TESTS ON
THE LIQUEFACTION OF CANADIAN COALS
BY HYDROGENATION

By

T. E. Warren and K. W. Bowles

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PREFACE

During the past twenty years the rate of the world's consumption of petroleum has greatly increased. In 1937 it was twice that of 1924, and three times that of 1920. About 2 billion barrels was produced in 1937, and at the end of that year it was estimated that the reserve in proved fields was about 20 billion barrels[1]. This reserve, in itself, assures an adequate supply for only a short time because a deficiency can occur while a great deal of recoverable petroleum remains underground. The reason for this is that the rate of production cannot economically be raised beyond certain limits.[2] Therefore, the maintenance of a sufficient supply to meet the expanding requirement is dependent upon the discovery of new fields.

The rate at which new petroleum reserves will be discovered is uncertain, and a review of the recent trend is not reassuring. According to a report prepared by A. W. McCoy for the Marland conference on oil conservation, the rate of discovery in the United States has declined more than 50 per cent during the past seven years as compared with the rate from 1924 through 1930.[3] The amount of petroleum consumed in that country during the past few years has been reported to exceed the amount discovered in new fields.[4] In spite of this, the proportion of the total world output produced in the United States has been increasing slightly and is now about 63 per cent.[5]

It is not too soon, therefore, to take under consideration the consequences of a possible deficiency of petroleum. As the demand approaches the potential supply it is probable that the price of petroleum products will rise until substitutes can compete economically. The proportion of these will then gradually increase as petroleum production diminishes. For this reason the raw materials and methods employed in the manufacture of substitutes are of considerable interest.

The principal raw materials available for the production of substitute liquid fuels are coal and the natural bitumens occurring in shales and sands. These materials differ from the fluid hydrocarbons in that they contain a smaller proportion of hydrogen. For example, a bituminous coal has about 5 to 6 per cent of hydrogen, whereas gasoline has about 14 per cent. Accordingly, any method which is to produce a large yield of gasoline from coal must be based on the addition of hydrogen.

There are two outstanding processes for the manufacture of liquid fuels from coal. The first combines hydrogen chemically with the coal through the agency of high pressure, and is called direct hydrogenation. With the second process, coal is first converted to water gas, hydrogen is then added, and the mixture is catalytically converted to hydrocarbons and

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[1] Oil and Gas Journal—Vol. 36, No. 33, page 52.

[2] Snider, L. C. and Brooks, B. T.: Bulletin of the American Association of Petroleum Geologists, Vol. 20, No. 1, page 15.

[3] National Petroleum News, Vol. 30, No. 40, page 16 (October 5, 1938).

[4] Dijk, K., and Mulders, K.: Petroleum Times, Vol. 39, No. 995, page 162 (February 1938).

[5] World Petroleum, Vol. 9, No. 2, page 67 (February 1938).

water. This is called indirect hydrogenation, or more often the Fischer-Tropsch process. The former has been more extensively employed in Europe than the latter, and is probably more economical. It produces gasoline having a high knock rating, and is applicable to such materials as bitumen as well as to coal. It can also be used to make benzene, toluene, phenol, and other tar acids from coal if more of these are required than can be supplied from coal tar. However, this process has so far not lent itself successfully to the production of a first rate Diesel fuel or lubricating oil from coal. Further, it is not applicable to all coals. The Fischer-Tropsch process has the advantage that it can be applied to any raw material from which water gas can be made. It also makes a good Diesel fuel, and is a possible means of making lubricating oil. Its principal disadvantage, aside from cost, is that the gasoline fraction has a very low knock rating.

Of these two processes, only direct hydrogenation has been studied experimentally at the Fuel Research Laboratories. This is principally because the applications of indirect hydrogenation to different materials vary only in the method of making water gas, and on that account there is less need for a survey of raw materials.

In this report are presented the results of hydrogenation tests on a series of coals of varying rank, and mostly of Canadian origin. The coals selected originated in widely separated districts from which more than 60 per cent of Canada's coal output is produced. The selection covers a wide range in the classification by rank, so as to indicate the kinds of coal on which further investigations can best be made. A sample of coal processed in the commercial hydrogenation plant of Imperial Chemical Industries at Billingham, England, has been included as a standard of comparison for the Canadian coals, with respect to their suitability to hydrogenation.

The coals selected varied in rank from medium-volatile bituminous to lignite; tests were also made on a representative Canadian peat. It was found that all the fuels could be partly liquefied, but the extent to which they could be converted into valuable liquid and gaseous products varied considerably. The yields of primary oil, for example, were between 44 and 77 per cent of the weight of the coal on the ash-and-moisture-free basis. The variation was mainly accounted for by difference in rank. Two of the Canadian coals gave higher oil yields than the standard English coal.

The hydrogenation investigation herein reported was conducted at the Fuel Research Laboratories by T. E. Warren and K. W. Bowles, who were assisted by other members of the technical staff of the Division. Assistance was also rendered by the plant foreman and machinists in the design and maintenance of the high-pressure reaction chamber and other parts of the testing apparatus which were constructed in the machine shop of the Bureau of Mines. The analyses of the coals, peat, and their solid residual products were carried out in the Coal Analysis Laboratory, and the analytical determinations of the constituents in the ash of the raw fuels tested were made in the Chemical Laboratory of the Metallic Minerals Division. The analyses of the oil and gaseous products were partly performed by analysts in the Oils and Gas Section of the Fuel Research Laboratories. Material assistance was also rendered by R. E. Gilmore,

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Senior Engineer, Division of Fuels, who was consulted from time to time on the selection of coals and other matters.

Acknowledgment is due the following companies who, upon request, provided the coals for the tests: The Dominion Coal Company, Ltd., Sydney, N.S.; The Crow's Nest Pass Coal Company, Ltd., Fernie, B.C.; Canadian Collieries (Dunsmuir), Ltd., Nanaimo, B.C.; Middlesboro Collieries, Ltd., Merritt, B.C.; Alexo Coal Company, Ltd., Alexo, Alta.; Rosedale Collieries, Ltd., Rosedale, Alta.; The Great West Coal Company, Ltd., Edmonton, Alta.; Dominion Briquettes and Chemicals, Ltd., Bienfait, Sask.; and Imperial Chemical Industries, Ltd., Billingham, England.

Ottawa, Ont.
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B. F. Haanel,
Chief, Division of Fuels.

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Tests on the Liquefaction of Canadian Coals by Hydrogenation

CHAPTER I

PREVIOUS WORK AT THE FUEL RESEARCH LABORATORIES

When the work on hydrogenation was begun at the Fuel Research Laboratories the objective was, broadly, to determine what use could be made of the process as applied to Canadian raw materials. It was apparent that a great deal of technical information would have to be obtained in order to reach any well-founded conclusions. There were three available sources of information, namely, the results of research and development elsewhere, theoretical studies, and experimental investigations.

The results of other investigators on the subject are in part available through the published literature and patents, and this information has been supplemented by direct correspondence. Unfortunately, the large commercial organizations associated in the International Hydrogenation Patents Company have published only incomplete descriptions of their processes and have not been willing to give details of small-scale testing equipment. The published literature has, therefore, been the greatest source of outside information.

A bibliography has been compiled on hydrogenation and closely related subjects covering the years from 1914, when Bergius' first patents were taken out, up to the present. Up to the end of 1937, abstracts of about 3,400 references had been classified. It is interesting to note that during the past few years, publications on hydrogenation and related subjects, including patents, have appeared at the rate of more than one per day.

Theoretical studies have not been very fruitful. Both coal and its liquid products are so complex that thermodynamics has only a very limited application. Data on hydrogen requirements are obtainable by the use of material balances, but these involve assumptions regarding the yields of the various products. It was soon apparent, therefore, that most of the desired information would have to be obtained by experimental methods.

At the beginning of the experimental work it was decided to conduct discontinuous tests in a small autoclave[1]. The method of operation was to place the charge in the autoclave, fasten the cover in position, and introduce hydrogen at 1,470 pounds per square inch (one hundred atmospheres) pressure. The autoclave was then heated to reaction temperature, in the neighbourhood of 770° F. (410° C.), at which the pressure was about 3,000 pounds per square inch. The temperature was maintained for an arbitrary period, after which the apparatus was cooled and the product removed. This procedure may be designated as batch operation.

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[1] Warren, T. E.: Mines Branch, Dept. of Mines, Canada, Rept. 725-1.

Alberta bitumen was chosen for the first experimental program because of its future commercial possibilities; also, as it was easier to treat than coal, more general information could be obtained with less effort and time. Three series of tests were made in which were determined the effects of varying, in turn, the temperature, pressure, and duration, in the absence of catalysts. In another series, a number of catalytic materials were tested under comparable conditions.

Using the same apparatus and procedure, experiments were made with tar produced by low-temperature carbonization of Nova Scotia coal. This material was selected largely for the reason that it had some of the chemical properties of coal, but its treatment presented fewer experimental difficulties.[1]

With both raw materials, it was found possible to convert the original charge into a comparatively light oil with no appreciable loss as coke, and with a loss as gas of about 10 per cent. However, the yields of light oil boiling in the gasoline range were low, and it was apparent that recycling or some other form of secondary treatment was necessary.

One disadvantage of the batch method of hydrogenation, as used in this work, was that the hydrogen concentration was reduced as the reaction proceeded. This could only be avoided by charging very small amounts, or by renewing the hydrogen. For this reason the autoclave was next used as a semi-continuous apparatus by passing a stream of hydrogen continuously through the charge which was not renewed. This necessitated the addition of a condenser and high-pressure receiver to remove volatile oil from the stream of hydrogen as it came from the autoclave. This method had the advantages of maintaining the hydrogen at a higher degree of purity than before, and of removing the light oil before secondary cracking could take place. It was first applied to coal tar.[2] Later it was used for experiments on the production of lubricating oil from bitumen, and more recently to make comparative tests on a series of Canadian coals[3].

The principal fault with the semi-continuous technique was that, after a large proportion of the charge had been removed as vapour, a high concentration of solids was left in the bottom of the autoclave, which resulted in poor heat transfer conditions and consequent over-heating and coking of part of the charge. This difficulty can be overcome by using a continuous method of operation. This procedure is used commercially and differs fundamentally from batch operation. For instance, in continuous hydrogenation of coal paste the composition of the reacting material is constant, the catalyst and ash are concentrated, and volatile material is treated for a shorter time than non-volatile material. On the other hand, in batch operation the composition of the reacting material changes continuously throughout the run, the concentrations of catalyst and ash are low, and the material charged is all treated for the same period of time. The semi-continuous procedure corrected these differences only in part. It was thought advisable, therefore, to construct a continuous apparatus in which the conditions of commercial operation could be more closely approached.

The first continuous apparatus has been described in detail in a previous report.[4] The reaction chamber was a short, vertical, 4-litre auto-

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[1] Warren, T. E., and Williams, A. R.: Mines Branch, Dept. of Mines, Canada, Rept. 737-3.
[2] Warren, T. E., and Bowles, K. W.: Mines Branch, Dept. of Mines, Canada, Rept. 737-3.
[3] The results of these later experiments have not been published.
[4] Warren, T. E., and Bowles, K. W.: Op. cit.

clave with a mechanical stirring paddle. A small compressor and a motor-driven pump charged hydrogen and liquid feed respectively to the chamber. The product was removed both directly as liquid and also as vapour in the stream of hydrogen. The hydrogen was recirculated either with a magnetic pump or by means of the compressor.

Tests were made with this equipment on low-temperature coal tar, bitumen, and a paste of bituminous coal in coal tar. The catalyst used was an oxide of molybdenum supported on coke. With all three materials, coke formation was avoided and the total losses, as gas and vapour, were about 10 per cent of the material charged. The yields of distillate oil were about 80 per cent by weight of the net charge. The hydrogen used amounted to about 6 to 7 per cent of the liquid feed.

The results obtained with this apparatus clearly demonstrated the advantages of continuous operation, but the equipment itself, especially the autoclave, gave a great deal of mechanical trouble. For this reason, a new reaction chamber was constructed, a better means of hydrogen recirculation installed, and numerous other changes made.[1] This new equipment has been used for the present tests and will, therefore, be described in detail.

[1] Warren, T. E., and Gilmore, R. E.: Ind. Eng. Chem., Vol. 29, p. 353.

CHAPTER II

DESCRIPTION OF CONTINUOUS PLANT
AND EQUIPMENT USED

The scale of operations of an experimental continuous plant may be varied within wide limits. In general, as the scale of operations is increased the information obtained is more applicable to commercial operations, and is more accurate, especially as regards energy consumption. In the present case, however, selection of a scale on which to begin operations was not entirely a matter of balancing the value of the results against the cost. In such work, it is usually advisable to obtain preliminary information with small-scale equipment before attempting the design of a larger and more expensive plant. Accordingly, the capacity of the new plant was chosen at about 100 pounds of coal per day, or near the lower limit set by the removal of samples and handling losses from the cycle.

In a sequence of operations, such as coal hydrogenation, the reliability of the equipment is important, for the whole plant may be shut down by a minor failure of one unit. This is well illustrated by the following quotation from a report of the (British) Fuel Research Board[1] referring to a Bergius experimental plant: "If a delay occurred which lasted half an hour, due, for example, to trouble with the compressor valves, it was necessary to stop feeding the paste. Then, on attempting to restart the paste pump, it would be found that the paste had set in the pump to an immovable mass, which in turn necessitated dismantling the pump. Meanwhile the stagnant material in the converter often formed a coke mass and jammed the stirrers so that ultimately, as a result of a comparatively trivial failure in one part of the plant, the whole would have to be dismantled, cleared, and a fresh start made." It is apparent that much work is justifiable in order to prevent even a comparatively short delay in any one of the series of mechanical operations. In the detailed description of the plant, concessions to reliability at the expense of mechanical efficiency will be noticed.

The description of the mechanical features of the plant can best be followed by referring to Figure 1.

Hydrogen production is an integral part of the commercial process, but it has not been included in the experimental plant chiefly because personnel for the operation of the generating equipment has not been available. The hydrogen is, therefore, purchased in cylinders.

Hydrogen from the shipping cylinders is measured through a wet meter into a small gas-holder. This is the only measurement of inlet hydrogen, so that any subsequent loss can not be distinguished from the hydrogen combined in the reaction. For this reason reference is made in this report to hydrogen "charged" rather than "combined". From the gas-holder, hydrogen is drawn into a three-stage compressor. It was found that the third-stage valves of the compressor were the cause of frequent interruptions, so that a spare third-stage head, complete with valves, is kept ready for replacement. It is possible to change valve-heads in about 10 minutes. Although it has given much less trouble, the second-stage

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[1] Fuel Research Board Technical Report No. 42, pp. 34–35.
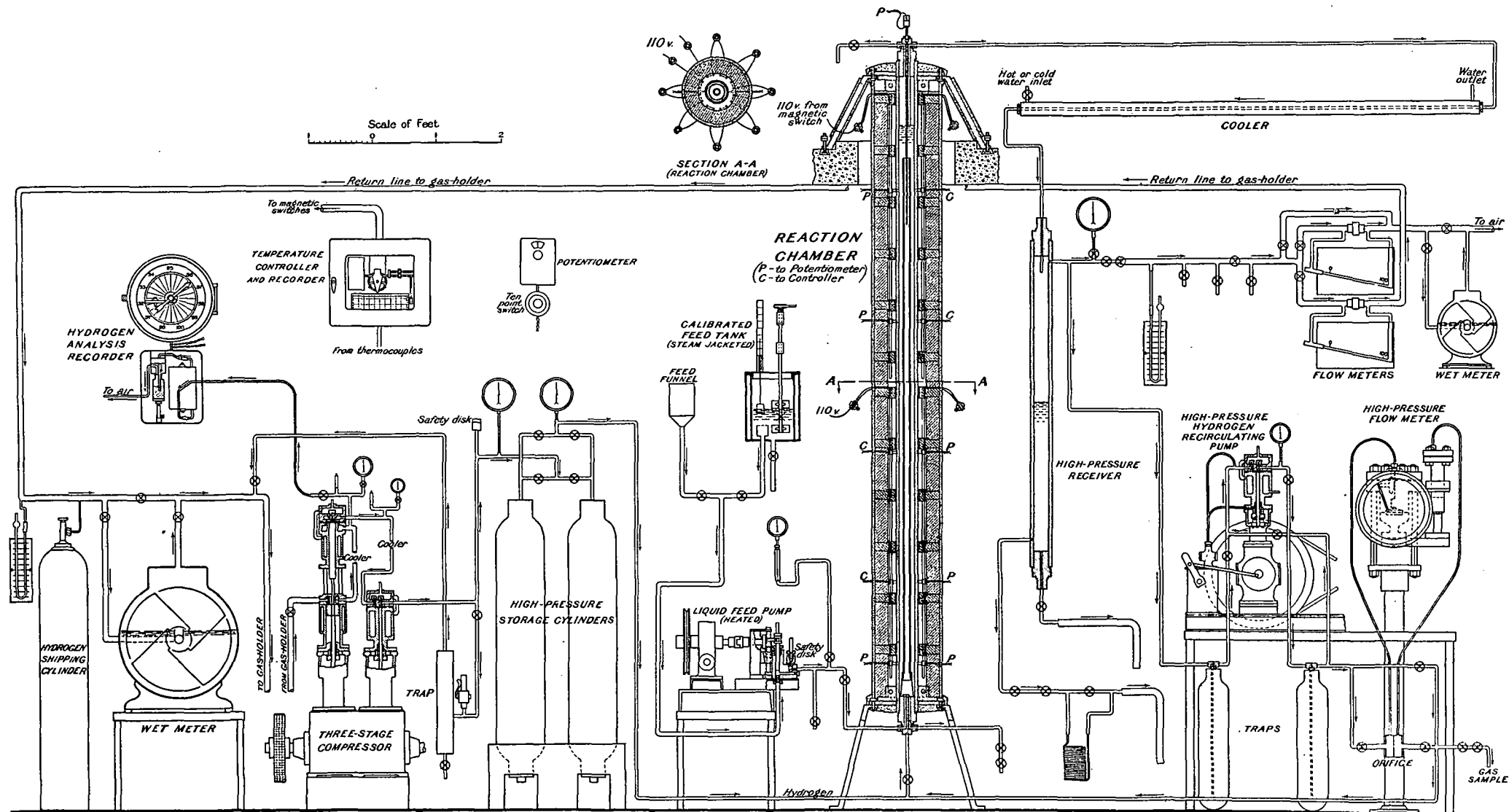
Figure 1.   Hydrogenation plant at the Fuel Research Laboratories.

head has also been duplicated and can be changed in the same way. The compressor is operated by a 5 horse-power motor connected through a variable speed drive. The speed of the compressor can, therefore, be varied so as to maintain a constant pressure. In order to ensure that no appreciable quantity of oxygen or air enters the high-pressure system, a hydrogen analysis recorder has been attached to the inlet of the second-stage of the compressor, and a bell rings when the concentration of hydrogen falls to 95 per cent. Beyond the second stage the hydrogen is under pressure so that there is no possibility of air leaking in. Should there be any oxygen in the cylinder as purchased, it would be detected by the recorder.

Hydrogen from the compressor is stored in two cylinders having a combined volume of about 1·8 cubic feet. During operation these are connected to the high-pressure system, and because of their relatively large capacity they reduce pressure fluctuations.

The production and charging of the paste of powdered coal and oil may be considered as another unit operation in the process. The properties of the paste, especially its viscosity, have an important influence on the performance of the pump. Because of this it was thought advisable, before beginning the program of continuous tests, to study some of the properties of pastes made from coal powdered to different sizes, having different proportions of coal to oil, and suspended in oils of different types.

Most of this work was devoted to measurements of the viscosities of pastes made by mixing coal passing a 200-mesh sieve with four liquid media covering a wide range of viscosity and specific gravity. Starting with the pure medium, coal was added in small increments and the corresponding viscosities were measured until the paste was no longer fluid. It was found that as the percentage of coal was increased the viscosity of the paste at first rose very slowly, but at a point which varied with the different media the viscosity began to rise more rapidly, so that the curve of viscosity plotted against coal percentage finally became almost vertical. The nearly-vertical part of the curve was in the neighbourhood of 50 per cent by weight of coal. These data have some bearing on the selection of a pump. As the viscosity of the paste is low up to about 50 per cent of coal, a simple type of pump is suitable for handling it up to this point. A further increase of 10 per cent in the proportion of coal so increases the viscosity that it would be difficult to charge the paste with any form of injector. It was concluded that for charging pastes of fine coal in a comparatively fluid medium there is very little advantage in using a pump especially designed for handling viscous material.

The liquid feed pump is shown in Figure 1, at the left of the reaction chamber. The body of the pump is made of stainless steel. The check valves are $\frac{7}{16}$-inch balls. The stroke is variable, so that the rate of pumping is adjustable from 0 to about 5 gallons per hour. A frangible disk placed at the high-pressure outlet relieves the pressure in case of an obstruction in the feed line to the reaction chamber, and a pressure gauge on the outlet serves to indicate the pulsations of the pump as well as the pressure on the feed line. At first, difficulty was experienced in using the gauge because it was easily obstructed with paste, but more recently it has been kept clear by injecting one or two cubic centimetres of oil per hour into the gauge line by means of a small hand-operated injector not shown in

Figure 1. It has not been necessary to heat the pump or the line leading from it to the reaction chamber, because the pastes charged up to the present have been sufficiently fluid at room temperature.

The reaction chamber is the most important single unit in the apparatus. Its volume was determined by the required throughput capacity, which is about 100 pounds of coal per day. Previous work indicated that the volume of reaction space required for this throughput would have to be about 600 cubic inches. The shape of the reaction chamber was largely determined by the method of stirring to be employed, by the necessary rate of heat transfer from the external heater, and by the availability and convenience of standard sizes of pipe. Taking these factors into consideration, the most suitable dimensions were considered to be a length of 10 feet, an inside diameter of 2·74 inches, and an outside diameter of 4 inches.

It was desirable to use the reaction chamber at a working pressure of 3,000 pounds per square inch for most of the experimental work, but occasionally pressures up to 5,000 pounds per square inch might be employed. The temperatures to be used were to vary between 752 and 932° F. (400 and 500° C.). The highest temperature to be used, more than any other factor, determined the thickness and the composition of the steel in the reaction chamber, which because of the high temperature had to be designed so as to resist plastic deformation, or "creep". The steel used, therefore, was an austenitic alloy containing 18 per cent of chromium, 8 per cent of nickel, and less than 0·07 per cent of carbon. The purpose of the low-carbon content is to prevent embrittlement due to decarburization by hydrogen. The chamber seems to have been amply strong for the service to which it has been put up to date, because no measureable increase has taken place in the external diameter.

The method of closure of the reaction chamber is based on the principle of the unsupported area, according to which the internal pressure in a vessel is used to force the closure outwards against a wedge-shaped gasket of comparatively small area. The pressure in the gasket is intensified by the ratio of the area of the closure to the projected area of the gasket. A drawing of the closure is shown in Figure 2. The gasket in this case is copper. The design was especially suitable for the present reaction chamber because it was unnecessary to use flanges. As a consequence, heat is dissipated at a comparatively slow rate from the heads, and it has been possible to maintain uniform temperatures throughout the full depth of the reacting charge. The design could, however, be improved by making the contact between the gasket and the wall of the tube slightly conical, so as to facilitate removal of the gasket.

Inside the reaction chamber, and concentric with it, is placed a 1-inch pipe having a funnel-shaped opening at the bottom. This stands on three short legs on the bottom closure of the reaction chamber and extends upward to about 14 inches from the upper closure. The function of this pipe is to stir the reacting liquid. Hydrogen bubbles into the funnel-shaped opening at the bottom from the hydrogen inlet and passes upward through the pipe carrying with it a stream of liquid in the same way as an air-lift pump. The liquid then flows down the annular space outside the pipe and is recirculated. This system of stirring possesses decided advantages in reliability over any mechanically-operated device and has proved very effective in maintaining a uniform temperature throughout the charge.

The composition of the solids carried out of the top of the reaction chamber with the liquid product, however, is not quite identical with that of the solids in the chamber.

Scale of Inches

Section A-A

Section B-B

Sectional elevation
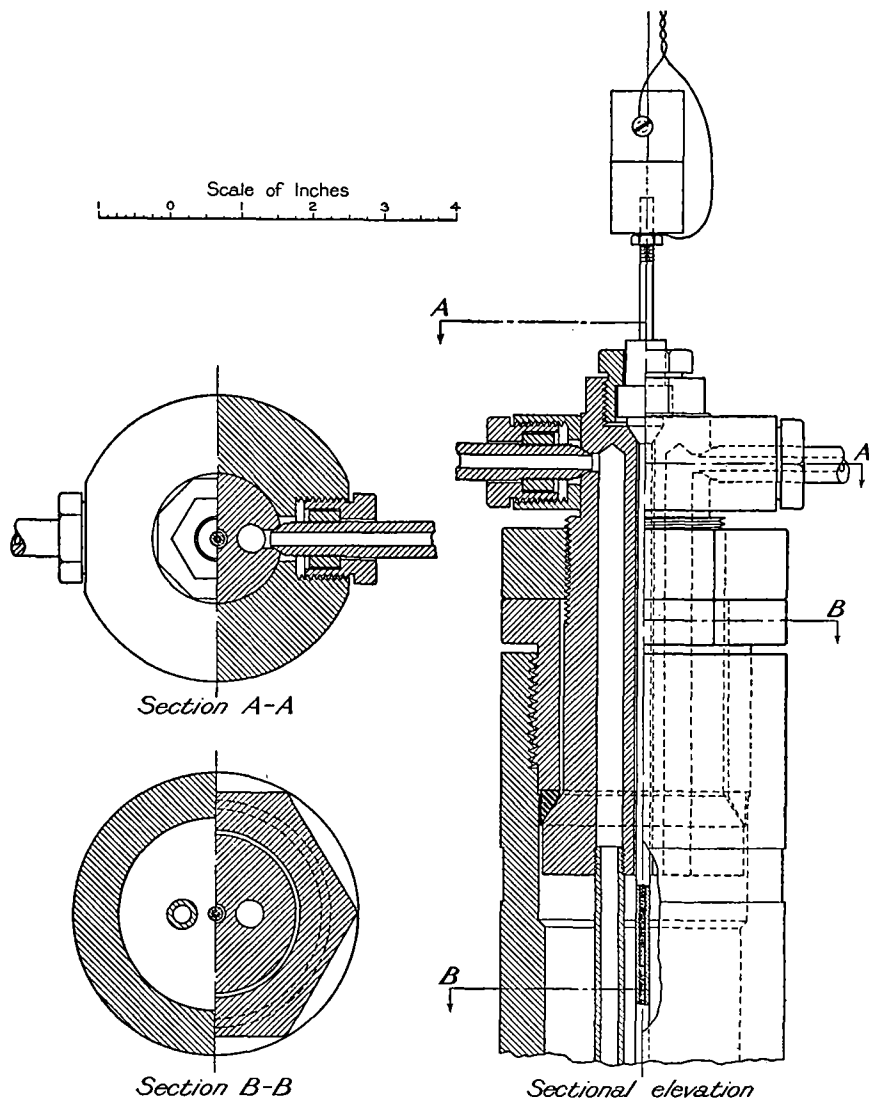
Figure 2.   Closure of reaction chamber.

