

Principal aspects of investigations into fluidised carbonisation undertaken at the Marienau Experimental Station

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IN 1948, CERCHAR, (the Centre d'Etudes et de Recherches des Charbonnages de France), started investigations on the application of fluidisation to the carbonisation of coal-fines.

Preliminary investigations led to the construction of a 100 g/hr capacity fluidiser and subsequently a pilot plant having a capacity of 1 ton/hr was jointly set up by CERCHAR and IRSID (Institut de Recherches de la Siderurgie, de France), at Marienau in the Lorraine coal fields. The present paper describes the experiments carried out with this pilot plant.

Facilities used for the investigation

The following equipment was used for the experiments:

(a) *1 ton per hour Carbonisation Unit*: This unit (Fig. 1) incorporates the new advances in the field of carbonisation and provides a number of alternative operating conditions, as will be described later.

(b) *Pre-heater unit*: For fluidised-carbonisation, the coal is generally preheated to about 300°–350°C, (i.e. just below the point of thermal decomposition) to effect economy in the process, and in view of the ease of operation, inspection and control.

A fluidiser of 30 cm internal diameter was used in these experiments; a counter-flow of hot-gases provided the fluidising current. It was possible to treat 200–500 kg/hr of coal in this unit.

This unit has also been used to study the following problems, not directly connected with fluidised carbonisation:

1. Drying of coal or other powdered materials (e.g. fine iron ore). This unit gives better results than are possible with conventional driers.
2. The oxidation of coal, which is sometimes necessary, especially in certain processes for preparing carbonised briquettes. Based on the results obtained with this unit, a successful pilot plant for the process has been constructed.
3. Preheating of coal, before charging into normal coke ovens. The process of dry charging developed at Marienau¹ results in an improvement in the quality of coke as also in increased

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¹ P. Foch "Possibilities of using slightly-coking coals, provided by previously drying the coal-blend"—Paper presented at the symposium on "Carbonisation" at the Central Fuel Research Institute, Jealgora (India) in March, 1957.

production. A considerable improvement in production is also effected by charging coal preheated to just a little below the point of thermal decomposition viz. 300–350°C. This process has been successfully tried on a semi-industrial scale, and the results have led us to conclude that they should be pursued on a larger scale.

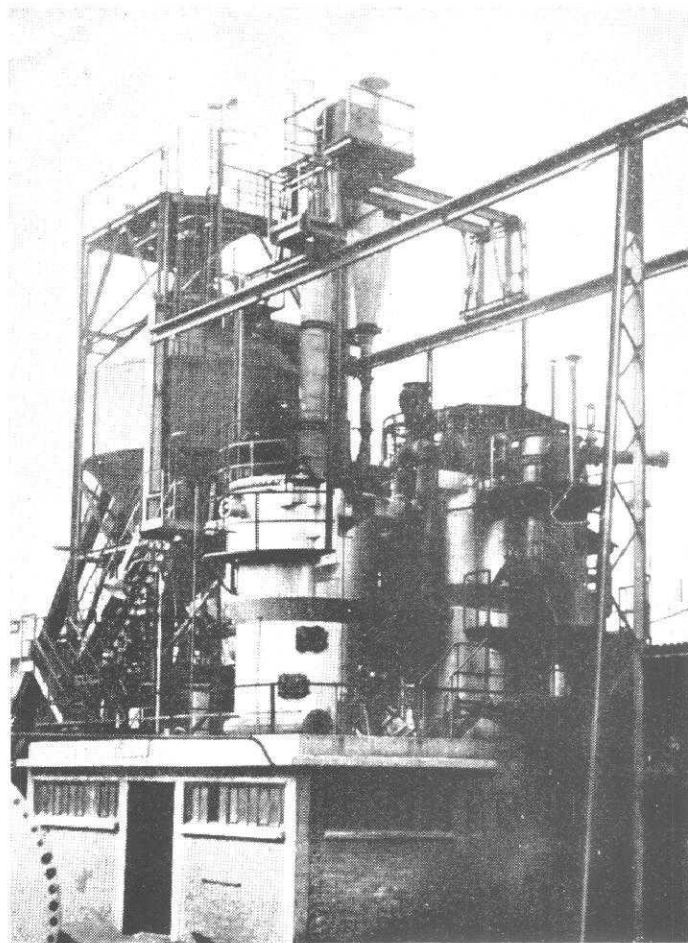


Fig. 1.
1 ton/hr. fluidised-carbonisation plant at the Marienau Experimental Station.

