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# FUEL RESEARCH INSTITUTE

OF SOUTH AFRICA

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TECHNICAL

MEMORANDUM

NO. 2 OF 1975

POSSIBLE ALTERNATIVES TO THE CONVENTIONAL  
OIL-FROM-COAL PROCESS.

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## S Y N O P S I S

After briefly discussing the conventional oil-from-coal process (Sasol), an account is presented of investigations, mainly in the United States of America, on alternative oil-from-coal processes, and a summary is given of the work done at the F.R.I. on fluidized-bed carbonization of coal.

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1. INTRODUCTION

The world we live in today is one with a seemingly insatiable demand for energy. This rightly is so because without an increasing consumption of energy no progress, with regard to both technology and society in general, is conceivable. Different sources of energy are available, for example rivers and tidal forces (which may be utilized to generate electricity), wind, the sun itself, nuclear energy, fossil fuels, etc. Fossil fuels are the source of energy used to a great extent at present. Fossil fuels are those fuels of organic origin found in the mantle of the earth, namely coal, natural oil and natural gas.

It is a human characteristic of man that he wishes to make life as convenient as possible for himself; this also applies to the use of fuels. Liquid fuels are convenient sources of energy, and with the exploitation of crude oil sources the use of oil and natural gas became more and more important. This resulted in the state of affairs in which the world relied increasingly on crude oil to satisfy its energy needs. Despite the fact that the world's energy reserve in coal is much larger than that in natural oil and gas, coal became more unpopular because it was less convenient to handle and produced unwanted ash. The other energy sources were virtually left out of the picture. With the realization that the supplies of crude oil and natural gas were becoming depleted rather rapidly, the other energy sources (coal, nuclear energy, tidal force, etc.) assumed renewed importance.

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\* A talk presented at an industrial chemistry students' seminar at the University of the Witwatersrand on 6th March, 1975.

Because the world has become accustomed to the convenience of using oil and gas as energy sources and because they are still generally available, these fuels will continue to play an important role in the future. When crude oil and natural gas become scarce and thus very expensive, the demand for oil and gas could conceivably still be partly satisfied by converting coal to oil and gas.

One fact that should be borne in mind is that it is possible, but only while coal is available, to manufacture a substitute for oil and natural gas.

## 2. COAL

Glenn and Rose<sup>1)</sup> describe coal as follows:- "Coal is a combustible solid resulting from the accumulation and burial of vegetation in previous geological ages. Subsequent pressure and temperature effects have altered these deposits to coal. The microscopic examination of coal reveals plant cell structures in abundant detail. The degree of coalification determines rank in the natural series with increasing carbon content - lignite, sub-bituminous coal, bituminous coal, and anthracite.

"Coal is composed chiefly of carbon, hydrogen and oxygen, with minor amounts of nitrogen and sulphur, and varying amounts of moisture and mineral impurities. Coal is neither carbon nor hydrocarbon. It consists chiefly of three-dimensional condensed cyclic structures of high molecular mass in which six- and five-membered carbocyclic and heterocyclic rings predominate. The proportion of carbon in ring structures increases with rank."

Depending on the rank of a coal, it contains from about 75 to more than 92,5 per cent carbon, about 3 to 6 per cent hydrogen, about 3 to 20 per cent oxygen, and about 0,5 to 4 per cent of nitrogen and sulphur.

Coal is generally regarded as a fuel, but its most important asset is that it is a major source of carbon.

## 3. CONVENTIONAL OIL-FROM-COAL PROCESS

The best known and the only commercial oil-from-coal process in the western world is the one used by Sasol. Sasol produces oil from coal basically in two stages<sup>2)</sup>- firstly, the production of purified gas from coal, and secondly

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the synthesis of liquid fuels from such gas. The Sasol process can be briefly described as follows: Synthesis gas, a mixture of hydrogen and carbon monoxide in specific proportions, is produced by gasifying coal in pressure gasifiers by means of steam and oxygen. From these gasifiers, distillation products such as ammonia and creosote are also obtained. The crude synthesis gas is purified in units where unwanted components are removed. The purified gas mixture is the raw material for a synthesis process.

