

The SRC-II Process [and Discussion]

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The SRC-II process

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Extensive laboratory and pilot plant experimental work on the Solvent Refined Coal process by Gulf Oil Corporation over the past 18 years, sponsored by the Fossil Fuel Division of the United States Department of Energy and its predecessor agencies, has led to the development of an improved version of the process known as SRC-II. This work has shown considerable promise in recent years and plans are now being made to demonstrate the SRC-II process with commercial size equipment in a 6000 ton/day (5440 t/day) plant to be located near Morgantown, West Virginia. On the basis of recent economic studies, the products (both liquid and gas) from a future large-scale commercial plant are expected to have an overall selling price of \$4.25–4.75/GJ (first quarter 1980 basis).

The major product of the primary process is distillate fuel oil of less than 0.3% sulphur for use largely as a non-polluting fuel for generating electrical power and steam, especially in the east where utilities and industry are currently using petroleum products. In such applications, SRC-II fuel oil is expected to be competitive with petroleum-derived fuels within the next decade. During this period, SRC-II fuel oil should be economically attractive compared with coal combustion with flue gas desulphurization in electric utility and industrial boilers, particularly in the major metropolitan areas.

Naphtha produced by the SRC-II process can be upgraded to a high-octane unleaded gasoline to supplement petroleum-derived supplies. Significant quantities of pipeline gas are also produced at a cost that should be competitive with s.n.g. from direct coal gasification. Light hydrocarbons (ethane, propane) from the process may be effectively converted to ethylene. In addition, certain fractions of the fuel oil might also be used in medium-speed diesel engines and automotive gas turbines.

For many of these applications, the fuel oil and other products from the SRC-II process would displace high-quality petroleum fractions, which could then be used for production of diesel fuels, jet fuels, home heating oil and gasoline by conventional refinery processes.

1. Introduction

The Solvent Refined Coal process dissolves coal in a process-derived solvent at elevated temperature and pressure in the presence of hydrogen, separates the undissolved mineral residue (ash plus insoluble carbon), then recovers the original solvent by vacuum distillation. The residue is an ashless, low-sulphur pitch-like material, solid at room temperature, known as Solvent Refined Coal. Research and development work on the Solvent Refined Coal process was begun in 1962 at Gulf's Merriam Laboratory (then part of Spencer Chemical Company) under the sponsorship of a government agency now part of the U.S. Department of Energy.

The encouraging results obtained in batch laboratory work and in continuous developmental units led to the signing of a contract with the government for design, construction and operation of a 50 ton/day† pilot plant at Fort Lewis, Washington. The pilot plant was started up in 1974 and has been operating for the last 6 years.

Laboratory work with continuous bench-scale units has proceeded simultaneously with the

† 1 U.S. ton = 0.9072 t.

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pilot plant programme, and led to the development of an improved version of the process, known as SRC-II. The development of the process has been discussed in a number of recent technical papers (Schmid et al. 1976, 1977; Jackson et al. 1978, 1979).

This work has shown considerable promise, and plans are now being made to design, construct and operate a 6000 t/day demonstration plant near Morgantown, West Virginia. The laboratory, pilot plant and demonstration projects are being supported by the Fossil Fuel Division of the U.S. Department of Energy.

The SRC-II process not only dissolves the coal in a process-derived solvent, but also hydrocracks the dissolved coal to liquids and gases. The hydrocracking is achieved by recycling the product slurry and by increasing the residence time in the reactor. The product slurry is recycled for mixing with the feed coal, rather than using a distillate solvent for this purpose. The recycle slurry consists of three primary components: (1) a distillate liquid corresponding to the coal derived solvent used in the original process, boiling in the range 200–480 °C, (2) normally solid dissolved coal with a boiling range above 480 °C, and (3) mineral residue, which consists of the inorganic portion of the coal plus a minor quantity of insoluble carbonaceous material which is not dissolved under the conditions prevailing in the hydrogenation reactor.

The mineral residue is known to be an active catalyst for the hydrogenation and hydrocracking reactions of the process, and the increase in concentration of the mineral residue in the reactor achieved by recycling this material significantly increases the extent of hydrocracking attainable. Furthermore, recycling the dissolved coal not yet reacted increases the effective residence time of this component in the reactor, for the same residence time per pass. In addition, the residence time per pass is increased to achieve even more extensive hydrocracking.

