Exxon Donor Solvent Liquefaction Process [and Discussion]

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Exxon Donor Solvent liquefaction process

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The Exxon Donor Solvent (EDS) coal liquefaction system is a direct liquefaction procedure being developed by the Exxon Corporation. Coal is chemically reacted and dissolved in a recycle solvent that is hydrogenated between passes to the liquefaction reactor. More than 2.6 barrels (0.41 m^3) of a synthetic crude boiling below 1000 °F (538 °C) are produced per ton (0.907 t) of dry, high volatile coal feed. Other ranks of coal can be effectively liquefied. The process development has proceeded to a 250 ton/day (227 t/day) pilot plant stage that went into operation in June 1980. The presentation addresses the chemical reactions and process conditions that result in ease of operability and flexibility of the EDS process.

Introduction

'Through hydrogenation it now is shown to be practicable to convert coal into liquid hydrocarbons at a cost which, although above prevailing oil prices, is not prohibitive. Thus, the coal reserves of the world become supplemental to the crude oil reserves' (W. C. Teagle, President of Standard Oil Company of New Jersey, quoted in *Scientific American*, November 1930).

Half a century later, there seems to be a general consensus that the commercial conversion of coal to hydrocarbon liquids is required. However, there is little agreement on the best way to achieve this objective. The process referred to by Walter Teagle, who was president of what is now the Exxon Corporation, was the Bergius process, a high-pressure catalytic process used extensively by the Germans during World War II. That the Bergius process was a technological success is unquestionable; whether it was an economic success is debatable. Only one alternative process for making hydrocarbon liquids from coal has proved to be a commercial success, and that is the Fischer–Tropsch process for catalytically converting the CO and H_2 of synthesis gas obtained from steam gasification of coal into hydrocarbon liquids. The Sasol group employs this procedure, but as with the German wartime efforts, economic data are difficult to obtain.

The Fischer–Tropsch process is an indirect coal liquefaction system. Most coal liquefaction procedures currently being investigated are, however, like the Bergius process, direct liquefaction processes. In these processes, the molecular structure of the coal is at least partly preserved, in contrast to its complete destruction in a preliminary gasification step. Supercritical extraction, being developed by the U.K. National Coal Board, and various pyrolysis processes do fall into this general category. In these processes, however, only a relatively small proportion of the coal is extracted as or converted to liquids. Most realistically, only those processes in which the principal product is a coal-derived liquid should be referred to as direct liquefaction processes.

All of the direct liquefaction processes that are under development and that have reasonable probability for commercial success are built around the concept of stewing coal in its own juice. That is, as shown in figure 1, powdered or crushed coal is slurried in a fraction of coal-derived liquid and heated to about 400–450 °C. A portion of the coal decomposes and is dissolved in the slurrying vehicle. A vehicle fraction is then separated from the products and recycled with fresh coal. Each of the direct coal liquefaction processes being developed in the United States employs

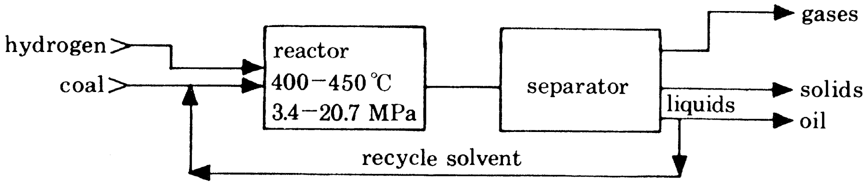


FIGURE 1. Basic elements of direct coal liquefaction process.

this same basic flowsheet, but with some additions or modifications. In the Exxon Donor Solvent (EDS) process, the vehicle is partly hydrogenated over a conventional petroleum hydrogenation catalyst. The vehicle then serves as a hydrogen donor for the chemical reactions that occur during the liquefaction process. To understand the significance of the hydrogen donor vehicle in the EDS process, it is necessary to understand what the structure of coal is, and how it is converted from a solid substance to a hydrocarbon liquid.

THE STRUCTURE OF COAL

Coal is, of course, a heterogeneous substance about which it is difficult to generalize. Coals are sedimentary rocks composed predominantly of microscopically differentiable, physically discrete and chemically distinctive organic entities referred to as macerals. Variable proportions of inorganic substances, minerals and water, are also found in coals. Three principal classes of

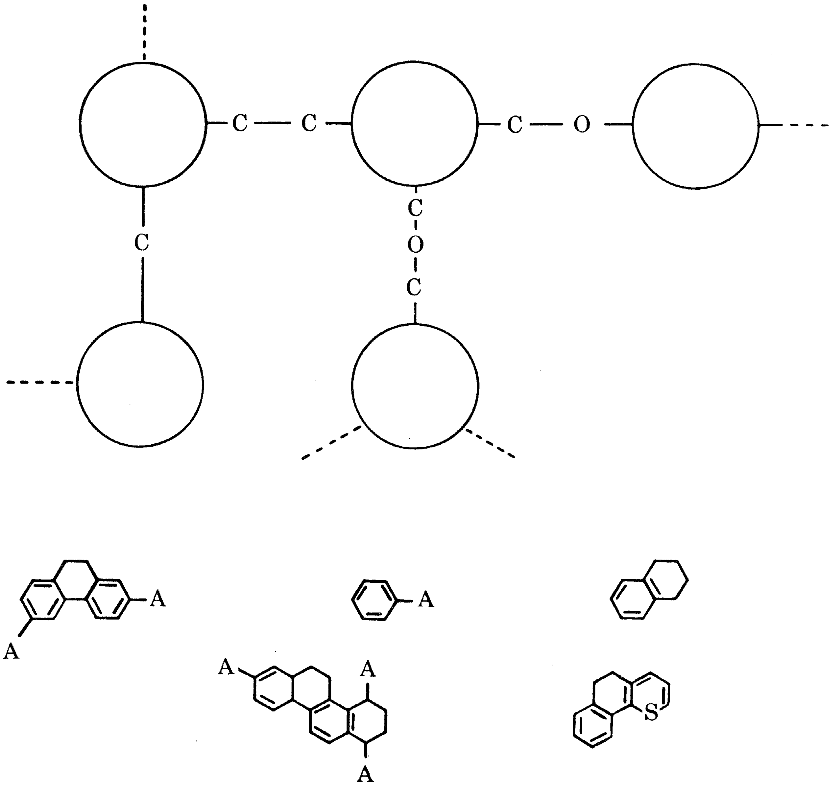


FIGURE 2. Coal modelled as cyclical units linked by 'bridges'. Circles in the upper diagram represent molecular units such as those shown in the lower, where A = $-OH$, $-CH_3$, $-CHOH$ or $-SH$.