Two synthesis processes are applied. In one of them (Synthol process) a powdered catalyst is swept along by the gas stream; in the other process (Arge) the gas passes through catalyst granules packed into a fixed bed in a reactor. The Synthol process produces mainly low boiling-point materials such as propane, butane, petrol, and considerable amounts of chemicals such as benzol, toluol, heavy naphta, acetone, alcohol, etc. The Arge process produces mainly higher boiling materials such as waxes, fuel oil, kerosene, diesel oil and smaller amounts of petrol, propane, butane and chemicals.

The unique combination of the two processes yields virtually the full range of products normally derived from crude oil, in addition to the oxygenated products usually manufactured in petrochemical plants. These processes also provide raw materials for producing nitrogenous fertilizers, butadiene and styrene for synthetic rubber manufacture.

#### 4. POSSIBLE ALTERNATIVE OIL-FROM-COAL PROCESSES

According to Harrison<sup>3)</sup>, the most striking difference in chemical composition of coal and oil is in the hydrogen content. In general, the higher the rank of a coal, the less hydrogen it contains. A typical petrol contains about 14 per cent of hydrogen, whereas coal contains only about 5 per cent of hydrogen on a dry ash-free basis. Thus, when converting coal to oil the proportion of hydrogen has to be increased.

Active research is being carried out, mainly in the United States of America, to produce oil from coal. To co-ordinate research on coal in the U.S.A., the Office of Coal Research (O.C.R.) was established as a section of the Department of the Interior. Private enterprise is supported financially by O.C.R. to carry out research on different aspects of coal utilization. One of the main projects is the one dealing with synthetic

liquid/...

liquid fuels from coal. It should be pointed out that the O.C.R. itself does not undertake any research - it merely acts as co-ordinator and leader.

According to Roger C.B. Morton<sup>4)</sup>, U.S. Secretary for the Interior, " ... coal liquefaction processes are expected to make major contributions to cleaner air standards, conserve chemicals prior to combustion, and help meet the increasing demand for petroleum and liquefied fuels with reduced reliance on imported fuels. The coal-to-liquids programme, though still in the pilot stage of testing, is already paying dividends in a practical way."

The following programmes are sponsored by the O.C.R., namely C.O.E.D., H-Coal, Solvent Refined Coal, C.S.F., Clean coke, Intermediate Coal Hydrogenation.

In addition or supplementary to these projects, other coal liquefaction processes have been developed or are being considered by different American firms or bodies. They include the following: United States Bureau of Mines, Gulf Research and Development Co., Exxon Research and Engineering Co., COGAS Development Co., Edison Electric Institute and Southern Company System, Coalcon, and Universal Oil Products Co.

A brief description of the different processes and the latest state of development is given below.

#### 4.1 Main coal liquefaction processes sponsored by O.C.R.

##### 4.1.1 COED (Char Oil Energy Development)<sup>5,6)</sup>

This process, developed by the F.M.C. Corporation, has reached the stage where a 36 t/d pilot plant at Princeton, N.J., operated successfully over extended periods of time and with different coal feedstocks. According to O.C.R., this is the most advanced coal-to-oil conversion process at present. The process converts coal to high-grade synthetic crude oil, pipeline quality gas, and char, by pyrolysis of coal in multi-stage fluidized beds. The yield of crude oil is about 22 per cent by mass.

The synthetic oil was tested successfully, as Project Seacoal I, by the U.S. Navy in the boilers of a destroyer during a 24 h voyage at sea during November, 1973.

An American firm, Ralph M. Parsons Co., is under contract to the O.C.R. to start a commercial design for a COED complex.

#### 4.1.2 H-Coal process<sup>7,8)</sup>

Hydrocarbon Research Incorporated developed the H-Coal process for converting coal to liquid fuel. A test unit at Trenton, N.J., having a capacity of 6 t of coal per day, is available for studies. The O.C.R. announced during November, 1974 that a 600 t/d pilot plant was to be erected in Catlettsbury, Ky.