The outlet from the reaction chamber is a pipe extending downward from the upper closure for about 9 inches.   This leaves a gas space above the upper surface of the liquid that prevents the pressure from rising very suddenly if the outlet line becomes obstructed and the liquid feed pump is

running. Another safety device is the location of check valves inside the bottom closure, to prevent the contents of the reaction chamber from discharging into the room should a break occur in the inlet line from the feed pump or in the hydrogen inlet.

The heater of the reaction chamber is in two parts, on the upper and lower halves of the tube respectively. Each of the heater units consists of a series of vertical loops of nichrome wire radiating directly to the outer wall of the reaction chamber and at a distance of about one inch from it. Outside the heater wires are specially made bricks in which diatomaceous earth is incorporated with the cement. These serve both as a frame and a heat insulator for the unit.

Temperatures of the reaction chamber and its contents are measured at thirteen points by means of iron-constantan thermocouples. Ten of the thermocouples are made by screwing No. 8 gauge iron and constantan wires into holes tapped into the wall of the reaction chamber, iron and constantan leads being used to connect them to the measuring instruments. Three other thermocouples are inserted in a well, approximately 9 feet long, extending down from the upper head of the reaction chamber; these are made of 28-gauge wire, and pass through 6-hole ceramic insulators having an outside diameter of only about $\frac{1}{10}$ inch. The tips of these thermocouples are near the top, middle, and bottom of the reaction chamber respectively. The well is curved so as to be outside the hydrogen lift pipe, the axis of which is the same as that of the reaction chamber. Several modifications of technique have been used in measuring temperatures inside the reaction chamber, but the one just described is the most recent and best. The instruments used for temperature measurements are of the potentiometer type, consisting of a two-point recording controller, an indicator with a multiple switch, and a three-point recorder. The controller is activated by two of the thermocouples tapped into the reaction chamber wall. Although it is the charge within the chamber that is to be maintained at constant temperature, it would be unsafe to control from thermocouples placed inside as the wall of the reaction chamber might become weakened by overheating.

Pressure is one of the important measurements to be made in all of the tests. Direct measurements are made with Bourdon gauges, usually having ranges of 0 to 8,000 pounds per square inch, and placed as shown in Figure 1. A recording gauge is also connected with the high-pressure receiver. The primary standard used for calibration is a piston (dead weight) gauge. The diameter of the piston and the magnitude of the weights were determined in the laboratories of the Bureau of Mines, and the instrument was also calibrated by the makers using measurements made at the United States Bureau of Standards. It has been found that the Bourdon gauges usually require small corrections when received, and that they change with use, sometimes as much as 100 to 200 pounds per square inch.

The equipment for cooling, collecting, and releasing the liquid product has required a considerable amount of development. The cooler is a tube 7 feet long, made from $\frac{1}{4}$-inch extra strong pipe of the 18-8 chrome-nickel alloy. It is surrounded by a water jacket through which hot or cold water may be run. With most coals it has been found necessary to maintain the temperature of the water at about 140° F. (60° C.) in order to prevent

PLATE I
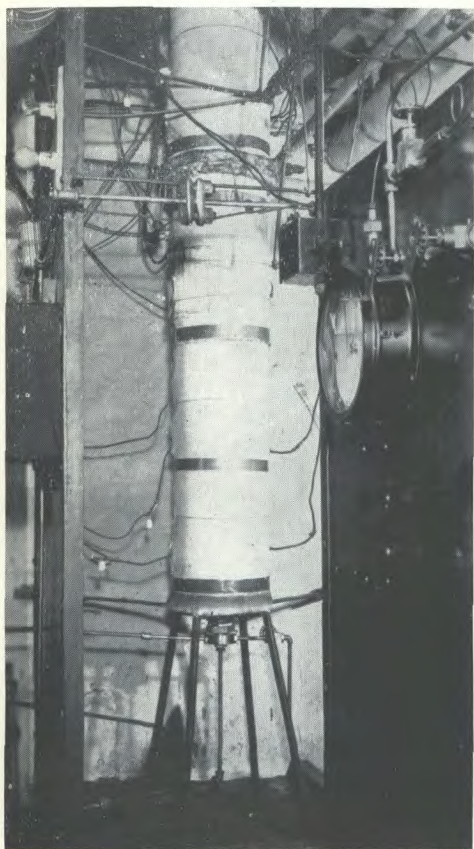
A. Reaction chamber and protecting wall.
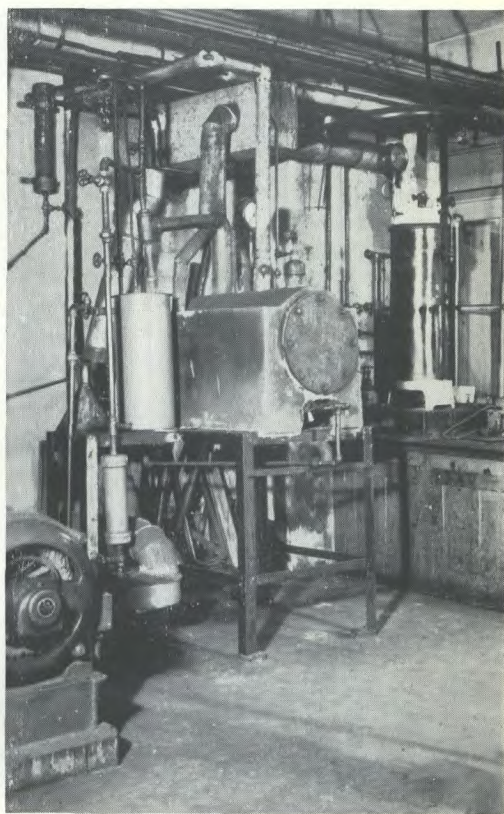
B. Large still used for preparation of vehicle.

PLATE 11
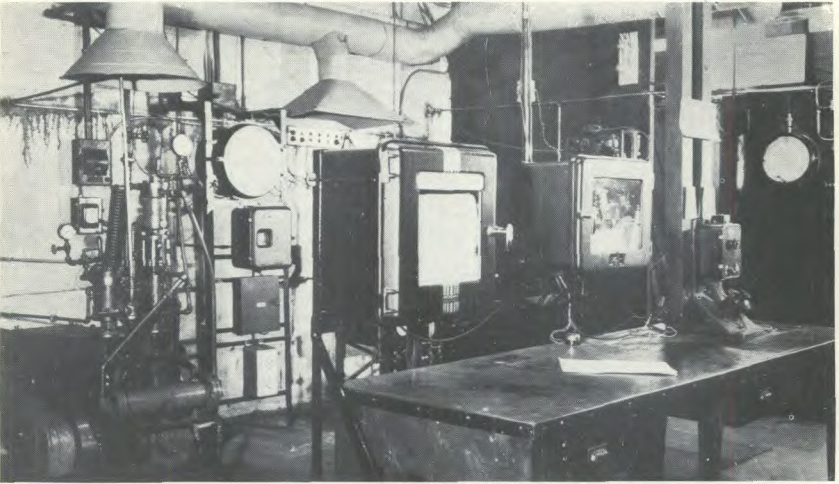
A. View in laboratory showing temperature recording and controlling instruments in right foreground.
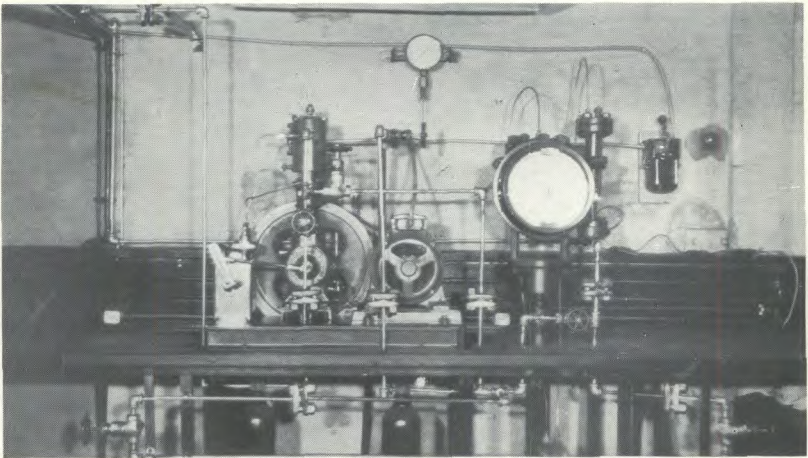
B. Hydrogen recirculating pump and high-pressure flow meter.

pitch from solidifying in the cooler. The high-pressure receiver, into which the cooled products are discharged, is made from tubing of the same composition and having the same diameter as the reaction chamber. It is 5 feet long, and the ends are closed in the same way as those of the reaction chamber. The lower half of the receiver and the outlet valves are heated by a coil of copper tubing through which hot water is circulated.

At first the removal of the liquid product from the high-pressure receiver presented a serious difficulty. The liquid product contains suspended solids which have a strong abrasive action when it flows rapidly through a valve. As the pressure differential across the release valve was 3,000 pounds per square inch the velocity of the liquid product was very high, and consequently the valve seat became so scored that, after only one or two gallons of product had been removed, the flow could not be stopped. This difficulty was finally overcome by placing a coil of tubing in series with the outlet valve, which could then be fully opened and the entire drop in pressure taken up in the coil. The coil is made from copper tubing having an internal diameter of $\frac{1}{16}$ inch and a length of 20 feet. It is kept at 212° F. (100° C.) in a water bath to prevent obstruction by lumps of undissolved pitch, which sometimes are to be found in the liquid product. The reliability of the coil is demonstrated by the fact that it has been used for more than a year without having to remove or clean it.

The hydrogen recirculating system is also an important unit in the continuous plant. Hydrogen passes from the top of the high-pressure receiver through a trap to a recirculating pump which passes it through the orifice of a high-pressure flow meter, whence it enters the bottom of the reaction chamber. The amount of hydrogen circulated in this way is about 420 cubic feet per hour measured at room temperature and atmospheric pressure, which is ten times the amount of make-up hydrogen charged to the gas-holder. It is desirable that the hydrogen be recirculated through the reaction chamber throughout the entire run, including heating and cooling periods, without a shut-down of more than 15 minutes' duration. Accordingly, the smallest mechanical difficulty usually results in a spoiled test.

One source of trouble in this system has been the accumulation of a deposit of ammonium bicarbonate in the line leading from the high-pressure receiver to the recirculating pump and also in the valves of the recirculating pump itself. Fortunately, this material is very soluble in water and can be easily removed.

The recirculating pump is a small one-stage booster compressor. Some alterations were necessary to overcome corrosion. As the valve heads and especially the valve springs were being attacked by the recirculating gas, the valve heads and valves and springs have been made of stainless steel.

A high-pressure orifice meter is used to maintain a constant rate of flow of gas through the reaction chamber. In order to measure the rate of flow of a gas by means of an orifice meter, it is necessary to know the density of the gas. The density of the recirculating hydrogen varies throughout a test because of the accumulation of methane and other gases in it. For some time samples of gas were withdrawn periodically from the recirculating system and their density measured by means of a Schilling apparatus, but more recently a gas density recorder has been installed that records con-

tinuously the density of the gas in the recirculating system. Gas samples are also taken for complete analysis in a Burrell apparatus.

The tubing used in connecting various parts of the equipment is nearly all $\frac{1}{4}$-inch extra strong pipe. This size was selected principally for the reason that it is the smallest in which extra strong fittings are commercially available. Ordinary pipe-thread connections are used with carbon steel pipe wherever the pipe is not heated and does not carry corrosive materials. For hot or corrosive service, 18-8 chrome-nickel pipe in the same size is used. It is difficult, however, to make up tight pipe-thread connections when both pipe and fittings are of this steel, and several other connections have been tried. Where small or flexible connections are needed, copper tubing having a bore of $\frac{1}{16}$-inch and an outside diameter of $\frac{3}{16}$-inch has been used and is connected by means of bronze cones which are tightened against the wall of the tube by conical seats.

In designing such equipment as this, possible risks of injury to the operators and apparatus must be reduced to a minimum. In the present work, the principal danger is the possibility of the formation of hydrogen-air, or hydrogen-oxygen mixtures and their subsequent ignition. Such explosive mixtures might form either in the high-pressure system, where they would be particularly hazardous, or in the room housing the equipment. The hydrogen analysis recorder previously mentioned prevents explosive mixtures from being inadvertently charged to the apparatus.

The forming of hydrogen-air mixtures in the room is not so easy to guard against, but the danger from explosive gas mixtures can be reduced by two diametrically opposed means. One is to maintain small gas flames at a number of points throughout the room so that the explosive mixture will be ignited before any large and dangerous accumulation takes place. The other is to rule out all possible sources of ignition in the room, and this has been the method used in the present work; it is especially effective in conjunction with an efficient ventilating system. In accordance with this principle, a rule is made to allow no smoking, to have no gas flames, to use only induction motors with oil-immersed switches, and to place control switches outside the room. An exhaust fan and air ducts are used for ventilation.

Another possible danger is the rupture of the apparatus, in which case there would be the subsequent danger of burns from hot oil, or of the ignition of oil or of hydrogen. For this reason the hot, high-pressure equipment has been surrounded by a steel wall. The inlet lines to the reaction chamber are protected by check valves inside the lower head of the chamber itself. A large carbon-dioxide fire extinguisher is kept just outside the room. A shower is also conveniently located. Further, as a precaution against rupture of the equipment, each piece of high-pressure apparatus is tested with water at a pressure higher than that to which it will be subjected under working conditions. The compressors are exempted from this rule, but they contain only small volumes under high pressure. All equipment to be subjected to both heat and pressure is made of 18-8 chrome-nickel steel. As previously stated, the temperature of the heated wall of the reaction chamber is measured at ten points to prevent overheating. Closed low-pressure lines, connected through valves to the high-pressure system, are protected by blow-off valves. Valves, which permit only a slow flow of gas and automatically close if a surge occurs, are placed in the lines leading to the gauges.

CHAPTER III

METHOD OF TESTING

In working out a method for conducting experimental tests, it is desirable to follow as closely as possible the procedure of commercial operations. In any commercial coal hydrogenation plant, operations would continue for several weeks or months without a stop. After such a plant had been running for some time, steady conditions would be reached, that is to say, there would be no difference in the composition of samples taken at different times. It is, therefore, necessary to run an experimental plant only until steady conditions have been reached and a further interval over which to take the samples has elapsed. At the beginning of the present work, the time required to reach steady conditions was not even approximately known. Tests were, however, limited to a single shift by the small number of operators available. A shift of 12 to 13 hours was long enough to make a run in which the operating variables, temperature, pressure, rate of charging, etc., were held constant for 6 to 8 hours. If longer tests were required, it was necessary to repeat a number of these 6- to 8-hour runs.

The principal factor involved in reaching steady conditions is that of bringing the reacting pitch in the reaction chamber to a constant analysis. It was found by experiment that the amounts of solid and inorganic material in the pitch were substantially constant after one 6- to 8-hour period. Subsequent runs have, therefore, been taken as representative of continuous operation over an indefinitely extended time. It is not certain, however, that progressive changes have not been taking place in other properties of the reacting pitch at a rate too slow to be observed in the present tests. It is, therefore, desirable to make longer runs to determine the time required to reach a completely steady state.

The usual procedure employed in making a hydrogenation test on a coal is as follows.

A quantity of 200 pounds, or more, of the coal is crushed in a rotary crusher to a maximum size of about ¼-inch. A sample is taken, and the ash and moisture contents are determined.

If the ash content be more than 5 per cent a float and sink test is made to determine the extent to which it can be reduced by washing. Data from this test can be interpreted in terms of commercial-scale washing practice.[1] If the test shows that the ash can be substantially reduced, the bulk sample for hydrogenation is separated in a liquid of suitable specific gravity. Usually the liquid used is a mixture of carbon tetrachloride and gasoline. After washing, the coal is air-dried to allow the washing liquid to evaporate.

If the moisture content of the coal is high the washed bulk sample is dried by heating it in an oven at 230° F. (110° C.) for a period of two or three days. During this time the coal is stirred frequently. The oven is internally fired with gas.

The cleaned and dried coal is passed through rolls which reduce its maximum size to about ⅛ inch. A sample is taken for a grindability test

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[1] Bird, B. M.: Second International Conference on Bituminous Coal, Vol. II, p. 82, Carnegie Inst. of Technology, Pittsburgh, Pa.

which indicates the amount of grinding necessary.[1] The bulk sample is pulverized in steel ball mills and, after mixing, is stored in covered cans. Just prior to mixing with the vehicle, a sample is taken for proximate, ultimate, sieve, and ash analyses, and for determination of the calorific value.

About 30 pounds of the prepared coal is mixed with an equal weight of oil, usually the fraction of the product from the preceding run boiling above 446° F. (230° C.). The catalyst, 5 per cent of stannous oxide, based on the weight of the coal, including ash and moisture, is incorporated in the paste of coal and oil.

The pitch, which was removed from the reaction chamber at the end of the previous run, is weighed and diluted with benzol until it is fluid enough to be pumped. Hydrogen is compressed into the system to a pressure of 3,000 pounds per square inch. The heaters are then turned on, and while the reaction chamber is heating the pitch and benzol are pumped into it. Hydrogen is recirculated through the reaction chamber, and the benzol is distilled into the high-pressure receiver, whence it is recovered and weighed. This procedure is followed because it is necessary to analyse the pitch in order to know if progressive changes are taking place in its composition, and in such short runs a small change in the weight of the pitch or in its content of insoluble material makes an appreciable difference in the yields. The cold undiluted pitch is usually a brittle solid, and owing to settling it does not have the same composition throughout the reaction chamber. It therefore has to be melted and run out, both for the purpose of weighing it accurately and of obtaining a representative sample for analysis. It is not possible, with the present pump, to reintroduce it into the reaction chamber in the hot molten state, and the simplest alternative is to add about half its weight of benzol as a diluent. As the benzol is almost all recovered and is entirely accounted for, this procedure does not affect the yields.

As soon as the benzol has all been removed, readings are taken of the gauges and meters that indicate the amount of hydrogen in the system. This is for the purpose of making a hydrogen balance. Subsequently, all the hydrogen introduced into the system is measured through the inlet meter, and a similar determination of the hydrogen in the system is made at the end of the run, when the temperature is below 662° F. (350° C.). The temperature, when the hydrogen balance is begun, is usually between 662 and 752° F. (350 and 400° C.), and as there is little or no hydrogen absorption below this temperature substantially all the reacting hydrogen is included in the balance. Some hydrogen is, however, lost by leaks around the packings of the compressors, by sampling for the hydrogen analysis recorder, by solution in the liquid product, and by surging through the liquid product valve when the level of the product in the high-pressure receiver falls below the outlet connection.

After the benzol has been removed and the gauges and meters read, the reaction chamber is allowed to heat as quickly as possible to reaction temperature. About $3\frac{1}{2}$ hours are required to heat the reaction chamber from room temperature to 824° F. (440° C.). Throughout the run, the temperature of the wall of the reaction chamber is usually controlled to within plus or minus 9° F. (5° C.) of this value. The temperature of the

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[1] A.S.T.M. Designation D 409-35 T (Hardgrove-Machine Method).

pitch in the reaction chamber fluctuates, owing to intermittent charging, 7° F. (4° C.) above and below a mean, which is usually about 36° F. (20° C.) lower than the temperature of the wall of the chamber. The temperature of the reaction chamber wall, as recorded, is the mean of the readings of thermocouples Nos. 4 and 8, taken at intervals of one hour throughout the run. These thermocouples are located at the points, marked "P", lying nearest to the centre of the chamber. (*See* Figure 1.)

The pressure throughout the run is kept constant, usually at 3,000 pounds per square inch, by adjusting the speed of the compressor. The withdrawal of the product, in batches of about one litre, at 20-minute intervals, causes temporary fluctuations of about 50 pounds per square inch in the system.

The rate of charging is maintained at about 9 pounds per hour by dividing the hourly charge into three equal quantities, of about one litre each, and charging them at intervals of 20 minutes. The rate of the feed pump is adjusted so that only about 10 minutes is required to charge one litre. The feed pump is thus idle long enough to make any necessary adjustments without interfering with the average rate of charging.

The fourth operating variable of importance is the rate of recirculation of hydrogen. Variations in this rate affect the degree of stirring in the chamber and probably also the rate of solution of hydrogen in the pitch. The rate is measured by means of a high-pressure flow meter, which indicates the pressure drop across an orifice through which the recirculating hydrogen flows. As mentioned previously, this pressure differential is a function both of the rate of flow of the gas and of its density, and a sample is continuously removed from the recirculating system and its density recorded. From the density of the gas the desired pressure differential is calculated, and the rate of flow is adjusted by means of a by-pass valve until this pressure differential is indicated by the flow meter. The rate of flow, so maintained, is usually 5 cubic feet per hour calculated at the conditions of reaction, namely, 3,000 pounds per square inch and about 788° F. (420° C.). The recirculating hydrogen is sampled for analysis at the beginning and end of the period over which the hydrogen balance is made, at the beginning and end of the charging period, and 3 hours after charging is started. It contains increasing amounts of hydrocarbons and oxides of carbon as the run progresses, especially when low-rank coals are being tested.

When all of the paste prepared for the run has been charged, the heating current is turned off and the rate of hydrogen recirculation is increased in order to cool the reaction chamber as rapidly as possible to 662° F. (350° C.). At this, or a lower temperature, the meters and gauges are again read for the purpose of making the hydrogen balance. The hydrogen is then released from the reaction chamber, while the pitch is still fluid. If the hydrogen be removed when the pitch is cool, some of it remains dissolved in the semi-solid and causes frothing. On the other hand, the temperature at which the pressure is released must not be too high, or a considerable proportion of the pitch may be carried away as vapour. Usually on the day following a run, the pitch is again heated and taken out of the reaction chamber. It is then weighed, stirred, sampled, and diluted with benzene in preparation for the subsequent run.

Samples of the liquid product and of the pitch from the reaction chamber are extracted in a Soxhlet apparatus with carbon tetrachloride

to determine the total amount of oil they contain, including material not ordinarily liquid but soluble in carbon tetrachloride. Ash is determined in the residue after extraction. A sample of the liquid product is also distilled in a Hempel apparatus[1] to determine the amount of water in the product and also the boiling range of the fractions up to 572° F. (300°C.). The liquid products have not been subjected to an extensive chemical study, although this would probably be a profitable field for investigation.

The liquid product is distilled in a large gas-fired iron still without rectification, in order to produce the vehicle for the subsequent run. Fractions up to 338° F. (170° C.) and between 338 and 446° F. (170 and 230° C.) are removed and the remainder of the liquid is distilled down to coke, using steam to carry off the heaviest fraction. All the oil recovered above 446° F. (230° C.) is used as vehicle. This method was adopted at the beginning of the present survey of Canadian coals and was, therefore, used until the series of tests was completed. Its principal fault is that the highest boiling fractions of the oil are cracked to coke in the still because of over-heating at the end of the distillation period. Because of the loss of this considerable quantity of oil, the vehicle has to be augmented by including fractions having a lower boiling range than those that would be used in commercial practice. The large still has no rectifying column, so that a considerable proportion of the vehicle boils below 446° F. (230° C.) in the Hempel apparatus.

Two other methods of removing solids from the vehicle, namely filtering and centrifuging, have recently been tested. The product of either of these operations is distilled to remove the fractions boiling up to 446° F. (230° C.), and the residuum is used as vehicle. There is no loss as coke in these operations because the distillation temperatures are low.

A sample of the vehicle is distilled in the Hempel apparatus to determine its boiling range and, recently, determinations of the viscosity of the vehicle have been made so as to have data on the effect of this variable on the behaviour of the paste pump. When the vehicle has been prepared by filtering or centrifuging the liquid product it also contains small amounts of solids, which are determined by extraction in a Soxhlet apparatus with carbon tetrachloride.

After the analytical results have been obtained, a table is prepared in which the in-going materials are classified under the headings: oil, ash-and-moisture-free coal, hydrogen, inorganic material, combustible solid material, water, and gas. The materials so classified are: vehicle, coal, catalyst, pitch, and hydrogen. The out-going materials are classified under the same headings, and include: liquid product, pitch, and gas plus loss. Distillation loss is considered as gas. A balance is struck between the in-going and out-going quantities under these various heads, which are then reduced to percentages of the ash-and-moisture-free coal. The increase of outlet over inlet oil, according to this system, is the net yield of oil. In other words, it is the yield which would be obtained in continued operation if no outside source of oil were available. This is also true of the yields of combustible solid and water. The yields of gas, as given in the table however, are not representative of the overall yield that would be obtained in commercial practice, because on a commercial scale

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[1] A.S.T.M. Designation D 285-36.

the greater part of the gas produced by the reaction would be used for the manufacture of hydrogen and so would not be available for other purposes. The quantities given in the tables are not more accurate than the analyses, and are therefore correct to only three significant figures. Additional figures have been retained in the tables to facilitate checking the calculations by addition.

The inorganic materials charged and removed do not always balance, and although small discrepancies may be accounted for by errors in analysis, it is probable that in some cases inorganic material is lost. As the yields are reported as per cent of the ash-and-moisture-free coal, the sum of the yields is not exactly 100 plus the percentage of hydrogen charged, unless the amount of inorganic materials charged is equal to the amount removed. The values under the heading "mean" are weighted in proportion to the durations of the runs indicated.

CHAPTER IV

HIGH-VOLATILE A BITUMINOUS COAL FROM SYDNEY DISTRICT, NOVA SCOTIA

The Sydney coal was selected for a comparative hydrogenation test because it is representative of the High-volatile A group of the bituminous class of the A.S.T.M. classification; also the production from this district is greater than from any other in Canada.

DESCRIPTION AND PRE-TREATMENT OF SAMPLE

The sample used for the test was produced in the Princess mine. It was slack coal which had been stored for some time at the Fuel Research Laboratories.

The ash content had been reduced by washing before shipment from 5·0 to 3·1 per cent on the dry basis, and further washing was not considered necessary.

The bulk sample was pulverized in a paddle-type pulverizer in two batches. The first of these was used up to and including Run 4, and the second for all the others. The size distribution of the first batch is as follows:

Sieve Analysis

U.S. Series Equivalent No.	Per cent
Through 16 on 30 mesh	0·5
" 30 " 50 "	3·2
" 50 " 100 "	16·7
" 100 " 200 "	25·4
" 200 mesh	54·2
Total	100·0

After pulverizing, the first batch contained 2·1 per cent of moisture and 3·0 per cent of ash, and the second 2·4 per cent of moisture and 3·3 per cent of ash. The complete analyses of the first batch are given below.

	As used in tests	Dry basis	Ash-and-moisture-free basis
Proximate Analysis—			
Moisture .... %	2·1		
Ash .... %	3·0	3·1	
Volatile matter .... %	37·3	38·1	39·3
Fixed carbon .... %	57·6	58·8	60·7
Ultimate Analysis:			
Carbon .... %	79·9	81·6	84·2
Hydrogen .... %	5·7	5·6	5·8
Ash .... %	3·0	3·1	
Sulphur .... %	1·1	1·1	1·1
Nitrogen .... %	1·7	1·7	1·8
Oxygen .... %	8·6	6·9	7·1
Calorific Value—			
B.T.U. per pound, gross	14,370	14,680	15,150

The analysis of the ash in the coal as charged is as follows:

	Per cent
Ignition loss at 1292° F. (700° C.)	2·29
SiO₂	25·76
Al₂O₃	17·72
Fe₂O₃	35·44
TiO₂	0·86
CaO	6·65
MgO	0·94
Na₂O	4·09
K₂O	0·49
SO₃	4·77
MnO	0·20
P₂O₅	0·44
Total	99·65

HYDROGENATION TEST

The hydrogenation test consisted of fifteen runs. The first three of these were made under standard conditions and are comparable to the tests on the other coals. Runs 4 to 9 were devoted to a brief investigation of the pressure, temperature, and rate variables. Runs 10 to 14 were used to investigate the effect of catalysts, and Run 15 was made to demonstrate the method of operation. For convenience in description, the runs have been divided into three groups comprising Runs 1 to 3, 4 to 9, and 10 to 15.

