

# ON THE MINERALOGICAL AND CHEMICAL EVOLUTION OF STONY METEORITES

by

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## SUMMARY

Reviewing the latest results of investigations on chondritic meteorites, the author reasserts the view that *in situ* reduction is an important factor in the evolutionary process. The most rudimentary form of matter of asteroid origin is represented by type I. carbonaceous chondrites. All stony meteorites that have fallen upon the Earth are in various stages of a gradual process of reduction starting from the carbonaceous. As regards the interpretation of the phenomena, the author accepts the necessity of assuming an "internal atmosphere" which gives a satisfactory explanation for the polymerization of hydrocarbons, the formation of hydrosilicates and the problems of the degrees of crystallinity and the formation of metallic iron. However, the rates of change of these processes of evolution are far from constant, the intensities vary in time; one product of a sudden intense reduction is the enstatite-chondrite, of peculiar mineralogical and chemical composition. In a novel evolutionary diagram presented here, there are two principal lines of evolution, as before, but the two lines, rather than being parallel, tend to imply the formation of the meteorites out of a uniform primordial mass.

The numerous papers published in the last few decades on meteorites mostly contain analytical results. Research involves in our days quite a wide variety of methods and instruments, which have furnished numerous novel data, some of them rather unexpected, and have inevitably led to the recognition of certain chemical, physical and textural similarities, "kinships", and as a result to the distinguishing of several new groups and categories.

The fundamental classification has not, of course, changed: the groups of stony iron- and iron-meteorites (aerolites, siderolites and siderites) are still valid, but it is generally recognized that stony meteorites are the most numerous of all and that, within this latter group, that of the chondrites is most populous. Some 86 per cent of all cosmic bodies fallen upon the Earth are chondrites. This is why a great majority of research is aimed at this group or at the meteorites belonging to this group. At the same time this implies that extraterrestrial space is favourable to the formation of chondrites, and this is why attention tends to be focused upon the chondrites at times when new results raise problems of genetic or rather evolutionary nature.

### Common chondrites

The outlines of the chemical nature of the cosmic material known as chondrites have been drawn by *Prior* (1916). According to him, the bulk chemical composition of chondrites is approximately uniform, which means that the amount of nickel-iron and the Fe content of Fe-Mg minerals are presumably in a contravariant relation to each other. In an up-to-date formulation, this signifies that any differences between the chondrites of nearly identical chemical composition are largely due to deviations in the state of oxidation of iron. In 1953, *Urey* and *Craig* examined the validity of this relationship in a comprehensive paper, thus once more focussing attention on this problem; on the other hand, the authors in question made some tentative cosmochemical suggestions, which gave a renewed impetus to speculation as to the origin of the chondrites. The above-mentioned paper is essentially based on a review according to certain principles of the 350 or so chemical analyses then available; on 94 selected, "superior" analyses, considered as reliable or corrected by computation, it establishes the proportions of metallic (+sulfidic) vs. oxidized iron in the chondrites. The distribution between the two

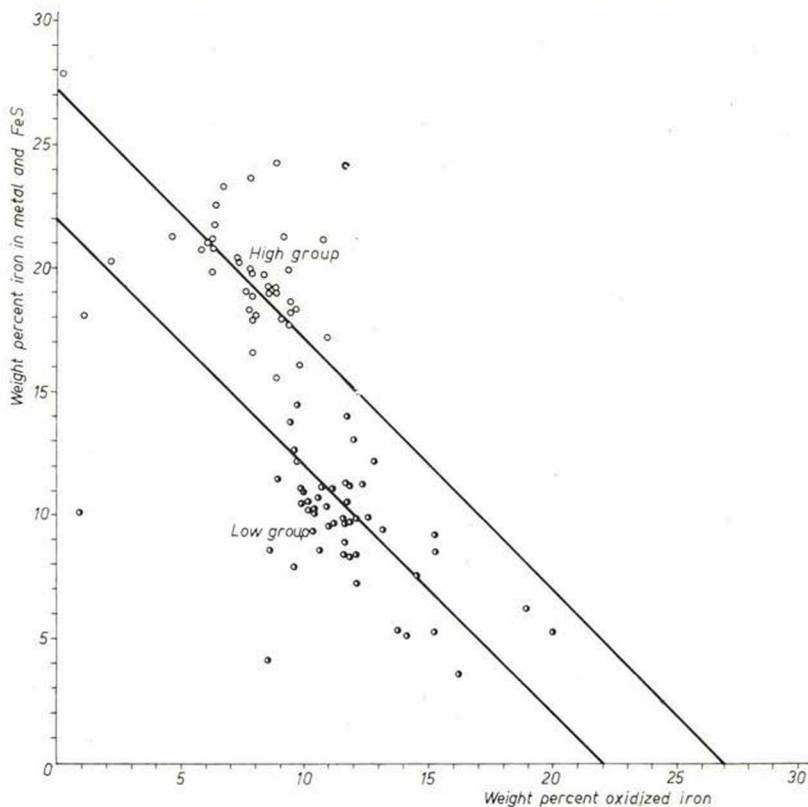


Fig. 1. Relationship between oxidized iron and iron as metal and sulfide in 94 selected analyses of chondritic meteorites (*H. C. Urey* and *H. Craig*).