In this process, coal is catalytically hydrogenated in an oil suspension in an ebullated bed reactor system. The synthetic crude oil is processed to petrol and fuel oil. The yield of liquid products is about 550 l per ton of coal, that is, about 47 per cent by mass.

#### 4.1.3 Solvent Refined Coal (S.R.C.) process<sup>9,10,11,12)</sup>

This process, originally investigated by Spencer Chemicals Corp., was developed further by the Pittsburg and Midway Coal Mining Co. (P. & M.), instructed by the Gulf Oil Corporation. P. & M. have proved that the S.R.C. process is technically feasible. The erection of a pilot plant to treat 50 t of coal per day was started at Tacoma (Fort Lewis), Wa., and was scheduled for completion during February, 1974.

The process is briefly as follows: Raw coal is pulverized and mixed with coal-based solvent. The "slurry" is heated to 437°C in the presence of hydrogen at a pressure of about 7000 kPa. The ash and undissolved material is removed from the resultant coal solution by filtration or centrifugation. The solvent is subsequently removed to yield a virtually ash-free coal extract that is a solid mass having a softening point of about 200°C. The coal extract may be converted into liquid fuels by hydrocracking. The yield of fuels from this process is stated to be about 56 per cent.

#### 4.1.4 C.S.F. process<sup>5,13)</sup>

The Consol Synthetic Fuel process was developed by the Consolidation Coal Co. and led to the erection of a pilot plant to process 40 t of coal per day in Cresap, W. Virg., in 1967. Called Project Gasoline, this work was terminated in 1970 because of mechanical problems in applying the process.

The process consists of the solvent extraction of coal under mild conditions. The ash is removed from the extract and the latter is then hydrogenated to

yield/...



yield a "crude oil" that can be refined further. The yield is stated to be about 380 l of petrol per ton of coal (about 30 per cent by mass).

#### 4.1.5 Clean Coke process<sup>14,15)</sup>

The Clean Coke process, developed by the United States Steel Corporation, is a multi-product process that combines carbonization and hydrogenation to convert non-metallurgical grade coals to low-sulphur metallurgical coke, chemical feedstocks, and liquid and gaseous fuels.

A contract by the O.C.R. provides for the design, construction and operation of process development units capable of processing 0,5 t of coal per day to permit evaluation tests to be carried out. These facilities were expected to be completed during 1974.

#### 4.1.6 Intermediate Coal Hydrogenation process<sup>4)</sup>

A process being investigated by the University of Utah is called the Intermediate Coal Hydrogenation process. A 23 kg of coal per hour hydrogenation unit was to be commissioned during 1974 under contract to the O.C.R.

### 4.2 Other processes

#### 4.2.1 Synthoil process<sup>12,14,16,17,18)</sup>

The United States Bureau of Mines developed the Synthoil process to convert high-sulphur coals to fuel oil low in ash and sulphur content. Their pilot plant treats 0,5 t of coal per day to yield about 240 l of clean fuel oil (yield about 480 l/t, that is about 41 per cent by mass). During April, 1974, it was stated that the U.S. Department of the Interior was ready to contract for an 8 t of coal per day pilot plant.

In this process, pulverized coal is slurried with some of its own product oil, pumped into a reactor together with hydrogen at high velocity to create turbulent conditions. The reactor is filled with immobilized catalyst pellets. The raw oil is centrifuged to remove ash and yields a low-ash, low-sulphur oil, easily refined to gasoline or diesel.

#### 4.2.2 COSTEAM<sup>18)</sup>

The United States Bureau of Mines also developed the COSTEAM process, which  
consists/...

consists of the reaction of carbon monoxide and steam with solid fuels at elevated temperature and pressure to yield a liquefied product. At present, work is carried out on a relatively small scale, namely in a 0,5 kg/h continuous unit.

#### 4.2.3 Catalytic Coal Liquefaction (CCL)<sup>19,20)</sup>

The Catalytic Coal Liquefaction process, investigated by Gulf Research & Development Co., is a variation of the S.R.C. process. Two bench-scale units were in operation and a 1 ton of coal per day pilot plant at Harmarville, Pa., was scheduled for completion in January, 1975.