Runs 1 to 3: Standard Test

Characteristics of Materials Charged. The coal as charged has been described.

The vehicle used in Run 1 was a fraction of high-temperature tar boiling above 446° F. (230° C.). The vehicle for Run 2 was the residuum after topping the product of Run 1 at 446° F. (230° C.). This residuum contained 0·85 per cent of solids, which were assumed to be one-half ash. The vehicle for Run 3 was the fraction of the product of Run 2 boiling above 446° F. (230° C.) in the large still with steam.

The catalyst in each run was stannous oxide, in amount equal to 5 per cent of the coal charged.

No pitch was charged in Run 1. While the reaction chamber was being heated part of the paste was pumped into it. In Runs 2 and 3 the pitch was produced, in each case, in the preceding run.

Operating Conditions. The temperature, pressure, charging rate, and duration of Runs 2 and 3 are shown below. In Run 1, the "hydrogen lift" was not used, and the temperature was not so uniform throughout the chamber as in the other runs. The rate of hydrogen recirculation varied

in each run, with varying density between 4 and 9 cubic feet per hour at the temperature and pressure in the reaction chamber. At the beginning of Run 3, the recirculating gas contained 93·4 per cent of hydrogen, and at the end 84·5 per cent. Gas was discharged from the recirculating system at an average rate of 34·0 cubic feet per hour.

—		Run 2	Run 3
Average wall temperature, thermocouples 4 and 8	°F.	831	829
	°C.	444	443
Pressure, lb./sq. in.		2940	2940
Average charging rate, lb. paste/hr.		9·3	8·8
Duration of run, hr.		5·33	7·45

Characteristics and Quantities of Materials Produced. The results of the analytical distillations of the liquid products of Runs 2 and 3 in the Hempel apparatus are:

—	Per cent by weight	
	Run 2	Run 3
Water	4·3	4·8
Up to 338° F. (170° C.)	4·9	4·3
338 to 446° F. (170 to 230° C.)	22·5	17·0
446 to 572° F. (230 to 300° C.)	} 67·5 {	36·4
Residuum		37·0
Loss	0·8	0·5

The results of extraction of the liquid products and pitches of Runs 2 and 3 with carbon tetrachloride, and the determination of ash in the solid residues are given below:

—	Liquid product		Pitch	
	Run 2	Run 3	Run 2	Run 3
Solid insoluble in CCl₄ .... %	3·88	6·36	31·3	32·6
Ash in insoluble solids .... %	59·5	57·9	50·2	52·0

The material balances of Runs 2 and 3 are given in Table I. In order to complete the balance for Run 2, it has been assumed that the hydrogen input was the same as in Run 3, that the input and output of inorganic material were equal, and that half the solid material in the vehicle was inorganic.

TABLE I

Material Balance, Quantities in Grammes

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total

Run 2

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Vehicle	10,863			47	47			10,957
Additional tar	942							942
Coal		10,398		329		230		10,957
Catalyst				548				548
Pitch	6,660			1,009	1,188			8,857
Hydrogen			832					832
Total input	18,465	10,398	832	1,933	1,235	230		33,093
Liquid product	20,203			513	349	961	178	22,204
Pitch	6,211			1,420	1,410			9,041
Recovery								31,245
Gas plus loss							1,848	1,848
Total output	26,414			1,933	1,759	961	2,026	33,093
Increase	7,949			0	524	731	2,026	11,230
Per cent increase	76·45				5·04	7·03	19·48	108·00
Decrease		10,398	832					11,230
Per cent decrease		100·00	8·00	0·00				108·00

Run 3

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Vehicle	14,560							14,560
Coal		13,817		437		306		14,560
Catalyst				728				728
Pitch	5,912			1,352	1,341			8,605
Hydrogen			1,110					1,110
Total input	20,472	13,817	1,110	2,517	1,341	306		39,563
Liquid product	24,962			1,040	756	1,369	132	28,259
Pitch	6,244			1,571	1,450			9,265
Recovery								37,524
Gas plus loss							2,039	2,039
Total output	31,206			2,611	2,206	1,369	2,171	39,563
Increase	10,734			94	865	1,063	2,171	14,927
Per cent increase	77·69			0·68	6·26	7·693	15·712	108·03
Decrease		13,817	1,110					14,927
Per cent decrease		100·00	8·03					108·03

*A-M-F coal: ash-and-moisture-free coal.

The yields as per cent of the ash-and-moisture-free coal are as follows:

—	Run 2	Run 3	Mean of Runs 2 and 3
Oil.. %	76·5	77·7	77·2
Combustible solid.. %	5·0	6·2	5·7
Water.. %	7·0	7·7	7·4
Gas.. %	19·5	15·7	17·3
Hydrogen charged.. %	8·0	8·0	8·0

Runs 4 to 9: Variation of Pressure, Rate of Charging, and Temperature

The object of Runs 4 to 9 was to determine the influence of altering the operating variables: temperature, pressure, and rate of charging. It was not attempted to reach steady conditions after each change in an operating variable, but rather to note the trend of the results after a single run. Accordingly, starting from the standard conditions of Run 3, Run 4 was made at a lower pressure. Run 5 was made at standard conditions again to restore the pitch to its normal state, and Run 6 was made at a slow rate of charging. Run 7 was a normalizing run at ordinary conditions, and Run 9 was made at a high temperature, Run 8 having been spoiled by a mechanical failure.

Characteristics of Materials Charged. The coal as charged has been described.

The vehicles for Runs 5, 6, and 9 were the fractions of the preceding runs boiling above 446° F. (230° C.) in the large still with steam. The vehicle for Run 4 was produced by distillation of the product of Run 3 to coke without steam, discarding the products boiling below 446° F. (230° C.). The vehicle for Run 7 was produced by filtering the product of Run 6 and removing the fraction of the filtrate boiling below 446° F. (230° C.) in the large still.

The catalyst in each run was stannous oxide, in amount equal to 5 per cent of the coal charged.

The pitch charged at the beginning of the run was, in each case, produced in the preceding run. The proportions of solids insoluble in carbon tetrachloride in the pitches, and of ash in the insoluble solids are given below:

—	Run 4	Run 5	Run 6	Run 7	Run 8	Run 9
Solids insoluble in CCl₄.................. %	32·6	30·8	35·3	50·9	33·1	33·9
Ash in insoluble solids.................. %	52·0	56·4	49·1	40·8	45·8	45·9

Operating Conditions. The temperature, pressure, charging rate, and duration of Runs 4, 6, 7, and 9 are given below. As in Runs 1 to 3, the rate of recirculation varied with density between 4 and 9 cubic feet per

hour at the conditions in the reaction chamber. Gas was discharged from the recirculating system in Runs 4, 5, and 6 at the average rate of 38·5, 18·8, and 16·3 cubic feet per hour respectively. In later runs no gas was discharged because this means of maintaining a high concentration of hydrogen in the recirculating gas was not effective.

—		Run 4	Run 6	Run 7	Run 9
Average wall temperature, thermocouples 4 and 8	°F.	833	831	832	857
	°C.	445	444	444	458
Pressure, lb./sq. in.		2430	2940	2940	2940
Average charging rate, lb. paste/hr.		9·0	6·0	8·9	8·7
Duration of run, hr.		6·8	8·0	6·5	7·8

The proportions of hydrogen in the gas recirculating through the reaction chamber at the beginning and end of the charging periods of the runs were as follows:

—	Run 4	Run 5	Run 6	Run 7	Run 9
Beginning of charging period	97·3*	98·3	94·9	96·2	92·1
End of charging period	85·1	86·7	82·8	78·7	68·2

*In Run 4 the hydrogen samples were taken during the heating and cooling periods.

Characteristics and Quantities of Materials Produced. The results of the analytical distillations of the liquid products in the Hempel apparatus are as follows:

—	Per cent by weight			
	Run 4	Run 6	Run 7	Run 9
Water	5·0	5·7	4·8	5·6
Up to 338° F. (170° C.)	5·9	7·3	4·4	6·9
338 to 446° F. (170 to 230° C.)	15·8	15·8	11·9	13·6
446 to 572° F. (230 to 300° C.)	29·8	26·9	29·6	27·9
Residuum	42·6	43·3	48·4	45·5
Loss	0·9	1·0	0·9	0·5

The results of extraction of the liquid products and pitches with carbon tetrachloride, and of the determination of ash in the solid residues are given below:

—	Liquid product				Pitch			
	Run 4	Run 6	Run 7	Run 9	Run 4	Run 6	Run 7	Run 9
Solids insoluble in CCl₄. %	5·76	3·90	13·62	3·56	30·8	50·9	33·1	53·58
Ash in insoluble solids.. %	56·6	46·5	49·6	41·9	56·4	40·8	45·8	36·3

The material balances are given in Table II.

TABLE II

Material Balance, Quantities in Grammes

—	Oil	A-M-F coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Run 4								
Vehicle.....................	13,443	....	....	403	....	282	....	13,443
Coal........................	....	12,758	....	403	....	282	....	13,443
Catalyst....................	....	....	....	672	....	....	....	672
Pitch.......................	6,038	....	....	1,518	1,401	....	....	8,957
Hydrogen...................	....	....	909	....	....	....	....	909
Total input............	19,481	12,758	909	2,593	1,401	282	....	37,424
Liquid product..............	21,764	....	....	803	616	1,227	219	24,629
Pitch.......................	6,964	....	....	1,745	1,349	....	....	10,058
Recovery...................							....	34,687
Gas plus loss...............							2,736	2,736
Total output............	28,728	....	....	2,548	1,965	1,227	2,955	37,423
Increase....................	9,247	....	....	....	564	945	2,955	13,711
Per cent increase...........	72·48	....	....	....	4·42	7·41	23·16	107·47
Decrease....................	....	12,758	909	45	....	....	....	13,712
Per cent decrease...........	....	100·00	7·12	0·35	....	....	....	107·47
Run 6								
Vehicle.....................	10,657	....	....	351	....	256	....	10,657
Coal........................	....	10,050	....	351	....	256	....	10,657
Catalyst....................	....	....	....	532	....	....	....	532
Pitch.......................	6,612	....	....	1,771	1,836	....	....	10,219
Hydrogen...................	....	....	1,063	....	....	....	....	1,063
Total input............	17,269	10,050	1,063	2,654	1,836	256	....	33,128
Liquid product..............	17,476	....	....	355	408	1,109	201	19,549
Pitch.......................	5,189	....	....	2,191	3,186	....	....	10,566
Recovery...................							....	30,115
Gas plus loss...............							3,013	3,013
Total output............	22,665	....	....	2,546	3,594	1,109	3,214	33,128
Increase....................	5,396	....	....	....	1,758	853	3,214	11,221
Per cent increase...........	53·69	....	....	....	17·49	8·49	31·98	111·65
Decrease....................	....	10,050	1,063	108	....	....	....	11,221
Per cent decrease...........	....	100·00	10·58	1·07	....	....	....	111·65

TABLE II—*Concluded*

Material Balance, Quantities in Grammes—*Concluded*

—	Oil	A-M-F coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Run 7								
Vehicle	12,811							12,811
Coal		12,081		423		307		12,811
Catalyst				641				641
Pitch	4,984			2,103	3,058			10,145
Hydrogen			732					732
Total input	17,795	12,081	732	3,167	3,058	307		37,140
Liquid product	19,940			1,670	1,697	1,187	227	24,721
Pitch	6,674			1,508	1,788			9,970
Recovery								34,691
Gas plus loss							2,449	2,449
Total output	26,614			3,178	3,485	1,187	2,676	37,140
Increase	8,819			11	427	880	2,676	12,813
Per cent increase	73·00			0·09	3·53	7·28	22·15	106·05
Decrease		12,081	732					12,813
Per cent decrease		100·00	6·06					106·06
Run 9								
Vehicle	15,079							15,079
Coal		14,219		498		362		15,079
Catalyst				754				754
Pitch	6,198			1,459	1,720			9,377
Hydrogen			1,175					1,175
Total input	21,277	14,219	1,175	2,711	1,720	362		41,464
Liquid product	24,292			401	556	1,492	134	26,875
Pitch	4,792			2,008	3,522			10,322
Recovery								37,197
Gas plus loss							4,267	4,267
Total output	29,084			2,409	4,078	1,492	4,401	41,464
Increase	7,807				2,358	1,130	4,401	15,696
Per cent increase	54·91				16·58	7·95	30·95	110·39
Decrease		14,219	1,175	302				15,696
Per cent decrease		100·00	8·26	2·12				110·38

The yields as per cent of the ash-and-moisture-free coal are as follows:

—		Run 4	Run 6	Run 7	Run 9
Oil	%	72·5	53·7	73·0	54·9
Combustible solid	%	4·4	17·5	3·5	16·6
Water	%	7·4	8·5	7·3	8·0
Gas	%	23·2	32·0	22·2	31·0
Hydrogen charged	%	7·1	10	6·1	8·3

These experiments do not indicate any advantage in operating at conditions other than those which were used for Runs 1 to 3. However, these are not necessarily optimal as further experimentation may demonstrate.

Runs 10 to 15: Variation of Catalyst

Characteristics of Materials Charged. The coal as charged has been described.

With the exception of Run 15, all the vehicles were, as usual, the fractions of the products of the preceding runs boiling above 446° F. (230° C.) in the large still with steam. The vehicle for Run 15 was produced from a high-temperature tar oil which was topped at 473° F. (245° C.).

No catalyst was added to the coal paste in Runs 10 and 11. In Runs 12, 13, and 14, lead acetate, $Pb(C_2H_3O_2)_2 . 3H_2O$, in amount equal to 5 per cent of the coal charged, was added to the coal paste. The catalyst for Run 15 was, as usual, 5 per cent of stannous oxide.

The pitch for Run 10 was produced in Run 9 and was diluted with benzol, filtered to remove the previous catalyst, and added to a fraction of high-temperature tar oil, which had been topped at 482° F. (250° C.). The mixture was topped at 527° F. (275° C.) to remove light oil. The pitch so produced contained 1·5 per cent of solid and 0·029 per cent of tin. Part of the pitch from Run 12 was lost, and part of the vehicle not used in Run 11 was topped at 554° F. (290° C.), and the residuum added to make up the pitch requirement for Run 13. The pitch for Run 15 was made by adding a high-temperature tar fraction topped at 473° F. (245° C.) to part of the pitch from the third run with Crowsnest coal. The pitches for Runs 11, 12, and 14 were, as usual, produced entirely in the preceding runs.

Operating Conditions. Run 12 was stopped when a valve on the outlet line from the high-pressure receiver could not be closed. The temperature, pressure, charging rate, and duration of the other runs are shown below:

—		Run 10	Run 11	Run 13	Run 14	Run 15
Average wall temperature, thermocouples 4 and 8	°F.	828	842	833	837	817
	°C.	442	450	445	447	436
Pressure, lb./sq. in.		2940	2940	2940	2940	2940
Average charging rate, lb. paste/hr.		8·4	9·5	9·1	9·5	10·1
Rate of hydrogen recirculation, at temperature and pressure of reaction chamber, cu. ft./hr.		5	5	5	5	......
Duration of run, hr.		7·7	6·5	5·6	6·7	6·2

The analyses of samples of the gas recirculating through the reaction chamber at the beginning and end of the charging period of each run are as follows:

	Run 10		Run 11		Run 13		Run 14	
	Start	End	Start	End	Start	End	Start	End
Carbon dioxide......... %	0·2	0·4	0·3	0·6	0·2	0·4	0·5	0·6
Oxygen................. %	0·3	0·4	0·4	0·4	0·2	0·3	0·2	0·3
Illuminants............ %	0·2	0·4	0·2	0·6	0·2	0·4	0·2	0·5
Carbon monoxide....... %	1·0	2·4	1·3	2·6	0·9	2·3	0·9	2·6
Hydrogen.............. %	89·6	64·2	88·8	63·3	96·1	68·3	92·5	62·1
Methane............... %	5·3	20·9	4·3	19·7	0·7	16·5	3·3	20·3
Ethane................ %	0·5	8·1	1·4	8·6	0·1	8·4	0·8	10·2
Nitrogen (by difference) %	2·9	3·2	3·3	4·2	1·6	3·4	1·6	3·4

Characteristics and Quantities of Materials Produced. The results of the analytical distillations of the liquid products in the Hempel apparatus are given below:

	Per cent by weight				
	Run 10	Run 11	Run 13	Run 14	Run 15
Water........	5·2	5·2	5·0	6·0	3·2
Up to 338° F. (170° C.)........	4·1	5·5	5·6	5·8	9·7
338 to 446° F. (170 to 230° C.)........	13·2	14·4	12·7	13·0	21·0
446 to 572° F. (230 to 300° C.)........	35·8	30·4	31·1	28·0	32·9
Residuum........	40·9	43·8	45·1	46·6	32·8
Loss........	0·8	0·7	0·5	0·6	0·4

The results of extraction of the liquid products and pitches with carbon tetrachloride, and of the determination of ash in the solid residues are given below:

Solids in Liquid Product and Ash in Solids

	Liquid product					Pitch				
	Run 10	Run 11	Run 13	Run 14	Run 15	Run 10	Run 11	Run 13	Run 14	Run 15
Solids insoluble in CCl₄..... %	1·96	3·31	2·71	4·56	2·31	24·92	30·28	31·19	47·70	35·71
Ash in insoluble solids....... %	24·1	27·6	32·7	35·7	52·8	22·7	23·4	33·5	25·5	38·7

The material balances are given in Table III.

TABLE III

Material Balance, Quantities in Grammes

—	Oil	A-M-F coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
			Run 10					
Vehicle	14,551							14,551
Coal		13,722		480		349		14,551
Catalyst								
Pitch	8,097				123			8,220
Hydrogen			1,083					1,083
Total input	22,648	13,722	1,083	480	123	349		38,405
Liquid product	26,293			138	434	1,474	237	28,576
Pitch	5,926			446	1,521			7,893
Recovery								36,469
Gas and loss							1,936	1,936
Total output	32,219			584	1,955	1,474	2,173	38,405
Increase	9,571			104	1,832	1,125	2,173	14,805
Per cent increase	69·75			0·76	13·35	8·20	15·83	107·89
Decrease		13,722	1,083					14,805
Per cent decrease		100·00	7·89					107·89
			Run 11					
Vehicle	14,066							14,066
Coal		13,264		464		338		14,066
Catalyst								
Pitch	5,196			391	1,333			6,920
Hydrogen			990					990
Total input	19,262	13,264	990	855	1,333	338		36,042
Liquid product	21,145			213	558	1,206	168	23,290
Pitch	6,343			645	2,110			9,098
Recovery								32,388
Gas and loss							3,654	3,654
Total output	27,488			858	2,668	1,206	3,822	36,042
Increase	8,226			3	1,335	868	3,822	14,254
Per cent increase	62·02			0·02	10·06	6·54	28·82	107·46
Decrease		13,264	990					14,254
Per cent decrease		100·00	7·46					107·46

TABLE III.—*Continued*
Material Balance, Quantities in Grammes—*Continued*

—	Oil	A-M-F coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Run 13								
Vehicle	11,250							11,250
Coal		10,609		371		270		11,250
Catalyst				483		80		563
Pitch	6,711			241	1,012			7,964
Hydrogen			772					772
Total input	17,961	10,609	772	1,095	1,012	350		31,799
Liquid product	18,617			180	370	1,015	112	20,294
Pitch	6,024			915	1,816			8,755
Recovery								29,049
Gas and loss							2,750	2,750
Total output	24,641			1,095	2,186	1,015	2,862	31,799
Increase	6,680			0*	1,174	665	2,862	11,381
Per cent increase	62·97				11·07	6·27	26·98	·107·29
Decrease		10,609	772					11,381
Per cent decrease		100·00	7·28					107·28

* Assumed

—	Oil	A-M-F coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Run 14								
Vehicle	14,064							14,064
Coal		13,262		464		338		14,064
Catalyst				603		100		703
Pitch	5,141			781	1,550			7,472
Hydrogen			923					923
Total input	19,205	13,262	923	1,848	1,550	438		37,226
Liquid product	21,113			387	696	1,415	133	23,744
Pitch	5,169			1,202	3,512			9,883
Recovery								33,627
Gas and loss							3,599	3,599
Total output	26,282			1,589	4,208	1,415	3,732	37,226
Increase	7,077				2,658	977	3,732	14,444
Per cent increase	53·36				20·04	7·37	28·14	108·91
Decrease		13,262	923	259				14,444
Per cent decrease		100·00	6·96	1·95				108·91

TABLE III—*Concluded*

Material Balance, Quantities in Grammes—*Concluded*

—	Oil	A-M-F coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
				Run 15				
Vehicle.....................	13,740	....	....	....	....	....	....	13,740
Coal........................	....	12,957	....	453	....	330	....	13,740
Catalyst....................	....	....	....	688	....	....	....	688
Pitch.......................	6,821	....	....	589	1,594	....	....	9,004
Hydrogen...................	....	....	787	....	....	....	....	787
Total input.............	20,561	12,957	787	1,730	1,594	330	....	37,959
Liquid product.............	24,704	....	....	320	287	840	105	26,256
Pitch......................	6,349	....	....	1,365	2,162	....	....	9,876
Recovery...................	....	....	....	....	....	....	....	36,132
Gas and loss...............	....	....	....	....	....	....	1,827	1,827
Total output...........	31,053	....	....	1,685	2,449	840	1,932	37,959
Increase....................	10,492	....	....	....	855	510	1,932	13,789
Per cent increase...........	80·97	....	....	....	6·60	3·94	14·91	106·42
Decrease....................	....	12,957	787	45	....	....	....	13,789
Per cent decrease...........	....	100·00	6·07	0·35	....	....	....	106·42

The yields as per cent of the ash-and-moisture-free coal are as follows:

—	Run 10	Run 11	Run 13	Run 14	Run 15
Oil...	69·8	62·0	63·0	53·4	81·0
Combustible solid........................	13·4	10·1	11·1	20·0	6·6
Water......................................	8·2	6·5	6·3	7·4	3·9
Gas..	15·8	28·8	27·0	28·1	14·9
Hydrogen charged................................	7·9	7·5	7·3	7·0	6·1

It is apparent from these yields that the stannous oxide catalyst (Run 15) gives better results than no catalyst (Runs 10 and 11) or a lead acetate catalyst (Runs 13 and 14).

CHAPTER V

MEDIUM-VOLATILE BITUMINOUS COAL FROM THE CROWSNEST DISTRICT IN BRITISH COLUMBIA

The Crowsnest (B.C.) coal was selected for a comparative hydrogenation test because it is representative of the Medium-volatile group of the bituminous class of the A.S.T.M. classification. The production from the district is large, and the coal is comparatively low in inherent ash.

DESCRIPTION AND PRE-TREATMENT OF SAMPLE

The sample used for the test was produced in the Michel colliery. It was slack coal which had been stored for some time at the Fuel Research Laboratories. As removed from storage it contained 1·0 per cent of moisture and 6·2 per cent of ash.

Float and sink tests had previously been made in these laboratories on coal from the Michel mine, and for this reason a complete test was not made on the sample for hydrogenation. The bulk sample was separated in a mixture of gasoline and carbon tetrachloride having a specific gravity of 1·40, and air-dried. The ash content of the washed coal was 3·7 per cent on the dry basis. In commercial practice it would be necessary to employ a "very efficient process" to reduce the ash to this extent.

After washing, the bulk sample was pulverized in the ball mill. The size distribution of the pulverized bulk sample is as follows:

Sieve Analysis

U.S. Series Equivalent No.	Per cent
Through 16 on 30 mesh	0·0
" 30 " 50 "	0·6
" 50 " 100 "	2·6
" 100 " 200 "	16·6
" 200 " 300 "	11·2
" 300 mesh	69·0
Total	100·0

Analyses of the coal after pulverizing are given below:

—	As used in tests	Dry basis	A-M-F basis
Proximate Analysis—			
Moisture.. %	1·0		
Ash.. %	3·6	3·7	
Volatile matter............................... %	26·4	26·6	27·6
Fixed carbon.................................. %	69·0	69·7	72·4
Ultimate Analysis—			
Carbon.. %	84·1	84·9	88·1
Hydrogen....................................... %	5·0	5·0	5·2
Ash.. %	3·6	3·7	
Sulphur... %	0·7	0·7	0·7
Nitrogen.. %	1·5	1·5	1·6
Oxygen.. %	5·1	4·2	4·4
Calorific Value—			
B.T.U. per pound, gross..................	14,730	14,880	15,450

The analysis of the ash in the coal as charged is as follows:

	Per cent
SiO$_2$	43·87
Al$_2$O$_3$	28·08
Fe$_2$O$_3$	12·87
TiO$_2$	1·20
CaO	7·22
MgO	0·81
Na$_2$O	0·52
K$_2$O	0·56
SO$_3$	2·24
MnO	0·07
P$_2$O$_5$	2·80
Total	100·24

HYDROGENATION TEST

The hydrogenation test consisted of eight runs. The first three of these were made with the temperature controller set to maintain the wall of the reaction chamber at 842° F. (450° C.). The next two runs were made with the controller set at 824° F. (440° C.) and 806° F. (430° C.) respectively, to observe the effect of lowering the temperature. In the last three runs the coal was treated with hydrochloric acid, and carbon tetrachloride was added to the catalyst. It had been stated in the literature that this procedure gave improved results.[1] For convenience in description, the runs have been divided into two groups comprising Nos. 1 to 5, and Nos. 6 to 8.

Runs 1 to 5: Variation of Temperature

Characteristics of Materials Charged. The coal as charged has been described.

The vehicle used in each of the runs was, as usual, the fraction of the product of the preceding run boiling above 446° F. (230° C.) in the large still with steam. The vehicle used in Run 1 was from Run 14 on Sydney coal. The distillation ranges of the vehicles are given below:

	Per cent by weight				
	Run 1	Run 2	Run 3	Run 4	Run 5
Water	0·4	Trace	Trace	0·9	0·4
Up to 338° F. (170° C.)	0·6	1·0	1·0	0·5	1·2
338 to 446° F. (170 to 230° C.)	14·6	14·3	13·9	14·6	13·1
446 to 572° F. (230 to 300° C.)	36·3	37·7	37·3	36·7	35·3
Above 572° F. (300° C.)	48·1	46·9	47·8	47·0	50·0
Loss	0·0	0·1	0·0	0·3	0·0

The catalyst in each run, as usual, was stannous oxide, in amount equal to 5 per cent of the coal charged.

[1] Gordon, K.: Journal of the Institute of Fuel, December, 1935.