maceral ingredients are recognized, i.e. vitrinite, inertinite and liptinite. The first two are derived from lignocellulose that constitutes plant cell-wall structures; the latter is derived from fatty acids that serve as protective tissues on plants. Vitrinite is by far the dominant maceral material composing typical coal seams, and hence its properties interest us most when considering the mechanism of liquefaction.

The specific properties of vitrinite in any given coal are largely controlled by the extent to which the woody source material has been metamorphosed or coalified by geological agencies of heat and pressure. The more severe the metamorphic processes that operated upon the coal, the higher the rank of the coal. Only the vitrinite (and the much less abundant liptinite) of lignitic and bituminous rank coals is readily amenable to liquefaction. The following discussion refers only to the vitrinitic material.

The chemical structure of vitrinite of bituminous rank coal is envisioned as being composed of highly substituted, cyclical carbon structures (variably 1, 2, 3 or more condensed rings) linked by alkyl or etheric (O, S) bridges (figure 2). Lower ranked coals contain more oxygenated functionalities, whereas the higher the rank of the coal, the fewer the functional groups and the more polycondensed the ring system.

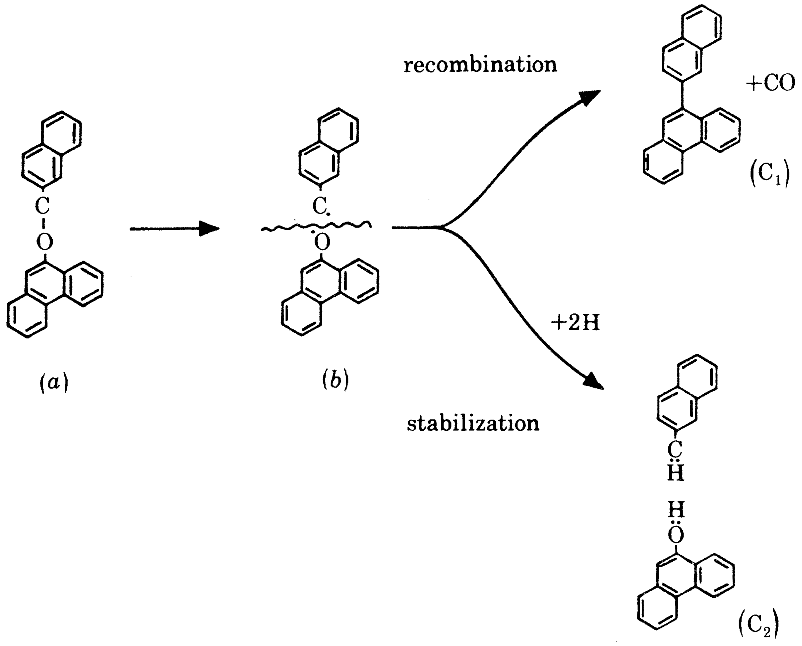


FIGURE 3. Influence of hydrogen on the fate of thermally formed free radicals.

LIQUEFACTION MECHANISM

Liquefaction is viewed as a pyrolytic process carried out at temperatures around 400–450 °C in which the bridges linking the cyclical structures are thermally ruptured. The process produces molecular fragments of various sizes and compositions from what was originally a highly extended structure composed of units like those shown in figure 2. In this process, the vehicle in which the coal is slurried acts not only as a dispersant for the molecular fragments, but also as a chemical reactant.

The thermally induced rupture of covalent bonds leads to the formation of pairs of unshared electrons on free radicals such as that depicted in figure 3*b*. These electrons are exceptionally reactive and may recombine or may avidly attack other molecular sites in a form of repolymerization (figure 3 c_1). Ideally, of course, one would like to see each unshared electron stabilized by a hydrogen atom to minimize the molecular mass of the resultant product. Both molecular hydrogen and the vehicle contribute hydrogen to this reaction.

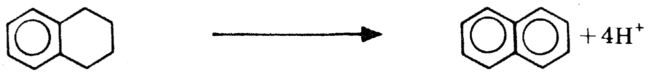
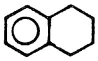
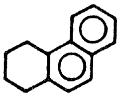
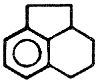
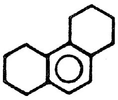
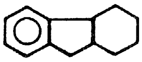
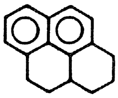


FIGURE 4. Model reaction for provision of hydrogen by hydroaromatics for coal liquefaction.