Product slurry from the SRC-II process is vacuum distilled to recover the material boiling below 900 °F. The residue from this distillation is then sent to an oxygen-blown gasifier. The gasifier produces synthesis gas, which is then converted to hydrogen and process fuel gas. Because of the extensive hydrocracking reactions, the quantity of dissolved coal (over 480 °C) remaining unconverted in the hydrogenation step is reduced to a practical level for a viable process, i.e. a level that produces little or no synthesis gas in excess of that required for production of hydrogen and fuel for the process. This process sequence eliminates the liquid–solids separation step required in the original Solvent Refined Coal technology.

Economic evaluations indicate that elimination of the liquid-solids separation step, together with a higher efficiency in integration of the high pressure gasification step into the process, offsets the cost of the additional hydrogenation involved in SRC-II. The SRC-II process is projected to produce a distillate liquid containing less than 0.3% sulphur for about the same cost as involved in the production of solid Solvent Refined Coal of 0.7–1.0% sulphur by the original SRC process. Experience at the Fort Lewis Pilot Plant indicates that the operability of the SRC-II process is attractive, with less maintenance and less downtime required than for the original version of the SRC process. The Fort Lewis Pilot Plant has been operated extensively in both modes since the startup in 1974.

2. PROCESS DESCRIPTION

A schematic flow diagram of the SRC-II process as it is currently visualized for large-scale demonstration or a commercial plant is shown in figure 1. Raw coal is pulverized and dried,

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then mixed with hot recycle slurry from the process. The coal–recycle slurry mixture is pumped, together with hydrogen, through a preheater to a reactor maintained at a pressure of about 1.4 MPa. In the preheater, the coal is almost completely dissolved in the solvent portion of the recycle slurry. The hydrocracking reactions occur primarily in the reactor, and the heat generated by these reactions raises the temperature of the reactants to the range 438–466 °C at the reactor outlet. Hydrogen quench is injected at various points in the reactor to control the temperature and alleviate the impact of the exothermic reactions.

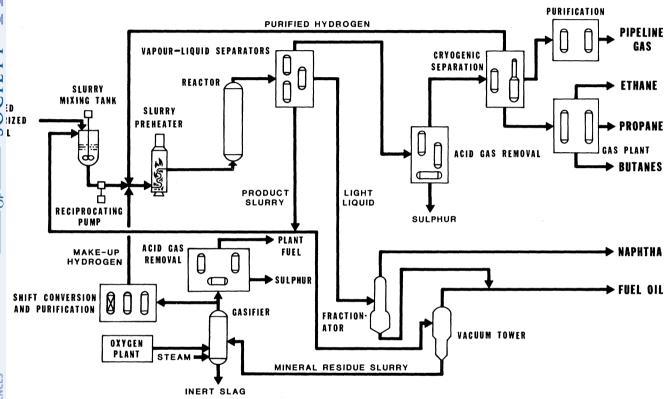


FIGURE 1. SRC-II process.

The reactor effluent then flows through a series of vapour-liquid separators, where it is separated into process gas, light hydrocarbon liquid and product slurry. The gas, consisting primarily of hydrogen and gaseous hydrocarbons, together with minor amounts of H₂S and CO₂, first goes through an acid gas removal step for removal of the H₂S and CO₂. The treated gas then goes to a cryogenic separation step for removal of the hydrocarbons. The purified hydrogen is recycled to the process, while the recovered hydrocarbons become by-products of the process. The C₁ fraction is sent to a methanator to convert any remaining CO to methane, then is sold as pipeline gas. The other light hydrocarbon gases are fractionated to produce ethane, propane and a mixed butane stream. The light hydrocarbon liquid goes to a fractionator where it is separated into naphtha (C₅-177 °C nominal boiling range) and a middle distillate.

The product slurry is split, with one portion being recycled to the process for slurrying with the feed coal. The other portion of the product slurry goes to a vacuum tower where a heavy 132

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distillate is removed overhead. The heavy distillate, together with middle distillate from the fractionation step, makes up the total fuel oil product of the process.

The residue from the vacuum tower is sent to a high-pressure slagging gasifier for production of synthesis gas. A portion of the synthesis gas goes through shift conversion and acid gas removal steps to produce pure hydrogen for the process. The synthesis gas in excess of that required for hydrogen production is passed through a separate acid gas removal step for removal of CO_2 and H_2S , then through a power recovery turbine, and is finally burned as plant fuel.

