

# TECHNOLOGY STATUS REPORT

## COAL LIQUEFACTION

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Department of Trade and Industry

CLEANER COAL TECHNOLOGY PROGRAMME

TECHNOLOGY  
STATUS REPORT

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# COAL LIQUEFACTION

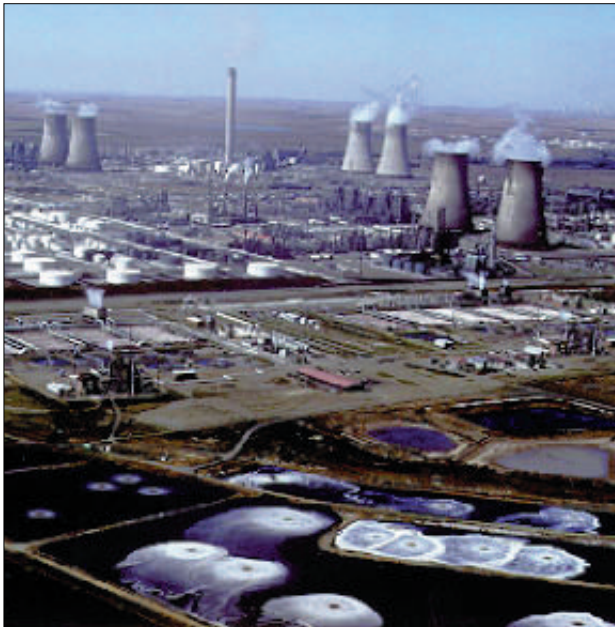


Figure 1. Sasol synthetic fuels plant at Secunda, South Africa

## SUMMARY

Coal is a solid with a high carbon content but with a hydrogen content of ~5%. Compared with liquid fuels (derived from crude oil) it is inconvenient to handle and unsuited to some applications, eg transportation.

Coal may be used to produce liquid fuels suitable for transportation applications by removal of carbon or addition of hydrogen, either directly or indirectly. The first approach is usually known as carbonisation or pyrolysis and the second as liquefaction. As the cost of converting coal into useful liquid fuels is higher than the cost of refining crude oil, it is the relative price of the raw feedstocks that provides the main incentive to pursue the technology.

Given the gradual depletion of oil reserves, at some stage in the future alternative sources of liquid fuels will be required. Coal liquefaction is one source, backed by large recoverable coal reserves globally.

Significant coal liquefaction research and development (R&D) was started up in the early 1970s, particularly in the USA, the UK and Japan, in response to various oil price shocks. Since the 1980s, though, developments have been largely put on hold with the notable exception of in South Africa. Here, with large reserves of coal but no oil or gas, trade embargoes over three decades to the mid-1980s drove large-scale application; up to 60% of transportation fuel requirements have been met from coal.

Many different 'direct' processes have been developed, but most are closely related in terms of the underlying reaction chemistry. Common features are the dissolution of a high proportion of coal in a solvent at elevated temperature and pressure, followed by hydrocracking of the dissolved coal with hydrogen gas ( $H_2$ ) and catalyst.

Direct liquefaction is the most efficient route currently available. Liquid yields in excess of 70% by weight of the dry, mineral matter-free coal feed have been demonstrated in favourable circumstances. Overall thermal efficiencies (% calorific value of the input fuel converted to finished products) for modern processes are generally in the range 60-70% if allowance is made for generating losses and other non-coal energy imports.

These processes generally have been developed to process development unit (PDU) or pilot plant scale and the main technical issues have been resolved. However, no demonstration or commercial-scale plant has yet been built.

The only operating process for the 'indirect' liquefaction of coal is South Africa's Sasol process, with three operating plants. The only 'core' unit specific to indirect liquefaction is the synthesis reaction step and the majority of recent work has therefore been concentrated on the development of improved catalysts, which need not be specific to a particular process.

With lower oil prices since 1985, interest in coal liquefaction for the production of transportation fuels has declined. At present, only Japan is active in large-scale process development, with a 150 tonnes/day plant in operation, whilst China (increasingly a net oil importer and containing areas remote from sources of oil) presents the strongest adoption prospect. China is currently co-operating with each of the USA, Japan and Germany on feasibility studies.

The commercial viability of coal liquefaction rests with the overall economics of the process. This depends on the availability of significant quantities of poor quality, low cost coal, and the unavailability or otherwise relatively high cost of oil (and gas). It remains to be seen whether a future increase in oil prices will result in renewed interest and possibly in full-scale commercialisation.

## BENEFITS OF THE TECHNOLOGY

- Coal liquefaction offers the following benefits:
- Largely proven technology for the manufacture of useful liquid products.
- Ability to manufacture transportation fuels from abundant coal.
- Insurance against depleting oil stocks and oil supply problems.

## DEPARTMENT OF TRADE AND INDUSTRY SUPPORT

Since 1990, the Department of Trade and Industry (DTI) has supported five projects associated with coal liquefaction, contributing £5.4M to a total projects cost of £40.6M.

## INTRODUCTION

Coal is a solid with a high carbon content but a hydrogen content of typically 5% and never more than 6%. In comparison with liquid fuels it is inconvenient to handle and unsuited to some applications. Most notably, it cannot be used directly to fuel the internal combustion (IC) engines and turbines that dominate transportation infrastructure worldwide. Transportation fuels for these are derived overwhelmingly from crude oil, which has about twice the hydrogen content of coal. They account for over 50% of total oil consumption.

The hydrogen content of these fuels varies from ~12.5% in some gasolines to 14.5% in aviation turbine fuels. For coal to replace them, it must be converted to liquids with similar hydrogen content. This can be achieved either by removing carbon or by adding hydrogen, either directly or indirectly. The first approach is known as carbonisation or pyrolysis and the second as liquefaction.

Liquid fuels have long been produced from coal. As the cost of converting coal into useful liquid fuels is higher than the cost of refining crude oil, it is the relative price of the raw feedstocks that has provided the main incentive to pursue the technology. The major exceptions to this principle resulted directly from the isolation of a country from reliable, secure sources of crude oil. Germany produced substantial amounts of coal-derived fuels during the Second World War, as did embargoed South Africa between the mid-1950s and 1980s.

With demand for oil products continuing to grow, and oil stocks gradually becoming depleted, there will come a time (between 2010 and 2020 by some estimates) when demand begins to outstrip supply. Price rises are then highly likely. For the time being, oil prices remain relatively very low.

The existing infrastructure of the developed world is extremely dependent on oil. Therefore, in the event of a shortfall occurring, alternative sources of liquid fuels will be required. Coal liquefaction is one source, and is backed by large recoverable coal reserves globally. These reserves are indeed significantly greater than for other fossil fuels.

## Development History

The production of coal-derived liquids as a by-product of coke-making commenced in Germany and the UK in the 1840s. These liquids had a variety of uses including solvents, wood preservatives, and fuels, and from the 1850s, were used as the basis of the coal-tar dyes still in current use. They formed the feedstocks for the developing petrochemical industry, where coal-derived aromatics were used in significant quantities until comparatively recently.

Most development derives from the early 1900s, when two distinct approaches were pursued. The earliest process route involved high-temperature and high-pressure dissolution of coal in a solvent to produce high boiling point (bp) liquids. No hydrogen or catalyst was used at this time. This approach, known as direct liquefaction, was patented by Bergius in 1913 and commercialised in the early 1920s; it also became known as either the Pott-Broche or I G Farben process.

In 1925 Fischer and Tropsch patented an alternative process known as indirect liquefaction. This involved the gasification of coal to produce a synthesis gas containing H<sub>2</sub> and carbon monoxide (CO). These were then reacted over a cobalt catalyst to produce liquids. The process was commercialised in the 1930s, initially for the production of chemical feedstocks rather than liquid fuels.

In 1935, at Billingham in the UK, a commercial-scale direct liquefaction plant came into operation processing coal and creosote oil to produce a total of 150,000 tonnes/year of gasoline. At the start of the Second World War, both Germany and the UK had operational coal liquefaction plant. By the end of the war, in Germany nine indirect and 18 direct liquefaction plant were producing almost 4 million tonnes/year of gasoline, 90% of German consumption.

Following the war the liquefaction plant in Germany and elsewhere were generally closed down. Although there was some further process development in the USA in the early and mid-1950s, the price of oil was falling relative to coal and the economics of liquefaction became increasingly unattractive. Large discoveries of new oil reserves were being made in the Middle East at this time that reduced the perceived need for strategic R&D programmes and reduced interest in coal liquefaction in all countries except South Africa.

South Africa was a special case as, due to its apartheid policies, it became increasingly isolated politically through the three decades from the mid-1950s to the mid-1980s. Unable to trade freely in oil and oil products and having very large coal reserves, but no indigenous oil, South Africa further developed and used coal liquefaction. Indirect liquefaction was selected as it was thought better suited to South African coals. The first plant, Sasol 1, was built in the 1950s. Two much larger plant were built in 1980 and 1982, using the same basic process chemistry but employing improved catalyst formulations and reactor designs. In the mid-1980s these plant together are estimated to have been producing up to 10 million tonnes/year of transportation fuels, or 60% of South Africa's requirements. All three plant are still in operation. Outside South Africa, interest in the production of transportation fuels from coal remained low until the early 1970s.

From the mid-1960s, at a time of mounting environmental concern over emissions from power generation, there was continued interest in developments of the original uncatalysed Pott-Broche process. Most work was carried out in the USA, where the more developed examples were the SRC-I (Solvent Refined Coal) and SRC-II processes, although other processes were also developed to a smaller scale in Japan and the UK. The SRC-I process was very similar in concept to the Pott-Broche process and aimed to upgrade coal to produce a clean boiler fuel with a much lower ash and sulphur content than the original coal. The SRC-II process,

however, was to produce distillate products: the distinguishing characteristic of the process was the recycle of vacuum bottoms.