TABLE 1. SOME POTENTIAL DONOR COMPONENTS IN SOLVENT

compound	structure	compound	structure
tetralin		tetrahydrophenanthrene	
tetrahydroacenaphthene		octahydrophenanthrene	
hexahydrofluorene		hexahydropyrene	

The vehicle is commonly modelled as the compound tetrahydronaphthalene (tetralin) shown in figure 4. That is not to say that the vehicle is tetralin, but rather that many compounds in the ideal vehicle have partly saturated structures such as that of tetralin. A number of these are shown in table 1. The hydroaromatic hydrogens are readily abstracted from such compounds, providing a facile means of stabilizing the free radicals formed by coal decomposition. The unpaired electron on the vehicle created by the abstraction of a hydrogen atom enters the

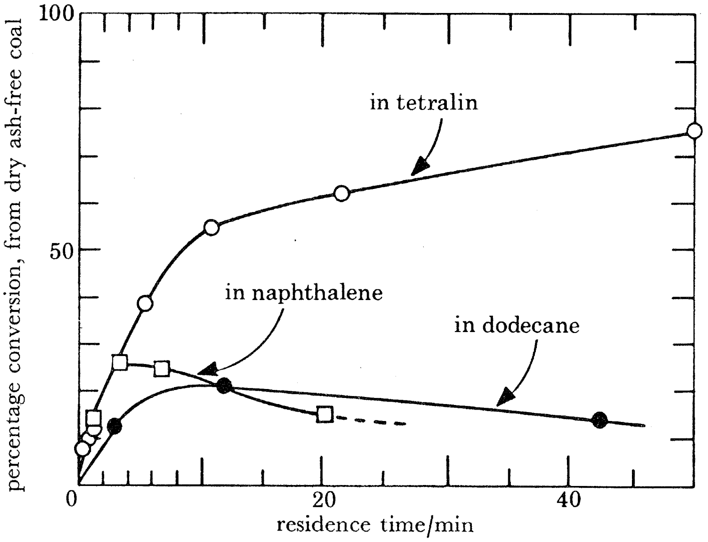


FIGURE 5. Influence of nature of vehicle on conversion of coal to benzene-solubles and gases.

aromatic π system, where it is far less likely to react with other molecular species. In this fashion, the partly hydrogenated vehicular compounds are converted to stable aromatic compounds, while at the same time the depolymerization fragments from coal are stabilized to compounds of relatively low molecular mass, some of which are liquids at ambient or elevated temperatures.

Early on in the reaction, coal is quite reactive and decomposes to materials that are soluble in solvents such as benzene or toluene (though the products still may not be distillable liquids). Indigenous hydrogen is transferred initially from the coal structure itself to stabilize free radicals. At some point, as shown by the dodecane and naphthalene curves in figure 5, inherent transferable hydrogen is consumed, and subsequently formed free radicals then repolymerize into insoluble compounds. If, however, donatable hydrogen is supplied to the system via an appropriate vehicle (e.g. tetralin), there is a progressive conversion of the coal to material of lower molecular mass material. The rate of formation of coal liquids appears to be dependent upon the rates of cleavage of various bridging systems and hence is coal-specific.

It is common to have fairly high hydrogen partial pressures in the liquefaction systems. It was commonly presumed that molecular hydrogen simply served to rehydrogenate the vehicle in the liquefaction reactor. However, L. W. Vernon of Exxon Research has recently shown that not only does molecular hydrogen stabilize free radicals, but it also initiates hydrocracking reactions that serve to reduce further the molecular mass of the coal structure. His experimental results are summarized in the following paragraphs.

Table 2. Pyrolysis of dibenzyl at 450 °C for 30 min with tetralin and molecular hydrogen

feed/mmol			products (ex 'solids') (percentages by mass)		
dibenzyl	2.8	2.8	benzene	0.1	7.1
tetralin	3.8	3.8	toluene	43.1	41.5
hydrogen	—	70	ethylbenzene	0.9	8.2
			dibenzyl	52.6	42.2

Model components containing the types of carbon–carbon bonds thought to be present in coal were pyrolysed in the presence of tetralin and molecular hydrogen at coal liquefaction conditions (450 °C, 30 min). The relative rates of conversion of the model compounds are predictable from the bond dissociation energies of the compounds. When the model structure, dibenzyl, is pyrolysed at coal liquefaction conditions, the distribution of major products depends upon the molecular hydrogen pressure. Table 2 shows the major products formed when dibenzyl was pyrolysed for 30 min at 450 °C. When dibenzyl was heated with an excess of a good donor solvent, such as tetralin, the conversion was 47 % and the only major product was toluene. Very little benzene and ethyl benzene were produced. When the same experiment was carried out in the presence of high-pressure hydrogen (about 11 MPa) the conversion was increased to 58 %. Toluene was still the primary product, but significant amounts of benzene and ethyl benzene were also produced in the presence of molecular hydrogen.

Conversion of dibenzyl in the presence of both molecular hydrogen and tetralin or in the presence of hydrogen alone proceeds along two parallel reaction paths. Toluene is produced by a thermal cracking reaction in which the rate controlling step is the thermal cleavage of the β-bond in dibenzyl. The free radical produced in the thermal cracking step abstracts a hydrogen atom either from the donor or from molecular hydrogen. Benzene and ethyl benzene, on the other hand, are produced by a hydrocracking reaction in which a hydrogen atom (from dissociation of molecular hydrogen via free-radical abstraction) attacks the α-bond of the alkyl aromatic. This two-path reaction mechanism predicts that the ratio of hydrocracking products (benzene plus ethyl benzene) to the thermal cracking product (toluene) should be directly proportional to hydrogen pressure. Figure 6 shows that the experimental data are consistent with this prediction.

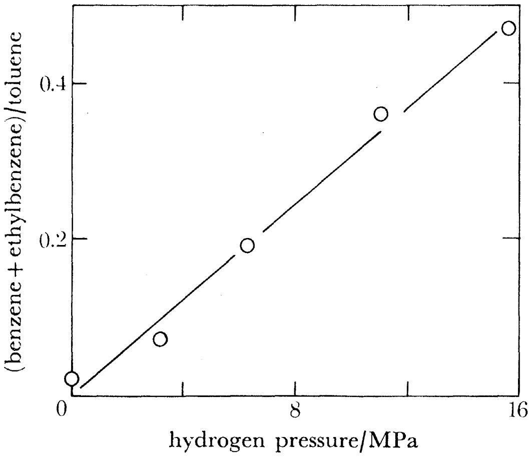


Figure 6. Effect of hydrogen pressure on yield of hydrocracking products from dibenzyl (450 °C, 30 min).