above-named groups of "total Fe" given in weight percentages turned out to be of two kinds, so that a "High" and "Low" group of chondrites could be distinguished on the basis of the iron content. As an average, 28.6 percent of iron was found for the *H* type and 22.3 percent for the *L* type. From this, Urey and Craig drew two essential conclusions; that the two groups imply two distinct modes of origin ("two parent substances") for the chondrites; and further that since the selected analyses gave two distinct "total iron levels", the general validity of Prior's rule is discutable and the rule itself is due to the inadequacies of early analyses. (Fig. 1.)

As it was revealed by an earlier critical evaluation (Sztróka et al., 1961.), the individual points of the diagram are rather intensely scattered, so that there is, as a matter of fact, no sharp boundary between the two groups, *H* and *L*. A further remarkable circumstance is that the straight line of the point sets is a line of approximately 45° slope, none other than the line of the arithmetical mean, necessarily due to a correlation between the values of the iron content.

The statements based on the selected analyses have intrigued a number of workers; e. g. Ringwood (1961) and Mason (1962) made attempts at eliminating the scatter and interpenetration of the two fields partly by omitting analyses performed on unfresh, obviously altered, oxidized ancient finds, and partly by applying the criterion that the  $\text{FeO}/\text{FeO} + \text{MgO}$  ratio in olivine and in the pyroxenes has to agree approximately with the ratio derived from the bulk analysis of the chondrites. Not only did these measures eliminate the interpenetration, but they established five distinct fields (enstatite, olivine-bronzite, olivine-hypersthene, olivine-pigeonite and carbonaceous chondrites). This refining of criteria further entailed a transfer of all chondrites containing iron-rich silicates (hypersthene) into the *L*-group, whereas the olivine-bronzite chondrites clustered in the *H*-group. It was remarkable, however, that the carbonaceous chondrites, left out of consideration by Urey and Craig, fell between the two fields, along a horizontal coordinate line which indicates a content of purely oxidized iron, whereas the enstatite chondrites of extreme metallic iron content formed, beyond a hiatus of some breadth, a band drawn out along the ordinate (Fig. 2.). Not only does this mean that a single set of criteria applied to the chemical constituents and to the total iron content produces a fairish number of distinct groups, but that the original diagram has itself undergone substantial changes. The two parallel lines came substantially closer to each other; between them, there cropped up the group, disregarded until then, of carbonaceous chondrites, which failed to conform to any one of the two Urey - Craig groups. Another group, that of amphoteric chondrites, a peculiar sub-group of olivine-hypersthene chondrites, came to be distinguished shortly after. In the meanwhile, further analytical treatment of carbonaceous meteorites by Wik (1956) resulted in the distinction of three well-defined sub-groups within that category. However, these three sub-groups constituted categories which in the following could readily be fitted into the line of the chondrites.

All these elements gradually began to fall into a comprehensive genetic pattern, a first outline of which has been presented in 1959 to the Geochemical Conference in Budapest, in connexion with the investigation of the Kaba mete-

orite (Sztróky et al. 1961). At the time of publication of this latter paper and afterwards, the problem was attacked by numerous workers (B. Mason, A. E. Ringwood, E. Anders, H. C. Urey, B. S. Levin, I. A.