Two other direct coal liquefaction processes were under development in the USA at the same time: the Exxon Donor Solvent (EDS) process and the H-Coal process. The distinguishing feature of the EDS process was a separate solvent hydrogenation step to carefully control the hydrogen donor characteristics of the solvent, and the most important feature of the H-Coal process was its use of an ebullated bed reactor.

In the early 1970s, political changes in the major oil-producing regions resulted in dramatic oil price rises and interest switched once more to the production of transportation fuels from coal. Oil prices continued to increase throughout the 1970s and remained high until late 1985. Significant R&D programmes were started, mostly based on variants of direct liquefaction. The main participants were the USA, Japan and the UK, which had been developing coal-upgrading processes, and Germany. Coal liquefaction, via indirect processes, attracted less interest, although there were parallel developments in synthesis reaction catalysis and technology for other purposes. Almost all of the current liquefaction processes originate from this period.

## CARBONISATION AND PYROLYSIS

High-temperature carbonisation is the oldest route for the production of liquids from coal, wherein hydrocarbon liquid is predominantly a by-product of coke-making. The low yields (<-5%) of liquid product and relatively high upgrading costs mean that traditional high-temperature carbonisation is not an option for the production of liquid fuels on a commercial basis.

Mild pyrolysis is also a carbonisation technology but with less severe operating conditions. Mild pyrolysis consists of heating the coal to a temperature in the range -450-650°C (compared with -950°C in high-temperature carbonisation), driving off volatile matter from the original coal and generating other volatile organic compounds formed by thermal decomposition during the treatment. Liquid yields are higher than for high-temperature carbonisation, but are still no more than 15-20% at most. The main product is a char with a reduced hydrogen and heteroatom content. The USA has led the development of this process, primarily as a means to upgrade low-rank sub-bituminous coals and lignites, to increase calorific value (CV) by rejection of the coal's oxygen as carbon dioxide (CO<sub>2</sub>) and reduce the sulphur content. At least one process has been developed to a semi-commercial scale. Given the large potential market for these technologies as fuel-upgrading processes, there is significant potential for these processes to contribute to the global supply of liquid fuels, despite their low liquid yields.

A higher yield of liquids can be obtained by rapid pyrolysis. These processes operate at temperatures up to 1200°C, but the residence time of the coal is significantly reduced, to a few seconds at most. Rapid pyrolysis is aimed at producing chemical feedstocks rather than liquid fuels and process economics are likely to be highly unfavourable for production of liquid fuels. There also appear to be unresolved engineering difficulties.

The disadvantage of all pyrolysis and carbonisation processes is that, if the hydrogen content of the feedstock coal is to be increased to the point at which it becomes a distillable liquid, the hydrogen content of the remaining solid must be reduced. In any case, the liquids produced are still of low quality and require, at least, additional treatment to remove solid contaminants and water. The resulting liquid products can then be blended to produce heating fuels and fuels for stationary turbine use. For the raw products to be used unblended, or to be used in transport fuels, they require still further treatment. Blending and processing with conventional refinery feedstocks is likely to be needed for this to be economically viable. This has not been successfully demonstrated to date.

Most of the recent interest in these processes has been in the USA as a method for upgrading coals. At least three technologies have been developed to pilot scale. Processes differ mainly in the design of the pyrolysing reactor. One process, the Liquids from Coal (LFC) process, has been in commercial-scale operation since 1992.

The LFC process (Figure 2) is a mild pyrolysis method for upgrading coal and was developed by SGI International. There are two saleable products: a low-sulphur, high heating-value solid known as 'process-derived fuel'

(PDF) and a hydrocarbon liquid known as 'coal-derived liquid' (CDL). PDF yields are considerably higher than CDL yields.

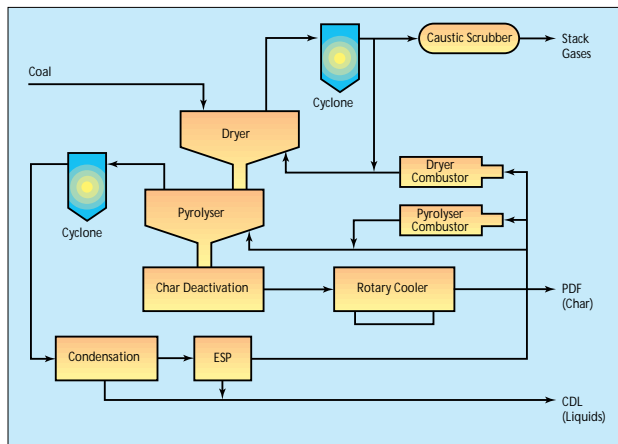


Figure 2. Encoal's LFC process

A demonstration plant owned by Encoal Corporation is located near Gillette, Wyoming, built with support from the United States Department of Energy's (USDOE's) Clean Coal Technology Demonstration Program. This was commissioned in 1992 and has a maximum capacity of 1000 tonnes/day of sub-bituminous Powder River Basin coal with a high moisture and low sulphur content.

The coal is crushed and screened and then heated by a hot gas stream on a rotary grate dryer. Control of the solid bulk temperature is critical at this point, as the intention is to reduce the moisture content without initiating structural changes in the coal, thereby minimising premature gas production. The dried coal is then fed into the main rotary grate pyrolyser where it is heated to ~540°C by a hot recycled gas stream. Again, accurate control of the solid heating rate and the residence time is required, as these parameters affect the product properties. On discharge from the pyrolyser the solids are cooled and pass to a deactivation step, consisting of treatment in a vibrating fluidised bed with a gas stream of controlled oxygen content. The PDF is thereby stabilised by slight oxidation, reducing the risk of subsequent pyrophoric behaviour.

The gas stream leaving the pyrolyser is cooled in a quench tower, condensing CDL but leaving water in the gas phase. Most of the residual gas is recycled to the pyrolyser, some being burned in the pyrolyser combustor to provide the necessary heat. The remaining gas is burned in the dryer combustor and passes into the dryer gas recycle loop. The purge from this loop is wet-scrubbed to remove particulates and sulphur oxides. Purge liquor from the scrubbers is discharged to ponds for evaporation.

After being stabilised by mild oxidation, the PDF, a low-sulphur reactive fuel suitable for pulverised coal-fired boilers, is shipped by rail to power plant. CDL, roughly equivalent to a No.6 fuel oil, is shipped by rail to a fuel oil distributor.

In 1996 SGI signed contracts for studies on commercial LFC demonstration plant in Indonesia, Japan and China. Plans for a 6 million tons/year plant, to be built in the USA, were announced in 1997.

## DIRECT LIQUEFACTION

Direct liquefaction processes aim to add hydrogen to the organic structure of the coal, breaking it down only as far as is necessary to produce distillable liquids. Many different processes have been developed, but most are closely related in terms of underlying reaction chemistry. Common features are the dissolution of a high proportion of coal in a solvent at elevated temperature and pressure, followed by the hydrocracking of the dissolved coal with H<sub>2</sub> and a catalyst. Direct liquefaction is the most efficient route currently available. Liquid yields in excess of 70% by weight of the dry, mineral matter-free coal feed have been demonstrated for some processes in favourable circumstances. Overall thermal efficiencies for modern processes are generally in the range 60-70% if allowance is made for generating losses and other non-coal energy imports.

The liquid products from direct liquefaction processes are of much higher quality than those from pyrolysis processes and can be used unblended for

most stationary fuel applications. They do, however, require further upgrading before they can be used directly as transportation fuels. This upgrading utilises standard petroleum industry techniques, allowing the products from a liquefaction plant to be blended into the feedstock streams of a petroleum refinery.

Direct liquefaction processes can conveniently be divided into two main groups, depending on whether the initial dissolution of the coal is separated from the conversion of the dissolved coal into distillable products:

- A single-stage direct liquefaction process gives distillates via one primary reactor or a train of reactors in series. Such processes may include an integrated on-line hydrotreating reactor, which is intended to upgrade the primary distillates without directly increasing the overall conversion.
- A two-stage direct liquefaction process is designed to give distillate products via two reactors or reactor trains in series. The primary function of the first stage is coal dissolution and is operated either without a catalyst or with only a low-activity disposable catalyst. The heavy coal liquids produced in this way are hydrotreated in the second stage in the presence of a high-activity catalyst to produce additional distillate.

Some processes were designed specifically to co-process coal with petroleum-derived oils and these may fall into either group. Also, coal liquefaction processes from both groups have been adapted for co-processing.

## Single-stage Processes

In the mid- to late 1960s, as interest was growing, all of the available processes were single-stage. Most development therefore continued to adopt a single-stage approach. Some developers added a second stage during the 1970s, following the oil crisis, to increase the production of light oils. The single-stage processes developed furthest are:

- Kohleol (Ruhrkohle, Germany)
- NEDOL (NEDO, Japan)
- H-Coal (HRI, USA)
- Exxon Donor Solvent (EDS) (Exxon, USA)
- SRC-I and II (Gulf Oil, USA)
- Imhausen high-pressure (Germany)
- Conoco zinc chloride (Conoco, USA).

Most of these have since been superseded and abandoned. Two exceptions are the Kohleol and NEDOL processes, both of which are considered ready for commercialisation by their developers.

Several other, less important, processes were developed to a modest scale in the USA. Other countries, notably Russia and Poland, also carried out R&D on single-stage liquefaction at a significant scale; the approaches adopted are believed to be similar in most respects to the Kohleol process.

### The Kohleol Process

The Kohleol process (Figure 3) with Integrated Gross Oil Refining (IGOR+) is a relatively recent development, by Ruhrkohle AG and VEBA OEL AG, of the process used on a commercial scale in Germany until 1945. Development proceeded via a 0.5 tonnes/day and 0.2 tonnes/day continuous unit at Bergbau-Forschung (now DMT) and a 200 tonnes/day plant at Bottrop. The Bottrop plant operated from 1981 to 1987, producing over 85,000t of distillate products from 170,000t of coal over approximately 22,000 operating hours. The technology can therefore be considered to be fully demonstrated.

