3

FUEL OIL FROM COAL

Low Temperature Carbonisation and Hydrogenation in Great Britain.

The extension of the use of home produced fuels by production of liquid fuels from coal is of great importance not only from the economic aspect, but also as relieving to some extent our dependence on foreign fuel in time of war. Some account of the present position as regards the two main processes involved, viz., Low Temperature Carbonisation and Hydrogenation may therefore be of interest.

It is not proposed in this article to discuss the economic aspect of the various processes, except that it may be stated that the distillation of crude petroleum oils for fuel products is likely to remain a cheaper process than extracting similar products from coal provided the supply of crude petroleum is adequately maintained, and also, other things being equal, it is obviously the most economical course to concentrate on the products which command the highest price in the market (at present, motor spirits).

Low Temperature Carbonisation.—Much of the early work in the low temperature field was inspired by an appreciation of the evils and waste resulting from the burning of raw coal for domestic and general uses. Quite independent of the medical aspect of the smoke problem, those who remember London during the great coal strike of 1926 will have carried away unforgettable memories of what our Capital might be. It is difficult to evaluate the damage caused by the smoke produced from domestic chimneys, but a semi-official statement has placed the figure as high as 10*s.* per ton of coal consumed. In the case of the Houses of Parliament alone the cost of repair due to corrosion of the stone work, consequential on the attack of atmospheric acids, has been estimated by the Office of Works at over one million pounds.

On an average some 35 per cent. of the total coal mined in Great Britain reaches the pit head in the form of smalls and dust. This small coal has the same calorific value, and therefore the same intrinsic value as large-size coal from the same seam, and is cheap only on account of its size. When a suitably-designed low-temperature retort is charged with certain kinds of small coal the heat will cause the charge to swell and form itself into large pieces. The process, therefore, to all intents and purposes, converts the coal dust into synthetic lumps of anthracite, the latter being more suitable for open fires as well as central heating stoves.

Low-temperature carbonisation has been acclaimed as the potential saviour of the coal industry and as a means by which oil may be obtained from coal thus rendering the country independent of imported petroleum. To view the situation in its true perspective, it must be realised that oil is only a by-product of this process. When 1 ton of coal is carbonized at so-called low temperatures, some 14 cwt. of smokeless fuel are produced, the remaining 6 cwt. of by-products include crude tar, motor spirit, rich gas, sulphate of ammonia, ash, etc.

The art of low-temperature carbonisation is flexible. There are already several types of process working on a sound basis. The chief processes being worked in Great Britain have for their objects the following :—

(1) Provision of smokeless fuel for domestic use. By-products : Crude coal oil, petrol, and a small quantity of rich gas.
(2) The same as No. 1, but accompanied by the production of larger quantities of gas of lower calorific value.
(3) Provision of low-temperature cokes for industrial purposes, including pulverised fuel.
(4) The production of low-temperature cokes for blending with coal for the purpose of improving the ultimate coke, whether gas or metallurgical.
(5) For the distillation of low-grade non-coking coal, lignites, cannel, shale, etc., mainly for the production of oil.

There are three main types of process in use, the chief difference being the nature of the retort and the manner in which the actual carbonisation is carried out. The three methods are briefly as follows :—

(1) Stationary coal heated in a stationary retort.
(2) Coal slowly moving heated in a stationary retort.
(3) Coal moving through retorts which themselves move or have moving parts.

Methods 1 and 2 yield domestic smokeless fuel as their chief product. Method 3 yields coke in a form suitable for industrial purposes, such as blending or for pulverised fuel, while, in cases where cannel coal is treated, the chief product is tar.

The usual carbonising temperature is between 500° C. and 600° C. with the different systems, and the time of carbonising varies from four hours in the case of the stationary coal in stationary retorts to above 22 hours in large stationary retorts in which the charge slowly passes through different heat zones. It is interesting to note that these plants operate continuously over long periods at, or very close to their maximum capacity. One plant in particular has been in continuous operation for over four years. This shows that the problems of gas handling and keeping the plant clear have been solved, and apart from the time taken for renewals as may be required, there is no need to close down for a single day.