(c) *Small-scale unit for room temperature study:* A special unit, similar in dimensions to the above unit (see 'b' above) but made in part of a transparent material, (such as perspex), was used for a detailed study of the properties of fluidised beds and other connected problems e.g. the distributor grids and dedusting cyclones, which can be economically studied at room temperature.

Fluidised carbonisation

The problem, nature of the product and possible uses: The demand of coke-breeze has increased in France in recent years. It is expected that within a year or two, it will be in a sizeable shortage, particularly because of its increased use in two processes undergoing rapid developments at Lorraine, and their adoption in the coal and steel industries:

- (a) Coke-breeze is used as a diluent in coke oven plants, which employ tamped charging. Considering that most coke oven plants do not produce sufficient coke-breeze for their own requirements, and the developments at present in progress, it can be estimated that an additional 100,000 tons of coke-breeze per year will be required for this purpose.
- (b) Coke-breeze is also used as a fuel in the sintering of iron ore. This process is now being adopted by more and more plants in France and it is estimated that the additional requirements of coke-breeze for this use alone would be at least 600,000 tons per year.

Thus the shortage of coke-breeze, in France, would be of the order of 700,000 tons per year at the end of two years. Similar requirements have been estimated for other countries, particularly those which are poor in coking coals and where direct reduction of iron ore is being considered.

It is, therefore, desirable to find ways and means to fill this gap. Various alternatives have been suggested for the purpose, such as, coals of different origin, semi-coke from a rotary-furnace, small crushed coke, and imported breeze. It seems that the best alternative, both from economic and technical considerations, would be a coke-breeze obtained by fluidisation of inferior coal. This breeze closely resembles the breeze from normal coking coals, and complete equivalence of the two, in the application under consideration, has been proved by semi-industrial and industrial scale trials. These trials made use of the coke (volatile matter 1-2%) which was obtained by treating a dry long-flame coal* in the form of 0-6 and 0-10 mm fines in the 1 ton/hr unit. The unit is quite flexible; it is possible to obtain in it cokes with volatile content varying between 1 and 18% by controlling the temperature of treatment.

The principles of a fluidised-carbonisation unit: Carbonisation takes place in a fluidisation reactor (Fig. 2) continuously fed with coal fines and providing

* Volatile matter=39% on pure coal basis: AFNOR swelling index (equivalent to BS Crucible Swelling Number)=1; non-coking.

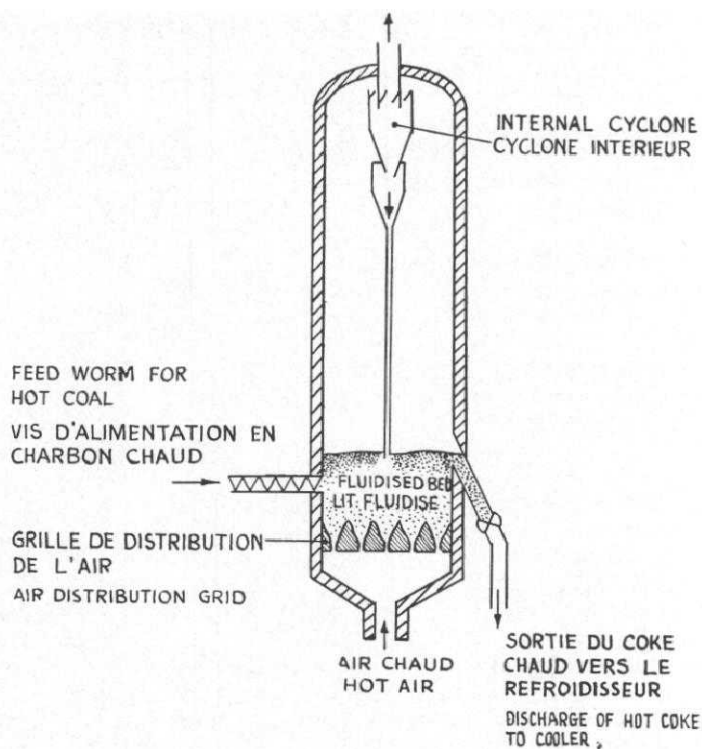


Fig. 2.