In 1997 the China Coal Research Institute (CCRI) signed a two-year agreement with Germany to carry out a feasibility study for a 5000 tonnes/day demonstration plant. The suitability of sites for a liquefaction plant in Yunnan Province was investigated, including the potential market for products.

Coal is slurried with a process-derived recycle solvent and a 'red mud' disposable iron catalyst, pressurised and preheated. H<sub>2</sub> is added and the

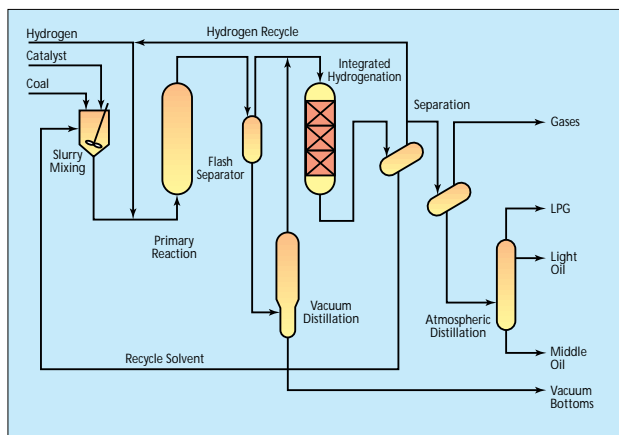


Figure 3. The Kohleol process

mixture passed to an up-flow tubular reactor, operating typically at 300bar and 470°C. The specific coal feed rate to this reactor is in the range 0.5-0.65 tonnes/m<sup>3</sup>/hour. Products from the top of the reactor pass to a hot separator. The overheads from this separator remain in the gas phase and are hydrotreated at a temperature of 350-420°C in a fixed-bed reactor at the same pressure as the main reactor. The hydrotreated products are depressurised and cooled in two stages. The liquid product from the first of these stages is recycled to the slurring step as part of the solvent. The liquid product from the second stage is routed to an atmospheric distillation column, yielding a light oil (C<sub>5</sub> - 200°C bp) and a medium oil (200-325°C bp) product.

The bottoms from the original hot separator pass to a vacuum distillation column to recover distillable liquids. These are added to the hydrotreating reactor feed, and are subsequently largely recycled as solvent. The vacuum column bottoms consist of pitch, mineral matter, unreacted coal and catalyst, and in commercial operation would be used as a gasifier feedstock for H<sub>2</sub> production.

Greater than 90% conversion can be obtained when processing bituminous coals, with liquid yields in the range 50-60% on dry ash-free coal. Process yields and quality, when using Prosper, a German bituminous coal, are summarised in Table 1.

Table 1. Kohleol process yields and product quality with Prosper coal

Process yields	Yield
Hydrocarbon gases (C <sub>1</sub> -C <sub>4</sub> )	19.0
Light oil (C <sub>5</sub> -200°C)	25.3
Medium oil (200-325°C)	32.6
Unreacted coal and pitch	22.1

Product quality	Light Oil	Medium Oil
Hydrogen (%)	13.6	11.9
Nitrogen (ppm)	39	174
Oxygen (ppm)	153	84
Sulphur (ppm)	12	<5
Density (kg m <sup>-3</sup> )	772	912

## The NEDOL Process

From 1978 to 1983, three direct coal liquefaction processes were developed by Japanese companies Nippon Kokan, Sumitomo Metals Industries and Mitsubishi Heavy Industries under a Japanese Government initiative. The initiative was managed by the New Energy and Industrial Technology Development Organisation (NEDO). By 1983 these processes had been tested at scales ranging from 0.1 tonnes/day to 2.4 tonnes/day. Rather than support each individually, NEDO amalgamated features of all three processes to produce the NEDOL process (Figure 4), targeted principally at sub-bituminous and low-rank bituminous coals. A consortium of 20 companies was then established, under the name Nippon Coal Oil Company Ltd, to design, build and operate a 250 tonnes/day pilot plant. However, the project was terminated in 1987 due to budgetary constraints.

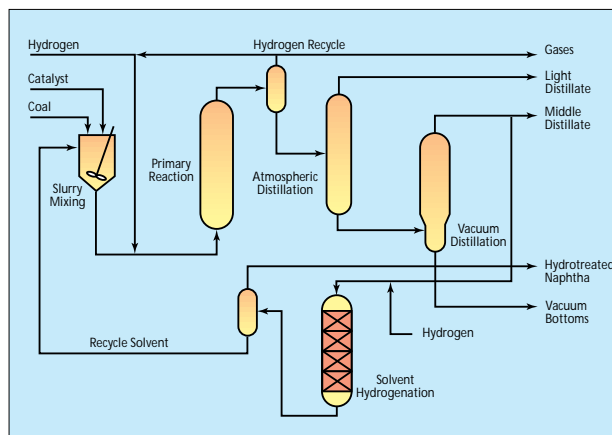


Figure 4. The NEDOL process

A 1 tonnes/day process support unit (PSU) was completed to schedule in 1988, at a cost of ~US\$30 million, and has operated on an intermittent campaign basis since that time. A redesign of the pilot plant, at a reduced scale of 150 tonnes/day, started in 1988. Construction of this plant commenced in October 1991 at Kashima, and was completed early in 1996.

The pilot plant was commissioned, followed by five operation runs, from March 1997 to September 1998. During these runs, three types of coal of different rank (Tanito Harum and Adaro coals from Indonesia and Ikeshima coal from Japan) were liquefied without serious problems. Many results were obtained, including the achievement of 80 days' continuous coal-charging operation, 58% by weight (dry ash-free coal basis) of oil yield, the successful use of slurry with a concentration of 50% and 6200 hours of cumulative operation.

CCRI signed an agreement with NEDO and the Centre for Coal Utilisation, Japan, to carry out a feasibility study for a 5000 tonnes/day demonstration plant. Several tests have been completed at laboratory- and PDU-scale with Yilan coal. At these scales, oil product yields of 62% were predicted. A pyrite from Heilongjiang province was used as the catalyst. Process evaluation, environmental impact assessment and economic analysis will be carried out in the next stage of the project, which is due for completion in 2000.

Coal is ground with 2-4% by weight of a synthetic iron-based catalyst and slurred with a recycled solvent. The slurry is mixed with H<sub>2</sub>, preheated and fed to the primary reaction stage. The reactors are simple tubular up-flow reactors operating at temperatures in the range 430-465°C and pressures in the range 150-200bar. The nominal slurry residence time is ~1 hour, with actual liquid-phase residence times in the range 90-150 minutes. The products from this primary reaction step are cooled, depressurised and distilled in an atmospheric column to take off a light distillate product.

The atmospheric column bottoms pass to a vacuum distillation column, where a middle distillate and a heavy distillate product are removed. Most of the middle distillate and all of the heavy distillate are recycled as solvent via a hydrogenation step. The vacuum column bottoms, which contain unreacted coal, mineral matter and catalyst, are discharged and in commercial operation would be used as a gasifier feedstock for H<sub>2</sub> production. The maximum solids loading which can be achieved in this stream is ~50%, but in practice it is understood that a loading of 35% is more typical. Since the pitch discharged with the solids represents a substantial loss of potential product, the process is limited to coal of relatively low ash content.

After mixing, the middle and heavy distillates from the vacuum column are fed to solvent re-hydrogenation reactors. These are down-flow packed catalyst beds operating at 320-400°C and 100-150bar. The catalysts used are variants of those used in the conventional petroleum industry for the hydrodesulphurisation of crude oil fractions. The nominal feed residence time is ~1 hour. The reactor products are depressurised at temperature into a flash distillation vessel, where a hydrotreated naphtha product is taken off. The liquid product from the flash distillation is recycled to the slurring step as the solvent.

Product yields vary with the type of coal being processed, although the primary reaction operating conditions are adjusted to minimise this. Distillate product yields of 50-55% (dry ash-free basis) are obtained with all these coals.

The liquid products are of relatively low quality and require more substantial upgrading than those from other processes, although this is not likely to present new technical difficulties.

### The H-Coal Process

The H-Coal process (Figure 5) was developed by HRI (now Hydrocarbon Technologies Incorporated, HTI) from the commercialised H-Oil process used to upgrade heavy oils. The process was developed to a 200 tons/day pilot plant, which was built at Catlettsburg, Kentucky, in 1980 and operated until 1983. Designs were subsequently prepared for a commercial-scale plant, to be built at Breckinridge, Kentucky. The H-Coal process remained the basis of most subsequent process development sponsored by the USDOE and was effectively incorporated into the Catalytic Two-stage Liquefaction (CTSL) process.

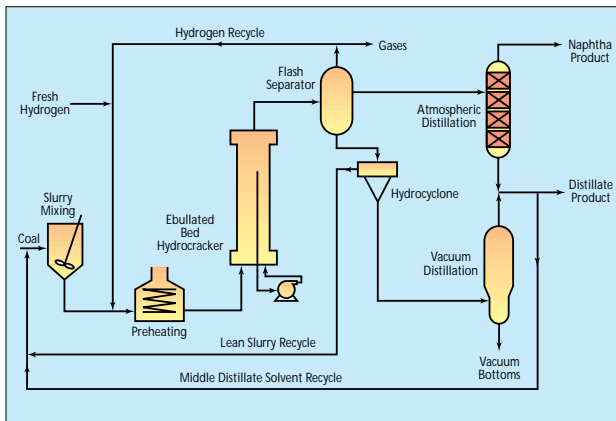


Figure 5. Schematic diagram of the H-Coal process

Coal is slurried with a recycle solvent that consists of a mixture of a solids-containing hydrocracker product with heavy and middle distillates obtained by product fractionation.  $H_2$  is added and the mixture is preheated and fed to an ebullated bed hydrocracker, which is the distinguishing feature of the process. This reactor operates at temperatures of 425-455°C and a pressure of 200bar. It contains a conventional supported hydrotreating catalyst, either nickel-molybdenum or cobalt-molybdenum on alumina. The catalyst is fluidised by  $H_2$  and a pumped internal recycle stream, for which the intake is positioned above the upper limit of the expanded bed of catalyst but still within the reactor liquid zone. This recycle stream contains unreacted coal solids.

