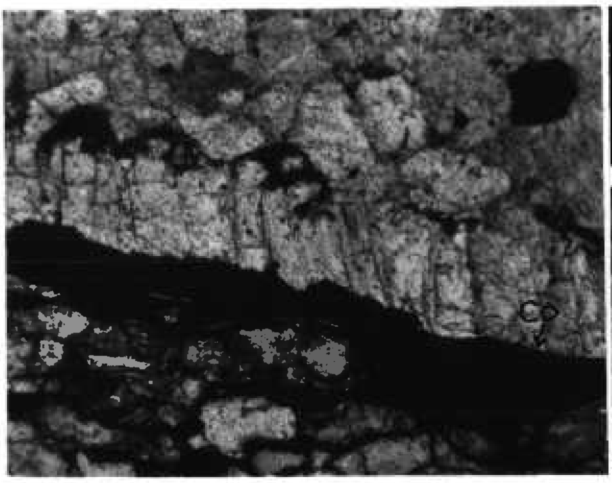

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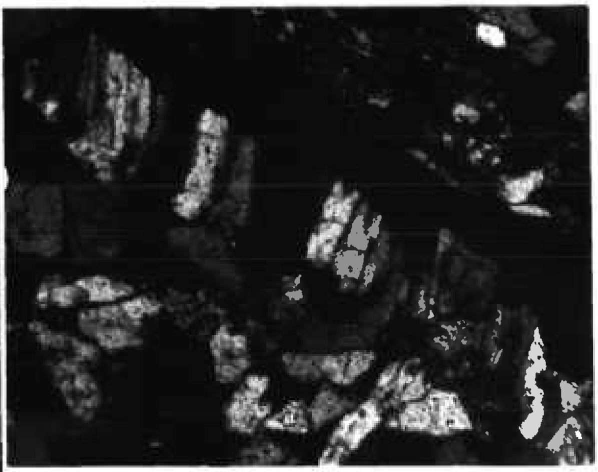

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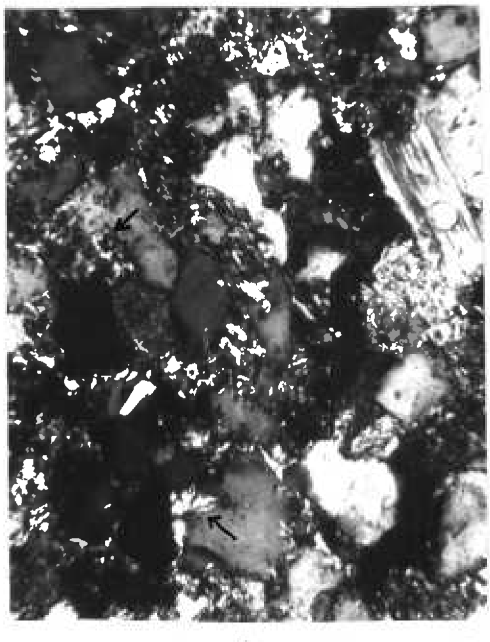

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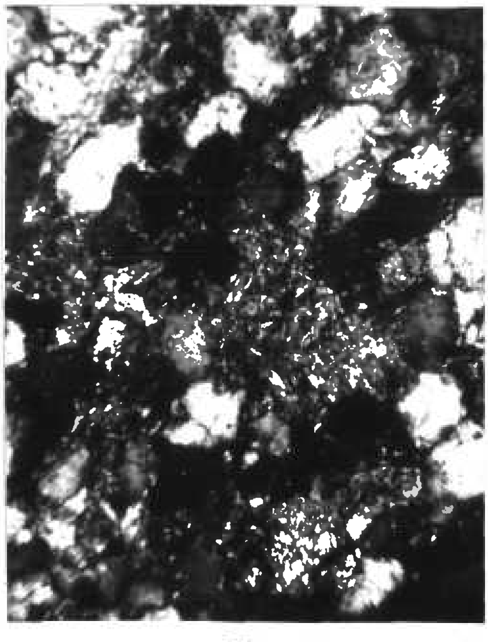

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CHAPTER VII

DIAGENESIS AND INCIPIENT METAMORPHISM OF THE CLAY MINERALS

DIAGENESIS AND INCIPIENT METAMORPHISM OF THE CLAY MINERALS

1. INTRODUCTION

This chapter deals with the study of clay minerals present in the various lithotypes of the Carboniferous rocks of the South Wales Coalfield. Because very little work was published on the clay mineralogy of these rocks, it was necessary to study these minerals in detail. There are two main objectives in carrying out this study. The first is to give an idea of the composition of the clay minerals in each of the studied lithotypes to complete the petrographical description. The second objective is concerned with the diagenesis and incipient metamorphism of the clay minerals.

For these purposes, insoluble clay fraction separated from the carbonate rocks of the Carboniferous Limestone Series, and clay fraction of Millstone Grit and Coal Measures terrigenous rocks were qualitatively and quantitatively analysed by means of X-ray diffraction.

METHODS OF STUDY

1. Sample Preparation:

(a) Separation of the $< 2\mu$ fraction:

The technique used for the separation of the $< 2\mu$ fraction is dependent on the nature of the studied rocks.

(a.i) Carbonate rocks:

A sample of approximately 200 gm. was washed and scrubbed in distilled water to remove possible adhering clay; the sample was then allowed to dry. It was then crushed to pass a 60-mesh sieve. About 100 gm. of the crushed material was treated by 0.5 M acetic acid until almost all of the carbonate material had been dissolved, (Ostrom, 1961; and Carroll and Starkey, 1971). The insoluble residue was then washed by distilled water three times, and the $<2\mu$ fraction was separated using the sedimentation method.

(a.ii) Terrigenous rocks:

A sample of about 100 gm. was washed and scrubbed in distilled water to remove possible adhering clay; the sample was then allowed to dry. After that, it was crushed to pass a 60-mesh sieve. Crushed material was soaked in distilled water for about three days. The $<2\mu$ fraction has been separated by sedimentation method.

(b) Preparation of Oriented Specimens:

The method applied by Shaw (1971) for the preparation of oriented clay mineral specimens for X-ray diffraction analysis was followed in the present study. The method involved filtering a clay suspension under suction through a suitable ceramic medium so that the clay material was deposited on the ceramic. The apparatus used consists of a cylindrical stainless steel filter funnel, attached to a conical flask jointed to a vacuum pump. A ceramic disc of about 1.5" in diameter and ¼" thick was used as a filter bed. The ceramic disc filter beds are drilled with a rock corer of standard unglazed biscuit tiles. A battery of three filter funnels was used in the present study (Fig. 7.1).

(c) Preparation of Non-oriented Specimen:

This technique was used for the X-ray analysis of the whole rock material of the argillaceous rocks. The rock specimens were washed in distilled water to remove possible adhering clay. Specimens were allowed to dry and then crushed to pass 200-mesh sieve. The powdered sample was back loaded into an aluminium cavity holder for X-ray analysis.

2. Analytical Methods:

A Philips PW 1060 diffractometer equipped with a linear recorder was used for the analysis. The following experimental conditions were used: Cobalt radiation (CoK_α) with an iron filter placed before the receiving slit to eliminate possible β radiation. As supply, 38 Kv and 24 mA were chosen, and 1 Kw generator was used to give high intensity

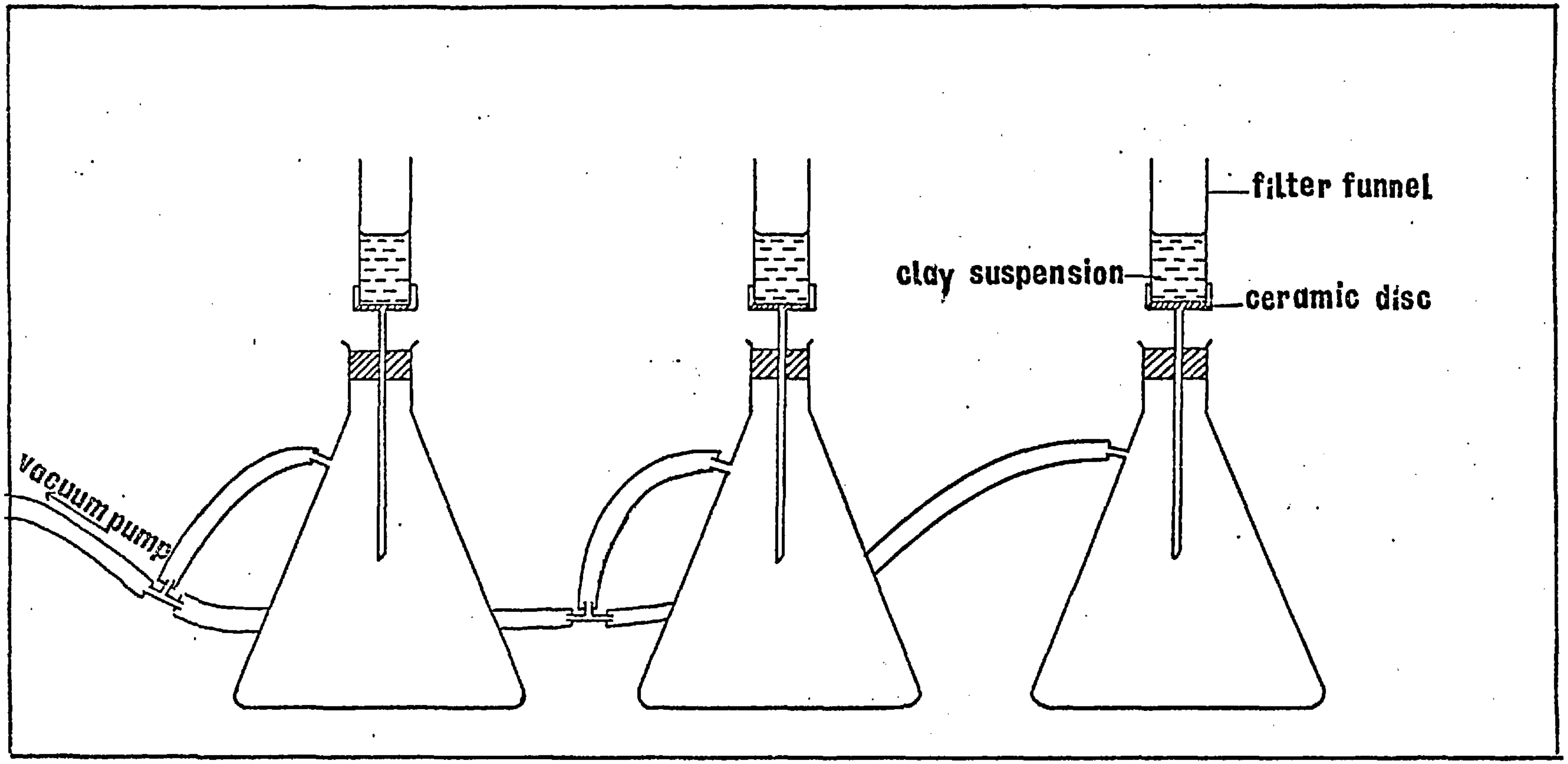


FIG. 7.1 — SUCTION ONTO CERAMIC DISC METHOD OF SAMPLE PREPARATION

and a good peak to background ratio. Scanning speed 1° 2θ/min., divergence and scatter slit both $\frac{1}{2}^\circ$ in the case of the oriented specimens, and 1° in the case of the non-oriented specimens, receiving slit 0.1 mm.

Generally, identification of the clay minerals by means of X-ray diffraction is facilitated by pre-treatment of the oriented mount in various ways before X-ray analysis (Brown, 1961).

In examining the clay fraction of the studied rocks the following experimental procedure was carried out:-

(a) Air-dried specimen mounted on a ceramic disc and scanned from 2°-34° 2θ.

(b) Specimen glycerated by spreading a thin film of liquid glycerol over the clay specimen and allowing it to dry. Scanned from 2°-20° 2θ.

(c) Specimen heated in muffled furnace at 550°C for one hour. Scanned from 4°-15° 2θ.

3. <u>Clay Mineral Identification</u>:

Examination of the clay fraction of the rocks studied shows that the principal clay minerals present in most of the samples are illite, mixed layer clays, kaolinite and chlorite.

Pyrophyllite has been identified in some Coal Measures rocks in the anthracite area.

The characteristic X-ray properties of these clay minerals have been discussed in detail in the literature, such as Brindley (1951); Grim (1953); MacEwan (1956); Weaver (1956); Sudo (1959) Brown (1961); Molloy and Kerr (1961); Warshaw and Roy (1961); Mackenzie and Mitchell (1966). The following is the general X-ray properties used for the identification of the present clay minerals.

Illite:

The term illite is used as a collective name denoting all 10 Å non-expanding clay minerals (Gaudette et al., 1966). In oriented mount it shows a relatively sharp basal reflection which becomes slightly larger after heat treatment. When the mineral is associated with illite-montmorillonite mixed layer mineral, which is usually the case in the rocks studied, the first-order basal reflection of both minerals partially overlap, forming a combined peak characterised by asymetrical tailing towards the lower diffraction angle. Illite has a marked second-order reflection of about 5 Å, though usually weaker than that of muscovite; in the trioctahedral 10 Å members of the mica minerals such as biotite it is almost absent.

Illite polymorphs:

Polymorphism for mica minerals was defined by Hendricks and Jefferson (1939). The muscovites and natural illites exist only in the 1M, 2M and 3T, plus a completely disordered 1Md form. The 3T form is practically unknown among the illites. Stability increases in the sequence 1Md - 1M - 2M, and an increase in temperature favours this evolution (Yoder and Eugster, 1954; and Velde, 1965). The determination of the polymorphic type of the illite in the studied samples was very difficult due to the presence of other inseparable clay minerals. In some samples it was impossible to determine the polymorphic type of illite. The method proposed by Maxwell and Hower (1967) was used for the determination of the polymorphic type of illite. In this method the polymorphic type is determined by measuring in non-oriented sample the intensity of the 2.80 Å peak which represents the 2M polymorph and of the 2.58 Å peak which represents the total dioctahedral illite, 1M and 2M (Fig. 7.2).

Mixed-layer clay minerals:

Two varieties of mixed-layer clay minerals were identified. These are, randomly interstratified illite-montmorillonite and regular interstratified illite-montmorillonite. Randomly interstratified illite-montmorillonite mineral was

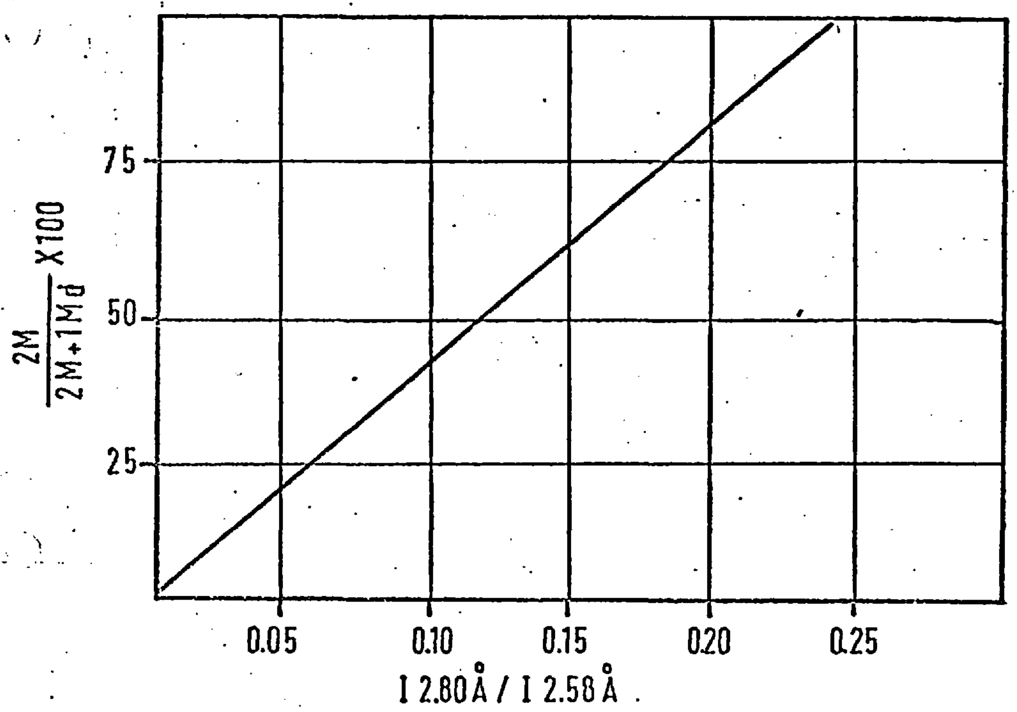


FIG. 7.2 THE RELATIONSHIP BETWEEN 2M POLYMORPH % AND I2.80 Å/I2.58 Å (AFTER MAXWELL AND HOWER, 1967)

identified on the basis of its broad peak which occupies the area between 13 Å and 10 Å in the case of the untreated sample. It shifts to lower diffraction angle on glyceration and contracts to 10 Å on heat treatment. This mineral gives non-integral series of basal reflections. The d spacing of its basal reflection (001) depends on the percentage of the swelling montmorillonitic layers within the structure. Brown and MacEwan (1950) calculate the (001) d spacing of a randomly interstratified illite-montmorillonite after glyceration and showed how it varied depending on the relative proportions of each mineral component. Percentage of expandable (montmorillonite) layers in the randomly interstratified illite-montmorillonite mixed layer mineral was determined by using a curve based on Weaver's data (1956), (Fig. 7.3).

Regular interstratified illite-montmorillonite mineral was identified in some samples from the Coal Measures Series. In contrast to the randomly interstratified illite-montmorillonite mineral, this mineral is characterised by a regular progression of its basal reflection. Fig. 7.4 shows an example of the X-ray diffraction pattern of both minerals.

Kaolinite:

Usually kaolinite can be identified directly from its strong diffraction maxima at 7.1 Å, 3.5 Å and 2.38 Å. On

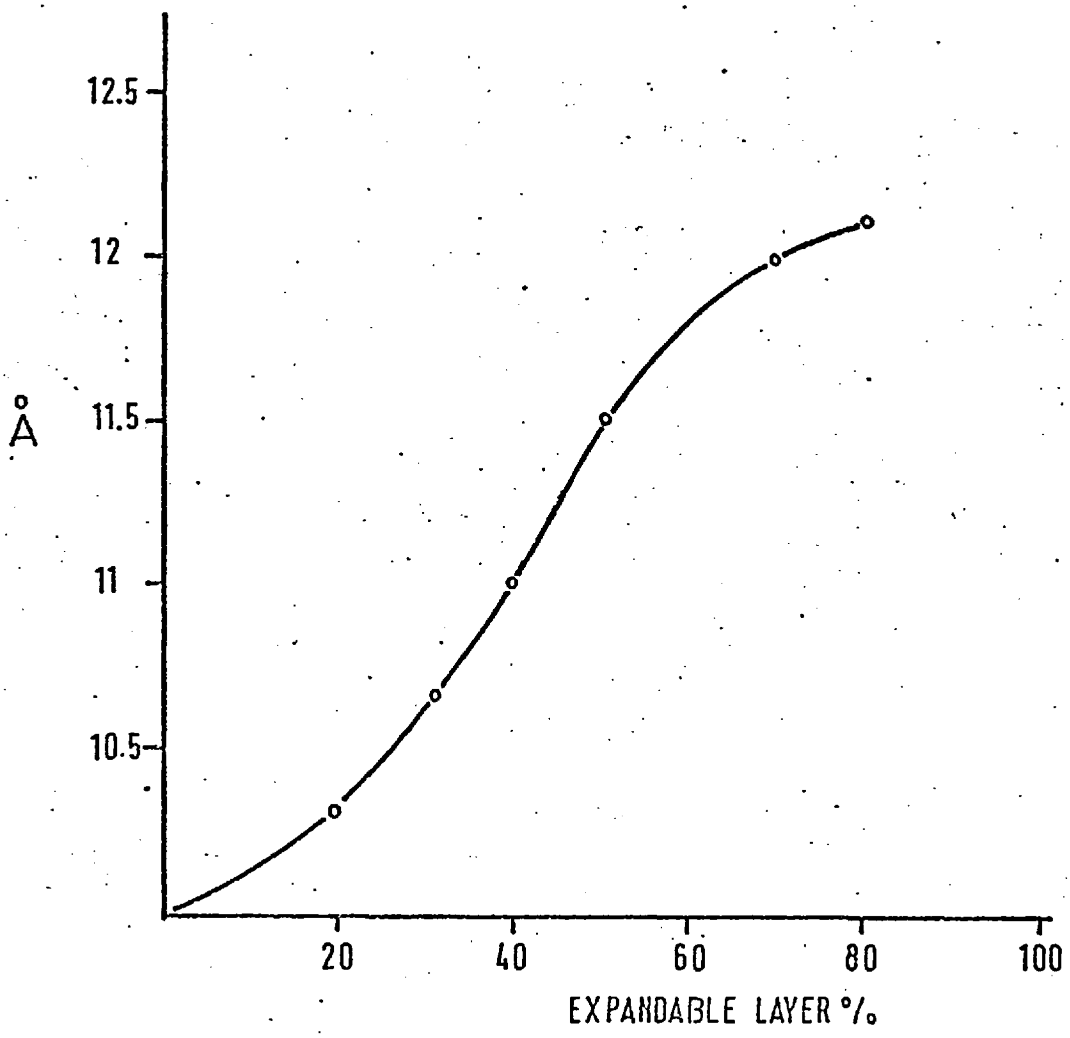


FIG.7.3 CURVE SHOWING MIGRATION OF 001/001 PEAKS OF RANDOMLY INTERSTRATIFIED 10 Å AND 12.4 Å LAYERS

(Data after WEAVER, 1956)

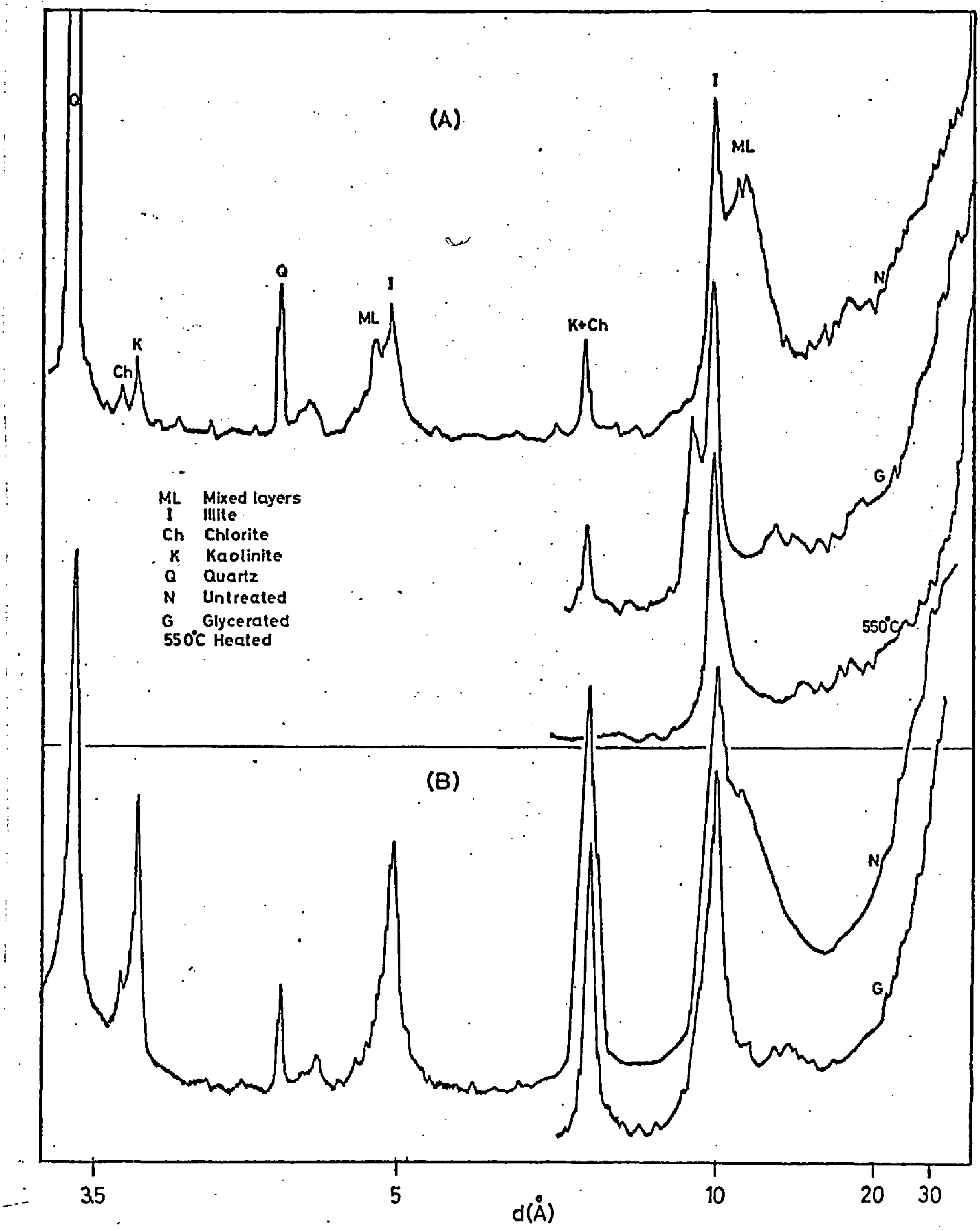


FIG. 7·4 X-ray diffractograms show the characteristic peaks of regular interstratified illite-montmorillonite mixed layer(A) sample no.908, and random interstratified illite-montmorillonite mixed layer(B) sample no. 2493-19.

heating at 550°C the kaolinite structure will break down and all the reflections due to kaolinite will disappear. Chlorite will give peaks at or near the position of the strongest basal reflection of kaolinite.

In the rocks studied it was easy in most cases to identify kaolinite in the presence of chlorite even in the untreated samples. It was possible to determine the state of crystallinity of the kaolinite from the X-ray diffractograms. Disorder in the kaolinite lattice produces blurred and weak reflections. It also shows an asymmetric (002) band with sharp head at 4.45 Å, fading away towards higher angles. On the other hand, well ordered kaolinite shows little or no band in this region and the reflections at 4.18 and 4.13 Å are resolved (Brindley, in Brown, 1961).

In some limestones and sandstones, dickite was identified. It is better crystallised than kaolinite and shows sharper reflections (Fig. 7.5). Unlike kaolinite, dickite is not affected by heating to 500°C. For dickite, the peak temperature lies about 100°C higher than that for kaolinite (Richardson, in Brown, 1961).

Chlorite:

Chlorite exhibits an integral series of basal reflections with a first order reflection of 14 Å. The

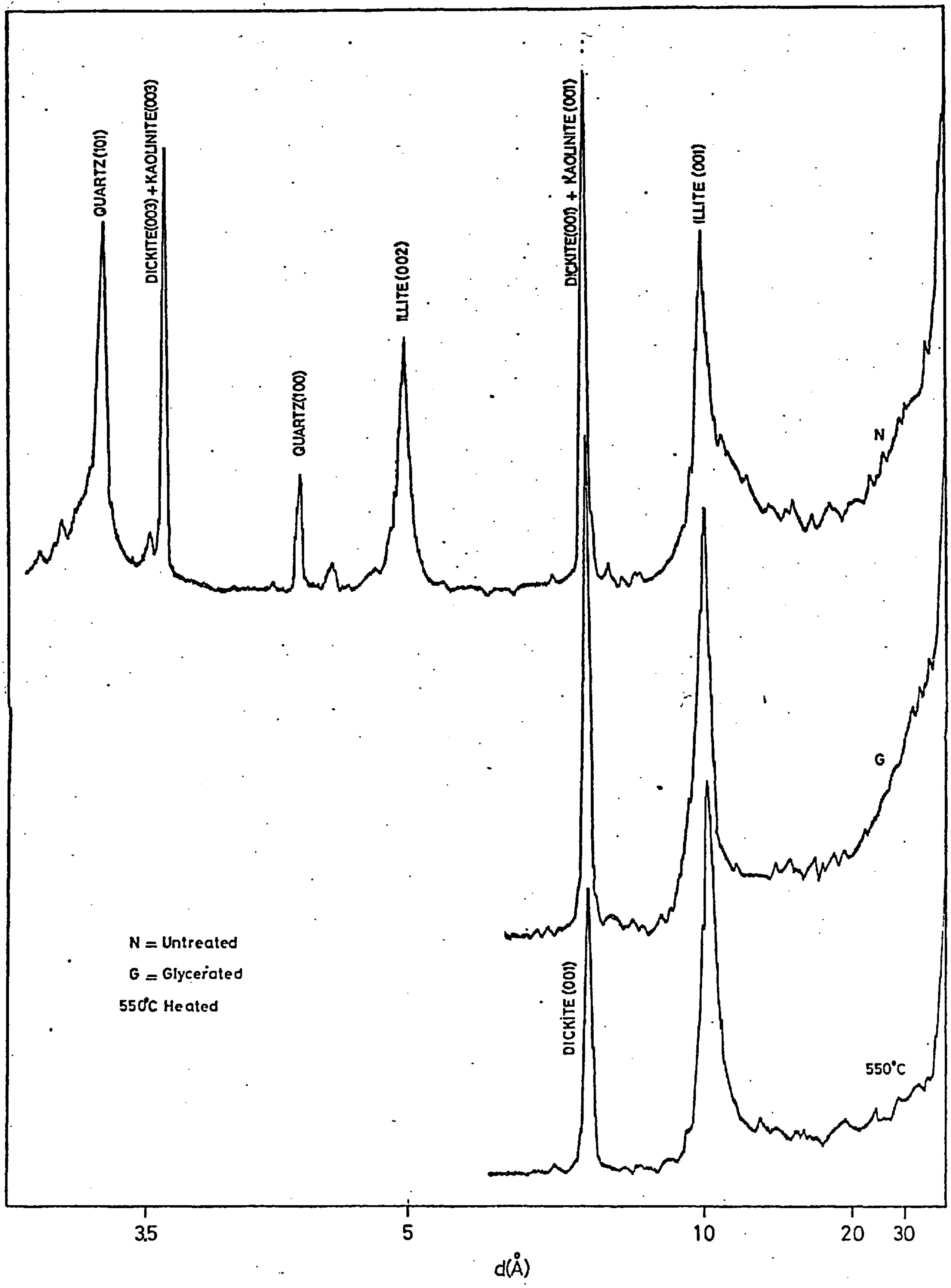


FIG.7.5 X-ray diffractograms of the clay fraction of sample no. 601 show the main diffraction peaks of dickite, well crystalline kaolinite and illite.

spacings are not generally affected by glyceration. Carefully executed heating can yield valuable data for identifying chlorite. Generally, upon heating the intensity of the (001) chlorite reflection increases, combined with a slight peak shift to higher angles, while the (002), (003) and (004) reflections decrease as the brucite layer decomposes.

It is well known that chlorites are represented by four polymorphs. Polymorphism of chlorites has been examined in detail by Brown and Bailey (1962, 1963); Shirozu and Bailey (1965) and Lister and Bailey (1967). The identification of chlorite polymorphs in the rocks studied was very difficult. This is due to the overlapping of the characteristic peaks of such polymorphs by the peaks of illite and kaolinite. Swelling chlorite was identified in few samples. This type of chlorite has been described by Stephen and MacEwan (1951). It shows a 13.6 Å basal spacing and generally behaves like other chlorites on heating, except for the basal spacing which expends on glyceration to 14.2 Å.

Pyrophyllite:

This mineral was identified on the basis of its (002) reflection at 9.16 Å mainly. It is not affected either by glyceration nor by heating (Fig. 7.6).

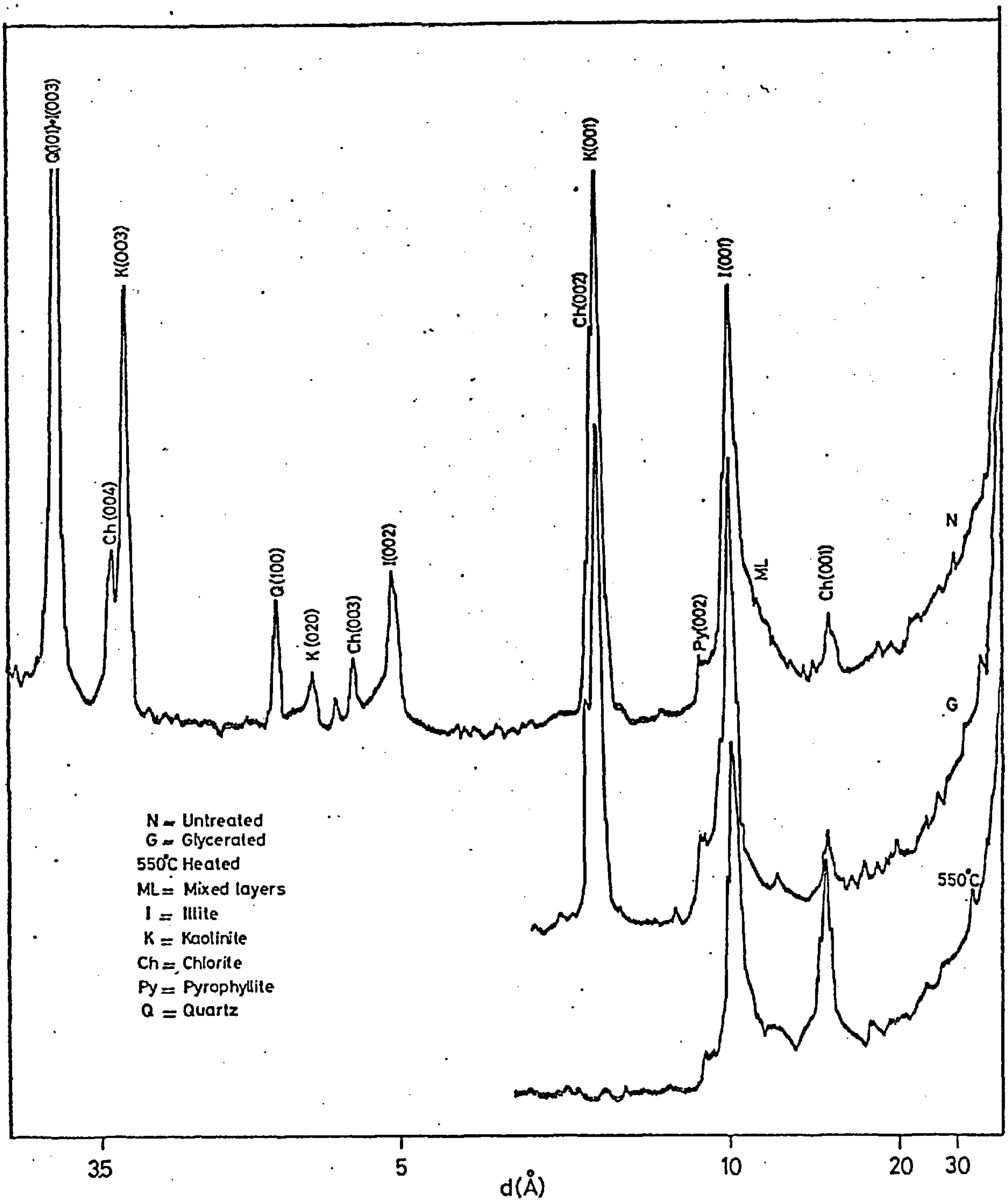


FIG.7.6 X-ray diffractograms of the clay fraction of sample no.815

4. Semi-quantitative Analysis of Clay Minerals in Oriented Mount:

Quantitative X-ray diffraction analysis of mineralogical mixtures involves comparison of the intensities of strong reflections characteristic of each mineral present in the mixture. In the last twenty years many methods of quantitative X-ray diffraction analysis of clay minerals have been published; Gibbs (1967), Pierce and Siegel (1969), Schultz (1964), Jones et al. (1954), Devine et al. (1972), Biscaye (1965) and Shaw (1971).

Quantitative analysis of the clay minerals in the present study was done by an adoption of the methods described by Schultz (1964) and Shaw (1971).

The following procedure was used:-

(a) Measure the peak areas of the 10 Å, 7 Å and 3.58 Å in the untreated air-dried oriented sample. The 10 Å peak area will include the area of the low angle shoulder. Peak area of the 10 Å reflection will be equivalent to the intensity of the (001) illite reflection plus the (001) reflection of the illite-montmorillonite mixed layer mineral. The 7 Å peak area will be equivalent to the integrated intensity of the (001) kaolinite and (002) chlorite reflections.

The 3.58 Å peak area will be equivalent to the intensity of the (002) kaolinite reflection.

(b) Measure the peak area of the 10 Å reflection after glyceration of the sample. This will be equivalent to the integrated intensity of the (001) illite reflection only.

(c) The (002) kaolinite reflection was adjusted to (001) equivalent by multiplying by 1.25 (Bucke and Mankine, 1971). Also, the peak area of 10 Å in the untreated sample was increased threefold to allow direct comparison with the peak areas of the chlorite and kaolinite.

The following equations were used in the calculations:-

$$\%\ \text{Illite} + \text{illite-montmorillonite mixed layer} = \frac{I\ 10\ \text{Å untreated} \times 3}{I\ 10\ \text{Å untreated} \times 3 + I\ 7\ \text{Å}} \times 100$$

$$\%\ \text{Illite} = \frac{I\ 10\ \text{A glycerated} \times 3}{I\ 10\ \text{Å untreated} \times 3 + I\ 7\ \text{Å}} \times 100$$

$$\%\ \text{Illite-montmorillonite} = \%(\text{Illite} + \text{illite-montmorillonite}) - \%\ \text{Illite}$$

$$\%\ \text{Kaolinite} + \text{chlorite} = 100 - \%(\text{Illite} + \text{illite-montmorillonite})$$

$$\%\ \text{Kaolinite} = \frac{I\ 3.58\ \text{Å} \times 1.25}{I\ 7\ \text{Å}} \times \%(\text{kaolinite} + \text{chlorite})$$

$$\%\ \text{Chlorite} = \%(\text{kaolinite} + \text{chlorite}) - \%\ \text{kaolinite}$$

Because of the semi-quantitative nature of the analysis, the relative clay mineral contents of the studies samples are only quoted to the nearest 1%.

The areas under the peaks were measured using a polar planimeter. In measuring the peak areas the background level beneath the peaks has to be projected by joining up the background levels on either side.

5. Quantitative Analysis of the whole Rock Material:

The whole rock material of the argillaceous rocks of Millstone Grit and Coal Measures Series were quantitatively analysed. Analysis was carried out on the diffractograms of randomly oriented mount. The principal mineral constituents found in the samples are: quartz, siderite, dolomite, calcite, feldspars, muscovite and 'clay minerals'. The clay minerals present are the same as in the $< 2\mu$ fraction, i.e. illite, mixed layer minerals, chlorite and kaolinite.

A semi-quantitative estimation of the relative amounts of these minerals was made based on the method outlined by Schultz (1964). Briefly, this involved comparing the intensities of particular diffraction peaks of various minerals when present as 100% of the specimen with the intensities of the same peaks in the samples being analysed.

The areas of the characteristic peaks of the minerals present were measured and will be equivalent to their intensities. They were then multiplied by their correction factors before direct comparison. Table 7.1 gives the characteristic peaks of each mineral and its correction factor after Schultz (1964).

TABLE 7.1 Factors for X-ray semi-quantitative estimation of minerals (after Schultz, 1964).

Mineral	hkl	d(hkl)Å	Intensity Factor (Cps per 100%)	Correction Factor
Quartz	101	3.34	2000	x 1
Feldspars	040	3.18	1000	x 2
Calcite	104	3.04	1000	x 2
Dolomite	104	2.88	1000	x 2
Siderite	104	2.74	1000	x 2

It was difficult to measure the peak area of the (001) muscovite due to the fact that it is superimposed on the broad diffuse (001) illite and illite-montmorillonite mixed layer mineral reflection. Therefore it is calculated with the clay minerals.