Diagrammatic layout of the fluidisation-carbonisation unit.

a counter-current of air. The heat required for the reaction is provided by partial combustion, and the temperature of the bed can be controlled by varying the ratio of air to coal. This temperature determines the volatile content of the products obtained. Thus, starting with the dry long-flame coal (which we use most frequently), the volatile content falls from 18% to 1% as the temperature of the bed rises from 450° to 850°.

Heating by partial combustion has the advantage of providing a good thermal balance, but has a disadvantage in that the gas produced is diluted by atmospheric nitrogen. It is, therefore, advisable to use pre-heated air and coal. Air can be easily pre-heated to 500-600°C by using a simple metallic heat exchanger. Preheating of the coal can be done with advantage in a fluidiser supplied with hot gases from a pressurised combustion chamber.

The product obtained, coke or semi-coke, is discharged from the apparatus at the temperature of the bed. It must be cooled, before further handling. Spraying with water in the open air would be effective, but would produce too much dust. A simpler method would be to apply the fluidisation technique. The hot semi-coke is fed into a special fluidiser, where the flow of gases keeps the solid matter in a state of incipient fluidisation. Injection of water into the middle of the bed produces cooling by evaporation. The supply of water can be controlled easily (and

Yields from and features of various operating conditions

	Single-stage carbonisation				Carbonisation with auto-thermal cracking		Two-stage carbonisation			
	Temperature of carbonisation : 500°C		Temperature of carbonisation: 800°C							
(1) Characteristics of products										
Initial coal	Dry Flame coal	10% moisture						
			8% ash							
Volatile content of coke	14%	2-3%		2-3%		2-3%		
Approximate upper calorific value of gas (before benzole removal)	2200 cal/cu. m.	2600 cal/cu. m.		2000 cal/cu. m.		3000 cal/cu. m.		
			N.T.P.	N.T.P.		N.T.P.		N.T.P.		
Ash content of coke	10-11%	12-13%		12-13%		12-13%		
			Weight or volume	Thermies	Weight or volume	Thermies	Weight or volume	Thermies	Weight or volume	
(2) Industrial yields per (metric) ton of dry coal										
Coke (kg)	770	5350	625	4500	630	4550	630	4550
Pitch (kg)	68	640	35	330	0	0	68	640
Dust (kg)	14		10		0	0	14	
Oils (kg)	40	320	20	140			40	320
Gas before benzole removal (cu. m. N.T.P.)	280	630	600	1600	1000	2000	400	1200
Preheating gas (subtract from gas produced)		-300		-300		-60		-300
(3) Overall industrial thermal efficiency (1) (Carbonising unit plus preheating unit)			92-93%		87-88%		90-91%		89-90%	

(1) Determined by: Upper calorific value of products discharged (semi-coke, coke, gas, tar).
Upper calorific value of material charged (gas, coal).

N.T.P.—Normal temperature and pressure i.e. 0°C and 760 mm. Hg. pressure.

is very well suited to automatic control). The solid product is then discharged practically cold (70 to 80°C) and dry.

It may be emphasised that each of the above fluidisers is fitted with a series of de-dusting cyclones.

Types of installation used at the Marienau Station: Based on the typical layout described above, three alternative types of installation have been envisaged, the choice between them being governed by the nature and quantity of the products required i.e. coke or semi-coke, on the one hand and tars and gas on the other (See Table).

(a) *Single-stage carbonisation*, which produces gas, tar and coke breeze, of a volatile content between 1 and 18 per cent according to operating conditions.