These studies on model coal structures offer firm evidence that molecular hydrogen can participate directly in free radical reactions under coal liquefaction conditions. Some of the reactions that are accelerated by molecular hydrogen are desirable in a coal liquefaction process and some are not. High-pressure hydrogen can promote the hydrocracking of some carbon–carbon bonds in the coal structure that are too strong to break thermally, and lead to higher distillable liquid yields. However, hydrogen also promotes ring opening of hydroaromatic molecules and accelerates the dealkylation of alkyl aromatic compounds; these two reactions are undesirable in a coal liquefaction process, the former causing donor solvent degradation and the latter leading to high gas yields and high hydrogen consumption. These studies suggest that the most hydrogen-efficient way to produce liquids from coal is to obtain a proper balance between the donor reactions and the molecular hydrogen reactions. Use of an efficient donor solvent along with molecular hydrogen in the coal liquefaction process produces high liquid yields at moderate hydrogen pressures and with moderate hydrogen consumption.

The Exxon Donor Solvent process

The Exxon Donor Solvent liquefaction process capitalizes both on the reactions with the hydrogen-donating solvent and the reactions with molecular hydrogen. The system flowsheet for the EDS process is shown in figure 7.

The advantageous characteristics of the EDS process are:

(1) it makes maximum use of petroleum refining technology;

(2) it is applicable to a wide range of coals (coals ranging in rank from lignitic to bituminous have been successfully processed);

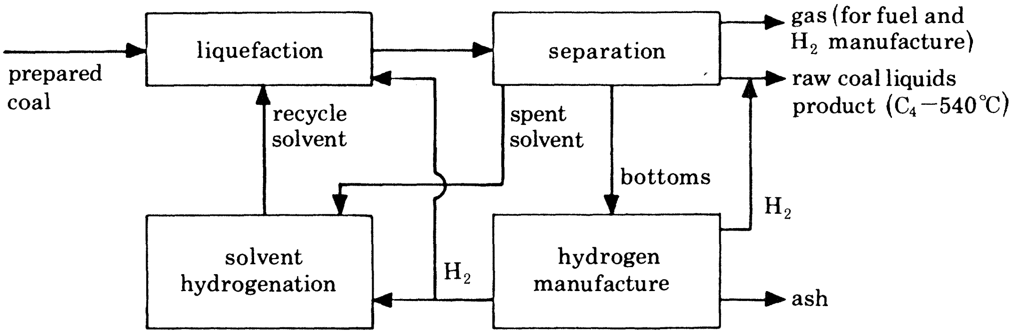


Figure 7. Simplified block diagram of the EDS process.

(3) by varying the processing conditions, it is possible to produce a variety of liquid products suitable for a variety of end uses;

(4) if necessary, liquid products can be upgraded by using conventional petroleum refining technology to relatively low sulphur, oxygen, and nitrogen contents.

The key feature that differentiates the process from others under development is the separate solvent hydrogenation step. In the hydrogenation unit, which is a simple trickle bed reactor containing conventional petroleum hydrogenation catalyst, solvent that has donated its hydrogen during liquefaction is rehydrogenated for recycling to the liquefaction reactor. Considerable experimentation was required to establish the preferred level of hydrogenation; underhydrogenation results in a lesser ability to donate hydrogen and thereby stabilize free radicals, whereas overhydrogenation forms saturated molecular species that do not serve as adequate hydrogen donors and crack to hydrocarbon gas under liquefaction conditions. In model terms, naphthalene is not a donor because it has no hydroaromatic hydrogens to donate; decalin is not a donor because it is completely saturated.

Table 3. Comparison of principal features of liquefaction test units

	tubing bomb	r.c.l.u.	c.l.p.p.	E.c.l.p.
coal quantity	5 g/run	34 kg/day	907 kg/day	226 t/day
key features	batch	continuous	continuous	continuous
	low cost			commercial scale
	flexible	flexible		
	1 day/run	1–2 weeks/run	1–2 weeks/run	
key results	conversion	process yields	process yields	process yields
	donor H transfer	product quality	product quality, operability	product quality, operability

Most of the research that has been carried out on the EDS process over the last 15 years has been devoted to defining preferred conditions for the maximization of liquid yields from various ranks of coals. A number of test units have been employed; they are compared in table 3. The tubing bombs have been employed to test process-variable effects and to screen coals, solvents and novel reaction conditions. Their advantage is that experiments can be carried out rapidly on small quantities of samples at relatively low cost. The recycle coal liquefaction units (r.c.l.us) are the real workhorses of the process development. These units are completely integrated systems that are small enough to give relatively rapid turnaround on tests, yet large enough to test concepts and process yields in a realistic continuous system with solvent recycle. They make sufficient liquid product to provide samples for characterization. The coal liquefaction pilot plant (c.l.p.p.) is of sufficient size that operations can be carried out at a scale that provides semi-commercial experience and provides adequate quantities of product samples for external testing. The Exxon coal liquefaction plant (E.c.l.p.), that started up in June 1980, is of a scale to test commercial-sized equipment.

After screening studies in tubing bombs to define the broad effects of processing variables on coal conversion, an extensive process variables study is conducted with the r.c.l.u. Upon defining preferred conditions, the c.l.p.p. is used to confirm yields, to test for operability concerns, and to generate product samples. The remainder of this paper will deal with the results from 15 years of research and development that have advanced the EDS process to a state of commercial readiness. The flexibility to process a wide range of coals and to produce a range of products will be stressed.

Effects of processing variables

Two coals have been studied intensively in the EDS development programme, one a Carboniferous-age, bituminous rank coal from Illinois (Monterey), the other a Palaeocene-age, sub-bituminous coal from Wyoming (Wyodak). Some of their properties, with properties of some other tested coals, are indicated in table 4.

Table 4. Selected properties of coals studied in EDS process

(Mass percentages of dry coal, free of mineral matter.)

	volatiles	carbon	oxygen	sulphur
Ireland	43	84	6	4.3
Burning Star	43	80	11	3.1
Monterey	46	78	12	4.3
Wandoan	55	78	15	0.2
Wyodak	48	74	18	0.5
Big Brown	52	74	20	1.2

The Illinois coal, from Exxon's Monterey no. 1 mine, is a typical, mid-continent, high volatile C bituminous coal. Sulphur content is high, as is typical of mid-continent coals. The Wyoming coal is a typical sub-bituminous coal from the Great Plains province. It is, in all important respects, similar to most western U.S. brown coals.