The pitch charged to the reaction chamber before the run was, in each case, produced in the preceding run. The pitch for Run 1 was from Run 14 on Sydney coal. This pitch contained lead compounds formed from the lead acetate that had been used as catalyst instead of stannous oxide. The proportions of solids insoluble in carbon tetrachloride in the pitches, and of ash in the insoluble solids are as follows:

—	Run 1	Run 2	Run 3	Run 4	Run 5
Solids insoluble in CCl₄............................ %	47·70	52·41	49·66	48·27	38·04
Ash in insoluble solids........................... %	25·5	28·5	31·5	27·0	32·5

Operating Conditions. The temperature, pressure, charging rate, rate of recirculation of hydrogen, and duration of each of the runs are as follows:

—	Run 1	Run 2	Run 3	Run 4	Run 5
Average wall temperature, thermocouples 4 and 8 { °F.	838	840	831	815	799
{ °C.	448	449	444	435	426
Pressure, lb./sq. in.......................................	2940	2940	2940	2940	2940
Average charging rate, lb. paste/hr...................	8·7	8·9	9·0	8·3	9·3
Rate of hydrogen recirculation, at temperature and pressure of reaction chamber, cu. ft./hr.........	6	5	5	6	6
Duration of run, hr....................................	5·83	7·50	6·33	7·83	6·50

Analyses of samples of the gas recirculating through the reaction chamber at the beginning and end of the charging period of each run are given below:

—	Run 1		Run 2		Run 3		Run 4		Run 5	
	Start	End	Start	End	Start	End	Start	End	Start	End
Carbon dioxide.. %	0·1	0·4	0·3	0·6	0·5	0·8	0·2	0·5	0·5	0·8
Oxygen.......... %	0·2	0·3	0·3	0·3	0·5	0·3	0·3	0·2	0·3	0·3
Illuminants ..... %	0·1	0·4	0·2	0·6	0·2	0·5	0·2	0·5	0·3	0·9
Carbon monoxide %	1·0	2·5	0·7	2·8	1·5	3·3	1·1	2·8	0·9	2·4
Hydrogen........ %	92·6	67·3	93·5	55·4	89·4	61·3	90·7	61·2	94·0	75·8
Methane........ %	3·0	16·6	2·3	22·8	4·7	20·0	4·3	20·1	1·6	11·2
Ethane.......... %	0·2	9·0	0·3	13·3	0·9	11·6	1·2	9·4	0·3	6·0
Nitrogen........ % (by difference)	2·8	3·5	2·4	4·2	2·3	2·2	2·0	5·3	2·1	2·6

Characteristics and Quantities of Materials Produced. The results of the distillations of the liquid products in the large still with steam are as follows:

—	Per cent by weight				
	Run 1	Run 2	Run 3	Run 4	Run 5
Water..	2·7	1·8	2·2	} 6·5	
Up to 338° F. (170° C.)...................	2·5	1·4	2·5		
338 to 446° F. (170 to 230° C.)...........	4·2	3·8	4·3	4·3	
Above 446° F. (230° C.)..................	63·8	64·7	65·4	64·4	49·5
Coke......................................	22·1	24·7	24·6	22·1	38·6
Loss......................................	4·7	3·6	1·0	2·7	

The results of the analytical distillation of the liquid products in the Hempel apparatus are given below:

—	Per cent by weight				
	Run 1	Run 2	Run 3	Run 4	Run 5
Water..	2·7	2·2	2·5	1·5	1·3
Up to 338° F. (170° C.)...................	4·5	3·3	3·6	3·6	2·7
338 to 446° F. (170 to 230° C.)...........	12·1	11·7	12·8	11·3	10·0
446 to 572° F. (230 to 300° C.)...........	25·9	26·4	26·5	26·8	25·9
Residuum..................................	54·0	55·8	53·5	56·1	59·1
Loss......................................	0·8	0·6	1·1	0·7	1·0

The results of extraction of the liquid products and pitches with carbon tetrachloride, and the determination of ash in the solid residues are given below:

—	Liquid product					Pitch				
	Run 1	Run 2	Run 3	Run 4	Run 5	Run 1	Run 2	Run 3	Run 4	Run 5
Solids insoluble in CCl$_4$..... %	9·64	11·49	8·30	10·16	14·59	52·41	49·66	48·27	38·04	44·34
Ash in insoluble solids........ %	36·5	38·5	35·7	35·5	31·4	28·5	31·5	27·0	32·5	26·5

The material balances of Runs 1 to 5 are given in Table I.

TABLE I
Material Balance, Quantities in Grammes

—	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Run 1								
Vehicle	11,243							11,243
Coal		10,726		405		112		11,243
Catalyst					562			562
Pitch	4,896			1,139	3,327			9,362
Hydrogen			554					554
Total input	16,139	10,726	554	2,106	3,327	112		32,964
Liquid product	16,945			686	1,195	527	156	19,509
Pitch	4,885			1,533	3,847			10,265
Recovery								29,774
Gas and loss							3,190	3,190
Total output	21,830			2,219	5,042	527	3,346	32,964
Increase	5,691			113	1,715	415	3,346	11,280
Per cent increase	53·06			1·05	15·99	3·87	31·20	105·17
Decrease		10,726	554					11,280
Per cent decrease		100·00	5·17					105·17
Run 2								
Vehicle	14,860							14,860
Coal		14,176		535		149		14,860
Catalyst					743			743
Pitch	4,389			1,378	3,456			9,223
Hydrogen			690					690
Total input	19,249	14,176	690	2,656	3,456	149		40,376
Liquid product	21,713			1,121	1,790	557	152	25,333
Pitch	5,289			1,643	3,574			10,506
Recovery								35,839
Gas and loss							4,537	4,537
Total output	27,002			2,764	5,364	557	4,689	40,376
Increase	7,753			108	1,908	408	4,689	14,866
Per cent increase	54·69			0·76	13·46	2·88	33·08	104·87
Decrease		14,176	690					14,866
Per cent decrease		100·00	4·87					104·87

* A-M-F coal: ash-and-moisture-free coal.

TABLE I—*Continued*
Material Balance, Quantities in Grammes—*Continued*

Run 3

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Vehicle	12,638							12,638
Coal		12,057		455		126		12,638
Catalyst				632				632
Pitch	4,119			1,280	2,784			8,183
Hydrogen			602					602
Total input	16,757	12,057	602	2,367	2,784	126		34,693
Liquid product	18,264			614	1,107	518	228	20,731
Pitch	5,351			1,348	3,645			10,344
Recovery								31,075
Gas and loss							3,618	3,618
Total output	23,615			1,962	4,752		3,846	34,693
Increase	6,858				1,968	392	3,846	13,064
Per cent increase	56·88				16·32	3·25	31·90	108·35
Decrease		12,057	602	405				13,064
Per cent decrease		100·00	4·99	3·36				108·35

Run 4

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Vehicle	14,206					129		14,335
Coal		13,676		516		143		14,335
Catalyst				717				717
Pitch	5,634			592	2,733			8,959
Hydrogen			618					618
Total input	19,840	13,676	618	1,825	2,733	272		38,964
Liquid product	22,882			941	1,711	392	183	26,109
Pitch	6,013			1,200	2,491			9,704
Recovery								35,813
Gas and loss							3,151	3,151
Total output	28,895			2,141	4,202	392	3,334	38,964
Increase	9,055			316	1,469	120	3,334	14,294
Per cent increase	66·21			2·31	10·74	0·88	24·37	104·52
Decrease		13,676	618					14,294
Per cent decrease		100·00	4·52					104·52

* A-M-F coal: ash-and-moisture-free coal.

TABLE I—*Concluded*

Material Balance, Quantities in Grammes—*Concluded*

—	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Run 5								
Vehicle....................	13,352					54		13,406
Coal......................		12,789		483		134		13,406
Catalyst..................				670				670
Pitch.....................	5,696			1,137	2,360			9,193
Hydrogen.................			509					509
Total input............	19,048	12,789	509	2,290	2,360	188		37,184
Liquid product............	20,439			1,126	2,462	320	246	24,593
Pitch.....................	5,298			1,119	3,102			9,519
Recovery..................								34,112
Gas and loss..............							3,072	3,072
Total output...........	25,737			2,245	5,564	320	3,318	37,184
Increase..................	6,689				3,204	132	3,318	13,343
Per cent increase..........	52.30				25.04	1.03	25.94	104.31
Decrease..................		12,789	509	45				13,343
Per cent decrease..........		100.00	3.98	0.35				104.33

* A-M-F coal: ash-and-moisture-free coal.

The yields as per cent of the ash-and-moisture-free coal are given below. The mean of Runs 3 and 4 has been taken for comparison with other coals, because the mean temperature of these two runs is near that used for the other coals.

—	Run 1	Run 2	Run 3	Run 4	Run 5	Mean of Runs 3 and 4
Oil................................... %	53.1	54.7	56.9	66.2	52.3	62.0
Combustible solid...................... %	16.0	13.5	16.3	10.7	25.0	13.2
Water................................. %	3.9	2.9	3.3	0.9	1.0	2.0
Gas................................... %	31.2	33.1	31.9	24.4	25.9	27.8
Hydrogen charged...................... %	5.2	4.9	5.0	4.5	4.0	4.7

There is apparently an optimum temperature at about 815° F. (435° C.) as measured by thermocouples 4 and 8 on the wall of the reaction chamber.

Runs 6 to 8: Variation of Catalyst

Characteristics of Materials Charged. The pulverized coal, as previously described, was thoroughly wetted with 7 per cent hydrochloric acid, dried at 230° F. (110° C.), and ground in the ball mill to break up loosely bonded lumps. After this treatment the coal retained about 0·2 per cent of hydrochloric acid, which could be washed off with water, in addition to that which had reacted with the coal or ash. The analysis of the coal so treated is as follows:

Per cent

Moisture	1·1
Ash	4·0
Volatile matter	25·5
Fixed carbon	69·4
Sulphur	0·6
Calorific value	13,950
Coking properties	Non-coking

The vehicle used in Run 6 was the fraction of the product of the preceding run boiling above 446° F. (230° C.) in the large still with steam, together with a small proportion of the similar fraction from Run 4. The vehicle for Run 7 was produced from the product of Run 15 on Sydney coal, which was filtered and distilled to 446° F. (230° C.) in the large still. The vehicle for Run 8 was largely oil produced by steam distillation of the product of Run 7 in the usual way, but it also contained some of the filtered oil from Run 15 on Sydney coal. The reason for these irregularities in the vehicles is that there was no product from Run 6, and Runs 5 and 7 did not produce sufficient quantities of product for vehicle in subsequent runs.

The catalyst in each of the three runs was stannous oxide and carbon tetrachloride, amounting to 5 and 1 per cent, respectively, of the coal charged.

The pitch charged to the reaction chamber for Run 6 was produced in Run 5. The pitch for Run 7 was produced in Run 15 on Sydney coal. The pitch for Run 8 was from Run 7. The proportions of solids insoluble in carbon tetrachloride, and of ash in the insoluble solids, are shown below:

—	Run 6	Run 7	Run 8
Solids insoluble in CCl₄ %	44·34	35·71	52·4
Ash in insoluble solids %	26·5	38·7	19·9

Operating Conditions. The temperature, pressure, charging rate, rate of recirculation of hydrogen, and duration of Runs 7 and 8 are given below. Run 6 was interrupted after about two hours of charging by a deposit of ammonium bicarbonate which obstructed the line between the high-pressure receiver and the recirculating pump. This deposit had probably accumulated to a large extent in previous operation.

—		Run 7	Run 8
Average wall temperature, thermocouples 4 and 8	°F.	817	817
	°C.	436	436
Pressure, lb./sq. in		2940	2940
Average charging rate, lb. paste/hr		9·1	8·7
Rate of hydrogen recirculation, cu. ft./hr. at temperature and pressure of reaction chamber		5·5	5·4
Duration of run, hr		6·5	6·0

The hydrogen lift pipe became obstructed during Run 8, so that agitation of the reacting charge was poor.

Analyses of the gas recirculating through the reaction chamber at the beginning and end of the charging periods of Runs 7 and 8 are as follows:

—		Run 7		Run 8	
		Start	End	Start	End
Carbon dioxide	%	0·5	0·4	0·3	0·3
Oxygen	%	0·2	0·3	0·1	0·3
Illuminants	%	0·3	0·5	0·3	0·3
Carbon monoxide	%	1·3	2·7	1·1	3·3
Hydrogen	%	90·6	70·9	92·9	65·5
Methane	%	3·9	15·6	2·9	21·7
Ethane	%	0·7	6·1	0·8	4·6
Nitrogen (by difference)	%	2·5	3·5	1·6	4·0

Characteristics and Quantities of Materials Produced. The results of the distillations of the liquid products in the large still with steam are as follows:

—	Per cent by weight	
	Run 7	Run 8
Water	3·0	4·3
Up to 338° F. (170° C.)	3·5	7·0
338 to 446° F. (170 to 230° C.)	5·4	4·1
Above 446° F. (230° C.)	53·7	64·0
Coke	31·9	15·4
Loss	2·5	5·2

The results of the analytical distillations of the liquid products in the Hempel apparatus are given below:

—	Per cent by weight	
	Run 7	Run 8
Water	1·3	1·7
Up to 338° F. (170° C.)	4·9	9·8
338 to 446° F. (170 to 230° C.)	12·7	15·6
446 to 572° F. (230 to 300° C.)	35·5	31·2
Residuum	44·1	40·5
Loss	1·5	1·2

The results of extraction of the liquid products and pitches with carbon tetrachloride, and the determination of ash in the solid residues are given below:

	Liquid product		Pitch	
	Run 7	Run 8	Run 7	Run 8
Solids insoluble in CCl₄................................... %	3·81	5·04	5.·4	68·5
Ash in insoluble solids..................................... %	33·9	27·3	19·9	17·3

Although the pitch was fluid when melted, it was not so readily soluble in carbon tetrachloride and benzol as that from the runs without hydrochloric acid. For this reason it was difficult to obtain a satisfactory analysis.

The material balances are given in Table II. It is to be noticed that there was an unusually large decrease in inorganic material during both runs. This may be due to volatilization of tin chloride in the determination of ash.

TABLE II

Material Balance, Quantities in Grammes

—	Oil	A-M-F coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
				Run 7				
Vehicle......................	13,175	....	....	10	8	17	....	13,210*
Coal........................	....	12,288	....	536	....	255	....	13,079
Catalyst....................	....	....	....	654	....	....	....	654
Pitch.......................	5,634	....	....	1,211	1,919	....	....	8,764
Hydrogen...................	....	....	832	....	....	....	....	832
Total input............	18,809	12,288	832	2,411	1,927	272	....	36,539
Liquid product............	22,739	....	....	315	613	317	365	24,349
Pitch......................	4,292	....	....	939	3,778	....	....	9,009
Recovery...................	....	....	....	....	....	....	....	33,358
Gas and loss...............	....	....	....	....	....	....	3,181	3,181
Total output...........	27,031	....	....	1,254	4,391	317	3,546	36,539
Increase...................	8,222	....	....	....	2,464	45	3,546	14,277
Per cent increase...........	66·91	....	....	....	20·05	0·37	28·86	116·19
Decrease...................	....	12,288	832	1,157	....	....	....	14,277
Per cent decrease..........	....	100·00	6·77	9·41	....	....	....	116·18

TABLE II—*Concluded*

Material Balance, Quantities in Grammes—*Concluded*

—	Oil	A-M-F coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Run 8								
Vehicle....................	13,352	....	....	4	4	23	....	13,383
Coal.......................	....	12,326	....	575	....	482	....	13,383
Catalyst...................	134	....	....	669	....	....	....	803*
Pitch......................	4,559	....	....	848	3,079	....	....	8,486
Hydrogen..................	....	....	815	....	....	....	....	815
Total input.............	18,045	12,326	815	2,096	3,083	505	....	36,870
Liquid product..............	21,657	....	....	324	862	400	282	23,525
Pitch......................	3,012	....	....	1,131	5,405	....	....	9,548
Recovery..................	....	....	....	....	....	....	....	33,073
Gas and loss...............	....	....	....	....	....	....	3,797	3,797
Total output............	24,669	....	....	1,455	6,267	400	4,079	36,870
Increase....................	6,624	....	....	....	3,184	....	4,079	13,887
Per cent increase...........	53·74	....	....	....	25·83	....	33·09	112·66
Decrease...................	....	12,326	815	641	....	105	....	13,887
Per cent decrease..........	....	100·00	6·61	5·20	....	0·85	....	112·66

* Including carbon tetrachloride.

The yields as per cent of the ash-and-moisture-free coal are as follows:

—		Run 7	Run 8
Oil...	%	66·9	53·7
Combustible solid...	%	20·0	25·8
Water..	%	0·4	−0·8
Gas..	%	28·9	33·1
Hydrogen charged...	%	6·8	6·6

CHAPTER VI

HIGH-VOLATILE A BITUMINOUS COAL FROM VANCOUVER ISLAND, B.C.

The Vancouver Island coal was selected for a comparative hydrogenation test although it is classified in the same group as two of the other coals in the series. The purpose of this duplication of tests within the High-volatile A group of the bituminous class was to find if properties other than those used for the A.S.T.M. classification had an appreciable influence on amenability to hydrogenation.

DESCRIPTION AND PRE-TREATMENT OF SAMPLE

The sample used for the test was mined in No. 5 Mine of the Comox colliery at Cumberland, B.C. It was washed prior to shipment. As received it was pea-slack in size, and had 0·9 per cent of moisture and 10·8 per cent of ash.

Float and sink tests had previously been made in these laboratories on coals from the same area, and for this reason a complete test was not made on the sample for hydrogenation. The bulk sample was separated in a solution of zinc chloride having a specific gravity of 1·50. This specific gravity could be employed in commercial practice without difficulty. After flotation it was washed with water to remove the zinc chloride. It was then dried in an oven at 212° F. (100° C.). The ash content of the washed bulk sample was 7·5 per cent on the dry basis.

After washing, the bulk sample was pulverized in the ball mill. The size distribution of the pulverized bulk sample is given below:

Sieve Analysis

U.S. Series Equivalent No.	Per cent
Through 16 on 30 mesh	0·0
" 30 " 50 "	0·3
" 50 " 100 "	10·5
" 100 " 200 "	33·1
" 200 " 300 "	3·6
" 300 mesh	52·5
Total	100·0

Analyses of the pulverized coal are given below:

		As used in tests	Dry basis	A-M-F basis
Proximate Analysis—				
Moisture	%	1·1		
Ash	%	7·5	7·5	
Volatile matter	%	34·2	34·6	37·4
Fixed carbon	%	57·2	57·9	62·6
Ultimate Analysis—				
Carbon	%	77·9	78·8	85·2
Hydrogen	%	5·3	5·2	5·6
Ash	%	7·5	7·5	
Sulphur	%	2·0	2·0	2·2
Nitrogen	%	1·2	1·2	1·3
Oxygen	%	6·1	5·3	5·7
Calorific Value—				
B.T.U. per pound, gross		13,880	14,040	15,180

The analysis of the ash in the coal as charged is given below:

	Per cent
SiO_2	22·21
Al_2O_3	10·89
Fe_2O_3	26·68
TiO_2	1·19
CaO	15·69
MgO	1·26
Na_2O	0·68
K_2O	0·47
SO_3	18·70
MnO	0·03
P_2O_5	1·91
Total	99·71

HYDROGENATION TEST

The hydrogenation test consisted of four runs employing the usual equipment and method. The results of the third run were apparently in error, so that a fourth run was necessary.

Characteristics of Materials Charged. The coal, as charged, has been described.

The vehicle used in each of the runs was, as usual, the fraction of the product of the preceding run boiling above 446° F. (230° C.) in the large still with steam. The vehicle used in Run 1 was from Run 8 on Crowsnest coal. The distillation ranges of the vehicles are as follows:

—	Per cent by weight			
	Run 1	Run 2	Run 3	Run 4
Water	0·4	0·2	0·2	0·3
Up to 338° F. (170° C.)	1·7	0·9	0·8	0·8
338 to 446° F. (170 to 230° C.)	18·2	13·6	12·7	12·4
446 to 572° F. (230 to 300° C.)	40·0	42·0	38·1	37·1
Above 572° F. (300° C.)	39·2	43·3	48·1	49·0
Loss	0·5	0·0	0·1	0·4

The catalyst in each run was stannous oxide, in amount equal to 5 per cent of the coal charged.

The pitch charged to the reaction chamber before Runs 2, 3, and 4 was in each case produced in the preceding run. The pitch for the first run was a semi-solid fraction of low-temperature tar. The proportions of solids insoluble in carbon tetrachloride in the pitches, and of ash in the insoluble solids are shown below:

—	Run 1	Run 2	Run 3	Run 4
Solids insoluble in CCl₄ .... %	17·82	35·33	38·58	35·30
Ash in insoluble solids .... %	7·3	43·2	43·6	46·0

Operating Conditions. The temperature, pressure, charging rate, rate of recirculation of hydrogen, and duration of each of the runs are shown below:

—		Run 1	Run 2	Run 3	Run 4
Average wall temperature, thermocouples 4 and 8	°F.	819	817	810	808
	°C.	437	436	432	431
Pressure, lb./sq. in		3000	3000	3000	3000
Average charging rate, lb. paste/hr		8·7	8·6	8·9	8·8
Rate of hydrogen recirculation, at temperature and pressure of reaction chamber, cu. ft./hr		5	6	6	6
Duration of run, hr		6·83	7·10	7·17	7·50

Analyses of samples of the gas recirculating through the reaction chamber at the beginning and end of the charging period of each run are given below:

—		Run 1		Run 2		Run 3		Run 4	
		Start	End	Start	End	Start	End	Start	End
Carbon dioxide	%	0·8	0·7	0·2	0·5	0·2	0·4	0·1	0·2
Oxygen	%	0·2	0·2	0·2	0·2	0·2	0·2	0·2	0·2
Illuminants	%	0·2	0·3	0·2	0·4	0·1	0·2	0·1	0·2
Carbon monoxide	%	1·5	2·9	1·1	1·6	1·0	2·4	0·4	1·3
Hydrogen	%	89·2	71·8	92·5	78·5	93·5	78·4	96·1	78·8
Methane	%	4·4	15·3	3·1	12·8	2·7	10·9	0·7	10·7
Ethane	%	1·2	5·7	0·7	4·4	0·5	4·9	0·2	5·7
Nitrogen (by difference)	%	2·5	3·1	2·0	1·6	1·8	2·6	2·2	2·9

Characteristics and Quantities of Materials Produced. The results of the distillations of the liquid products in the large still with steam are given below:

—	Per cent by weight			
	Run 1	Run 2	Run 3	Run 4
Water	3·2	1·7	3·0	} 2·3
Up to 338° F. (170° C.)	3·0	1·7	2·6	
338 to 446° F. (170 to 230° C.)	7·6	5·8	2·3	2·7
Above 446° F. (230° C.)	65·0	61·2	57·3	60·7
Coke	18·0	26·6	31·5	30·8
Loss	3·2	3·0	3·3	3·5

A sample of the coke produced in the distillation of the product from Run 3 had the analysis given below:

	Per cent	
—	As received	Dry basis
Moisture	0·5	
Ash	22·5	22·6
Volatile matter	26·9	27·1
Fixed carbon (by difference)	50·1	50·3
Sulphur	3·2	3·2
Calorific value	12,460	12,520

The results of the analytical distillations of the liquid products in the Hempel apparatus are given below:

	Per cent by weight			
—	Run 1	Run 2	Run 3	Run 4
Water	3·1	2·1	2·3	1·6
Up to 338° F. (170° C.)	5·3	4·3	4·1	3·2
338 to 446° F. (170 to 230° C.)	13·6	10·5	8·7	7·1
446 to 572° F. (230 to 300° C)	29·4	23·1	25·3	22·2
Residuum	48·1	59·5	58·9	65·2
Loss	0·5	0·5	0·7	0·7

The results of extraction of the liquid products and pitches with carbon tetrachloride, and the determination of ash in the solid residues are as follows:

	Liquid product				Pitch			
—	Run 1	Run 2	Run 3	Run 4	Run 1	Run 2	Run 3	Run 4
Solids insoluble in CCl₄ %	5·34	12·05	13·85	14·62	35·33	38·58	35·30	36·48
Ash in insoluble solids.. %	41·9	48·5	48·1	46·5	43·2	43·6	46·0	42·5

The material balances of the runs are given in Table I.

TABLE I
Material Balance, Quantities in Grammes

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Run 1								
Vehicle	13,511					54		13,565
Coal		12,399		1,017		149		13,565
Catalyst				678				678
Pitch	6,835			108	1,374			8,317
Hydrogen			691					691
Total input	20,346	12,399	691	1,803	1,374	203		36,816
Liquid product	22,440			551	765	764	123	24,643
Pitch	6,105			1,441	1,894			9,440
Recovery								34,083
Gas and loss							2,733	2,733
Total output	28,545			1,992	2,659	764	2,856	36,816
Increase	8,199			189	1,285	561	2.856	13,090
Per cent increase	66·13			1·52	10·36	4·52	23·03	105·56
Decrease		12,399	691					13,090
Per cent decrease		100·00	5·57					105·57
Run 2								
Vehicle	13,767					28		13,795
Coal		12,608		1,035		152		13,795
Catalyst				689				689
Pitch	5,987			1,398	1,837			9,222
Hydrogen			533					533
Total input	19,754	12,608	533	3,122	1,837	180		38,034
Liquid product	22,165			1,518	1,611	545	130	25,969
Pitch	6,113			1,674	2,165			9,952
Recovery								35,921
Gas and loss							2,113	2,113
Total output	28,278			3,192	3,776	545	2,243	38,034
Increase	8,524			70	1,939	365	2,243	13,141
Per cent increase	67·61			0·56	15·38	2·89	17·79	104·23
Decrease		12,608	533					13,141
Per cent decrease		100·00	4·23					104·23

* A-M-F coal: ash- and-moisture-free coal.

TABLE I—*Concluded*
Material Balance, Quantities in Grammes—*Concluded*

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total

Run 3

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Vehicle	14,090					28		14,118
Coal		12,904		1,059		155		14,118
Catalyst				706				706
Pitch	5,773			1,581	2,046			9,400
Hydrogen			554					554
Total input	19,863	12,904	554	3,346	2,046	183		38,896
Liquid product	23,426			1,877	2,025	648	197	28,173
Pitch	6,273			1,574	1,848			9,695
Recovery								37,868
Gas and loss							1,028	1,028
Total output	29,699			3,451	3,873	648	1,225	38,896
Increase	9,836			105	1,827	465	1,225	13,458
Per cent increase	76·22			0·81	14·16	3·60	9·49	104·28
Decrease		12,904	554					13,458
Per cent decrease		100·00	4·29					104·29

Run 4

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Vehicle	14,514					44		14,558
Coal		13,306		1,092		160		14,558
Catalyst				728				728
Pitch	6·010			1,498	1,758			9,266
Hydrogen			679					679
Total input	20,524	13,306	679	3,318	1,758	204		39,789
Liquid product	23,312			1,907	2,195	449	196	28,059
Pitch	5,998			1,464	1,980			9,442
Recovery								37,501
Gas and loss							2,288	2,288
Total output	29,310			3,371	4,175	449	2,484	39,789
Increase	8,786			53	2,417	245	2,484	13,985
Per cent increase	66·03			0·40	18·16	1·84	18·67	105·10
Decrease		13,306	679					13,985
Per cent decrease		100·00	5·10					105·10

* A-M-F coal: ash-and-moisture-free coal.

The yields as per cent of the ash-and-moisture-free coal are as follows:

—		Run 1	Run 2	Run 3	Run 4	Mean of Runs 2 and 4
Oil	%	66·1	67·6	76·2	66·0	66·8
Combustible solid	%	10·4	15·4	14·2	18·2	16·8
Water	%	4·5	2·9	3·6	1·8	2·3
Gas	%	23·0	17·8	9·5	18·7	18·3
Hydrogen charged	%	5·6	4·2	4·3	5·1	4·7

The very low gas yield of Run 3 is not compatible with the analyses of recirculating gas, and for this reason Run 3 has not been included in the mean.