(b) *Carbonisation with post-combustion*, yielding only gas and coke breeze, with a volatile content of 1 to 2%.

(c) *Two-stage carbonisation*, producing low-temperature tar in considerable quantities, together with gas and coke breeze with a volatile content of 1 to 2%.

(a) *Single-stage carbonisation:* The layout of this unit is based on the principles set out earlier. Fig. 3

shows the carbonising unit, which is supplied with air and coal, preheated in the appropriate apparatus, together with the cooling unit. The treatment of the gas—which contains tar, water and fine dust, which has not been trapped in the cyclones—poses a difficult problem, since abrupt cooling would bring down a ternary mixture, almost impossible to decant. This difficulty has been got over by carrying out condensation in two stages. A preliminary cooling is carried out by spraying with recycled oil. This removes the pitch and almost the entire quantity of fine dust. The lighter oils which remain, together with the water, are then condensed and separated by decantation.

Detailed experiments, spread over a long period, have been carried out in the 1 ton/hr plant, described earlier. It was possible to extend operations over several days which permitted the production of sizeable quantities of coke or semi-coke for determining the proper working conditions, and to study the economics of the process. From these studies, it has been concluded that whereas this type of installation would be suitable for the preparation of semi-coke with a volatile content of 14-15%, and would also give a high yield of low temperature tar, it would be less attractive for the production of coke with 1-2% volatile matter, since,

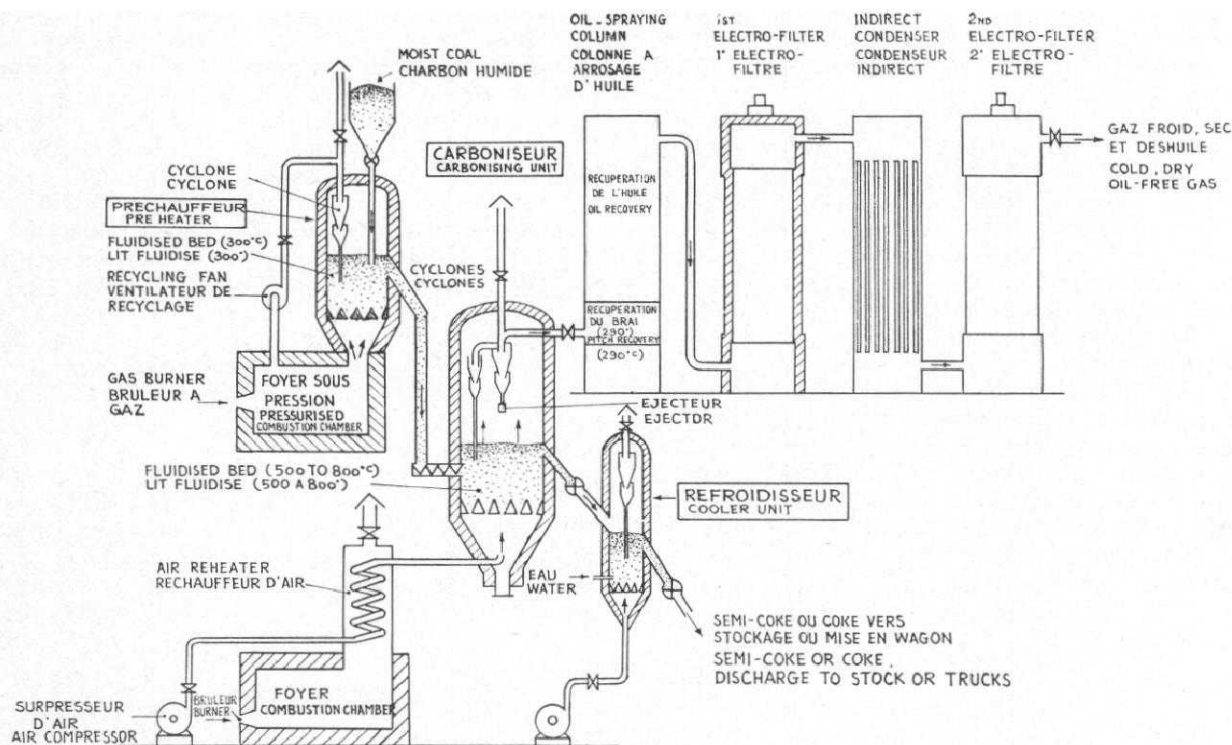


Fig. 3.
Single-stage carbonisation.

in this case, the tar yield is lower and the thermal balance not so good.