The amount of by-products produced varies somewhat according to the process, but the main factor in determining the amount is the nature of the coal used. The average yield of crude tar from British bituminous coal may be taken at from 18–20 gallons per ton of coal carbonised. This may seem small in comparison, but when it is considered that if the 40,000,000 tons of coal used annually in this country for domestic purposes were to be carbonised, the annual yield of tar would approximate to 3,500,000 tons, of which rather less than half would be ultimately suitable for Naval use as a fuel. This crude tar is now treated by distillation and washing for the production of various materials required by the chemical industry, including light spirit, naphtha, solvents, cresyllic and carbolic acid, crude dye stuffs, disinfectants, synthetic resin, creosote and pitch.

The heavier neutral distillate is then topped to reduce flash point, moisture content, etc., and constitutes a fuel oil, which, from its analysis, is seen to compare favourably with petroleum fuel oil.

Specific gravity at 60° F.		0·974
Flash point (Pensky-Martens)		178° F
Viscosity on Redwoods Patt. 1—		
In seconds at 60° F.		58
In seconds at 32° F.		120
Sulphur	per cent.	0·85
Water by volume	,,	0·10
Inorganic acidity		Nil
Sediment (total insoluble in benzine)	per cent.	0·02
Naphthalene	,,	1·5
Anthracene	,,	Negligible trace.
Pitch (residue on distillation)	,,	2·2
Crude coal tar acids (by volume)	,,	2·0
Paraffin wax		Trace.
Ash content		0·03
Spontaneous ignition temperature		323° C.
Calorific value ; B.Th.Us.	per lb.	17·750 (max. fig.).

It is interesting to note that although the calorific value of coal oil is somewhat lower than that of the average petroleum fuel oil, this is compensated to some extent by the former having a much higher specific gravity.

Thus :—

Petroleum Oil (density, 0·91)—

Weight of 1 gallon .. = 9·1 lb.
Calorific value .. .. = say 19,000 B.Th.Us. per lb.
B.Th.Us. per gallon .. = 19,000 × 9·1.
= 172,900.

Coal oil (density, 0·974)—
Weight of 1 gallon .. = 9·74 lb.
Calorific value .. .. = 17,750 B.Th.Us. per lb.
B.Th.Us. per gallon .. = 17,750 × 9·74.
= 172,885.

One of the principal features of low-temperature oils is their complete freedom from the offending solids naphthalene and anthracene. Neither of these materials is formed in the crude tar provided the temperature at which the coal is originally carbonised is not raised above 600° C. Another feature which cannot be overlooked is its miscibility with petroleum oils, provided that the pitch content is not more than about 2 per cent.

Trials have been carried out using oil obtained by means of the high temperature carbonisation process, such as is used in the production of coal gas, but it was found that this creosote oil had many disadvantages as compared with petroleum. The oil was burned in admixture with Trinidad fuel (one part creosote to two parts fuel oil) on a number of ships. It was found that the mixture burned quite satisfactorily with no unusual amount of carbon deposit and no excessive choking of the filters. It is known that precautions in handling were necessary in order to avoid skin irritation, probably due to the anthracene content; and there existed the danger of choking of the oil system with precipitated naphthalene and anthracene. The extent of dilution of the creosote which is necessary to prevent this, is indicated by the experimental fact that anthracene and naphthalene are deposited from mixtures of creosote and petroleum oil when the proportions in the mixture exceed about 0·25 and 7 per cent. respectively. Similarly, pitch is deposited if its amount in the creosote exceeds 2 per cent.

Hydrogenation.—The fact that coal can be chemically converted into oil has been known for a considerable time, but the early methods employed expensive chemicals, and were regarded as interesting scientific experiments. It was not until Bergius in 1912, demonstrated that coal could be converted into oil by the action of the gas hydrogen under conditions of heat and pressure, that the matter began to have practical importance.

Bituminous coal consists essentially of carbon, hydrogen and oxygen, with small quantities of nitrogen, sulphur and other bodies. The hydrogen contents of bituminous coals range chiefly between 4½ to 5½ per cent., and for rough calculations we may safely assume that the available hydrogen in coal, over and above that necessary to combine with the oxygen in the coal, is of the order of 5 per cent. Mineral oil is much richer in hydrogen, containing some 14 to 15 per cent. It might reasonably be assumed, and is in fact the case, that if about 10 per cent. more hydrogen can be added to the coal a liquid fuel can be produced.