3. Economics: required product selling price

Under contract with the U.S. Department of Energy, Gulf has completed a market assessment for SRC-II products and a preliminary design and economic evaluation for a conceptual commercial SRC-II plant. The plant would process West Virginia area coal at a rate of 34 000 tons per stream day. The net daily output of products from such a plant is shown in table 1.

Table 1. Products from typical commercial plant processing 33 500 tons per stream day, using West Virginia coal

methane $1.4 \times 10^6 \text{ m}^3/\text{day}$ at s.t.p. ethane and propane 3000 ton/day butane 300 ton/day naphtha $17\,000 \text{ barrels/day}$ (2700 m³/day) fuel oil $56\,000 \text{ barrels/day}$ (8900 m³/day)

The investment and operating costs for the conceptual commercial plant were based on a design developed by assuming that the key process steps will be successfully demonstrated in the mid-1980s in the 6000 ton/day demonstration plant, and that the first commercial plants will be built and operating in the early 1990s. Projected investment costs are given in table 2

Table 2. Estimated conceptual commercial plant investment costs for a plant processing 33 500 tons per stream day, using West Virginia coal

(Based on first quarter of 1980.)

	costs/M\$
coal and ash handling	86
hydrogenation	707
hydrogen production	480
product recovery	255
utilities and general facilities	266
	1794
indirect investment	190
total	1984

and estimated operating costs in table 3. Additional assumptions used in the economic evaluation are given in table 4. On the basis of these assumptions, the required selling price for all SRC-II products is calculated to be about \$4.25-4.75/GJ (first quarter 1980 basis).

It should be noted that these prices are nearly competitive with current prices for petroleum-based fuel oils. By the time commercial SRC-II plants could be in operation, these prices may well be more attractive than the price of comparable petroleum fuels. It must be appreciated,

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TABLE 3. ESTIMATED ANNUAL OPERATING COSTS

(Based on first quarter of 1980.)

	costs/M
coal	370
operations	22
maintenance	54
overhead	34
total	480

Table 4. Major assumptions used in economic evaluation

coal cost

\$1.09/GI

depreciation

13 years double declining balance, with a switch to straight line when this becomes

income tax rate

 $50\,\%$

investment tax credit

10 %

rate of return

15% d.c.f. on 75% equity; 9% interest on 25% debt

cost escalation

6% per year

however, that these estimates are preliminary and will not be firm until the demonstration project provides definitive performance and cost data.

4. BOILER FUEL TESTS

The major product from the SRC-II process is distillate fuel oil with an approximate boiling range between 180 and 480 °C. Other typical properties are given in table 5. Primary use of this material in the near future is expected to be as fuel oil for large oil-burning facilities, and perhaps later in combined cycle combustion turbines, especially those located near major

Table 5. Properties of SRC-II fuel oil

metropolitan areas. The major question involving burning characteristics of coal liquids relates to the higher nitrogen content compared with petroleum fuel oils and the potential effects on NO_x emissions. Since NO_x emissions are sensitive to burning conditions, actual burning tests are being conducted under various conditions to assess the effects.

Several burning test programmes have been carried out to confirm that the SRC-II fuel oil could be successfully used in conventional power plants and that emission levels of potential atmospheric contaminants could be controlled.

The first burning test for the liquid fuel oil was conducted in a 3.2 GJ/h test boiler. With a viscosity comparable to no. 2 fuel oil, no preheating was required and handling characteristics were attractive. The SRC-II fuel oil was used interchangeably with no. 2 fuel oil without forming sediments. Cold boiler light-offs were made without incident. Although the fuel oil has the relatively high organic nitrogen content characteristic of coal-derived liquids, combustion control technologies were effective in decreasing NO_x emissions and smoke to environmentally acceptable levels.

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In the fall of 1978 a full-scale test programme was pursued in a commercial power plant of the Consolidated Edison Company in New York City. The test was conducted in three phases in Con Edison's 74th Street power station utilizing a 225 ton/h steam electric Combustion Engineering tangentially fired boiler (KVB Inc. 1979).

Work in the first phase involved preliminary checking of equipment and instruments for measuring emissions, as well as establishment of NO_x reduction trends with staged combustion techniques, while burning the current power plant fuel, a low-sulphur no. 6 fuel oil. The purpose of this phase was to reduce the time necessary to carry out the subsequent SRC-II tests and to achieve minimum NO_x levels with the limited supply (4500 barrels; 715 m³) of SRC-II fuel oil. Properties of the test fuels used in this programme are given in table 6.