CHAPTER VII

HIGH-VOLATILE A BITUMINOUS COAL FROM COUNTY DURHAM, ENGLAND

The Durham coal was included in the series of comparative hydrogenation tests because it is processed in the commercial hydrogenation plant of Imperial Chemical Industries at Billingham, England. It is, therefore, a suitable standard of comparison for the Canadian coals with respect to amenability to hydrogenation.

DESCRIPTION AND PRE-TREATMENT OF SAMPLE

The sample used for the test was produced in the Elemore colliery in Durham County, England. Before shipment to these laboratories, the ash content had been reduced from 13 to 2·5 per cent by washing, and the moisture content was only 2·7 per cent, so that neither washing nor drying was necessary.

The bulk sample was pulverized in the ball mill in the usual way. The size distribution of the pulverized bulk sample is given below:

U.S. Series Equivalent No.	Per cent
Through 16 on 30 mesh	0·0
" 30 " 50 "	2·2
" 50 " 100 "	21·5
" 100 " 200 "	24·5
" 200 " 300 "	10·2
" 300 mesh	41·6
Total	100·0

Analyses of the coal after pulverizing are given as follows:

—		As used in tests	Dry basis	A-M-F basis
Proximate Analysis:				
Moisture	%	2·7		
Ash	%	2·5	2·6	
Volatile matter	%	34·2	35·2	36·1
Fixed carbon	%	60·6	62·2	63·9
Ultimate Analysis—				
Carbon	%	81·1	83·4	85·6
Hydrogen	%	5·5	5·3	5·4
Ash	%	2·5	2·6	
Sulphur	%	1·4	1·4	1·4
Nitrogen	%	1·7	1·8	1·9
Oxygen	%	7·8	5·5	5·7
Calorific Value—				
B.T.U. per pound, gross		14,390	14,790	15,180

The analysis of the ash in the coal as charged is given below:

	Per cent
SiO₂	37·05
Al₂O₃	19·40
Fe₂O₃	32·20
TiO₂	1·84
CaO	2·66
MgO	Trace
Na₂O	1·12
K₂O	0·86
SO₃	4·88
MnO	0·04
P₂O₅	0·23
Total	100·28

HYDROGENATION TEST

The hydrogenation test consisted of four runs, using the equipment and method previously described. The fourth run was made because the second and third were not in good agreement.

Characteristics of Materials Charged. The coal, as charged, has been described.

The vehicle used in each of the runs was the fraction of the product of the preceding run boiling above 446° F. (230° C.) in the large still with steam. The vehicle for the first run was from the fourth run with Vancouver Island coal. The distillation ranges of the vehicles are given below:

	Per cent by weight			
	Run 1	Run 2	Run 3	Run 4
Water	0·6	0·2	0·2	0·2
Up to 338° F. (170° C.)	0·9	0·8	1·0	0·9
338 to 446° F. (170 to 230° C.)	10·7	12·5	13·2	14·9
446 to 572° F. (230 to 300° C.)	34·3	32·0	34·9	32·4
Above 572° F. (300° C.)	53·4	54·3	50·7	51·6
Loss	0·1	0·2	0·0	0·0

The catalyst in each run was stannous oxide, in amount equal to 5 per cent of the coal charged.

The pitch charged to the reaction chamber before the run was, in each case, produced in the preceding run. The pitch for the first run was produced in the fourth run with Vancouver Island coal. The proportions of solids insoluble in carbon tetrachloride in the pitches and of ash in the insoluble solids are shown below:

	Run 1	Run 2	Run 3	Run 4
Solids insoluble in CCl₄ %	36·48	32·56	32·32	29·52
Ash in insoluble solids %	42·5	40·5	37·3	41·0

Operating Conditions. The temperature, pressure, charging rate, rate of recirculation of hydrogen, and duration of each of the runs are as follows:

—		Run 1	Run 2	Run 3	Run 4
Average wall temperature, thermocouples 4 and 8	°F.	828	828	831	820
	°C.	442	442	444	438
Pressure, lb./sq. in.		3000	3000	3000	3000
Average charging rate, lb. paste/hr.		8·7	9·0	8·8	8·7
Rate of hydrogen recirculation, at temperature and pressure of reaction chamber, cu. ft./hr.		5	5	5	6
Duration of run, hr.		7·00	7·30	7·00	7·58

The analyses of samples of the gas recirculating through the reaction chamber at the beginning and end of the charging period of each run are:

		Run 1		Run 2		Run 3		Run 4	
—		Start	End	Start	End	Start	End	Start	End
Carbon dioxide	%	0·32	0·47	0·21	0·43	0·22	0·13	0·16	0·28
Oxygen	%	0·48	0·39	0·14	0·23	0·11	0·13	0·32	0·43
Illuminants	%	0·08	0·47	0·22	0·49	0·00	0·26	0·32	0·53
Carbon monoxide	%	0·48	2·69	0·99	2·51	1·00	2·92	1·05	2·55
Hydrogen	%	92·60	67·62	90·50	68·50	87·20	67·20	94·70	74·30
Methane	%	3·82	19·10	4·00	17·26	5·64	24·36	1·44	13·58
Ethane	%	0·05	6·38	0·81	8·28	0·11	3·04	0·21	4·54
Nitrogen (by difference)	%	2·17	2·88	3·13	2·30	5·72	1·96	1·80	3·79

Characteristics and Quantities of Materials Produced. The results of distillation of the liquid products of Runs 1, 2, and 3, in the large still with steam, are given below:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water	6·8	2·9	4·0
Up to 338° F. (170° C.)		3·1	2·8
338 to 446° F. (170 to 230° C.)	2·2	3·1	3·0
Above 446° F. (230° C.)	65·5	56·6	62·9
Coke	22·2	30·7	24·5
Loss	3·3	3·6	2·8

The liquid product from Run 4 was distilled up to 338° F. (170° C.) to remove water, filtered using benzol as a diluent, and again distilled to remove the benzol and the fraction boiling between 338 and 446 ° F. (170 and 230° C.)

The results of the analytical distillations of the liquid products in the Hempel apparatus are given as follows:

—	Per cent by weight			
	Run 1	Run 2	Run 3	Run 4
Water..	2·9	3·2	4·4	3·5
Up to 338° F. (170° C.)..	3·3	4·0	4·0	4·7
338 to 446° F. (170 to 230° C.).................................	9·8	9·3	11·3	10·4
446 to 572° F. (230 to 300° C.).................................	23·4	21·2	27·3	23·9
Residuum..	60·1	61·5	52·4	56·9
Loss...	0·5	0·8	0·6	0·6

The results of extraction of the liquid products and pitches with carbon tetrachloride and the determination of ash in the residues are given as follows:

—	Liquid product				Pitch			
	Run 1	Run 2	Run 3	Run 4	Run 1	Run 2	Run 3	Run 4
Solids insoluble in CCl₄ %	7·03	10·15	6·99	8·13	32·56	32·32	29·52	30·55
Ash in insoluble solids.. %	47·1	47·6	52·1	49·6	40·5	37·3	41·0	40·0

The material balances of the four runs are given in Table I.

TABLE I

Material Balance, Quantities in Grammes

—	Oil	A-M-F* coal	Hydro- gen	Inor- ganic	Com- bustible solid	Water	Gas	Total
				Run 1				
Vehicle.....................	13,366					81		13,447
Coal.......................		12,748		336		363		13,447
Catalyst....................				672				672
Pitch......................	5,641			1,346	1,820			8,807
Hydrogen..................			656					656
Total input.............	19,007	12,748	656	2,354	1,820	444		37,029
Liquid product............	22,264			823	924	721	124	24,856
Pitch.....................	6,412			1,253	1,842			9,507
Recovery..................								34,363
Gas and loss..............							2,666	2,666
Total output...........	28,676			2,076	2,766	721	2,790	37,029
Increase....................	9,669				946	277	2,790	13,682
Per cent increase...........	75·85				7·42	2·17	21·89	107·33
Decrease...................		12,748	656	278				13,682
Per cent decrease...........		100·00	5·15	2·18				107·33

* A-M-F coal: ash- and-moisture-free coal.

TABLE I—*Continued*

Material Balance, Quantities in Grammes—*Continued*

—	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Run 2								
Vehicle	14,483					29		14,512
Coal		13,757		363		392		14,512
Catalyst				726				726
Pitch	5,913			1,140	1,676			8,729
Hydrogen			768					768
Total input	20,396	13,757	768	2,229	1,676	421		39,247
Liquid product	23,535			1,325	1,458	877	219	27,414
Pitch	6,074			1,082	1,819			8,975
Recovery								36,389
Gas and loss							2,858	2,858
Total output	29,609			2,407	3,277	877	3,077	39,247
Increase	9,213			178	1,601	456	3,077	14,525
Per cent increase	66·97			1·29	11·64	3·31	22·37	105·58
Decrease		13,757	768					14,525
Per cent decrease		100·00	5·58					105·58
Run 3								
Vehicle	13,552					27		13,579
Coal		12,873		339		367		13,579
Catalyst				678				678
Pitch	5,588			995	1,673			8,256
Hydrogen			754					754
Total input	19,140	12,873	754	2,012	1,673	394		36,846
Liquid product	22,306			923	849	1,115	152	25,345
Pitch	6,271			1,077	1,550			8,898
Recovery								34,243
Gas and loss							2,603	2,603
Total output	28,577			2,000	2,399	1,115	2,755	36,846
Increase	9,437				726	721	2,755	13,639
Per cent increase	73·31				5·64	5·60	21·40	105·95
Decrease		12,873	754	12				13,639
Per cent decrease		100·00	5·86	0·09				105·95

* A-M-F coal: ash- and-moisture-free coal.

TABLE I—*Concluded*

Material Balance, Quantities in Grammes

—	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Run 4								
Vehicle.....................	14,497	....	....	....	....	20	....	14,526
Coal.........................	....	13,771	....	363	....	392	....	14,526
Catalyst.....................	....	....	....	726	....	....	....	726
Pitch........................	5,670	....	....	974	1,401	....	....	8,045
Hydrogen...................	....	....	764	....	....	....	....	764
Total input.............	20,167	13,771	764	2,063	1,401	421	....	38,587
Liquid product..............	24,068	....	....	1,106	1,123	960	165	27,422
Pitch......................	6,335	....	....	1,115	1,672	....	....	9,122
Recovery..................	....	....	....	....	....	....	....	36,544
Gas and loss..............	....	....	....	....	....	....	2,043	2,043
Total output...........	30,403	....	....	2,221	2,795	960	2,208	38,587
Increase....................	10,236	....	....	158	1,394	539	2,208	14,535
Per cent increase..........	74·33	....	....	1·15	10·12	3·91	16·04	105·55
Decrease...................	....	13,771	764	....	....	....	....	14,535
Per cent decrease..........	....	100·00	5·55	....	....	....	....	105·55

* A-M-F coal: ash-and-moisture-free coal.

The yields as per cent of the ash-and-moisture-free coal are as follows:

—		Run 1	Run 2	Run 3	Run 4	Mean of Runs 2, 3, and 4
Oil...........................	%	75·9	67·0	73·3	74·3	71·5
Combustible solid....................	%	7·4	11·6	5·6	10·1	9·2
Water........................	%	2·2	3·3	5·6	3·9	4·2
Gas..........................	%	21·9	22·4	21·4	16·0	19·9
Hydrogen charged..............	%	5·2	5·6	5·9	5·6	5·7

CHAPTER VIII

HIGH-VOLATILE B BITUMINOUS COAL FROM THE NICOLA DISTRICT IN BRITISH COLUMBIA

Nicola coal was selected for a hydrogenation test because it is representative of the B group of the High-volatile bituminous class of the A.S.T.M. classification.

DESCRIPTION AND PRE-TREATMENT OF SAMPLE

The sample used for the test was produced in the Middlesboro collieries at Merritt, B.C. It consisted of lumps of mixed sizes with some fines. There was a comparatively large amount of white ankerite on the surface of the lumps, and also some small pieces of resin. As received, the sample had 6·7 per cent of moisture and 11·3 per cent of ash.

The bulk sample was crushed and a float and sink test made to determine the extent to which ash could be reduced by washing. Part of the data from this test is given below:

Specific gravity	Floats, per cent dry basis	Ash in floats, per cent dry basis
Less than 1·30	19·4	3·8
1·30 to 1·35	68·1	7·0
1·35 to 1·40	82·4	8·2
1·40 to 1·50	94·4	9·9
1·50 to 1·60	97·5	10·6
Greater than 1·60	100·0	12·1

From the results of the float and sink test the percentages floated and their ash contents corresponding to different degrees of difficulty in commercial washing were calculated. This is summarized below:

Designation of commercial process required	Specific gravity of washing medium	Floats, per cent dry basis	Ash in floats, per cent dry basis
"Efficient process, high tonnage"	1·533	96·0	9·7
"Very efficient process, low tonnage"	1·458	90·8	8·8

Because the ash content of the unwashed coal was high and a considerable reduction could be made by washing, the bulk sample for the test was washed. The specific gravity of the solution used, which was a mixture of gasoline and carbon tetrachloride, was 1·40. The ash content of the washed bulk sample was 7·7 per cent on the dry basis. Referring

to the washing data on page 53, it will be noted that the limiting specific gravity in commercial practice would be about 1·46, which corresponds to an ash content of 8·8 per cent.

After washing, the bulk sample was pulverized in the ball mill. The size distribution of the pulverized bulk sample is given below:

U.S. Series Equivalent No.	Per cent
Through 16 on 30 mesh	3·4
" 30 " 50 "	23·64
" 50 " 100 "	26·36
" 100 " 200 "	18·80
" 200 " 300 "	5·50
" 300 mesh	22·30
Total	100·00

Analyses of the coal after pulverizing are given as follows:

		As used in tests	Dry basis	A-M-F basis
Proximate Analysis—				
Moisture	%	5·6		
Ash	%	7·3	7·7	
Volatile matter	%	36·9	39·1	42·4
Fixed carbon	%	50·2	53·2	57·6
Ultimate Analysis—				
Carbon	%	69·8	73·9	80·1
Hydrogen	%	5·8	5·5	6·0
Ash	%	7·3	7·7	
Sulphur	%	0·6	0·6	0·6
Nitrogen	%	1·6	1·7	1·8
Oxygen	%	14·9	10·6	11·5
Calorific value, B.T.U. per pound, gross		12,400	13,140	14,240

The analysis of the ash in the coal as charged is given below:

	Per cent
SiO_2	50·68
Al_2O_3	34·16
Fe_2O_3	7·51
TiO_2	1·17
CaO	2·13
MgO	1·07
Na_2O	0·24
K_2O	1·26
SO_3	0·46
MnO	0·05
P_2O_5	1·03
Total	99·76

HYDROGENATION TEST

The hydrogenation test consisted of three runs, employing the usual equipment and method.

Characteristics of Materials Charged. The coal as charged has been described.

The vehicles used in Runs 2 and 3 were the fractions of the products of the preceding runs, boiling above 446° F. (230° C.) in the large still with

steam. The vehicle used in Run 1 was from Run 4 with Durham coal. The liquid product was distilled up to 338° F. (170° C.) to remove water and was filtered, using benzol as a diluent. It was then distilled up to 446° F. (230° C.), and the residuum used as a vehicle. This residuum contained 3·99 per cent of solid insoluble in carbon tetrachloride, of which 49·6 per cent was estimated to be ash. The distillation ranges of the vehicles are given below:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water	0·0	0·2	0·0
Up to 338° F. (170° C.)	0·6	0·5	0·8
338 to 446° F. (170 to 230° C.)	8·1	13·3	13·5
446 to 572° F. (230 to 300° C.)	26·8	30·9	32·6
Above 572° F. (300° C.)	64·0	54·6	52·9
Loss	0·5	0·5	0·2

The catalyst in each run was stannous oxide, in amount equal to 5 per cent of the coal charged.

The pitch charged to the reaction chamber before the run was, in each case, produced in the preceding run. The pitch for Run 1 was produced in Run 4 on Durham coal. The proportions of solids insoluble in carbon tetrachloride in the pitches and of ash in the insoluble solids are shown below:

—	Run 1	Run 2	Run 3
Solids insoluble in CCl₄ .... %	30·55	23·84	23·20
Ash in insoluble solids .... %	40·0	58·9	71·3

The average purity of the hydrogen charged, as read from the recorder chart, is shown as:

—	Run 1	Run 2	Run 3
Average per cent H₂	97·4	97·5	97·5

Operating Conditions. The temperature, pressure, charging rate, rate of recirculation of hydrogen, and duration of each of the runs are shown below:

—		Run 1	Run 2	Run 3
Average wall temperature, thermocouples 4 and 8 .... { °F.		820	819	813
{ °C.		438	437	434
Pressure, lb./sq. in.		3000	3000	3000
Average charging rate, lb. paste/hr.		8·7	8·9	8·9
Rate of hydrogen recirculation at temperature and pressure of reaction chamber, cu. ft./hr.		5	5	5
Duration of run, hr.		7·67	7·17	7·00

The analyses of samples of the gas recirculating through the reaction chamber at the beginning and end of the charging period of each run are given as:

—	Run 1		Run 2		Run 3	
	Start	End	Start	End	Start	End
Carbon dioxide.......................... %	0·12	0·33	0·20	0·28	0·37	0·46
Oxygen.................................. %	0·26	0·38	0·10	0·14	0·10	0·48
Illuminants.............................. %	0·18	0·44	0·10	0·00	0·10	0·40
Carbon monoxide...................... %	1·15	2·89	1·30	2·13	1·24	2·20
Hydrogen............................... %	91·20	75·02	88·60	75·43	89·60	80·20
Methane................................ %	4·39	13·77	5·20	16·44	5·28	13·85
Ethane................................. %	0·17	5·20	0·11	1·19	0·20	0·73
Nitrogen (by difference)................ %	2·53	1·97	4·39	4·39	3·11	1·68

Characteristics and Quantities of Materials Produced. The results of distillation of the liquid products in the large still with steam are given below:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water......................................	6·0	6·5	6·6
Up to 338° F. (170° C.)....................	3·0 ⎫	4·3	3·7
338 to 446° F. (170 to 230° C.).............	2·2 ⎬		2·4
Above 446° F. (230° C.)....................	57·0 ⎭	55·9	58·5
Coke......................................	31·8	29·4	27·5
Loss......................................	0·0	3·9	1·3

The results of the analytical distillations of the liquid products in the Hempel apparatus are given below:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water......................................	6·1	6·2	6·4
Up to 338° F. (170° C.)....................	4·5	4·1	5·2
338 to 446° F. (170 to 230° C.).............	7·3	8·8	8·7
446 to 572° F. (230 to 300° C.).............	22·7	24·9	26·4
Residuum..................................	57·8	55·2	52·9
Loss......................................	1·6	0·8	0·4

The results of extraction of the liquid products and pitches with carbon tetrachloride, and the determination of ash in the residues, are given as follows:

—	Liquid product			Pitch		
	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
Solids insoluble in CCl₄.................. %	9·05	9·37	9·97	23·84	23·20	22·32
Ash in insoluble solids.................. %	63·6	66·9	66·2	58·9	71·3	69·0

The material balances of the three runs are given in Table I.

TABLE I

Material Balance, Quantities in Grammes

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Run 1								
Vehicle	14,091	....	....	291	295	....	....	14,677
Coal	....	12,784	....	1,071	....	822	....	14,677
Catalyst	....	....	....	....	....	....	....	734
Pitch	5,955	....	....	1,048	1,572	....	....	8,575
Hydrogen	....	....	914	....	....	....	....	914
Total input	20,046	12,784	914	3,144	1,867	822	....	39,577
Liquid product	22,975	....	....	1,589	909	1,684	442	27,599
Pitch	6,997	....	....	1,290	900	....	....	9,187
Recovery								36,786
Gas and loss							2,791	2,791
Total output	29,972	....	....	2,879	1,809	1,684	3,233	39,577
Increase	9,926	....	....			862	3,233	14,021
Per cent increase	77·64	....	....			6·74	25·29	109·67
Decrease	....	12,784	914	265	58	....	....	14,021
Per cent decrease	....	100·00	7·15	2·07	0·45	....	....	109·67
Run 2								
Vehicle	14,013	....	....	....	....	28	....	14,041
Coal	....	12,230	....	1,025	....	786	....	14,041
Catalyst	....	....	....	701	....	....	....	701
Pitch	6,589	....	....	1,215	847	....	....	8,651
Hydrogen	....	....	664	....	....	....	....	664
Total input	20,602	12,230	664	2,941	847	814	....	38,098
Liquid product	22,969	....	....	1,721	852	1,703	220	27,465
Pitch	6,709	....	....	1,445	582	....	....	8,736
Recovery								36,201
Gas and loss	....			....	....	....	1,897	1,897
Total output	29,678	....	....	3,166	1,434	1,703	2,117	38,098
Increase	9,076	....	....	225	587	889	2,117	12,894
Per cent increase	74·21	....	....	1·84	4·80	7·27	17·31	105·43
Decrease	....	12,230	664	....	....	....	....	12,894
Per cent decrease	....	100·00	5·43	....	....	....	....	105·43

* A-M-F coal: ash-and-moisture-free coal.

TABLE I—*Concluded*

Material Balance, Quantities in Grammes—*Concluded*

—	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total

Run 3

—	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Vehicle	13,760							13,760
Coal		11,985		1,004		771		13,760
Catalyst				688				688
Pitch	6,152			1,325	533			8,010
Hydrogen			677					677
Total input	19,912	11,985	677	3,017	533	771		36,895
Liquid product	21,897			1,736	887	1,684	105	26,309
Pitch	6,800			1,348	606			8,754
Recovery								35,063
Gas and loss							1,832	1,832
Total output	28,697			3,084	1,493	1,684	1,937	36,895
Increase	8,785			67	960	913	1,937	12,662
Per cent increase	73·30			0·56	8·01	7·62	16·16	105·65
Decrease		11,985	677					12,662
Per cent decrease		100·00	5·65					105·65

* A-M-F coal: ash-and-moisture-free coal.

The yields as per cent of the ash-and-moisture-free coal are as follows:

—		Run 1	Run 2	Run 3	Mean of Runs 2 and 3
Oil	%	77·6	74·2	73·3	73·8
Combustible solid	%	0·5	4·8	8·0	6·4
Water	%	6·7	7·3	7·6	7·4
Gas	%	25·3	17·3	16·2	16·8
Hydrogen charged	%	7·2	5·4	5·7	5·5

CHAPTER IX

HIGH-VOLATILE C BITUMINOUS COAL FROM THE SAUNDERS DISTRICT IN ALBERTA

Saunders coal was included in the series of comparative hydrogenation tests because it is representative of the C group of the High-volatile bituminous class in the A.S.T.M. classification.

DESCRIPTION AND PRE-TREATMENT OF SAMPLE

The sample was from run-of-mine and comprised lumps of mixed sizes and fines. A comparatively large amount of fusain and some white ankerite were present. As received the sample had 7·4 per cent of moisture and 7·1 per cent of ash.

The bulk sample was crushed, and a grindability determination made. The grindability index of the dried coal was 52·5.

A float and sink test was made to determine the extent to which ash could be reduced by washing. Part of the data from this test is given below:

Specific gravity	Floats, per cent, dry basis	Ash in floats, per cent, dry basis
Less than 1·30	0·0	
1·30 to 1·35	58·6	3·6
1·35 to 1·40	82·9	4·5
1·40 to 1·45	88·6	4·8
1·45 to 1·50	91·9	5·1
1·50 to 1·60	94·3	5·4
Greater than 1·60	100·0	7·7

As the ash content of the unwashed coal was fairly low and could not be greatly reduced by flotation, the bulk sample for the hydrogenation test was not washed.

The bulk sample was not dried. It was pulverized in a ball mill in the usual way. The size distribution of the pulverized bulk sample is given as follows:

U.S. Series Equivalent No.	Per cent
Through 16 on 30 mesh	0·2
" 30 " 50 "	13·4
" 50 " 100 "	30·4
" 100 " 140 "	12·7
" 140 " 200 "	9·2
" 200 " 300 "	6·3
" 300 mesh	27·8
Total	100·0

The analyses of the coal after pulverizing are given below:

—	As used in tests†	Dry basis	A-M-F basis
Proximate Analysis—			
Moisture... %	7·5		
Ash.. %	6·8	7·3	
Volatile matter.................................. %	32·3	34·9	37·6
Fixed carbon..................................... %	53·4	57·8	62·4
Ultimate Analysis—			
Carbon... %	68·6	74·2	80·0
Hydrogen... %	5·0	4·6	5·0
Ash.. %	6·8	7·3	
Sulphur.. %	0·3	0·3	0·3
Nitrogen... %	1·1	1·2	1·3
Oxygen... %	18·2	12·4	13·4
Calorific Value—			
B.T.U. per pound, gross...........................	11,640	12,590	13,580

† Coal used in first run had moisture content of 6·13 and ash content of 7·14.

The analysis of the ash in the coal as charged is as follows:

	Per cent
SiO_2...	28·48
Al_2O_3...	15·48
Fe_2O_3...	9·53
TiO_2...	0·17
CaO...	28·22
MgO...	2·63
Na_2O...	2·62
K_2O...	0·36
SO_3...	10·01
MnO...	0·05
P_2O_5...	2·07
Total...	99·62

HYDROGENATION TEST

The hydrogenation test consisted of three runs, employing the usual equipment and method.

Characteristics of Materials Charged. The coal as charged has been described.

The vehicle used in each of the runs was the fraction of the product of the preceding run boiling above 446° F. (230° C.) in the large still with

steam. The vehicle used in the first run was from the third run with Nicola coal. The distillation ranges of the vehicles are given as follows:

	Per cent by weight		
	Run 1	Run 2	Run 3
Water	0·2	0·2	0·2
Up to 338° F. (170° C.)	0·8	1·5	1·1
338 to 446° F. (170 to 230° C.)	13·2	15·8	15·1
446 to 572° F. (230 to 300° C.)	36·6	34·0	37·3
Above 572° F. (300° C.)	48·8	48·3	45·8
Loss	0·4	0·2	0·5

The catalyst in each run was stannous oxide, in amount equal to 5 per cent of the coal charged.

The pitch charged to the reaction chamber before the run was, in each case, produced in the preceding run. The pitch for the first run was produced in the third run on Nicola coal. The proportions of solids insoluble in carbon tetrachloride in the pitches, and of ash in the insoluble solids, are shown below:

	Run 1	Run 2	Run 3
Solids insoluble in CCl₄ .... %	22·32	37·89	41·74
Ash in insoluble solids .... %	69·0	43·2	40·2

The average purity of the hydrogen charged, as read from the recorder chart, is shown as:

	Run 1	Run 2	Run 3
Average per cent H₂	97·6	97·2	97·8

Operating Conditions. The temperature, pressure, charging rate, rate of recirculation of hydrogen, and duration of each of the runs, are as follows:

	Run 1	Run 2	Run 3
Average wall temperature, thermocouples 4 and 8 ...... °F.	819	815	817
°C.	437	435	436
Pressure, lb./sq. in.	3000	3000	3000
Average charging rate, lb. paste/hr.	9·0	8·8	8·6
Rate of hydrogen recirculation, at temperature and pressure of reaction chamber, cu. ft./hr.	5	5	5
Duration of run, hr.	6·67	7·00	6·50

The analyses of samples of the gas recirculating through the reaction chamber at the beginning and end of the charging period of each run are given below:

		Run 1		Run 2		Run 3	
		Start	End	Start	End	Start	End
Carbon dioxide	%	0·24	1·67	0·18	2·29	0·26	1·89
Oxygen	%	0·28	0·24	0·09	0·14	0·18	0·12
Illuminants	%	0·18	0·24	0·09	0·14	0·00	0·24
Carbon monoxide	%	1·23	2·86	1·22	3·15	1·14	2·72
Hydrogen	%	91·90	76·50	91·00	71·90	91·30	75·40
Methane	%	3·00	15·18	2·19	15·83	2·11	13·45
Ethane	%	0·05	1·98	0·18	1·17	0·13	2·20
Nitrogen (by difference)	%	3·12	1·33	5·05	5·38	4·88	3·98

Characteristics and Quantities of Materials Produced. The results of the distillations of the liquid products in the large still with steam are given as follows:

	Per cent by weight		
	Run 1	Run 2	Run 3
Water	7·7⎫	12·0*	8·5
Up to 338° F. (170° C.)	5·1⎭		3·7
338 to 446° F. (170 to 230° C.)	1·8	2·6	2·9
Above 446° F. (230° C.)	60·7	55·1	59·4
Coke	20·9	26·4	22·3
Loss	3·8	3·9	3·2

* Water and oil could not be accurately separated by decantation.