For the estimation of the "clay minerals" present in the "whole rock" samples, the variation made by Shaw (1971) to the Schultz method was used. This involved measuring the intensities of the illite and muscovite (001) reflections, the kaolinite (001) and the chlorite (002) reflections, correcting them for differences in diffracting ability relative to each other, and then comparing the total corrected intensity directly with that of the quartz (101) reflections.

DISTRIBUTION OF THE CLAY MINERALS

1. Clay Minerals in the Carboniferous Limestone rocks:

Clay minerals have been separated from 44 samples representing five lithotypes. These are: 22 samples of biomicrites, 3 samples of micrites, 13 samples of biosparites and oosparites, 9 samples of dolomites and 7 samples of dolomicrites. Results of qualitative and quantitative analyses of the clay minerals in the $< 2\mu$ fraction of these rocks are given in Table 7.2. Fig. 7.7 shows the average relative frequencies of the clay minerals in the various lithotypes.

Petrographic studies of these carbonate rocks revealed that clay minerals are present in two forms. The first form, and the most common, is as very minute particles mixed with micrite. These clay particles may be responsible in part for the cloudy appearance of the micrite. In dolomites (late diagenetic dolomite) clay particles are present as inclusions in the dolomite crystals. The second form is as cavity fillings.

The following clay minerals were identified in most of the carbonate rocks studied: illite, illite-montmorillonite

TABLE 7.2 CLAY MINERAL COMPOSITION AND ILLITE CHARACTERISTICS IN THE CARBONIFEROUS LIMESTONE LITHOTYPES

Lithotype	Locality	Sample No.	Illite %	Mixed Layer %	Kaolinite %	Chlorite %	Expendable Layer %	Sharpness Ratio	$\frac{I(002)}{I(001)}$
BIOMICRITES	Gower	616	92	8	-	-	20	4.6	1.0
		625	100	-	-	-	-	2.2	0.3
		622	65	20	15	-	30	3.5	0.4
	Kidwelly	674	100	-	-	-	-	3.0	0.6
	Cilwrchen	655	75	22	-	3	-	3.5	0.63
		654	86	14	-	-	-	3.6	0.7
		659	80	10	-	10	20	3.2	0.7
	Penwyllt	635	100	-	-	-	-	-	-
		633	72	20	-	8	50	2.8	0.8
		630	60	10	30*	-	40	2.0	0.5
	Merthyr Tydfil	105	80	20	-	-	35	2.7	1.0
		102	50	50	-	-	-	2.0	-
		103	70	18	10*	2	40	2.8	0.9
	Ystrad	76	60	31	9	-	30	2.0	0.6
		77	30	60	-	10	25	1.1	1.0
		BY55	45	45	10	-	30-50	2.5	0.7
		BY44	40	55	5	5	40-50	2.0	0.6
		BY37	75	10	-	15	25-40	2.7	0.6
		BY48	40	50	10	-	25-50	3.2	0.7
		BY54	55	35	-	10	25-50	2.0	0.6
	Ruthin	C04	55	30	-	15	35	3.7	1.0
		M5	50	50	-	-	50	2.5	0.8
Micrite	Gower	601	70	10	20*	-	25	5.0	0.8
	Ruthin	C02	100	-	-	-	-	2.5	0.45
		C01	90	10	-	-	30	2.2	0.7
BIOSPARITE	Gower	609	80	-	20	-	-	4.7	0.6
	Kidwelly	671	95	-	5	+	-	5.0	0.5
	Torcoed	666	97	-	3	-	-	3.5	0.5
	Penwyllt	626	90	10	-	-	20	2.2	0.45
	Ystrad	BY64	60	36	4	-	35	1.7	0.6
		BY62	93	-	2	5	-	1.8	0.6
	Brynmawr	BY29	90	10	-	-	25	2.4	1.0
		BY32	55	40	-	5	25-40	4.2	0.6
		BY43	75	-	-	25	-	2.8	0.5
	Ruthin	RU3	70	20	-	10	-	-	-
		M2	100	-	-	-	-	2.0	0.6
		LNS2	55	40	5	-	25-40	2.2	0.6
		B01	100	-	-	-	-	2.2	0.5

TABLE 7.2 (Cont.)

Lithotype	Locality	Sample No.	Illite %	Mixed Layer %	Kaolinite %	Chlorite %	Expandable Layer %	Sharpness Ratio	$\frac{I(002)}{I(001)}$
DOLOMITES	Gower	608	100	-	-	-	-	1.6	0.5
	Brynmawr	By22	50	40	10	-	50	1.5	0.6
	Risca	RI3	70	-	30	-	-	1.7	0.6
		RI1	95	-	5	-	-	2.8	0.8
		Cum3	100	-	-	-	-	-	-
	Tongwynlais	A7	87	10	3	-	20-40	2.0	0.55
		SA17	100	-	-	-	-	-	-
	Ruthin	M1	56	44	-	-	50	1.3	0.6
	Forest of Dean	1025	95	5	-	-	-	1.9	0.42
DOLOMICRITE	Brynmawr	BY30	60	40	-	-	30	3.0	0.7
		BY24	100	-	-	-	-	1.8	0.7
	Ystrad	BY65	62	30	8	-	30	4.4	0.85
	Tongwynlias	A13	95	5	-	-	20	1.9	0.85
		A9	65	30	-	5	30	2.1	0.5
	Ruthin	AR4	60	40	-	-	25	1.8	0.97
		AR2	50	20	30	-	20	1.3	1.0

* Dickite present.

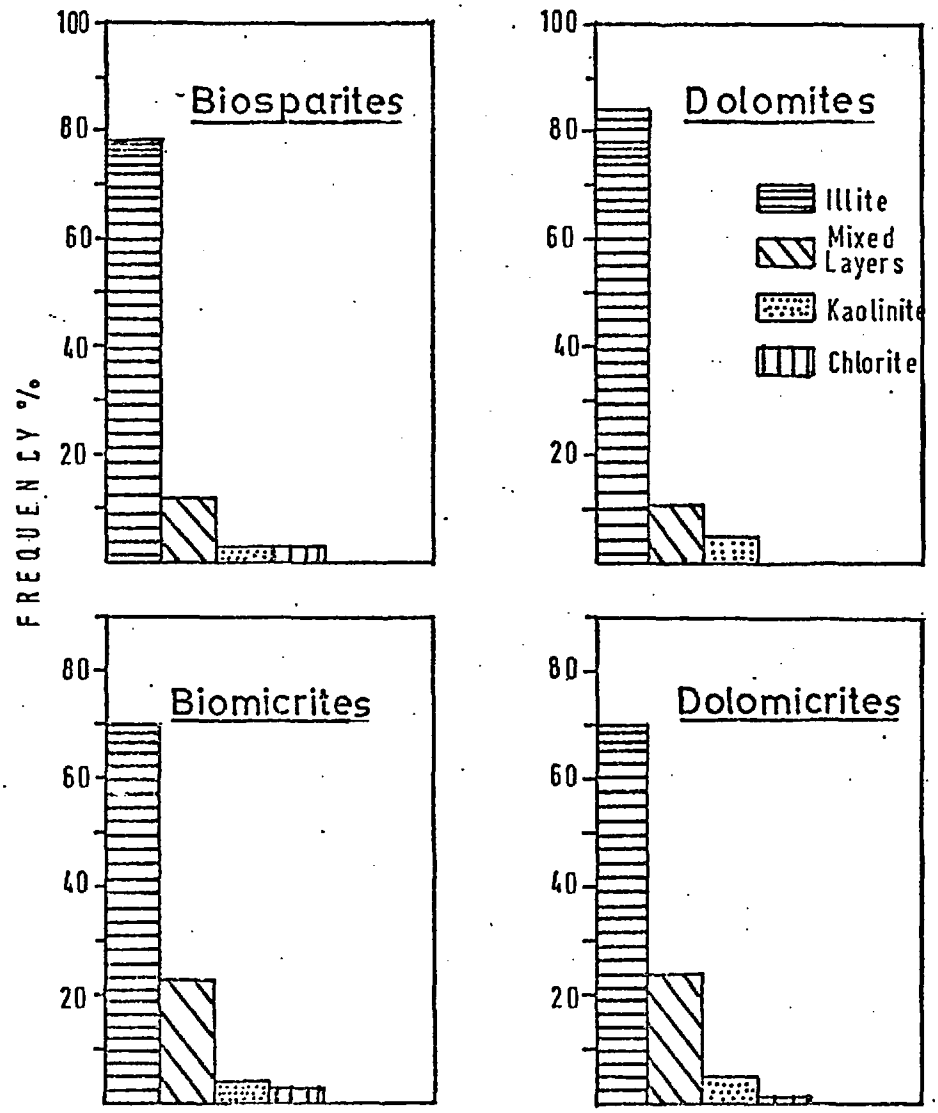


FIG.7.7 Average frequency% of the clay minerals in the carbonate lithotypes of the Carboniferous Limestone

mixed layer, chlorite and kaolinite. Dickite was identified in few samples.

Illite:

Illite is the most frequent clay mineral in all of the carbonate lithotypes. There is no significant difference in the illite frequency in the different lithotypes. It averages about 70% in micrites, biomicrites and dolomicrites; about 78% in biosparites and oosparites; and 84% in dolomites. Interpretation of the X-ray diffractograms of randomly oriented mounts of 10 samples has revealed that illite is present as 2M form.

Illite-montmorillonite mixed layer:

It is represented by broad diffuse peak with d spacing ranging from 10.3 Å to 11.7 Å. Its basal reflections do not give rational indices, so that random interstratification can be assumed. Illite-montmorillonite mixed layer is present with an average of 12% in biosparites and oosparites, 11% in dolomicrites, 23% in micrites and biomicrites, and 24% in dolomicrites. Thus, this mineral is more frequent in micritic rocks rather than in sparites and dolomites.

Expendable layer percentages were calculated by using Weaver's data (1956), Fig. 7.3 . It was found that they range from 20% to 50%. Illite-montmorillonite mixed layer clays having various amounts of expendable layers were found to be present together in the same sample.

Kaolinite:

It is present in some of the samples studied and is generally represented by small amounts. It was found that kaolinite ranges in average from 3 to 5% in the various carbonate lithotypes.

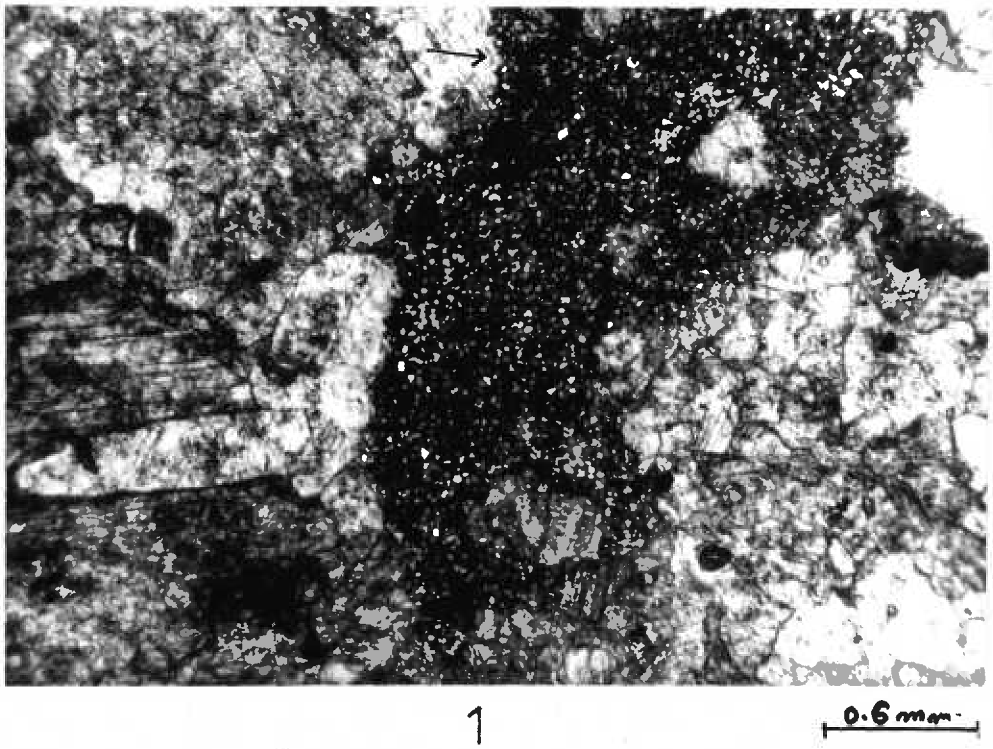
Petrographic studies showed that well crystalline vermicular kaolinite is present as a cavity filling in some biomicrite and dismicrite in the anthracite zone. X-ray analysis confirmed the occurrence of this mineral and also revealed that kaolinite is mixed with dickite in these rocks (Plate 35).

Chlorite:

Generally chlorite is present in small amounts in some of the rocks studied. It occurs with an average of about 3% in biomicrites, biosparites and oosparites, and dolomicrites. Chlorite was found to be completely absent in the dolomites.

2. Clay Minerals in the Millstone Grit Rocks:

Quantitative and qualitative analysis of clay minerals in the $< 2\mu$ fraction has been carried out on 16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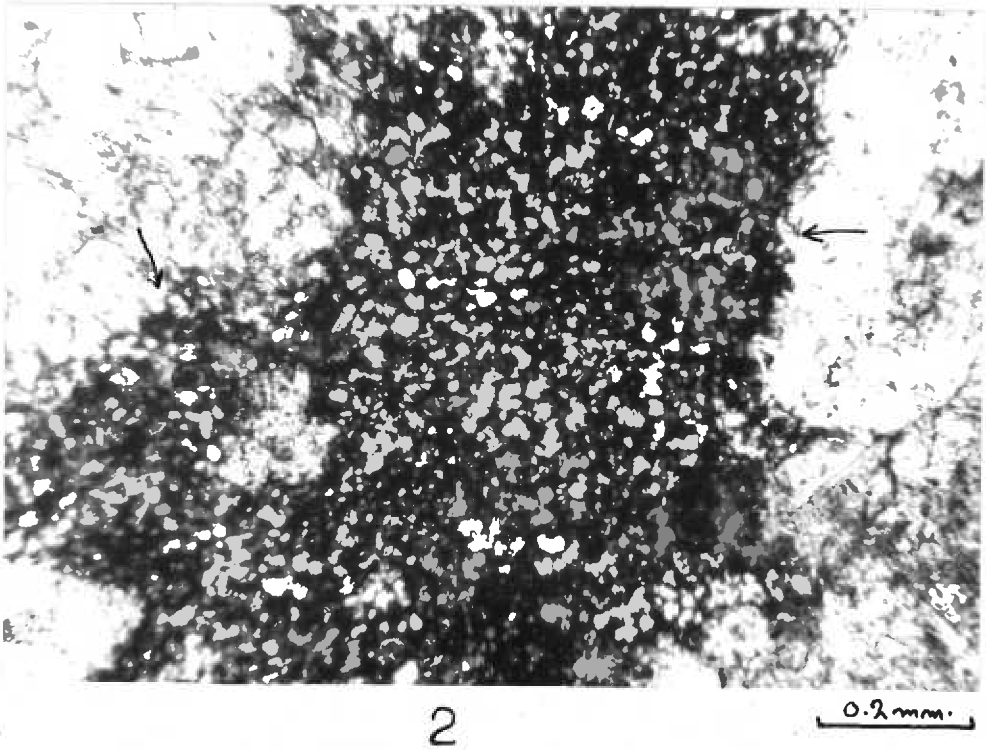
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PLATE 35 Figs. 1 and 2

Kaolinite and dickite filling cavity in recrystallized biomicrite. Note corrosion of calcite by kaolinite (arrow). Sample 615.

samples representing the main three lithotypes of the Millstone Grit Series (sandstones, siltstones and argillaceous rocks). Litharenites were omitted because it was impossible to separate the clay matrix in the presence of the detrital argillaceous rock fragments. The whole rock material of the argillaceous rocks was also examined.

a. Argillaceous Rocks:

Argillaceous rocks of the Millstone Grit Series are represented mainly by shales. The following is the distribution of clay and non-clay minerals in these rocks.

X-ray analysis of the whole rock material of these rocks revealed that they consist of clay minerals and muscovite, quartz, feldspars, siderite and dolomite, Table 7.3. Clay

TABLE 7.3 WHOLE ROCK COMPOSITION OF THE SHALES OF THE MILLSTONE GRIT SERIES

Sample No.	Clay %	Quartz %	Feldspars %	Siderite %	Dolomite %
88	62	11	12	15	-
614	65	12	10	2	11
623	65	30	-	5	-
678	63	34	-	3	-
681	68	21	8	3	-
679	60	29	11	-	-
661	59	34	7	-	-
663	66	34	-	-	-

minerals are the most frequent and range from 59 to 68%, with an average of 63%. Quartz ranges from 11 to 34%, with an average of 24%. Feldspars are present in some samples with an average of 10%. Siderite is present in most of the samples with an average of 5%, and dolomite was identified in one sample only.

Clay minerals present in the $< 2\mu$ fraction of the shales are: illite, illite-montmorillonite mixed layer, kaolinite and chlorite, Table 7.4. Illite (2M) is the most frequent and is present with an average of 56%. Illite-montmorillonite,contains from 20 to 50% of expendable layer averaging 35%. Kaolinite is present with an average of 3%, and chlorite averages 6%.

b. Siltstones and Sandstones:

By contrast to the argillaceous rocks, the clay fraction of these rocks is rich in chlorite and kaolinite. They are present with an average of 12% and 15%, respectively, (Fig. 7.9). Occurrance of kaolinite and chlorite has been confirmed by the petrographic studies. In most siltstones and sandstones, especially quartz arenite, vermicular kaolinite is present filling pore spaces.

TABLE 7.4 CLAY MINERAL COMPOSITION AND ILLITE CHARACTERISTICS IN THE $< 2\mu$ FRACTION OF THE VARIOUS LITHOTYPES OF THE MILLSTONE GRIT SERIES

Lithology	Sample No.	Illite %	Mixed Layer %	Kaolinite %	Chlorite %	Expandable Layer %	Sharpness Ratio	$\frac{I(002)}{I(001)}$
SANDSTONE	652	65	10	-	25	25	-	0.48
	91	66	20	-	14	25-40	5.0	0.40
	631	56	20	24	-	-	-	0.40
	94	84	16	-	-	25	-	-
	84	55	20	15	10	30-50	3.3	0.50
	BY17	16	24	20	40	25	3.0	0.65
SILTSTONE	BY3	54	-	32	14	-	4.3	0.38
	81	41	41	-	18	25-35	3.8	0.48
	BY18	25	45	20	10	30	2.5	0.71
	662	75	13	12	-	20	-	0.50
SHALE	88	75	5	-	20	20	3.5	0.35
	614	45	30	10	15	20-40	3.2	0.60
	623	40	50	-	10	40	2.5	0.60
	678	68	32	-	-	25	2.7	0.25
	681	57	33	8	2	25	4.0	0.50
	679	55	45	-	-	20	2.0	0.54
	661	59	41	-	-	25	2.0	0.22
	663	52	46	2	-	25	3.3	0.57

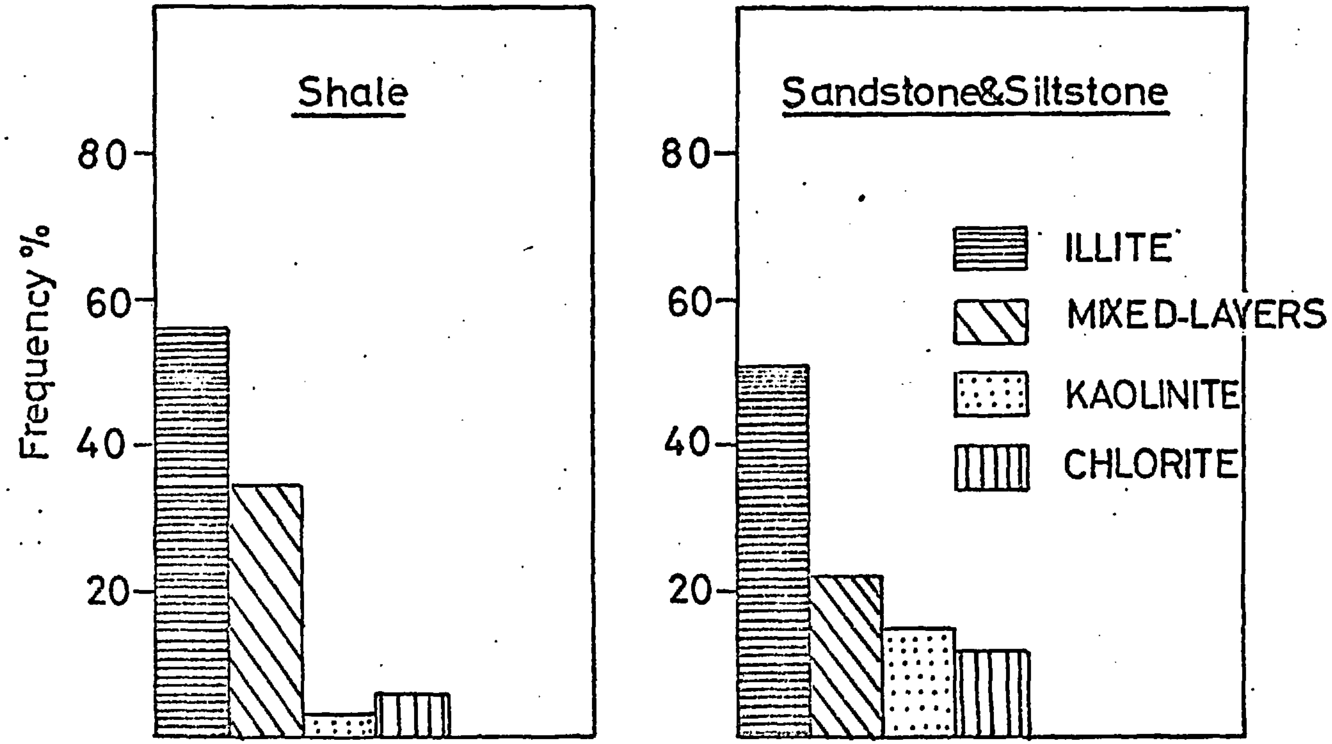


FIG.7.9 Average frequency% of the clay minerals in the Millstone Grit.

3. Clay Minerals in the Coal Measure rocks:

Very little work has been done on the clay mineralogy of the Coal Measure rocks of the South Wales Coalfields. Nagelschmidt and Hicks (1943) studied some shales from the Coal Measures in South Wales Coalfields and they found that they consisted mainly of illite with about 10% kaolinite and 1% quartz. Wilson (1963) studied the clay mineralogy of the underclays of the Eastern part of the coalfields. He found that they consist mainly of kaolinite and illite with smaller amounts of chlorite and mixed layer minerals.

In the present study 147 samples representing the various lithotypes have been studied. The clay minerals in the $< 2\mu$ fraction of all of the samples were quantitatively and qualitatively analysed, and the results are tabulated in Table 7.5. The whole rock material of the argillaceous rocks has also been studied and the results are given in Table 7.6.

a. Argillaceous Rocks:

Whole rock mineralogy:

The minerals identified in the whole rock material of the argillaceous rocks are; clay minerals (and muscovite), quartz, feldspars, siderite, dolomites and calcite. Clay minerals (including muscovite) are the main mineral constituent

TABLE 7.5 CLAY MINERAL COMPOSITION AND ILLITE CHARACTERISTICS IN THE $< 2\mu$ FRACTION OF THE VARIOUS LITHOTYPES OF THE COAL MEASURES SERIES.

	Locality	Lithology	Sample No.	Illite %	Mixed Layer %	Kaolinite %	Chlorite %	Sharpness Ratio	$\frac{I(002)}{I(001)}$
1.	Brynamman	Litharenite	834*	50	-	30	20	9.9	0.45
			830	42	20	18	20	8.0	0.47
			818*	51	15	26	8	6.5	0.50
		Quartzarenite	911	34	44	22	0	-	0.65
			810	38	50	12	T	4	0.52
			829	37	50	-	13	4	0.41
			815*	35	23	31	11	7.7	0.37
		Mudstone	833*	50	21	16	13	5.5	0.35
			822*	50	16	14	20	4	0.45
			824	45	40	12	3	3.7	0.76
			800	40	35	13	12	4.5	0.56
			804	54	36	4	6	4.5	0.60
		Underclay	816*	45	20	29	6	2.6	0.45
			865*	44	33	12	11	6.0	0.45
			910	30	30	40	T	5.0	0.53
			781	52	40	8	T	2.7	0.66
			774	40	43	4	3	2.7	-
			766	55	33	-	12	4.0	0.56
2.	Rhydy Meardy	Quartzarenite	908	40	55	3	2	7	0.44
			858	35	25	30	10	5.5	0.44
		Mudstone	900	93	-	4	3	4.7	0.29
			903	40	48	10	2	2.8	0.68
			907	76	20	2	2	4.2	0.54
			859	45	40	10	5	5.0	0.59
		Underclay	871	55	40	-	5	5.5	0.70
3.	Pantyffnon	Quartzwacke	262	40	50	8	2	4.3	0.50
		Mudstone	260	48	40	10	2	4.5	0.56
			263	53	30	15	2	4.5	0.54
4.	Cwmgwilli	Sandstone	744	60	40	-	-	6.0	0.50

T = Traces.

TABLE 7.5 (Cont.)

	Locality	Lithology	Sample No.	Illite %	Mixed Layer %	Kaolinite %	Chlorite %	Sharpness Ratio	$\frac{I(002)}{I(001)}$
5.	Abernant	Quartzwacke	709	40	55	2	3	4.5	-
		Mudstone	711	55	35	4	6	5.5	-
		Shale	708	57	34	4	5	2.7	0.41
6.	Morlais	Litharenite	749	50	34	14	2	3.2	0.41
			745	76	5	-	19	6.7	0.43
		Quartzwacke	748	60	14	20	6	4.2	0.43
		Mudstone	746	59	34	-	6	5.5	0.47
7.	Brynlliw	Quartzwacke	715	50	10	40	-	5.0	0.37
		Siltstone	713	51	26	13	11	5.5	0.29
8.	Bleanant	Litharenite	722	63	23	-	14	6.6	0.31
			728	67	17	-	16	5.0	0.41
		Mudstone	724	46	34	6	14	5.0	0.56
			727	44	30	20	6	2.0	0.23
			732	50	29	10	11	4.0	0.41
			725	70	-	30	-	4.0	0.27
9.	Treforgan	Mudstone	754	54	30	8	8	4.2	0.47
			759	56	30	7	7	3.5	0.43
10.	Dillwyn	Litharenite	276	34	30	30	6	6.0	0.40
		Mudstone	280	42	33	7	18	5.0	0.40
11.	Bleangwrach	Siltstone	197	45	20	32	3	2.3	0.41
		Mudstone	190	48	30	10	2	2.3	0.42
		Shale	194	55	25	15	5	3.2	0.26
		Underclay	204	58	30	10	2	3.0	0.32
			202	55	33	10	2	3.5	0.26
			200	55	35	8	2	3.0	0.22
			199	65	15	20	-	3.0	-
12.	Mardy	Litharenite	305	72	-	-	28	4.6	0.35
			309	60	15	-	25	7.0	0.31
			307	59	18	-	28	6.7	0.47
		Quartzarenite	354	43	27	28	2	5.6	0.50
			355	45	7	48	-	4.0	0.42
		Siltstone	360	45	25	25	5	3.0	0.24
		Mudstone	350	50	22	16	12	3.6	0.33
			357	60	28	12	-	3.2	0.27

TABLE 7.5 (Cont.)

	Locality	Lithology	Sample No.	Illite %	Mixed Layer %	Kaolinite %	Chlorite %	Sharpness Ratio	$\frac{I(002)}{I(001)}$
13.	Tylorstown	Litharenite	304	34	38	20	8	2.0	0.35
		Siltstone	304A	45	20	20	15	6.0	0.39
		Mudstone	302	46	20	20	13	4.5	0.44
14.	Penrikyber	Mudstone	115	50	38	10	2	3.0	0.42
			114	61	20	15	4	2.0	0.41
			118	60	25	10	5	2.1	0.43
		Shale	116	35	35	30	T	1.5	0.40
			113	65	25	10	T	2.0	0.41
15.	Abercynon	Mudstone	173	60	10	30	-	3.3	0.28
			171	51	31	14	4	3.5	0.31
		Shale	172	40	35	20	5	2.0	0.32
16.	Penallta	Quartzarenite	179	29	21	50	T	2.0	-
		Quartzwacke	181	33	26	41	T	3.0	0.38
		Siltstone	182	50	16	34	-	5.0	0.56
		Shale	188	61	25	14	-	2.0	0.27
17.	Ffaldau	Siltstone	223	47	30	19	4	3.5	0.45
			231	50	32	16	2	3.2	0.31
		Mudstone	229	38	27	16	19	4.0	0.30
18.	Coegnant	Quartzwacke	219	50	20	30	T	3.0	0.43
		Siltstone	213	59	26	15	-	3.4	0.36
		Mudstone	216	60	25	15	T	2.5	0.72
		Underclay	215	40	40	16	2	3.0	0.37
			211	45	30	23	2	3.0	0.39
19.	Marine	Quartzarenite	121	-	-	100	-	-	-
		Mudstone	127	43	42	15	T	2.8	0.45
			134	50	31	16	3	2.2	0.35
			131	45	25	25	5	1.5	0.30
			132	44	32	20	4	2.0	0.32
		Underclay	123	41	40	15	4	2.2	0.36
			130	50	30	17	3	2.3	0.24

TABLE 7.5 (Cont.)

	Locality	Lithology	Sample No.	Illite %	Mixed Layer %	Kaolinite %	Chlorite %	Sharpness Ratio	$\frac{I(002)}{I(001)}$
20.	Bedwellty	Litharenite	560	85	-	-	15	5.5	0.31
		Quartzwacke	568	20	10	65	5	-	0.36
			577	52	30	15	3	1.5	0.18
		Quartzarenite	570	35	45	20	T	1.9	0.16
			576	35	25	30	10	3.0	0.51
			585	30	10	60	T	1.9	0.50
		Siltstone	573	45	30	20	5	1.8	0.31
			582	40	30	20	10	2.0	0.25
			584	40	30	30	T	1.4	0.16
		Mudstone	562	40	25	31	4	1.8	0.15
			580	30	30	30	10	2.5	0.53
			587	40	45	7	8	1.7	0.25
			588	30	35	25	10	2.0	0.39
21.	Bedwas	Quartzarenite	241	32	30	24	4	2.5	0.40
		Quartzwacke	243	22	12	60	4	2.5	0.28
		Siltstone	249	41	9	33	17	3.2	0.38
			235	25	20	53	2	3.8	0.37
		Mudstone	247	50	27	18	5	3.5	0.36
			237	61	18	18	3	2.9	0.38
			234	40	40	18	2	3.5	0.31
			250	50	32	15	3	3.0	0.43
22.	Six Bells	Quartzarenite	154	10	-	90	T	1.9	0.24
			141	30	10	60	T	1.8	0.30
		Siltstone	140	35	20	40	5	2.0	0.27
		Mudstone	153	38	30	28	4	2.2	0.25
			152	50	33	15	2	2.5	0.25
			147	50	32	15	3	2.0	0.38
		Underclay	150	30	37	30	8	2.3	0.47
			144	30	20	50	-	2.0	0.56
			145	40	20	40	T	1.9	0.41
23.	Park Slip	Quartzarenite	1479-3	30	30	40	T	2.0	0.5
		Underclay	2493-10	40	35	20	5	2.5	0.5
			2493-19	40	40	16	4	2.2	0.6

TABLE 7.5 (Cont.)

Locality	Lithology	Sample No.	Illite %	Mixed Layer %	Kaolinite %	Chlorite %	Sharpness Ratio	$\frac{I(002)}{I(001)}$
24. Pentrych	Sandstone	388-2	30	10	45	15	3.0	0.5
		388-5	60	20	20	T	2.0	0.25
		388-6	30	20	50	-	1.8	-
		420-3	50	25	25	-	2.5	0.31
		295-5	30	10	60	-	1.8	-
		224-2	50	25	25	T	1.8	0.24
	Siltstone	388-4	50	25	20	5	2.0	0.60
		420-6	35	25	40	T	1.6	0.40
		224-3	40	30	30	T	1.8	0.30
		224-4	42	30	25	3	1.5	0.37
	Mudstone	388-1	20	15	50	15	1.3	-
		420-1	51	30	15	4	3.0	0.30
		420-4	59	30	8	3	2.0	0.28
		420-5	50	40	10	-	1.8	0.30
		296-1	50	30	20	-	1.9	0.54
		895-1	50	38	10	2	2.5	0.50
	Shale	295-4	56	30	12	2	2.3	0.30
	Underclay	420-2	60	20	18	2	2.0	0.26
		295-3	68	20	10	2	2.2	0.34

T = Traces.

TABLE 7.6 WHOLE ROCK MINERAL COMPOSITION OF THE ARGILLACEOUS ROCKS OF THE COAL MEASURES SERIES

Locality	Sample No.	Lithology	Quartz %	Clay %	Feldspars %	Siderite %	Dolomite %	Calcite %
Brynamman	833	Mudstone	23	62	7	-	8	-
	800	Mudstone	21	65	6	-	8	-
	824	Mudstone	32	60	7	-	-	-
	822	Mudstone	23	66	6	-	5	-
	804	Mudstone	17	57	5	20	-	-
	781	Underclay	17	57	7	12	7	-
	766	Underclay	26	62	12	-	-	-
	816	Underclay	23	66	4	7	-	-
	865	Underclay	16	60	10	10	4	-
	774	Underclay	19	72	9	-	-	-
Rhydy Meardy	900	Mudstone	19	70	6	-	5	-
	907	Mudstone	23	66	5	-	5	-
	903	Mudstone	24	76	-	-	-	-
	859	Mudstone	16	78	6	-	-	-
Pantyffynon	260	Mudstone	22	72	3	-	3	-
	259	Mudstone	21	62	3	14	-	-
Abernant	708	Shale	31	60	9	-	-	-
	711	Mudstone	23	66	5	6	-	-
Morlais	746	Mudstone	16	57	7	14	3	3
Bleanant	724	Mudstone	22	68	5	-	5	-
	732	Mudstone	23	62	4	7	4	-
	727	Mudstone	24	76	-	-	-	-
	725	Mudstone	20	54	9	17	-	-
Treforgan	754	Mudstone	23	50	5	17	5	-
	759	Mudstone	22	65	3	10	-	-
Bedwellty	566	Mudstone	33	67	-	-	-	-
	588	Mudstone	23	77	-	-	-	-
	580	Mudstone	34	66	-	-	-	-
Bedwas	250	Mudstone	18	55	4	23	-	-
	237	Mudstone	23	56	6	9	6	-
	234	Mudstone	24	32	-	44	-	-

TABLE 7.6 (Cont.)

Locality	Sample No.	Lithology	Quartz %	Clay %	Feldspars %	Siderite %	Dolomite %	Calcite %
Six Bells	153	Mudstone	25	72	3	-	-	-
	150	Underclay	22	48	2	24	2	2
Park Slip	2493-19	Underclay	20	68	4	6	2	-
	2493-10	Underclay	15	77	2	5	1	-
Pentrych	895-1	Mudstone	24	66	7	-	3	-
Dillwyn	280	Mudstone	34	56	10	-	-	-
Bleangwrach	190	Mudstone	17	49	6	25	3	-
	202	Underclay	20	46	4	30	-	-
Mardy	350	Mudstone	17	53	9	21	-	-
Tylorstown	302	Mudstone	18	55	9	18	-	-
Penrikyber	115	Mudstone	11	60	4	25	-	-
Abercynon	171	Mudstone	29	57	14	-	-	-
	173	Mudstone	12	22	5	61	-	-
Penallta	188	Shale	6	89	5	-	-	-
Ffaldua	221	Mudstone	14	44	5	34	3	-
	216	Mudstone	30	63	7	-	-	-
	211	Underclay	33	65	5	-	-	-
	215	Underclay	15	82	3	-	-	-
Coegnant	229	Mudstone	21	62	17	-	-	-
	225	Mudstone	24	70	6	-	-	-
Marine	127	Mudstone	20	60	-	15	5	-
	132	Mudstone	28	72	-	-	-	-
	134	Mudstone	28	51	9	9	3	-
	123	Underclay	5	83	5	-	7	-
	130	Underclay	27	57	4	8	4	-

of these rocks and have an average of 63%. Quartz is present with an average of 21%. Feldspars, siderite and dolomite are present in small amounts. They have an average of 5%, 10% and 2%, respectively. Calcite was identified in two samples only. Fig. 7.10 shows the average mineral composition of the whole rock material of the various argillaceous lithotypes.

Clay mineralogy:

The clay minerals in the $< 2\mu$ fraction of the argillaceous rocks consist mainly of illite, regularly interstratified illite-montmorillonite, randomly interstratified illite-montmorillonite, kaolinite and chlorite. Pyrophyllite was identified in some rocks. Illite and illite-montmorillonite are the most frequent minerals; they average about 50% and 30%, respectively. Kaolinite averages about 16%, while chlorite is the least frequent mineral and has an average of about 4%.

b. Siltstones and Sandstones:

Clay minerals separated from siltstones, quartzarenites, and quartzwackestones were found similar to those in the argillaceous rocks. The main difference between the argillaceous rocks, and sandstones and siltstones is the abundance of kaolinite in the latter rocks (Fig. 7.11). Kaolinite has an average of about 40% in the quartzarenites. Abundance of kaolinite in these rocks was confirmed by the

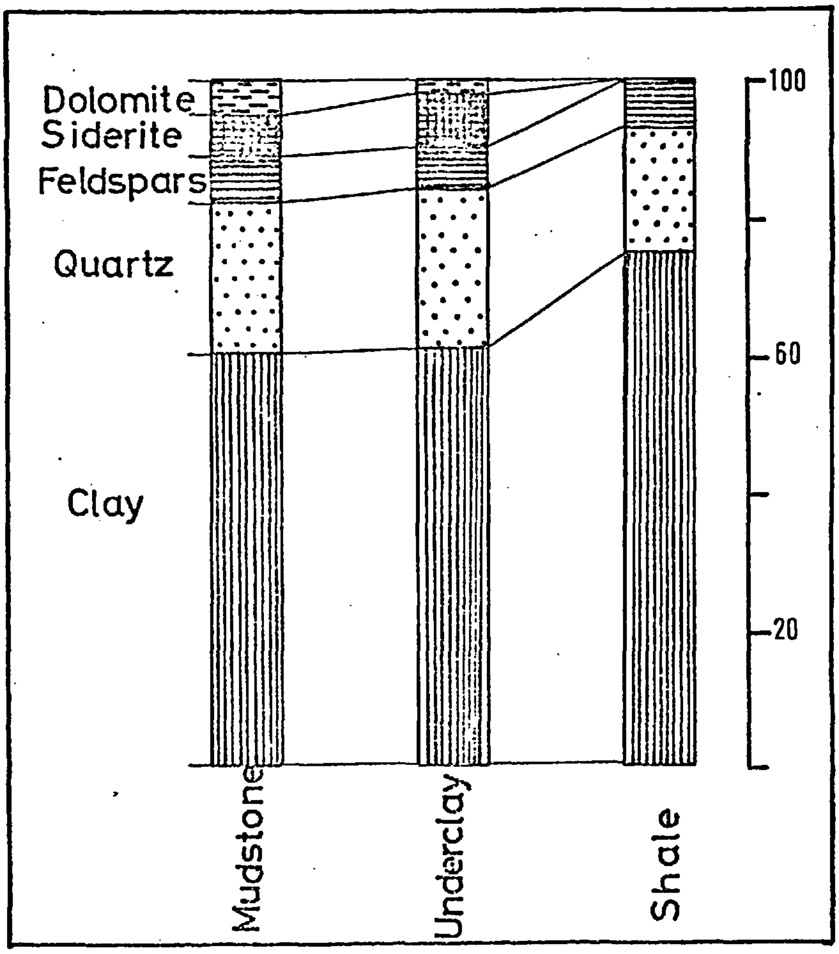


FIG.7.10 Average frequency of the various minerals forming the whole rock material of the argillaceous rocks of the Coal Measures.