TABLE 6. PROPERTIES OF TEST FUELS (Based on average analysis of samples taken during test programme.)

	no. 6 fuel oil	SRC-II fuel oil
viscosity		
at 37.8 °C	enturina	40 S.U.s
at 50 °C	300-700 S.U.s	-
nitrogen (by mass)	0.23%	1.02%
sulphur (by mass)	0.24%	0.22%

The second phase involved a 6 day test of the SRC-II fuel oil to determine its combustion performance and emission levels under various operating conditions. Tests were made at full load, three-quarter load and one-half load while using normal combustion (baseline) and staged combustion techniques. The staged combustion tests were made to evaluate the possibility for substantially decreasing NO_x emission levels.

The third phase of testing involved measuring the combustion performance and emission levels while using the low-sulphur no. 6 fuel oil, with the boiler operating as close as possible to the operating conditions used during the second phase.

TABLE 7. LARGE-SCALE SRC-II FUEL OIL BURN TEST AT CON EDISON

	E.P.A. requirements	test burn results
NO_x (parts/106)	400	175–300
sulphur	85% removal	95% removal
particulates (g/GJ)	13	<13 (no precipitator)
boiler efficiency		comparable with
		petroleum fuel oil

The Consolidated Edison test results, as shown in table 7, indicated the suitability of SRC-II coal liquids as a high-quality boiler fuel. No operational problems were encountered and no deposits were observed. Combustion efficiency was comparable to that for the low-sulphur no. 6 fuel oil, as were the levels of carbon monoxide and hydrocarbon emissions. Modifications to burner equipment required to handle the SRC-II fuel oil were considered to be no more extensive than those required for similar variations in petroleum fuels.

All tests were run with no smoking and less than 13 g/GJ total particulates. While the higher nitrogen content SRC-II fuel oil produced higher NO_x emission levels than the low-sulphur no. 6 fuel oil, the difference was substantially less than would be expected from the nitrogen contents of the two fuels. SRC-II fuel oil produced approximately 70% more NO_x than no. 6 fuel oil, though its nitrogen content was more than four times as high.

Based on the overall test results, it is expected that a boiler currently capable of meeting the E.P.A. requirements of 130 g NO_x/GJ for petroleum fuels will be capable of satisfying the proposed standard for coal-derived liquids (215 g/GJ, equivalent to 400 parts/106) using the SRC-II fuel oil. Furthermore, the tests showed that NO_x formation could be reduced sub-

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stantially for both fuels (on the order of 35%) by staged combustion. Additional tests on other

boiler designs are planned during 1980.

5. MARKET APPLICATIONS

(a) Electric utility boilers

Recent studies have compared the potential use of SRC-II fuel oil with the direct burning of coal accompanied by 'scrubbing' or flue gas desulphurization, for large East Coast utilities. These studies indicate that SRC-II fuel oil is likely to be a competitive fuel for a portion of the electric power requirements, particularly in the populous regions of the United States.

Conducted with the involvement of Consolidated Edison of New York, Baltimore Gas and Electric, Boston Edison and Florida Power Corporation, the studies considered a broad range of applications, including (1) replacing oil in existing oil-fired boilers as well as in new oil-fired boilers, (2) using new combined cycle combustion turbines at today's level of technology, and (3) using advanced combined cycle combustion turbines at expected levels of technology in the future.

Results of these studies show that in certain cases coal-derived fuel oil from SRC-II could be an attractive alternate when compared with other means of using coal. In most of these situations, the lower cost of coal - compared with SRC-II fuel oil - is not sufficient to offset the higher capital costs of coal-fired plants with flue gas desulphurization.

(b) Electric utility combustion turbines

As part of a contract with D.O.E. to evaluate markets and economics for SRC-II products, the potential use of SRC-II fuel oil in gas turbines with heat recovery boilers (combined cycle units) has been investigated. This study, based on data from two different actual utility company systems, indicates that:

- (1) advanced combined cycle units using SRC-II fuel oil are more attractive economically than direct firing of coal in conventional boilers with flue gas desulphurization;
- (2) use of SRC-II fuel oil in advanced combined cycle units would remain an attractive generating scheme when the system also includes coal gasification integrated with advanced combined cycle units;
- (3) potential use of SRC-II fuel oil to generate electric power through advanced combustion turbine units with heat recovery boilers could be substantial.

(c) Industrial boilers

For boilers used by industries other than electric utilities, SRC-II fuel oil is expected to be competitive over a wide range of boiler sizes, in both new and existing plants.