(b) *Carbonisation with post-combustion*: This installation, intended solely for carbonisation at 800° to 850°C, yields gas which does not contain any tar vapours. On leaving the carbonisation bed, the mixture of gas and tar vapours is cracked by partial combustion—by injection of secondary air—thus raising its temperature to about 450°C. The gas can now be used for the drying and preheating of moist coal. The process effects an appreciable improvement in the thermal balance, because it eliminates the need of producing hot gases in a separate combustion chamber. The layout, shown in Fig. 4, indicates the relative positions of the different stages. The coal passes through the following circuit.

After drying and preheating to 300°C in the upper bed by the gas from the cracking stage, the coal passes down into the lower bed, where it is subjected to carbonisation at 800°C by partial combustion with air. The hot coke obtained then passes into a cooling unit, similar to that in the single stage plant. On leaving the upper bed the gases are de-dusted by batteries of internal cyclones and are then sent to a scrubber, eliminating the expensive tar-recovery units. The process improves the thermal efficiency by three to four units in comparison with that of the single stage installation (c.f. column 2 in the Table).

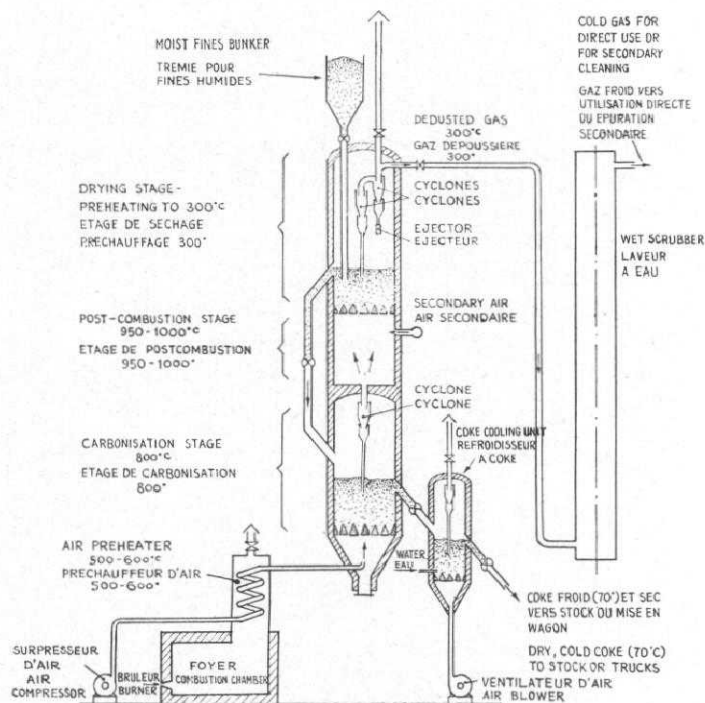


Fig. 4.
Carbonisation with auto-thermal cracking.

Developmental studies are at present in progress on the 1 ton/hr plant, but sufficient information is already available to plan the construction of a 10 ton/hr unit.

(c) *Two-stage carbonisation*: This installation, intended for the production of tar, makes use of the available heat of the gases leaving the 800°C carbonisation stage to partially carbonise the coal at 500°C, transforming it into semi-coke. It is necessary to provide an independent installation for preheating the coal, with a combustion chamber which produces hot gases, as in the single stage unit (a). The installation (Fig. 5) would operate in the following manner:

The coal is dried and preheated in the first fluidising unit. It then passes into the upper bed of the carboniser, where it is transformed into semi-coke having a volatile content of 14-15%. This semi-coke overflows and passes down into the lower bed, where carbonisation at 800°C turns it into coke (volatile content 1-2%). This coke is cooled in the last fluidising unit, using the same principles as in the other two installations. The

gases from the lower stage of the carboniser provides the heat for partial carbonisation and after being de-dusted by cyclones, is fed to a condensation system similar to that in the one stage unit.