Since the hydrocracking reactions required to produce distillable liquids are highly exothermic, accurate temperature control is crucial for engineering scale-up. The ebullated-bed reactor system offers substantial advantages over fixed-bed reactors in this, as the reactor contents are well mixed and temperature monitoring and control is more easily effected. Also, ebullated-bed reactors allow catalyst to be replaced while the reactor remains in operation, enabling a constant catalyst activity to be maintained. This is particularly important with supported catalysts as, although these have a high initial activity, they deactivate relatively rapidly at coal liquefaction conditions.

The reactor products pass to a flash separator. Liquids in the overheads are condensed and routed to an atmospheric distillation column, producing naphtha and middle distillate. The flash bottoms are fed to a bank of hydrocyclones. The overheads stream, which contains 1-2% solids, is recycled to the slurrying stage. The underflow is routed to a vacuum distillation column. Solids are removed with the vacuum column bottoms, while the vacuum distillate forms part of the product for export.

As with other processes, yields are dependent on the coal. >95% overall conversion can be obtained with suitable coals, with liquid yields up to 50% (dry basis).

### The Exxon Donor Solvent Process

Exxon Corporation started EDS process development in the 1970s and progressed to the construction of a 250 tons/day pilot plant at Baytown, Texas, in 1980. At this point Exxon considered that the process (Figure 6)

was ready for commercialisation, although development was discontinued. Liquid yields were lower than in more recent processes. As a result the process showed relatively high specific capital costs and apparently uncompetitive economics. The pilot plant was operated until 1982, with further research continued until at least 1985.

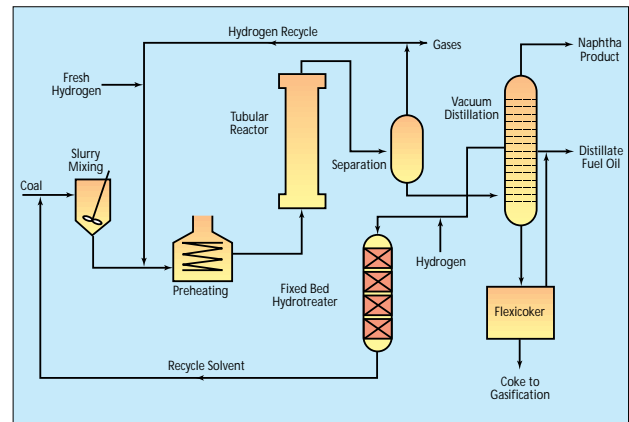


Figure 6. Schematic diagram of the EDS process

Coal is slurried with a distillable recycled solvent that has been rehydrogenated to restore its hydrogen donation capacity. This improves the effectiveness of the solvent, and this is the key distinguishing feature of the process.

The slurry is mixed with  $H_2$ , preheated and fed to a simple up-flow tubular reactor that operates at 425-450°C and 175bar. No catalyst is added. The reactor effluent passes to gas-liquid separators, from which the liquid product is fed to a vacuum distillation column. Naphtha and middle distillate products are recovered, although most of the middle distillate is recombined with the heavy distillate to form the basis for the recycle solvent. The vacuum column bottoms, containing the solid residues, are discharged and fed to a proprietary Exxon 'Flexicoker' unit. This combines pyrolysis and gasification steps to produce additional distillate product and a fuel gas, which would be used for  $H_2$  production. The pyrolysis step is carried out at temperatures in the range 485-650°C. Flexicoking is now in commercial use.

Rehydrogenation of the recycle solvent is carried out in a fixed-bed catalytic reactor, using either nickel-molybdenum or cobalt-molybdenum on an alumina support. The reactor is operated at conditions in the region of 370°C/110bar, although conditions are varied to control the degree of hydrogenation of the solvent and thus maintain its quality.

Process yields are closely related to the characteristics of the coal being processed, but can be varied to some extent by altering conditions. Typically, overall liquid yields (including the liquids produced by Flexicoking) are ~36% for lignites, ~38% for sub-bituminous coals and ~39-46% for bituminous coals (all dry ash-free basis). Liquid yields can be increased further by recycling part of the vacuum bottoms stream to the slurrying step, although this was not tested on the 250 tons/day plant. Using this technique, yields of up to 47% for lignites, 50% for sub-bituminous coals and 60% for bituminous coals could be achieved.

The boiling range distribution of the liquid products can also be varied within a wide range, depending on market requirements.

### The SRC-I and SRC-II Processes

The SRC processes were originally developed to produce cleaner boiler fuels from coal. A 0.5 tons/day plant was built in 1965 and scaled up in 1974 to two separate pilot plant. These were located at Wilsonville (SRC-I, 6 tons/day see Figure 7) and Fort Lewis, Washington (SRC-I, 50 tons/day). The Fort Lewis plant was later converted to a SRC-II unit, although because of the more severe conditions required for SRC-II, the capacity was downgraded to about 25 tons/day. The objective of the SRC-II process is to produce distillate products. Detailed designs for large-scale plant were subsequently prepared, although these plant were not built. The Wilsonville plant continued to be funded by the USDOE until 1992 as a pilot-scale test facility for the whole US direct liquefaction development programme. The SRC processes have now been abandoned in their original form, but elements have been incorporated in more recent US processes.



Figure 7. SRC-1 6 tons/day direct liquefaction plant at Wilsonville

### The Imhausen High-pressure Process

The development of the Imhausen high-pressure process commenced in 1982 and a 100kg/day PDU was commissioned in 1984. The process operating conditions appear very severe (470-505 °C and 600-1000bar). As a consequence, it seems unlikely that the process could be successfully commercialised unless yields were exceptionally high.

### The Conoco Zinc Chloride Process

In the late 1970s and early 1980s Conoco worked on the development of a process which uses molten zinc chloride to hydrocrack coal directly to give good yields of gasoline in a single step. This process is one of the very few direct liquefaction processes that is not a direct derivative of pre-war technology. The process was taken to the 1 ton/day pilot plant scale, although this was operated for only a short period and with limited success. Major metallurgical difficulties were experienced as a result of the highly corrosive nature of zinc chloride and other chloride salts that formed in the system. These difficulties remain, but if they could be resolved this is one area in which further development might have the potential to make a significant improvement in liquefaction economics.

## Two-stage processes

Most two-stage direct liquefaction processes were developed in response to the oil price rises of the early 1970s, often as a development of earlier single-stage processes. Work was carried out in many different countries, but relatively few processes were developed beyond the laboratory scale and many were generically very similar. Processes include:

- Catalytic Two-Stage Liquefaction (CTSL) (USDOE and HRI, now HTI, USA)
- Liquid Solvent Extraction (LSE) (British Coal Corporation, UK)
- Brown Coal Liquefaction (BCL) (NEDO, Japan)
- Consol Synthetic Fuel (CSF) (Consolidation Coal Co, USA)
- Lummus ITSL (Lummus Crest, USA)
- Chevron Coal Liquefaction (CCLP) (Chevron, USA)
- Kerr-McGee ITSL (Kerr-McGee, USA)
- Mitsubishi Solvolysis (Mitsubishi Heavy Industries, Japan)
- Pyrosol (Saarbergwerke, Germany)
- Amoco CC-TSL (Amoco, USA)
- Supercritical Gas Extraction (SGE) (British Coal Corporation, UK).

Only the CTSL, LSE and BCL processes continued in development beyond the late 1980s.

### The Catalytic Two-stage Liquefaction Process

The CTSL process is a development of the H-Coal single-stage process. In the pilot plant at the Wilsonville liquefaction test facility, the process was further developed over a period of almost 15 years. This facility was closed in 1992, but development work funded by the USDOE continues,

including continuous bench-scale and PDU-scale testing at HTI's Lawrenceville facility. The process has now evolved into a generic composite of much of the liquefaction development work funded by the USDOE in the 1980s and 1990s. Most recently, a close-coupled configuration has been adopted in which both stages use an active supported catalyst.

Earlier configurations included a thermal first dissolution stage, sometimes assisted by a low-activity disposable catalyst similar to that used in many single-stage processes. Both short contact and longer residence times were tested. In recent work HTI has included an in-line hydrotreater after the second-stage reactor to improve product quality.

In 1997, HTI signed a two-year agreement with CCRI to carry out a feasibility study for a direct coal liquefaction plant using Senhwa coal. HTI has support for this from the USDOE. Laboratory-scale tests and pre-feasibility studies were undertaken and potential sites and product markets investigated in the Shenfu Dongsheng area.

A schematic diagram for the most recent version of the process is given in Figure 8. Coal is slurried with a process-derived recycle solvent, preheated, mixed with H<sub>2</sub> and fed to the bottom of an ebullating bed reactor. This reactor contains a supported catalyst, generally nickel-molybdenum on alumina, which is fluidised by an internal recycle in the reactor. The reactor, therefore, has the characteristics of a uniform-temperature continuous-stirred tank reactor.