A recent study (Doherty 1978) based on a General Electric Co. manufacturing plant suggests that, compared with a plant using SRC-II fuel oil, the higher investment cost for a coal-burning plant equipped for flue gas desulphurization cannot be justified for new manufacturing plants ranging in size from 50 to 250 ton/h of steam-generating capacity, with typical capacity use.

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(Plants are normally run at 50% capacity or less.) With an existing oil-fired plant, the attractiveness of burning SRC-II fuel oil is significant for even somewhat larger units.

An economic evaluation based on a specialty chemicals plant of Rohm & Haas Co. in Philadelphia has shown similar results. The capacity factor (normal percentage of use) for the steam boiler in this chemical processing plant study was high (55%) and the steam-generating capacity was 135 ton/h. For either new or existing boilers in the 50 ton/h size range, the investment in facilities for burning coal with flue gas desulphurization, or for an atmospheric fluidized-bed combustion system, was not justified if SRC-II fuel oil were available at or near projected prices.

(d) Petroleum substitution and displacement

(i) Light hydrocarbons

A typical 33500 ton/stream day commercial SRC-II plant would produce about 1.4×10^6 m³ (at s.t.p.) of methane (pipeline gas) per day. This methane should be competitive with pipeline gas produced by direct gasification of coal and could be a substitute for home heating oil. The quantity of methane produced by an SRC-II commercial plant could vary significantly, depending on the downstream processing steps used and the selection of plant fuel.

Although ethane and propane can be sold directly or converted to methane, their use as a chemical feedstock is also of interest. The quantity of ethane and propane projected to be available from domestic natural gas is not sufficient to supply future ethylene plants. Without new supplies of ethane and propane, the difference would have to be made up by petroleum-derived naphtha and gas oil. Use of coal-derived ethane and propane for ethylene production should indirectly make more naphtha and gas oil available for the production of gasoline at petroleum refineries.

(ii) Naphtha to gasoline

The naphtha from the SRC-II process (C_5 –177 °C) can be hydrotreated to remove nitrogen, then reformed to produce high-octane gasoline. The properties of raw and hydrotreated SRC-II naphtha are shown in table 8. The high content of cyclic hydrocarbons (76.2% aromatics plus

TABLE 8. PROPERTIES OF RAW AND PRETREATED SRC-II NAPHTHAS

	raw	hydrogenated
sulphur (parts/106)	2400	0.08
nitrogen (parts/106)	8800	0.2
hydrocarbon analysis (by volume)		
paraffins	21%	24%
cycloparaffins	45%	62%
aromatics	34%	14 %

cycloparaffins) in the hydrotreated naphtha make it an attractive charge stock for catalytic reforming. This is reflected in the high yield (89.6% by volume) when reformed to a research clear octane number of 98. Thus, the SRC-II naphtha will be an attractive source of unleaded gasoline.

The price of raw SRC-II naphtha used in the overall SRC-II economic evaluation was based strictly on heat value. A study has recently been made to establish a value based on its use as a supplemental feedstock to a petroleum refinery. The basic assumptions of the study were as follows.

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- (1) Naphtha from 33500 ton/stream day SRC-II plant is severely pretreated to remove nitrogen, then reformed at the SRC-II plant to produce high-octane gasoline. The reformate is then transported to an existing 150000 barrels per day (24000 m³/day) petroleum refinery.
- (2) The value of the raw SRC-II naphtha is calculated to give the same return on total investment (refinery plus naphtha upgrading facilities located at the SRC-II plant) as set for the base refinery.

By using the above assumptions, two different cases were calculated:

- (1) a west Texas crude refinery with conventional facilities for producing gasoline and other typical refinery products;
- (2) a Kuwait crude refinery using hydrodesulphurization of atmospheric residue followed by fluid catalytic cracking of the treated residue, with otherwise conventional facilities.

In both cases, the SRC-II reformate is used to increase the total quantity of gasoline produced at the same crude charge rate. The results are shown in table 9 as relative values. The

Table 9. Value of raw SRC-II naphtha to refineries

	west Texas crude refinery	
relative cost of crude oil	1.00	1.00
relative value of raw SRC-II naphtha	0.985	1.01

absolute value of the crude oil used for this study has no effect on the conclusions reached regarding the relative value of the crude oil and the SRC-II raw naphtha. The significant conclusion is that the value of the raw naphtha is about equal to the value of crude oil as a refinery feedstock.