In the EDS process, there are complex interactions between temperature, residence time of the slurry in the liquefaction reactor, pressure, and solvent quality. The influences of residence time and temperature on the product slate from the two principally studied coals are shown diagrammatically in figures 8 and 9. Total conversion (to liquids and gases) is roughly comparable for these coals, and increases with both residence time and temperature. However, more oxygenated gases (including water) are produced from the Wyodak coal.

At the lowest practicable operating temperature (about 430 °C), longer residence time leads to increased production of liquids at the expense of non-distillable bottoms (> 538 °C material). At higher temperatures, longer residence time results in increased conversion of coal, but the additional products are predominantly hydrocarbon gases. It is presumed that the increased yield of gases results from naphthenic ring opening and subsequent dealkylation of reactants.

Experiences with the effects of pressure are, at present, rather minimal. As would be expected from the model compound studies discussed earlier, higher hydrogen partial pressures do result in some additional conversion. However, the increases have not been dramatic for the coals

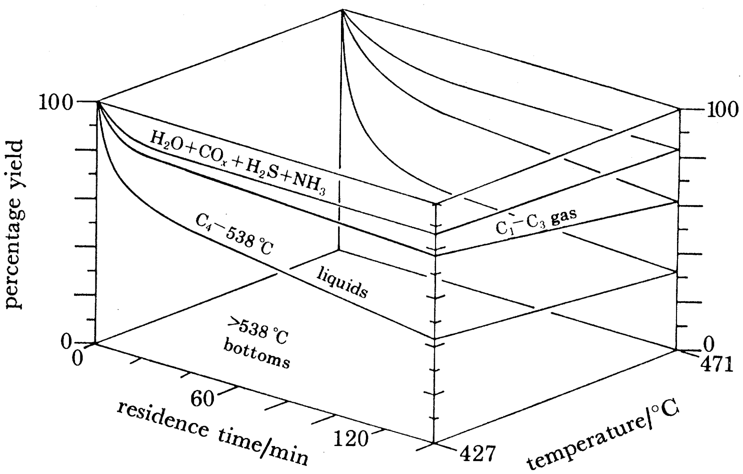


FIGURE 8. Increase in conversion of Monterey coal with increasing temperature and time.

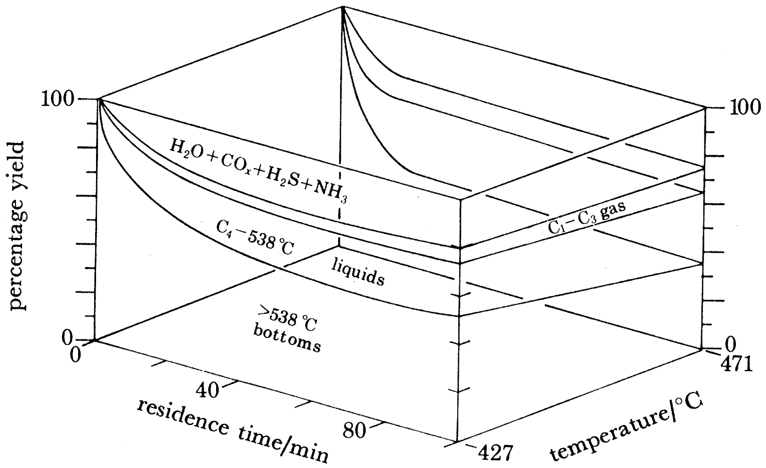


FIGURE 9. Increase in conversion of Wyodak coal with increasing temperature and time.

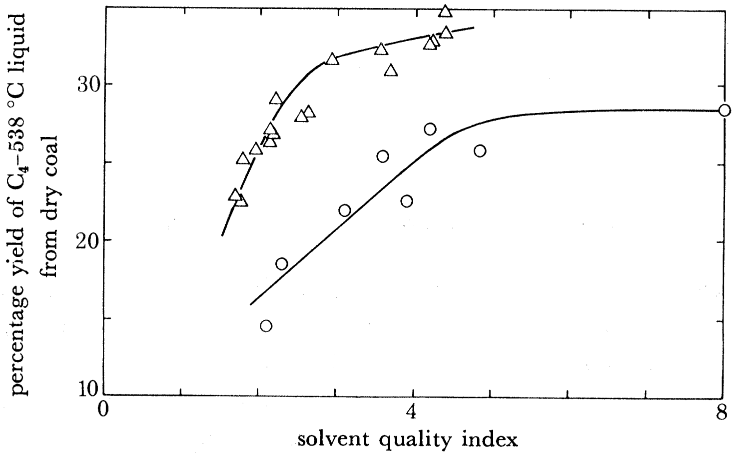


FIGURE 10. Effects of solvent quality on liquid yield for Monterey (△) and Wyodak (○) coals.

investigated in the EDS process so far. The normal operating condition is 10.3 MPa, a pressure readily achievable without resorting to esoteric metallurgies or reactor systems, and yet one that maintains a substantial portion of the solvent in the liquid phase.