It will be observed from the table (column 4) that in this process the yield of tars is higher, and that of gas is lower, but the gas itself has a slightly higher calorific value. The thermal balance is nearly equal to that of installation (b).

This type of plant is to be tried out shortly at the Marienau Station; it is probable that, taking into account the technological information obtained with the other installations, we shall rapidly obtain the data necessary for constructing a 10 ton/hr plant of this type.

Conclusions

We have examined three types of installations for fluidised carbonisation of inferior coals envisaged at the Marienau Experimental Station. It would seem that, under the prevailing conditions in France, the industry will be particularly interested

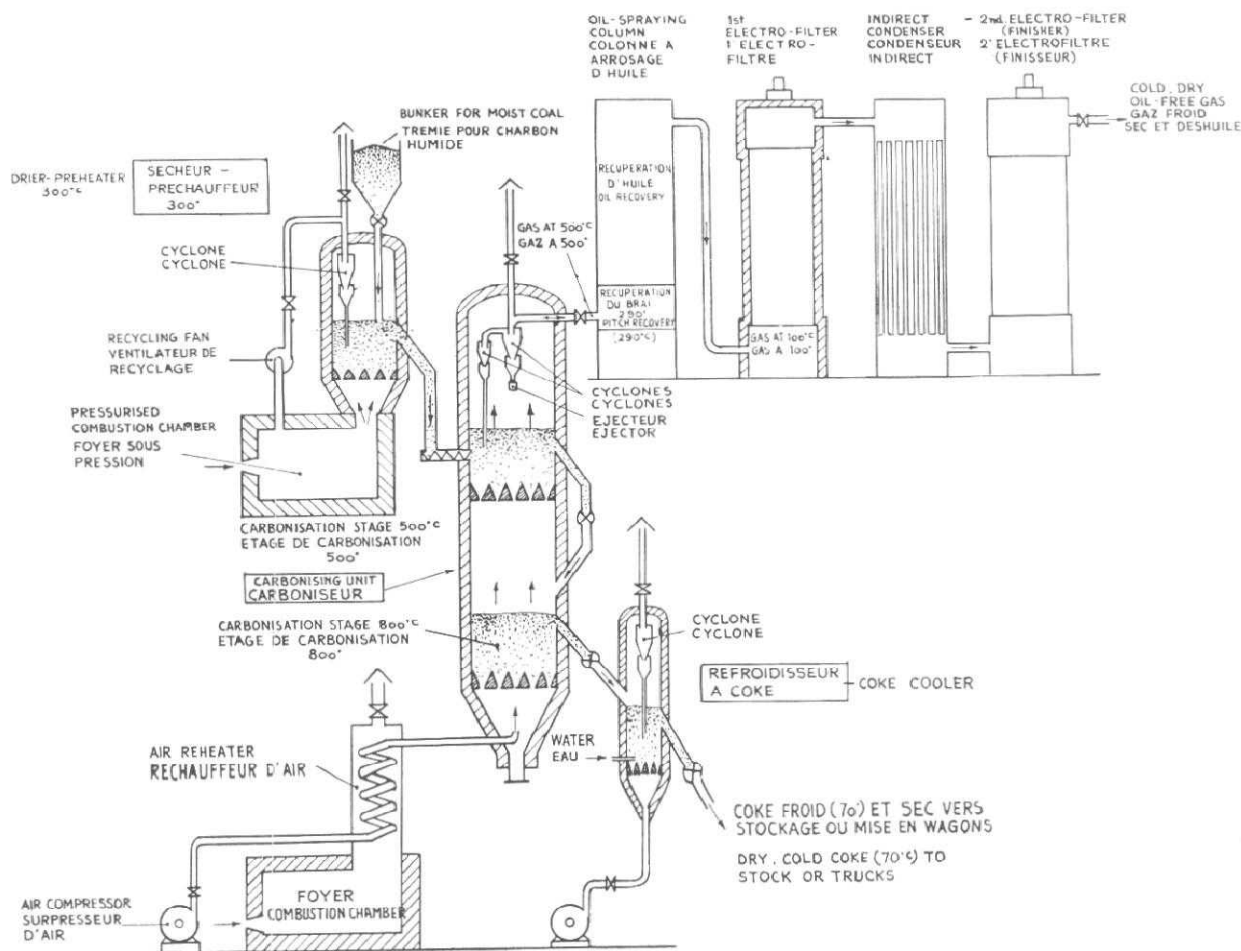


Fig. 5.

Two-stage carbonisation with preheating of the air and the coal.

in the latter two installations described.

The installation using post-combustion—which needs the lowest investment outlay and has the lowest requirements in power and labour—is of the greatest interest for the steel-making industry which requires coke breeze for the sintering plants; they would be able to find a market for the lean gas produced by feeding it into the internal gas mains. On the other hand, the tars produced are of little interest to the steel industry.

The economic returns from the two-stage carbonisation plant are essentially based on the market

for by-products. It is, therefore, of most interest to those companies which are able profitably to exploit the tars obtained.

This naturally leaves the first type of installation, which simultaneously produces gas and tar, for use only where a semi-coke with a high volatile content is required.

Various foreign countries might be able to resolve certain problems in the treatment of local coals by choosing one of these three types of installations, thereby facilitating the use of local coals in their coal or steel-making industries.

DISCUSSIONS

Mr. Das Gupta, C.F.R.I., Jealgora: May I request Prof. Crussard for some more particulars about the tar recovered from the process he has described, particularly its dust content? It was stated that a non-flaming coal with 40% volatile matters was used. May we know whether it was of an absolutely non-caking type?

We are not yet certain as to whether such a process will be useful in India, as our problem in this country is to make a low temperature char suitable for blending with some of our high volatile coals to produce metallurgical coke. Comparative coking tests using blends containing high temperature coke with 2 to 3% volatile matter in one case and low temperature char with 10 to 13% volatile matter in another case, have shown that while the latter used in 15% of the charge makes metallurgical coke, not more than 3 to 4% of the former coke can be added to most Indian coking coals without impairing the physical properties