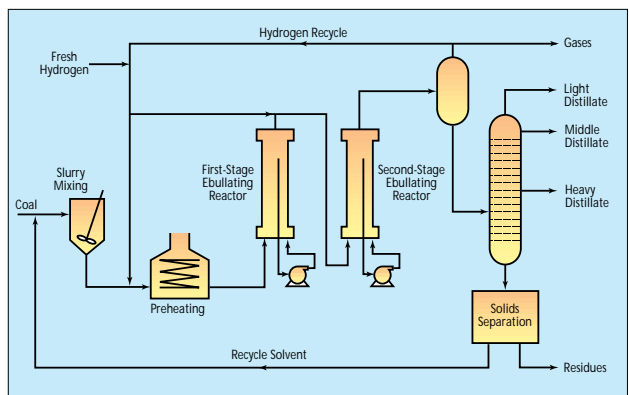


Figure 8. The CTSL process

The solvent acts as a hydrogen donor and solubilises the coal by breaking down its structure to a substantial extent in the first reactor. This first reactor substantially rehydrogenates the solvent.

Typically, operating conditions of 170bar and a temperature of 400-410°C are used when processing bituminous coals. With sub-bituminous coals, higher temperatures may be required to effect this structural breakdown and the hydrogen donor effect becomes of less importance. In one process variant, a dispersed molybdenum or iron catalyst may be used in the first stage in these circumstances.

The reactor products pass directly into the base of a second, ebullating-bed reactor stage, operating at the same pressure as the first stage but at a higher temperature (~430-440°C). This reactor also contains a supported catalyst, generally but not necessarily the same as that in the first reactor.

After separation and depressurisation steps, the products from the second reactor enter an atmospheric distillation column, where distillate products boiling up to 400°C are removed. The bottom stream from this column contains solvent, unreacted coal and mineral matter. These solids are removed by one of several possible techniques and the solvent is recycled to the slurrying step. In some process variants, only part of the atmospheric column bottom stream is routed to the solids removal step, resulting in the recycle solvent containing mineral matter and any dispersed catalyst that may have been used. The process is designed to operate without the need to remove a separate pitch stream as a product. Most operating experience at the Wilsonville plant was gained with the Kerr-McGee critical solvent de-ashing (CSD) process, also known as the residual oil solvent extraction (ROSE) process.

Since the process has now been taken back to the developmental stage, recently reported yields vary considerably depending on the coal processed, the process configuration adopted and the particular processing



conditions. Distillate product yields of 65% or higher on dry ash-free coal can be obtained, although the product is relatively high-boiling. The residue of unconverted coal and the heavy preasphaltenic material rejected with the CSD ash concentrate is approximately 20%, in equal proportions. The final operations at Wilsonville provided a well-defined base case for process conditions, yields and product quality.

### The Liquid Solvent Extraction Process

The LSE process was developed by British Coal Corporation between 1973 and 1995. A 2.5 tonnes/day pilot plant was built and operated for four years at Point of Ayr, North Wales (Figure 9), but has since been decommissioned. An outline design for a 65 tonnes/day demonstration plant has been produced in sufficient detail to allow a contractor to proceed directly to the detailed design stage. However, there are no current plans to construct such a plant.



Figure 9. LSE pilot plant facility at Point of Ayr

Figure 10 shows a schematic diagram of the process. Coal is slurried with a process-derived recycle solvent, preheated and passed to a non-catalytic digestion step, which consists of two or more continuous-stirred tank reactors in series. These reactors operate at a temperature of 410-440°C and a pressure of 10-20bar, required solely to reduce solvent vaporisation. No H<sub>2</sub> is used in this step, but the solvent acts as a hydrogen donor, transferring up to 2% by weight of hydrogen to the coal.

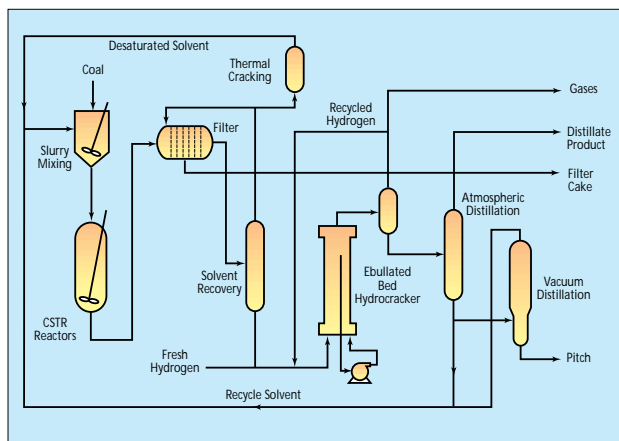


Figure 10. The LSE process

The digester product is partially cooled and filtered in a vertical-leaf pressure filter to remove unreacted coal and ash. The filter cake is washed with a light recycle oil fraction to recover product and dried under vacuum. The dried filter cake contains only a small proportion of residual non-distillable liquids; the process is therefore relatively insensitive to the ash content of the feed coal or the extent to which it can be dissolved. In commercial operation the filter cake would be gasified to produce H<sub>2</sub>.

The filtered coal extract passes to a distillation column to recover the light oil wash solvent and is then preheated, mixed with H<sub>2</sub> and routed to one or more ebullating-bed reactors in series. There is no inter-stage separation and the reactors operate at nominally the same conditions: ~200bar, 400-440°C and a space velocity in the range 0.5-1.0h<sup>-1</sup> (kg feed

per kg catalyst per hour). The reactor products are cooled, depressurised and passed to an atmospheric distillation column to recover a distillate product. The cut-point of this column is adjusted to maintain solvent balance, resulting in a product that typically boils below 300°C. The column bottoms are routed partially to a vacuum distillation column, used to control the level of pitch in the recycle solvent. The overheads from this column are recombined with the main atmospheric column bottoms stream and recycled as the solvent to the slurrying step.

A significant proportion of saturated species may build up in the recycle solvent under certain conditions, reducing the effectiveness of the extraction step. Thermal cracking is used to control this aspect of solvent quality.

The pilot plant operating programme concluded with a steady-state demonstration run. The results from this run indicate that it would be possible to operate the process in an all-distillate product mode, removing the requirement for a vacuum column. Solvent quality was maintained without the need for a separate, thermal cracking step. Primary product-quality data from this demonstration period are summarised in Table 2. The total distillate product yield is in the range 60-65% (dry ash-free coal), most of which boils below 300°C. The total filter cake yield includes ~7% of undistillable pitch.

Table 2. LSE process conditions, yields and product quality using Point of Ayr coal

Operating conditions	
Solvent/coal ratio	2.2
Digestion pressure (bar)	15
Digestion temperature (°C)	431
Nominal residence time (min)	50
Hydrocracking pressure (bar)	200
Hydrocracking temperature (°C)	434
Space velocity (kg feed/kg cat/hour)	0.76
Product yield	
C <sub>1</sub> -C <sub>4</sub> hydrocarbon gases (% daf coal)	15.4
C <sub>5</sub> -300°C distillate product (% daf coal)	49.9
300-450°C solvent surplus (% daf coal)	12.4
Pitch (>450°C) (% daf coal)	0.8
Filter cake organics (% daf coal)	23.9
Product analysis	
Hydrogen (wt%)	12.14
Nitrogen (wt%)	0.14
Sulphur (wt%)	0.04

daf: dry ash-free

### The Brown Coal Liquefaction Process

The BCL process (Figure 11) was developed by NEDO of Japan to a 50 tonnes/day pilot-plant scale, constructed at Morwell in Victoria, Australia. It was operated over the period 1985-1990, processing a total of ~60,000t of coal. Operations ceased in October 1990. The plant was decommissioned in 1991 and demolished in 1992.

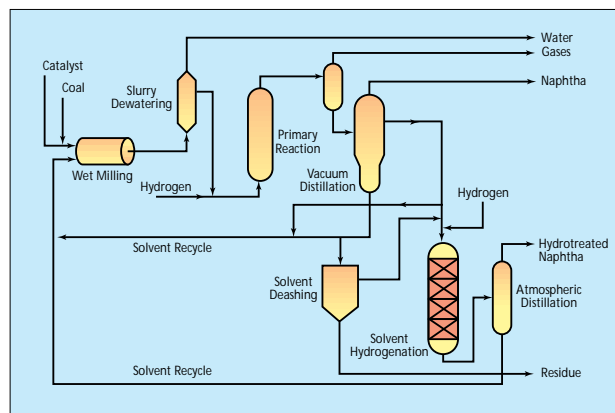


Figure 11. The BCL process

The process is designed specifically to handle very low-rank coals such as those found in the Latrobe Valley of Victoria, which may contain >60% moisture. As a result, a crucial aspect is the efficient drying of the coal. The 50 tonnes/day rated throughput of the pilot plant required ~170 tonnes/day of raw coal to be processed.

Following extensive pilot plant operation, R&D using a 0.1 tonnes/day bench-scale continuous liquefaction test facility and related equipment was carried out until 1997 to improve the reliability, economics and environmental compatibility of the coal liquefaction process. Based on the R&D results an improved BCL process was proposed. This comprises slurry de-watering, liquefaction, in-line hydrotreating, and de-ashing, with the following features:

- use of a high-active and inexpensive catalyst such as limonite ore pulverised in oil
- use of a heavy fraction solvent (bp 300-420°C)
- adoption of coal liquid bottom (CLB) bp>420°C recycling.

Compared with the results of the pilot plant, the increase of oil yield, improvement of product oil quality and suppression of scale formation in reactors were proved using the bench-scale unit with <1% (dry ash-free coal) catalyst addition. It was estimated that the improved process could decrease the crude oil equivalent nominal price by 24% compared with the BCL process at the Australian pilot plant.

## Co-processing

'Co-processing' is generally a variant on other direct liquefaction processes.

It involves simultaneous upgrading of coal and of a non coal-derived liquid hydrocarbon. The liquid hydrocarbon also serves as the slurrying and transport medium for the coal. This is usually a low-value high-boiling-point material, such as bitumen, an ultra-heavy crude oil or a distillation residue or tar from conventional crude oil processing. There is no solvent recycle loop and the underlying process may be either single- or two-stage. In general, co-processing technologies are based on adaptations of pre-existing direct liquefaction processes to a once-through non-recycling basis. In these processes most of the liquid product derives from the oil rather than from the coal.