The Bergius method for the conversion of coal into oil is in essence quite simple, though the technical difficulties which must be overcome are great. The coal is ground to about the same degree of fineness as is employed for coal dust firing and mixed with about 40 per cent. of its own weight of the heavy oil produced during an earlier operation. To this mixture is added a special catalyst which greatly assists the chemical reactions.

After intimate mixing this paste was pumped into a heated vessel through which hydrogen was passing under a pressure varying from 150 to 250 atmospheres, according to the nature of the work in hand. The vessel was externally heated by gas flames, and its contents were kept agitated by means of revolving scrapers. At the exit end of the vessel the now comparatively thin fluid tar-like product was discharged through a valve into a receiver, where the liquid separated from the gas.

It will be realized that the high temperatures and pressures required for the process leads to considerable mechanical difficulties, these difficulties can, however, be overcome and, technically, the process is quite practicable.

The crude tar derived from high or low temperature carbonisation may also be hydrogenated to produce motor spirit and Diesel oils, and the process is described below.

It was found that certain coals were not amenable to this process. As an illustration, with certain Scotch coals reaction did not start until, say, 480° C. under 200 atmospheres pressure, and at 482° C. the autoclave contents might be converted into a more or less coherent mass of coke.

Hydrogenation of Crude Coal Oil.—The chemical difference between crude coal tar and the various grades of petrol lies principally in the quantity of hydrogen present in relation to carbon. In round figures, the one contains some 8 per cent. and the other some 15 per cent. of the hydrogen element. It will thus be seen that to convert coal oil into petrol it is necessary to add roughly 7 per cent. of hydrogen. A further small quantity of hydrogen is required to combine with the oxygen, sulphur and nitrogen present in the coal oil, transforming them into water, sulphuretted hydrogen gas and ammonia respectively. In this form they are free and can be removed.

In this process also the presence of a suitable catalyst is essential if the conversion of a high percentage of coal oil into petrol is to be achieved.

In view of the promising results from many experiments the Fuel Research Station, in 1929, designed and erected a plant for the continuous treatment of five gallons of low temperature tar at 200 atmospheres pressure and at temperatures of 450° C. to 460° C., and 480° C. to 490° C. in the liquid and vapour phases respectively.

The chemical change takes place in a vertical pressure converter, the latter being heated externally by an electric furnace. Tar and hydrogen under pressure are fed in at the top of the converter and pass down through an internal pipe to the bottom. The tar and hydrogen are thus pre-heated before entering the catalyst space. The catalyst is contained in a cage which just fits the reaction vessel, the vapour and liquid phases being separated by a perforated plate. In the liquid phase compartment there is a baffle plate about 1 in. from the bottom which prevents the formation of a stagnant layer of liquid at the bottom of the converter, and thus avoids unnecessary coke formation.

The pre-heated tar and hydrogen pass from the bottom through the liquid and liquid phase catalyst. The liquid in the reaction vessel is maintained at a constant level by a standpipe from which " heavy product " is withdrawn intermittently. From the liquid phase the low-boiling portion of the tar vapourises, together with the fresh low boiling material formed. The mixture of vapours and residual hydrogen pass upwards through the bed of vapour phase catalyst. The products then pass through a condenser to a receiver where permanent gas and liquid " light product " are separated.

The results from 20 gallons of tar using impregnated charcoal containing 25 per cent. of ammonium molybdate as catalyst are given below :—

Motor spirit to 200° C.	11·6	20·2 gallons.
Diesel oil	8·6	

With an expenditure of 2,400 cu. ft. of hydrogen.

A consideration of the markets for motor spirit and Diesel oil would decide whether this high-boiling material should be reprocessed or not.

With reprocessing the yield of spirit (0 to 200° C.) in one stage was 33 per cent., and its composition was similar to that of the spirit obtained in the first treatment of the tar. The spirit produced, after refining with 70 per cent. sulphuric acid and caustic soda, was water-white, and had a specific gravity of 0·80. Its distillation range was 18 per cent. to 100° C., 31 per cent. 100° to 120° C., and 48·5 per cent. 120° to 200° C. The anti-knock value of the spirit was high, 1·3 ratios above Summer Shell.