Solvent quality is an important variable, and is defined on the basis of an index referred to as the solvent quality index (s.q.i.). As shown in figure 10, liquid yields from both standard coals increase with increasing s.q.i. to a point where the yield becomes asymptotic. The optimum s.q.i., however, for each of the coals is different. It is assumed that the differences in response of the coals to solvent quality are related to differences in their oxygen content. Oxygen occurs dominantly as oxygenated functional groups (principally carboxylic) that are readily decomposed at typical liquefaction temperatures. Although decomposition of functional groups does not lead to significant reduction in molecular mass, it does result in the formation of free

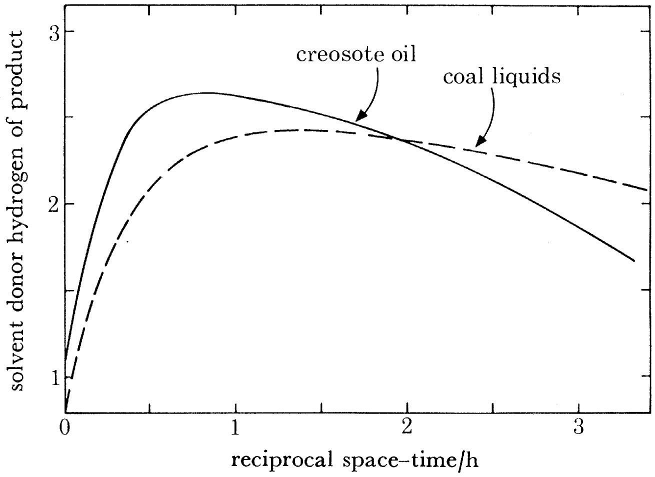


Figure 11. Effect of space–time in hydrotreater on solvent donor hydrogen content. Space–time is the mass of feed per hour per mass of catalyst in the reactor, and the values have been divided by a reference value.

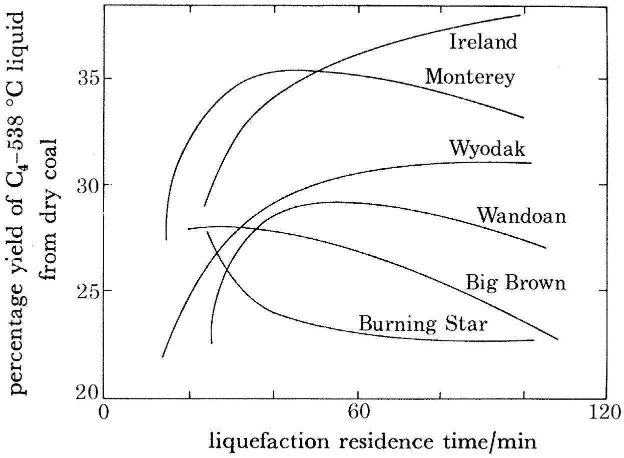


Figure 12. Liquid yield responses for different coals treated at 10.3 MPa and 448 °C (r.c.l.u. pilot plant data).

radicals that must be stabilized by donor hydrogen. It is the ability to tailor the solvent quality to the coal being liquefied that allows EDS to process a broad range of coal ranks optimally.

The solvent is fractionated (distilled) from the total stream emerging from the liquefaction reactor. Control of hydrogen donor characteristics of the solvent is exerted principally by the residence time (actually space–time) of the solvent in the hydrotreater. The effect of space–time on donatable hydrogen is shown diagrammatically in figure 11. Note that after having reached a maximum value, the donatable hydrogen actually decreases with increasing severity of treatment. Increasing residence time produces a solvent with higher hydrogen content; however, at some point the added hydrogen serves to saturate completely some molecular species. As indicated earlier, saturated cyclical molecular species (e.g. decalin) are poor hydrogen donors. It could be expected, in addition, that saturated rings would tend to crack in the subsequent liquefaction step leading to degradation of the solvent.

In figures 8 and 9 the effect of residence time on the yield slates of the Monterey and Wyodak coals was depicted. A total of 24 other coals have been studied in various degrees of detail.

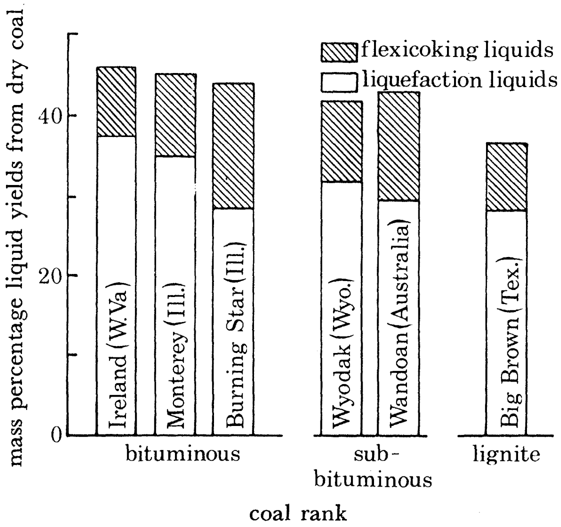


FIGURE 13. Comparability of preferred liquefaction and coking liquid yields in EDS process for most coals tested.

Table 4 lists some of the properties of those studied most intensively. Each coal studied has its own unique response in the EDS process, as shown in figure 12. In general, there is an optimum residence time for the production of liquids, beyond which time the liquid yield falls off. This progressive decrease in liquid yield is due primarily to increased production of hydrocarbon gases as the liquid products decompose into alkylated species, which then proceed to dealkylate. However, in several instances, loss of liquids may be attributable to so-called repolymerization reactions.

Although the responses to liquefaction residence time are complex and still not reconciled completely to coal properties, maximum liquid yields under preferred conditions of liquefaction followed by coking of bottoms are remarkably similar except for the Big Brown lignite (see figure 13). We are presently attempting to integrate available data into a comprehensive correlation system that relates EDS response to coal properties. Only partial success has, however, been achieved. Considerably more work is needed to understand the mechanistic influences of coal properties on EDS responses.

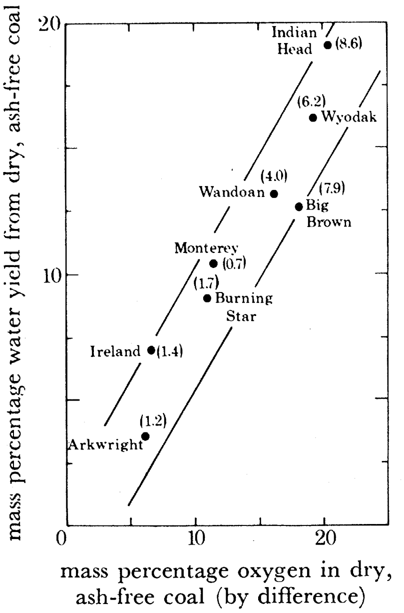


FIGURE 14. Effect of oxygen in coal on water and carbon oxide yields (carbon oxide yields shown in parentheses). Conditions: 448 °C, 10.3 MPa.