The overall aim of co-processing is to upgrade the petroleum-derived solvent at the same time as the coal is liquefied, thereby reducing capital and operating costs per unit of product. However, the non coal-derived solvents are both poor physical solvents for coal and poor hydrogen donors. This results in a relatively low conversion of the coal to liquid products. The economics of co-processing, therefore, depend predominantly on the differential between the heavy liquid feedstock cost and the price of conventional crude oil. The addition of a low-price coal to the feed improves the process economics by reducing the average feedstock cost. Compared with other liquefaction routes, capital costs are generally significantly lower per unit of product, since most of the product is derived from the oil feedstock. In practice, the true competitors for co-processing are likely to be heavy oil upgrading processes.

Although some co-processing technologies have been developed to several tonnes/day PDU, or pilot plant scale, they have not been developed to the same degree as other liquefaction processes. None has been demonstrated at significant (~100 tonnes/day) scale.

Processes include:

- MITI Mark I (Japan)
- MITI Mark II (Japan)
- the Cherry P Process (Osaka Gas Co., Japan)
- Solvolysis (Mitsubishi Heavy Industries, Japan)
- Mobil (USA)
- Pyrosol (Saarbergwerke, Germany)
- Chevron (USA)
- Lummus Crest (USA)
- Alberta Research Council (ARC, Canada)

- CANMET AOSTRA (Canada)
- Rheinbraun (Germany)
- TUC (Technical University of Clausthal, Germany)
- UOP Slurry-catalysed (UOP, USA)
- HTI (USA).

The most important of these are ARC, CANMET, HTI and Lummus Crest.

### Lummus Crest Co-processing

Lummus Crest Co-processing was developed to a 0.25 tons/day PDU in the early and mid-1980s. It is a derivative of the Lummus ITSL process. A key feature of the process is that the petroleum heavy oil is hydrogenated prior to its use as slurrying solvent for the coal. This generates some hydrogen donor capability, increasing the extent to which the coal is dissolved and reducing repolymerisation and coking reactions.

Coal is slurried with the hydrogenated petroleum residue and reacted in an uncatalysed, short contact time reactor at a temperature of 430-450°C and a hydrogen pressure of 140bar. The reactor products pass directly to the second stage LC-Finer ebullated-bed reactor, which operates at the same pressure and a temperature of 400-435°C with a supported hydrotreating catalyst. As with many other co-processing options, the intention appears to be to integrate the process within an existing oil refinery. ~90% of the coal is dissolved in the first stage on a dry ash-free basis, with overall conversions approaching 95%. The overall conversion of heavy material in the petroleum residue is 70-80%. The total net yield of distillable products is in the range 50-55% on fresh feed.

### Alberta Research Council Co-processing

ARC, in collaboration with Canadian Energy Developments, originally developed the two-stage Counter-flow Reactor (CFR) process for upgrading tar-sand bitumen. Subsequently, the process was adapted to co-process sub-bituminous coals with bitumen. Incorporation of coal is said to increase distillable oil yields in comparison with those obtainable from bitumen alone. The process is unusual as it uses a CFR and, in place of H<sub>2</sub>, uses CO and water in the first stage. The first stage has been tested at a scale of 0.25 tons/day for co-processing and 5 tons/day for bitumen alone.

Coal is first cleaned by an oil agglomeration technique and then slurried with bitumen, water and a disposable alkali metal catalyst. The mixture is fed to the top of the counter-flow reactor, which operates at 380-400°C and 87bar. CO is fed to the base of this reactor and travels upwards, the shift reaction generating H<sub>2</sub>. The high oxygen content of sub-bituminous coals is reduced by use of CO and steam and the process is claimed to be more effective and lower cost than the direct use of H<sub>2</sub>.

The second stage, in principle, consists of a second CFR reactor system operated at ~420-480°C/175bar. Either H<sub>2</sub> or CO/steam could be used in this stage. There is no recycle of product from the second stage.

The conversion of the coal depends primarily on the coal characteristics, but conversions of up to 98% on dry, ash-free coal can be obtained in some cases. The overall product yield from the two stages is approximately 70% on the combined weight, dry ash-free coal and bitumen-fed.

### CANMET Co-processing

The CANMET hydrocracking process was intended to hydrocrack heavy oils and was developed to a 5000 barrels (bbl)/day commercial scale at the Petro Canada Montreal refinery by 1985. A variation of the process was adapted for co-processing and was taken to the 0.5 tons/day pilot plant scale in a three-year R&D consortium programme sponsored by Rheinbraun AG, Amoco Corporation and the Alberta Oil Sands Technology and Research Authority (AOSTRA). CANMET discontinued R&D in 1993.

Coal and a disposable coal-based catalyst are slurried with a petroleum vacuum residue or bitumen, mixed with H<sub>2</sub> and fed to a single-stage up-flow reactor. Typical operating conditions are reactor temperatures from 440-460°C, pressures from 10-15MPa with feedstock coal concentrations of 30-40wt% (mineral matter-free basis).

The reactor product is separated and fractionated to recover distillate products and an undistillable residue. The extent to which coal is converted is highly dependent on coal characteristics, but may be as high

as 98% on a dry ash-free basis. The conversion of high-boiling material in the bitumen or vacuum residue may be up to 70%, depending on reaction severity. Overall net distillable oil yields of up to 80% on dry ash-free slurry feed are reported.

### HTI Co-processing

HTI (previously HRI) has carried out test work on co-processing since 1985, treating it as a simplified version of the mainstream two-stage direct CTSL coal liquefaction process. It differs only in that there is no recycle solvent loop. Most work has been carried out with lignites and other low-rank coals. Recent work includes the use of HTIs GelCat™, an iron-based dispersed catalyst.

Coal conversions of up to 91% (dry ash-free basis) have been reported. The conversion of heavy material in the petroleum residue varied from 80% to 90%, with overall distillable product yields in the range 77-86% by weight on the total feed.

## INDIRECT LIQUEFACTION

The only 'core' unit specific to indirect liquefaction is the synthesis reaction step. Here, a consensus has developed that slurry-phase fluidised-bed reactors are preferable. The majority of recent work in the field has therefore concentrated on the development of improved catalysts, which need not be specific to a particular process.

Synthesis reaction technologies have applications outside coal liquefaction. In particular, there has been substantial R&D on processes intended to convert natural gas to liquids. Since these processes all involve the preliminary partial oxidation or steam reforming of natural gas to produce synthesis gas, there seems no reason in principle why they could not be applied to a synthesis gas derived from coal. Two of these, developed by Mobil and Shell, have been put into commercial-scale operation. Others have been tested only to a pilot plant scale, including processes from BP-Kvaerner, Exxon and Syntroleum Corporation, all similar to the Shell process (or the Sasol process) except for their use of different and proprietary catalysts. Sasol also offers a natural gas synthesis process, the Sasol Slurry Phase Distillate (SSPD) process, based on its Sasol process synthesis technology.

Indirect liquefaction involves, as a first step, the complete breakdown of the coal structure by gasification with steam. The composition of the gasification products is then adjusted to give the required mixture of H<sub>2</sub> and CO, and to remove sulphur-containing catalyst poisons. The resulting 'synthesis gas' is reacted over a catalyst at relatively low pressure and temperature. The products may be paraffins, olefinic hydrocarbons or alcohols (particularly methanol), depending on the catalyst selected and the reaction conditions used.

The only commercial-scale coal liquefaction process currently in operation is the indirect Sasol process of South Africa. Sasol produces gasoline, diesel fuel and a wide range of chemical feedstocks and waxes from three plant.

### The Sasol Process

The Sasol process is based on the Fischer-Tropsch (FT) liquefaction process operated on a large scale in Germany and elsewhere in the 1930s and 1940s, although over 40 years' continued development by Sasol has resulted in substantial improvements in the synthesis step. The production of synthesis gas by the Lurgi gasification of coal has remained relatively unchanged except for an increase in gasifier size. The first plant, Sasol 1, was built at Sasolburg in South Africa in the mid-1950s with a capacity of ~6000bbl/day of gasoline. The much larger Sasol 2 and 3 plant were completed at Secunda in 1980 and 1982 respectively. These plant were each designed to produce 50,000bbl/day of gasoline, together with substantial quantities of other products for use as chemical feedstocks, from the processing of 30,000 tons/day of coal.

A schematic diagram of the Sasol process, based on the Sasol 2 and 3 plant, is shown in Figure 12.

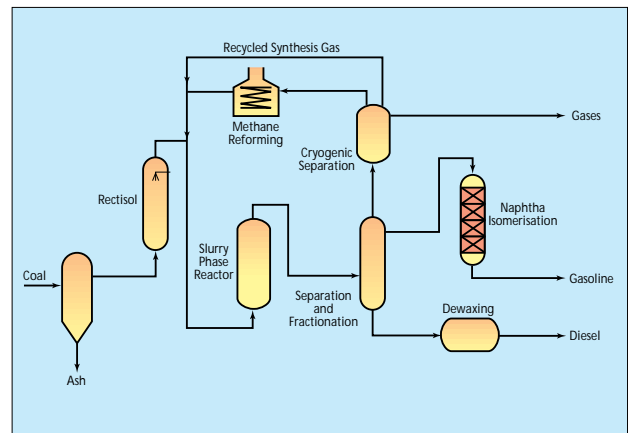


Figure 12. The Sasol process

The developments by Sasol have resulted in several changes to the processes now used. Sasol uses low-temperature Fischer Tropsch (LTFT) and high-temperature Fischer Tropsch (HTFT) for synthesis. LTFT is used exclusively at Sasolburg and comprises the older Arge fixed-bed technology as well as the newer-generation slurry-phase FT process. At Secunda the HTFT process is used exclusively and comprises older circulating fluidised-bed technology as well as new-generation Sasol Advanced Synthol (SAS) technology. SAS technology, first used in Secunda in 1995, gives the Sasol 2 and Sasol 3 plant a capacity of ~150,000bbl/day of crude equivalent of white products plus chemicals.