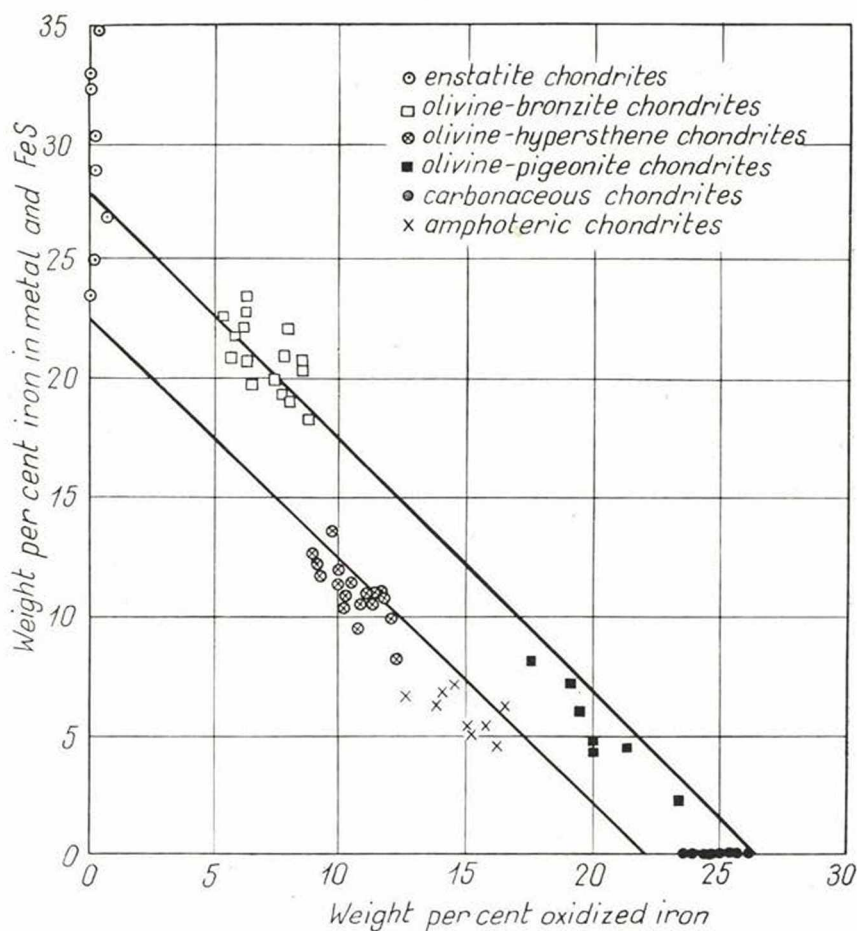


Fig. 2. Relationship between oxidized and reduced iron in chondrites (adapted from B. Mason, 1952).

Wood, C. B. Moore) and its scope widened from a purely meteoritical to a cosmochemical one. It can confidently be stated that the majority of the papers on meteorites published in our days represent a common viewpoint on the fundamental concepts of meteorite genesis, if not on the details of its processes. These fundamental concepts are as follows.

a) Radioactive dating and cosmochemical investigations concur in proving that the solid meteoritic bodies come from the solar system and that their origin was also common with that of the solar system.

b) Astronomical analysis and orbital determinations based on the best visual observations tend to prove that meteorites move on short-period orbits and enter the vicinity of the Earth from the zone of the asteroids. This hypothesis was further confirmed by the first accurate orbital computation, performed on photographs of the Příbram fall (1959) by Czechoslovak workers (C e p l e c h a, 1961).

c) The asteroids revolve on intersecting orbits substantially perturbed by the planets. Friction and collisions cause them to break up, resulting in brecciated textures and angular forms. The regular, sudden changes in the luminosity of the asteroids, observed by means of satellites and lately also of earth-based telescopes, implies unequivocally the irregular angular shape of these celestial bodies. The statistic process of collision and expulsion from the original orbit results in occasional deviations towards and impacts upon Earth and Mars.

d) As regards mineralogical and chemical features, the principal components ( $\text{SiO}_2$ ,  $\text{MgO}$ ,  $\text{FeO}$  and  $\text{Fe}$ ) reveal a considerable variability, most of which is, however, only apparent and is due to variations in the state of oxidation of iron. Thus the  $\text{FeO}$  content ranges from little more than zero (e.g. S. Sauveur, 0.21 percent) to almost 26 per cent (25.60, Mokoia); at the same time, the content of metallic  $\text{Fe}$  decreases from 25.73 per cent (S. Sauveur) to 0.00 per cent (Mokoia). A further fact to be considered, established some time ago by P r i o r, is that the  $\text{Ni}$  content of the metallic phase varies inversely as the percentage of the metallic phase within the meteorite: the less the nickel-iron constituent, the higher is its  $\text{Ni}$  content. A corollary of this feature is that the richer in  $\text{Ni}$  the metallic phase of a chondrite, the more iron oxide there is in the  $\text{Mg}$  silicates. All this means, however, only differences in the distribution among the various phases of the same elements, whose overall abundance is almost the same in all chondrites.

e) As we have seen above, the mineralogical constitution of the chondrites is, with the exception of the carbonaceous ones, fairly uniform. The main constituents are olivine and pyroxene, adding up to a total of 60 to 80 weight per cent. The amount of  $\text{FeNi}$  may reach 27 per cent; troilite amounts to 6 per cent on the average, plagioclase (or glass of plagioclastic composition: maskelynite) to about 8 to 10 percent. Chromite and an accessory phosphate (apatite or merilite) may further be mentioned. An important observation is that the relative composition of the two main constituents is almost invariable: olivine contains slightly more  $\text{Fe}$  than associated pyroxene. This tallies well with the equilibrium conditions of the  $\text{FeO-MgO-SiO}_2$  ternary system. In short, the chemical and mineralogical constitution of the individual chondrite groups fits into a remarkably coherent series, which proves that the meteorites did not come to exist as the products of a series of random, uncoordinated events.