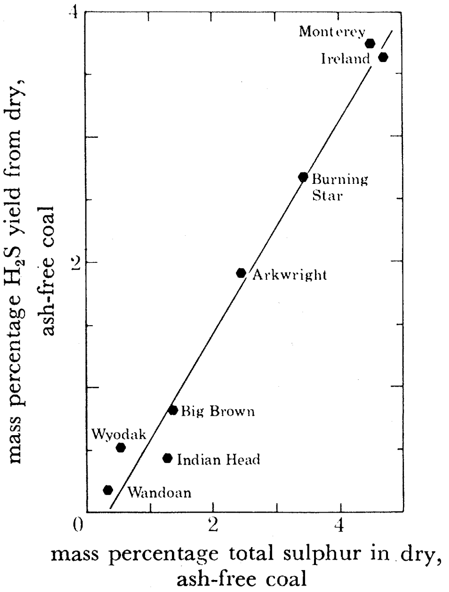


FIGURE 15. Effect of sulphur in coal on hydrogen sulphide yields. Conditions as for figure 14.

We do know that yields of water and carbon oxide gases are closely correlative with the organic oxygen content of the coal (see figure 14), and that H_2S yield is strongly correlative with the total sulphur content of the coal (see figure 15). Coal oxygen content is rank-related, whereas sulphur content is not related to coal rank. Thus, these responses to coal composition are due to entirely different sources of coal property variations.

OPERABILITY

The EDS process has been operated for many thousands of hours in integrated, continuous units at a scale that implies that commercial operability is readily achievable. Only two potential operating problems have been identified, and solutions to both have been developed.

One of the requirements for smooth operation is that the vacuum fractionator bottoms (namely, the non-distillable product) must have a low enough viscosity (while still hot) to be pumped from the fractionator. Bottoms viscosity for all ranks of coal is readily controlled by several means, among which, as shown in figure 16, is adjustment of liquefaction reactor process conditions.

Another potential operability problem inherent to low-rank coals results from the formation of $CaCO_3$ in the liquefaction reactor. Many lignitic and sub-bituminous coals contain up to 3% (by mass) of calcium distributed throughout the coal as cations exchanged to acidic (principally carboxylic) functional groups. As the coal is liquefied, these oxygenated functional groups decompose, freeing calcium oxide (CaO) which combines with CO_2 in the system to form $CaCO_3$. The $CaCO_3$ forms a deposit on the reactor wall surface, and also forms spheres referred to as oolites; these are commonly caviare-sized but may grow to sufficient size that they accumulate in the bottoms of the upflow reactor and in downstream equipment. Numerous potential solutions to this problem have been tested, and several promising techniques have been discovered. The currently favoured solution is a simple solids withdrawal procedure. A novel fallback procedure is to pretreat the moist feed coal with gaseous sulphur dioxide, which forms sulphurous acid in the coal pores. In turn, the acid exchanges a proton for the Ca^{2+} ion, and forms stable calcium salts such as calcium sulphate, which pass through the reactor without agglomerating into oolites or scale. It is worth noting in this connection that any calcite ($CaCO_3$) indigenous to the feed coal is stable in the system and does not contribute to the formation of deposits.

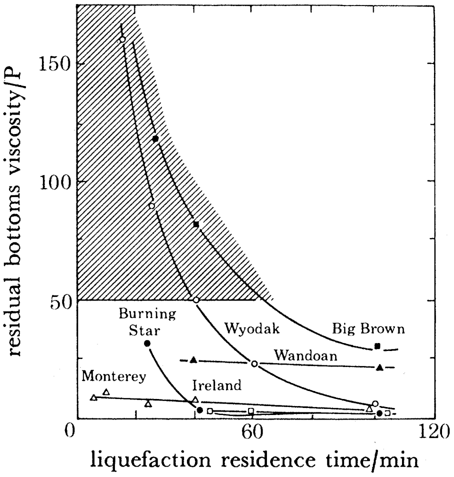


FIGURE 16. Control of residual liquefaction bottoms viscosity at 287 °C ($10\ s^{-1}$ shear rate) for different rank coals. The shaded area is the region of operability problems due to high viscosity. Conditions as for figure 14.

PROCESS IMPROVEMENTS

Two variations on the basic process that result in increased yields have been discovered. It is known that for a given carbon content, the higher the sulphur content of the coal, the higher the conversion. The precise role of the sulphur is still not understood, but two hypotheses have been proposed. Some organic sulphur may exist as thermally unstable thioethers that bridge stable molecular moieties. These could serve as weak links in the molecular structure, leading to enhanced decomposition. Alternatively, H_2S, whose partial pressure in the system is correlated with sulphur content of the coal, may act as a hydrogen transfer agent. Its presence could thereby increase the effective donor hydrogen concentration in the reactor. Based on this supposition, we have found that the addition of H_2S to the reactor when coals with low sulphur content are being liquefied increases their conversion significantly (see table 5).

A recent innovation is referred to as bottoms recycle, a process in which the non-distillable (> 538 °C) fraction of the product is recycled to the liquefaction reactor with fresh coal and solvent. As shown in table 6, significant conversion of the bottoms occurs by recycling, probably because of additional residence time in contact with fresh donor solvent. Numerous tests to define the preferred conditions for the recycle mode are being carried out, and this is an area that continues to receive considerable attention. Not only is conversion increased, but, as shown in table 6, there is considerable enhancement of the conversion to naphtha range material (C_4–204 °C boiling range, a desirable motor gasoline feedstock).