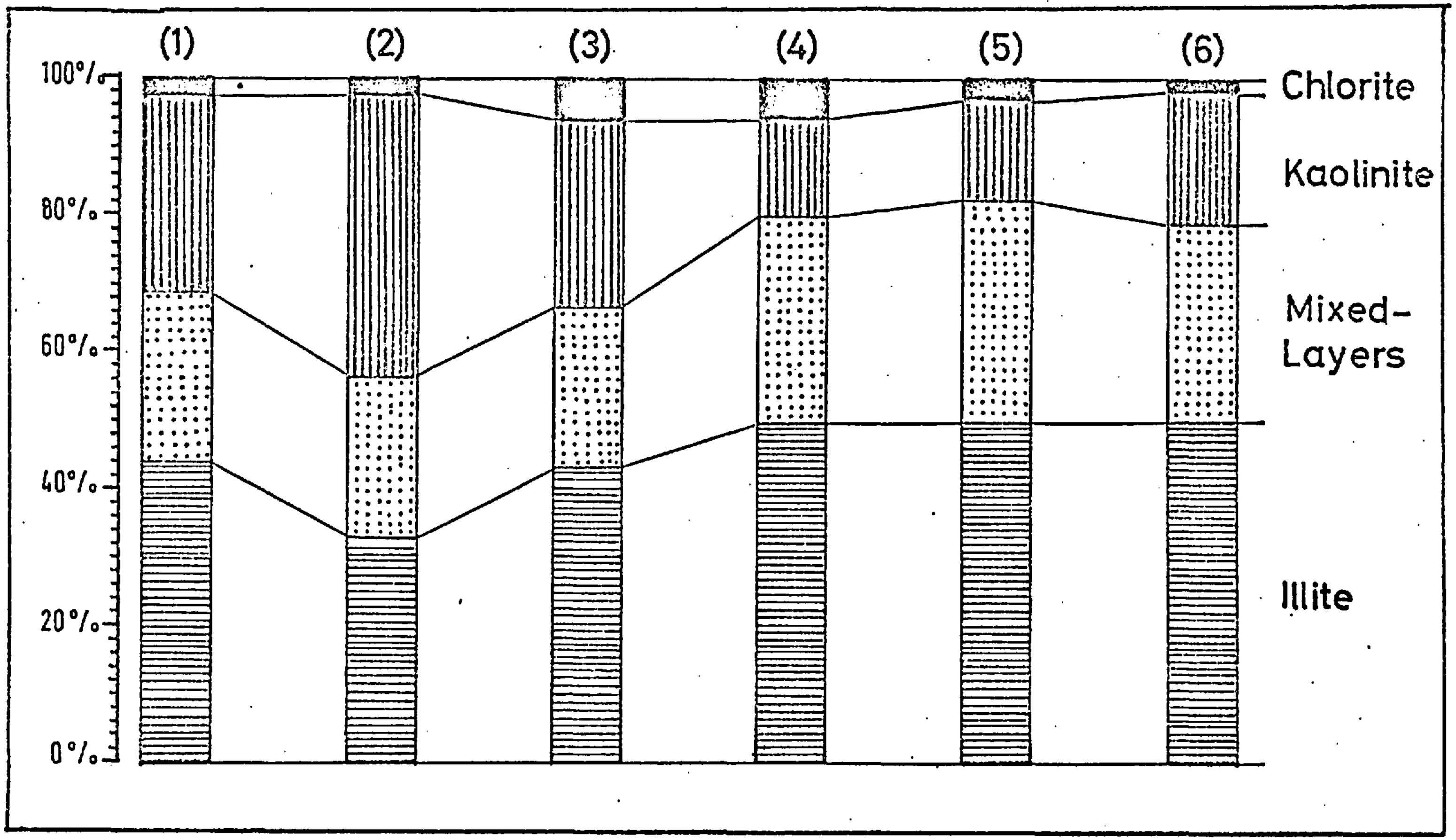


FIG.7.11 Average frequency % of the clay minerals in the <2μ fraction of the various lithotypes of the Coal Measures.

1) Quartzwackes (Av. of 17 samples)	4) Mudstones (Av. of 53 samples)
2) Quartzaremites (Av. of 12 samples)	5) Underclays (Av. of 18 samples)
3) Siltstones (Av. of 17 samples)	6) Shales (Av. of 6 samples)

petrographic studies, where vermicular kaolinite was found to be present as pore-filling cement in most of the quartzarenites. The quartzwackestones usually contain less amounts of kaolinite (24%).

DIAGENESIS OF CLAY MINERALS

Clay minerals in the rocks studied can be of two main genetic types. These are: Allochtonous and Autochtonous clay minerals.

Allochtonous Clay Minerals:

Detrital clays are predominant in most of the rocks studied. These clays could be derived from weathering horizons and soils of the parent rocks. Detrital clays in the carbonate rocks could be in part derived from the weathering horizons of the shales of the lower Limestone Shale Group. They could also be derived from soils which covered limestone exposures in the Carboniferous Limestone time. These soils are preserved in some localities as thin beds of seatearth within the carbonate sequence.

In Millstone Grit and Coal Measures rocks, argillaceous rocks of the older beds were most probably the source of the detrital clays of the younger beds.

Autochtonous Clay Minerals:

These are clay minerals which have been formed after the deposition and during the diagenesis of the sediments. Two processes were responsible for the formation of these clay minerals, namely; by transformation of detrital clay and non-clay mineral particles, and by direct precipitation from pore solutions.

(a) Transformation processes:

These processes took place just after the deposition of the detrital components of the sediments, and before their complete lithification. By these processes some of the non-clay minerals were altered to clay minerals, and detrital clays could be altered to another clay mineral. Two processes were distinguished, namely; degradational alteration and aggradational alteration.

Degradational alteration is responsible for the transformation of high temperature metamorphic and igneous minerals to clay minerals. It is represented in the rocks studied by: (1) transformation of muscovite to illite, and (2) transformation of muscovite to kaolinite. These processes have been described petrographically in Chapter V . It is assumed that a great quantity of the illite and mixed layer minerals in the terrigenous rocks of the Millstone Grit and Coal Measure rocks could have been formed by the degradation of detrital muscovite during the early stage of diagenesis. Smoot (1960) showed that mixed layers become more abundant during the diagenesis of American Pennsylvanian sandstones. In the micaceous siltstones kaolinitisation of muscovite flakes is suggested as the most important process responsible for the abundance of the kaolinite in these rocks.

Aggradational alteration mechanism was described by Weaver (1959). It is suggested that this process is responsible for the transformation of formerly deposited montmorillonite to illite and/or illite-montmorillonite mixed layer. This can be the answer for the absence of montmorillonite in all of the Carboniferous rocks of South Wales. Aggradational transformation can take place during sedimentation and diagenesis. It was very difficult to distinguish that which took place during sedimentation from that formed during diagenesis, because the two are superimposed.

(b) Chemical precipitation process:

This process is responsible for the precipitation of clay minerals from pore solution during the diagenesis. Clay minerals formed by this process are frequent in both the carbonate and non-carbonate rocks. It was found that vermicular kaolinite is the only mineral formed by this process.

In the carbonate rocks, petrographical studies revealed that kaolinite is present as pore-filling mineral. It occurs as well crystallized coarse vermicular aggregates. It was noticed also that these cavities are formed partially by dissolution of calcite. Schroeder et al. (1968) reported the occurrence of similar kaolinite and dickite in the Pennsylvanian limestone of South-eastern Kansas. They suggested that these minerals have been precipitated from heated groundwater, possibly mixed with magmatic waters. Authigenic kaolinite and dickite in the studied carbonate rocks are most probably precipitated from acidic hot water, which has partially dissolved the carbonate firstly forming the vugs and then precipitated kaolinite and dickite.

INCIPIENT METAMORPHISM OF CLAY MINERALS

Introduction

In the last few years many authors have studied the alteration of the clay minerals during the incipient metamorphism stage. Kossovskaya and Shutov (1970) summarised some of the Russian work on the alteration of terrigenous rocks during deep burial. Desegonzac (1970) in his review of the transformation of clay minerals during diagenesis and low-grade metamorphism has mentioned most of the important works in this field of study.

Formation of new clay minerals and recrystallization of illite are the main changes in the clay minerals that have taken place during incipient metamorphism of the rocks studied.

(1) Mineralogical Changes:

X-ray analysis of clays separated from various lithotypes revealed that there is a close relationship between authigenesis of clay minerals and the degree of incipient metamorphism of the rocks studied, as indicated from coal ranks.

The following changes in the clay minerals were observed in the rocks studied:-

(i) In the carbonate rocks, the expandable layer percentages of the randomly interstratified illite-montmorillonite clay minerals decrease in the direction of increasing coal rank and therefore the degree of incipient metamorphism. The same phenomenon was reported by Burst (1959) in the clay minerals of the Eocene of the Gulf Coast (Texas and Louisiana). He found that the ability of the illite-montmorillonite mixed layers to expand decreased gradually with increase in depth until it almost disappeared below 4,500 m., and only illite and chlorite were observed.

(ii) In the terrigenous rocks of the Coal Measures it was found that regular mixed layer illite-montmorillonite (Allevardite) started to appear in the anthracite zone. It is most probable that the abundant randomly interstratified illite-montmorillonite has been altered to allevardite by increasing the grade of metamorphism. Long and Neglia (1968) reported in the Miocene of Gagliano (Sicily) that illite-montmorillonite mixed layer is re-ordered gradually at depth from 2,000 m. to 3,000 m. This was also observed by Desegonzac (1969) in other places.

(iii) The appearance of pyrophyllite in small amounts in the terrigenous rocks of the Coal Measures in the anthracite area. This mineral was reported in the anchiozone of the Paleozoic rocks in the Sahara, Desegonzac (1969) and Chemaux et al. (1970).

(iv) Well crystalline kaolinite and dickite is observed in the carbonate rocks of the anthracite zone.

(v) No evident relationship is noted between variation in the clay minerals and the depth of burial in the area studied.

(2) Crystallinity of Illite:

Introduction:

Illite crystallinity is a parameter sufficiently sensitive to give evidence of metamorphic recrystallisation well before classical petrographical analysis, as shown by Weaver (1960). This technique was applied to the Jurassic "terres noires" of the Alps (Desogonzac et al. (1966), Desogonzac (1969)), the Northern Pyrenees (Kubler, 1966) and the Paleozoic of Northern France (Desegonzac et al., 1968). The use of statistical crystallinity measurements is a certain index of the zone of thermal influence of metamorphism.

Measurements of the degree of crystallinity:

The degree of crystallinity of illite was used by many authors as an indication of the degree of metamorphism in sedimentary rocks. Weaver (1960) quantified the crystallinity of illite by measuring the thickness of the 10 Å peak. He calculated it by dividing the peak height at 10 Å by the peak height at 10.5 Å, and called it the sharpness ratio. Weaver used the sharpness ratio of the 10 Å peak to determine the relative degree of metamorphism in the paleozioc rocks of the Quachita mountain, U.S.A. Table 7.7 shows Weaver's sharpness ratio of 10 Å peak and the related degree of metamorphism.

TABLE 7.7 THE RELATIONSHIP BETWEEN SHARPNESS RATIO OF 10 Å PEAK AND DEGREE OF METAMORPHISM.

Degree of Metamorphism	Sharpness Ratio
Unmetamorphosed rocks	2.3
Incipient metamorphism	2.3
Incipient to weak metamorphism	4.5
Weak metamorphism	6.3
Low-grade metamorphism	12.1

Kubler (1966) has quantified the crystallinity of illite by measuring the width of the 10 Å peak at its half height. This measure is known as illite crystallinity index. Unfortunately Kubler's crystallinity index is dependent upon the experimental conditions, which vary as an inverse function of the crystallinity. In the present work Weaver's sharpness ratio has been used. The measured sharpness ratios of the studied samples are tabulated in Tables 7.2, 7.4 and 7.5.

Sharpness ratio of illite in the rocks studied:

Carbonate rocks:

Sharpness ratio of illite in the carbonate rocks has been calculated, Table 7.2 (page 358). The samples used in this study were collected from several localities in a way to represent the different grades of incipient metamorphism. The average of illite sharpness ratio in the samples studied at each locality was plotted on a map. The data have been approximately contoured and an isosharpness ratio map of the illite in the Carboniferous Limestone rocks of South Wales is produced, Fig. 7.12. Approximation in this map is due to the fact that the sampling points are controlled by the distribution of the outcrops.

It was found that sharpness ratio of illite in these rocks ranges from 1.1 to 5. From the illite isosharpness map it is concluded that sharpness ratios progressively increase, from the east to the west, i.e. with increase in the coal rank.

Terrigenous rocks:

(i) Millstone Grit:

The sharpness ratios of illite in the terrigenous rocks of the Millstone Grit range from 2.0 to 4.3. Most of the Millstone Grit samples are collected from the anthracite area, and they have sharpness ratios over 2.3.

(ii) Coal Measures rock:

Illite sharpness ratio has been calculated in samples representing the various lithotypes of the Coal Measures Series, and the various grades of incipient metamorphism as indicated by coal rank. Data are tabulated in

FIG.7.12 ILLITE ISOSHARPNESS RATIO MAP OF THE CARBONIFEROUS LIMESTONE.

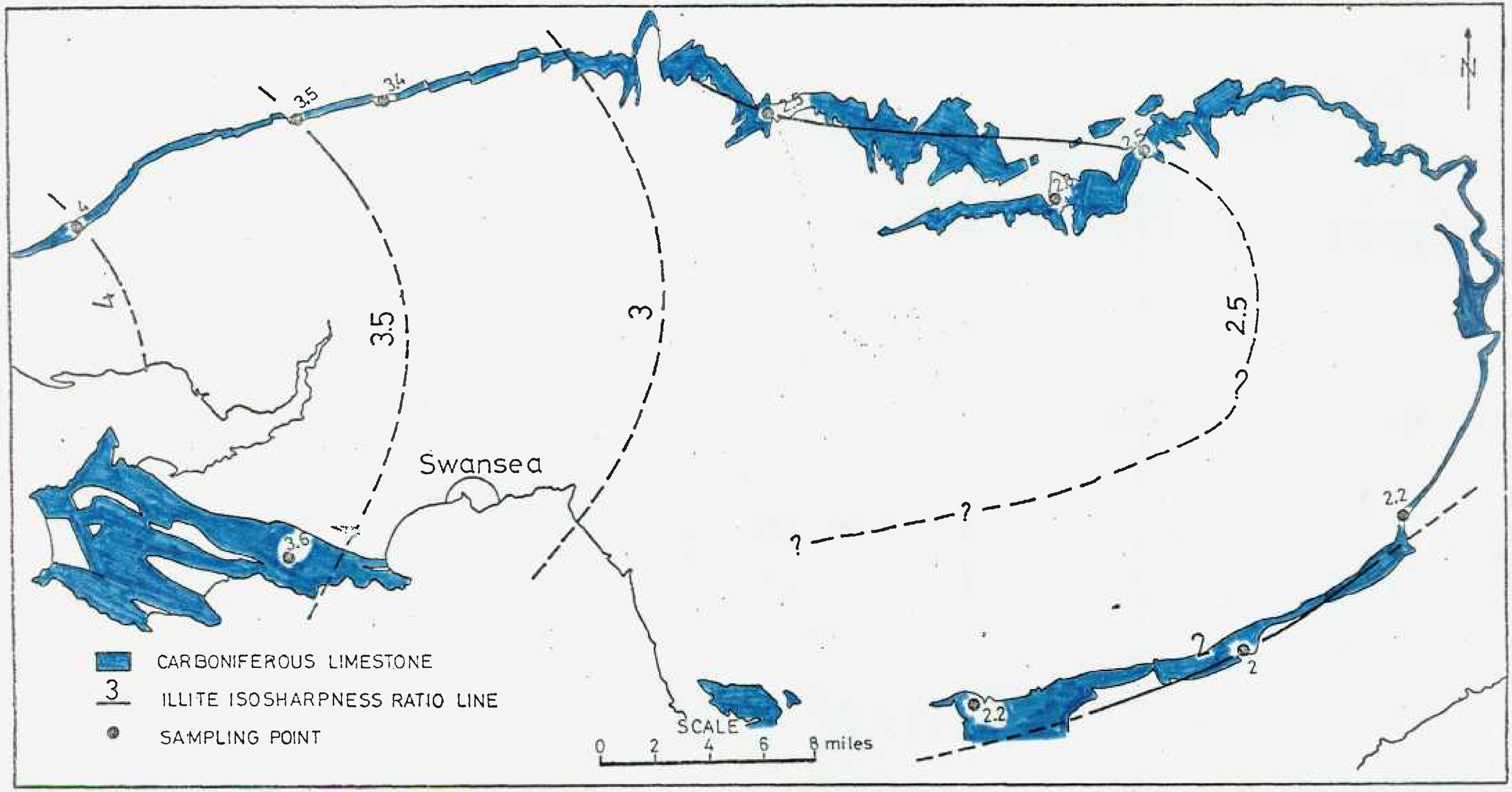


Table 7.5 (page). Fig. 7.13 is the isosharpness map of illite in the Coal Measures rocks. It shows a progressive increase of the illite sharpness ratio towards the west of the coalfield. It reaches up to 6 in the anthracite area. Fig. 7.14 shows the relationship between illite sharpness ratio and the fixed carbon %. From this diagram it can be concluded that sharpness ratio increases with the increase in the fixed carbon % of the associated coal seams. It was also found that sharpness ratio of illite in sandstones and siltstones is often higher than that in the associated argillaceous rocks.

Generally it was found that there is no evident relationship between the illite sharpness ratio and the depth of burial in the localities studied.

(3) Intensity Ratio (002)/(001) of Illite:

The relative intensity of (001) reflections is related to chemical composition of the octahedral layers in micas and chlorites. Nagelschmidt (1937) has shown that the 5 Å reflection was much stronger for dioctahedral micas of the muscovite type than for the trioctahedral micas of the biotite or phlogopite type. Klingebiel and Latauche (1962) have used the intensity ratio of (002)/(001) to differentiate between aluminous and ferromagnesian illites. Esquevin (1969)

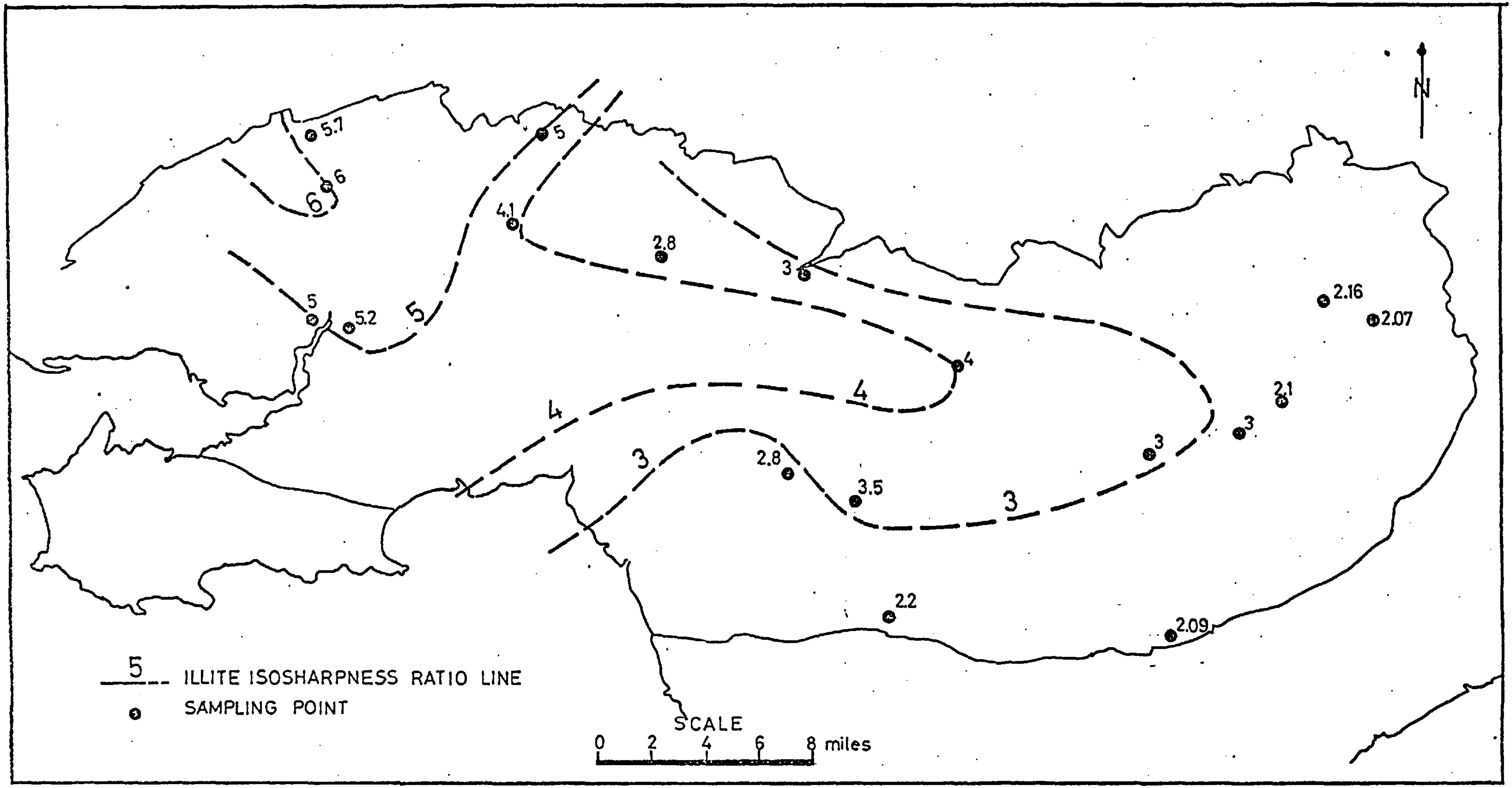


FIG.7.13 ILLITE ISOSHARPNESS RATIO MAP OF THE COAL MEASURES

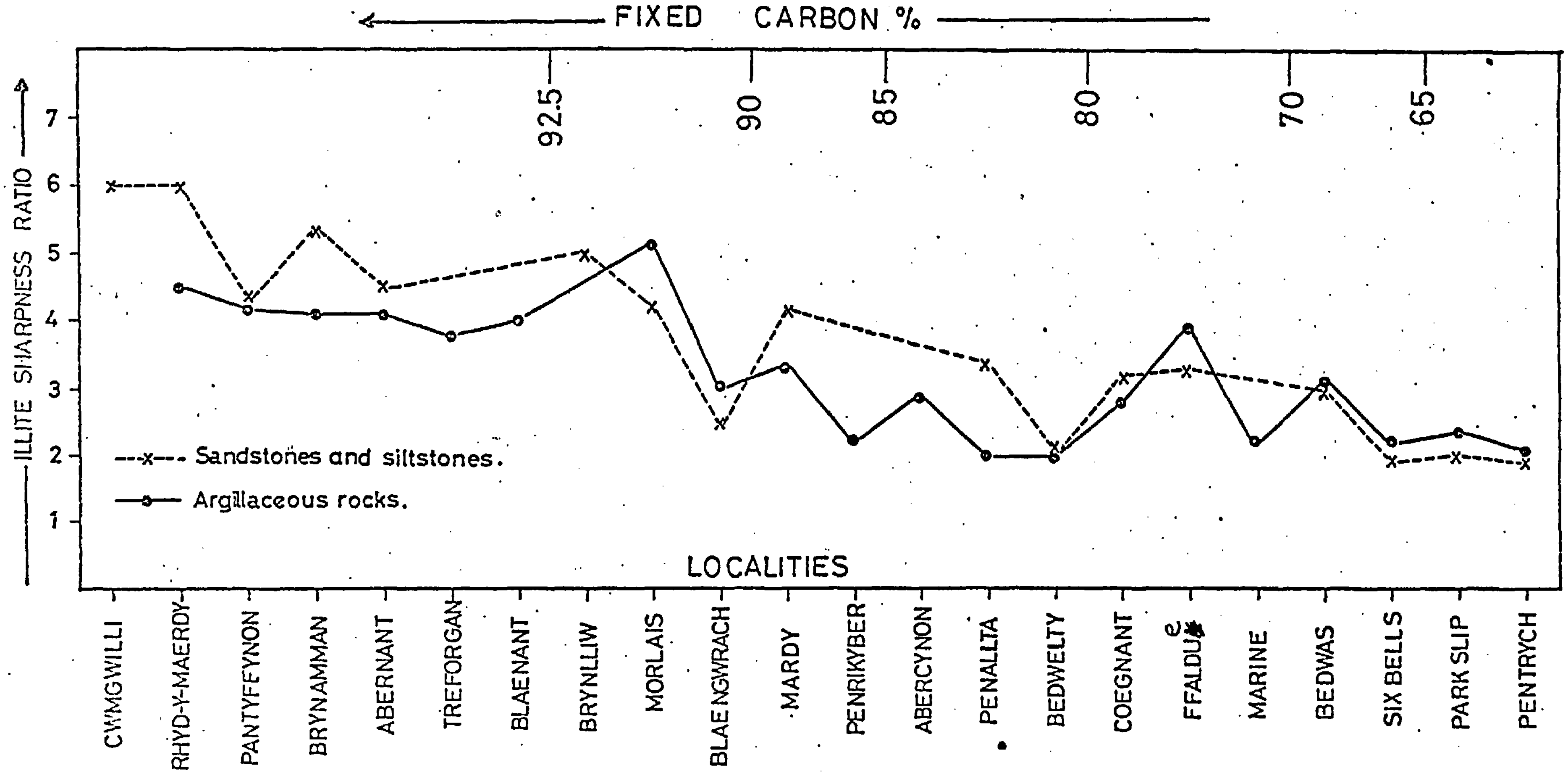


FIG.7.14 THE RELATIONSHIP BETWEEN ILLITE SHARPNESS RATIO IN THE COAL MEASURES ROCKS AND THE FIXED CARBON% IN THE ASSOCIATED COAL.

suspected that the crystallinity of illites depended not only on the temperature of recrystallization, but also on the chemical composition. He studied the relationship between the crystallinity of illite and the intensity ratio of the (002) and (001) peaks. The same relation has been studied by Desegonzac (1969) in many samples representing various grades of diagenesis and metamorphism. He concluded that increase in metamorphism was accompanied by increase in the I(002)/I(001), i.e. was accompanied by increasing in the Al/Mg+Fe. Fig. 7.15 summarises the relationship between crystallinity and I(002)/(001) of illites after Desegonzac (1969).

Illite crystallinity represented by the sharpness ratio has been plotted against the I(002)/I(001) in the Carboniferous Limestone and Coal Measures rocks (Figs. 7.16 and 7.17), respectively.

In the case of the Carboniferous Limestone rocks, it was found that I(002)/I(001) of illite ranges from about 0.4 to 0.95. No significant relation between the illite crystallinity and I(002)/I(001) of illite has been noticed.

In the illites of the Coal Measures rocks a relationship between crystallinity represented by the sharpness ratio, Al/Fe+Mg represented by I(002)/I(001) and the fixed carbon % of the associated coal seams has been recognized, Fig. 7.17.

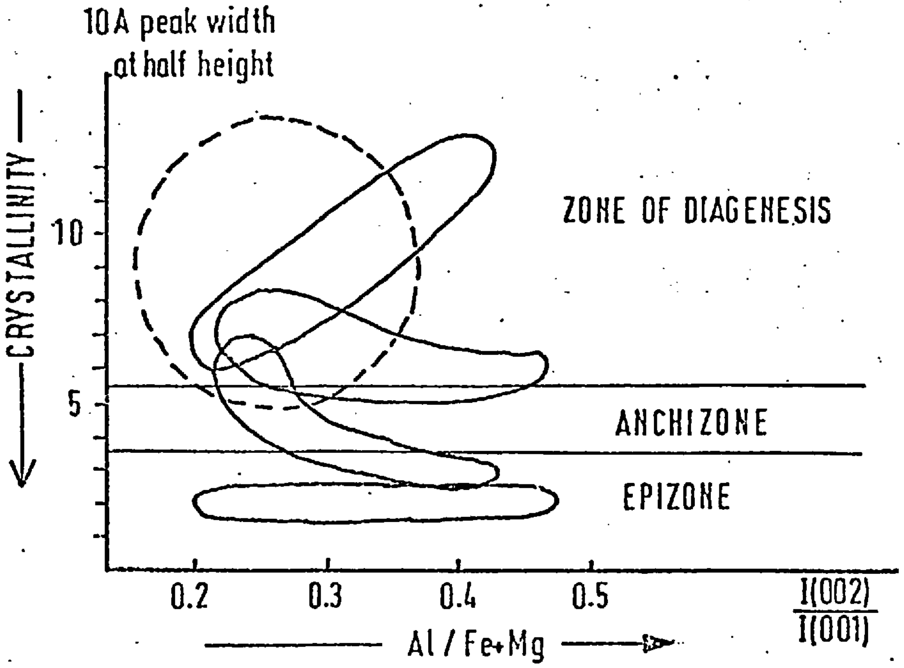


FIG.7.15 TYPES OF RELATION BETWEEN CRYSTALLINITY AND INTENSITY RATIO 002/001, IN AN ILLITE POPULATION,(after DESEGONZAC,1969).

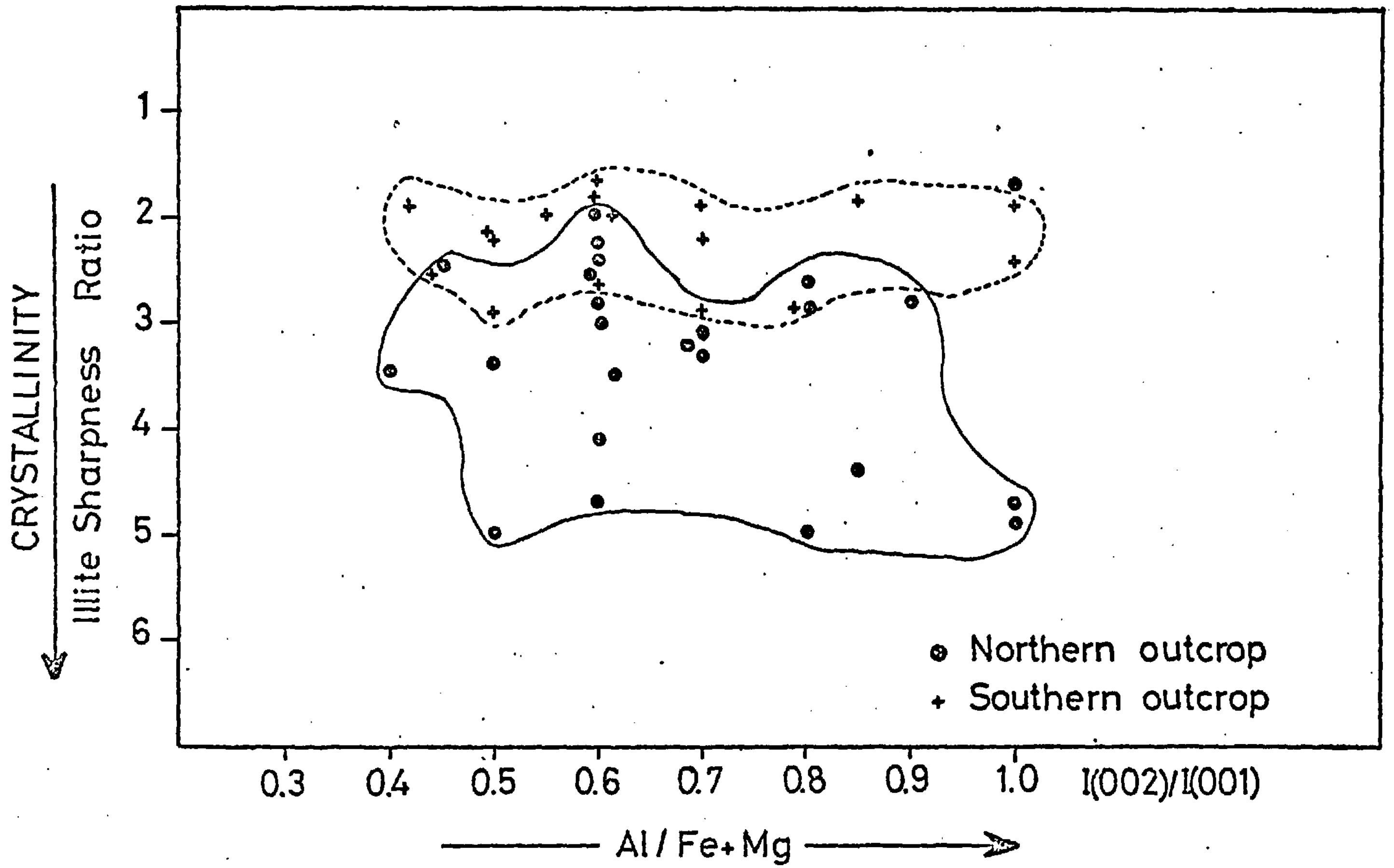


FIG. 7.16 RELATIONSHIP BETWEEN ILLITE CRYSTALLINITY AND I(002)/I(001) IN THE CARBONIFEROUS LIMESTONES

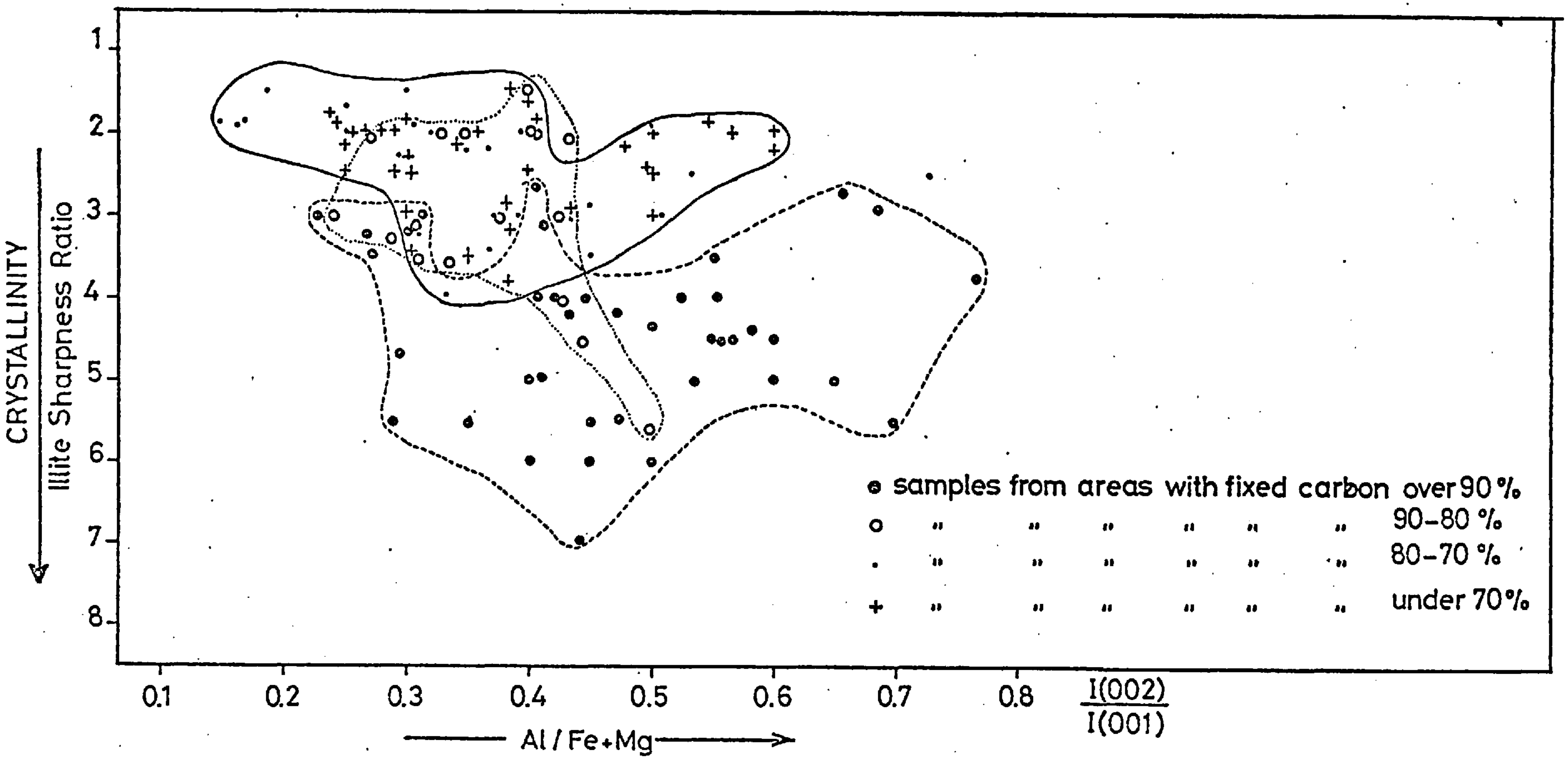


FIG. 7.17 RELATIONSHIP BETWEEN ILLITE CRYSTALLINITY AND I(002)/I(001) IN THE COAL MEASURES ROCKS.

It was found that increasing the fixed carbon % of the associated coal seams (i.e. increasing of the grade of metamorphism) is accompanied by increase in illite crystallinity and increase in the Al/Fe+Mg of the illites. It can be concluded that by increasing the grade of incipient metamorphism of the Carboniferous rocks of South Wales, illites become aluminous.

CHAPTER VIII

INCIPIENT METAMORPHISM OF THE ORGANIC MATTER

INCIPIENT METAMORPHISM OF THE ORGANIC MATTER

A. INTRODUCTION

The metamorphism of organic deposits and dispersed organic matter in sedimentary and meta-sedimentary rocks has attracted the attention of many geologists and geochemists for about a century. This is due to the fact that there is a clear relationship between degree of metamorphism of organic matter and occurrence of hydrocarbons on a regional scale. Study of incipient metamorphism of organic matter in sedimentary rocks is of twofold importance. The first is to study the evolution of the hydrocarbons and their generation from the original rock sources. The second is to study the different variations in the organic matter properties during progressive metamorphism and therefore their application as a palaeogeothermometer.

Metamorphism of the organic matter is a result of the effects of temperature, pressure and time. The main consequences of the metamorphism of the organic matter is the progressive increase in their fixed carbon content and decrease in their volatile contents. These chemical changes are usually accompanied by variations in the physical and optical properties of the organic matter. Among these properties are reflectivity, refractive index, bireflectance, colour and crystallinity.

Regarding the crystallinity, it was found that with metamorphism, progressive molecular reorganisation of the organic matter can be observed in the X-ray diffraction pattern (French, 1964; Kisch, 1966; Rutland, 1968; Griffin, 1967; and Landis , 1971). The samples of low rank have a very diffuse pattern with a very low wide peak poorly differentiated from the general background. As the rank increases, the pattern tends towards that of graphite. It was found also that with increase in temperature, the dimension of the organic crystallites (especially in the a-axis) increases (Blayden et al., 1944; and Goodarzi and Murchison, 1972).

In the present study, two techniques have been applied in studying the metamorphism of the organic matter. These are; the examination of the organic matter in reflected light and in transmitted light.

B. EXAMINATION OF THE ORGANIC MATTER IN REFLECTED LIGHT

GENERAL STATEMENT

This part deals with the petrography of the dispersed organic matter in the rocks studied and their reflectivity measurements. Reflectivity measurement is a well-established technique for the determination of coal rank and the thermal alteration of coal (Brown and Taylor, 1961; Francis, 1961; Taylor, 1961; Murchison, 1964; Chandra, 1965 McCartney and Ergun, 1967; and Bennet and Taylor, 1970). It is generally well known that the percentage of reflectivity of polished vitrinite surfaces increases with increasing the grade of metamorphism. Reflectivity can be applied to spores as well as vitrinite but less exactly (Alpern, 1967). The refractive index of the organic matter was found to be dependent on the grade of metamorphism, but it is not as widely applicable as reflectivity (Forsman, 1963).