In the CCL process, a coal-oil slurry is forced through a series of fixed-bed catalysts in the presence of hydrogen at a minimum pressure of about 14 MPa. The catalyst enhances the ability of the product to accept hydrogen and thus to become a high-grade fuel. The yield of liquid fuel in this process is about 480 l/t, that is about 41 per cent by mass.

#### 4.2.4 COGAS process<sup>19,21,22)</sup>

The COGAS process is really a further step to the COED process. The COGAS Development Co. was established by the following firms: F.M.C. Corporation, Consolidation National Gas Co., Panhandle Eastern Pipeline Co., Republic Steel Corp., Rocky Mountain Energy Co., and Tennessee Gas Pipeline Co., to develop a proprietary process for the gasification of char from coal pyrolysis. The COGAS complex thus produces synthetic crude oil as well as pipeline quality gas. A 100 t of coal per day pilot plant was to be started up during June, 1974 at Leatherhead in England.

#### 4.2.5 Exxon's process<sup>14,19)</sup>

After seven years of basic research, Exxon Research & Engineering Co. developed a process to make synthetic crude oil. Esso has operated a small pilot plant, and the next step will be the erection of a 0,5 t of coal per day pilot plant. A 300 t of coal per day liquefaction pilot plant is being designed.

The process consists of first making oil from the coal, then hydrogenating the oil to produce a hydrogen-rich solvent. The solvent is brought together with more coal and fed to a reactor, where the coal is liquefied by addition

of/...

of the hydrogen present in the solvent to the coal. A yield of 320 to 480 l of oil per ton of coal is expected (about 27 to 41 per cent by mass).

#### 4.2.6 Edison Electric Institute and The Southern Company System<sup>23)</sup>

The two companies, Edison Electric Institute and The Southern Company System, have announced a \$6 million joint research project to yield clean coal. The project calls for the construction and operation of a 6 t/d pilot plant at Wilsonville, Ala., for studying the key steps in solvent refining of coal (S.R.C. process).

The development programme is designed so that advancements made at the Wilsonville pilot plant will supplement the development of a 50 t/d S.R.C. pilot plant at Tacoma, Wa.

#### 4.2.7 Coalcon<sup>24)</sup>

A new company, called Coalcon, was established during 1974 to design, engineer and construct plants for the conversion of coal to liquid and gaseous fuels and to chemical feedstock. Coalcon was formed as a joint venture by Union Carbide and Chemical Construction Corp. (Chemico).

#### 4.2.8 U.O.P. process<sup>25)</sup>

The firm Universal Oil Products developed a process whereby pulverized coal in a solvent is treated with hydrogen at elevated temperature and pressure. An oil is formed, which can be processed to petrol and diesel.

### 4.3 Comments on efforts in the U.S.A. to develop alternative processes

The above survey of the state of development in the U.S.A. shows that a lively interest is shown in coal liquefaction processes. Some of these processes have been developed to the stage where large-scale plants are being planned. Other processes are not developed to such an extent - some of them are only in the initial stage. A striking aspect is the interest and active participation shown by various private firms in the development of oil-from-coal processes. Another notable feature is the huge amounts of money spent, both by the U.S. Government and by private enterprise, on the various projects.

### 4.4 Efforts by Sasol to develop alternative processes

In the Republic of South Africa, Sasol is investigating the production of solvent/...

solvent refined coal in a pilot plant. The liquefaction of coal and coal extracts is also being investigated by Sasol in collaboration with the Potchefstroom University for C.H.E.

## 5. WORK DONE AT THE FUEL RESEARCH INSTITUTE ON COAL CONVERSION

### 5.1 Research projects

The F.R.I. initiated research on various aspects of coal conversion during 1974, aimed particularly at obtaining liquid fuels and base feedstock for the chemical industry.