All in all, it is important to bear in mind that, firstly, in the olivine-bronzite and olivine-pyroxene chondrites the main constituents reflect something like a chemical equilibrium, and that, secondly, the  $\text{Fe}$  content of olivine as has been established by B. M a s o n is nearly constant. It seems to us that Prior's rule should essentially be identified with this relationship (or with its average over the entire chondrite field).

The properties of meteorites have lately been studied in detail by Wood (1963) another comprehensive critical study of the topic was published by Anders (1964); These papers put forward the common view that the chondrite texture must have undergone several phases of thermal metamorphism, reflected by differences in crystallinity, so much so that in profoundly metamorphized chondrites there remains hardly any difference between the matrix and the crystalline chondrules. The chemical-mineralogical uniformity attains in some instances such a degree that the resulting chondrites are comparable with Earth rocks, particularly if the sum total of non-volatile elements is considered, i.e. if the analysis is recalculated for atom percentages on a  $H_2O$ -, C-, O- and S-free basis.

The proportions of the elements as established by reliable analyses, including also the trace elements and results determined by activation analysis, reveal the chondrites to have undergone a considerable amount of fractionation, involving the depletion of some elements and the enrichment of others. This is particularly obvious, as will be expounded below, in the case of the carbonaceous chondrites.

The problem thus carries us over into the chondrite group which has so far been dealt with only passim and which does not bear out the above general relationships.

### Carbonaceous chondrites

There are, as has been mentioned above, three sub-groups of carbonaceous chondrites which differ particularly in their physical and mineralogical properties but also in their chemical composition, from the common chondrites. The composition of group I. reflects very closely the primordial state of condensed, solidified matter. Its elemental association is approximately the same as that of "cosmic matter". The loose aggregates, lacking any chondrules and significant crystallinity, may be regarded as an ancient (primitive) stage from which the terrestrial planets were presumably also evolved. Its density is the lowest of all (2.2), it consists largely of amorphous hydrosilicates with some magnetite. Its carbon content is 3.5 percent; moreover, it contains 20.1 percent chemically bound water, free sulfur and even sulfate compounds. The carbon is roentgenographically amorphous and the rest of the substances is not crystalline, either. The X-ray patterns of a typical specimen of this group (Orgueil, 1869, France) show the lines only of magnetite\*. In spite of 22 percent of  $SiO_2$ , no silicate line whatsoever is visible on the diagram.

The carbonaceous chondrites of group II are somewhat denser than the former (2.5 to 2.9). They include serpentine and chlorite varieties as silicatic ingredients: indeed, X-ray analysis has revealed that in some of them (Boriskino, Alais, Mighei, Nogoya, Santa Cruz) a mineral of chloritic or serpentinitic structure is dominant. The mean carbon content is 2.46 percent, the water content 13.35 per cent.

\*According to a verbal communication by J. Orzel minute crystals of magnetite have lately been demonstrated by electron microscopy in the Orgueil meteorite.

Group III (the so-called ornansite group) already contains a great deal of olivine and accessory pigeonite, as well as magnetite and a small amount of layered latticed silicates. Occasionally there is some metallic iron and the texture is decidedly chondritic. The density is 3.4 to 3.5; the mean carbon content is 0.5 per cent.

Carbonaceous chondrites nowadays stand in the focus of interest. The idea that carbonaceous chondrites necessarily represent an earlier, initial state of the evolution of meteorites seems more and more convincing to most workers. These bodies represent a well-defined sequence of evolutionary stages. This means that the individual sub-groups as well as the group as a whole represent certain transitory events in the process of evolution. In a word, the sequence of carbonaceous chondrites is fundamentally a sequence of reduction beginning with group I. Recent evidence in favour of this hypothesis shall be presented below.

Plots of the weight percentages of the individual elements calculated from H. B. Wick's data (1956) indicate gradual transitions. The first diagram shows the relative increase of iron content vs. the decrease of the oxygen content. (Fig. 3) The correlation is so uniform that the graph has a slope of almost exactly  $45^\circ$ . Fig. 4., showing the variation of carbon and hydrogen content, likewise exhibits a monotonous variation. The graphs tend toward zero. (Both elements are highly volatile.)