METHODS OF STUDY

1. Samples:

For the purposes of the present study, 54 samples were chosen to represent the various rock types encountered in the studied basin, as well as the different grades of metamorphism. Table 8.1 shows the distribution of these samples.

TABLE 8.1 - Distribution of the samples examined in reflected light.

Series	Lithology	No. of Samples
Carboniferous Limestone	Biomicrite	8
	Biosparite	1
	Dolomicrite	2
	Shale	2
Millstone Grit	Sandstone	2
	Siltstone	1
	Shale	3
Coal Measures	Sandstone	17
	Siltstone	2
	Argillaceous Rocks	15

2. Polished section preparation:

Preparation of normal polished section has been discussed in detail by Ramdohr and Rehwald (in Freund, 1966). Since the purpose of these polished sections is the study of the organic matter in the rocks, so the notes published by Murchison and Boult (1961) on the polished surfaces of the coal materials were taken into consideration. The following is the procedure used in the preparation of the rock polished-surface in the present study.

a. Impregnation:

Most of the argillaceous rocks and some of the quartzwackestone samples have been impregnated. Araldite resins AY18 and hardener HZ18 (Ciba Co., Oxford and Cambridge), proportion 100:75 by weight, have been used for impregnation. Fresh, dry and clean rock specimens of one cubic inch in size were immersed in the prepared resin for 48 hours at 40°C under vacuum.

b. Cutting:

Rock specimens were cut at right angles to the bedding in the form of slices of about ¾" x ¾" and of about ¼" to ⅜" in thickness. One face of the rock slice was ground on a wet glass plate with 280 carborundum.

c. Mounting:

Material to be ground and polished on an automatic machine must be in a suitable form. This was achieved by mounting the specimen in artificial resin. The rock slices were placed into plastic moulds and cold setting artificial resin poured into them, (Griffin and Gregory embedding resin). The resin was left to cure for about 24 hours. The face of the mounted specimen was ground on a high speed rotating diamond wheel with water as a lubricant. The back side of the mounted specimens were also ground for levelling.

d. Grinding:

Specimens were first ground on an automatic labing machine using 800 Silicon Carbide suspended in water for about three hours. This produced an optically flat semi-polished surface. The specimens were then thoroughly cleaned to remove all of the abrasive traces.

e. Pre-polishing stage:

Grinding was followed by polishing on a rotating lab covered in linen using 850 aluminium oxide and water. The time for polishing ranges from 5 to 20 minutes.

f. Polishing:

The actual polishing of the specimens followed in four stages. In all of these stages pure mineral oil was used as a lubricant.

(i) 6-12μ diamond paste abrasive. The lab covered with hyperocel pellon - polishing time ranges from ½ to 2 hours.

(ii) 1-5μ diamond paste abrasive. The lab covered with hyperocel pellon - polishing time ranges from ½ to 2 hours.

(iii) 0-2μ diamond paste abrasive. The lab covered with wet cloth - polishing time ranges from ½ to 1 hour.

(iv) 0-½μ diamond paste abrasive. The lab covered with wet cloth - polishing time ranges from 10 to 45 minutes.

Remarks:

(i) The specimens have been thoroughly washed with hot water and detergent and then dried after finishing from every stage and before starting the next. In some cases ultrasonic vibrator was used to remove the abrasive particles from the fine pits and cracks.

(ii) The polishing time for each stage depends on the nature of the specimen, especially the lithology. Carbonate rocks were the easiest and quickest to polish, while quartzarenites took longer.

(iii) In some of the argillaceous rock specimens, especially underclays, large areas of the surface were blocking out very badly during the grinding and the pre-polishing stages. In this case the specimens have been impregnated again. Impregnation was carried out on the mounted specimen by covering its surface with araldite resin.

(iv) The rotating speed of the lab as well as the weight loading on the specimen were changeable according to the nature of the specimens.

3. Equipment and method employed for reflectivity measurements:

The equipment used for reflectivity measurements and its arrangement is shown in Fig. 8.1. Its main parts comprised Zetopan (Richert) incident light microscope with auxiliary magnification system; a photomultiplier tube with a spectral response extending from 200 to 800 nm, ; a DC amplifier; and a pen recorder with 10 mV full scale deflection. Measurements with this equipment can be made using polarising air and oil objectives on field areas as small as one micron diameter. Reflectivities were measured at 546 nm. in air and in oil (n oil = 1.515 at 24°C).

The procedures used are those described by Bayer (1967) for the A.S.T.M. Sub-Committee on Petrographic Analysis of Coal. Briefly, they are as follows:

i) The various components of the apparatus are carefully aligned, which is necessary because of the great sensitivity of the instruments employed. They are affected by variations in electrical current, deviations in the optical path, and instability due to changes in humidity.

ii) The specimen and the standard are placed on the revolving microscope stage. They must be optically levelled. Two standards have been used, namely, Zeiss

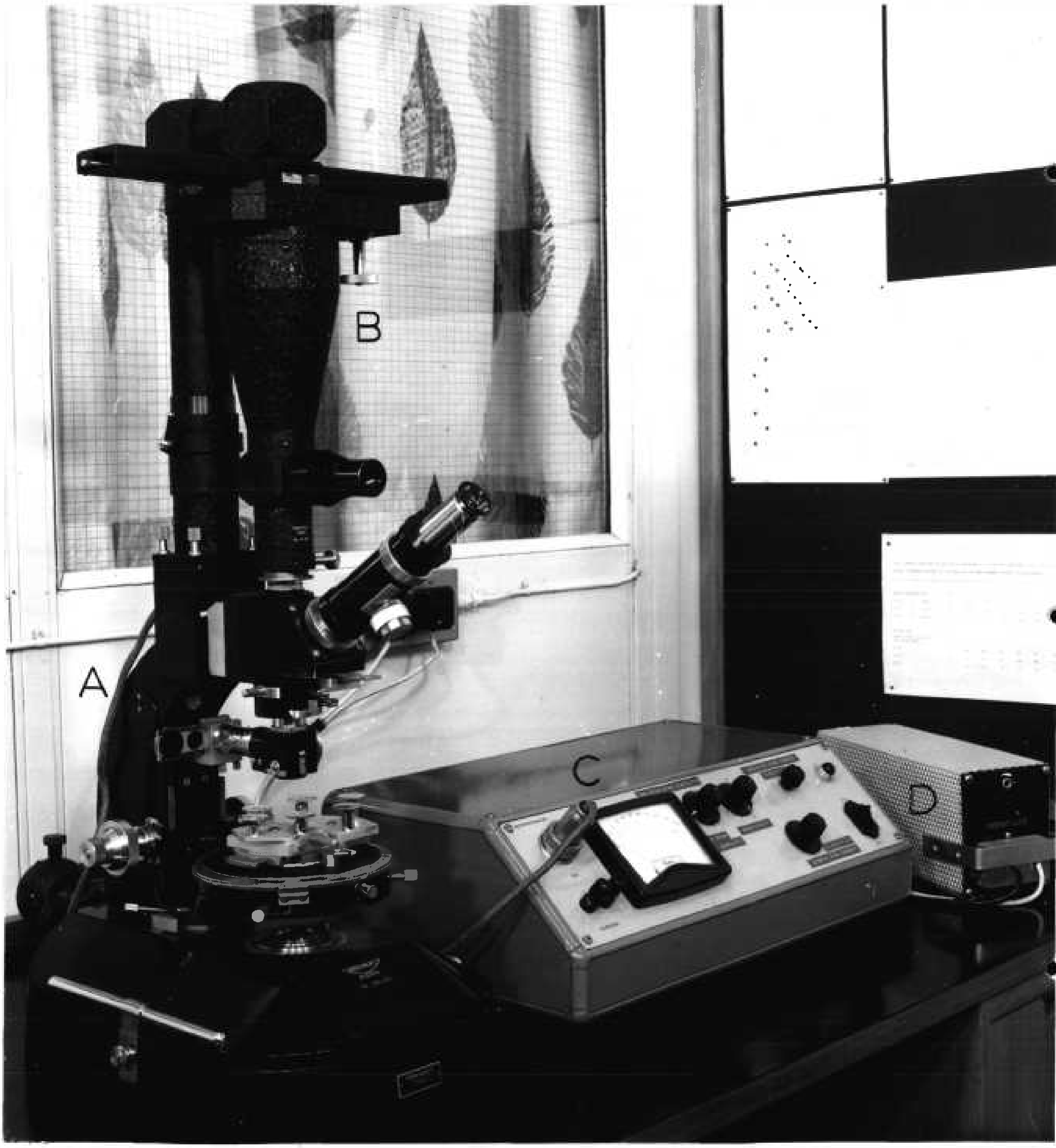


FIG. 8.1 Apparatus used in reflectance measurements.

A. Reflected light microscope.

B. Photomultiplier tube.

C. Recorder.

D. Constant current supply.

reflectance standard NGI (R air % 5.33 at 546 nm.) and Zeiss reflectance standard SiC (R air % 20.9 and R oil % 7.7 at 546 nm.).

iii) The microscopic stage is then slowly rotated through 360^{o} and the highest reading on the recorder is noted (A). The reading represents the maximum reflectance of the organic matter under observation. Then the specimen mount is moved and the standards are examined and the reading on the recorder is observed (B). The procedure has been repeated at least three times for each grain.

iv) For the calculation of the reflectivity % of the specimen, the following equation has been applied:

$$\frac{R}{R_1} = \frac{A}{B}$$

where, R = the reflectivity % of the specimen.

R_1 = the reflectivity % of the standard.

A = the recorder reading of the specimen.

B = the recorder reading of the standard.

The reflectivity percentage in air and in oil has been measured in the studied samples.

PETROGRAPHY OF THE DISPERSED ORGANIC MATTER

Because the reflectivity varies with the variation of the organic matter type, so it was necessary to identify the organic matter used for the measurements.

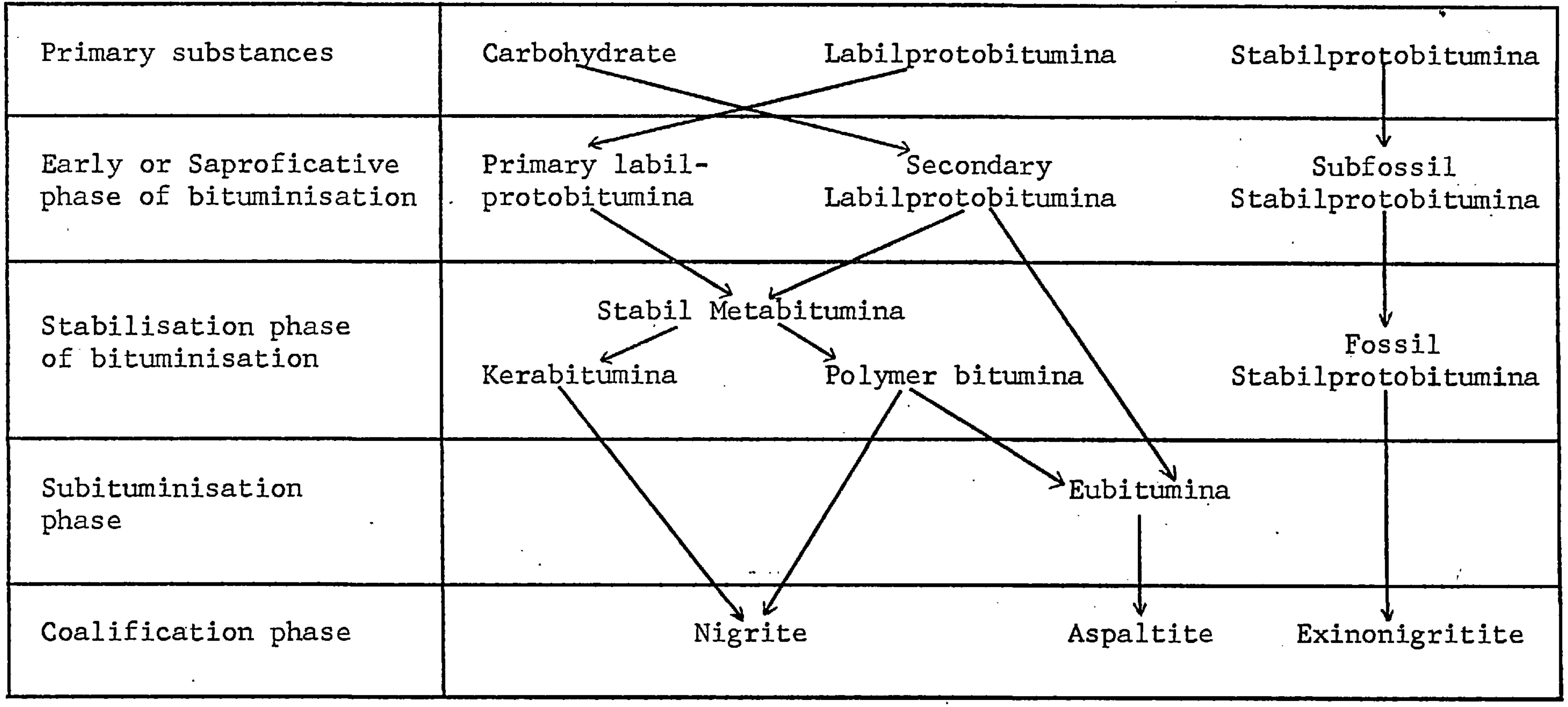
Classification of the organic matter:

Classification of the organic matter, both as dispersed grains in sedimentary rocks and as organic deposits, has been studied by many coal and oil geologists (Stopes, 1935; Potonie, 1951; Bitterli, 1963; Muratov, 1963; Spackman and Thompson, 1963; Alpern, 1970; and Correia, 1971).

Potonie (1951) classified bitumen petrographically into eleven types. Table 8.2 shows these types and their genetic classification. Bitterli (1963) has classified the deposited organic matter on the basis of its chemical properties into three groups, namely; (a) naptha bitumen (substantially soluble in carbon disulphide); (b) Kerabitumen (substantially insoluble); and (c) coal (substantially insoluble).

Muratov (1963) has designed a genetic classification of the organic matter. He considered the organic remains as minerals and used the term "organite" for the organic matter, and the term "anorganite" for the inorganic mineral constituents of the rock. His classification is based on the following

TABLE 8.2 POTONIE'S CLASSIFICATION OF BITUMEN



Primary substances	Carbohydrate Labilprotobitumina Stabilprotobitumina
Early or Saproficative phase of bituminisation	Primary labil-protobitumina Secondary Labilprotobitumina Subfossil Stabilprotobitumina
Stabilisation phase of bituminisation	Stabil Metabitumina Kerabitumina Polymer bitumina Fossil Stabilprotobitumina
Subituminisation phase	Eubitumina
Coalification phase	Nigrite Aspaltite Exinonigritite

basis; (a) character of the initial material; (b) the mechanism of its accumulation and fossilisation; (c) the nature of the subsequent alterations that led to the formation of the organite in question; and (d) the degree of manifestation of the main factors of the transformation. Muratov's classification is summarised in Table 8.3.

Correia (1971) in his study of the diagenesis of the organic matter in subsidiary rocks has classified the acid insoluble organic matter into three groups. These are:

(a) Insoluble organic matter with definite morphological shapes, which include fossil micro-organisms (such as spores and pollen grains, dinoflagellatesm acritarchs, chitinozoans, scolecodonts and microforaminifera), organic debris (such as cuticles, tracheids, sporangial debris and other plant and animal remains debris).

(b) Insoluble organic matter with poorly defined outlines including fine organic matter which may be colloidal.

(c) Soluble organic matter.

Alpern (1970) has recently suggested a classification for the organic matter in the sedimentary rocks based mainly on their petrographic characteristics in reflected light. He used the term "organolite" to define all types of the microscopic

TABLE 8.3 MURATOV'S CLASSIFICATION OF ORGANIC MATTER

TYPE	SUBTYPE	DIVISION OR GROUP	CLASS	SUBCLASS	ORDER	FAMILY
1. HUMO-ORGANITE	Humites		Cata humites			Brown Coal Hard Coal
			Meta humites			Anthracite Para anthracite Graphite
			Epi humites	True epihumites		Oxidised brown coal Secondary brown coal
				Oxy-humites		Oxalates Metallates Humates
	Liptites	Tar liptite Cuticular liptite Spore liptite Suberim liptite				
	Humo-liptites					
2. SAPRO-ORGANITE		Macro saprites (boghead)	Cata boghead			Ortho boghead Boghead hard coal
			Meta-boghead			Boghead anthracite Boghead para-anthracite Apo-boghead (graphite)
			Epi-boghead			Bogheadous secondary brown coal
			Proto-napthoids Lipto-napthoids Pyro-napthoids Tecto-napthoids			
				Epi-ntpthides	Products of subareal weathering	Kirs Humino kerites
					Products of subsurface weathering	Maltlas Asphalts
					Products of microbiological weathering	Algarites
				Cata-napthides		Gilstones Grahamites
				Meta-napthides		Shungites Graphite
				Schizo-napthides	Napth-filtrates Gas condensate Ozo Kerites Hatchellites	
3. SAPRO-HUMO-ORGANITES	Sapro-humites Sapro-liptites					
4. PYRO-ORGANITES						

organic matter dispersed or congregated in the sedimentary rocks. On the basis of the morphographic characteristics he distinguished seven groups of organolites. These are: (1) Textites (tissues); (2) Gelites (humic gels; (3) Detrites (microfragments and intercalated gels); (4) Bitumes (asphalts and bitumens); (5) Spherolites (excretions and inclusions); (6) Sporites (spores and pollen grains); (7) Organites (different organisms). On the basis of compaction, gelification or oxidation, homogeneity and reflectivity, each group has been further divided into subgroups and classes. Table 8.4 summarises Alpern's organolite classification.

Identification and nomenclature of the dispersed organic matter in the studied rocks is based mainly on Alpern's classification.

Identification of the organic matter in polished sections:

Five organolite groups have been identified in the polished sections of the rocks studied, namely, Textites, Gelites, Detrites, Bitumes and Sporites.

a. Textites:

Organic fragments belonging to this group are very frequent in most of the samples studied. Two subgroups have been identified, namely; xylotextite and fungitextite. Xylo-

TABLE 8.4 ALPERN'S CLASSIFICATION OF ORGANOLITES

TEXTITES	Xylotextite	Gelitextite	Xylinite Xylotelinite Telinite (Cryptotelinite)
		Fusi Textite	Biofusinite Pyrofusinite
	Exotextite	Suberitextite Cutitextite	
	Fungitextite		Geli Funginite Fusi Funginite
GELITES		Gelinite	Porigelinite Levigelinite
		Collinite	Meso Collinite Cata Collinite
		Oxygelite	Dessica Gelite Perioxy Gelite
DETRITES		Epi Detrite	Pori Detrite Densi Detrite
		Meso Detrite	Hetero Collinite Polymacerite
		Cata Detrite	Iso Detrite Aniso Detrite
BITUMITES		Norma Bitumite	Epi Bitumite Meso Bitumite Cata Bitumite
		Oxy Bitumite	
SPHEROLITES		Spherite Vacuolite	Resi Spherite Geli Spherite Fusi Spherite Oxy Spherite (Cito Spherite) (Resinite)
SPORITES			Epi Sporite Meso Sporite Cata Sporite
ORGANITES		Algae Chitinozoa Acritarche Cholecodont Membrano Detrite	

textites are those of the fossils of the lignocellulosic tissues while the fungitextites are the pseudo tissues of the fungi. The latter are usually characterised by walls of irregular thickness and irregular shapes and dimensions.

a.1 Xylotextites:

Two main types of xylotextite have been identified, namely, gelitextite and fusitextite.

a.1.1 Gelitextite:

These are plant tissues which have been decomposed chemically and biochemically through various complicated processes such as humification, gelification and calcification. The texture of these debris reflects the following phenomena: (i) expansion of the cell wall; (ii) obliteration of pores and cavities; (iii) increase in reflectance. As a whole, these phenomena provoke an increase in homogeneity, compaction and lustre. Two varieties of gelitextite have been identified, namely, xylotelinite and telinite.

a.1.1.1 Xylotelinite:

The xylotelinite fragments have a distinguished tissue structure with the cell wall slightly gelified. They are most common in sandstones and in some argillaceous rocks.

Usually they are of elongated shape and lie parallel to the bedding. They are generally compacted, so that the cell cavities tend to be elongated and lensoidal in shape with their long diameter parallel to the bedding (Plate 36, Figs. 1, 2, 3 and 4). Combined grains of zylotelinite and cryptotelinite are present. Gradational transformation from xylotelinite to cryptotelinite can be recognised in many fragments (Plate 36 , Figs. 3 and 4).

a.1.1.2 Telinite:

Telinite debris show an advanced state of gelification of the xylotelinite. Compaction of the tissue is almost total, but the structure can still be recognised (Plate 36 , Figs. 5 and 6).

a.1.2 Fusitextite:

Fusitextite grains are common in most of the sandstones and the argillaceous rocks. They are usually characterised by their high reflectivity which does not change much with coalification. Two varieties of fusitextite have been identified, namely, biofusinite and pyrofusinite.

a.1.2.1 Biofusinite:

It is characterised by collapsed cellular tissue. Biofusinite fragments are commonly present associated with the gelitextite debris. This type of fusitextite is commonly produced by the biochemical decomposition of the plant debris in partial aerobic environment.

a.1.2.2 Pyrofusinite:

It is characterised by well-preserved cellular tissue and a relatively high reflectivity. It is usually present as fine-grained debris and is less common than the biofusinite debris. Pyrofusinite is fossil charcoal (Plate 37 , Figs. 1, 2, 3, 4, 5 and 6).

a.2 Fungitextites:

This type is less common than the other organolite types. It is usually present in carbonate rocks. Fungitextites are present as globular bodies with irregular cavities inside. According to their reflectivity they mostly belong to the fusifunginite type (Plate 37 , Fig. 7).

b. Gelites:

They are present in almost all of the studied lithotypes. On the basis of reflectivity, gelites are represented only by the collinite type which is characterised by maximum homogeneity. Two types of collinite debris have been distinguished, namely, autochtonous collinite and allochtonous collinite. Autochtonous collinite debris is usually present as well preserved rootlets in the underclays, and as thin laminae parallel to the bedding (similar to algal mats) in the siltstones and mudstones (Plate 38 , Figs. 1 and 2).

c. <u>Detrites</u>:

Organolite fragments belonging to this group are characterised by an accumulation of microfragments with or without identified shape floating in a groundmass of humic gels. The identified detrites debris is mainly of the densidetrite type. It is commonly present in the coarse-grained sandstones and in some carbonate rocks. In the sandstones, densidetrite is present as coarse-grained fragments consisting generally of low reflectivity lensoidal debris floating in a relatively high reflectivity collinite groundmass (Plate 38 Fig. 3). In carbonate rocks, densidetrite is present as coal streaks trapped between the allochems in some biomicrites. These coal streaks consist of fine-grained elongated debris of collinite, fusinite and fusifungite floating in a groundmass of micrinite and gelinite (Plate 38 , Fig. 4). Densidetrite has also been recognised as stylolitic seams in some dolomicrites (Plate 39 , Fig. 6). So it is an autochtonous densidetrite formed by the concentration of the organolite debris along the stylolites.

PETROGRAPHY OF THE ORGANIC MATTER (in reflected light)

(Plates 36 - 39)

PLATE 36

Fig. 1 Xylotelinite, cell cavities are elongated and filled with resinite (?). Coal Measures Sandstone. Sample 170.

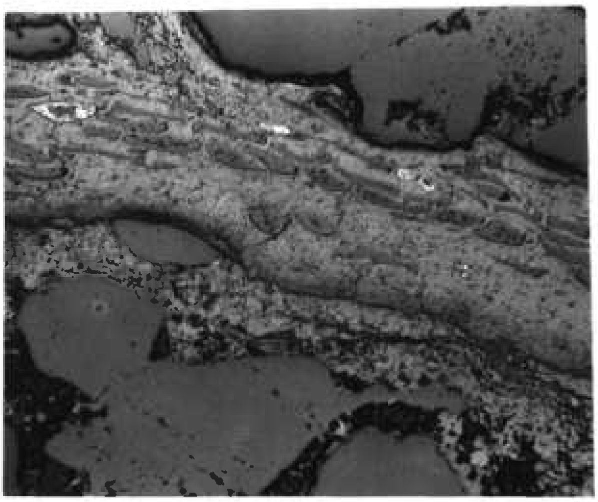
Fig. 2 Xylotelinite, cell cavities filled with siderite. Sideritized lithicarenite of the Upper Coal Measures. Sample 733.

Fig. 3 Xylotelinite (Xt) progressively altered to Cryptotelinite (Ct) and associated with fusinite (F). Coal Measures Sandstone. Sample 249.

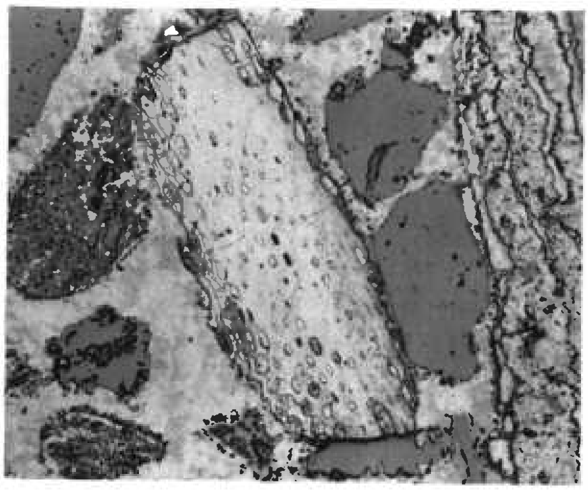
Fig. 4 Xylotelinite (Xt) altered to Cryptotelinite (Ct). Note cellular cavities partially filled with pyrite. Coal Measures Sandstone. Sample 170.

Fig. 5 Telinite (T). Coal Measures Sandstone. Sample 338-6.

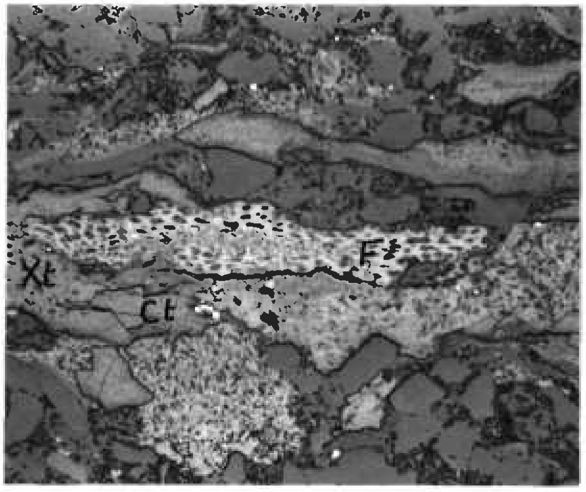
Fig. 6 Telinite (T). Note cellular cavities are compacted and transferred to discontinuous elliptical cavities. Lithicarenite of the Millstone Grit. Sample 90.



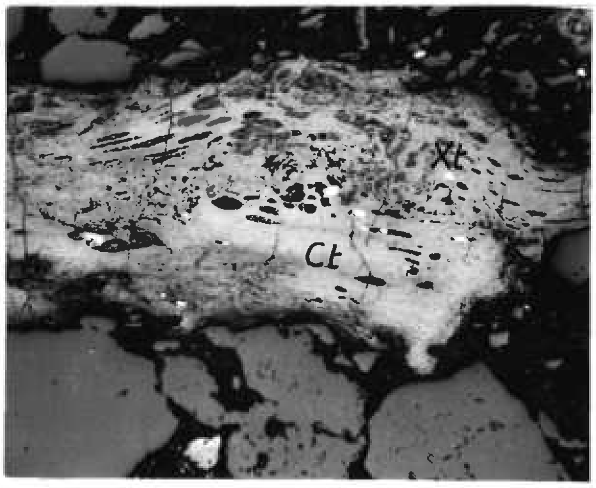
1 100μ



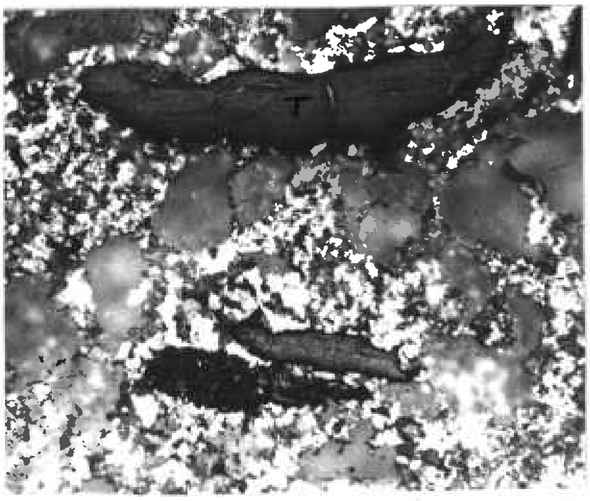
2 100μ



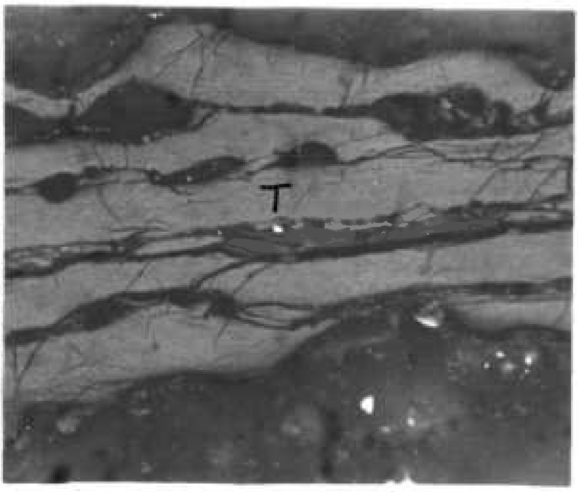
3 100μ



4 100μ



5 100μ



6 50μ

PLATE 37

Fig. 1 Pyrofusinite. Note the well preserved cellular tissue. Coal Measures Shale. Sample 131.

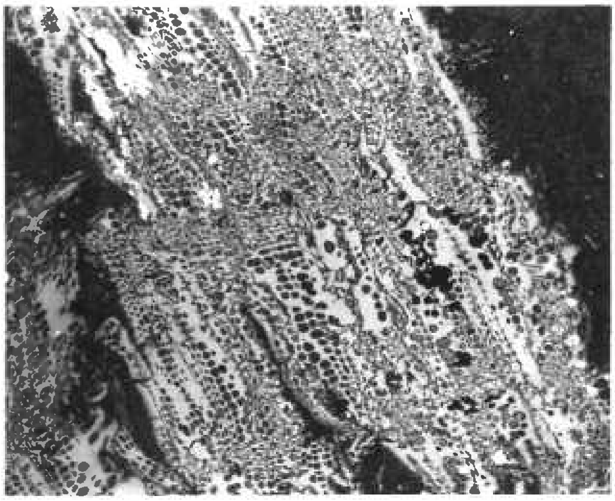
Fig. 2 Pyrofusinite (F) with a collapsed cellular tissue and telinite (T). Note the difference in reflectivity. Coal Measures Sandstone. Sample 170.

Figs. 3 & 4 Pyrofusinite (F) and telinite (T) in Coal Measures Sandstone. Sample 719.

Fig. 5 Pyrofusinite. Coal Measures Sandstone. Sample 338-6.

Fig. 6 Slightly deformed pyrofusinite fragment. Coal Measures Sandstone. Sample 249.

Fig. 7 Fusifunginite (arrow). The funginite tissue has been distructed by the force of crystallization of the siderite infilling (S). Note the presence of framboidal pyrite. Millstone Grit Shales. Sample 88.



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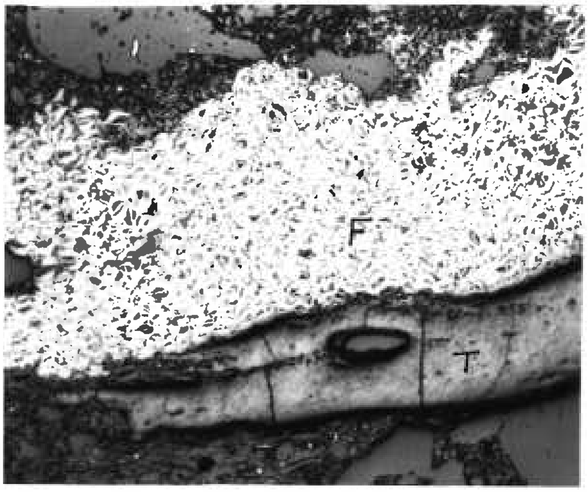

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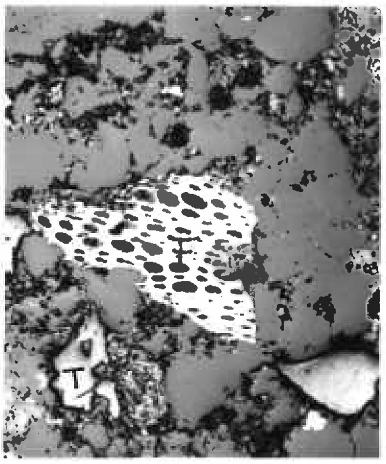

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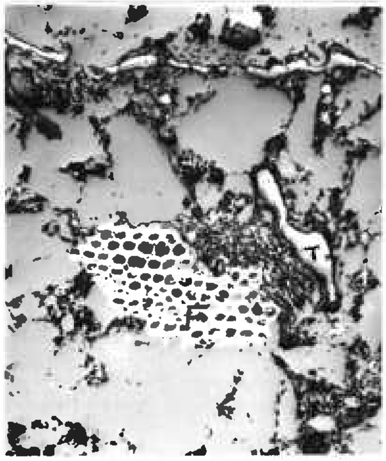

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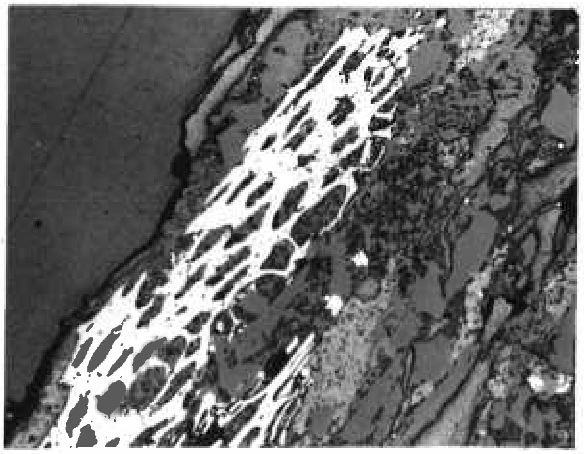

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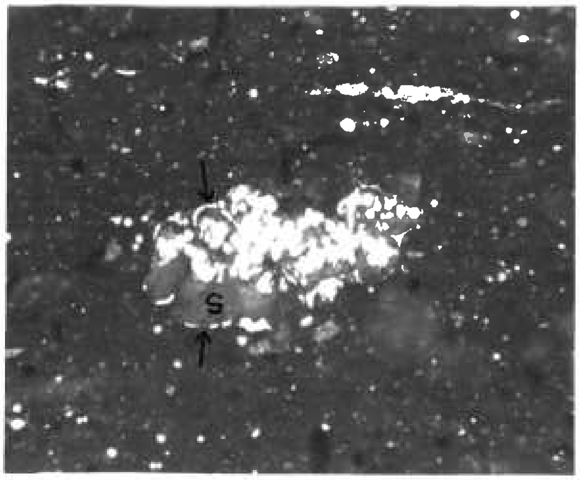

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PLATE 38

Fig. 1 Cata collinite fragment. Coal Measures Sandstone. Sample 170.

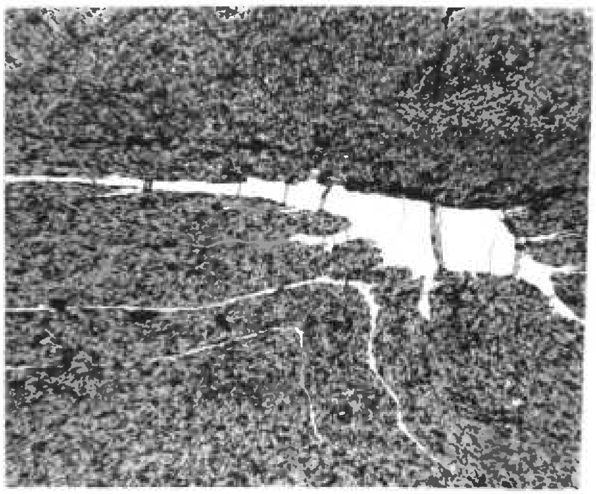
Fig. 2 A plant rootlet preserved as Cata collinite in a Coal Measures Underclay. Sample 907.

Fig. 3 Detrite (D) and Collinite (C). Note displacement of a fractured collinite fragment due to the force of crystallization of pyrite (arrow). Coal Measures Sandstone. Sample 388-6.

Fig. 4 Detrite streaks floating in fibrous calcite. Note detrite consists of collinite fragments embedded in a micrinite groundmass. Biomicrite. Sample 615.

1

100 µ



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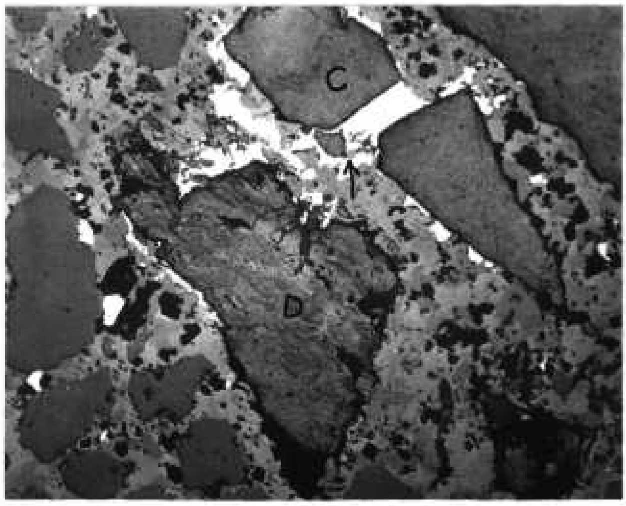
150 µ



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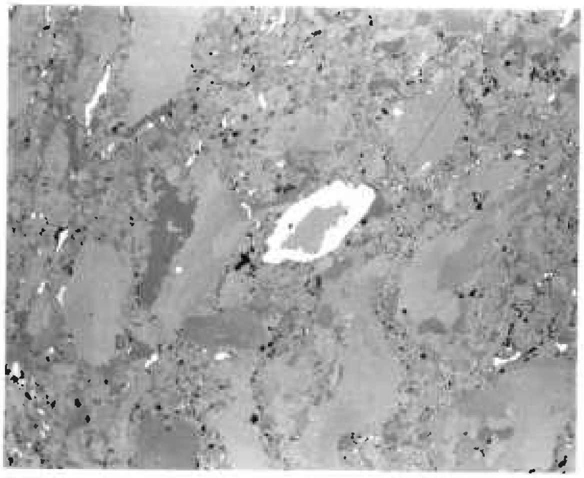
100 µ

4

120 µ

PLATE 39

Fig. 1	Sporite floating in the matrix of a biomicrite. Sample 622.
Fig. 2	Sporite floating in a detrite fragment. Biomicrite. Sample 615.
Fig. 3	Sporite. Millstone Grit Shale. Sample 661.
Fig. 4	Sporite. Biomicrite. Sample 614.
Fig. 5	Sporite. Note the destruction of the original spore by the force of crystallization of the siderite infilling (S). Millstone Grit Shale. Sample 88.
Fig. 6	Sporite floating in a lump of organic matter which has been concentrated in a stylolite seam. Dolomicrite. Sample BY65.
Fig. 7	Bitumite lump. Note the concentric fractures. Coal Measures Sandstone. Sample 388-6.