The following are the research projects receiving attention:-

- 5.1.1 Pyrolysis of coal, primarily to yield liquid fuels and hydrocarbon gases.
- 5.1.2 The activation of hydrogen to facilitate the hydrogenation of coal.
- 5.1.3 Gas extraction of coal and coal products to yield chemical compounds.
- 5.1.4 The manufacture of acetylene directly from coal in an electric-arc plasma.
- 5.1.5 The study of certain catalysts which perform an important function in rendering coal and coal derivatives into usable chemicals.

### 5.2 Coal pyrolysis

The activities of the F.R.I. on coal pyrolysis will be described rather more fully, as this is regarded to be one of the major alternatives to the conventional oil-from-coal process. Coal pyrolysis as investigated at the F.R.I. consists of the investigation of low-temperature fluidized-bed carbonization of coal.

5.2.1 Carbonization is a term applied to the heat processing of carbonaceous materials. In this treatment, the volatile compounds are removed usually with the main purpose of obtaining a solid residue with a higher fixed-carbon content than the original material. In this way the carbonization of, for instance, wood, yields charcoal, and coal yields coke or char, depending on the original material, the equipment, and the final temperature.

5.2.2/...

5.2.2 Low-Temperature carbonization is defined as the heat treatment of coal at a final temperature ranging between 450 and 700°C.

Four major products are obtained from low-temperature carbonization of coal, namely char (the solid residue), tar, aqueous liquid, and gas. Char and tar are the most important of these as far as useful products are concerned.

5.2.3 Fluidization. The term fluidization means to cause a bed of finely divided solid particles to behave like a fluid under certain conditions.

To obtain a concept of fluidization in its widest meaning, a bed of finely divided granular particles resting on a grid inside a tube is observed. When a fluid (a liquid or a gas) is passed through the tube from below the grid, very little happens at low flow rates - the fluid only passes through the voids between the particles. When the velocity of flow is increased gradually, a point is reached where the solid particles are only just being suspended in the fluid. At this point the force of friction between the particles is equal to the weight of the particles and occasionally a particle moves away vertically from the top surface and almost immediately falls back into the bed. A bed in this condition is known as a bed at minimum fluidization.

When the fluid velocity is increased still further in the case of a gas-solid system, instabilities with bubbling and channelling of gas arise. At still higher flow rates, severe fluctuations take place and the solid particles move vigorously. Such a bed is called a gas-fluidized bed.

A dense-phase fluidized bed displays a clearly observable top boundary. It behaves much the same as a boiling liquid and displays liquid characteristics; for example, a light object will float on top of the fluidized bed, the top level remains horizontal when the container is tipped, etc.

5.2.4 Development of fluidized-bed carbonization

There has been interest in the application of fluidized-bed carbonization of coal as far back as 1934, and various processes have since been introduced. The main purpose of fluidized-bed carbonization prior to 1960 was to obtain the solid product, that is the char, which was used as a boiler fuel or an additive to coke-oven charges.

Subsequently/...

Subsequently, the Consolidation Coal Company in the U.S.A. developed a new approach, namely the use of fluidized-bed carbonization as a primary step in a process for the manufacture of synthetic liquid fuel. This meant that the valuable recoverable volatile matter in the coal would be recovered prior to the gasification or combustion of the coal.

With the development of the COED process, in which coal is carbonized in a series of reactors, the practicability of fluidized-bed carbonization as a means of producing synthetic liquid fuels from coal was proved to be technically feasible.

### 5.3 Fluidized-bed carbonization of South African coals

#### 5.3.1 General

During 1972, the F.R.I. decided to start an investigation of the fluidized-bed carbonization of South African coals. The aim was twofold, namely

- (i) to find a way to supplement the tar and pitch supply in this country,
- (ii) to investigate the possibility of producing substitute liquid fuels.

As a result of the subsequent developments regarding the supply of crude oil, the emphasis at the Institute is now on synthetic liquid fuel production.