The results of the analytical distillations of the liquid products in the Hempel apparatus are given below:

	Per cent by weight		
	Run 1	Run 2	Run 3
Water	7·6	8·1	8·2
Up to 338° F. (170° C.)	7·3	5·7	9·6
338 to 446° F. (170 to 230° C.)	11·5	10·8	7·5
446 to 572° F. (230 to 300° C.)	23·5	22·3	20·9
Residuum	49·7	52·6	52·9
Loss	0·4	0·5	0·9

The results of extraction of the liquid products and pitches with carbon tetrachloride, and the determination of ash in the residues are given as follows:

		Liquid product			Pitch		
		Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
Solids insoluble in CCl_4	%	9·96	12·78	12·62	37·89	41·74	48·21
Ash in insoluble solids	%	53·4	45·1	43·8	43·2	40·2	33·1

The material balances of the three runs are given in Table I.

TABLE I
Material Balance, Quantities in Grammes

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Run 1								
Vehicle	13,244					27		13,271
Coal		11,519		942		810		13,271
Catalyst				664				664
Pitch	6,568			1,288	579			8,435
Hydrogen			575					575
Total input	19,812	11,519	575	2,894	579	837		36,216
Liquid product	20,467			1,327	1,158	1,896	100	24,948
Pitch	5,746			1,515	1,991			9,252
Recovery								34,200
Gas and loss							2,016	2,016
Total output	26,213			2,842	3,149	1,896	2,116	36,216
Increase	6,401				2,570	1,059	2,116	12,146
Per cent increase	55·57				22·31	9·19	18·37	105·44
Decrease		11,519	575	52				12,146
Per cent decrease		100·00	4·99	0·45				105·44
Run 2								
Vehicle	13,645					27		13,672
Coal		11,717		930		1,025		13,672
Catalyst				684				684
Pitch	5,381			1,418	1,865			8,664
Hydrogen			559					559
Total input	19,026	11,717	559	3,032	1,865	1,052		37,251
Liquid product	20,185			1,480	1,801	2,080	128	25,674
Pitch	5,451			1,570	2,336			9,357
Recovery								35,031
Gas and loss							2,220	2,220
Total output	25,636			3,050	4,137	2,080	2,348	37,251
Increase	6,610			18	2,272	1,028	2,348	12,276
Per cent increase	56·42			0·15	19·39	8·77	20·04	104·77
Decrease		11,717	559					12,276
Per cent decrease		100·00	4·77					104·77

* A-M-F coal: ash-and-moisture-free coal.

TABLE I—*Concluded*
Material Balance, Quantities in Grammes—*Concluded*

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Run 3								
Vehicle	12,375					25		12,400
Coal		10,627		843		930		12,400
Catalyst				620				620
Pitch	4,996			1,439	2,140			8,575
Hydrogen			552					552
Total input	17,371	10,627	552	2,902	2,140	955		34,547
Liquid product	17,612			1,244	1,596	1,845	203	22,500
Pitch	5,069			1,562	3,156			9,787
Recovery								32,287
Gas and loss							2,260	2,260
Total output	22,681			2,806	4,752	1,845	2,463	34,547
Increase	5,310				2,612	890	2,463	11,275
Per cent increase	49·96				24·58	8·37	23·18	106·09
Decrease		10,627	552	96				11,275
Per cent decrease		100·00	5·19	0·90				106·09

* A-M-F coal: ash-and-moisture-free coal.

The yields as per cent of the ash-and-moisture-free coal are as follows:

—	Run 1	Run 2	Run 3	Mean of Runs 2 and 3
Oil............%	55·6	56·4	50·0	53·3
Combustible solid............%	22·3	19·4	24·6	21·9
Water............%	9·2	8·8	8·4	8·6
Gas............%	18·4	20·0	23·2	21·5
Hydrogen charged............%	5·0	4·8	5·2	5·0

CHAPTER X

SUB-BITUMINOUS B COAL FROM THE DRUMHELLER
DISTRICT IN ALBERTA

Drumheller coal was selected because it is representative of the B group of the sub-bituminous class in the A.S.T.M. classification; and the district is one of Canada's important producing areas.

DESCRIPTION AND PRE-TREATMENT OF SAMPLE

The sample used for the test was produced in the Rosedale colliery. As received, it consisted of 2½-inch lumps, dull in colour, and containing no fusain.   There were small amounts of white ankerite and shale present. The coal contained 15·0 per cent of moisture and 6·3 per cent of ash.

The bulk sample was crushed, and a float and sink test was made before drying, to determine the extent to which ash could be reduced by washing.   Part of the data from this test is given below:

Specific gravity	Floats, per cent dry basis	Ash in floats, per cent dry basis
Less than 1·30	3·1	5·3
1·30 to 1·35	71·7	5·5
1·35 to 1·40	94·4	6·3
1·40 to 1·45	97·2	6·6
1·45 to 1·50	98·4	6·7
1·50 to 1·60	99·2	7·0
Greater than 1·60	100·0	7·4

It is apparent that the ash is largely inherent and cannot be greatly reduced by flotation.   Accordingly, the bulk sample for the hydrogenation test was not washed.

The grindability index of the undried coal was 39·3.

The bulk sample was dried in the usual way to a moisture content of 2·1 per cent.

The analyses of the coal after drying are given below:

		As used in tests	Dry basis	A-M-F basis
Proximate Analysis—				
Moisture	%	2·1		
Ash	%	7·5	7·7	
Volatile matter	%	37·6	38·4	41·6
Fixed carbon	%	52·8	53·9	58·4
Ultimate Analysis—				
Carbon	%	68·1	69·5	75·3
Hydrogen	%	4·5	4·4	4·8
Ash	%	7·5	7·7	
Sulphur	%	0·7	0·7	0·8
Nitrogen	%	1·6	1·6	1·7
Oxygen	%	17·6	16·1	17·4
Calorific Value—				
B.T.U. per pound, gross		11,480	11,730	12,710

The analysis of the ash in the coal as charged is given below .

	Per cent
SiO_2	39·31
Al_2O_3	20·11
Fe_2O_3	6·73
TiO_2	0·32
CaO	11·81
MgO	1·92
Na_2O	6·91
K_2O	0·45
SO_3	10·92
MnO	0·01
P_2O_5	1·30
Total	99·79

HYDROGENATION TEST

Four runs were made using the equipment and method previously described. The fourth run was made because Runs 2 and 3 were not in good agreement.

Characteristics of Materials Charged. The coal for each run was taken from the dried bulk sample and pulverized separately. The sieve analyses of the coal, as pulverized for the four runs, are given below:

—		Run 1	Run 2	Run 3	Run 4
Through 16 on 30 mesh	%	0·0	0·0	0·0	0·0
" 30 " 50 "	%	0·2	0·0	0·0	0·0
" 50 " 100 "	%	0·7	5·5	8·5	7·3
" 100 " 140 "	%	6·5	15·6	17·5	16·6
" 140 " 200 "	%	12·5	14·9	15·1	16·6
" 200 " 300 "	%	25·4	22·1	18·6	16·9
" 300 mesh	%	54·7	41·9	40·3	42·6
Total	%	100·0	100·0	100·0	100·0

The ash and moisture contents were determined from samples taken after the coal was pulverized. They are shown below:

—		Run 1	Run 2	Run 3	Run 4
Moisture	%	2·1	1·88	2·02	1·88
Ash	%	7·5	7·28	7·14	7·48
Ash on dry basis	%	7·7	7·42	7·29	7·60

There was not enough of the fraction above 446° F. (230° C.) from the third run with the Saunders coal for vehicle for Run 1. For this reason, some of the fraction produced by filtering and distilling to 446° F. (230° C.)

the product of the fifteenth run with Sydney coal was added. The proportions from these two sources were: Saunders 84·4 per cent, and Sydney 15·6 per cent. Since part of this oil had been filtered, the vehicle contained 0·3 per cent of solids, of which 52·8 per cent was estimated to be ash. The vehicles used in Runs 2, 3, and 4 were, as usual, the fractions of the products of the preceding runs boiling above 446° F. (230° C.) in the large still with steam.

The distillation ranges of the vehicles, determined with the Hempel apparatus, are given below:

—	Per cent by weigbt			
	Run 1	Run 2	Run 3	Run 4
Water	0·4	0·3	0·4	0·2
Up to 338° F. (170° C.)	0·4	0·4	0·7	0·4
338 to 446° F. (170 to 230° C.)	15·6	17·9	16·9	16·0
446 to 572° F. (230 to 300° C.)	35·7	35·0	32·4	30·8
Above 572° F. (300° C.)	47·8	46·2	49·4	52·5
Loss	0·1	0·2	0·2	0·1

The viscosities of the vehicles used for Runs 2 and 4 are given as:

—	Run 2	Run 4
Viscosity, Saybolt Universal at 100° F.	66	87

The catalyst in each run was stannous oxide in amount equal to 5 per cent of the coal charged.

The pitch from the run preceding Run 1 was spoiled, and low-temperature tar was substituted. Pitch for the other runs was, in each case, produced in the preceding run. The proportions of solids insoluble in carbon tetrachloride in the tar and pitches, and of ash in the insoluble solids are given as follows:

—	Run 1	Run 2	Run 3	Run 4
Solids insoluble in CCl₄ %	20·88	47·61	40·50	39·64
Asb in insoluble solids %	4·52	34·2	51·6	54·5

The average purity of the hydrogen charged, as read from the recorder chart, is shown below:

—	Run 1	Run 2	Run 3	Run 4
Average per cent H_2	97·7	97·7	97·8	97·9

Operating Conditions. The temperature, pressure, charging rate, rate of recirculation, and duration of each of the runs, are:

—		Run 1	Run 2	Run 3	Run 4
Average wall temperature, thermocouples 4 and 8........ { °F.		819	813	824	817
{ °C.		437	434	440	436
Average temperature inside reaction chamber*.......... } °F.		....	783	788	788
{ °C.		....	417	420	420
Pressure, lb./sq. in..		3000	3000	3000	3000
Average charging rate, lb. paste/hr...........................		7·4	8·6	8·9	8·7
Rate of hydrogen recirculation, at temperature and pressure of reaction chamber, cu. ft./hr............................		5	5	5	5
Duration of run, hr..		8·08	7·00	7·37	7·00

* Average of three thermocouples near top, middle, and bottom of reaction chamber.

The analyses of samples of the gas recirculating through the reaction chamber at the beginning and end of the charging period of each run are given below:

—	Run 1		Run 2		Run 3		Run 4	
	Start	End	Start	End	Start	End	Start	End
Carbon dioxide......... %	1·29	4·37	0·60	4·68	0·96	5·10	0·36	4·49
Oxygen.................. %	0·20	0·24	0·14	0·16	0·10	0·26	0·22	0·50
Illuminants............. %	0·20	0·36	0·52	0·58	0·13	0·46	0·18	0·75
Carbon monoxide....... %	1·68	6·07	2·20	6·86	2·04	6·43	1·32	6·42
Hydrogen.............. %	87·80	61·90	87·20	61·00	87·50	58·20	91·82	60·20
Methane............... %	3·47	20·40	4·44	18·80	3·93	20·73	1·87	18·35
Ethane................. %	0·20	2·75	0·22	4·12	0·19	4·65	0·23	3·72
Nitrogen (by difference) %	5·16	3·91	4·68	3·80	5·15	4·17	4·00	5·57

Characteristics and Quantities of Materials Produced. The results of the distillations of the liquid products of Runs 1, 2, and 3 in the large still with steam are:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water.......	8·8	8·0	8·1
Up to 338° F. (170° C.).....	5·8	4·8	3·8
338 to 446° F. (170 to 230° C.)........	4·6	3·0	4·7
Above 446° F. (230° C.).......	72·5	66·7	63·7
Coke.......	6·4	14·8	17·2
Loss.......	1·9	2·7	2·5

The product from Run 4 was centrifuged prior to distillation. The results of two passes through the centrifuge are given below:

	Per cent
Liquid recovered (less solids)	81·0
Oil and water removed with solids	6·5
Solids, oil-free, recovered	6·4
Solids remaining in liquid	3·1
Handling losses	3·0
	100·0

Distillation in the large still of the liquid passed twice through the centrifuge gave the results shown below:

	Per cent by weight
Water	6·8
Up to 338° F. (170° C.)	4·3
338 to 446° F. (170 to 230° C.)	3·8
Residuum above 446° F. (230° C.) containing solids	82·0
Distillation loss	3·1
	100·0

The results of the analytical distillation of the liquid products in the Hempel apparatus are given below:

—	Per cent by weight			
	Run 1	Run 2	Run 3	Run 4
Water	8·8	7·9	8·1	7·6
Up to 338° F. (170° C.)	7·1	6·6	5·0	6·1
338 to 446° F. (170 to 230° C.)	16·0	12·2	12·7	11·0
446 to 572° F. (230 to 300° C.)	25·0	23·4	22·5	21·2
Residuum	43·0	49·7	51·3	53·5
Loss	0·1	0·2	0·4	0·6

The results of the extraction of the liquid products and pitches with carbon tetrachloride, and the determination of ash in the residues are given below:

—	Liquid product				Pitch			
	Run 1	Run 2	Run 3	Run 4	Run 1	Run 2	Run 3	Run 4
Solids insoluble in CCl₄..%	2·25	6·61	7·90	9·46	47·61	40·50	39·64	40·90
Ash in insoluble solids...%	50·6	59·5	62·6	65·9	34·2	51·6	54·5	53·8

The material balances of the four runs are given in Table I.

TABLE I
Material Balance, Quantities in Grammes

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Run 1								
Vehicle	13,345			21	18	52		13,436
Coal		11,854		983		275		13,112
Catalyst				655				655
Pitch	6,096			75	1,595	232		7,998
Hydrogen			890					890
Total input	19,441	11,854	890	1,734	1,613	559		36,091
Liquid product	21,404			274	268	2,120	24	24,090
Pitch	4,646			1,444	2,779			8,869
Recovery								32,959
Gas and loss							3,132	3,132
Total output	26,050			1,718	3,047	2,120	3,156	36,091
Increase	6,609				1,434	1,561	3,156	12,760
Per cent increase	55·75				12·10	13·17	26·62	107·64
Decrease		11,854	890	16				12,760
Per cent decrease		100·00	7·51	0·13				107·64
Run 2								
Vehicle	13,272					40		13,312
Coal		12,087		972		253		13,312
Catalyst				666				666
Pitch	4,442			1,253	2,410			8,105
Hydrogen			825					825
Total input	17,714	12,087	825	2,891	2,410	293		36,220
Liquid product	20,563			948	646	1,905	48	24,110
Pitch	5,464			1,915	1,797			9,166
Recovery								33,276
Gas and loss							2,944	2,944
Total output	26,017			2,863	2,443	1,905	2,992	36,220
Increase	8,303				33	1,612	2,992	12,940
Per cent increase	68·69				0·27	13·34	24·75	107·05
Decrease		12,087	825	28				12,940
Per cent decrease		100·00	6·83	0·23				107·05

* A-M-F coal: ash-and-moisture-free coal.

TABLE I—*Concluded*
Material Balance, Quantities in Grammes—*Concluded*

Run 3

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Vehicle	14,418					58		14,476
Coal		13,158		1,028		290		14,476
Catalyst				723				723
Pitch	5,075			1,712	1,606			8,393
Hydrogen			908					908
Total input	19,493	13,158	908	3,463	1,606	348		38,976
Liquid product	21,762			1,287	769	2,108	104	26,030
Pitch	5,738			2,054	1,714			9,506
Recovery								35,536
Gas and loss							3,440	3,440
Total output	27,500			3,341	2,483	2,108	3,544	38,976
Increase	8,007				877	1,760	3,544	14,188
Per cent increase	60·85				6·67	13·38	26·93	107·83
Decrease		13,158	908	122				14,188
Per cent decrease		100·00	6·90	0·93				107·83

Run 4

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Vehicle	13,440					27		13,467
Coal		12,201		1,010		256		13,467
Catalyst				674				674
Pitch	5,580			1,913	1,598			9,091
Hydrogen			842					842
Total input	19,020	12,201	842	3,597	1,598	283		37,541
Liquid product	20,412			1,545	800	1,884	149	24,790
Pitch	5,671			2,112	1,813			9,596
Recovery								34,386
Gas and loss							3,155	3,155
Total output	26,083			3,657	2,613	1,884	3,304	37,541
Increase	7,063			60	1,015	1,601	3,304	13,043
Per cent increase	57·89			0·49	8·32	13·12	27·08	106·90
Decrease		12,201	842					13,043
Per cent decrease		100·00	6·90					106·90

* A-M-F coal: ash-and-moisture-free coal.

The yields as per cent of the ash-and-moisture-free coal are as follows:

—	Run 1	Run 2	Run 3	Run 4	Mean of Runs 2, 3, and 4
Oil..%	55·8	68·7	60·9	57·9	62·5
Combustible solid................................%	12·1	0·3	6·7	8·3	5·1
Water...%	13·2	13·3	13·4	13·1	13·3
Gas...%	26·6	24·8	26·9	27·1	26·3
Hydrogen charged..............................%	7·5	6·8	6·9	6·9	6·9

CHAPTER XI

SUB-BITUMINOUS C COAL FROM THE EDMONTON DISTRICT IN ALBERTA

Edmonton coal was selected for a hydrogenation test because it is representative of the C group of the sub-bituminous class in the A.S.T.M. classification.

DESCRIPTION AND PRE-TREATMENT OF SAMPLE

The sample used for the test was mined in the Black Diamond mine at Clover Bar. It consisted of large lumps, apparently containing no fusain. As received it had 24·2 per cent of moisture and 7·8 per cent of ash.

The bulk sample was crushed and dried in the usual way to a moisture content of 2·3 per cent.

The grindability index of the dried coal was 37·6.

A float and sink test was made to determine the extent to which ash could be reduced by washing. Part of the data from this test is given below:

Specific gravity	Floats, per cent dry basis	Ash in floats per cent dry basis
Less than 1·35	76·3	7·8
1·35 to 1·40	92·6	8·8
1·40 to 1·45	95·0	9·0
1·45 to 1·50	96·6	9·2
1·50 to 1·60	98·4	9·5
Greater than 1·60	100·0	10·6

It is apparent that the ash is largely inherent and cannot be greatly reduced by flotation. Accordingly, the bulk sample for the hydrogenation test was not washed.

The analyses of the coal after drying are given below:

—		As used in tests	Dry basis	A-M-F basis
Proximate Analysis—				
Moisture	%	2·3		
Ash	%	9·3	9·5	
Volatile matter	%	36·7	37·6	41·5
Fixed carbon	%	51·7	52·9	58·5
Ultimate Analysis—				
Carbon	%	65·1	66·6	73·6
Hydrogen	%	4·5	4·3	4·8
Ash	%	9·3	9·5	
Sulphur	%	0·4	0·5	0·5
Nitrogen	%	1·4	1·4	1·5
Oxygen	%	19·3	17·7	19·6
Calorific Value—				
B.T.U. per pound, gross		10,760	11,010	12,170

The analysis of the ash in the coal as charged is given as:

	Per cen
SiO_2	38·66
Al_2O_3	25·63
Fe_2O_3	4·89
TiO_2	0·50
CaO	14·32
MgO	1·53
Na_2O	4·78
K_2O	0·42
SO_3	6·39
MnO	0·02
P_2O_5	2·54
Total	99·68

HYDROGENATION TEST

The hydrogenation test consisted of three runs, using the equipment and method previously described.

Characteristics of Materials Charged. The coal for each run was taken from the dried bulk sample and pulverized separately. The sieve analyses of the coal as pulverized for the three runs are given below:

—		Run 1	Run 2	Run 3
Through 16 on 30 mesh	%	0·0	0·0	0·0
" 30 " 50 "	%	0·0	0·0	0·0
" 50 " 100 "	%	0·5	3·7	7·0
" 100 " 140 "	%	4·2	14·1	17·9
" 140 " 200 "	%	10·7	16·0	15·2
" 200 " 300 "	%	26·4	22·2	21·6
" 300 mesh	%	58·2	44·0	38·3
Total	%	100·0	100·0	100·0

The ash and moisture contents were determined from samples taken after the coal was pulverized. They are shown below:

—		Run 1	Run 2	Run 3
Moisture	%	2·28	2·65	2·27
Ash	%	9·29	9·83	9·57
Ash on dry basis	%	9·51	10·10	9·79

The vehicle for Run 1 was produced by centrifuging the liquid product from the Run 4 with Drumheller coal. The product was centrifuged twice and distilled to 446° F. (230° C.); the residuum boiling above this temperature in the large still was used as vehicle. This residuum contained 4·56 per cent of solids insoluble in carbon tetrachloride, of which 37·0 per

cent remained as ash after ignition. The viscosity of this vehicle was too high, and the paste had to be diluted with benzol. The vehicles used in Runs 2 and 3 were the fractions of the products of the preceding runs boiling above 446° F. (230° C.) in the large still with steam, excepting that 12 per cent of the vehicle for Run 3 was produced in Run 1 on Drumheller coal.

The distillation ranges of the vehicles for Runs 2 and 3, determined with the Hempel apparatus, are given below:

—	Per cent by weight	
	Run 2	Run 3*
Water	0·6	0·8
Up to 338° F. (170° C.)	0·6	0·7
338 to 446° F. (170 to 230° C.)	12·4	14·5
446 to 572° F. (230 to 300° C.)	32·1	35·0
Above 572° F. (300° C.)	54·1	48·8
Loss	0·2	0·2

* Distillate above 446° F. (230° C.) from Run 2.

The viscosities of the vehicles used for Runs 2 and 3 are:

—	Run 2	Run 3
Viscosity, Saybolt Universal at 100° F	92	75

The catalyst in each run was stannous oxide in amount equal to 5 per cent of the coal charged.

The pitch charged to the reaction chamber before the run was, in each case, produced in the preceding run. The pitch for Run 1 was produced in Run 4 on Drumheller coal. The proportions of solids insoluble in carbon tetrachloride in the pitches and of ash in the insoluble solids are shown below:

—	Run 1	Run 2	Run 3
Solids insoluble in CCl₄ %	40·90	43·04	40·93
Ash in insoluble solids %	53·8	44·8	56·3

The average purity of the hydrogen charged, as read from the recorder chart, is:

—	Run 1	Run 2	Run 3
Average per cent H_2	97·6	97·7	97·5

Operating Conditions. The temperature, pressure, charging rate, rate of recirculation of hydrogen, and duration of each of the runs, are shown below:

—		Run 1	Run 2	Run 3
Average wall temperature, thermocouples 4 and 8	°F.	813	820	819
	°C.	434	438	437
Average temperature inside reaction chamber*	°F.	779	790	777
	°C.	415	421	414
Pressure, lb./sq. in.		3000	3000	3000
Average charging rate, lb. paste/hr.		9·7	9·3	9·1
Rate of hydrogen recirculation, at temperature and pressure of reaction chamber, cu. ft./hr.		5	5	5
Duration of run, hr.		7·00	6·00	6·67

* Average of three thermocouples near top, middle, and bottom of reaction chamber.

The analyses of samples of the gas recirculating through the reaction chamber at the beginning and end of the charging period of each run are given below:

—		Run 1		Run 2		Run 3	
		Start	End	Start	End	Start	End
Carbon dioxide	%	0·43	5·56	0·65	5·28	0·72	6·14
Oxygen	%	0·61	0·19	0·08	0·21	0·19	0·22
Illuminants	%	0·00	0·47	0·10	0·63	0·23	0·63
Carbon monoxide	%	2·35	6·45	1·38	6·69	1·96	6·57
Hydrogen	%	86·00	65·00	90·20	63·80	88·00	63·20
Methane	%	8·64	15·62	2·67	17·42	4·06	16·17
Ethane	%	0·68	2·87	0·39	3·27	0·44	3·57
Nitrogen (by difference)	%	1·29	3·84	4·53	2·70	4·40	3·50

Characteristics and Quantities of Materials Produced. The results of the distillations of the liquid products in the large still with steam are given below:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water	6·9	8·4	8·4
Up to 338° F. (170° C.)	8·5	5·9	3·8
338 to 446° F. (170 to 230° C.)	2·5	2·8	2·6
Above 446° F. (230° C.)	53·0	59·4	61·9
Coke	26·5	20·5	20·5
Loss	2·6	3·0	2·8

The results of the analytical distillation of the liquid products in the Hempel apparatus are as follows:

	Per cent by weight		
	Run 1	Run 2	Run 3
Water	7·1	8·1	8·5
Up to 338° F. (170° C.)	9·8	7·4	4·8
338 to 446° F. (170 to 230° C.)	8·1	10·4	11·0
446 to 572° F. (230 to 300° C.)	17·6	21·9	21·7
Residuum	56·9	51·7	53·4
Loss	0·5	0·5	0·6

The results of extraction of the liquid products and pitches with carbon tetrachloride and the determination of ash in the residues are given below:

	Liquid product			Pitch		
	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
Solids insoluble in CCl_4 .... %	13·40	9·93	11·00	43·04	40·98	40·67
Ash in insoluble solids .... %	61·2	60·6	63·1	44·8	56·3	56·7

The material balances of the three runs are given in Table I.

TABLE I

Material Balance, Quantities in Grammes

	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Run 1								
Vehicle	15,674			232	396			**16,302
Coal		12,183		1,282		317		13,782
Catalyst				689				689
Pitch	5,003			1,863	1,599			8,465
Hydrogen			818					818
Total input	20,677	12,183	818	4,066	1,995	317		40,056
Liquid product	20,650			2,144	1,359	1,856	131	26,140
Pitch	5,521			1,869	2,302			9,692
Recovery								35,832
Gas and loss							4,224	4,224
Total output	26,171			4,013	3,661	1,856	4,355	40,056
Increase	5,494				1,666	1,539	4,355	13,054
Per cent increase	45·10				13·67	12·63	35·75	107·15
Decrease		12,183	818	53				13,054
Per cent decrease		100·00	6·71	0·44				107·15

* A-M-F coal: ash-and-moisture-free coal.
** Benzol was added to the paste to reduce its viscosity. The benzol has been included as vehicle.