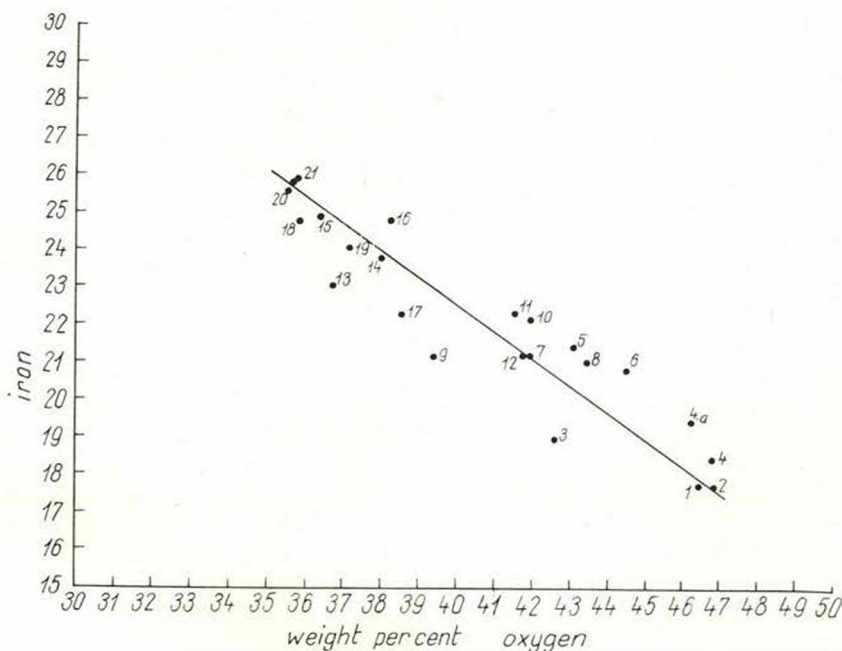


Fig. 3. Relationship between iron and oxygen content in carbonaceous chondrites. Chemical analyses expressed as elements, with oxygen added to make 100 per cent.

1. Alais; 2. Tonk; 3. Ivuna; 4. Orgueil I.; 4/a. Orgueil II.; 5. Nogoya; 6. Cold Bokeveld; 7. Mighei; 8. Nawapali; 9. Haripura; 10. Boriskino.; 11. Erakot; 12. Murray; 13. Santa Cruz; 14. Al Rais; 15. Renazzo; 16. Grosnaya; 17. Kaba; 18. Vigarano; 19. Mokoia; 20. Lance; 21. Felix; 22. Ornans.

The C vs. Fe plot is not less instructive. (Fig. 5.) The amount of carbon is seen to drop almost to zero at maximum iron content. The separation of Wiik's three types of carbonaceous chondrites is particularly apparent in this diagram.

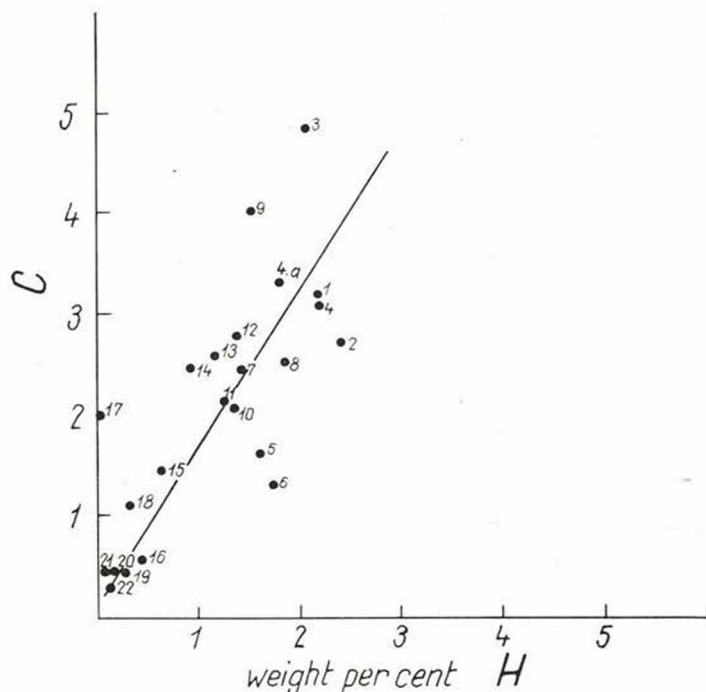


Fig. 4. Relationship between carbon and hydrogen content in carbonaceous chondrites (for key see Fig. 3.)

At the same time the uppermost group, that of the ornansites with 0.5 percent carbon, is seen to represent a gradual transition towards common chondrites. This suggests further that some traces of hydrocarbons or carbon must be present also in the common chondrites.