One reason why fluidized-bed carbonization is considered to be an attractive process, is that it can conceivably be combined with the conventional Sasol process, as mentioned in 5.2.4 (proposal by Consolidation Coal Co.). The line of thought is as follows: In the conventional Sasol process the coal as received is gasified to yield mainly the synthesis gas - that is, the coal is broken down completely to two basic compounds, namely hydrogen and carbon monoxide, which are then reacted to yield the required products. In this gasification process, the destruction of most of the volatile matter containing valuable compounds, such as tar acids, tar bases, neutral oils, pitch, etc., is unavoidable and these compounds are also largely converted to carbon monoxide and hydrogen.

The proposal is, therefore, that the primary step should be the carbonization of the coal in a fluidized bed in order to recover the useful liquids. The solid residue, that is the char, containing the fixed carbon, can then

be gasified to yield the synthesis gas. By means of the combination of pyrolysis, gasification and synthesis processes, more efficient utilization of the coal may be possible.

This process, although technically feasible, may not be economically viable, but the latter aspect should be investigated carefully.

### 5.3.2 Experimental work

A fluidized-bed unit capable of processing about 3 kg of coal per hour is being used to investigate the carbonization of coal with the aim of producing the maximum amount of liquid product (tar).

The apparatus consists of the following:

- (i) Electrically heated heat exchangers for heating the fluidization gas to operating temperature, that is to between 420 and 600°C.
- (ii) The reactor proper, consisting of lagged stainless steel pipe sections having an internal diameter of about 81 mm and a total height of about 1 110 mm. Pipes at different heights provide means for the discharge of char from the reactor at three different levels. A grid for distributing the fluidization gas evenly into the reactor is fitted to the base of the reactor.
- (iii) A products recovery train, consisting of a cyclone for fine coal collection, an electrostatic precipitator for tar collection, and two U-tubes kept in a low-temperature bath (about minus 40°C) to recover lighter liquids from the product gas stream.
- (iv) A bunker and variable wormfeeder to feed coal at the desired rate into the bottom of the reactor from its side.
- (v) An automatic gas sampler for sampling the product gases continuously during a run. The flow rate of the exhaust gases is measured by a rotameter, the outlet being at atmospheric pressure.

The apparatus is operated continuously, that is, coal is fed continuously and the products are removed continuously. The minimum time for a single run is one hour.

The orientative experiments have been successfully concluded, and the research programme proper is now aimed at finding the optimum conditions

for/...

for obtaining the maximum yield of tarry liquids from a particular coal. The factorial method is being applied in the planning of experiments in order to obtain the maximum amount of data from the experimental results.

The effect of the following variables on the yield of tarry liquids is being investigated:-

- (a) Type of coal.
- (b) Size of coal.
- (c) Type of fluidization gas.
- (d) Residence time of coal in the reactor.
- (e) Temperature of carbonization.

The carrying out of the actual programme is now well on its way. The apparatus functions satisfactorily. Preliminary results indicated that higher yields of tarry liquids are obtained with a high-volatile low-rank coal (volatile matter content about 40% d.a.f.) than with a typical Witbank middlings coal having a volatile matter content of about 28% d.a.f. The appearance of the tars also differed appreciably; the tar from the low-rank coal was a rather free-flowing black "oil", whilst the middlings coal produced a typical evil-smelling and relatively viscous tar.

#### 6. THE FUTURE OF OIL-FROM-COAL PROCESSES IN THE R.S.A.

To conclude this brief survey, one may speculate on the future of oil-from-coal processes in this country. For this purpose, it will be best to consider the opinions of authorities who have practical experience in this field, namely at Sasol. It has already been decided to build a second Sasol. Some time ago, Dr P.E. Rousseau<sup>26)</sup>, Chairman of Sasol, stated that any decision by South Africa to set up a second Sasol would be based on strategic considerations. He also forecast that a second Sasol would probably be ten times the size of the present one. Mr D.P. de Villiers, Managing Director of Sasol, inter alia made the following remarks<sup>27)</sup>: "The results of our own work on improving the existing oil-from-coal process and developing other processes are encouraging. I believe that processes which are not now being used will eventually be used because of their low capital cost and their higher yield of liquid fuel per ton of coal."

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From these remarks it could be concluded that the second oil-from-coal plant would use the same process as Sasol I, but that coal conversion plants after that might be based on other processes.

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