of the coke. I, therefore, feel that the coals to which these fluidised chars were added must have been of different types than ours. We are in fact confronted with the problem of disposing of the small amount of coke breeze produced in the present plants in India. Messrs Tata Iron and Steel Co. are now utilising the coke breeze in boilers and I feel that the rest of the coking industries could re-cycle for coking the little coke breeze they produce and by this their problem of coke breeze disposal will probably be solved and we may not have to produce artificially any high temperature coke breeze for blending. However, I would like to emphasise that addition of coke breeze produced by any manner is not a panacea for getting a metallurgical coke from high volatile coals in India.

Dr. A. Lahiri, Director, C.F.R.I., Jealgora: I feel that this process has got very great possibilities in India, apart from the question of making a char for blending with high volatile coal as was pointed out by Mr. Das Gupta. With certain

coals, for example, with the Dishergarh coal of the Ranigunj coal fields which has been so far used for steam raising by the railways and for export, and which has not been classified as a coking coal, we have been able to make a metallurgical coke with a blend of low temperature char prepared by fluidised carbonisation to the extent of 10 to 12%, but apart from this I think there is a very wide scope even otherwise. We have been experimenting on this for the last few years on a 4" reactor and we are now building a 12" reactor for making fluidised char for several purposes. It is now coming to be realised that burning coal in its raw state may be a wasteful process and the first attempt at this has been made in the Texas Aluminium Co. in America where they are carbonising lignite and using the char for generation of power; at the moment they do not know exactly what to do with the tar and are running it into a million gallon tank and carrying out research. Pittsburg Consolidated also has taken it up very seriously. In the United States, for example, they expect that by 1980 they will burn about 400 million tons of coal in power plants and they are thinking that even if half of it could be carbonised and burnt in the form of char a very huge quantity of tar could be obtained for processing both into petroleum as well as petro-chemicals, particularly for making middle distillates; the other aspect of it is that the fluidised char could be used for gasification. It is a very reactive fuel and it could be gasified with oxygen for the production of synthetic gas either for making oil and other hydro-carbons or for making fertilisers and at the same time a huge quantity of tar comes out, primary tar containing as much as 15 to 20% or even more of phenols which can be processed. Ultimately, the economics of this process depends on the cost of heat input for carbonisation and the treatment of tar; if the