Above mentioned is confirmed also by the fact that the trace element association of carbonaceous chondrites agrees with the "cosmic" abundances of trace elements, whereas in the common, stony meteorites the trace element content is considerably less. Furthermore, the carbonaceous meteorites contain a great deal of primordial noble gases. Systematic investigations have proved the presence of primordial gases in almost all common chondrites, but their abundance was found to be much less than in the carbonaceous chondrites.

The problems of fractionation and of the depletion of certain elements has been treated in some detail by Anders (1964). On the basis of the data available this author compared the abundances of the "cosmic association" of elements with the abundances established for carbonaceous meteorites. Part



of the results are shown as Fig. 6. The diagram needs no particular explanation: the trends exhibit a pronounced parallelism.

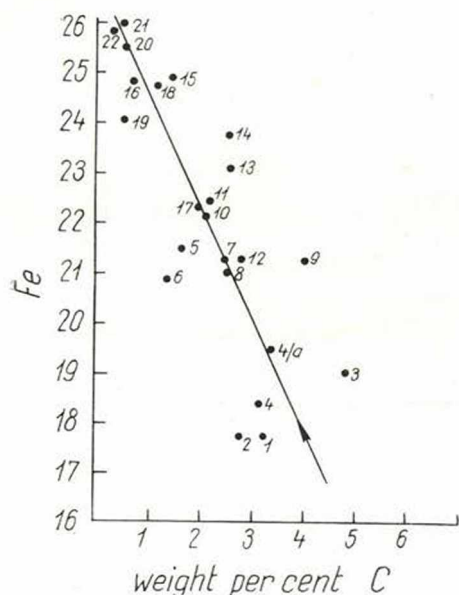


Fig. 5. Relationship between carbon and iron in carbonaceous chondrites (for key see Fig. 3).

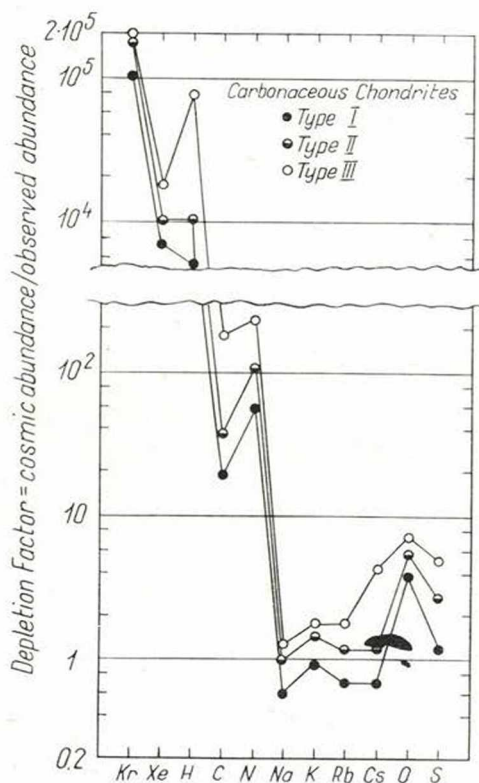


Fig. 6. Abundances for some depleted elements in carbonaceous chondrites (E. Anders).

The study of primordial gases is in a direct relationship with the determination of the age of meteorites and with the study of isotope anomalies. It is known that formerly an average age of  $4 \cdot 10^9$  years has been computed from the helium content and the concentration of radiogenic elements in meteorites. The method has, however, yielded data of considerable scatter, probably owing to inconstant helium loss. Series examinations with other methods have yielded  $4.5 \cdot 10^9$  years on an average. A few age determinations carried out lately on carbonaceous chondrites are listed in the following table.

Ages estimated from the cosmogenic  $^{21}\text{Ne}$  content are listed beside the K/Ar ages. These show about the same trend as the K/Ar data, but of course they give the time since the meteorite orbits as a body (or fragment) of at most metre size. The  $^{40}\text{K}/^{40}\text{Ar}$  data scatter within the above-mentioned range, but do not exceed the  $4.5 \cdot 10^9$  years obtained for common meteorites.