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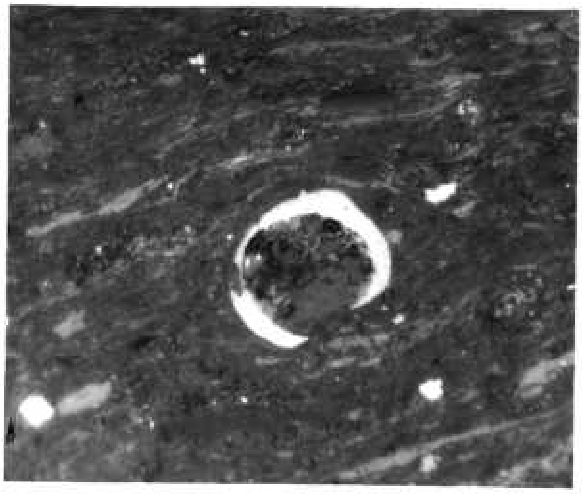

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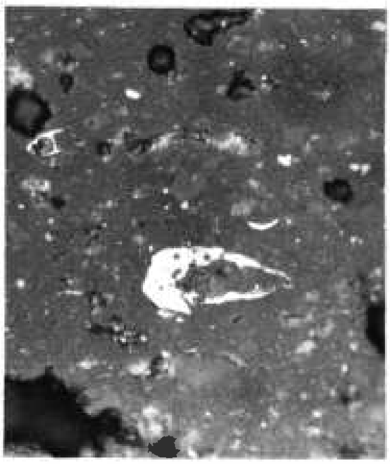

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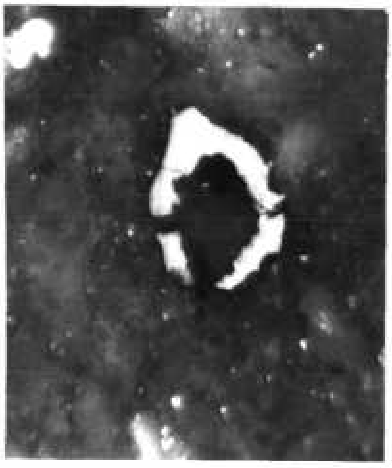

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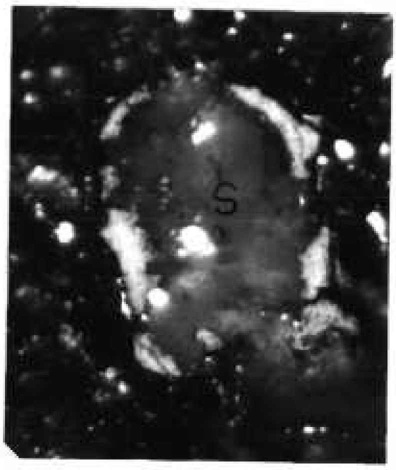

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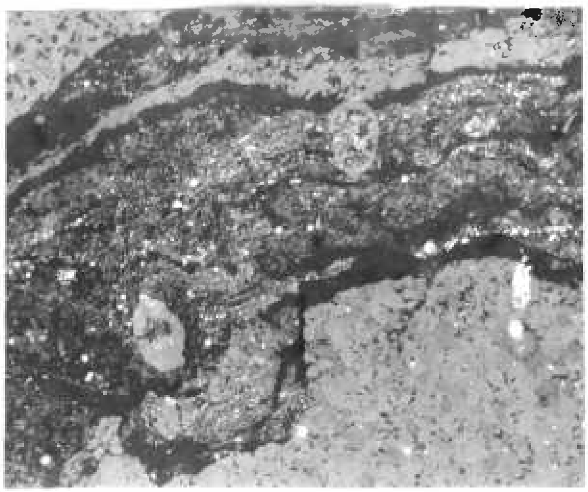

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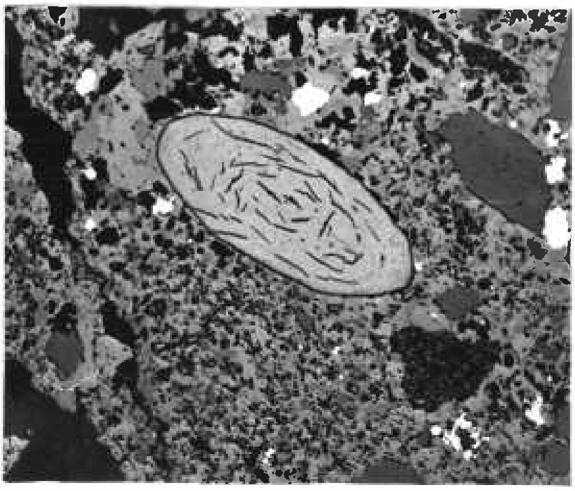

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d. Bitumens:

This is the least common type among the other organolites in the rocks studied. It is represented mainly by catabitumite debris which is present mainly in the carbonate rocks. It is present as shapeless, very fine-grained bodies sporadically distributed in the rock. In some rocks, catabitumites were found to be concentrated on stylolites. In some sandstones it is present as ovoidal-shaped bodies of high reflectivity with dessiccation cracks (Plate 39, Fig. 7).

e. Sporites:

They are present as spherical bodies with thin, regular walls. According to their reflectivity, Mesosporites and Catasporites have been identified (Plate 39, Figs. 1, 2, 3, 4, 5 and 6).

Mode of occurrence of the organic matter:

a. In sandstones and siltstones:

In the massive sandstones and siltstones, organic matter is present usually as detrital grains in the size range of the framework mineral grains (Plate 40 , Fig. 1). In the thinnly-bedded sandstones and siltstones, organic matter is commonly present as thin collinite laminae lying on the bedding.

b. In argillaceous rocks:

In shales, organic matter is usually present as fine silt-size grains sporadically distributed in the rock, sometimes as elongated debris lying parallel to the bedding. In the mudstones, organic matter is generally present as an algal-mat-like laminae of collinite (Plate 40 , Fig. 3). In the case of underclays, they are present in two forms, these are; (1) well preserved rootlets consisting of collinite (Plate 38 , Fig. 2) ; and (2) as extremely fine-grained debris and very thin fine-grained shreds.

c. In carbonate rocks:

In biomicrites and biosparites, organolites are present mainly as very fine-grained collinite and Catabitumite debris dispersed in the micrite matrix and between the calcite crystals forming the cement (Plate 41 , Figure 2). Occasionally, in some biomicrites thin coal streaks are present trapped between the allochems. In the dolomicrites bitumite is present as intergranular pore-filling. Concentrations of organic matter along the stylolites are a common phenomenon in the carbonate rocks (Plate 41 , Fig. 1).

Textures of the organic matter:

For descriptive purposes textures of the organic matter are grouped as; (a) deformational textures; and (b) textures showing the relationships between the organolite debris and the mineral matter.

a. Deformational textures:

a.1 Grain fracturing:

This texture is most evident in sandstones and carbonate rocks where collinite debris has been intensively fractured. Fracturing is commonly accompanied by spalled-off slivers of collinite, some of which may display rotation (Plate 42 , Figs. 5 & 6).On the basis of geometry, two types of fractures have been identified, namely; irregular fractures and concentric fractures. The former type is the most common and is usually displayed in collinite debris. Concentric fractures have been recognised only in Catabitumite debris and could be dessiccation fractures.

a.2 Grain crushing and collapsing of the cellular tissues:

This texture is commonly displayed in pyrofusinite debris. Where compaction has been intense, pyrofusinite grains were crushed and the fragments rotated (Plate 42 , Fig. 1). Compaction of the pyrofusinite grains led to the collapse of the cellular structure resulting in the reduction of the grain size (Plate 42 , Figs. 2 and 3).

b. Textures showing the relationships between the organolite debris and the mineral matter:

b.1 The grain boundaries of the organolites:

It was found that the shape of the organolite debris boundaries is dependent on the type of organolite, lithology and the diagenesis of the rock. In carbonate and argillaceous rocks the organolite grain boundaries are more or less regular. In the sandstones the boundaries are generally irregular and follow the grain boundaries of the surrounding framework grains (Plate 42 , Fig. 4). Gelite and gelitextite organolites, such as collinite, xylotelinite and telinite are found to be the least capable of preservation of their original grain boundaries. Fusinite debris usually preserves its original grain boundaries.

b.2 Replacement of the organolite by mineral matter:

Partial replacement of the organolite debris by authigenic minerals has been recognised in many of the rocks studied. Pyrite and siderite are the main minerals seen to replace the organolites.

b.2.1 Replacement of the organolites by siderite:

Replacement of organolite by siderite has been recognised in the sandstone where severe sideritisation has taken place. Plate 43 , Figs. 1-3 show the various stages of replacement of collinite debris by authigenic siderite. Siderite usually starts to replace the organolite along grain boundaries, fractures and cavities. Corrosion texture has been displayed in collinite fragments in the early stages of the siderite replacement. Severe sideritisation of collinite debris has resulted in the formation of siderite pseudomorphs after collinite with relict patches of collinite.

b.2.2 Replacement of organolite by pyrite:

Authigenic pyrite is frequently present in most of the biomicrites and the argillaceous rocks and in some sandstones. It is present in three forms, namely, framboidal, euhedral and subhedral crystalline aggregates; and in massive habit. Replacement of organolites by pyrite is present as fine-grained framboids dispersed in the collinite fragments (Plate 43 , Figure 5) . Pyrite framboids surrounding and replacing collinite of fossilised rootlets in underclays have been observed (Plate 43 , Fig. 4).

b.3 Mineral matter as infillings in the organolites

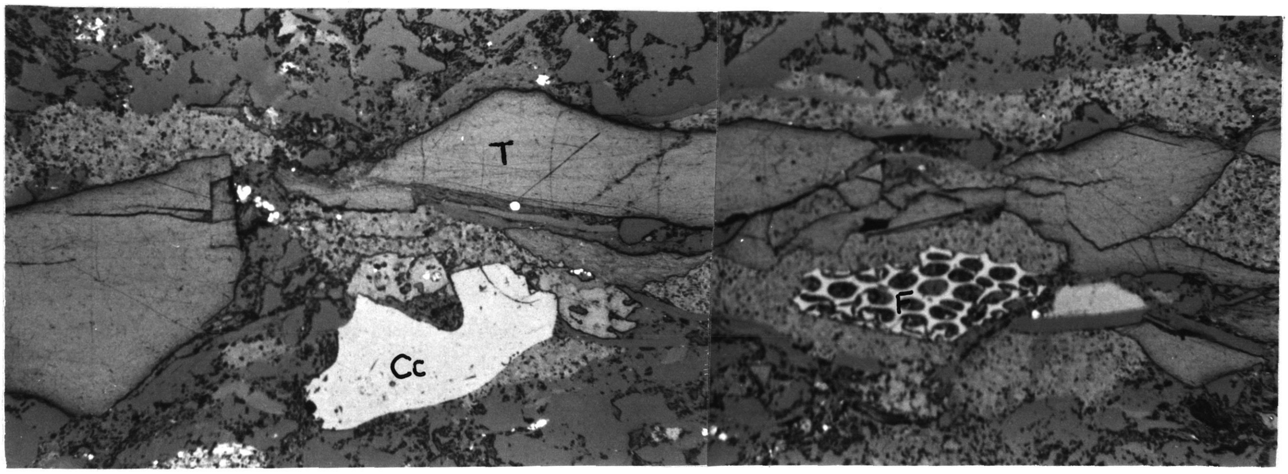
Authigenic as well as detrital mineral matter is commonly present as infillings of cavities and fractures of some organolite fragments. Among the authigenic minerals are

MODES OF OCCURRENCE OF THE ORGANOLITES

(Plates 40 - 41)

PLATE 40

Fig. 1	Organolite fragments concentrated in bands in a Coal Measures Sandstone. Cata Collinite (Cc), telinite (T), fusinite (F). Sample 249.
Fig. 2	Organolite debris scattered in a siltstone. Coal Measures Siltstone. Sample 356.
Fig. 3	Algal mats (?) preserved as Collinite (arrow) in a Coal Measures Siltstone. Sample 247.
Fig. 4	Organolite debris scattered in biomicrite. Sample 655.
Fig. 5	Organolite fragments in pyrite-rich shale. Pyrite (P), organolite (O). Sample 185.



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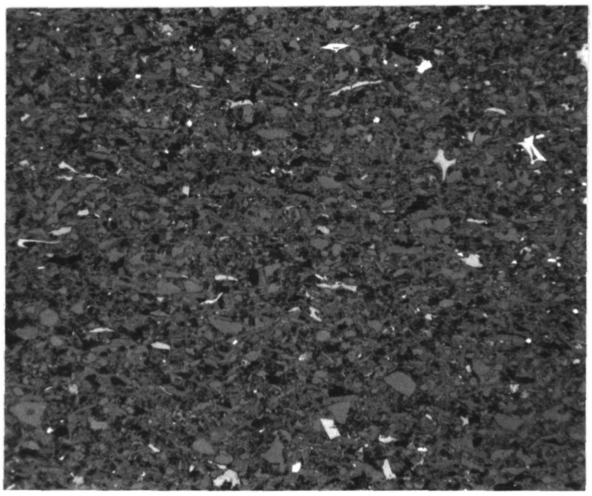

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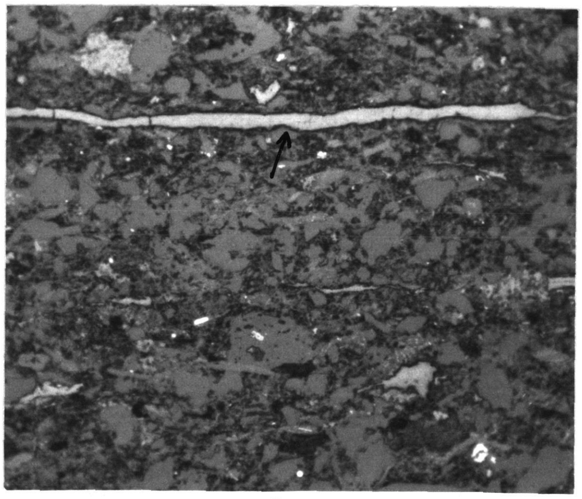

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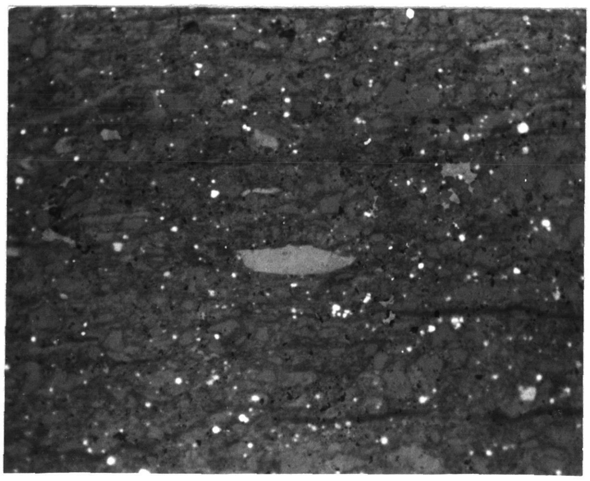

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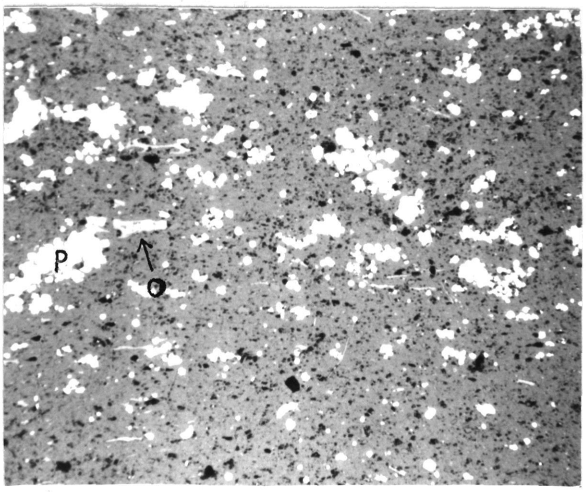

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PLATE 41

Fig. 1 Concentration of the organolite fragment on a stylolite seam (arrow). Biomicrite. Sample 633.

Fig. 2 Organolite fragments floating in the matrix of a biomicrite. Sample 659.

Fig. 3 Organolite debris in Coal Measures Sandstone. Sample 1002.

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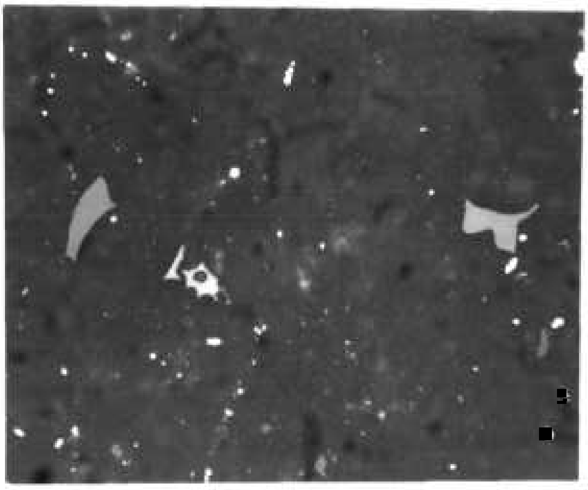

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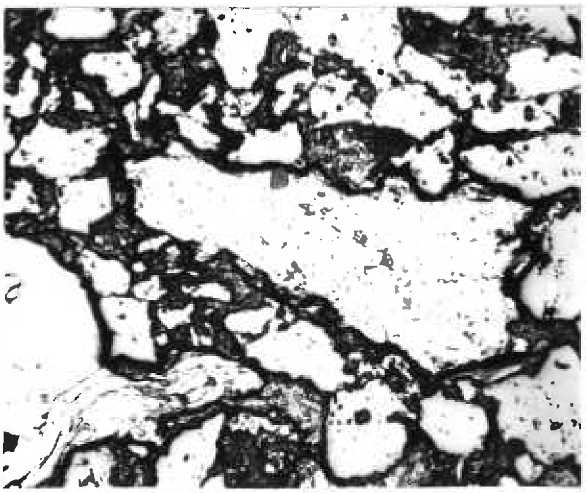

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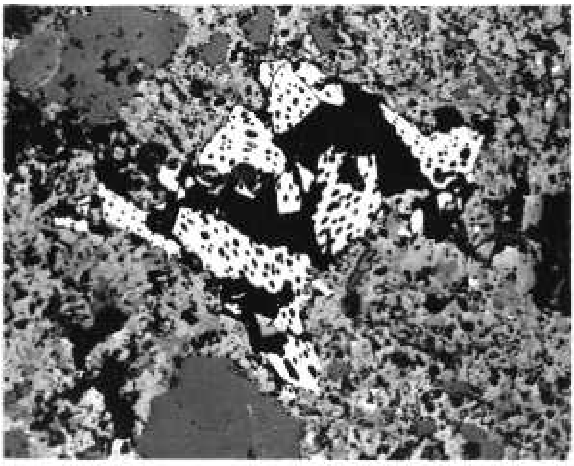
FABRICS OF THE ORGANOLITES

(Plates 42 - 43)

PLATE 42

Fig. 1	Crushed pyrofusinite fragment in sandstone. Coal Measures Sandstone. Sample 388-6.
Fig. 2	Collapsed pyrofusinite tissue. Note the pyrofusinite fragment squeezed between the quartz grains. Coal Measures Sandstone. Sample 719.
Fig. 3	Partially collapsed pyrofusinite tissue. Coal Measures Shale. Sample 131.
Fig. 4	Irregular grain boundaries of collinite debris in sandstone. Coal Measures Sandstone. Sample 719.
Fig. 5	Fractured collinite fragment. Note spalled-off slivers of collinite, and pyrite framboids (P) in the collinite. Biomicrite. Sample 633.
Fig. 6	Fractured collinite fragment. Note fractures filled with an authigenic mineral. Coal Measures Sandstone. Sample 760.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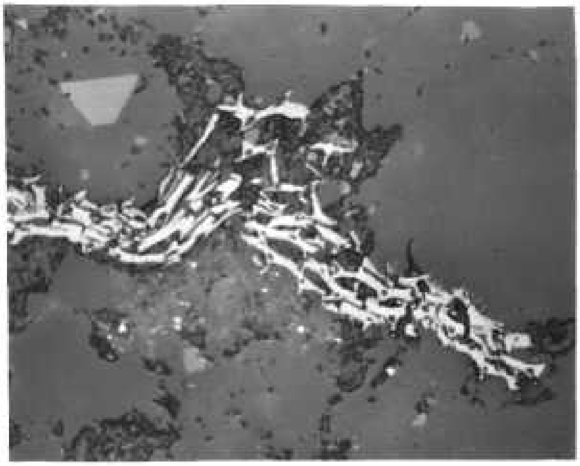
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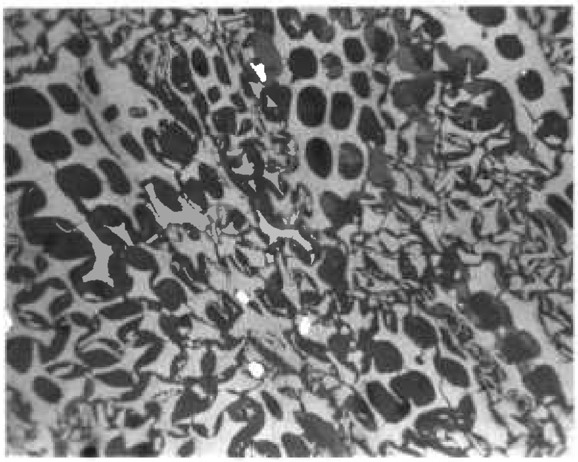
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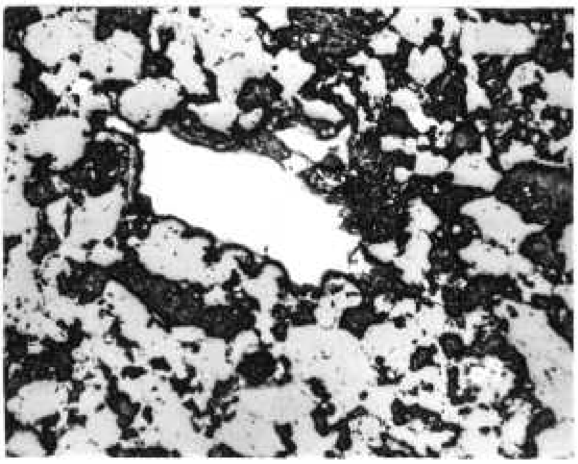
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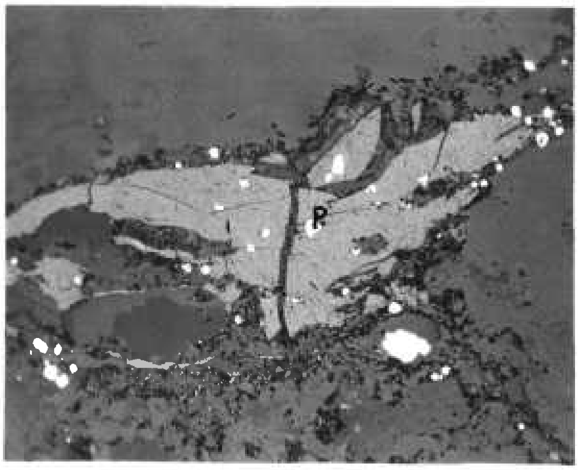

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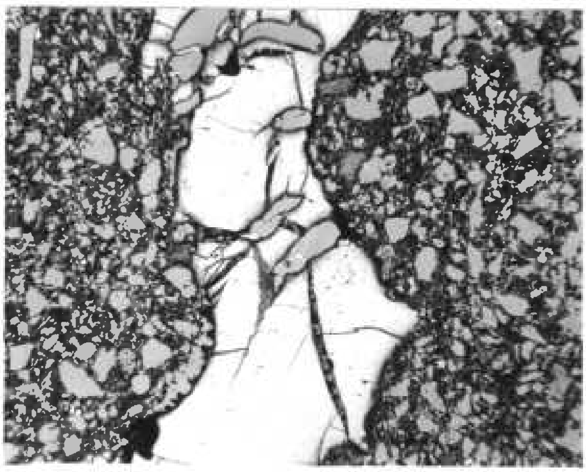
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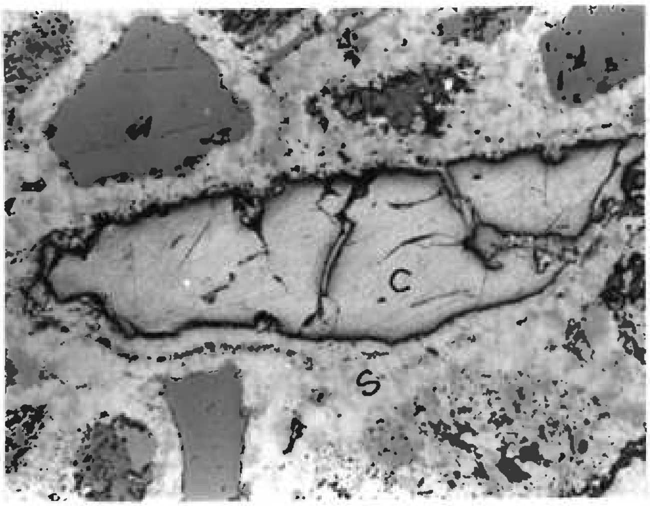
PLATE 43

Figs. 1, 2 & 3 Slow progressive replacement of a collinite (C) fragment by authigenic siderite (S). Coal Measures sideritized sandstone. Sample 733.

Fig. 4 Framboidal pyrite (P) surrounded and replacing a collinitized rootlet. Coal Measures Underclay. Sample 907.

Fig. 5 Pyrite partially replacing a collinite fragment. Biomicrite. Sample 633.



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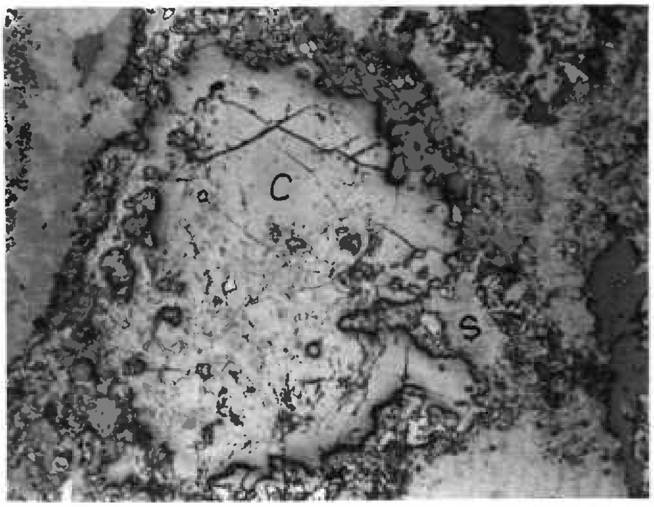
100μ



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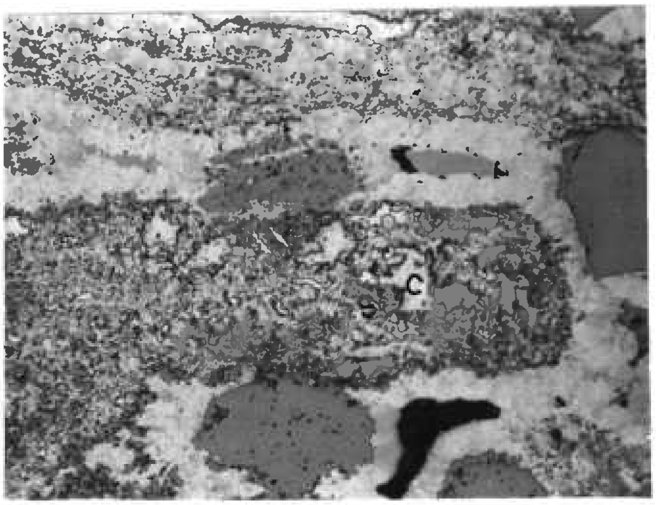

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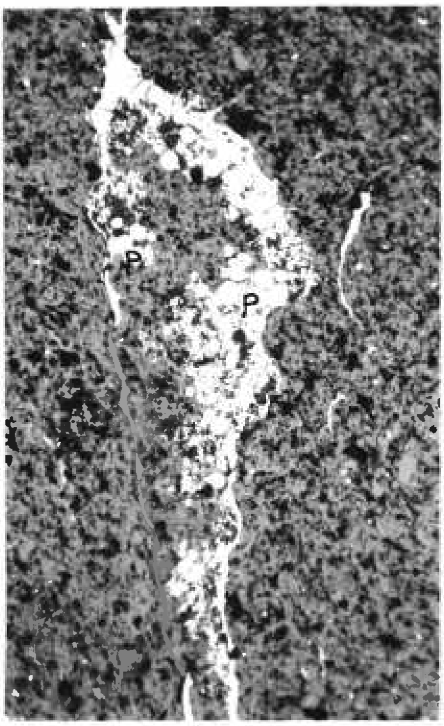

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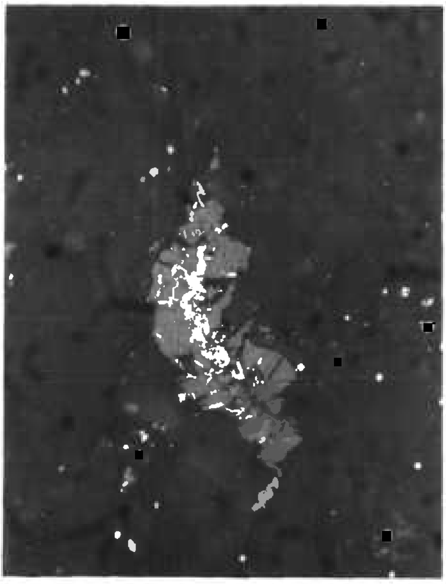

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100μ

pyrite, siderite, quartz and calcite. Massive pyrite is commonly present as infilling of cellular cavities of textite fragments (Plate 36 , Fig. 4), and some fractures in collinite debris. In carbonate rocks, calcite is the main infilling mineral of cavities and fractures of the organolite fragments. Pores and fractures in the organolite fragments occur in sandstones and in some argillaceous rocks, are commonly filled with siderite. In some cases the force of crystallisation of the infilling minerals results in the destruction of the original fabric of the organolite fragments (Plate 39 , Fig. 5).

REFLECTIVITY MEASUREMENTS

General Statement

Reflectivity is the primary quantitative parameter employed by coal petrologists to measure change of properties and to assess rank in the different coal constituents or macerals. Recently, reflectivity measurements of the organic matter in the sedimentary rocks have been applied by the oil geologists to determine the grades of incipient metamorphism, and therefore the occurrence of oil and gas. In the present study, reflectivity technique has been applied to distinguish the grades of incipient metamorphism in the carboniferous rocks of the South Wales Coalfield.

Effects of temperature and pressure on the reflectivity of organic matter:

Experimental work done by many coal petrologists has shown that the reflectivity of the coaly matter, especially that of vitrinite, increases with increase in the temperature of carbonization (Chandra, 1965; Ghosh, 1968 ' Marshall and Murchison, 1971; and Goodarzi and Murchison, 1972). Fig. 8.2 shows the relationship between the temperature of carbonization and the reflectivity of vitrinite.

Shapiro and Gray (1966) have examined the reflectance and other properties of a set of coal samples from Antarctica which were taken from strata approximately 1000 ft. thick underlying a thick diabase sill. The maximum oil reflectance falls from the highest value of 11.5% 52 ft. below the sill, to 2.8%, 1167 ft. below the sill. Chatterjee, Chandra and Ghosh (1964) concluded that the reflectance of the Poniati Seam, intruded by a meta-peridotite dyke, indicated a temperature of 600°C at the dyke-coal contact. This conclusion is based on comparison of the reflectance pattern from the intruded coals with that of laboratory carbonised coal.

Temperatures as high as 900°C close to the coal-dyke contact were estimated by Kisch (1966) from reflectance determinations on thermally metamorphosed semi-anthracitic vitrinite.

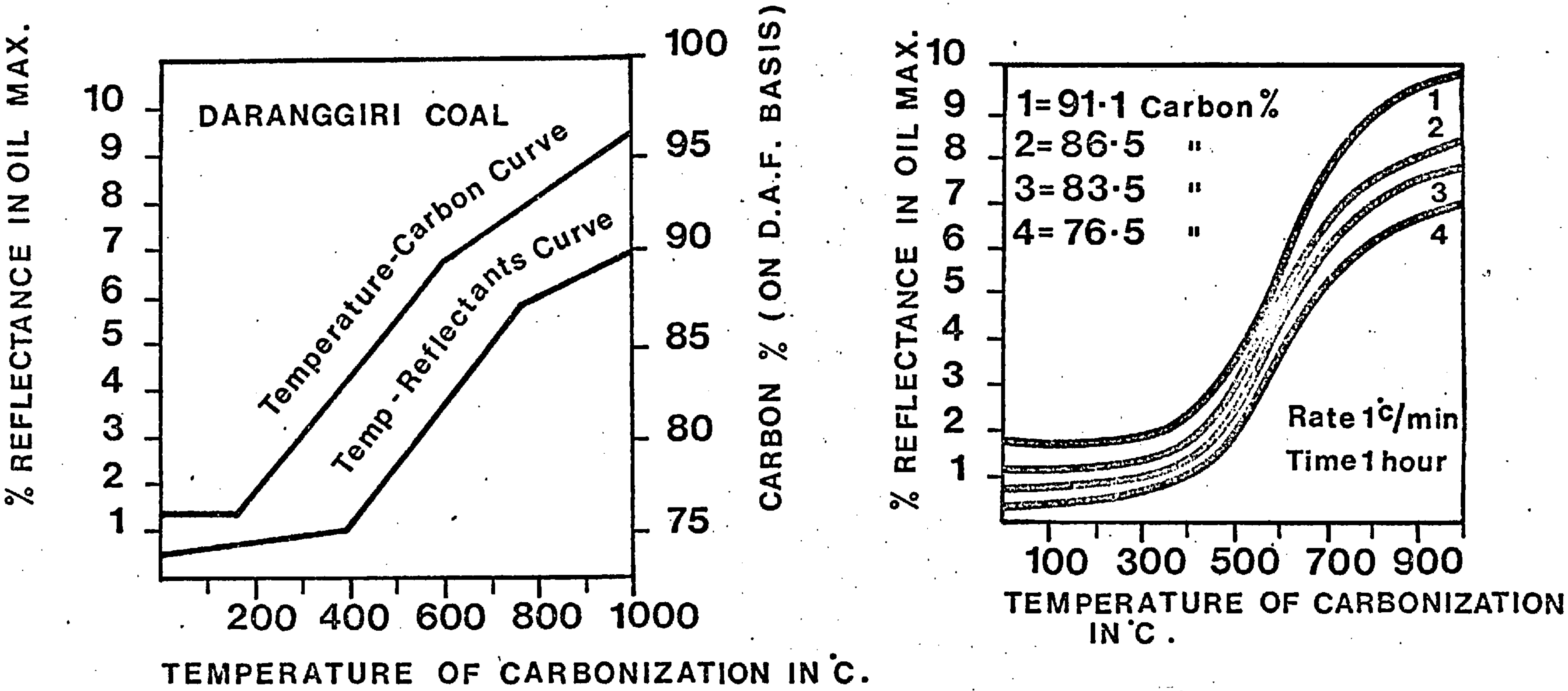


Fig. 8-2 Relationship between Temperature, Reflectance & Carbon content of Laboratory Carbonized Coals of the Daranggiri Coalfield India. After Gosh 1968

In another Australian example, in which a porphyrite was intruded into a medium-volatile bituminous coal, temperatures below 760°C were indicated from their reflectance (Kisch and Taylor, 1966).

Chandra (1965) studied the combined effect of temperature and pressure on a single coal. When the coal was heated at 325°C or 350°C under pressure up to 6000 atmospheres, reflectance increased and continued to do so with time. Chandra suggested that pressure was responsible for the rise. Pressure could modify the temperature level at which the reflectance increase took place and possibly its rate of change, but it does not seem likely that pressure alone could cause the reflectance rise, although it could have contributed to the development of anisotropy which was observed at the higher temperature.

This view is supported by the results of Hryckowian, Dutcher and Dachille (1967) who heated anthracites in the temperature range 400 to 900°C at pressure up to 20,000 lb./in^2. They showed that pressure alone produced little change in reflectance, and also demonstrated that rise of temperature caused an increase in reflectance which was greatest in the temperature range where there was a maximum evaluation of volatiles.

Response of the various types of organolites to reflectivity variations:

The various types of organolite are not equally sensitive to the same biochemical and physical conditions, e.g. exine extinction is different from that of tissues (Alpern, 1970). Fig. 8.3 shows the variation of sensitivity of the different organolite groups to carbonization. It is clear that vitrinite is the most sensitive mineral to the carbonization. For this reason, it has been recommended by many authors that vitrinite is the most suitable organolite type for reflectivity measurements.

Brown, Cook and Taylor (1964) have classified the vitrinite of a wide range of coals into two types, namely, vitrinite (A), and vitrinite (B). Vitrinite (A) has a higher reflectance and lower volatile content than vitrinite (B). They concluded that carbonization behaviour of coals could be correlated more precisely with the reflectance of vitrinite (A).

Recently, Benedict et al. (1968) have indicated that a poorly reactive fraction of what is recognised as vitrinite should be referred to as "pseudovitrinite". They suggested that pseudovitrinite is a separate maceral which can be distinguished from the remainder of the vitrinite in the coal on the basis of several characteristics, including a higher reflectance in oil.

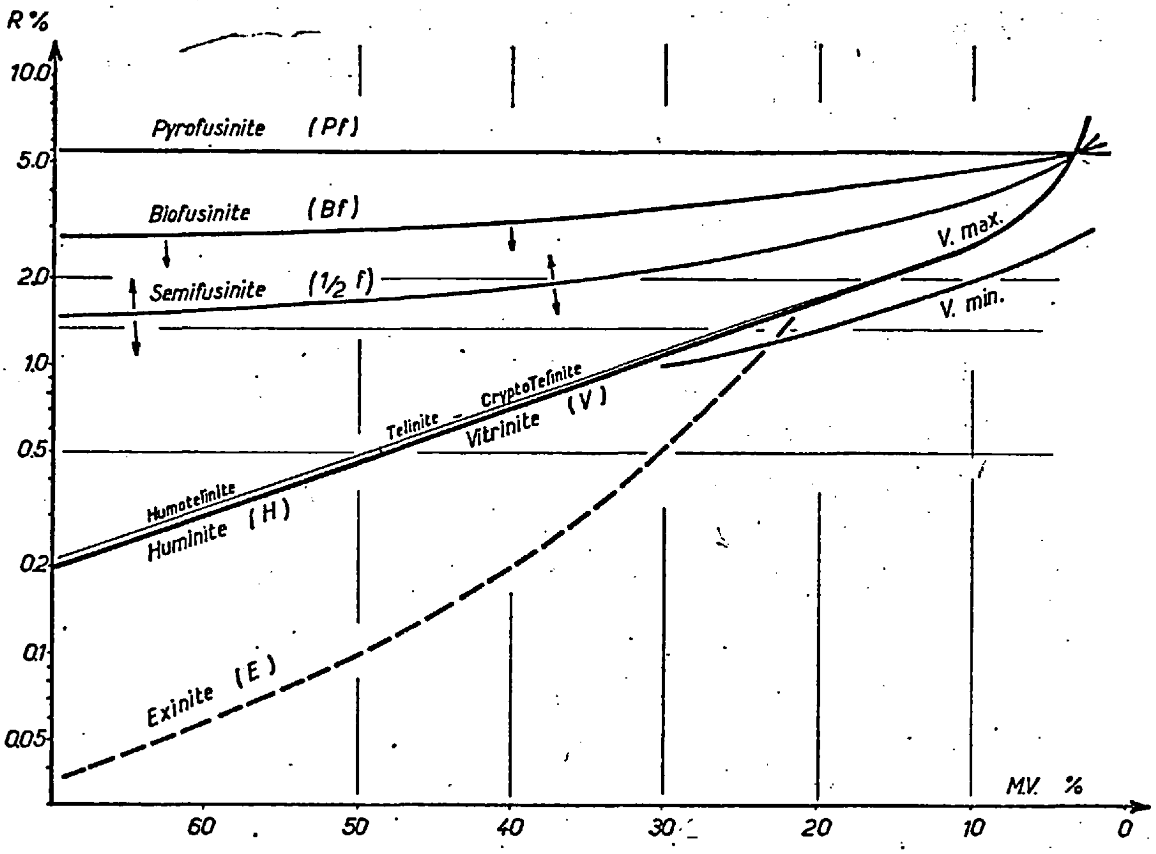


FIG. 8.3 EVOLUTION OF REFLECTIVITY OF THE VARIOUS MACERALS DURING CARBONIZATION (AFTER ALPERN, 1970)

Cryptotelinite in the studied samples is correlated with vitrinite (A) of Brown et al. (1964). Therefore it has been used for the reflectivity measurements.