cost of carbonisation can be met by the processing of the tar, then the char will be available at the same price as coal. I believe this process has one of the greatest possibilities, particularly in India where the exploitation of non-coking coal will be increasingly greater.

Prof. Charles Crussard, Director, IRSID, France: I wish to thank Dr. Lahiri for his precise comments on the position of India regarding the coke situation.

Regarding the question by Mr. Das Gupta about the by-products, I cannot give a definite answer, as the composition of the tar depends of the type of fluidising units chosen, on the operating conditions, and also on the way tar is recovered: a single-stage cooling would yield a complex mixture, almost impossible to decant; but by carrying out the condensation in two stages, the preliminary

cooling by spraying recycled oils brings down pitch and fine dust, while the lighter oils are recovered in the secondary stage.

The coal used for these trial runs was a dry flaming coal with about 40% volatile and 8% ash content; regarding its caking quality, it had a British Swelling Index 1 to 1.5. The dust content of the product was of course somewhat higher, about 12%.

Regarding the possible application of this process in India, I do not know exactly the situation in this country, but the point is that, if you only use with ordinary caking techniques a mixture of coke breeze with high volatile coals, it does not give always good coke; but, if you use preheating or tamping methods, combined with the mixing of coke breeze, you may have excellent products.

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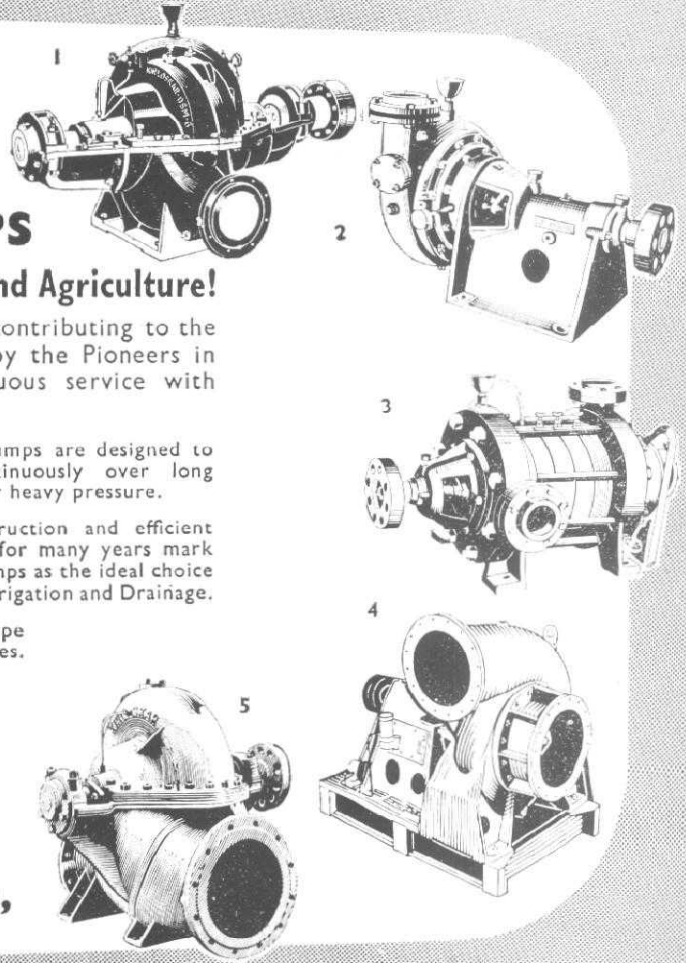
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