The good agreement of the data enumerated above carries, however, the germs of many a problem concerning the details. A confrontation of carbona-

Table

Carbonaceous meteorite	K/Ar age 10 <sup>7</sup> yrs	From <sup>21</sup> Ne content due to cosmic rays; 10 <sup>8</sup> yrs	Authors
Felix	4,5	56	Stauffer (1961)
Felix	4,1	48	Zähringer (1962)
Lancé	3,9	5	Stauffer (1961)
Mighei	4,3	—	Gerling & Rik (1956)
Mokoia	3,4	4	Stauffer (1961)
Murray	2,5	4	Stauffer (1961)

aceous chondrites with the common ones, which latter bear the traces of decidedly thermal changes of state, reveals the apparent contradiction that the carbonaceous chondrites contain compounds (hydrocarbons) which cannot possibly have withstood a heating above 300 to 350 degrees C. The problem of the *in situ* thermal alteration of the primordial mass is further complicated by the fact that the simple "cosmic" hydrocarbon (methane) has undergone a manifest polymerization, as the chondrites contain many terms of the paraffine-bitumen sequence. It is known that methane decomposes into carbon monoxide and hydrogen at 600 to 650 degrees C. Hence, the first accumulation must have passed through a stage where the small hydrocarbon molecule could lose a hydrogen atom and the radicals which thus gained a substantial chemical potential could unify into hydrocarbons having a higher number of carbon atoms. A further remarkable fact is that in the ornansites that still contain some hydrocarbons, there are chondrules of crystalline magnetite and associated Ni-magnetite (trevorite) (S z t r ó k a y, 1960); indeed, there are well-developed chondrules with crystalline olivine and sometimes pyroxene. On the other hand, some ornansites show traces of layered silicate lattices. (B e r n a l, 1961; K v a s h a, 1948, 1961).

### An outline of evolution history

Instead of a further enumeration of problems concerning certain details and of apparent contradictions let us give a brief summary of our present knowledge of the origin and evolution of meteorites. So much is obvious by now that the size of meteoritic bodies within the solar system could not possibly have exceeded the size of the asteroids. Even if this precludes any differentiation by gravity, certain thermal processes could result in a thermal or physical "zonation". According to some workers, the internal heating of the asteroids must have been due to radioactive decay of short half-life (<sup>26</sup>Al, <sup>60</sup>Fe). Such a mechanism could bring about temperatures above 1000 degrees C. in the planetoid-like body: however, these temperatures prevailed *only in the central part*, whereas the outer surface stayed below freezing point. There was consequently a zone, perhaps as much as a kilometre deep, in which the temperature was of necessity between 0 and 100 degrees C. That is, below the permanently frozen layer of the surface there was a zone where water was liquid. The frozen outer

crust and the "internal atmosphere" (A n d e r s, 1964) developed beneath it played an important role in holding back the noble gases, in the formation of hydrosilicates and in their preservation and, in a general way, in the conservation of the primordial oxidized components and the hydrocarbons, whose polymerisation could not take place anywhere else than in this zone. The hotter zones further inwards gave occasion to the in situ formation of chlorites and serpentines. M a s o n has proved experimentally (1960) that at a temperature of about 600 degrees C., dehydration and partial reduction produces olivine and pyroxene and minute grains of metallic iron out of chlorite and serpentine. Hence, an amorphous silicate of serpentine-like composition can be envisaged as the mother substance of the controversial process of chondrule formation. According to B e n n i n g t o n (1956) it can be proved thermochemically that olivine and pyroxene can form at relatively low temperatures out of the substance of carbonaceous chondrites, as olivine and serpentine attain the state of mutual thermal equilibrium at temperatures as low as 200 degrees C. Hence, in the environment visualized above, the dehydration of serpentine, the continuous formation of chondrules in the solid-phase substance and the reduction of iron is a self-explanatory process. The above image owes much to L e v i n (1965), who added diffusion in the solid and semi-solid state to the processes of evolution taking place in the celestial body of asteroid size. It was expounded by this author that the diffusion coefficient is a highly temperature-dependent parameter: under the same span of time, it produces a much more intense exchange of particles at higher temperatures than in the cold state. Provided that diffusion operates in a mass whose central part has had a temperature of 800 to 1000 degrees C. for at least a few hundred million years, the above mechanism yields all by itself the explanation of the formation of pallasites and also of a homogeneous iron agglomeration. It was seen above that even in the ornansite-type carbonaceous chondrites there are minute droplets of metallic iron and that part of nickel is present in an oxide bond (trevorite). In small grains, surface tension made the initial metallic products of reduction assume sponge-like shapes. Later on, in part of the meteoritic body where temperature reached 800 to 1000 degrees C., the Ni-Fe agglomeration gradually increased in bulk and grew into such an "inclusion" as is represented by the iron meteorites fallen upon the Earth. If the diffusion coefficient is assumed to be of the order of  $10^9$  to  $10^{10}$   $\text{cm}^2 \text{s}^{-1}$ , the time (about  $10^9$  years) elapsed since the origin of the celestial body was sufficient to permit the formation of quite large metallic domains, and even the conditions of formation of the macrocrystals of  $\gamma$  modification (octahedrites) were given. Fragmentation due to collisions led to a separation of the iron and stony iron "inclusions" from the stony matter: owing to differences in density and elastic properties, the stony fraction was liberated from the Fe-Ni fraction. This line of thought entails the view that stony iron meteorites are representative of such stages of the process of evolution outlined above in which the separation of the silicates from the metallic phase did not reach maturity.