TABLE I—*Concluded*

Material Balance, Quantities in Grammes—*Concluded*

Run 2

—	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Vehicle	12,271					74		12,345
Coal		10,802		1,210		333		12,345
Catalyst				617				617
Pitch	4,986			1,688	2,079			8,753
Hydrogen			874					874
Total input	17,257	10,802	874	3,515	2,079	407		34,934
Liquid product	18,192			1,344	873	1,809	112	22,330
Pitch	5,547			2,169	1,683			9,399
Recovery								31,729
Gas and loss							3,205	3,205
Total output	23,739			3,513	2,556	1,809	3,317	34,934
Increase	6,482				477	1,402	3,317	11,678
Per cent increase	60·00				4·42	12·98	30·71	108·11
Decrease		10,802	874	2				11,678
Per cent decrease		100·00	8·09	0·02				108·11

Run 3

—	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Vehicle	13,351					108		13,459
Coal		11,857		1,292		310		13,459
Catalyst				673				673
Pitch	5,110			1,975	1,533			8,618
Hydrogen			856					856
Total input	18,461	11,857	856	3,940	1,533	418		37,065
Liquid product	19,224			1,670	977	2,045	144	24,060
Pitch	5,842			2,270	1,734			9,846
Recovery								33,906
Gas and loss							3,159	3,159
Total output	25,066			3,940	2,711	2,045	3,303	37,065
Increase	6,605			0†	1,178	1,627	3,303	12,713
Per cent increase	55·71				9·93	13·72	27·86	107·22
Decrease		11,857	856					12,713
Per cent decrease		100·00	7·22					107·22

* A-M-F coal: ash-and-moisture-free coal.
† Assumed.

The yields as per cent of the ash- and-moisture-free coal are as follows:

—	Run 1	Run 2	Run 3	Mean of Runs 2 and 3
Oil......................................%	45·1	60·0	55·7	57·7
Combustible solid......................%	13·7	4·4	9·9	7·3
Water..................................%	12·6	13·0	13·7	13·4
Gas....................................%	35·8	30·7	27·9	29·2
Hydrogen charged......................%	6·7	8·1	7·2	7·6

CHAPTER XII

LIGNITE FROM THE BIENFAIT DISTRICT IN SASKATCHEWAN

Bienfait lignite was included in the series of tests because it is representative of the lignites occurring in the Souris area in Saskatchewan. This area produces the major part of the total output of that Province, which is about one million tons per year. It is well situated geographically, and the cost of mining is low.

DESCRIPTION AND PRE-TREATMENT OF SAMPLE

The sample tested was slightly higher in rank than the average from the whole area, and was very near the dividing line between the sub-bituminous and lignite groups according to the A.S.T.M. classification. There was some fusain, and also thin black bands of bright coal. As received, it had 33·5 per cent of moisture and 6·7 per cent of ash.

About 300 pounds were crushed and dried in an oven at 230° F. (110° C.) to a moisture content of 3 per cent. The dried lignite was then stored in covered containers.

The grindability index of the dried lignite was 51·1.

A float and sink test was made to determine the extent to which ash could be reduced by washing. Part of the data from this test is given as follows:—

Specific gravity	Floats, per cent dry basis	Ash in floats, per cent dry basis
Less than 1·20	0·3	5·2
1·20 to 1·30	65·0	7·9
1·30 to 1·35	81·1	8·3
1·35 to 1·45	93·9	8·9
1·45 to 1·50	96·8	9·3
1·50 to 1·60	98·8	9·5
Greater than 1·60	100·0	10·1

It is apparent that the ash is largely inherent and cannot be greatly reduced by flotation. Accordingly, the bulk sample for the hydrogenation test was not washed.

The analyses of the lignite after drying are:

—		As used in tests	Dry basis	A-M-F basis
Proximate Analysis—				
Moisture	%	3·0		
Ash	%	9·8	10·1	
Volatile matter	%	37·9	39·1	43·5
Fixed carbon	%	49·3	50·8	56·5
Ultimate Analysis—				
Carbon	%	65·2	67·2	74·8
Hydrogen	%	4·9	4·7	5·2
Ash	%	9·8	10·1	....
Sulphur	%	0·4	0·4	0·5
Nitrogen	%	1·3	1·4	1·5
Oxygen	%	18·4	16·2	18·0
Calorific Value—				
B.T.U. per pound, gross		10,880	11,220	12,480

The analysis of the ash in the lignite as charged is given as:

	Per cent
SiO_2	27·21
Al_2O_3	17·84
Fe_2O_3	6·76
TiO_2	0·49
CaO	19·77
MgO	4·98
Na_2O	9·88
K_2O	0·49
SO_3	11·25
MnO	0·01
P_2O_5	0·96
Total	99·64

HYDROGENATION TEST

The hydrogenation test consisted of three runs, using the equipment and method previously described.

Characteristics of Materials Charged. The lignite for each run was taken from the dried bulk sample and powdered separately. Sieve analyses of the lignite as pulverized for the three runs are given below:

U.S. Series Equivalent No.		Run 1	Run 2	Run 3
Through 16 on 30 mesh	%	0·0	0·0	0·0
" 30 " 50 "	%	0·0	0·0	0·0
" 50 " 100 "	%	10·2	6·9	7·6
" 100 " 140 "	%	18·2	16·1	16·1
" 140 " 200 "	%	14·2	16·5	15·9
" 200 " 300 "	%	18·9	21·4	21·1
" 300 mesh	%	38·5	39·1	39·3
Total	%	100·0	100·0	100·0

The ash and moisture contents of the lignite as charged were as shown.

—		Run 1	Run 2	Run 3
Moisture	%	2·95	2·60	3·19
Ash	%	9·29	9·43	10·16
Ash on dry basis	%	9·57	9·68	10·49

The vehicle used in each case was the fraction of the product of the preceding run boiling above 446° F. (230° C.) in the large still with steam. The vehicle for Run 1 was from the product of Run 3 on Edmonton coal. The distillation ranges of the vehicles, determined with the Hempel apparatus, are given below:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water	0·8	0·6	0·4
Up to 338° F. (170° C.)	0·5	0·3	0·5
338 to 446° F. (170 to 230° C.)	14·4	15·7	15·3
446 to 572° F. (230 to 300° C.)	35·5	32·7	33·8
Above 572° F. (300° C.)	48·7	50·5	49·9
Loss	0·1	0·2	0·1

The viscosities of the vehicles are given as:

—		Run 1	Run 2	Run 3
Viscosity, Saybolt Universal at 100° F		73	75	72

The catalyst in each run, as before, was stannous oxide, in amount equal to 5 per cent of the lignite charged.

The pitch charged to the reaction chamber before the run was in each case produced in the preceding run. The pitch for Run 1 was produced in Run 3 on Edmonton coal. The proportions of solids insoluble in carbon tetrachloride in the pitches and of ash in the insoluble solids are shown as:

—		Run 1	Run 2	Run 3
Solids insoluble in CCl₄	%	40·67	44·04	47·36
Ash in insoluble solids	%	56·7	56·1	58·1

The average purity of the hydrogen charged, as read from the recorder chart, is shown as

—	Run 1	Run 2	Run 3
Average per cent H₂...	97·8	97·7	97·5

Operating Conditions.   The operating conditions are given below:

—		Run 1	Run 2	Run 3
Average wall temperature, thermocouples 4 and 8...	°F.	826	819	815
	°C.	441	437	435
Average temperature inside reaction chamber...	°F.	792[1]	774[2]	783[3]
	°C.	422	412	417
Pressure, lb./sq. in...		3000	3000	3000
Average charging rate, lb. paste/hr...		9·0	8·6	9·0
Rate of hydrogen recirculation, at temperature and pressure of reaction chamber, cu. ft./hr...		5	5	5
Duration of run, hours...		6·67	6·92	6·67

[1] One thermocouple in annular space, 6 feet from top.
[2] One thermocouple in annular space, 3 feet from top.
[3] One thermocouple in annular space, about centre of reaction chamber.

The analyses of samples of the gas recirculating through the reaction chamber at the beginning and end of the charging period of each run are given as follows:

—		Run 1		Run 2		Run 3	
		Start	End	Start	End	Start	End
Carbon dioxide...	%	0·54	6·90	0·47	6·73	0·75	7·22
Oxygen...	%	0·11	0·15	0·09	0·45	0·22	0·22
Illuminants...	%	0·13	0·33	0·19	0·45	0·15	0·60
Carbon monoxide...	%	1·52	7·44	1·63	7·35	2·35	7·68
Hydrogen...	%	90·10	59·80	90·20	60·30	86·82	59·60
Methane...	%	2·63	21·18	2·06	18·07	3·98	18·47
Ethane...	%	0·13	2·41	0·54	3·83	0·13	3·04
Nitrogen (by difference)...	%	4·84	1·79	4·82	2·82	5·60	3·17

Characteristics and Quantities of Materials Produced.   The results of the distillations of the liquid products in the large still with steam are given below:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water...	9·2	8·5	8·5
Up to 338° F. (170° C.)...	4·2	5·3	5·2
338 to 446° F. (170 to 230° C.)...	3·6	3·0	3·1
Above 446° F. (230° C.)...	63·7	63·7	62·3
Coke...	16·2	17·0	17·6
Loss...	3·1	2·5	3·3

The results of the analytical distillations of the liquid products in the Hempel apparatus are given below:

	Per cent by weight		
	Run 1	Run 2	Run 3
Water	8·6	8·3	8·6
Up to 338° F. (170° C.)	5·7	6·7	6·3
338 to 446° F. (170 to 230° C.)	12·6	12·0	10·6
446 to 572° F. (230 to 300° C.)	22·2	22·4	23·7
Residuum	50·3	49·9	50·2
Loss	0·6	0·7	0·6

The results of the extractions of the liquid products and pitches with carbon tetrachloride and the determination of ash in the residues are given as follows:

	Liquid product			Pitch		
	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
Solids insoluble in CCl₄ ... %	8·45	9·77	10·95	44·04	47·36	43·14
Ash in insoluble solids ... %	63·4	65·6	61·8	56·1	58·1	59·0

The material balances of the three runs are shown in Table I.

TABLE I
Material Balance, Quantities in Grammes

	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Run 1								
Vehicle	13,143			1,232		106		13,249
Coal		11,620		1,232		397		13,249
Catalyst				663				663
Pitch	5,190			2,017	1,541			8,748
Hydrogen			847					847
Total input	18,333	11,620	847	3,912	1,541	503		36,756
Liquid product	19,089			1,242	717	1,993	139	23,180
Pitch	5,720			2,525	1,975			10,220
Recovery								33,400
Gas and loss							3,356	3,356
Total output	24,809			3,767	2,692	1,993	3,495	36,756
Increase	6,476				1,151	1,490	3,495	12,612
Per cent increase	55·73				9·91	12·82	30·08	108·54
Decrease		11,620	847	145				12,612
Per cent decrease		100·00	7·29	1·25				108·54

* A-M-F coal: ash- and moisture-free coal.

TABLE I—*Concluded*

Material Balance, Quantities in Grammes—*Concluded*

—	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
			Run 2					
Vehicle...................	13,126					79	....	13,205
Coal......................	....	11,621	....	1,241	....	343	....	13,205
Catalyst..................	....			661				661
Pitch....................	5,249			2,317	1,813	....	....	9,379
Hydrogen.................	....	....	870	....	....	....		870
Total input........	18,375	11,621	870	4,219	1,813	422	....	37,320
Liquid product.............	19,382	....	....	1,529	802	1,980	167	23,860
Pitch.....................	5,290	....	....	2,765	1,994	....	....	10,049
Recovery..................	....		....	....	....	....	....	33,909
Gas and loss..............						....	3,411	3,411
Total output.......	24,672	....	....	4,294	2,796	1,980	3,578	37,320
Increase..................	6,297	....	....	75	983	1,558	3,578	12,491
Per cent increase...........	54·19	....	....	0·64	8·46	13·41	30·79	107·49
Decrease..................	....	11,621	870	....	....	....	....	12,491
Per cent decrease..........	....	100·00	7·49	....	....	....	....	107·49
			Run 3					
Vehicle...................	13,196					53	....	13,249
Coal......................	....	11,474	....	1,351	....	424	....	13,249
Catalyst..................	....			662				662
Pitch....................	4,769		....	2,492	1,798	....		9,059
Hydrogen.................	....	....	806	....	....			806
Total input........	17,965	11,474	806	4,505	1,798	477	....	37,025
Liquid product.............	19,048	....	....	1,614	998	2,052	143	23,855
Pitch.....................	5,699	....	....	2,551	1,773	....	....	10,023
Recovery..................	....	....	....	....	....	....	....	33,878
Gas and loss..............						....	3,147	3,147
Total output.......	24,747	....	....	4,165	2,771	2,052	3,290	37,025
Increase..................	6,782	....	....	....	973	1,575	3,290	12,620
Per cent increase...........	59·11	....	....	....	8·48	13·73	28·67	109·99
Decrease..................	....	11,474	806	340	....	....	....	12,620
Per cent decrease..........	....	100·00	7·03	2·96	....	....	....	109·99

* A-M-F coal: ash-and-moisture-free coal.

The yields as per cent of the ash-and-moisture-free coal are as follows:

—	Run 1	Run 2	Run 3	Mean of Runs 2 and 3
Oil... %	55·7	54·2	59·1	56·6
Combustible solid............................. %	9·9	8·5	8·5	8·5
Water.. %	12·8	13·4	13·7	13·5
Gas.. %	30·1	30·8	28·7	29·8
Hydrogen charged............................. %	7·3	7·5	7·0	7·3

CHAPTER XIII

LIGNITE FROM THE ONAKAWANA DISTRICT IN ONTARIO

Onakawana lignite is of lower rank than the other lignite tested (Bienfait). It is also of interest because the deposits are located in Ontario.

DESCRIPTION AND PRE-TREATMENT OF SAMPLE

The sample used had been in storage for some time at the Fuel Research Laboratories. It consisted of lumps of various sizes including fines.

After removal from storage, the bulk sample was crushed and passed over a ⅛-inch screen to remove the fines, which might ignite during the drying operation. After crushing and screening, the sample had a moisture content of 16·75 and an ash content of 7·58. This moisture content indicates that the sample had dried considerably in storage.

After crushing and screening, the bulk sample was dried in the usual way to a moisture content of about 5·7 per cent.

The grindability index of the dried lignite was 69·1.

A float and sink test was made on the dried lignite to determine the extent to which ash could be reduced by washing. Part of the data from this test is given below:

Specific gravity	Floats, per cent dry basis	Ash in floats, per cent dry basis
Less than 1·30	3·7	4·9
1·30 to 1·40	69·1	6·7
1·40 to 1·50	94·0	7·3
1·50 to 1·60	97·1	7·5
Greater than 1·60	100·0	8·1

It is apparent that the ash is largely inherent and cannot be greatly reduced by flotation. Accordingly, the bulk sample for the hydrogenation test was not washed.

The analyses of the bulk sample after drying are given below:

—		As used in tests	Dry basis	Ash and moisture-free basis
Proximate Analysis—				
Moisture	%	6·1	....	....
Ash	%	8·0	8·6	....
Volatile matter	%	43·3	46·1	50·4
Fixed carbon	%	42·6	45·3	49·6
Ultimate Analysis—				
Carbon	%	58·4	62·1	67·9
Hydrogen	%	4·8	4·4	4·8
Ash	%	8·0	8·6	....
Sulphur	%	0·8	0·8	0·9
Nitrogen	%	0·5	0·5	0·6
Oxygen	%	27·5	23·6	25·8
Calorific Value—				
B.T.U. per pound, gross		9,570	10,190	11,150

The analysis of the ash in the lignite as charged is given as follows:

	Per cent
SiO₂	19·15
Al₂O₃	12·38
Fe₂O₃	12·83
TiO₂	0·25
CaO	23·16
MgO	6·41
Na₂O	2·14
K₂O	0·58
SO₃	22·73
MnO	0·03
P₂O₅	0·10
Total	99·76

HYDROGENATION TEST

The hydrogenation test consisted of three runs using the equipment and method previously described.

Characteristics of Materials Charged. The lignite for each run was taken from the dried bulk sample and pulverized separately. The sieve analyses of the lignite as pulverized for the three runs are given below.

U.S. Series Equivalent No.		Run 1	Run 2	Run 3
Through 16 on 30 mesh	%	0·0	0·0	0·0
" 30 " 50 "	%	0·0	0·0	0·0
" 50 " 100 "	%	6·7	6·5	14·1
" 100 " 140 "	%	14·4	14·6	16·6
" 140 " 200 "	%	15·1	15·1	12·7
" 200 " 300 "	%	18·3	20·0	15·2
" 300 mesh	%	45·5	43·8	41·4
Total	%	100·0	100·0	100·0

The ash and moisture contents were determined from samples taken after the lignite was pulverized. They are shown below.

—		Run 1	Run 2	Run 3
Moisture	%	5·65	6·60	6·11
Ash	%	8·17	7·44	8·53
Ash on dry basis	%	8·66	7·97	9·09

The vehicle used in each of the runs was, as usual, the fraction of the product of the preceding run boiling above 446° F. (230° C.) in the large still with steam. The vehicle for the first run was made from the product of the third run with Bienfait lignite.

The distillation ranges of the vehicles, determined with the Hempel apparatus, are given below:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water...	0·4	0·4	0·2
Up to 338° F. (170° C.).....................	0·3	0·3	0·6
338 to 446° F. (170 to 230° C.).............	13·7	13·9	16·6
446 to 572° F. (230 to 300° C.).............	35·5	34·7	34·0
Above 572° F. (300° C.)....................	49·9	50·7	47·8
Loss...	0·2	0·0	0·8

The viscosities of the vehicles are:

—	Run 1	Run 2	Run 3
Saybolt Universal at 100° F..............................	76	74	72

The catalyst in each run, as usual, was stannous oxide, in amount equal to 5 per cent of the lignite charged.

The pitch charged to the reaction chamber before the run was, in each case, produced in the preceding run. The pitch for Run 1 was from Run 3 on Bienfait lignite. The proportions of solids insoluble in carbon tetrachloride in the pitches and of ash in the insoluble solids are shown below:

—		Run 1	Run 2	Run 3
Solids insoluble in CCl₄.............................. %		43·14	48·14	46·22
Ash in insoluble solids.............................. %		59·0	56·4	60·1

The average purity of the hydrogen charged, as read from the recorder chart, is shown below:

—	Run 1	Run 2	Run 3
Average per cent H₂...	97·8	97·7	97·8

Operating Conditions. The temperature, pressure, charging rate, rate of recirculation of hydrogen, and duration of each of the runs, are shown below.

—		Run 1	Run 2	Run 3
Average wall temperature, thermocouples 4 and 8.... { °F.		828	817	831
°C.		442	436	444
Average temperature inside reaction chamber*....... { °F.		768	779	793
°C.		409	415	423
Pressure, lb./sq. in...................................		3,000	3,000	3,000
Average charging rate, lb. paste/hr......................		8·5	8·5	8·5
Rate of hydrogen recirculation, at temperature and pressure of reaction chamber, cu. ft./hr......................		5	5	5
Duration of run, hours...............................		6·67	6·58	6·58

*One thermocouple near middle of reaction chamber.

Analyses of samples of the gas recirculating through the reaction chamber at the beginning and end of the charging period of each run are given below.

		Run 1		Run 2		Run 3	
		Start	End	Start	End	Start	End
Carbon dioxide	%	0·76	10·14	1·78	10·23	2·51	10·65
Oxygen	%	0·54	0·39	0·22	0·11	0·10	0·11
Illuminants	%	0·22	0·49	0·18	0·57	0·19	0·22
Carbon monoxide	%	2·39	7·87	2·18	7·05	2·04	7·52
Hydrogen	%	88·74	61·02	86·40	57·60	84·90	54·82
Methane	%	1·07	11·12	4·72	15·83	4·60	21·11
Ethane	%	0·37	5·12	0·34	3·62	0·33	3·07
Nitrogen (by difference)	%	5·91	3·85	4·18	4·99	5·33	2·50

Characteristics and Quantities of Materials Produced. The results of the distillations of the liquid products in the large still with steam are given as:

	Per cent by weight		
	Run 1	Run 2	Run 3
Water	11·2	12·1	11·7
Up to 338° F. (170° C.)	4·2	5·5	8·5
338 to 446° F. (170 to 230° C.)	2·6	2·3	3·2
Above 446° F. (230° C.)	62·7	67·4	68·2
Coke	16·4	10·6	6·3
Loss	2·9	2·1	2·1

The results of the analytical distillations of the liquid products in the Hempel apparatus are:

	Per cent by weight		
	Run 1	Run 2	Run 3
Water	11·4	12·1	12·5
Up to 338° F. (170° C.)	4·9	7·0	9·0
338 to 446° F. (170 to 230° C.)	11·8	13·7	13·5
446 to 572° F. (230 to 300° C.)	22·7	23·0	24·8
Residuum	48·8	43·9	39·9
Loss	0·4	0·3	0·3

The results of extraction of the liquid products and pitches with carbon tetrachloride, and the determination of ash in the residues are given below:

		Liquid product			Pitch		
		Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
Solids insoluble in CCl₄	%	9·95	6·68	3·61	48·14	46·22	4·28
Ash in insoluble solids	%	61·8	62·9	61·0	56·4	60·1	63·9

The material balances of the three runs are given in Table I.

TABLE I

Material Balance, Quantities in Grammes

—	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Run 1								
Vehicle.....................	13,116					53		13,169
Coal........................		11,338		1,080		751		13,169
Catalyst....................				659				659
Pitch.......................	5,235			2,343	1,628			9,206
Hydrogen...................			840					840
Total input.........	18,351	11,338	840	4,082	1,628	804		37,043
Liquid product.............	18,280			1,436	888	2,663	93	23,360
Pitch.......................	5,111			2,676	2,068			9,855
Recovery....................								33,215
Gas and loss...............							3,828	3,828
Total output.......	23,391			4,112	2,956	2,663	3,921	37,043
Increase....................	5,040			30	1,328	1,859	3,921	12,178
Per cent increase...........	44.45			0.27	11.71	16.40	34.58	107.41
Decrease....................		11,338	840					12,178
Per cent decrease...........		100.00	7.41					107.41
Run 2								
Vehicle.....................	12,873					52		12,925
Coal........................		11,116		956		853		12,925
Catalyst....................				647				647
Pitch.......................	4,575			2,395	1,851			8,821
Hydrogen...................			849					849
Total input.........	17,448	11,116	849	3,998	1,851	905		36,167
Liquid product.............	17,933			931	549	2,681	66	22,160
Pitch.......................	5,581			2,883	1,914			10,378
Recovery....................								32,538
Gas and loss...............							3,629	3,629
Total output.......	23,514			3,814	2,463	2,681	3,695	36,167
Increase....................	6,066				612	1,776	3,695	12,149
Per cent increase...........	54.57				5.51	15.98	33.24	109.30
Decrease....................		11,116	849	184				12,149
Per cent decrease...........		100.00	7.64	1.66				109.30

* A-M-F coal: ash and moisture-free coal.

TABLE I—*Concluded*
Material Balance, Quantities in Grammes—*Concluded*

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Run 3								
Vehicle	13,241					27		13,268
Coal		11,331		1,128		809		13,268
Catalyst				663				663
Pitch	4,710			2,433	1,615			8,758
Hydrogen			826					826
Total input	17,951	11,331	826	4,224	1,615	836		36,783
Liquid product	18,874			497	318	2,823	68	22,580
Pitch	5,429			3,238	1,830			10,497
Recovery								33,077
Gas and loss							3,706	3,706
Total output	24,303			3,735	2,148	2,823	3,774	36,783
Increase	6,352				533	1,987	3,774	12,646
Per cent increase	56·06				4·70	17·54	33·31	111·61
Decrease		11,331	826	489				12,646
Per cent decrease		100·00	7·29	4·32				111·61

* A-M-F coal: ash-and-moisture-free coal.

The yields as per cent of the ash-and-moisture-free coal are as follows:

—		Run 1	Run 2	Run 3	Mean of Runs 2 and 3
Oil	%	44·5	54·6	56·1	55·4
Combustible solid	%	11·7	5·5	4·7	5·1
Water	%	16·4	16·0	17·5	16·8
Gas	%	34·6	33·2	33·3	33·3
Hydrogen charged	%	7·4	7·6	7·3	7·5

CHAPTER XIV

PEAT FROM ALFRED, ONTARIO

Alfred peat was selected for a comparative hydrogenation test in order to complete the study of the solid fuels of low rank. The peat from this particular district is especially suitable for such a test because it has been produced on a large scale and its characteristics have been thoroughly investigated.

DESCRIPTION AND PRE-TREATMENT OF SAMPLE

The sample used had been in storage for some time at the Fuel Research Laboratories. It consisted of lumps of various sizes, brown in colour, earthy in appearance, and very soft, with some woody material. No surface moisture was present. The moisture in the sample as removed from storage was only 13·95 per cent, which indicates that the peat had dried considerably. The ash content was 3·57 per cent on the wet basis.

The bulk sample was dried in lump form in order to minimize the possibility of ignition. The moisture content of the dried peat was 5·4 per cent.

The grindability index of the dried peat was 42·1.

No washing test was made because the ash content was low.

The analyses of the peat after drying are given as follows:

—		As used in tests	Dry basis	A-M-F basis
Proximate Analysis—				
Moisture	%	5·4	....	....
Ash	%	4·2	4·4	....
Volatile matter	%	61·4	65·0	68·0
Fixed carbon	%	29·0	30·6	32·0
Ultimate Analysis—				
Carbon	%	53·2	56·3	58·9
Hydrogen	%	5·9	5·6	5·9
Ash	%	4·2	4·4	....
Sulphur	%	0·2	0·2	0·2
Nitrogen	%	2·0	2·1	2·2
Oxygen	%	34·5	31·4	32·8
Calorific Value—				
B.T.U. per pound, gross		9,180	9,710	10,160

The analysis of the ash in the peat as charged is given as:

	Per cent
SiO$_2$	25·81
Al$_2$O$_3$	8·12
Fe$_2$O$_3$	10·43
TiO$_2$	0·17
CaO	31·02
MgO	13·54
Na$_2$O	0·37
K$_2$O	0·67
SO$_3$	8·95
MnO	0·04
P$_2$O$_5$	1·34
Total	100·46

HYDROGENATION TEST

The hydrogenation test consisted of three runs, using the equipment and method previously described.

Characteristics of Materials Charged. The peat for each run was taken from the dried bulk sample and pulverized separately. The tests proved that peat required much more grinding than the other fuels, for instance, peat ground for 4½ hours and screened through a $\frac{1}{16}$-inch screen was too coarse to be pumped after mixing with the vehicle. The samples for the test had to be ground for 26 hours or more, with only 6 pounds instead of 12 in each mill, to be fine enough to be pumped. For comparison, the dried Onakawana lignite was ground only 2½ hours, with 12 pounds in each mill. The screen analyses of the peat, as pulverized for the three runs, are given below. The sample for the first run was ground in two batches, for 26 and 29 hours respectively, with about 6 pounds in each mill. The samples for the other two runs were each ground for 32 hours, with about 10 pounds in each mill.