The incoming coal is wet-screened and the <5mm fraction directed to the steam boiler whilst the >5mm fraction goes to the Lurgi gasifiers. Raw-gas clean-up on both sites is carried out using conventional Lurgi Rectisol with cold methanol as the wash medium. The LTFT process operates at 200-250°C and 20-30bar, and produces paraffins and waxes using an iron-based catalyst.

The HTFT process operates at 300-350°C and 20-30bar, again with an iron-based catalyst, and produces a lighter, more olefinic product slate including gasoline, petrochemicals and oxygenated chemicals. The gasoline produced by upgrading the primary products is of particularly good quality.

The SSPD process was developed for monetising natural gas and comprises natural gas reforming, slurry-phase FT and mild hydroprocessing to produce naphtha and an excellent diesel. The SSPD process uses a cobalt-based catalyst specially developed for the slurry-phase system. The naphtha, because of its paraffinic nature, has a low octane number and as such is poor quality for gasoline, but it is a very good cracker feedstock. Studies carried out by Sasol have shown that Haldor Topsøe autothermal reforming, used to reform the natural gas with oxygen, is the most appropriate for the FT process. Hydroprocessing of the products is very mild and the slurry-phase FT has been demonstrated commercially in a 2500bbl/day unit at Sasolburg. Sasol believes that this relatively simple three-step process is superior in all aspects and can be built and operated economically.

A schematic diagram of the SSPD process is shown in Figure 13.

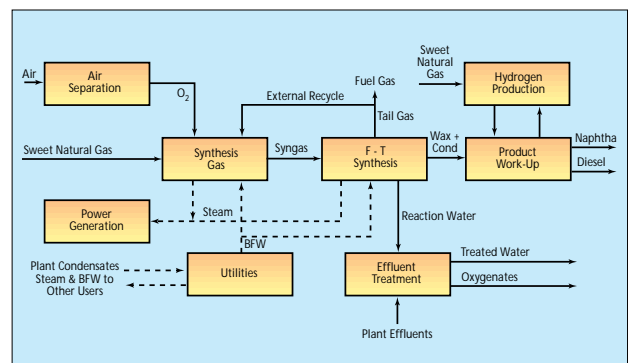


Figure 13. SSPD process

## The Mobil MTG Process

The Mobil methanol-to-gasoline (MTG) process produces gasoline from coal or natural gas in two distinct steps. The process has been taken to a commercial scale in a 12,500bbl/day plant built in New Zealand to process gas from the Maui field. Although this plant is still operating, it has recently been used solely for methanol production, for which the economics are currently more favourable.

In the first step, synthesis gas produced by steam reforming of natural gas or by coal gasification is reacted over a copper-based catalyst to produce methanol in near to 100% yield. The reaction is carried out at 260-350°C and 50-70bar.

The second step involves partial dehydration of methanol to dimethyl ether at 300°C over an activated alumina catalyst, followed by reaction over a fixed-bed zeolite ZSM-5 catalyst. These reactions are strongly exothermic, with the feed entering the reactor at 360°C and leaving at 415°C. The reactor pressure is 22bar. A series of reactions converts methanol and dimethyl ether to olefins and then to saturated hydrocarbons. Yields of material in the gasoline boiling range represent ~80% of the total hydrocarbon product. With alkylation of by-product propane and butane, total gasoline yields of 90% at 93.7 RON (octane number) were achieved at the New Zealand plant. The use of a fluidised-bed reactor offers advantages for temperature control and maintenance of constant catalytic activity over a fixed-bed system. The fluidised-bed reactor operates at an almost isothermal temperature of 410°C but at a pressure of only 3bar. Primary, gasoline-range liquid yields are lower, but there is little difference in final gasoline yields after alkylation.

## The Shell SMDS Process

The Shell Middle Distillate Synthesis (SMDS) process produces a high-quality diesel fuel from natural gas. It is a process being considered in many Gas-to-Liquids (GTL) processes in oil production. As the process produces liquids from synthesis gas, although the technology is primarily aimed at a natural gas feedstock, synthesis gas generated from coal gasification would, presumably, be equally suitable. A schematic diagram of the process is shown in Figure 14.

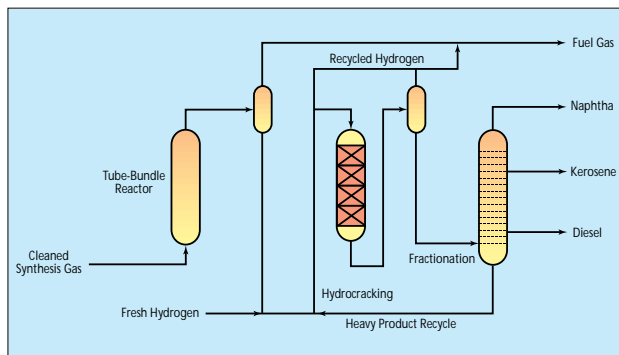


Figure 14. Schematic diagram of SMDS process

Natural gas is first partially oxidised in an oxygen-blown Shell gasifier to produce synthesis gas. This gasification approach is preferred over steam reforming, despite the considerably higher capital cost and lower thermal efficiency, because it produces a synthesis gas with the correct CO:H<sub>2</sub> ratio of 1:2. Steam reforming produces excess H<sub>2</sub>, which in a stand-alone operation can only be used as fuel.

The cleaned synthesis gas is then reacted over a proprietary Shell catalyst in a fixed-bed tube-bundle reactor that is cooled in boiling water. The product is almost exclusively paraffinic. The catalyst formulation and operating conditions in this step are deliberately chosen to give a much higher-boiling product than usual, since this minimises the production of hydrocarbon gases.

In the final step, the waxy heavy paraffin is catalytically hydrogenated, isomerised and hydrocracked in a single trickle-bed reactor over a proprietary catalyst to give products that are mainly middle distillates. The reactor operates at 300-350°C and 30-50bar. A high degree of product recycle is used to minimise the production of light products and to ensure that higher bp products are recycled to extinction. By varying the hydrocracking severity and the extent of recycle, the product distribution can be adjusted to give up to 60% diesel, with 25% kerosene and 15%

naphtha. Alternatively, up to 50% kerosene can be produced, with 25% each of naphtha and diesel.

## THERMAL EFFICIENCIES AND CO<sub>2</sub> EMISSIONS

The thermal efficiency of a plant is defined as the percentage of the CV of the input fuel that is in the finished products. Typically therefore (as shown in Figure 15), in an LSE plant, coal is the only energy input providing feedstock to reactors and utilities and the products are finished transport fuels. For the LSE process the thermal efficiency is 67.8%. This can be improved to just over 70% if integrated gasification combined-cycle power generation is used. Other single- and two-stage direct liquefaction schemes have values in the range 60-70%, lower values being usually caused by low-grade coal feeds which take up to 5% of the coal's energy to drive off its moisture.

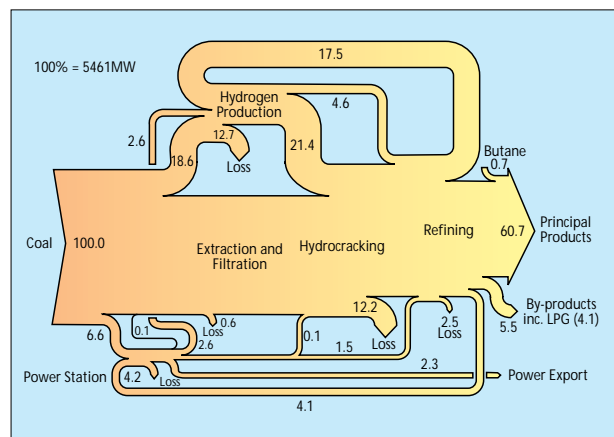


Figure 15. Energy flow diagram for a conceptual LSE plant

However, the only processes currently under consideration with multiple reactors are expected to give 65-70% to finished fuels; 67% is thus a fair average figure for direct liquefaction.

Indirect liquefaction of coal is much less efficient, Sasol 1 being only 37%. However, Sasol 2 and 3 are said to be much better, perhaps in the mid-50s%.

Pyrolysis is even less efficient because the solid product is only equivalent to coal, thus the thermal efficiency of finished transport fuels from the net coal input is likely to be 45% at most. However, in pyrolysis, the liquids are merely by-products of a solid fuel upgrading process.

For the purposes of deriving the CO<sub>2</sub> impact of liquefaction, 'standard' bituminous coal and crude oil can be selected (Table 3), each of which may be used to generate transport fuels.

Table 3. 'Standard' bituminous coal and crude oil

Standard fuels	Bituminous coal	Crude oil
Carbon (%)	86	86.1
Hydrogen (%)	5.5	11.8
Oxygen (%)	6.0	-
Nitrogen and sulphur (%)	2.5	2.1
Net CV (MWh/tonne)	9.66	11.49

The crude oil in Table 3 is similar to that marketed as heavy fuel oil.

Refineries with the necessary thermal or catalytic crackers have claimed thermal efficiencies of 93-94% when allowance is made for electrical power generation. Thus an efficiency of 90% may be assumed for a hypothetical refinery producing only transport fuel.

The standard coal may be converted to the standard crude oil, by direct and indirect liquefaction, at thermal efficiencies of 75% and 61% respectively, with all the carbon in the coal, not ending up in the 'crude oil', converted to CO<sub>2</sub> (either in power stations or furnaces). For direct or indirect liquefaction this amounts to 1.84 tonnes or 3.00tonnes CO<sub>2</sub>/tonne crude. Note that, even at 100% thermal efficiency, there is some CO<sub>2</sub> produced because of the release of 0.06 tonnes of hydrogen from coal.