Finally, returning once more to the properties of the stony group, let us mention a circumstance which merits further analysis. It seems rather peculiar that the most oxidized carbonaceous chondrites and the most reduced enstatite chondrites (which contain the highest percentage of metallic iron) (Fig. 2),

i. e. the meteorites at the two ends of the line of evolution, should contain one and the same "cosmic" association of trace elements, among others substantial amounts of primordial inert gases. Enstatite chondrites are very rare; no more than 10 or 12 falls are known so far, and their mineralogical composition is somewhat out of the ordinary. Iron is almost entirely metallic or sulfidic, implying a very high-degree reduction. Total iron content exceeds 35 percent and there frequently is a carbon content of about 0.5 percent. The silicate phase consists almost entirely of enstatite; olivine is totally absent; there is some quartz, tridymite, cristobalite, as well as oldhamite, alabandine and daubréelite as accessories. Pronounced chemical similarity to the carbonaceous chondrites suggests this rare group of chondrites to have formed by very sudden and energetic reduction. This hypothesis is borne out by the fact that the chondrules are in most cases rather underdeveloped and that the matrix is impregnated with metallic and sulfidic constituents. If one wishes to stay within the framework of the above-outlined hypothesis of evolution, the formation of this peculiar group must necessarily be interpreted by assuming that some of the meteorites of chondritic state evolved along a sideline. The presentation in atomic percentages of the chemical composition, particularly as shown by the latest analyses, results in the diagram of Fig. 7. This image, which differs from the Urey - Craig synthesis and also from the more recent ones, proves be-

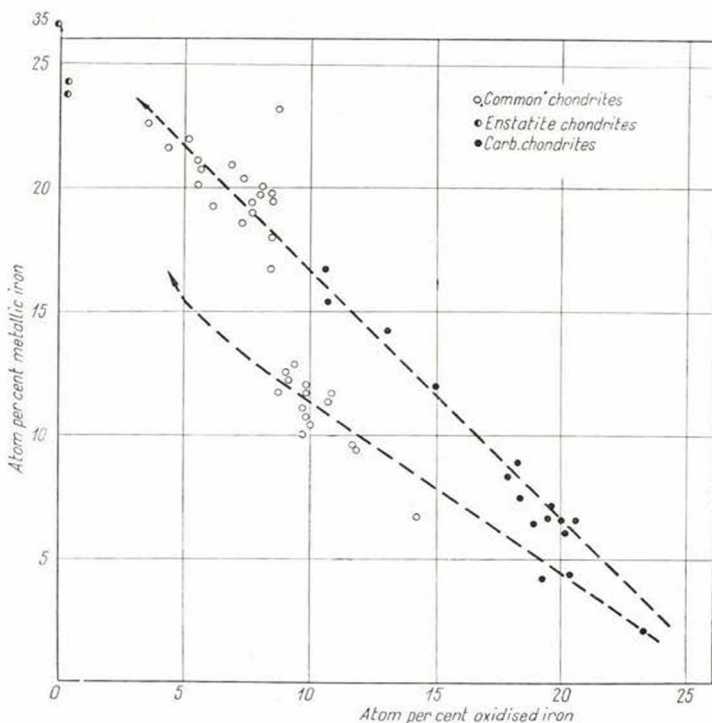


Fig. 7. Relationship between metallic and oxidized iron in 54 mainly new analyses re-calculated in atom percentages on volatile-free basis.

sides the continuity of the "main line" also a common point of origin, an evolution from a common aggregate of matter. In other words, two lines of evolution are traced: the one joins the olivine-pyroxene and olivine-bronzite chondrites, whereas the other, also issuing from the carbonaceous chondrites, passes across the lower amphoterite and olivine-hypersthene chondrites through a gap of some width towards the enstatite chondrites.

All this does not, of course, change the ideas presented above concerning the evolution of meteorites; it only enhances that even if we agree upon an origin within the solar system, and out of a common primordial mass, we must reckon with processes of unlike intensity. These differences in the intensity of the processes follow quite naturally from differences in size and consequent differences in temperature and "internal atmosphere". Internal diffusion, the conservation of volatiles and the intensity of reduction are all size-dependent processes.

In other words, fractionation in the solid state is, over the time and temperature range involved, a highly versatile process which, although evolving into essentially the same direction, may result in more or less important differences in the end products. This effect is further enhanced by the enormous time span of its duration, commensurable with the time elapsed since the origin of the solar system.

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