(iii) Other fuel oil applications

The SRC-II distillate product planned for use as a boiler fuel is a liquid with a wide boiling range (180–480 °C). This product has generally been divided into two major fractions: a middle distillate with a boiling range of about 180–290 °C and a heavy distillate with a boiling range of about 290–480 °C. Typical properties of each of these two fractions are given in table 10.

Table 10. Properties of SRC-II fuel oil fractions

	middle distillate	heavy distillate
boiling range	180–290 °C	290–480 °C
pour point	< −43 °C	7 °C
sulphur (by mass)	0.2%	0.4%
nitrogen (by mass)	0.9%	1.3%

Manufacturers of medium-speed diesel engines (used in railroad locomotives and domestic ships) and automotive gas turbine engines (being developed for trucks and buses) have been surveyed to determine whether SRC-II middle distillates could be used to fuel these engines. They have indicated that the middle distillate has potential as an acceptable alternative fuel for both of these engine types.

During the early years of a commercial synthetic fuels industry, the coal-derived middle and heavy distillates will probably be used primarily as produced, with little or no further upgrading. SRC-II coal liquids can be directly used as fuels for electric utility and industrial boilers, and potentially in stationary combustion turbines, medium-speed diesel engines and automotive gas turbines. Their widespread use would 'free up' supplies of liquid petroleum

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products, which would then be available for upgrading to premium diesel fuels, jet fuels and gasoline. In general, it is projected to be more efficient to produce these materials from petroleum stocks displaced by SRC-II products than to produce them directly from SRC-II fuel oil,

at least for the near future.

Some examples of methods by which this could be done are:

(1) no. 2 fuel oil replaced by SRC-II fuel oil could be used directly for home heating oil or processed in a petroleum refinery into jet fuel, diesel fuel and gasoline;

- (2) diesel fuel replaced in medium-speed diesels and automotive gas turbines by SRC-II middle distillate could be used to augment short supplies of diesel fuel for trucks and other high speed diesel applications;
- (3) residual fuel oil or selected fractions of this oil, replaced in the industrial and electric utility boiler market by SRC-II fuel oil, could be converted to gasoline, jet fuels or home heating oil by a variety of petroleum conversion processes.

6. Conclusions

Low-sulphur distillate fuel oil produced from coal by the SRC-II process could be economically competitive with petroleum fuels during the 1980s. In this time frame, the SRC-II fuel oil is also expected to be attractive compared with combustion of coal with flue gas desulphurization in the U.S.A.'s East Coast oil-burning power plants, as well as in small and medium-sized industrial boilers.

The SRC-II process produces substantial quantities of methane, light hydrocarbons and naphtha. These products have value as feedstocks for preparation of pipeline gas, ethylene and high-octane unleaded gasoline and will contribute to the overall attractiveness of the SRC-II process. Products of the SRC-II process can replace petroleum fractions in many applications, thereby making these fractions available for processing by conventional refinery methods to gasoline, diesel fuel, jet fuels and home heating oils.

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Discussion

S. P. S. Andrew, F.R.S. (I.C.I. Agricultural Division, Billingham, U.K.). During the development of coal liquefaction processes some 50 years ago, considerable difficulty was experienced in

burning the unreacted solid sludge. Have the authors any experience of this process and do they intend to use a fluidized-bed or an entrained-solids type gasifier?

- B. K. Schmid. We have not done any experimental work on burning the unconverted solids. Our conceptual studies indicate that burning of this material is a less attractive option than gasification. Gasification with a high-temperature, high-pressure entrained-bed gasifier appears to be optimum for our process, and such a gasifier is included in our current designs.
- H. Schulz (Engler-Bunte-Institut, Universität Karlsruhe, Germany). The Gulf SRC-II process is claimed to be non-catalytic; however a beneficial recycle of the residual matter is observed. It thus follows that, e.g. iron and sulphur in the coals to be processed act as catalysts. Could the authors please specify what amounts of these materials should be present in the feed coal for SRC-II and what results have been obtained with coals of particularly low sulphur and iron content.
- B. K. Schmid. We have found that iron sulphides have a catalytic effect in the hydrocracking of the dissolved coal to liquids and that the extent of hydrocracking generally increases with increasing contents of iron and pyritic sulphur in the coal. We have had satisfactory results with coals of about 1% iron and 1% pyritic sulphur. Although other factors are believed to influence the extent of hydrocracking, coals of lower iron and sulphur content can be processed with some penalty in economics and/or thermal efficiency.