Reflectivity variation:

(a) In the Carboniferous Limestone rocks:

Reflectivity measurements of vitrinite in the Carboniferous Limestone rocks are given in Table 8.5.

It was found that the maximum reflectivity in oil of the vitrinite in the Carboniferous Limestone rocks varies from 1.69% to 4.81%. The lowest reflectivity value has been determined in rocks from the Forest of Dean while the highest value has been attained in rocks from the Torcoed area in the western end of the South Wales Coalfield.

Fig. 8.4 shows the variation in the vitrinite reflectivity % in oil in the Carboniferous Limestone rocks. Almost regular increase in the reflectivity values going from the Forest of Dean toward the western end of the South Wales Coalfield can be seen. The reflectivity values in rocks from the Merthyr Tydfil area were found to be higher than expected. This may be due to the effect of the Neath disturbance which might increase the grade of metamorphism of the rocks in this area.

TABLE 8.5 REFLECTIVITY OF VITRINITE GRAINS IN THE CARBONIFEROUS LIMESTONES

Locality	Sample No.	Lithology	Av. R. air %	No. of readings	Av. R. oil %	No. of readings
Gower	614	Biomicrite	11.79	6	3.09	5
	622	Biomicrite	11.22	7	2.26	6
	615	Biomicrite	11.30	5	3.81	5
Torcoed	669	Biosparite	12.84	5	3.81	3
Cilwen	655	Biomicrite	10.17	4	2.31	4
	659	Biomicrite	11.54	3	2.10	5
Llanybei	664	Shale	11.23	4	2.12	3
Merthyr	105	Biomicrite	8.07	5	2.74	4
Ystrad	BY65	Dolomicrite	8.66	4	1.85	3
	BY37	Biomicrite	8.64	3	2.17	3
Tongwynlais	A9	Dolomicrite	8.70	5	1.92	4
	C02	Biomicrite	8.64	4	1.84	4
Forest of Dean	1069	Calc.Shale	8.66	5	1.69	6

TABLE 8.6 REFLECTIVITY OF VITRINITE GRAINS IN THE MILLSTONE GRIT ROCKS

Locality	Sample No.	Lithology	Av. R. air %	No. of readings	Av. R. oil %	No. of readings
Glynneath	81	Clayey Siltstone	9.09	4	2.68	6
	88	Shale	9.81	5	1.69	5
	84	Quartz Wackestone	8.95	4	2.00	4
	90	Litharenite	7.74	5	1.75	5
Gower	623	Shale	9.07	4	1.54	3
Kidwelly	661	Shale	9.38	4	2.65	5

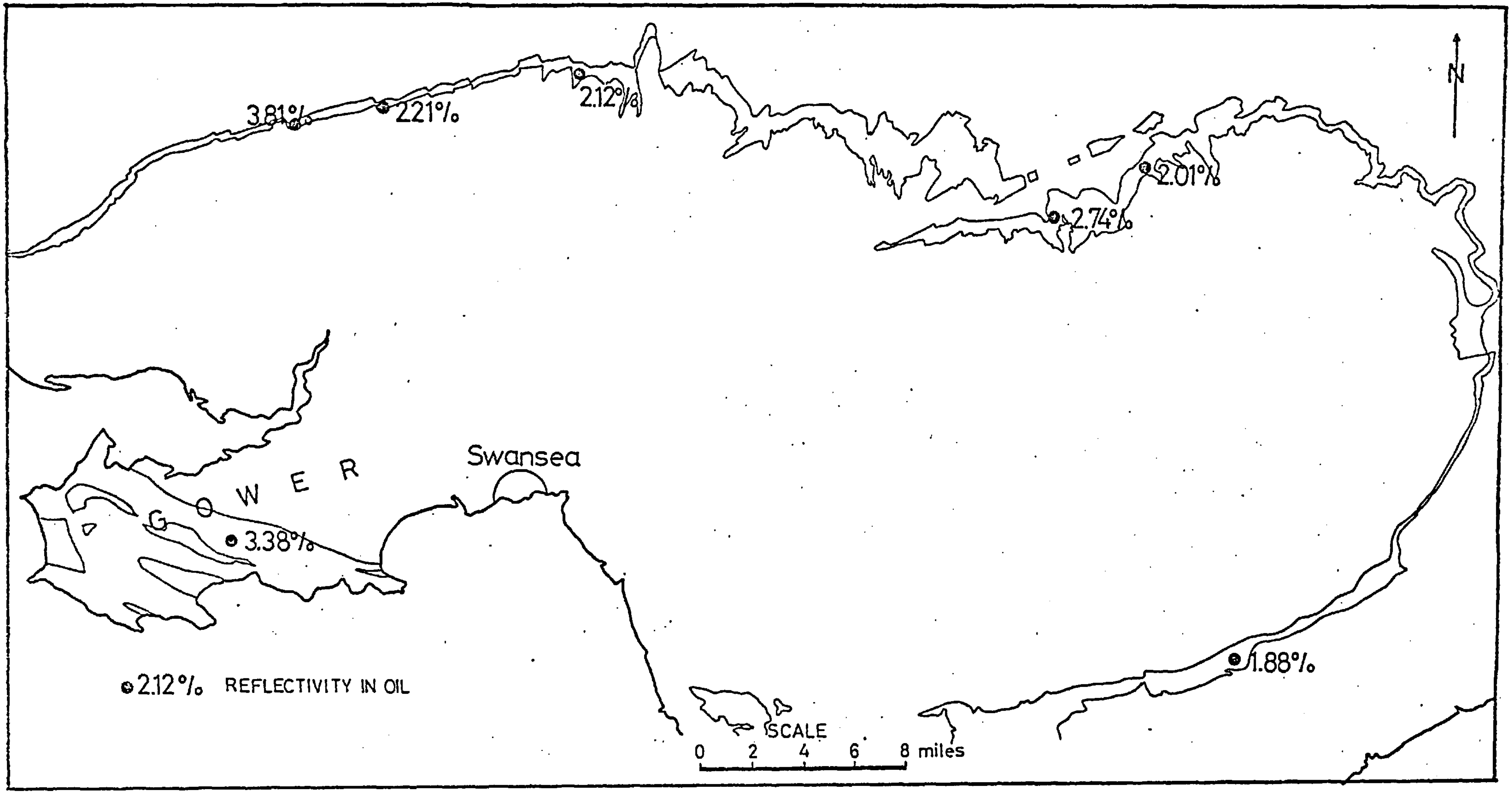


FIG.8.4 REGIONAL VARIATION IN THE REFLECTIVITY VALUES IN THE CARBONIFEROUS LIMESTONE

(b) In the Millstone Grit rocks:

Reflectivity has been measured in Millstone Grit rocks collected from three localities, namely, Glynneath, Gower and Kidwelly, Table 8.6. The average of the maximum reflectivity in oil were found to be 2.08%, 1.54% and 2.65% respectively. The trend of increase in reflectivity from east to west and from south to north of the coalfield can be noticed.

(c) In the Coal Measures rocks:

Reflectivity of vitrinite grains has been measured in Coal Measures rock samples collected from different localities and at various depths. Results are given in Table 8.7. At each locality, the average of the reflectivity in oil values has been calculated and they were plotted on a map. An isoreflectivity map has been produced by contouring these values, Fig. 8.5. It has been concluded that vitrinite grains in the samples studied range in reflectivity from 1.06% (at the eastern border of the coalfield) to 3.08% (at the north-west border of the coalfield). It was found also that reflectivity progressively increases westwards.

On the contrary, no regular variation in the reflectivity values was noticed in relation to the depth of burial.

TABLE 8.7 REFLECTIVITY OF VITRINITE GRAINS IN THE COAL MEASURES ROCKS

Locality	Sample No.	Lithology	Av. R air %	No. of readings	Av. R oil %	No. of readings
Brynamman	829	Sandstone	9.81	3	2.46	2
	826	Sandstone	8.51	3	2.44	3
	911	Sandstone	11.80	4	3.08	3
Rhydy-meardy	870	Quartzarenite	8.63	7	2.75	5
	871	Seatearth	10.82	8	3.02	5
	907	Claystone	9.38	7	2.77	4
Ammanford	719	Wackestone	9.23	8	2.31	7
Treforgan	760	Seatearth	7.20	8	2.84	6
	756	Seatearth	7.19	7	2.00	7
Bleanant	728	Litharenite	7.21	4	1.54	3
	733	Sandstone	10.68	6	3.06	4
	731	Claystone	7.20	7	2.55	7
Morlias	746	Claystone	10.10	8	2.77	7
	745	Litharenite	10.42	4	2.89	4
Abernant	706	Seatearth	8.62	8	2.55	5
Mardy	356	Siltstone	8.66	6	2.30	6
	360	Siltstone	9.22	6	2.70	5
	306	Litharenite	7.76	4	1.38	3
Marine	131	Claystone	7.21	9	1.40	7
Bedwas	249	Sandstone	5.77	10	1.07	7
	235	Sandstone	6.92	9	1.15	8
	247	Silty Claystone	7.93	11	1.07	8
Penallta	185	Mudstone	8.37	8	1.70	6
Penrikyber	116	Shale	7.67	8	2.03	7
Abercynon	170	Sandstone	7.56	8	1.23	6
Six Bells	153	Silty Claystone	5.77	7	0.92	7
	151	Sandstone	7.56	6	1.03	5
	143	Shale	8.22	8	1.54	8
	141	Sandstone	8.66	4	1.50	4
Forest of Dean	1005	Claystone	7.21	7	1.08	6
	1037	Litharenite	5.76	8	0.77	5
	1022	Quartz Wackestone	7.13	3	1.07	3
Parkship	2943	Claystone	7.21	7	1.06	5
Pentrych	388-6	Sandstone	7.30	5	1.23	5

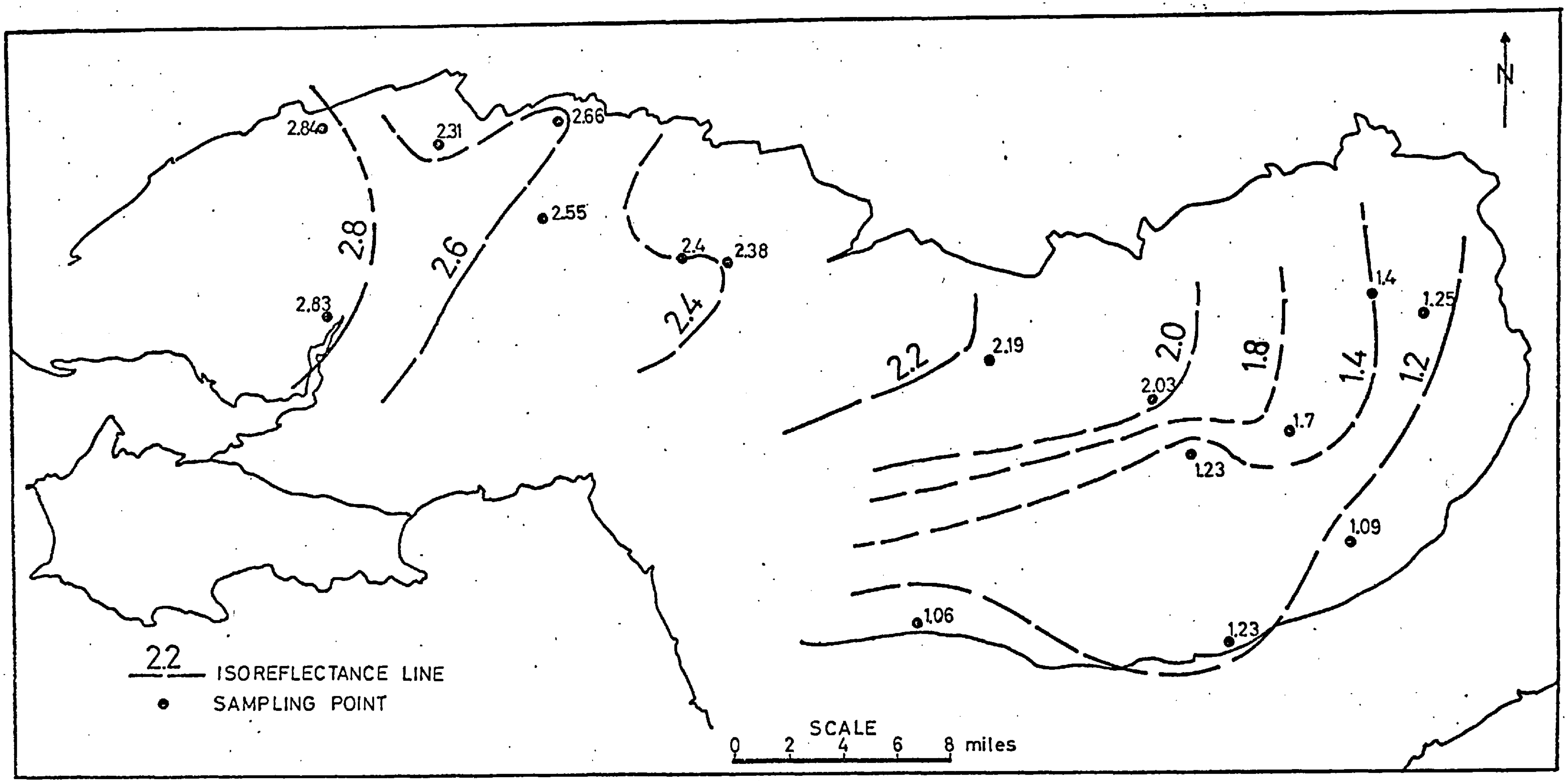


FIG.8.5 ISOREFLECTIVITY MAP OF THE COAL MEASURES.(R_{oil}%)

In Fig. 8.6 the vitrinite reflectivity % in air and in oil has been plotted against the fixed carbon % in the associated coal seams. It can be noticed that the reflectivity % increases with increase in the fixed carbon %, i.e. with increase in the grade of metamorphism.

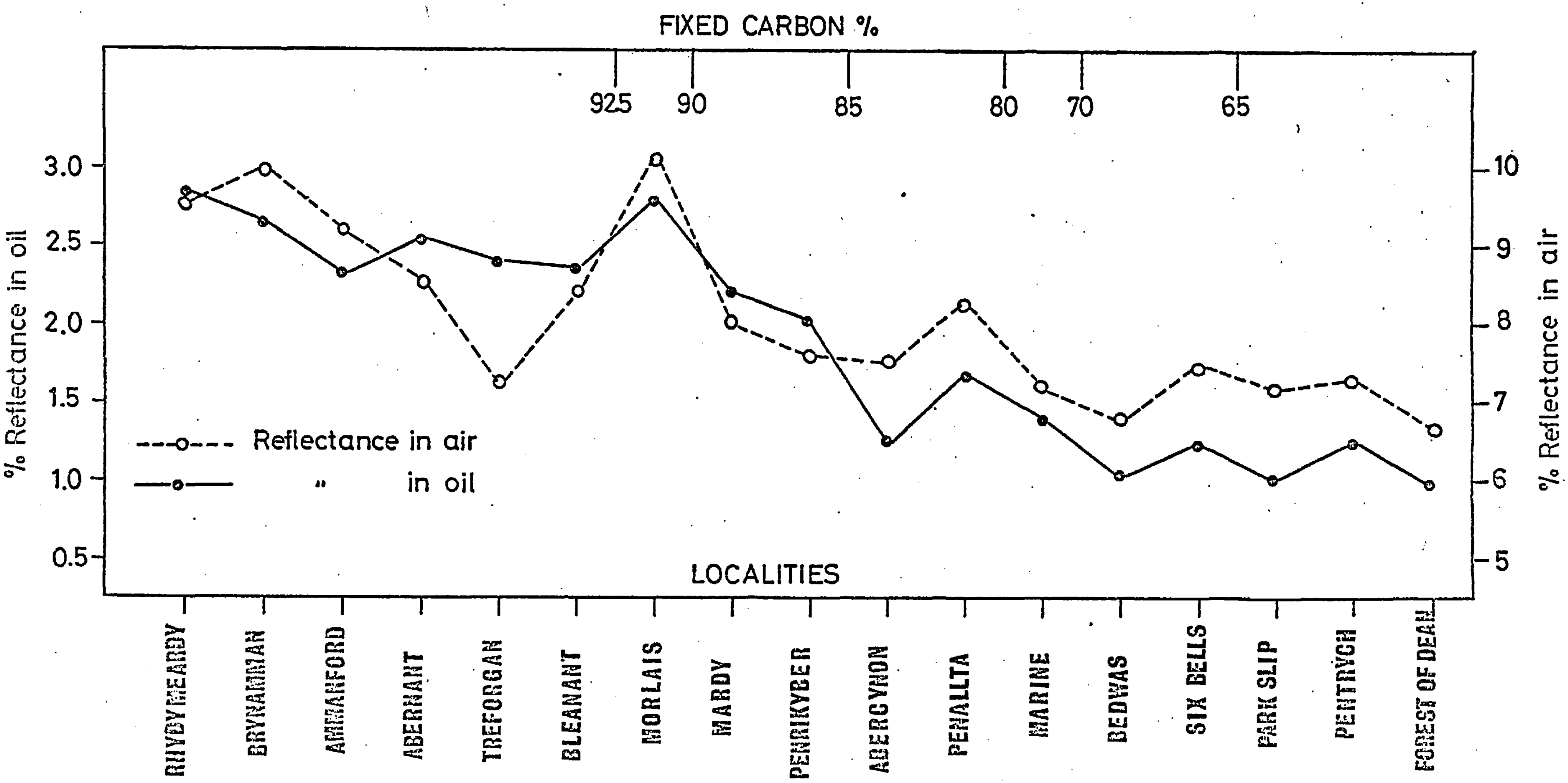


FIG.8.6 Relationship between reflectivity% of vitrinite grains in the Coal Measures rocks and the fixed carbon% in the associated coal seams.

C. EXAMINATION OF ORGANIC MATTER IN TRANSMITTED LIGHT

GENERAL STATEMENT

Pollen grains and spores in coals from different ranks studied under the miscoscope in transmitted light after maceration or in thin sections, gradually changes in colour from yellow to light brown to dark brown-black. Changes in colour of pollen grains and spores from other sedimentary rocks, such as shales, sandstones, etc. are similar. The term carbonization has been used by Gutjahr (1966) to denote this alteration process. The chief interest and importance of carbonization investigation of pollen grains and spores is that the carbonization measurements can be used as indices of the level of incipient metamorphism of the organic matter.

Stadinchenko (1931) studied the effect of temperature on fossil organic matter as well as on organic matter in Recent deposits. She found that when heat is gradually applied to thin sections of these sediments, they first change in colour, slowly turning creamy yellow, then golden yellow and orange. Fusion takes place at 380-390°C and the substance volatilises at about 425-430°C. She concluded also that carbonization of the plant substances results in raising the temperatures of fusion and volatilization and in change in optical characteristics from isotropic to anisotropic.

Wilson (1961) found a close relation between the coal rank and the degree of carbonization of spores and pollen grains in the Arkoma Basin of Oklahoma. In high-volatile coals (50 to 60% fixed carbon) spores are yellow to dark brown, while those in medium-volatile coals (60-70% fixed carbon) are dark brown to black.

Gutjahr (1966) found measurable effects on light transmission of Recent Quercus Robur pollen grains heated at 100°C for a week, drastic carbonization changes were observed at higher temperatures (viz. 150°C and 200°C).

Staplin (1969) studied the metamorphism of the organic matter with relation to oil and gas occurrence in British Columbia, Canada. He used mainly the colour of the insoluble organic matter as an indicator for the degree of metamorphism and therefore the variation in the nature of the associated hydrocarbons.

Correia (1971) studied the effects of depth of burial and therefore temperature, on the degree of preservation of the spores and pollen grains as well as the other organic remains in some sedimentary basins in France, Africa and South America. He found that the degree of preservation (represented by the translucency) of the organic matter decreases with increase in the depth of burial.

In the present study, the degree of carbonization of spores and pollen grains was used to determine the grades of incipient metamorphism of the carboniferous rocks in the South Wales Coalfield.

METHODS OF STUDY

1. Samples:

116 samples have been chosen for the study. They are distributed in the following order: 16 samples from the Carboniferous Limestone; 6 samples from the Millstone Grit; and 90 samples from the Coal Measures.

2. Sample preparation:

The procedure used for separation of the organic matter from the rocks has been recommended by Dr. M. Muir (personal communication).

(a) A fresh lump of the rock of about 2 cu. cm. was crushed to about 1 mm. grain size.

(b) Carbonate rock samples were treated with 10% HCl until almost all of the carbonate mineral matter had been dissolved. In some of the terrigenous rocks which contain considerable amounts of diagenetic siderite, the crushed sample was soaked in warm 20% HCl for about six hours with continuous stirring.

(c) Samples, both treated with HCl and untreated, were washed very thoroughly in distilled water three times.

(d) Samples were then washed in several liquids to make the sample pass from pure distilled water washed to pure acetone washed. These liquids are:

i) Acetone 25% + water 75%.
ii) Acetone 50% + water 50%.
iii) Acetone 75% + water 25%.
iv) Pure acetone.

(e) 2.2 sp. gr. heavy liquid (66% bromoform + 34% acetone) was used for the separation of the organic matter from the mineral matter. The heavy liquid was added to the acetone-washed sample, then shaken well and centrifuged. The heavy liquid with the light fraction, which is the organic matter, was filtered through a fine filter(2-4µ size) using a vacuum pump. The mineral matter is discarded.

(f) The filtrate (organic matter) was washed twice in acetone. Then it was washed again in the various mixtures of acetone and distilled water to pass from acetone-washed to pure distilled water-washed.

(g) The organic matter was then mounted on a glass slide using glycerin gel as a permanent mount media. Three slides are prepared for each sample.

MEASUREMENT OF THE DEGREE OF CARBONIZATION

Organic particles show metamorphic evolution by becoming progressively more opaque and the colour darkens, passing from transparent yellow to orange red, reddish-brown and finally to black. This colourimetric evolution is progressive, but manifests itself differently on various parts of individual microfossils of organic debris (Correia, 1971).

In order to determine the level of organic metamorhpism, two methods have been developed:

(a) The simplest and fastest of the two methods to measure the degree of carbonization of pollen grains and spores is the subjective evolution of the colour of the spores and pollen grains by their appearance under the microscope in transmitted light.

(b) Colour changes of pollen grains and spores with increasing incipient metamorphism are accompanied by a decrease of translucency to final opacity as observed with transmitted light. As a result of this observation, Gutjahr (1966) has designed a method of measuring the translucency of pollen grains and spores using a photo-electric cell.

In the present study the first method has been used. A numerical representation has been suggested to transfer the colour observed into number (carbonization index). The carbonization indices used are as follows:

Carbonization Index	Colour
1	Yellow
2	Orange
3	Golden Brown
4	Chocolate Brown
5	Dark Brown
6	Black

The results of the carbonization measurements of all the samples studied are tabulated in Table 8.8.

RESULTS

Fig. 8.7 shows the isocarbonization map of spores and pollen grains in the Coal Measures rocks of South Wales. The carbonization indices vary from 2 in the east and the south to 6 at the north-western end of the South Wales Coalfield. It was found that the degree of carbonization of the spores and pollen grains is progressively increasing in a regular pattern westwards. By comparing the isocarbonization contours with

TABLE 8.8 DEGREE OF CARBONIZATION OF SPORES AND POLLEN GRAINS

Locality	Sample No.	Lithology	Carbonization Index
(A)	CARBONIFEROUS LIMESTONE		
Gower	616'	Underclay	5
	606	Biomicrite	2
	622	Biomicrite	2
	609	Biosparite	2
Torcoed	664	Shale	2
	667	Biomicrite	3
Cilwrchen	655	Biomicrite	3
	659	Biomicrite	3
	654	Shale	3
Ystrad	BY52	Biomicrite	3
Brynmawr	BY37	Biomicrite	3
	BY32	Biosparite	2
	BY24	Dolomite	2
Tongwynlais	A9	Dolomicrite	2
	M7	Oosparite	2
Ruthin	C02	Micrite	3
	B03	Biomicrite	3
Forest of Dean	1030	Quartzarenite	2
	1013	Biosparite	2
	1041	Shale	2
(B)	MILLSTONE GRIT		
Gower	614	Shale	3
	623	Shale	3
Glynneath	88	Shale	3
	81	Siltstone	3
(B)	COAL MEASURES		
Forest of Dean	1005	Claystone	2-3
	1038	Underclay	2-3
	1001	Claystone	3
	1002	Quartzwacke-stone	2
	1037	Litharenite	2
Park Slip	2493-10	Underclay	3
	1479-12	Mudstone	2
	1480-4	Underclay	3
	1479-5	Shale	2
Pentrych	420-4	Mudstone	2-3
	40-3	Sandstone	2-3
	388-2	Siltstone	2
Sixbells	143	Mudstone	3
	153	Underclay	3
Bedwas	250	Mudstone	3
	241	Mudstone	3
	242	Mudstone	3-4
	243	Sandstone	4
	247	Siltstone	2-3
	249	Sandstone	4
Marine	131	Underclay	3
	120	Mudstone	3-4
Coegnant	216	Mudstone	4
	221	Mudstone	4
Faldna	227	Mudstone	5
	225	Underclay	4
	231	Underclay	4
Penallta	188	Shale	4
	186	Underclay	4
	181	Siltstone	4
Abercynon	171	Mudstone	4
	420	Underclay	4
Penrikyber	116	Shale	5
	113	Underclay	
Mardy	308	Underclay	6
	356	Underclay	6
	353	Mudstone	6
	313	Siltstone	6
Bleangwr-ach	189	Shale	6
	199	Underclay	5-6
	201	Shale	5-6
Treforgan	759	Mudstone	6
	756	Underclay	6
	760	Underclay	6
Bleanant	727	Shale	6
	731	Shale	6
	724	Underclay	6
	722	Quartzarenite	6
Morlias	749	Underclay	6
Brynwilli	716	Underclay	6
	716	Litharenite	6
Abernant	708	Claystone	6
Cumgorse	258	Sandstone	6
Brynamman	783	Mudstone	5
	777	Underclay	6
	790	Underclay	6
	865	Underclay	6
	817	Mudstone	6
	912	Mudstone	6
	803	Shale	6
	914	Shale	6
	829	Litharenite	5-6
	834	Litharenite	6
	825	Claystone	6
Ammanford	720	Shale	6
Pantyffnon	266	Sandstone	6
	265	Mudstone	6
Rhydymeardy	871	Underclay	5
	857	Mudstone	6
	874	Mudstone	6
	907	Mudstone	6
	902	Mudstone	6

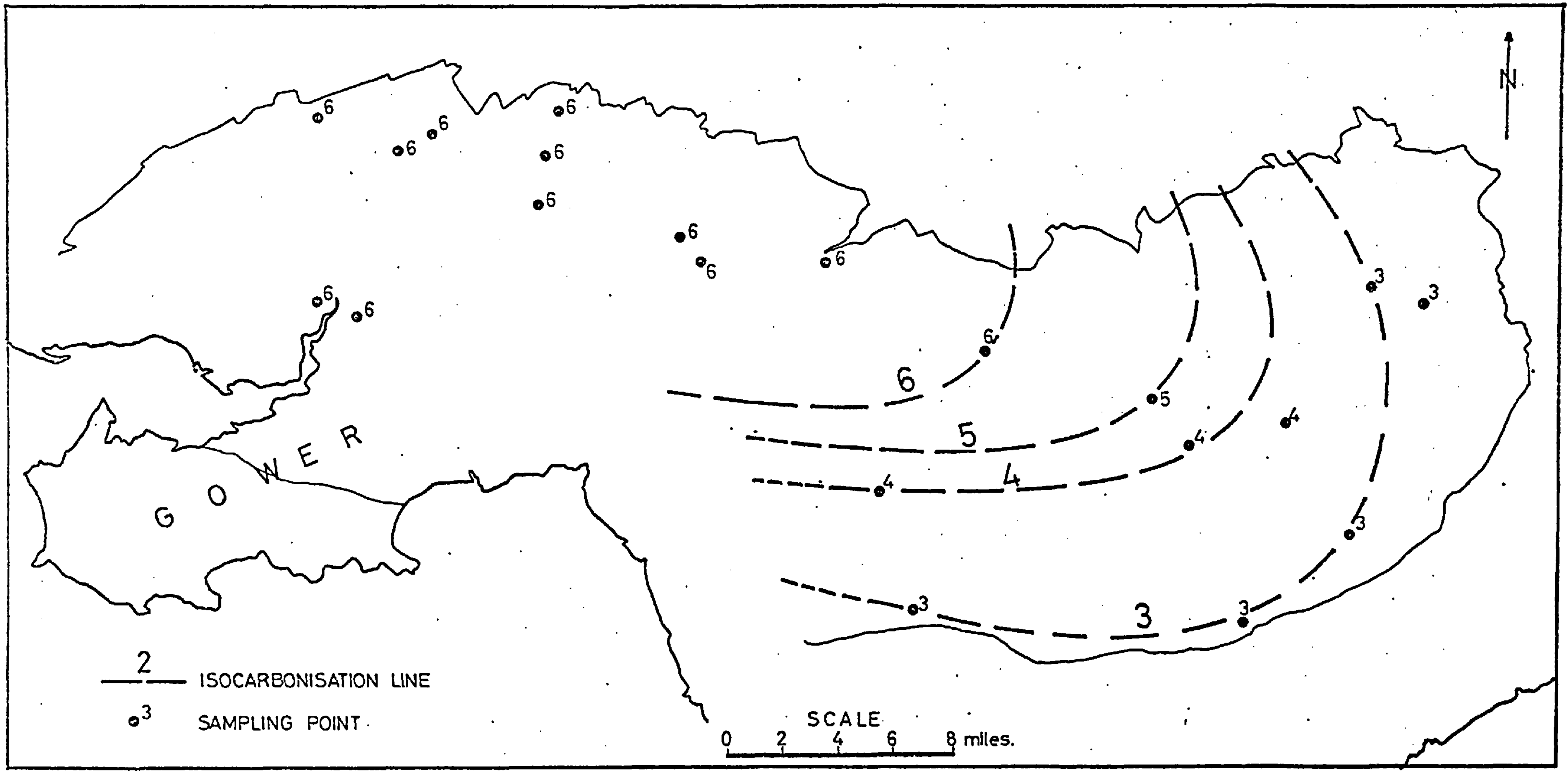


FIG.8.7 ISOCARBONISATION MAP OF THE COAL MEASURES.

the isovol contours of the South Wales coalfield, it was found that they are almost parallel to each other.

In the Carboniferous Limestone rocks it was found that the variation in the degree of carbonization of the spores and pollen grains is very limited. The carbonization indices range from 2 to 3. By comparing the degree of carbonization of spores and pollen grains in the terrigenous rocks of the Coal Measures with those in the carbonate rocks from the same metamorphic zone (anthracite zone), it has been noticed that they respond differently to the metamorphism. This observation indicates the effect of lithology on the metamorphic evolution of the organic matter.

In Fig. 8.8 the carbonization indices of the spores and pollen grains in the Coal Measures rocks are plotted against the fixed carbon % in the associated coal seams. It can be seen that the degree of carbonization of spores and pollen grains is progressively increasing with increase in the fixed carbon % of the associated coal seams.

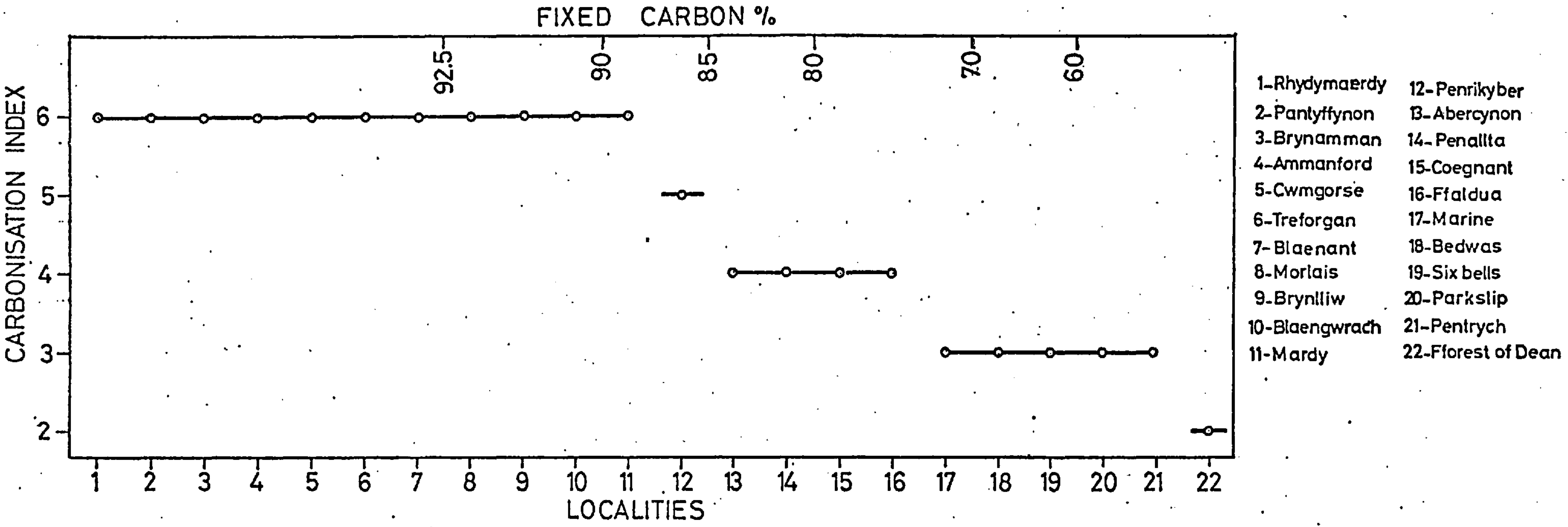


FIG.8.8 THE RELATIONSHIP BETWEEN THE DEGREE OF CARBONISATION OF SPORES AND POLLEN GRAINS IN THE COAL MEASURES ROCKS AND THE FIXED CARBON% OF THE ASSOCIATED COAL.

CHAPTER IX

DISCUSSION AND CONCLUSIONS

DISCUSSION AND CONCLUSIONS

DIAGENESIS AND INCIPIENT METAMORPHISM

One of the most problematic subjects facing both sedimentary and metamorphic petrologists is the definition of the transitional stage between diagenesis and true regional metamorphism. Broadly defined, diagenesis includes all those changes that occur in the sediment and rock up to the time that metamorphism commences. The point at which metamorphism can be said to start is somewhat vague. Sujowski (1958) has defined the term diagenesis to include all those processes which turn a fresh sediment into a stable rock of some hardness under conditions of pressure and temperature not widely removed from those existing on the earth's surface. On the other hand, Pettijohn et al. (1972) have defined diagenesis to include all those processes, chemical and physical, which affect the sediment after deposition and up to the lowest grade of metamorphism (the green schist facies).

The stage between diagenesis and the green schist metamorphic facies has been studied in the terrigenous rocks by many geologists and clay mineralogists, and many names have been given to this stage. Lands (1967) has termed this the stage of cometamorphism. The Russian geologists have studied this stage in detail in the terrigenous rock, and their work was mainly based on the clay mineral modifications. Logvinenko (1957) has used the terms eipgenesis and metagenesis, and Koplan (1971) has used the term catagenesis to indicate this stage. French geologists usually use the term anchiometamorphism (Kubler, 1964), while American geologists use the term burial metamorphism (Burst, 1959). Brown (1972) has studied the post-diagenetic modification in the carbonate rocks, and he used the term incipient metamorphism for these modifications.

In the present study incipient metamorphism is defined to include all those processes affecting both organic and inorganic rock constituents, between diagenesis and low-grade regional metamorphism (green schist facies).

DIAGENESIS OF THE CARBONATE ROCKS

Carbonate rocks forming most of the Carboniferous Limestone Series are represented by five main lithotypes, namely; micrites, biomicrites, biosparites, oolitic limestones and dolomites. Four diagenetic processes are responsible for the modifications of the carbonate sediments, namely; biological, physical, physio-chemical and chemical processes.

Biological processes, represented by micritization, algal borings and algal encrustation, have played an important role in the modifications of the original fabric and the textural parameters of most of the skeletal debris. Algal borings have severely affected some skeletal debris, resulting in their bio-fragmentation, while algal encrustations are responsible for coarsening of other skeletal debris. One of the most significant effects of the skeletal debris micritization is the replacement of their original fabrics (monocrystalline fabric of the echinoderm fragments, fibrous calcite and aragonite fabric of the bivalvia, etc.) by crypto-crystalline micrite.

Physiochemical processes are represented by the replacement of aragonite by calcite. Comparing the studied Carbonate rocks with their analogous Recent Sediments, it has been suggested that aragonite and high-Mg calcite which once formed the skeletal debris of some molluscans, oolites and part of the micrite matrix, have been completely replaced by low-Mg calcite. Two processes were suggested for the replacement of aragonite by low-Mg calcite; these are: (a) dissolution of the unstable aragonite and precipitation of the stable low-Mg calcite in the resulting cavities; and (b) in situ inversion of aragonite to calcite in most of the studied rocks. Inversion of aragonite to calcite in the studied rocks took place in the very early stage of diagenesis, approximately after the modification by the biological processes and before cementation.

Physical processes represented by compaction, mechanical deformation and pressure solution have affected the sediments at various stages in their diagenetic history. Compaction of some of the biomicrite sediments has modified the original shape of the allochems (especially pellets and oolites). In micrites compaction is responsible for the development of vugs and cavities. Compaction has affected the sediments in the early stage of diagenesis, before cementation has taken place.

Two stages of mechanical deformation have been noticed, namely: early diagenetic and late diagenetic. Early diagenetic mechanical deformation has taken place while the allochems were physically unstable under the pressure exerted by the overburden. This process is responsible for spalling-off of the outer shells of some oolites, collapsing of some micrite envelopes and the developments of crumbly fractures. It is suggested that this process affected the sediments after the dissolution of aragonite and before the complete cementation. Late diagenetic deformation has affected the sediments after they have been completely cemented and lithified. This process has led to the development of a large scale fractures which could be related to tectonism.

The Pressure Solution process has affected rocks at two stages. Early diagenetic pressure solution has taken place during the compaction and resulted in the development of stylotitic grain contacts. Late diagenetic pressure solution has affected the rocks after their complete cementation, and is responsible for the development of stylolites.

Chemical processes have played the most important role in the lithification and the modification of the carbonate lithotypes, and they are represented by cementation, dolomitization, dedolomitization and silicification.

Cementation is the most significant diagenetic modification affecting almost all of the carbonate lithotypes.