In refining crude oil to finished fuels there would be a further 0.32 tonnes CO<sub>2</sub>/tonne of crude oil processed, giving 10.31MWh of CV in the finished products.

In all, ~7-10 times as much CO<sub>2</sub> is emitted in converting coal to transport fuels, when compared with crude oil.

## PROSPECTS FOR IMPLEMENTATION

In 1985 oil prices fell suddenly and, with brief exceptions, have remained low since. Interest in coal liquefaction for the production of transportation fuels has declined accordingly. At present, only Japan is active in large-scale process development, with a 150 tonnes/day plant in operation at Kashima near Tokyo, and China is participating in several collaborative feasibility and process development studies. It remains to be seen whether a future increase in oil prices will result in renewed interest and possibly in full-scale commercialisation. In the meantime, the emphasis has returned to coal upgrading.

Coal liquefaction, by whichever route, is capital-intensive and therefore benefits substantially from economies of scale. Most studies on process economics have assumed that a full-scale commercial plant would produce 50,000-100,000bbl/day of liquid products. Such a plant would process 15,000-35,000 tonnes/day of bituminous coal or up to double that amount of sub-bituminous coal or lignite. At the lowest end of this range, the annual consumption would be 5 million tonnes/year of bituminous coal. The output from this plant would still be small relative to that from a typical modern crude oil refinery, where a throughput of >200,000bbl/day is common.

Countries that might implement coal liquefaction must, therefore, have ready access to large quantities of coal. The economics of liquefaction depend strongly on coal costs and this coal must be delivered to the plant at a low price. Since coal is more difficult to transport than oil, it would, as a general principle, be better for liquefaction to be carried out in the country of origin and preferably at the point of origin. There will, however, be exceptions to this generalisation, particularly where

construction costs near the point of origin are likely to be high due to remoteness or where there are already good rail links.

If liquefaction is to be carried out in the country of origin of the coal, there must be sufficient reserves to last for a project lifetime of ~25-30 years. The coal feedstock required for a commercial-scale liquefaction plant equates to 3-4 million tonnes oil equivalent (mtoe)/year. This means a minimum reserve of about 100mtoe or 150 million tonnes of coal or 300 million tonnes of lignite for the required reserves and minimum reserve/production ratio (R/P) of 25. Some 24 countries meet this criterion (Table 4).

The single most important factor behind the decision to implement coal liquefaction is the price of crude oil. The discussion and the analysis summarised in Table 4 provides a comparison of the likelihood of individual countries pursuing coal liquefaction given an economically-favourable crude oil price.

## FUTURE DEVELOPMENT

The scale of oil-refining industry operations is such that it would be likely to be impractical to produce the same amount of liquids via coal liquefaction. Liquefaction can only really offer a means of providing a supplementary source of hydrocarbon liquids. This is likely to be most attractive as a source of transportation fuels once the supply of crude oil can no longer meet demand for these products. Under these circumstances, it is likely that liquefaction-derived fuels would be distributed through the existing infrastructure which, inevitably, is controlled and operated by the oil industry. This would greatly simplify the distribution and marketing of liquefaction products. It is, therefore, likely that coal liquefaction will initially be implemented by the oil industry rather than by the coal industry. This trend is demonstrated by the fact that most of the private-sector process developments have been undertaken by oil companies.

Given this scenario, it is likely to be beneficial for a liquefaction plant to export its products directly to an existing refinery, whether as raw materials for further processing or as finished fuels for simple blending into the refinery product pool. As many of the ancillary requirements, such as

Table 4. Analysis of potential implementing countries

Country	Large coal reserve	Low cost coal production	Large oil demand	R/P Ratio**	Restriction on oil availability	Potential for implementation				
						Yes				No
USA *	√	√	√	9.8	Security of supply and possible political and environmental considerations	√				
Canada	√	√	√	9.2	Low reserves	√				
Mexico	√			33.6						√
Brazil	√		√	15.5	Net oil importer with low reserves			√		
Colombia	√	√		11.7	Increasing demand with low reserves	√				
Venezuela	√			59.5						√
Bulgaria	√			0	No oil or gas		√			
Czech Republic	√			0	No oil or gas				√	
Germany	√		√	0	No oil reserves		√			
Greece	√	√	√	0	No oil reserves	√				
Hungary	√			0	No oil reserves					√
Poland	√			0	No oil reserves					√
Romania	√			32.5						√
Spain	√	√	√	0	No oil reserves		√			
Turkey	√	√	√	0	No oil reserves		√			
UK	√		√	5.2	Low oil reserves	√				
Former Soviet Union	√			25	Political and economic considerations			√		
South Africa	√	√	√	0	No oil reserves	√				
Zimbabwe	√	√	?	0	No oil reserves but no demand; economic considerations					√
Australia	√	√	√	8	Low oil reserves	√				
China	√	√	√	20.5	Oil reserves remote from demand	√				
India	√	√	√	15.6	Oil reserves remote from demand	√				
Indonesia	√	√	√	9	Low oil reserves with increasing domestic demand	√				
Japan	√	√	√	0	No oil reserves	√				

\* Oil demand in USA is ten times that of most other countries

\*\* BP Statistical Review of World Energy 1998

power and chemical feedstock supplies, would be common to both the refining and liquefaction processes, there would be opportunities for sharing some of the necessary facilities. Equally, many of the unit operations are likely to be similar in both the refinery and the coal liquefaction plant. Again, this favours implementation by the oil industry.

Integration may involve simply a shared product-blending and export facility. Even this minimal integration of facilities would offer significant capital cost reductions. Studies have shown that a 120,000bbl/day refinery could accept the whole output from a 50,000bbl/day liquefaction plant with minimal change in overall product quality, so the typical refinery of 200,000bbl/day would cope easily.

A more complex integrated approach could involve the liquefaction plant and the refinery transferring products for further processing. Primary distillates from the liquefaction plant could be blended and processed with the equivalent feedstocks in the refinery. In addition to capital savings, the greatest benefits would come from exporting low-value products from the refinery to the liquefaction plant, for gasification to produce H<sub>2</sub>. These materials may include vistar from a visbreaking unit or high-sulphur coke from a delayed coker, for which the refiner may otherwise have no outlet. This also increases flexibility in determining the refinery product mix, possibly by making additional use of coking to increase distillate production. The liquefaction plant would no longer require additional coal as a supplemental utility fuel. The joint benefit has been estimated to be equivalent to reducing the cost of liquefaction by US\$2-3/bbl.

This level of integration does not risk compromising the operability of an oil refinery. Much more complex schemes are possible, although refiners may be reluctant to implement these until they have gained confidence in the liquefaction plant operation. Some examples are the use of common H<sub>2</sub> production and power generation facilities, steam, cooling and other utility systems. A total reduction in overall liquefaction costs of ~US\$5/bbl has been suggested as achievable.

There are a relatively small number of unit operations specific to a particular coal liquefaction process. These are generally serviced by a large number of ancillary units. The ancillary units and, in many cases, the core units are now well proven on a large scale in other applications. Since 60-75% of the total estimated capital costs is accounted for by these units, overall process costs have become comparatively well defined. This is one area in which liquefaction technologies have advanced significantly since the mid-1980s. Whilst this is a considerable advantage in some respects, the potential for dramatic reductions in overall costs are limited as the core processes have evolved slowly through incremental improvements. It seems likely that this trend will continue meaning that further cost reductions will probably result from general engineering advances as much as from genuine process improvements.

The cost of catalysts is so high that what appear quite expensive means for decreasing it could be worthwhile, eg increasing reactor size substantially in order to reduce the catalyst operating temperature.

## CONCLUSIONS

### Development Status

- Liquid transportation fuels are characterised as having a hydrogen content of ~12-15%, whilst typically coal has a 5% hydrogen content and higher carbon content. Coal-to-liquid conversion processes require hydrogen to be added or, alternatively, carbon to be removed. Numerous 'direct' and 'indirect' processes have been developed for this purpose.
- Considerable R&D into liquefaction processes was carried out during the 1970s and early 1980s, mainly in the USA, Japan, the UK and Germany spurred on by oil price shocks. Since then, development has largely been put on hold.
- South Africa is the only country presently operating liquefaction plant. It has large reserves of coal but no oil and gas. Trade embargoes over three decades to the mid-1980s drove large-scale application and up to 60% of transportation fuel requirements have been met from coal.

- Coal liquefaction is now a largely proven technology for the manufacture of useful liquid products.

### Benefits of the Technology

- Coal is abundant but, in comparison with liquid fuels, inconvenient to handle and unsuited to some applications particularly transport. Coal liquefaction provides the ability to manufacture transportation fuels from coal.
- The technology provides insurance against depleting oil stocks and oil supply problems.

### Disadvantages of the Technology

- Liquefaction processes typically achieve an energy conversion (% CV of the input fuel converted to finished products) of 65-70% (direct liquefaction) and 55% (indirect liquefaction).
- Converting coal to transportation fuels results in ~7-10 times as much CO<sub>2</sub> being emitted, compared with converting crude oil. This increase in CO<sub>2</sub> emissions at the processing stage has the effect of raising overall CO<sub>2</sub> emissions from transport by ~50%, compared with transport based on conventional, refined petroleum products.

### Market Prospects

- Presently, only Japan is active in large-scale process development, whilst China (increasingly a net oil importer and containing areas remote from sources of oil) presents the strongest adoption prospects.
- The commercial viability of coal liquefaction rests with the overall economics of the process. This depends on the availability of significant quantities of poor-quality, low-cost coal, and the unavailability or otherwise relatively high cost of oil (and gas). Appropriate market conditions are likely to emerge around 2010: many countries are then likely to be involved.

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