U.S. Series Equivalent No.	Run 1		Run 2	Run 3
	26 hrs.	29 hrs.	32 hrs.	32 hrs.
On 16 mesh	0·4	0·0	0·0	0·0
Through 16 on 30 mesh	0·0	0·0	0·0	0·0
" 30 " 50 "	0·0	0·0	6·6	0·0
" 50 " 100 "	2·6	2·0	14·3	14·9
" 100 " 140 "	6·6	42·9	18·7	13·7
" 140 " 200 "	33·4	24·0	27·4	19·4
" 200 " 300 "	37·3	19·9	19·8	25·5
" 300 mesh	19·7	11·2	13·2	26·5
Total	100·0	100·0	100·0	100·0

The ash and moisture contents were determined from samples taken after the peat was pulverized. They are shown below:

—		Run 1	Run 2	Run 3
Moisture	%	5·0	4·9	5·1
Ash	%	4·5	6·5	4·2
Ash on dry basis	%	4·7	6·9	4·4

The vehicle for Run 1 was prepared in an unusual way. The run was interrupted when the coarse peat, which had been ground for 4½ hours, could not be pumped. Accordingly, the paste that had been prepared was distilled in the large still with steam to recover as much of the vehicle as possible. To this was added a mixture of the oil distillates boiling above 446° F. (230° C.) from a number of previous runs, amounting to about 43 per cent of the total. The vehicles for Runs 2 and 3 were, as usual, the fractions of the products of the preceding runs boiling above 446° F. (230° C.) in the large still with steam.

The distillation ranges of the vehicles determined with the Hempe apparatus are given below:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water	4·7	0·5	0·2
Up to 338° F. (170° C.)	0·6	0·2	0·5
338 to 446° F. (170 to 230° C.)	9·7	13·8	15·1
446 to 572° F. (230 to 300° C.)	40·2	36·9	36·3
Above 572° F. (300° C.)	44·6	48·4	47·9
Loss	0·2	0·2	0·0

The viscosities of the vehicles are given as:

—	Run 1	Run 2	Run 3
Saybolt Universal at 100° F	72	74	70

The catalyst in each run, as usual, was stannous oxide, in amount equal to 5 per cent of the peat charged.

The pitch charged to the reaction chamber before the run was, in each case, produced in the preceding run. The pitch for the first run was produced in the third run on Onakawana lignite. The proportions of solids insoluble in carbon tetrachloride in the pitches and of ash in the insoluble solids are:

—	Run 1	Run 2	Run 3
Solids insoluble in CCl_4 %	48·28	42·69	46·19
Ash in insoluble solids %	63·9	77·1	79·0

The average purity of the hydrogen charged, as read from the recorder chart, is shown as:

—	Run 1	Run 2	Run 3
Average per cent H_2 %	98·5	97·6	97·8

Operating Conditions. A paste consisting of equal proportions of peat and vehicle was too viscous to pump. This is probably because peat has a lower density than coal and, therefore, has more interstitial space

per unit of weight. Accordingly, the ratio of the weight of peat to the weight of vehicle in each of the runs was 40 to 60. The temperature, pressure, charging rate, rate of recirculation of hydrogen, and duration of each of the runs are shown below:

—		Run 1	Run 2	Run 3
Average wall temperature, thermocouples 4 and 8	°F.	819	815	817
	°C.	437	435	436
Average temperature inside reaction chamber[1]	°F.	786	788	795
	°C.	419	420	424
Pressure, lb./sq. in.		3,000	3,000	3,000
Average charging rate, lb. paste/hr		8·1	8·2	8·2
Rate of hydrogen recirculation at temperature and pressure of reaction chamber, cu. ft./hr		5	5	5
Duration of run, hr.		6·57	6·25	6·33

[1] Average of three thermocouples near top, middle, and bottom of reaction chamber.

Analyses of samples of the gas recirculating through the reaction chamber at the beginning and end of the charging period of each run are given below:

—		Run 1		Run 2		Run 3	
		Start	End	Start	End	Start	End
Carbon dioxide	%	2·66	9·52	2·14	8·83	1·09	8·72
Oxygen	%	0·03	0·17	0·12	2·42	0·04	0·10
Illuminants	%	0·25	0·50	0·08	0·69	0·20	0·52
Carbon monoxide	%	1·71	6·61	2·47	7·15	1·80	6·22
Hydrogen	%	86·20	60·80	84·00	51·42	90·10	64·30
Methane	%	3·63	15·95	4·68	14·39	3·79	17·26
Ethane	%	0·38	2·59	0·56	3·16	0·23	2·05
Nitrogen (by difference)	%	5·14	3·86	5·95	11·94	2·75	0·83

Characteristics and Quantities of Materials Produced The results of the distillations of the liquid products in the large still with steam are given as follows:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water	14·7	12·1	11·4
Up to 338° F. (170° C.)	4·1	6·4	5·4
338 to 446° F. (170 to 230° C.)	2·8	3·2	3·8
Above 446° F. (230° C.)	69·8	73·8	71·7
Coke	5·9	2·2	4·5
Loss	2·7	2·3	3·2

The results of the analytical distillations of the liquid products in the Hempel apparatus are given as:

—	Per cent by weight		
	Run 1	Run 2	Run 3
Water	15·2	11·4	11·7
Up to 338° F. (170° C.)	5·0	7·5	6·7
338 to 446° F. (170 to 230° C.)	13·3	13·0	12·3
446 to 572° F. (230 to 300° C.)	24·9	29·8	29·5
Residuum	41·0	37·5	39·1
Loss	0·6	0·8	0·7

The results of extraction of the liquid products and pitches with carbon tetrachloride, and the determination of ash in the residues are given below:

—	Liquid product			Pitch		
	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
Solids insoluble in CCl₄ ... %	4·59	1·73	3·95	42·69	46·19	48·53
Ash in insoluble solids ... %	67·6	76·3	70·0	77·1	79·0	76·5

The material balances of the three runs are given below in Table I.

TABLE I
Material Balance, Quantities in Grammes

—	Oil	A-M-F* coal	Hydro-gen	Inor-ganic	Com-bustible solid	Water	Gas	Total
Run 1								
Vehicle	14,940			470		737		15,677
Coal		9,459		470		523		10,452
Catalyst				523				523
Pitch	4,697			2,802	1,583			9,082
Hydrogen			683					683
Total input	19,637	9,459	683	3,795	1,583	1,260		36,417
Liquid product	18,573			724	347	3,546	140	23,330
Pitch	5,513			3,166	941			9,620
Recovery								32,950
Gas and loss							3,467	3,467
Total output	24,086			3,890	1,288	3,546	3,607	36,417
Increase	4,449			95		2,286	3,607	10,437
Per cent increase	47·04			1·00		24·17	38·13	110·34
Decrease		9,459	683		295			10,437
Per cent decrease		100·00	7·22		3·12			110·34

* A-M-F coal: ash- and- moisture-free coal.

TABLE I—*Concluded*
Material Balance, Quantities in Grammes—*Concluded*

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total

Run 2

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Vehicle	14,671					74		14,745
Coal		8,702		642		486		9,830
Catalyst				492				492
Pitch	5,117			2,938	873			8,928
Hydrogen			656					656
Total input	19,788	8,702	656	4,072	873	560		34,651
Liquid product	18,574			285	88	2,460	173	21,580
Pitch	5,276			3,577	951			9,804
Recovery								31,384
Gas and loss							3,267	3,267
Total output	23,850			3,862	1,039	2,460	3,440	34,651
Increase	4,062				166	1,900	3,440	9,568
Per cent increase	46·68				1·91	21·83	39·53	109·95
Decrease		8,702	656	210				9,568
Per cent decrease		100·00	7·54	2·41				109·95

Run 3

—	Oil	A-M-F* coal	Hydrogen	Inorganic	Combustible solid	Water	Gas	Total
Vehicle	14,388					29		14,417
Coal		8,718		404		490		9,612
Catalyst				481				481
Pitch	4,778			3,240	861			8,879
Hydrogen			691					691
Total input	19,166	8,718	691	4,125	861	519		34,080
Liquid product	18,135			599	257	2,537	152	21,680
Pitch	4,630			3,339	1,027			8,996
Recovery								30,676
Gas and loss							3,404	3,404
Total output	22,765			3,938	1,284	2,537	3,556	34,080
Increase	3,599				423	2,018	3,556	9,596
Per cent increase	41·28				4·85	23·15	40·79	110·07
Decrease		8,718	691	187				9,596
Per cent decrease		100·00	7·93	2·14				110·07

* A-M-F coal: ash-and-moisture-free coal.

The yields as per cent of the ash-and-moisture-free coal are as follows:

—	Run 1	Run 2	Run 3	Mean of Runs 2 and 3
Oil.. %	47·0*	46·7	41·3	44·0
Combustible solid.......................... %	−3·1*	1·9	4·9	3·4
Water...................................... %	24·2	21·8	23·2	22·5
Gas.. %	38·1	39·5	40·8	40·2
Hydrogen charged.......................... %	7·2	7·5	7·9	7·7

* The apparent negative yield of combustible solid is due to liquefaction of some of the solid in the pitch, which was from the preceding run with Onakawana lignite.

CHAPTER XV

DISCUSSION OF RESULTS

In Chapters IV to XIV the tests have been described in chronological order. However, in the following discussion of the results, it is more convenient to number the coals in descending order of rank as shown in Table I.

TABLE I
Designation of Coals

No.	Province	District[1]	Chapter No.
1	British Columbia..................	Crowsnest............................	V
2	British Columbia..................	Vancouver Island.....................	VI
3	(Durham, England)................	(Durham, England).................	VII
4	Nova Scotia......................	Sydney..............................	IV
5	British Columbia..................	Nicola..............................	VIII
6	Alberta...........................	Saunders............................	IX
7	Alberta...........................	Drumheller..........................	X
8	Alberta...........................	Edmonton...........................	XI
9	Saskatchewan.....................	Bienfait............................	XII
10	Ontario...........................	Onakawana..........................	XIII
11	Ontario...........................	Alfred..............................	XIV

[1] An entire district is not always accurately represented by a single sample.

The system of classification of the American Society for Testing Materials[1], according to which the coals have been ranked, is shown in Table II.

[1] A.S.T.M. Designation D 388-38.

TABLE II
Classification of Coals by Rank*

Class	Group	Limits of fixed carbon or B.T.U. Mineral-matter-free basis	Requisite physical properties
I. Anthracitic.........	1. Meta-anthracite...........	Dry F.C., 98 per cent or more (Dry V.M., 2 per cent or less)	Non-agglomerating[1]
	2. Anthracite................	Dry F.C., 92 per cent or more and less than 98 per cent (Dry V.M., 8 per cent or less and more than 2 per cent)	
	3. Semi-anthracite............	Dry F.C., 86 per cent or more and less than 92 per cent (Dry V.M., 14 per cent or less and more than 8 per cent)	

F.C.=Fixed carbon.   V.M.=Volatile matter.   B.T.U.=British thermal units.

* This classification does not include a few coals which have unusual physical and chemical properties and which come within the limits of fixed carbon or B.T.U. of the High-volatile bituminous and sub-bituminous ranks. All of these coals either contain less than 48 per cent dry, mineral-matter-free fixed carbon, or have more than 15,500 moist, mineral-matter-free B.T.U.

[1] If agglomerating, classify in Low-volatile group of the bituminous class.

TABLE II—*Concluded*

Classification of Coals by Rank—*Concluded*

Class	Group	Limits of fixed carbon or B.T.U. Mineral-matter-free basis	Requisite physical properties
II. Bituminous[a]	1. Low-volatile bituminous coal.	Dry F.C., 78 per cent or more and less than 86 per cent (Dry V.M., 22 per cent or less and more than 14 per cent)	
	2. Medium-volatile bituminous coal.	Dry F.C., 69 per cent or more and less than 78 per cent (Dry V.M., 31 per cent or less and more than 22 per cent)	
	3. High-volatile A bituminous coal.	Dry F.C., less than 69 per cent (Dry V.M., more than 31 per cent); and moist[b] B.T.U., 14,000[d] or more.	
	4. High-volatile B bituminous coal.	Moist[b] B.T.U., 13,000 or more and less than 14,000[d].	
	5. High-volatile C bituminous coal	Moist B.T.U., 11,000 or more and less than 13,000[d].	Either agglomerating or non-weathering[e].
III. Sub-bituminous	1. Sub-bituminous A coal	Moist B.T.U., 11,000 or more and less than 13,000[d].	Both weathering and non-agglomerating.
	2. Sub-bituminous B coal	Moist B.T.U., 9,500 or more and less than 11,000[d].	
	3. Sub-bituminous C coal	Moist B.T.U., 8,300 or more and less than 9,500[d].	
IV. Lignitic	1. Lignite	Moist B.T.U., less than 8,300	Consolidated
	2. Brown coal	Moist B.T.U., less than 8,300	Unconsolidated

F.C.=Fixed carbon.   V.M.=Volatile matter.   B.T.U.=British thermal units.

[b]Moist B.T.U. refers to coal containing its natural bed moisture but not including visible water on the surface of the coal.

[a]It is recognized that there may be non-caking varieties in each group of the bituminous class.

[d]Coals having 69 per cent or more fixed carbon on the dry mineral-matter-free basis shall be classified according to fixed carbon, regardless of B.T.U.

[e]There are three varieties of coal in the High-volatile C bituminous coal group, namely: Variety 1, agglomerating and non-weathering; Variety 2, agglomerating and weathering; Variety 3, non-agglomerating and non-weathering.

The ranks of the coals according to this classification are given in Table III and are shown graphically in Figure 3. The equilibrium moisture, determined at 86° F. (30° C.) and 100 per cent humidity (extrapolated), as well as the fixed carbon on the dry, mineral-matter-free basis,

TABLE III

Rank of Coals by A.S.T.M. Classification

Coal No.	Class	Group	Mineral-matter-free		Moisture, 100 per cent humidity
			Dry, F.C.	Moist, B.T.U.	
1	Bituminous	Medium volatile	72·7	15,350	1·0
2	Bituminous	High-volatile A	63·2	14,950	2·3
3	Bituminous	High-volatile A	64·1	14,680	3·6
4	Bituminous	High-volatile A	60·9	14,660	3·5
5	Bituminous	High-volatile B	58·2	13,210	7·4
6	Bituminous	High-volatile C	62·9	12,350	9·1
7	Sub-bituminous	B	58·9	10,410	17·5
8	Sub-bituminous	C	59·1	9,090	24·0
9	Lignitic	Lignite	57·2	8,300	31·7
10	Lignitic	Brown coal	54·9	6,700	35·6*
11	Peat		32·6	6,540	34·4*

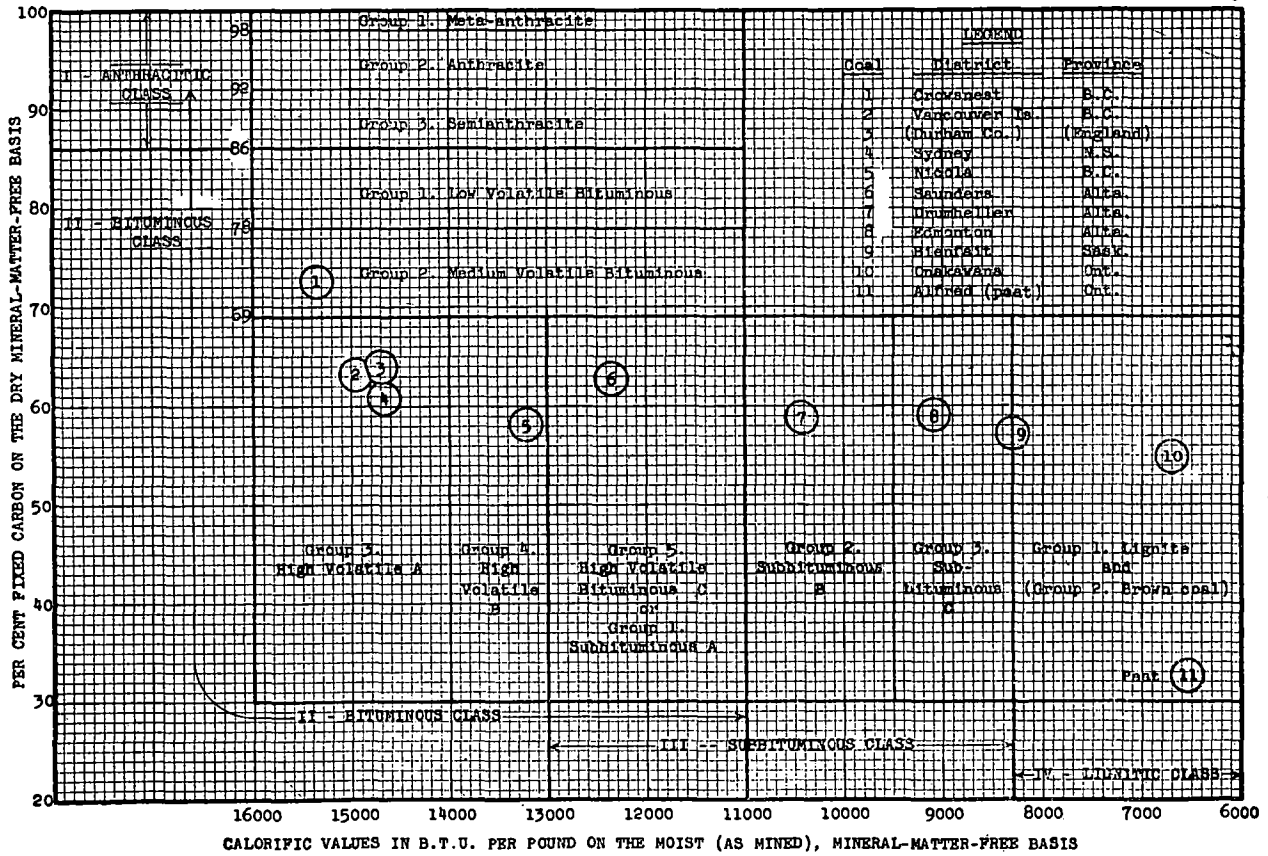* Determinations by dehydration on different samples from the same district.

Figure 3.　Position of the coals tested in the A.S.T.M. classification by rank.

and the calorific value on the moist, mineral-matter-free basis, have been included in the table to indicate the relative positions of the coals within the groups. Coals Nos. 1 to 4 are caking, No. 5 is weakly caking, and the others are both non-caking and non-agglomerating. No. 6 is ranked as High-volatile bituminous C because of the non-weathering properties of coals from this field.

The standard operating conditions which have been employed throughout the tests were first adopted as a result of previous work with other equipment and preliminary tests using the present plant. It was considered advisable, however, before making an extensive survey of different coals, to investigate very briefly the effect of altering the major operating conditions. Accordingly, after the standard test had been applied to coal No. 4 from the Sydney district (Chapter IV), further experiments were made by altering in turn the pressure, rate of charging, temperature, and catalyst. Coal No. 1 from the Crowsnest district (Chapter V) was also tested at different temperatures and with a different catalyst.

It was found that the reaction with the Sydney coal was not greatly affected by a reduction in pressure of about 500 pounds per square inch (Run 4, Chapter IV). However, decreasing the rate of charging by about 30 per cent, or increasing the temperature 25° F. (14° C.), lowered the yield of oil to about 70 per cent of that obtained under the usual conditions (Runs 6 and 9, Chapter IV).

The experiments on catalysts with the Sydney coal consisted of two consecutive runs in which no catalyst was charged with the paste, followed by two in which lead acetate was charged. All four of these runs (10, 11, 13, 14, page 28) gave lower yields of oil than those in which stannous oxide was used. Hydrochloric acid, as a catalytic agent, was tested with the Crowsnest coal (Runs 7 and 8, Chapter V). Its use resulted in an increased consumption of hydrogen, but the results were inconclusive because of difficulties in analysis.

These experiments were of such short duration that they serve only to show the trends resulting from changes in the conditions. As, however, they did not indicate any way in which the standard operating conditions could be improved these were not altered and have been maintained substantially as described in Chapter III.

In Table IV, the average yields from two or three runs with each coal are shown in relation to some of the properties of the coals.

Of the properties shown in Table IV, rank appears to have the major influence on the results. The oil yield increases with descending rank to a maximum with coal No. 4, and then decreases progressively. Coal No. 6 is the only exception to this general trend. The yields of combustible solid are comparatively high in coals Nos. 1, 2, and 6. This would normally be expected of coal No. 1, because of its high rank, but coals Nos. 2 and 6 are exceptional in this respect. The yields of water increase with descending rank, as would be expected from the increasing oxygen content of the coals. The yields of gas are lowest for coals Nos. 4 and 5, which factor contributes to their high yields of oil. For coals of both higher and lower rank, the yields of gas are progressively greater. The consumption of hydrogen is highest for coal No. 4, but is also high for the sub-bituminous coals, lignites, and peat.

TABLE IV

Comparison of Properties and Yields

Coal No	1	2	3	4	5	6	7	8	9	10	11
A.S.T.M. class	.... Bituminous						.. Sub-bituminous ..		...... Lignitic		.. Peat ..
A.S.T.M. group	Medium volatile	High-volatile A	High-volatile A	High-volatile A	High-volatile B	High-volatile C	B	C	Lignite	Brown Coal	
Geological period	Lower Cretaceous	Upper Cretaceous	Carbonaceous	Carbonaceous	Tertiary	Cretaceous	Upper Cretaceous	Upper Cretaceous	Tertiary	Tertiary	Tertiary
Nitrogen, per cent as charged	1·5	1·2	1·7	1·7	1·6	1·1	1·6	1·4	1·3	0·5	2·0
Carbon, A-M-F	88·1	85·2	85·6	84·2	80·1	80·0	75·3	73·6	74·8	67·9	58·9
Mean yields (per cent of A.M.F. coal)—											
Oil	62·0	66·8	71·5	77·2	73·8	53·3	62·5	57·7	56·6	55·4	44·0
Combustible solid	13·2	16·8	9·2	5·7	6·4	21·9	5·1	7·3	8·5	5·1	3·4
Water	2·0	2·3	4·2	7·4	7·4	8·6	13·3	13·4	13·5	16·8	22·5
Gas	27·8	18·3	19·9	17·3	16·8	21·5	26·3	29·2	29·8	33·3	40·2
Hydrogen charged, per cent of A-M-F. coal	4·7	4·7	5·7	8·0	5·5	5·0	6·9	7·6	7·3	7·5	7·7

It is difficult to evaluate the effects of characteristics other than rank, such as the geological age, the composition of the ash, and the proportions of nitrogen and sulphur, because they are largely obscured by the effects of rank. The exceptional behaviour of coals Nos. 2 and 6, however, gives some indication of the influence of these properties.

Some of the coals from Western Canada have been metamorphosed more rapidly than those from Nova Scotia and England, and, therefore, have attained the same rank in a shorter time. Thus, coal No. 2 is younger than coals Nos. 3 and 4, although they are all classified in the same group. The yield of combustible solid from coal No. 2 was higher, and the yield of oil lower than those from the other two coals, but more data would be required to justify the general conclusion that coals that have been rapidly metamorphosed are not easily liquefied.

Analyses of the ash constituents of all of the coals have been given previously. These are reduced to a common basis of comparison in Table V. The figures in the table are percentages of the indicated constituents in the coal as charged.

TABLE V

Ash Constituents as Per Cent of Coal Charged

Coal No.	1	2	3	4	5	6	7	8	9	10	11
SiO₂	1·58	1·67	0·93	0·77	3·70	1·94	2·95	3·60	2·67	1·53	1·08
Al₂O₃	1·01	0·82	0·48	0·53	2·49	1·05	1·51	2·38	1·75	0·99	0·34
CaO	0·26	1·18	0·07	0·20	0·16	1·92	0·89	1·33	1·94	1·85	1·30
MgO	0·03	0·09	......	0·03	0·08	0·18	0·14	0·14	0·49	0·51	0·57
Fe₂O₃	0·46	2·00	0·80	1·06	0·55	0·65	0·50	0·45	0·66	1·03	0·44
TiO₂	0·04	0·09	0·04	0·03	0·09	0·01	0·02	0·05	0·05	0·02	0·01
Na₂O	0·02	0·05	0·03	0·12	0·02	0·18	0·52	0·44	0·97	0·17	0·02
K₂O	0·02	0·04	0·02	0·01	0·09	0·03	0·03	0·04	0·05	0·05	0·03
MnO	.....	.....	.....	0·01	.....	.....	.....	.....	.....	.....	.....
P₂O₅	0·10	0·14	0·01	0·01	0·08	0·14	0·10	0·24	0·09	0·01	0·06
SO₃	0·08	1·40	0·12	0·14	0·03	0·68	0·82	0·59	1·10	1·82	0·38
Total ash	3·6	7·5	2·5	3·0	7·3	6·8	7·5	9·3	9·8	8·0	4·2

There is no evidence of catalytic activity in any constituent of the ashes. The ash of coals Nos. 2 and 6 might have been expected to hinder liquefaction. Yet, each ash constituent, of which both have a high proportion, is also present in high proportion in one or more of the other coals. On the other hand, there is no evidence of any catalytic action of the ash assisting liquefaction of the other coals. Coal No. 4, which gave the best yield of oil, has less of every constituent, excepting manganese oxide, than one or the other of coals Nos. 2 and 6. Manganese is present only in very small amounts, and in about equal proportion, in coals Nos. 5 and 6, which gave respectively high and low yields of oil.

Variation in the proportion of sulphur did not seem to have any consistent effect on the yields, but there is an apparent relationship between the nitrogen content of the coal and the yield of combustible solid. The coals that have high nitrogen contents in general give low yields of combustible solids and correspondingly high yields of oil. No. 10 is an ex-

ception to this rule in that it has a low nitrogen content and yet gives a low yield of combustible solids.

From the foregoing data, it appears that the best coals for hydrogenation are in the High-volatile A and B groups of the bituminous class, and are high in nitrogen.

A sample of the coal processed in the commercial hydrogenation plant of Imperial Chemical Industries at Billingham, England (No. 3), has been included in the series of tests as a standard of comparison for the Canadian coals. In comparing the coals, the amount of primary oil product has been taken as an index of their suitability for the production of gasoline. The complete process requires a second stage of hydrogenation in which some losses are incurred, so that the yields from the present tests are higher than they would have been if the final product had been gasoline.

The yields of primary oil are shown in Table VI as Imperial gallons per short ton of dry and ash-free coal. The average specific gravity of the primary oils has been taken as 1·0 in preparing this table.

TABLE VI

Yields of Primary Oil

	Designation of Coal		Imperial gallons per 2,000 lb., dry and ash-free basis
No.	Location	Rank	
1	Crowsnest............B.C.	Medium-volatile bituminous............	124
2	Vancouver Island......B.C.	High-volatile bituminous A.............	134
3	Durham..............Eng.*	" " A.............	143
4	Sydney...............N.S.	" " A.............	154
5	Nicola...............B.C.	" " B.............	148
6	Saunders.............Alta.	" " C.............	107
7	Drumheller..........Alta.	Sub-bituminous B......................	125
8	Edmonton...........Alta.	" C.....................	115
9	Bienfait.............Sask.	Lignite................................	113
10	Onakawana..........Ont.	Lignite................................	111
11	Alfred...............Ont.	Peat..................................	88

* Standard of comparison.

On the dry and ash-free basis all the coals, excepting the peat, yielded more than 100 gallons of primary oil per short ton. The highest yield was 154 gallons per ton, and the lowest 107 gallons. It should be noted, however, that the dry basis favours the low-rank coals, which would be handicapped commercially by the expense of drying. The ash-free basis favours the coals high in ash, and especially those having a high content of inherent ash. These would have the added expense of washing before the liquefaction process, or be subject to a considerable loss and expense in removing ash from the primary liquid product.

It will be noticed that two of the Canadian coals, from the Sydney district in Nova Scotia and the Nicola district in British Columbia respectively, gave higher yields of oil than the standard English coal. The sample of Sydney coal tested was particularly well suited for the process because of its low inherent ash.

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