It is responsible for the lithification and destruction of the original porosity of the sediments. Non-ferroan calcite, ferroan calcite and dolomite are the main minerals forming the cement. Non-ferroan calcite is the most common cement mineral, especially in the early stages of cementation. It is suggested that the Carbonate material forming the cement has been derived from the following sources; (a) the Carboniferous sea water, (b) the excess calcium carbonate released during the inversion of aragonite to low-Mg calcite, (c) calcium carbonates released during the early and late diagenetic stylolitization.

Four main fabric types have been identified among the calcite cement, namely; (a) fibrous drusy calcite, (b) radiaxial fibrous mosaic calcite, (c) equicrystalline mosaic cement, and (d) rim cement or syntaxial calcite overgrowths. It was found that two main factors have controlled the fabric of the cement, namely; (a) the nature of the place on which the cement has been precipitated, i.e. fracture, vug, mould, intergranular pores, etc., and (b) the nature of the substrata on which the cement has been precipitated, i.e. monocrystalline, polycrystalline, etc. Cementation has developed in the early stages of diagenesis and continued up to the late stages after the dolomitization and the tectonic deformation. Four stages of cementation have been recognised. Variations in the fabric and the mineralogy of the cement have been noticed in shifting from a certain stage to another.

Dolomitization has greatly modified both the composition and the textures of the limestone lithotypes, and it is very prominent in the eastern and south-eastern outcrops. Two stages of dolomitization have been recognised, namely; syndiagenetic and anadiagenetic. Syndiagenetic dolomitization has affected the sediments in the very early stage of diagenesis just after the sedimentation. In this stage dolomicrite has been developed by the interaction between the freshly deposited aragonite or calcite micrite and the dense brine flow downward from the supratidal zone into the main water mass. Anadiagenetic dolomitization has taken place through a long period starting at the early stages of diagenesis after the first stage of cementation and continued even after the complete cementation of the rocks. This type of dolomite has been developed as a result of the interaction between the supersaturated pore solutions and the allochems.

All of the stages of the anadiagenetic dolomitization from the incipiently dolomitized limestone to completely dolomitized limestones, have been recognised. Anadiagenetic dolomite is usually coarse-grained and zoned. In the partially dolomitized limestones, dolomite is generally preferentially replacing micrite matrix and the micrite bodies (pellets, oolites and the micrite envelopes). The source of magnesium needed for the dolomitization has been suggested to be: (a) the hypersaline solution developed by the evaporation of the Carboniferous sea water in the supratidal areas; (b) magnesium released during the inversion of high-Mg calcite to low-Mg calcite; (c) magnesium could be derived from the clay minerals, present as disseminated grains in the limestones or as shale beds intercalated with the limestones. Two processes have been suggested for the derivation of magnesium from the clay minerals, namely; by ion exchange, and by direct reaction of the calcite with the clay minerals.

Silicification has played a limited role in the modification of the limestone lithotypes. It is most prominent in the Lower Limestone Shale group and the lower part of the Main Limestone group. Silicification is represented by beekitized crinoidal debris, developments of secondary overgrowths on the oolite quartz nuclei, and by completely silicified biomicrite and calcarenite bands. It is suggested that silicification has taken place after the first stage of cementation. Two sources of silica needed for silicification of the rocks studied have been suggested, namely; (a) silica leached from the clay minerals, and (b) silica held by some algae.

The role played by dedolomitization in the modification of the studied rocks is very limited. On the basis of the petrography and field relations three stages of dedolomitization have been recognised, namely; (a) an early diagenetic dedolomitization affecting the partially dolomitized biomicrites after they have been silicified, (b) epidiagenetic dedolomitization which has affected some of the dolomitized limestone during the time they have been exposed in Carboniferous times, (c) dedolomitization which has affected dolomites and dolomitized limestone by the action of solution on their exposed surfaces in the recent times. The first two stages of dedolomitization, (a) and (b), have been termed fossil dedolomitizationn and the last stage,(c), Recent dedolomitization.

It has been noticed that dedolomitization is preferentially affecting ferroan dolomite. Dolomite fabrics are generally preserved after dedolomitization. Replacement of dolomite by calcite is most probably took place by an exchange of calcium for magnesium from the Ca-rich surface water in the case of the surface dedolomitization or from the calcium released during the silicification of the dolomitized biomicrite in the case of the early diagenetic dedolomitization. This in part agrees with Al-Hashimi and Hemmingway (1973) suggestion on the mechanism of dedolomitization of the Northumberland Carboniferous rocks.

From the detailed study of the diagenesis of the various lithotypes of the Carboniferous Limestone group, it was found that the diagenetic history and the diagenetic modifications were controlled mainly by the original texture and composition of the sediments as well as by the physiochemical conditions of the diagenetic environments. It was found also that the final result of the diagenesis of the Carboniferous limestones is the complete destruction of their original porosity and partial destruction of their secondary porosity (porosity developed by dolomitization).

Fig. 9.1 shows the time relationship of the various diagenetic processes which affected the Carboniferous Limestones.

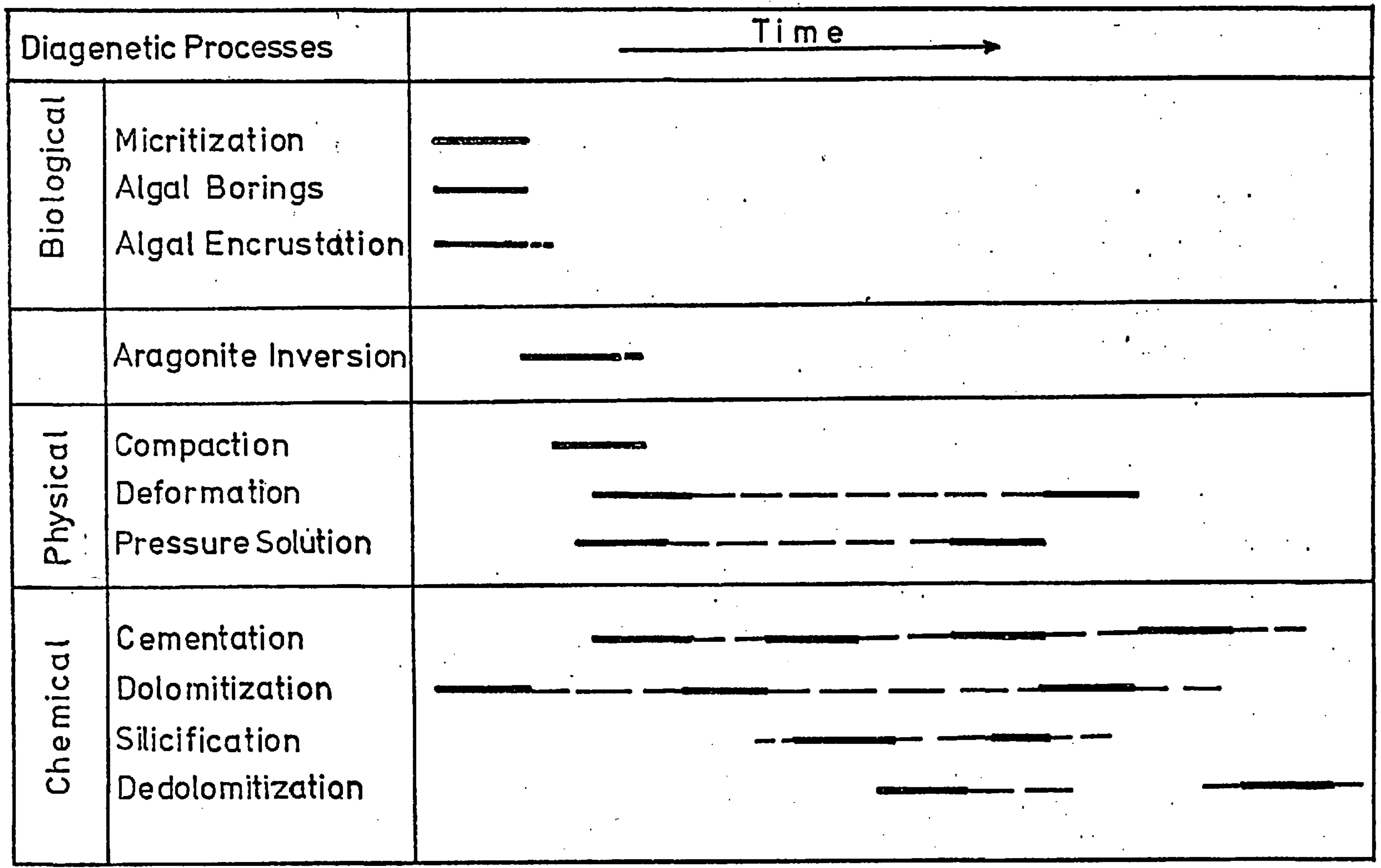


FIG. 9.1 – DIAGENETIC PROCESSES AND RELATIVE TIME RELATIONSHIPS — CARBONATE ROCKS

DIAGENESIS OF THE TERRIGENOUS ROCKS

Terrigenous rocks form the whole sequence of the Millstone Grit and the Coal Measures Series, and partly the Carboniferous Limestone Series. They are represented mainly by three sandstone lithotypes (quartzarenites, lithic arenites and quartz wackes), two siltstone lithotypes (quartzitic siltstone and clayey siltstone), and three argillaceous lithotypes (shale, mudstone and claystone.

Three main diagenetic processes are responsible for the textural and composition modifications of the rocks, namely; physical, physiochemical and chemical.

Physical processes have played an important role in the fabric modification of the framework grains of most of the siltstones and sandstones lithotypes. Grain fracturing and crushing, grain bending and grain squeezing are the most important modifications. Grain fracturing and crushing are usually displayed by the quartz and feldspar grains in the coarse-grained sandstones. Mica flakes in most of the siltstone and sandstone lithotypes are commonly bent between the rigid framework grains. In the quartzarenites and the quartzitic siltstones, mica flakes have frequently been squeezed between the quartz grains to form a lump of minute mica shreds filling the pore spaces. In the lithic arenites, the argillaceous rock fragments have been squeezed between the other rigid framework grains and they form a clayey pseudomatrix.

Pressure solution process has been effective in most of the lithic arenites and wackestones, while it is insignificant in quartzarenites and quartzitic siltstones.

Chemical processes have greatly modified the fabric and the composition of most of the terrigenous lithotypes. The following are the processes affecting the studied rocks; (a) precipitation of pore-filling minerals, (b) mineral replacement, and (c) chemical alteration.

Precipitation of pore-filling minerals process has played a significant role in the lithification of most of the siltstone and sandstone lithotypes, particularly quartzarenites and quartzitic siltstones. Quartz, dolomite, siderite and kaolinite are the main pore-filling minerals. Four types of silica cement have been recognised, namely; quartz overgrowths, microcrystalline quartz, spherulitic chalcedony and idiotopic quartz. Quartz overgrowths are the most important cement, especially in the quartzarenite and the quartzitic siltstones. Dolomite is represented mainly by the ferroan variety and has been recognised as pore-filling and fracture-filling cement in most of the sandstone and siltstone lithotypes. In some of the Coal Measures quartzarenites, it is present as the main cementing mineral. Siderite is present in most of the studied lithotypes as partially filling some pore spaces. Pore-filling kaolinite is commonly present as coarsely crystalline vermicules frequently occurring in the quartz-arenites and the quartzitic siltstones.

Mineral replacement process involves partial or complete replacement of either framework grains or newly-formed authigenic minerals. The active minerals recognised in this process are siderite, dolomite, microcrystalline quartz, kaolinite and pyrite. Siderite as a replacement mineral has played an important role in the diagenetic modification of almost all of the Millstone Grit and the Coal Measures lithotypes. In the argillaceous rocks, siderite nodules and spherules have been developed as a result of replacement of the clayey groundmass by the segregated siderite. Siderite has partially replaced quartz grains, mica flakes and lithic grains in the quartz wackes and lithic arenites of the Coal Measures. Dolomite is most active as a replacive mineral in the sandstones and siltstones. The role played by microcrystalline quartz, kaolinite, pyrite and francolite is very limited.

Besides the fact that precipitation of authigenic minerals has modified the original rock composition, they have also modified the fabric of the framework grains.

Disruption and expansion of mica flakes by the force of crystallization of siderite or dolomite has frequently been noticed. It was found also that the force of crystallization of the dolomite or siderite precipitated in the pore spaces of some quartzarenites, was responsible for the development of an open packing framework.

Chemical alteration processes are responsible to a great extent for the modification of the original mineralogy of most of the Millstone Grit and the Coal Measures lithotypes. Kaolinitization and illitization of the micas and the feldspars grains are frequently displayed in most of the studied rocks. Limonitization of siderite and ferroan dolomite occasionally takes place.

From the discussion of the origin and the conditions of formation of the authigenic minerals in the Millstone Grit and the Coal Measures lithotypes, the following conclusions have been reached:

(1) Authigenic quartz has been precipitated in the early stage of diagenesis from acidic water. Two main types of authigenic quartz have been developed, namely; quartz overgrowths and microcrystalline quartz. The former has precipitated earlier than the latter. Silica needed for the precipitation of the early diagenetic quartz

overgrowths is most probably derived from the silica released during the weathering of the detrital micas. Silica precipitated in later stages of cementation could be derived from two sources, namely; (a) from silica released during the transformation of the mica flakes to clays, (b) silica released during the replacement of the silicaceous mineral grains by dolomite or siderite.

(2) Two stages of siderite precipitation have been recognised, namely syndiagenetic and diagenetic. The abundance of the syndiagenetic siderite in the argillaceous lithotypes is due to the fact that the physiochemical conditions within the argillaceous sediments could be firmly separated from the water body above the water/sediment interface. This is assuming that the physiochemical conditions of the water body were not suitable for the formation of the siderite. Diagenetic siderite has been precipitated at shallow depth of burial. Two mechanisms are suggested for the development of this siderite, namely, by displacement and by replacement.

(3) Dolomite is frequently present in the siltstone and sandstone lithotypes. The presence of ferroan dolomite variety suggests that dolomite has been precipitated in reducing diagenetic environment at

shallow depth of burial. Magnesium and iron necessary for the formation of the ferroan dolomite are most probably derived from the clay minerals forming the matrix of some sandstones or forming the argillaceous rock fragments in the lithic arenites.

(4) Intercalations of pyrite-rich shales and siderite-rich shales could be due to the fall-off in available sulphates, which is itself a reflection of change in the salinity of the environment of deposition.

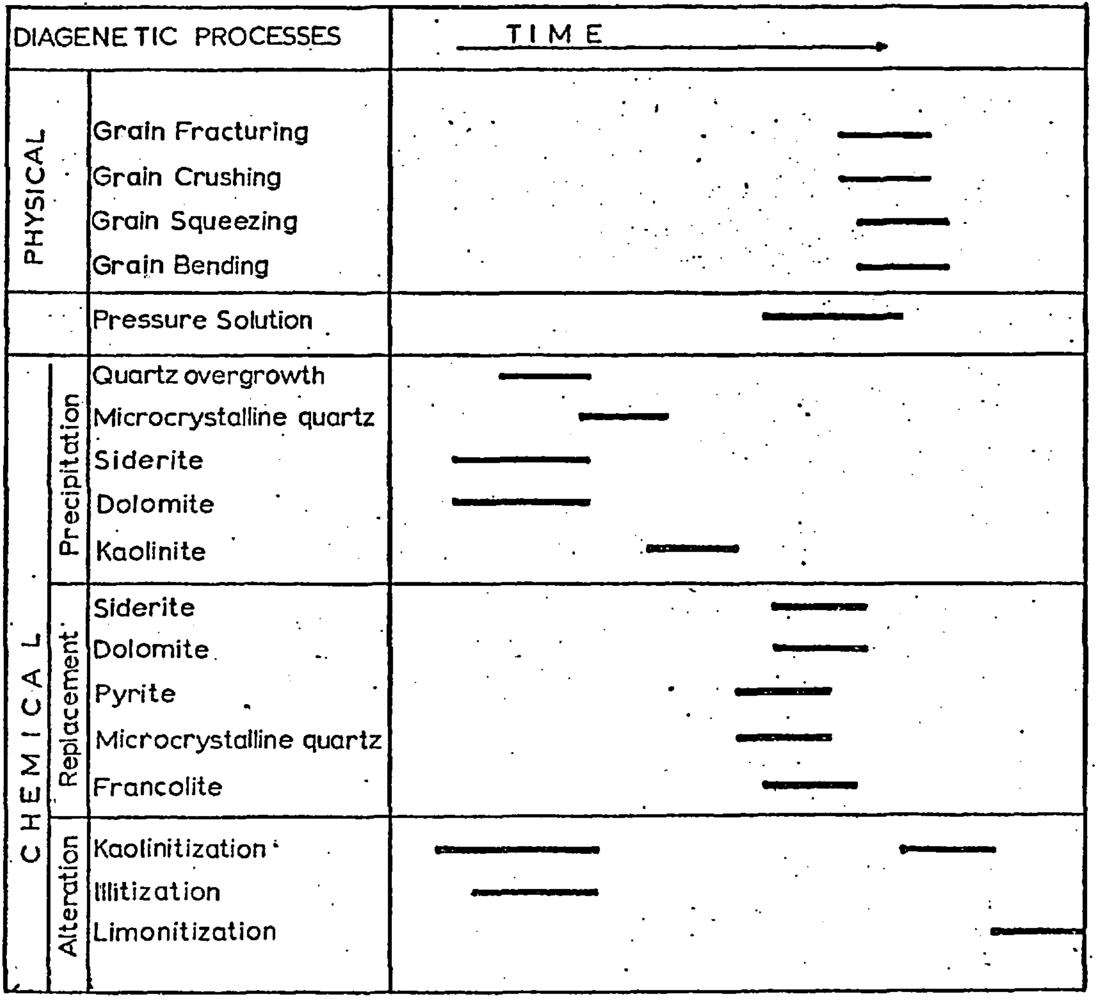
(5) Kaolinite has been developed by two main processes, namely; (a) by direct precipitation from supersaturated acidic pore solutions, and (b) by the kaolinitization of the mica flakes.

(6) Francolite is suggested to be formed by direct precipitation from supersaturated pore solutions. The phosphorous necessary for the formation of francolite might be derived from the plant tissue remains.

The end results of the diagenesis of the Millstone Grit and the Coal Measures terrigenous lithotypes is the destruction of their original porosity, either by the action of physical processes (compaction and grain squeezing) or by the precipitation of pore-filling cement or by the action of both processes together.

Fig. 9.2 shows the diagenetic processes that affected the Millstone Grit and the Coal Measures rocks and the relative time relationship.

FIG.9.2_DIAGENETIC PROCESSES AND RELATIVE TIME RELATIONSHIPS —— TERRIGENOUS ROCKS



CLAY MINERALOGY

Clay fractions separated from the carbonate lithotypes of the Carboniferous Limestone Series and from the Millstone Grit and Coal Measures lithotypes have been studied qualitatively and quantitatively by means of X-ray diffraction. Regarding the distribution of the clay minerals in the various lithotypes of Carboniferous rocks, the following conclusions have been reached.

(1) In almost all of the studied lithotypes, it was found that clay minerals are represented mainly by illite and illite-montmorillonite mixed layer and subordinate amounts of kaolinite and chlorite. Trace amounts of dickite and pyrophyllite have been detected.

(2) In the carbonate rocks, clays are present in two forms, namely; as very fine particles mixed with micrite, and as coarsely-crystalline vermicules of kaolinite and dickite filling cavities. Illite is the most frequent clay mineral in all of the Carbonate lithotypes and is present only in the 2M form. It averages 70% in micrite, biomicrites and dolomicrites, 78% in biosparites and oolitic limestones, and 84% in dolomite. Illite-montmorillonite mixed layer is abundant in micritic rocks

about (24%) more than in sparites and dolomites (about 12%). Kaolinite is commonly present in small amounts, about 5% on average. Chlorite is usually present in trace amounts in limestones, while it is completely absent in dolomites.

(3) In the terrigenous rocks of the Millstone Grit and the Coal Measures, illite-montmorillonite mixed layer clays are most abundant and illite is less frequent compared with clays in the carbonate rocks. It was found also that the distribution of kaolinite has been controlled by the lithology of the terrigenous rocks. Quartzarenites were found to be the lithotype richest in kaolinite. The abundance of kaolinite in the quartzarenites could be attributed to two reasons, namely; (a) these rocks were porous enough to allow the precipitation of a considerable amount of authigenic kaolinite, (b) the original porosity of the rock has facilitated the circulation of the pore solutions which therefore caused the kaolinitization of the mica flakes.

(4) Finally, it has been concluded that diagenesis controlled the clay mineral composition of the studied rocks to a great extent.

INCIPIENT METAMORPHISM

In the study of the incipient metamorphism of Carboniferous rocks of the South Wales Coalfield, four indicators have been used to identify the metamorphic grades of these rocks. These indicators are;

1. Fabric modifications as revealed from the petrographic study.
2. Modifications in clay mineral composition and in illite crystallinity as detected by means of X-ray diffraction.
3. Variation in the reflectivity percentage of the disseminated organic matter.
4. Variation in the degree of carbonization of spores and pollen grains.

1. Fabric Modifications

Detailed petrographic study of both carbonate and terrigenous rocks has revealed that in the western part of the coalfield (anthracite area) the original and the diagenetic fabrics have been modified. These newly formed fabrics are attributed mainly to the effect of temperature and pressure. The following is a summary of the incipient metamorphic fabrics recognised in the carbonate and the terrigenous rocks.

(a) Incipient metamorphic fabrics of the Carbonate rocks:

The main effects of incipient metamorphism are recrystallization and lattice deformation. Recrystallization is defined in the present work as a process which takes place commonly after complete lithification of the rock and is responsible for the modification of the original crystal size to sizes stable under certain conditions of temperature and pressure. Two types of recrystallization have been recognised in the Carboniferous Limestones of South Wales, namely; aggrading recrystallization and degrading recrystallization or crystal diminution. Aggrading recrystallization is responsible for an increase in the micrite crystal size and the developments of pseudospar crystals, followed by a progressive loss of original textures and ultimately partial obliteration of allochem debris. Degrading recrystallization or crystal diminution is responsible for the replacement of coarse crystals by a mosaic of finely crystalline pseudospar crystals. This type of recrystallization is displayed by the incipiently metamorphosed crinoidal debris and the coarsely crystalline fracture-filling cement.

Deformational fabrics are very common in the Carbonate rocks of the anthracite area and are represented by; (a) brush extinguished calcite and dolomite crystals, (b) bending of twin lamellae and cleavages in calcite and dolomite crystals, and (c) the development of deformed fibrous calcite veinlets.

There is a close correlation between fabric modification in the Carbonate rocks and coal rank. It was found that the development of the incipient metamorphic fabrics progressively increases towards the north-western part of the coalfield. The modifications are gradual and it is difficult to draw a borderline between metamorphosed and unmetamorphosed carbonate rocks. But, it is clear that incipient metamorphic fabrics are prominent in the area where the associated coal seams have a fixed carbon more than 80%. It has been noted that the fabric modifications are mainly lateral and no variation with increase in depth occurs.

It was found also that the fabric modifications have been controlled to some extent by the lithology of the metamorphosed rocks. Micritic rocks poor in organic matter are the most susceptible to recrystallization. Dolomite is the most stable lithotype in conditions of incipient metamorphism, the only modification noticed being the development of brush extinction.

(b) Incipient metamorphic fabrics of the terrigenous rocks:

Terrigenous rocks are found to be less susceptible to incipient metamorphism with respect to their fabric modification. The following are the main fabric modifications noticed in the incipiently metamorphosed terrigenous rocks: (a) reaction of the clayey matrix with the quartzitic framework grains, (b) the deformation of the argillaceous rock fragments and the development of pseudomatrix in the lithic arenites, and (c) the development of deformed fibrous quartz veinlets. All of these modifications occur in sandstone and siltstone lithotypes and it was difficult to notice any modifications in the fabrics of argillaceous lithotypes.

Fabric modifications in the terrigenous rocks are found to occur in the anthracite area (with fixed carbon of about 92.5%).

2. Modifications in Clay Minerals

Changes in the clay mineral composition and in the crystallinity of illite are the main results of the incipient metamorphism of the clay minerals. On a regional scale it was found that in the anthracite area some of the clay minerals become less frequent and some other new minerals have been developed. The following is the summary of these mineralogical changes.

Decrease in the percentage of the expandable layer in the randomly interstratified illite-montmorillonite mixed layer, especially in the carbonate rocks.

Appearance of regular mixed layer illite-montmorillonite (Allevradite).

Appearance of pyrophyllite and dickite.

One of the most important modifications of the clay
rals related to incipient metamorphism is the recrystall-
ion of illite in the direction of increase of regularity
heir crystal structure. Weaver's sharpness ratio has been
as an index for the determination of the illite crystall-
y. An approximate gradual increase in the crystallinity
llite towards the anthracite area of the coalfield has
noticed. Beside the fact that variation in the illite
allinity is related to the coal rank, it is found also
e controlled by the lithology. Illite in sandstones was
to be the most susceptible to crystallinity modification
r than that in argillaceous rocks and in carbonate rocks.
could be due to the fact that porosity of sandstones
itated circulation of pore water and therefore made
elements needed for the transformation of ill-crystalline
e to well crystalline illite accessible.

3. Metamorphism of Organic Matter

The disseminated organic matter in the Carboniferous rocks has been studied in reflected light and in transmitted light. The main purpose of the study of the organic matter in reflected light is the measurement of their reflectivity in polished surfaces. For this purpose, detailed petrographical study of the organic matter in polished sections has been made and Alpern's classification (1970) was used in this study. Five organolite groups have been identified, namely; Textites (xylotelinite, telinite, biofusinite, and pyrofusinite), gelites, detrites, bitumens and sporites. The mode of occurrence and the texture of the organic matter in the various lithotypes of the Carboniferous rocks have been described. Cryptotelinite grains have been used for the reflectivity measurements.

In the Carboniferous Limestone rocks no regular variation in the reflectivity percentage has been noticed, but generally it was found that in the south-eastern outcrop where the fixed carbon is about 70% the reflectivity in oil averages about 1.88%, in the northern outcrops where fixed carbon ranges from 70% to 92.5% reflectivity in oil ranges from 2.01% to 2.74%, while in the north-western outcrop and where the fixed caron is about 92.5% reflectivity in oil averages 3.81%. In the terrigenous rocks of the Coal

Measures the variation in reflectivity is regular and the isoreflectivity lines are more or less parallel to the isovol and the isocarb lines. Reflectivity in oil is progressively increased from east (R oil = 1.2%) to west (R oil = 2.8%).

For the purposes of study the organic matter in transmitted light, the organic matter has been separated from 116 samples representing the various lithotypes forming the carboniferous rock and distributed laterally and vertically to cover most of the coalfield. The main aim of this study is to determine the degree of carbonization of spores and pollen grains. As the variation in the colour of the spores and pollen grains is the most obvious optical phenomenon related to the process of carbonization, therefore the colour index was used in the present study to determine the degree of carbonization. The following numerical representation was used as carbonization index:

Yellow = 1, orange = 2, golden brown = 3,
chocolate brown = 4, dark brown = 5, and
black = 6.

The carbonization indices have been visually determined.

In the Coal Measures rocks, the carbonization index progressively increased from east (carbonization index = 2) to west (carbonization index = 6), and it was found that the isocarbonization lines are approximately parallel to the isovol and isocarb line of the associated coal seam (nine feet seam). In the case of the carbonate rocks the variation in the carbonization index was found to be very limited (2 and 3). In the anthracite area where the carbonization index of the Coal Measures rocks is 6 (spores and pollen grains are completely black), the carbonization index of the carbonate rocks in the same area is 3. This may be due to the fact that in the carbonate rocks spores and pollen grains have been sealed within the calcite cement which hinder the devolatilization of the gaseous elements and therefore the spores and pollen grains remained unmetamorphosed. This observation explains the effect of lithology on the susceptibility of the organic matter to metamorphism.

INCIPIENT METAMORPHIC ZONES OF SOUTH WALES COALFIELD

Using the previously described modifications in both organic and inorganic constituents of the carboniferous rocks, three zones of incipient metamorphism have been identified in the South Wales Coalfield, Fig. 9.3. A summary of each zone is given in Table 9.1.

Zone 1:

Rocks belonging to this zone occupy the south-eastern and the eastern part of the coalfield. The rocks, both the carbonates and the terrigenous, show no sign of any post-diagenetic modifications. The only fabrics preserved are those of diagenetic origin. The only modification observed in the clay minerals is the absence of discrete montmorillonite which has been attributed to be diagenetic. Therefore clay minerals present in this zone are illite, illite-montmorillonite, chlorite and kaolinite. The sharpness ratio of illite is approximately less than 3, which is characteristic of the unmetamorphosed rocks (Weaver, 1960 and Kubler, 1968). The vitrinite reflectivity in oil is generally less than 1.2% and the carbonization index of spores and pollen grains is less than 3 (of orange and yellow colour). Coals in this zone have fixed carbon less than 70%.

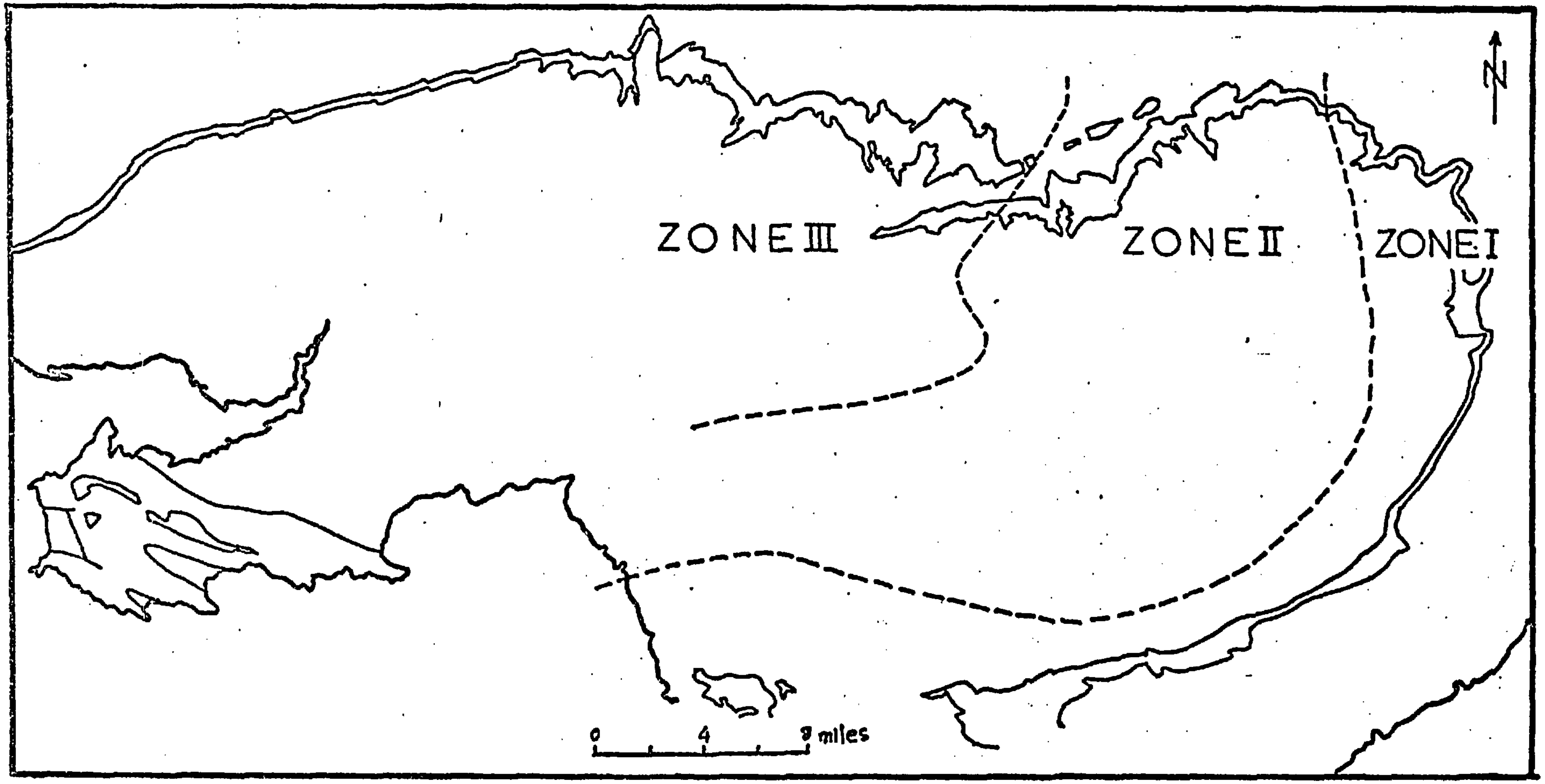


FIG. 9.3– Incipient metamorphic zones of the South Wales Coalfield

TABLE 9.1 CHARACTERISTICS OF THE ZONES OF INCIPIENT METAMORPHISM

ZONE	ROCK TYPE	ROCK FABRIC	CLAY MINERALS	Illite Sharpness Ratio	Vitrinite reflectivity in oil %	Carbonization Index	Fixed Carbon %
I Early Stage of Incipient Metamorphism	Carbonate rocks	No post-diagenetic fabric modification.	Illite, randomly interstratified illite-montmorillonite mixed layer, chlorite and kaolinite	2.5	1.8	3	70
	Terrigenous rocks	No post-diagenetic fabric modification.	Illite, randomly interstratified illite-montmorillonite mixed layer, chlorite and kaolinite	3.0	1.2	3	
II Middle Stage of Incipient Metamorphism	Carbonate rocks	Partial recrystallization of the micrite matrix and some of the oolites. Slight deformation of the sparry calcite represented by development of twin lamellae and by slight brush extinction.	The same mineral association as in Zone I. Decrease in the expandable layers in the mixed layer mineral.	2.5-3.0	2.01-2.74	3	70-87
	Terrigenous rocks	No post-diagenetic fabric modification	The same mineral association as in Zone I.	3.0-4.0	1.2-2.2	3-5	
III Late Stage of Incipient Metamorhpism	Carbonate rocks	Almost complete recrystallization of the micrite matrix and the foraminiferal tests and partial recrystallization of oolites and pelts. Degrading recrystallization of the monocrystalline crinoidal fragments and coarsely-crystalline fracture-filling calcite. Calcite and dolomite crystals are highly deformed and characterised by wide angle brush extinction, broad carved and fractured twin lamellae. Deformed fibrous calcite veinlets are common.	Appearance of dickite.	3.0-4.0	2.74-3.8	4	87-92.5
	Terrigenous rocks	In quartzwackes clays have been recrystallized to micaceous clays which commonly show signs of reaction with the quartz grains. Argillaceous rock fragments in litharenites are severely squeezed between the rigid framework grains and transferred to pseudomatrix. Deformed fibrous quartz veinlets are common.	Appearance of pyrophyllite and regular interstratified illite-montmorillonite.	4.0-6.0	2.2-2.8	6 (black)	

Zone II:

This zone occurs in the eastern central part of the coalfields. Carbonate rocks show slight modifications in their diagenetic fabrics. Micrite of the matrix of some biomicrites has been partially recrystallized to microcrystalline pseudospar calcite. Slight deformation fabrics have been developed, represented mainly by brush extinction in some calcite and dolomite crystals. No sign of post-diagenetic modification in the fabric of the terrigenous rocks has been observed. Clay minerals are represented by illite, illite-montmorillonite mixed layer, chlorite and kaolinite. Illite sharpness ratios range from 3 to 4 in the terrigenous rocks. The vitrinite reflectivity in oil ranges from 1.2% to 2.2% in the terrigenous rocks, and averages about 2.2% in the carbonate rocks. Carbonization index of spores and pollen grains ranges from 3 to 5 (of chocolate brown, golden brown and dark brown colour). Coals in this zone have fixed carbon ranging from 70% to 87%.

Zone III:

Rocks belonging to this zone occur in the western part of the coalfield. Both carbonate and terrigenous rocks are characterised by post-diagenetic fabric modification. In the carbonate rocks micrite matrix and micrite allochems (oolites and pellets) are almost completely recrystallized to pseudospar mosaic. On the other hand, monocrystalline skeletal debris (crinoidal fragments) and some of the coarsely crystalline fracture-filling calcite have been degradingly recrystallized to microcrystalline pseudospar mosaic. Deformational fabrics are clearly developed. Brush extinguished calcite and dolomite crystals, bent or crushed twin lamellae and deformed fibrous calcite veinlets are frequent. In the case of the terrigenous rocks particularly sandstones, post-diagenetic modifications are clearly displayed in the clay matrix and the argillaceous rock fragments. Argillaceous rock fragments, present in considerable amounts in the lithic arenites,have been completely deformed and transferred to pseudomatrix where the original fabrics have been entirely destroyed. On the other hand, clay minerals forming the matrix in the wackestones have been recrystallized and in many cases they started to react with the quartz grains and a hazey border between the matrix and the quartzitic framework grains has been developed.

New clay minerals have been formed in this zone, namely, pyrophyllite, dickite and regular mixed layer illite-montmorillonite. These minerals indicate high temperature and pressure conditions. Illite has a sharpness ratio of more than 4. Vitrinite reflectivity in oil is more than 2.2% in the terrigenous rocks, and spores and pollen grains are completely black (carbonization index = 6). Coals in this zone have more than 87% fixed carbon.

From the present study of the incipient metamorphism of the carboniferous rocks of the South Wales coalfield, the following remarks may be made:

(1) Incipient metamorphism affects both organic and inorganic constituents of the rocks. Metamorphism of the organic materials commonly modified their chemical composition and structure which consequently affects their optical characteristics (reflectivity and transparency). While the incipient metamorphism of the mineral matter has mainly modified their physical and physio-chemical properties, such modifications manifest themselves by changes in the fabrics and in the crystal structure.

(2) Organic and inorganic matter forming the rocks respond differently to incipient metamorphism. Organic matter is more sensitive to the effect of temperature and pressure.

(3) The susceptibility of the various minerals to incipient metamorphism varies. Carbonate and clay

minerals are more susceptible to metamorphism than siliceous minerals.

(4) The metamorphism of organic matter is dependent to some extent on the lithology of the host rock.

Comparison with Other Studies

Because the present work deals with the study of the incipient metamorphism of two main rock types, namely: carbonates and terrigenous rocks, and the modifications of both the organic and inorganic constituents have been studied, it was difficult to correlate the incipient metamorphic zones of the South Wales Carboniferous rocks with those described by many authors at various localities in the world. This is due to the fact that in most of the available published literature, the authors have usually dealt with one rock type which is commonly terrigenous rocks, and have used one indicator, clay minerals, reflectivity or degree of carbonization.

On the basis of illite crystallinity, zone I in the present work is correlated with the unmetamorphosed rocks of Weaver (1960) and Kubler (1968), while zones II and III are correlated with the anchiometamorphic zone of Kubler (1968) and the incipient to weak metamorphic zone of Weaver

(1960). With regard to the fabric modifications in terrigenous rocks and modifications in clay minerals, zones I, II and III of the present work could be correlated with the early stage, middle stage and late stage of catagenesis respectively, of the Russian geologists (Kossovskaya et al., 1957 and Koplan, 1971).

The carbonate rocks of Zone III in the present work could be correlated with the eometamorphosed carbonate rocks of the central Appalachians, U.S.A. (Brown, 1972). On the basis of the colour variation of spores and pollen grains, Staplin (1969) has recognised three main thermal zones in the Palaeozoic rocks of the British Columbia, Canada. These are zones of none or slight thermal alteration, zone of moderate thermal alteration and zone of strong and severe thermal alteration, which could be correlated with Zones I, II and III of the present study, respectively. Comparison of the zones of incipient metamorphism of South Wales Carboniferous rocks with those described by other authors is given in Table 9.2.