BOTTOMS PROCESSING

No matter how the liquefaction system is optimized, there is still a residual carbonaceous material (bottoms) that can not be recovered overhead in the fractionator. In order to maximize the efficiency of the process, this bottoms fraction must be utilized. The most favourable options for the utilization of the bottoms fraction involve its conversion to both fuel gas and some other product. If the proprietary Exxon Flexicoker system is employed, additional liquid products are recovered. If a more straightforward gasification system is used, the products are fuel gas and hydrogen. Although present plans are to include the Flexicoker option in any commercial plant, this decision is subject to revision.

TABLE 5. INCREASE OF LIQUID YIELD FROM LOW-SULPHUR COAL BY ADDED H_2S

(All percentages by mass.)

	Illinois (4.2% S)		Wyoming coal (0.5% S)	
H_2S added (% on coal)	0	4	0	4
yields (% on coal)				
C_1–C_3 gas	4.1	4.3	4.1	4.2
C_4–538 °C liquid	34	35	31	37
hydrogen consumed	2.5	2.5	2.5	2.7

TABLE 6. INCREASED LIQUID YIELDS THROUGH EDS BOTTOMS RECYCLING

(All percentages by mass.)

	Illinois (Monterey)		Pittsburgh (Ireland)		Wyoming (Wyodak)	
	only coal	bottoms recycling	only coal	bottoms recycling	only coal	bottoms recycling
liquefaction conversion†	56	75	60	75	56	75
C_4–538 °C†	35	44	38	48	32	45
C_4–204 °C liquids‡	66	80	69	88	82	78

† As a percentage of dry coal.
‡ As a percentage of C_4–538 °C liquids.

PRODUCTS

Products from the EDS Liquefaction process are summarized in table 7. The proportions of the total product in each category and their precise properties are dependent upon both the processing configuration and the feedstock coal. The C_4–177 °C naphtha is an excellent reformer feedstock from which a motor gasoline blendstock material with 105 research octane number is obtained in an 85% volume yield. Alternatively, a yield of about 50% of benzene, toluene and xylenes can be obtained by standard petroleum separation processes.

Saturates from the 177–204 °C distillate can be recovered by extraction, and these meet all specifications for jetfuel A. With no treatment, the 177–204 °C distillate has been found by Westinghouse Corporation to be an acceptable turbine fuel. The 204–371 °C cut meets all specifications for home fuel oil except A.P.I. gravity. Alternatively, it is a good fuel oil blendstock. The 399–538 °C vacuum gas oil is roughly equivalent to a low-sulphur fuel oil, except that nitrogen tends to be higher than in comparable petroleum-derived products. It is not directly compatible with pétroleum stocks. The coker gas oil and scrubber liquids are similar to the vacuum gas oil.

All product liquids are amenable to upgrading by using conventional petroleum processing techniques. Such upgrading is consumptive of hydrogen, and whether or to what degree it is practised depends upon relative processing costs and product values.

SUMMARY

During the 15 years of research and development that led to the present Exxon Donor Solvent liquefaction process, many optional process configurations for direct coal liquefaction have been investigated. The present process represents the features that have been proved to be technologically viable and most economically attractive. The developmental work has defined the preferred process, rather than a processing conception having defined the direction of the development. The characteristics of the present process are that it maximizes the use of conventional technology; it is at an advanced stage of development; it is flexible enough to process a wide range of coal ranks; and it can be tailored to produce a variety of products.

The process is still being subjected to an intensive programme of development; modifications, such as bottoms recycling, are incorporated as they are proved feasible (and economic). Additional developments and refinements can be expected as the programme proceeds toward commercial development.

TABLE 7. EDS PRODUCTS AND THEIR DISPOSITION

stream	disposition	yield (percentage of liquids)
C_4–177 °C naphtha	motor gasoline chemicals	40–80
177–204 °C distillate 204–399 °C solvent	jet fuel turbine fuel home heating oil fuel oil blendstock	10–30
399–538 °C vacuum gas oil	fuel oil blendstock	10–30
coker gas oil scrubber liquids	fuel oil blendstock	

Many persons have contributed to the research programme through the years. To name all of them is virtually impossible. I am especially indebted to the following EDS personnel who assisted me in the preparation of this paper: R. F. Bauman, D. F. Ryan, J. W. Taunton, K. L. Trachte, L. W. Vernon, R. D. Williams and S. Zaczepinski.

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Discussion

C. F. KNIGHTS (*Harwell Research, Didcot, Oxon., U.K.*). Does Dr Neavel expect problems in the scale-up of the pressure vessel used in the liquefaction step?

R. C. NEAVEL. If by 'pressure vessel' Dr Knights is referring to the reactor in which the liquefaction takes place, we do not expect any scale-up problems. In the EDS process, this reactor is simply a 'holding tank' with no internal contrivances. Its design and construction are well within present technological capabilities.

H. SCHULZ (*Engler-Bunte-Institut, Universität Karlsruhe, Germany*). Dr Neavel has described the fundamental chemistry of the EDS process as a thermal cleavage of, for example, carbon–carbon bonds and a successive saturation of the radical fragments with hydrogen from the solvent. In a catalytic process, however, one will have many steps of desulphurization accompanying unselective thermal steps, which both contribute to molecular mass reduction. Could Dr Neavel please give some data that show the degree of, for example, desulphurization when liquefying coals with hydrogen transferring solvents?

R. C. NEAVEL. Because mid-distillate range product (204–399 °C b.p.) passes through the solvent hydrotreater in the EDS process, its contents of sulphur and nitrogen are quite low. Naphtha (C_4–204 °C) and vacuum gas oil (371–538 °C) which do not pass through the hydrotreater, contain somewhat more S and N than the mid-distillate. Typical results (percentages by mass) from liquefaction of a high and a low sulphur coal are shown in table 8.

TABLE 8

	coal		naphtha		mid-distillate		vacuum gas oil	
coal name	S	N	S	N	S	N	S	N
Monterey	4.1	1.2	0.58	0.17	0.004	0.04	1.0	1.4
Wyodak	0.5	1.1	0.06	0.16	0.005	0.08	0.05	0.7