TABLE 9.2 COMPARISON OF ZONES OF INCIPIENT METAMORPHISM OR CATAGENESIS AS UNDERSTOOD BY VARIOUS AUTHORS (IN PART AFTER KOPLAN, 1971)

<table>
<tr><th>Kossovskaya A.G. & Shutov V.D. (1955)</th><th>Ruhkin L.B. (1956)</th><th>Logvinenko N.V. (1957)</th><th colspan="3">Kossovskaya A.G. Logvinenko N.V. & Shutov V.D. (1957)</th><th>Vassoyevich N.B. (1957, 1964)</th><th colspan="2">Koplan M.Y. (1971)</th><th>Kubler B. (1966)</th><th>Present Work</th></tr>
<tr><td></td><td></td><td></td><td colspan="3"></td><td></td><td colspan="2">DIAGENESIS</td><td>DIAGENESIS</td><td>DIAGENESIS</td></tr>
<tr><td rowspan="3">Zone of Clay Matrix</td><td>Zone of weak Epigenesis</td><td rowspan="4">Zone of Normal Epigenesis</td><td rowspan="5">Epigenesis</td><td rowspan="3" colspan="2">Initial Epigenesis</td><td rowspan="3">Early Catagenesis or Proto Catagenesis</td><td rowspan="12">CATAGENESIS</td><td rowspan="4">Early Stage of Catagenesis</td><td rowspan="12">ANCHIMETAMORPHISM</td><td rowspan="2">I
Early Stage of Incipient Metamorphism</td></tr>
<tr><td rowspan="2">Zone of moderate Epigenesis</td></tr>
<tr><td rowspan="6">II
Middle Stage of Incipient Metamorphism</td></tr>
<tr><td>Zone of Chlorite-Silica Matrix</td><td rowspan="8">Zone of Strong Epigenesis</td><td rowspan="2" colspan="2">Deep Seated Epigenesis</td><td rowspan="7">Middle Catagenesis or Meso Catagenesis</td></tr>
<tr><td rowspan="2">Zone of regenerated Quartz Matrix</td><td rowspan="3">Zone of progressive Epigenesis</td><td rowspan="6">Middle Stage of Catagenesis</td></tr>
<tr><td rowspan="6">Metagenesis</td><td rowspan="2" colspan="2">Early Metagenesis</td></tr>
<tr><td rowspan="3">Zone of regenerated Quartz and Micaceous Matrix</td></tr>
<tr><td rowspan="6">Zone of Initial Metamorphism</td><td rowspan="6">Green Schist Facies</td><td rowspan="4">Late Metagenesis</td></tr>
<tr><td rowspan="4">III
Late Stage of Incipient Metamorphism</td></tr>
<tr><td rowspan="4">?</td></tr>
<tr><td rowspan="2">Late Catagenesis or Apo Catagenesis
?</td><td rowspan="2">Late Stage of Catagenesis</td></tr>
<tr><td rowspan="2">Zone of very Strong Epigenesis or Initial Metamorphism</td><td rowspan="2">Regional Metamorphism</td><td>Muscovite-Chlorite Zone</td></tr>
<tr><td>Biotite Zone</td><td>Early Metagenesis
Proto Metagenesis</td><td colspan="2">METAMORPHISM</td><td>METAMORHPISM</td><td>METAMORPHISM</td></tr>
</table>

CAUSES OF COALIFICATION AND INCIPIENT METAMORPHISM

From the present study it has been concluded that the metamorphism of the Carboniferous rocks follows the same pattern as the coal seam metamorphism or carbonization. Therefore the causes of the coal metamorphism could generally be considered to be the causes of the incipient metamorphism of the associated rocks. Metamorphism or carbonization of coal is indicated by its progressive increase in the fixed carbon % and in the volatile constituents.

From a physio-chemical point of view, the coalification process can be subdivided into three stages (Huck and Karweil, 1962). In the beginning of the coalification process complicated chemical reactions are caused by fungi and bacteria. In the second stage (the formation from peat to highly volatile bituminous coals) the bacterial and chemical changes decrease in favour of a mechanical compression of sediments which causes expulsion of water. The last stage (the formation from highly volatile bituminous coal to anthracite) is a combination of physical and chemical changes. The chemical metamorphism of coal is characterised by an increasing aromatization of humic substances. During this process molecular groups of non-aromatic character rich in hydrogen and oxygen are released. At the same time the remaining molecules of aromatic character are growing together (M. and R. Teichmüller, 1954). Among the factors causing carbonization reported in literature are temperature, pressure and time. Very convincing are the results of experiments on artificial coalification undertaken by Huck and Karweil (1962) and by Huck and Patteisky (1964, and reported in M. and R. Teichmüller, 1966). They show that under vacuum conditions at a temperature of 350 °C a highly volatile bituminous coal increased in rank from 67 to 82%. By contrast, when a pressure of 8000 atm. was applied at 350 °C, the fixed carbon remained unchanged.

An example of slight temperature exposure in nature during a considerable time interval is the Lower Carboniferous brown coal in the basin of Moscow. This deposit has never been buried deeply and it has been exposed to low temperature only. Examples of high temperature exposure in nature are the severe carbonization zones near small or large igneous bodies. Examples are the pliocene brown coals of Palembang, Sumatra, Indonesia (Mukherjee, 1955 in Gutjahr, 1966) which by contact metamorphism are partly changed into anthracite, and the organic metamorphic zones around the Bramsche massif, German Federal Republic (M. and R. Teichmüller, 1958). These and other studies suggest that temperature is the predominant cause of carbonization.

With respect to pressure, White (1925) emphasised the importance of the "dynamic thrust" as the cause of coal metamorphism. However, Lesley (1879) had earlier proposed that the regional change in the rank of coal in Pennsylvania was due to the higher coal rank having been buried more deeply. He pointed out that the difference in the amount of the original overburden could easily explain why disturbed Belgian coals are not anthracite and why Arkansas anthracites are not disturbed. M. and R. Teichmüller (1954) mentioned a brown coal in a folded belt in Pakistan which was greatly deformed by dynamic thrusting but was only slightly altered. The western part of the Prokop'evsk-Kissel area (Kuznetsk Basin, Russia) is characterised by steeply folded and faulted coal measures (Ammosov, 1961). These coals are less coalified than the coals in the less disturbed parts of the basin.

Hoffman (1936) has subjected various coal samples for four days to a pressure of 18,000 kg. per square centimeter. He noted an average decrease in volatile matter of 1.5%, indicating very slight carbonization. According to Hoffman (1936) and Bergiues (1913) coalification by pressure is successful only if accompanied by relatively high temperature. This was also observed by Gropp and Bode (1932) who noticed a decrease of volatile matter of immature coals when subjected to 300 °C and 1800 atm. The above examples show that pressure alone apparently does not increase the rank of the coal.

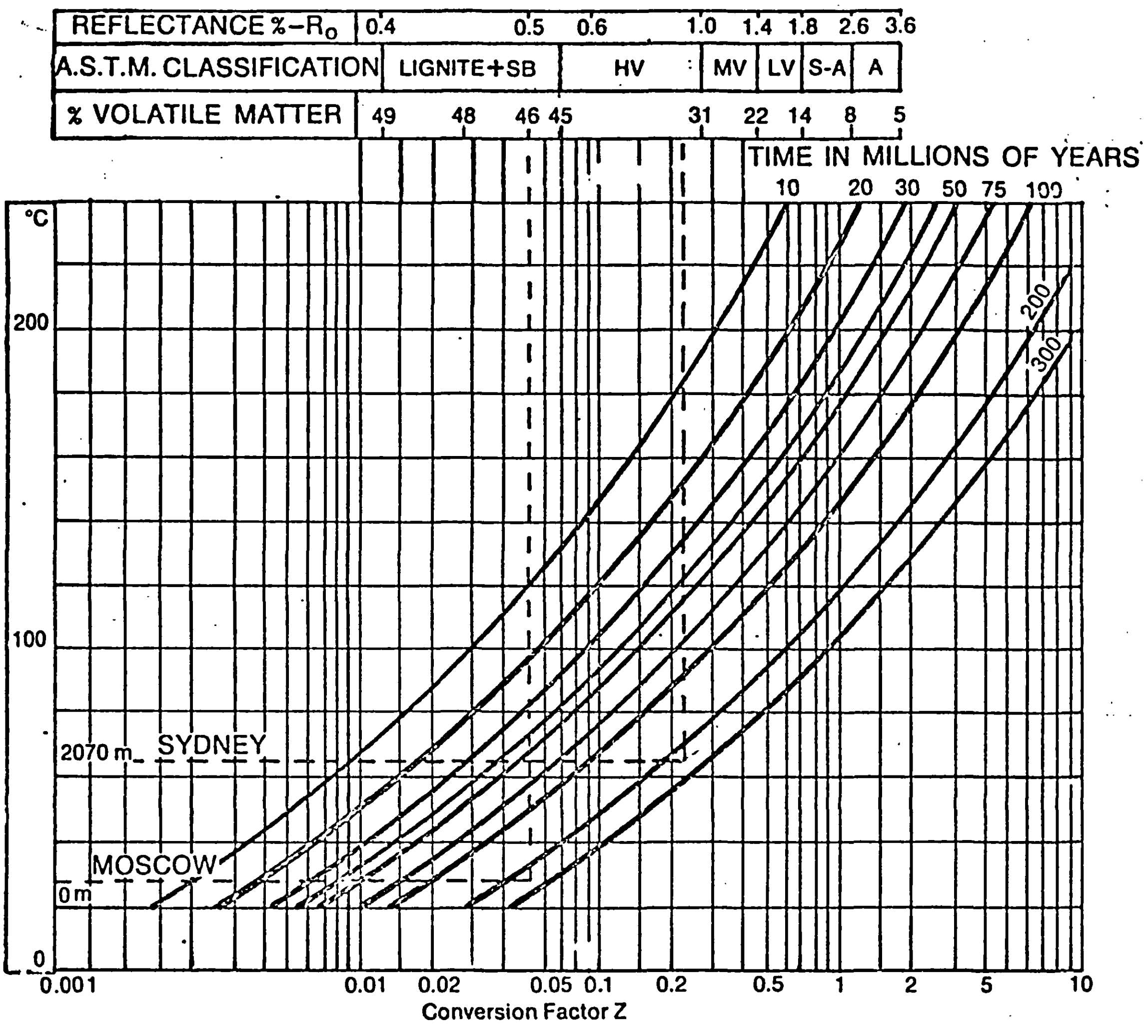


FIG. 9.4 RELATIONSHIP BETWEEN RANK, TEMPERATURE AND TIME OF COALIFICATION (AFTER KARWEIL, 1956).

The effect of exposure time on the carbonization of organic matter has been discussed by many authors. Gutjahr (1966) has concluded from his experimental study of the effect of time and temperature on the degree of carbonization of Recent pollen grains and spores that the effect of time at low temperature is not important, while time at higher temperatures is of importance. One of the best examples in nature is the effect of exposure time on Lower Carboniferous brown coal of Moscow. Apparently time alone could not carbonize this carbonaceous material beyond the brown coal rank (M. and R. Teichmüller, 1954).

Karweil (1956) was able to show a correlation between coal rank, rock temperature and duration of heating. This correlation is illustrated in Fig. 9.4, where the rank is plotted on the abscissa as factor Z (which has been converted into appropriate rank parameters at the top of the diagram following Gutjahr, 1966), the rock temperature is shown on the ordinate in °C, and the duration of heating is portrayed by the successive curves in millions of years. The diagram shows that two coals subjected to the same temperature for different lengths of time will attain a different rank, e.g. at 100°C for 10 m.y. the rank will be lignite, but after 100 m.y. it will be a medium volatile bituminous coal.

There is a two-fold systematic variation in the development of the different kinds of coal in South Wales that shows features of unusual interest. On the one hand, in accordance with Hilt's Law*, the lower seams in the sequence at any one locality tend to be of higher rank than the higher seams. On the other hand, any one coal seam tends to become progressively more anthracitic as it is followed towards the north, north-west and west: thus the bituminous coals are mainly found along the southern and the eastern outcrops, the steam coals in the central part of the coalfield between the Taff and the Neath (particularly in the Rhondda) valleys, and the anthracites along the north crop westwards from the Neath valley, especially in the Gwendraeth valley and in Pembrokeshire.

The causes of the regional changes in the rank of South Wales coals are not certainly known. The effect of time is not obviously important, especially in relation to the lateral variations which exist in the same coal seams. Trotter (1948) argued that devolatilization and the corresponding increase of rank from gas coals to anthracite were products of shearing stress with incidental friction heat created by a major thrust that comes to outcrop as the [illegible]

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* Hilt's Law states that: "In coal seams there is a loss of volatile content with increasing depth". (Hilt, 1873)

Cennen disturbance on the north-west crop and that supposedly sinks to depths southwards underneath the coalfield (Fig. 9.5). He further suggested that the rank of a coal seam is then a function of its distance from the thrust plane. In contrast, Jones (1949) and Wellman (1950) have invoked a process of burial metamorphism that is an aspect of Hilt's Law, the weight of overlying sediment being the initial load, and rank then being a function of pressure and temperature at depth. Such an explanation, applied to the present anthracite area, implies that where the relatively thin representatives of the Lower and Middle Coal Measures of the Gwendraeth belt now lie there was formerly a very thick overburden - estimated at about 18,000 ft. (Jones, 1949). It also implies that the depositional basin carried its thickest accumulation of sediments in the region of the Gower and Llanelli synclines, where semi-anthracites occur as high in the sequence as the seams of the Swansea beds.

Mohafez (1966) studied the rank variation in the British Coals. He concluded that there is a broad and roughly quantitative relationship between the intensity of the deformation and the distribution of the fixed carbon in coals, with the exception of the Western area of Ireland and the South Wales Coalfield. He further assumed that the apparent reverse in the general trend of increasing carbon ratios in the north-

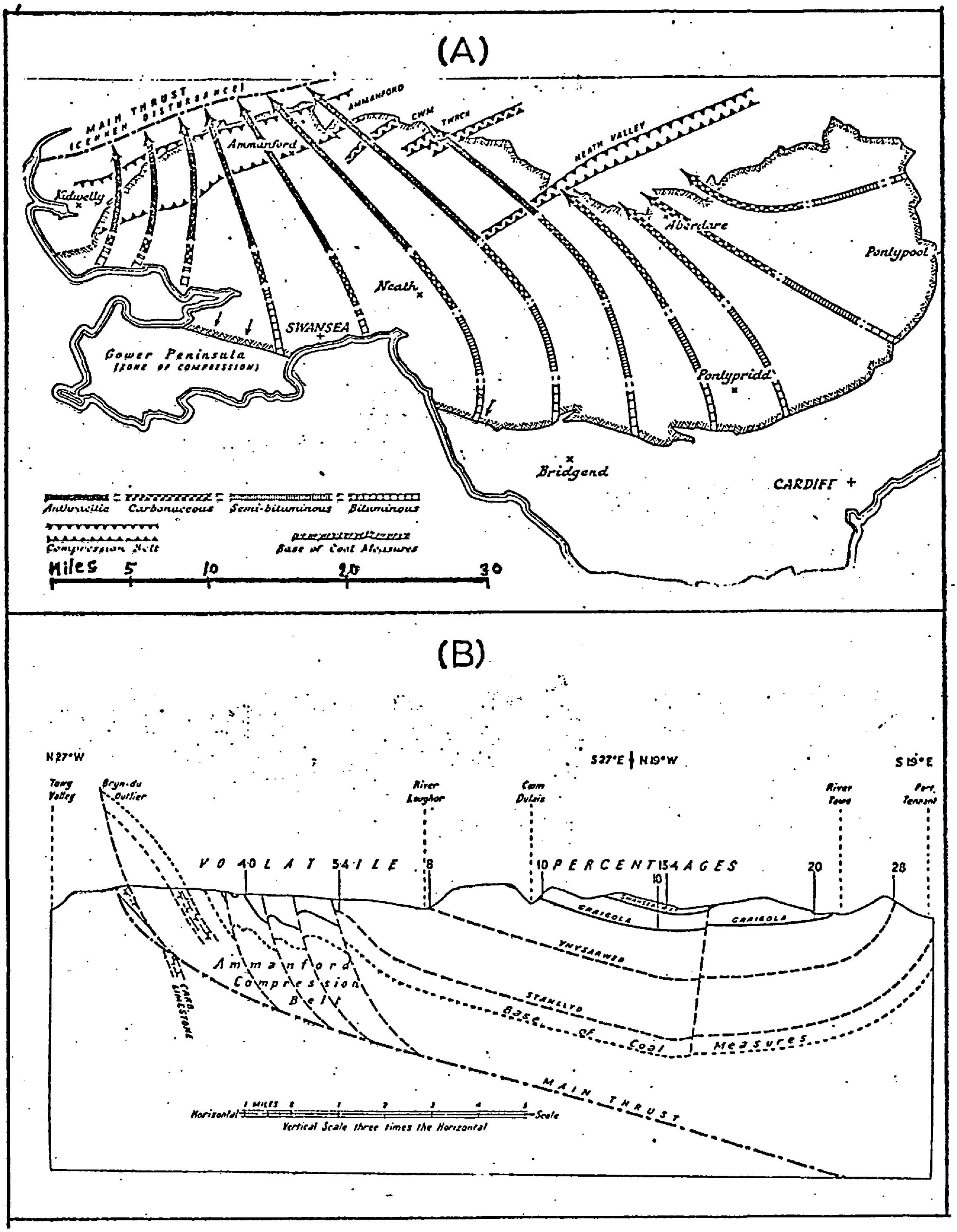

FIG. 9.5 - A map (A) and cross section (B) of the South Wales Coalfield, showing the relationship between the main thrust and devolatilization (after Trotter, 1948).

western area of the South Wales Coalfield is related to the reflection of the lateral pressure which was transmitted through the basement and had mostly affected the edge of St. George's Massif. With regard to Hilt's Law, Mohafez has demonstrated cases which show evidence contrary to Hilt's Law in the South Wales Coalfield.

With regard to the Carboniferous rocks, it was found that the trend of their incipient metamorphism is approximately parallel to that of the coal metamorphism. Therefore it is assumed that the causes of coal metamorphism are responsible for the rock metamorphism. On the basis of the experimental work of Chandra (1965), Ghosh (1968) and Goodarzi and Murchison (1972) on the effect of heat on vitrinite reflectivity, and Standnichenko (1931) on the effect of heat on the carbonization of spores and pollen grains, it has been concluded that the carboniferous rocks of South Wales in the anthracite area could have been subjected to a temperature of about 400 °C. This heat could be transmitted from unknown source, lies at depth beneath the carboniferous rocks to the north-west of the South Wales Coalfield. The frictional heat created during thrusting of the carboniferous rocks on the Careg Cennen fault (Trotter, 1948) could be one of the possibilities. On the other hand, deformational fabrics displayed by the carboniferous limestones outcropping at the north-western side of the coalfield could be attributed to the effect of tectonism. The western part of the coalfield is more tectonised than the eastern part (Neath disturbance, Tawe disturbance and Amman disturbance).

APPLICATION OF INCIPIENT METAMORPHISM ON HYDROCARBON EXPLORATION

The potential for oil and gas of a region depends on the presence of suitable organic substances that have been preserved under a low degree of metamorphism. This low-grade incipient metamorphism has been named eometamorphism by Lands (1967). It has a pronounced effect on organic matter, and for that reason has been referred to as organic metamorphism (Gutjahr 1966, Staplin 1969).

As early as 1865 Rogers was able to demonstrate that in the Appalachian basin there exists a correlation between coal composition, oil and gas occurrence, and the degree of metamorphism of the associated strata. This observation was later elaborated upon by David White, who in 1915 introduced his "carbon-ratio" theory. This theory uses the fixed carbon content of coal as an indicator of the character of the hydrocarbons in nearby reservoirs which becomes lighter and more volatile as the rank of coal increases. Thus, where the carbon content is high there will be gas or oil of high grade, and where it is low the oil will be heavy. However, there is a limit to the amount of metamorphism that can take place and still permit oil or gas to remain in the sediments so affected. This limit, which has been referred to as the Hydrocarbon "dead line" lies at about 65 to 70% fixed carbon (White 1920, Fuller 1919 and Ammosov 1961). The relationships between coal rank and the type and occurrence of oil and gas are summarised in Table 9.3.

Mohafez (1966) studied the relationship between the coal metamorphism and the occurrence of hydrocarbons in Great Britain. He plotted on the regional isocarb map the localities of British oil and gas fields. This map shows that commercial oil and gas fields which are concentrated in the eastern areas of the Midlands, Nottinghamshire, most apparently occur below the 60% isocarb line (ranging from 55% to 63%). Areas above 65% isocarb line are practically barren of commercial oil and gas. And areas with a high percentage of fixed carbon (anthracite) are entirely barren of oil and gas, Fig. 9.6. These observations show that the carbon ratio theory applies to Great Britain.

Other physical and chemical properties of organic matter have been used as rank parameters and therefore as indicators for the occurrence of oil and gas. Among these properties is the vitrinite reflectance values, which have been plotted in relation to the fixed carbon percentages according to the correlation established by Kötter (1960). The oil "dead line" at 65% fixed carbon corresponds to a reflectance of 0.8%, the most promising interval of organic metamorphism for the occurrence of oil and gas lies between

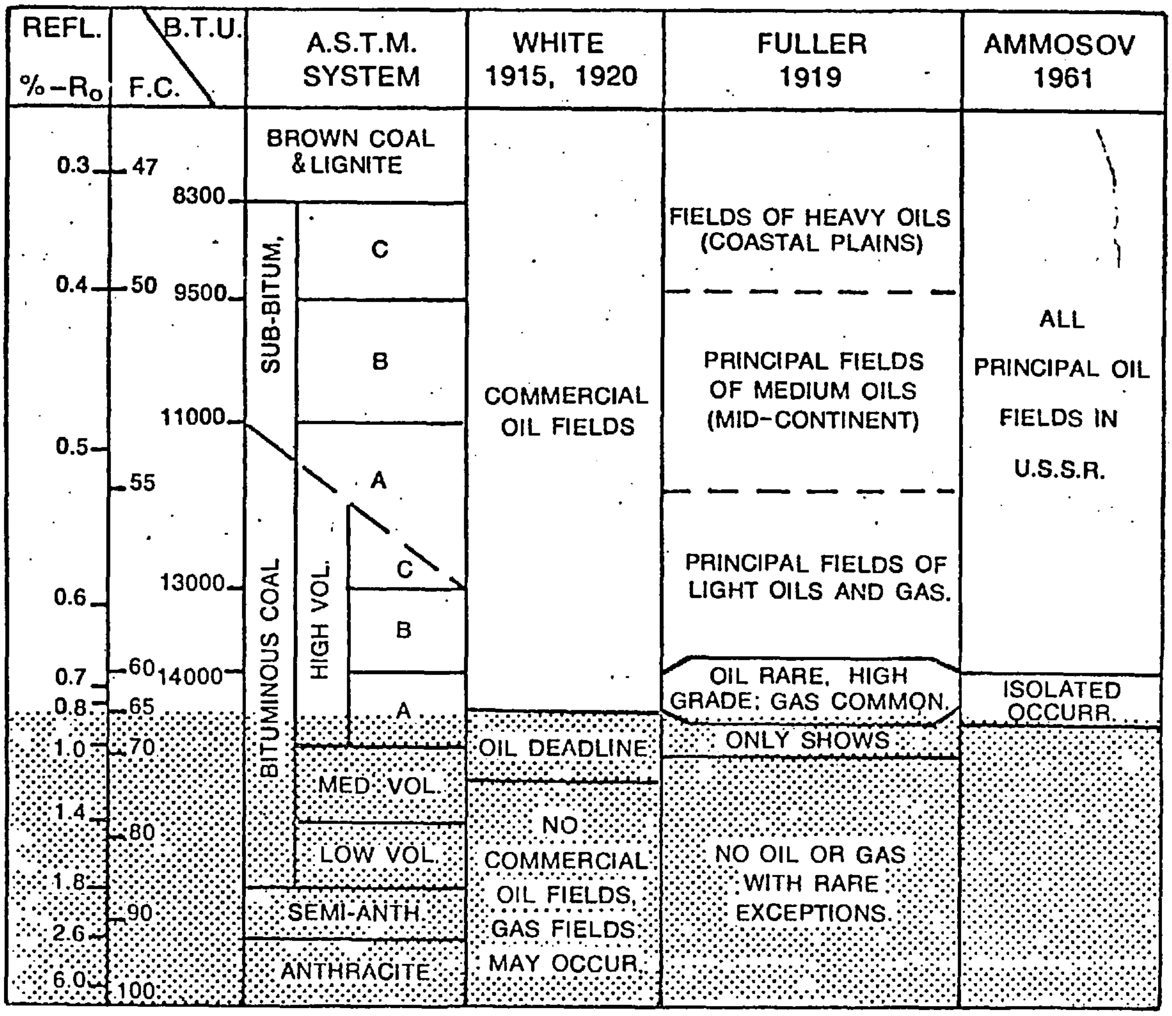

TABLE 9.3 RANK OF COAL AND OCCURRENCE OF OIL AND GAS FIELDS (AFTER HACQUEBARD AND DONALDSON, 1970).

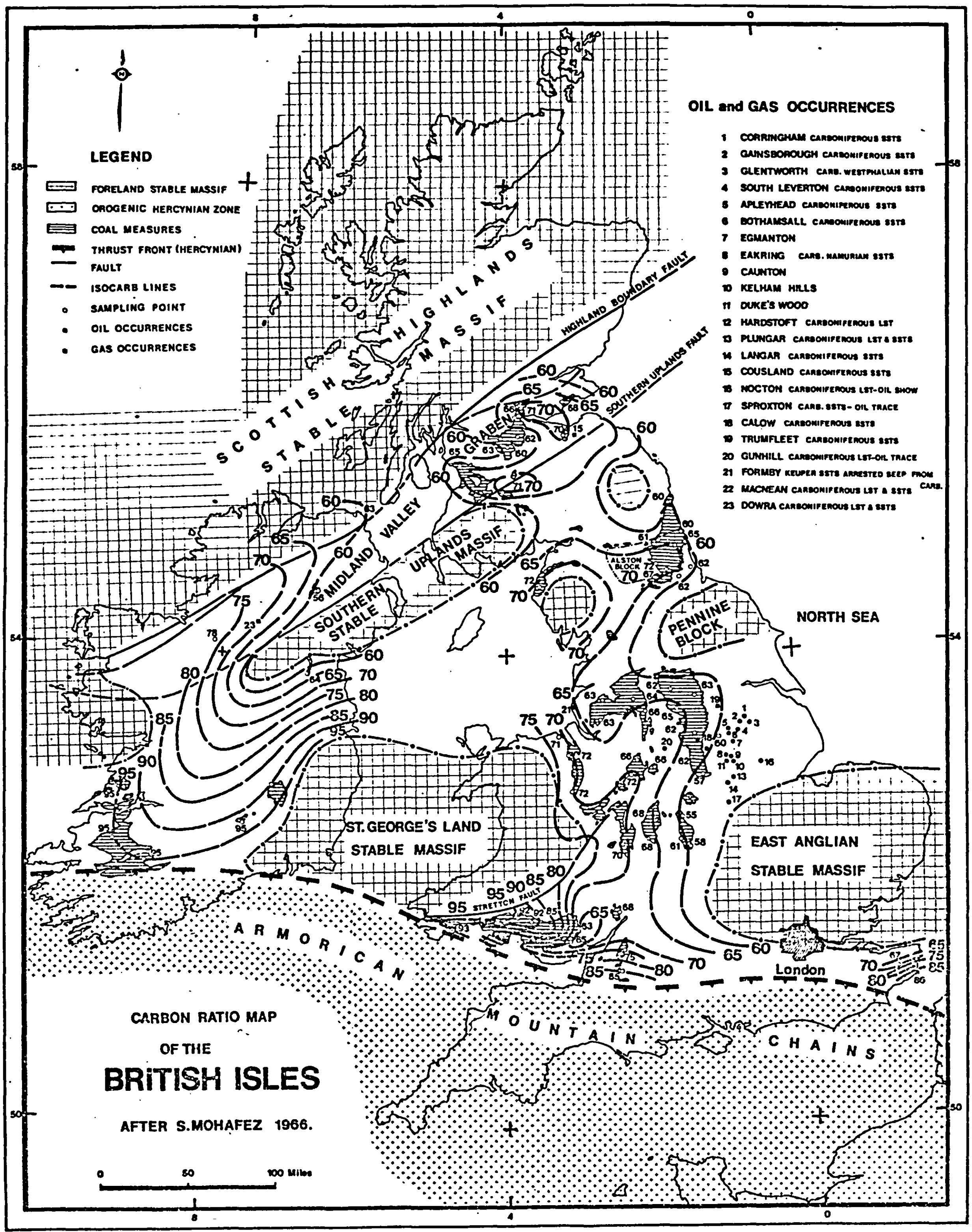
LEGEND
FORELAND STABLE MASSIF
OROGENIC HERCYNIAN ZONE
COAL MEASURES
THRUST FRONT (HERCYNIAN)
FAULT
ISOCARB LINES
SAMPLING POINT
OIL OCCURRENCES
GAS OCCURRENCES
OIL and GAS OCCURRENCES
1 CORRINGHAM CARBONIFEROUS SSTS
2 GAINSBOROUGH CARBONIFEROUS SSTS
3 GLENTWORTH CARB. WESTPHALIAN SSTS
4 SOUTH LEVERTON CARBONIFEROUS SSTS
5 APLEYHEAD CARBONIFEROUS SSTS
6 BOTHAMSALL CARBONIFEROUS SSTS
7 EGMANTON
8 EAKRING CARB. NAMURIAN SSTS
9 CAUNTON
10 KELHAM HILLS
11 DUKE'S WOOD
12 HARDSTOFT CARBONIFEROUS LST
13 PLUNGAR CARBONIFEROUS LST & SSTS
14 LANGAR CARBONIFEROUS SSTS
15 COUSLAND CARBONIFEROUS SSTS
16 NOCTON CARBONIFEROUS LST-OIL SHOW
17 SPROXTON CARB. SSTS- OIL TRACE
18 CALOW CARBONIFEROUS SSTS
19 TRUMFLEET CARBONIFEROUS SSTS
20 GUNHILL CARBONIFEROUS LST-OIL TRACE
21 FORMBY KEUPER SSTS ARRESTED SEEP FROM CARB.
22 MACNEAN CARBONIFEROUS LST & SSTS
23 DOWRA CARBONIFEROUS LST & SSTS
SCOTTISH HIGHLANDS STABLE MASSIF
HIGHLAND BOUNDARY FAULT
SOUTHERN UPLANDS FAULT
GRABEN
MIDLAND VALLEY
SOUTHERN UPLANDS MASSIF
SOUTHERN STABLE
ALSTON BLOCK
PENNINE BLOCK
NORTH SEA
ST. GEORGE'S LAND STABLE MASSIF
STRETTON FAULT
EAST ANGLIAN STABLE MASSIF
London
ARMORICAN MOUNTAIN CHAINS
CARBON RATIO MAP OF THE
BRITISH ISLES
AFTER S.MOHAFEZ 1966.
0 50 100 Miles
8
4
0
58
54
50

FIG.9.6

47 and 60% fixed carbon, or between 0.3 and 0.7% reflectance. The great value of using the reflectance of vitrinite was first pointed out by M. and R. Teichmüller (1958), who successfully employed this technique for oil and gas exploration in Lower Saxony, West Germany. Hacquebard and Donaldson (1970) have used the same technique for the determination of the approximate vertical position of the oil "dead line" in the Upper Paleozoic of the Atlantic provinces, Canada.

Gutjahr (1966) suggested that the degree of carbonization of pollen grains and spores in sedimentary rocks devoid of coals can be used instead of fixed carbon values to determine areas of equal organic metamorphic intensity and to provide a clue to the hydrocarbon potential of these areas. Correia (1969) and Staplin (1969) in their studies on the diagenesis and metamorphism of the organic matter disseminated in sedimentary rocks, have confirmed the relationship between the degree of carbonization of spores and pollen grains and the occurrende of oil and gas. Table 9.4 summarizes the relationships between the type of organic matter, degree of alteration and expected hydrocarbons as concluded by Staplin (1969).

From the geochemical point of view, Hunt and Jamison (1956) and Hunt (1961) in their study on the indigenous organic

TYPE OF ORGANIC MATTER, MATURED FACIES		ASSOCIATED HYDROCARBONS
a.	Sapropelic, amorphous	wet gas and oil
b.	Plant cuticle, charcoal	Dry gas
c.	Mixture of a and b	Wet gas and oil
ORGANIC METAMORPHISM		
a.	Blackening, increase in index of refraction	Wet or dry, depending on thermal alteration index
b.	Pyrobitumen	
c.	Graphite, mineralization	

THERMAL-ALTERATION INDEX		ORGANIC MATTER	ASSOCIATED HYDROCARBONS
1.	None	fresh, yellow	Wet or dry
2.	Slight	brownish yellow	Wet or dry
3.	Moderate	brown	Wet or dry
4.	Strong	black	Dry gas
5.	Severe	black, with additional evidence of rock metamorphism	Dry gas to barren

TABLE 9.4 RELATIONSHIP BETWEEN TYPE OF ORGANIC MATTER, DEGREE OF ALTERATION AND EXPECTED HYDROCARBONS (AFTER STAPLIN, 1969).

matter in sedimentary rocks, have concluded that no correlation was noted between the age or depth of burial of a sedimentary rock and the distribution of its various types of organic matter. On the other hand, they noticed that extreme metamorphism alters the organic material of a rock. They found that phyllite contains very little hydrocarbon or asphalt compared with shales. Baker and Claypool (1970) in their study of the effects of incipient metamorphism on organic matter in mud rock have concluded that metamorphism results in net loss of hydrocarbon. They found also that metamorphosed carbonaceous mudrock has a significantly high proportion of saturated, compared to aromatic, hydrocarbon than unmetamorphosed mudrock. This difference suggests that the net loss of aromatic hydrocarbon during incipient metamorphism is greater than that of saturated hydrocarbon.

Bordenave et al. (1966) and Giraud (1970) have studied the geochemical evolution of kerogen during the diagenesis and incipient metamorphism of the sedimentary rocks and they have used the degree of carbonization of spores and pollen grains to determine the level of incipient metamorphism in the studied rocks. They concluded that progressive increase in incipient metamorphism (as indicated by the degree of carbonization) followed by decrease in the ratio of degraded carbon to total carbon before pyrolysis (degradation factor, T_D). On the other hand, they found that the potentials in gas, oil and aromatic products vary regularly as a function of the degradation factor.

From the foregoing review, one can notice the close relationship between incipient metamorphism and the occurrence of hydrocarbons and therefore it emphasises the importance of the determination of the level of incipient metamorphism.

Based on the fixed carbon % of the coal seams of South Wales and on the modification in the physical and optical properties of the organic and inorganic constituents of the various lithotypes of the Carboniferous rocks, three zones of incipient metamorphism have been recognised. From this zonation the deadline for the occurrence of oil can be determined independently of the fixed carbon %. Table 9.5 shows the relationship between incipient metamorphic zones and the occurrence of hysrocarbons. The close relationship existing between the fixed carbon % of coals and the incipient modifications in the rocks shows that these rock modifications could be used for oil exploration instead of the fixed carbon % in the basins devoid of coals.

TABLE 9.5 RELATIONSHIP BETWEEN INCIPIENT METAMORPHISM OF ROCKS AND HYDROCARBON OCCURRENCE

ZONATION OF DIAGENESIS AND INCIPIENT METAMORPHISM IN PRESENT WORK					Fixed Carbon % of Coal	Staplin F.I. (1969)	Wassojewitch (1969, in Alpern 1970)	White D. (1915, 1920)
		Illite Sharpness Ratio	Carboniz- ation Index	Vitrinite Reflectivity in oil %				
	DIAGENESIS					Wet or dry gas	early diagenetic methane	Commercial oil fields
				0.5	50			
--?--	--------?-------							
INCIPIENT METAMORPHISM	Zone I Early Stage						Oil	
		3	3	1.2	70			Oil dead line
	Zone II Middle Stage						Wet gas	No commercial oil fields
		4	6	2.2	87			
	Zone III Late Stage		Black			Dry Gas	Methane	Gas fields may occur
		6	6	2.8	92.5	Dry Gas or barren		

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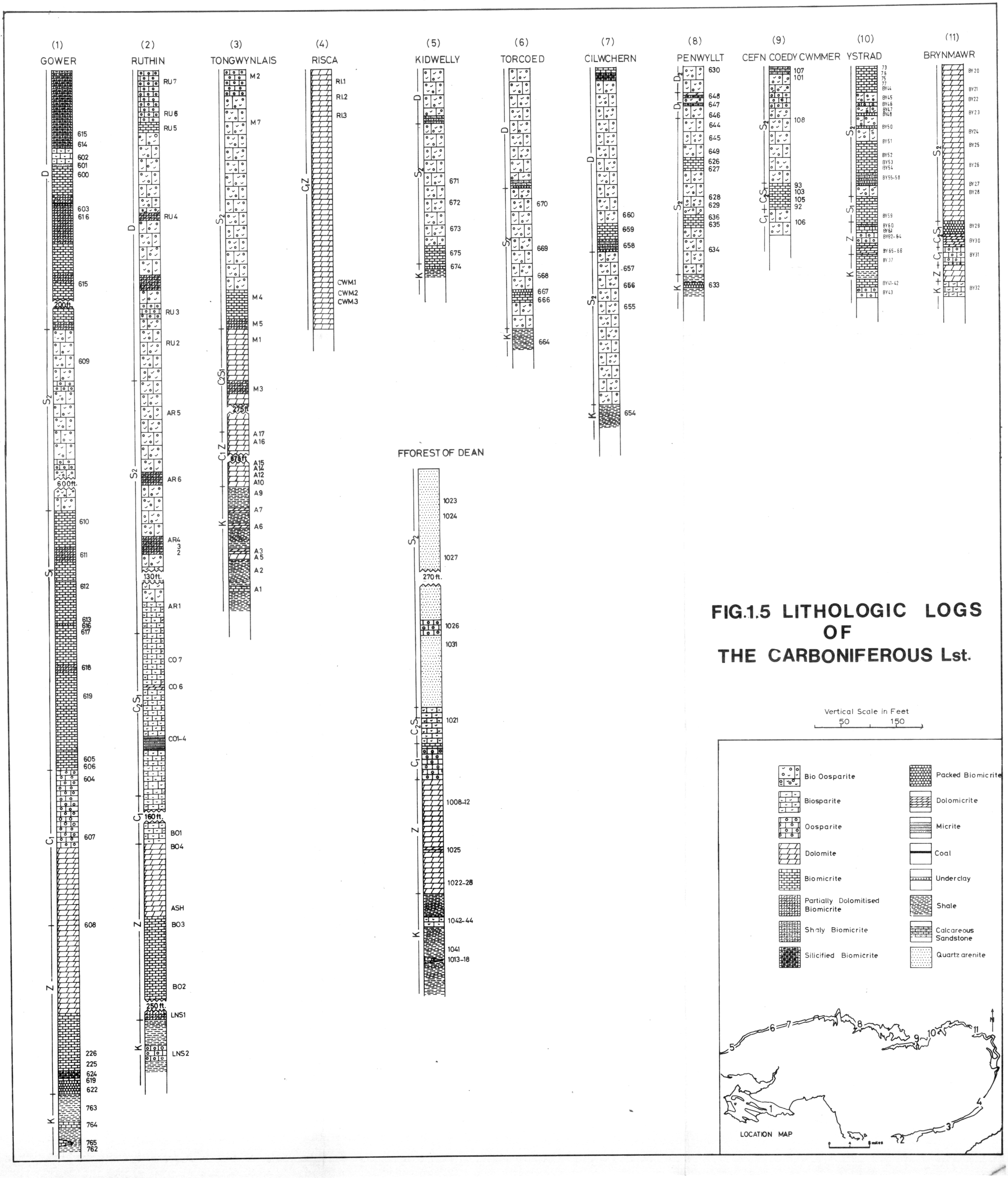

FIG.1.5 LITHOLOGIC LOGS OF THE CARBONIFEROUS Lst.

FIG.1.6 LITHOLOGIC LOGS OF THE CAOL MEASURES