# Recent Progress in the Direct Liquefaction of Coal

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**Interest in direct coal liquefaction steadily decreased during the 1980s as the price of crude oil dropped; there is now only one integrated coal liquefaction pilot plant active full time in the United States. The economics derived early in the decade established the price of transportation fuels from coal at $80 per barrel or higher. However, there have been dramatic improvements in the technology since 1983 that have not been widely appreciated. Recent designs and cost estimates show that a 60 percent decrease in the cost of liquid fuels from coal to an equivalent of $35 per barrel for crude oil. Although this cost is not low enough to justify immediate commercialization, additional improvements have been identified that could make direct liquefaction an attractive way to produce gasoline and other conventional fuels.**

IT SEEMS LIKELY THAT THE WORLD WILL CONTINUE TO RELY on liquid hydrocarbon fuels for transportation. The largest source by far for such fuels today is crude oil, and the United States has found and used up the cheap domestic oil it had. There are many alternative sources of liquid fuels (*1*), but all are likely to be more expensive. Relative economics will determine which sources are used and in what order, subject to political limitations. As long as oil can be obtained outside the United States for less than $20 per barrel, imports will replace domestic production. However, when consumption begins to approach production capacity worldwide, the OPEC cartel is likely to increase the price. A wide variety of options will become attractive; one of these could be conversion of coal to liquids. Coal liquefaction would be an attractive option for the United States because of its vast coal reserves.

Coal consists of complex macromolecules without repeating monomer units that are built primarily of carbon and hydrogen but also contain significant amounts of oxygen, sulfur, and nitrogen. The constituent units tend to be mostly substituted aromatics or hydroaromatics, and the degree of condensation increases as the coal matures. Because coal has a hydrogen-to-carbon ratio significantly lower than that for petroleum, converting coal into liquid transportation fuels requires either the addition of substantial amounts of hydrogen or the removal of excess carbon. Many methods of liquefaction have been proposed, tested, and even commercialized. One of the oldest commercial processes is direct liquefaction, discovered by Bergius in Germany in the 1920s (*2*).

Direct liquefaction involves addition of hydrogen to coal in a solvent slurry at elevated temperature and pressure. The solvent provides a convenient transportation medium for the coal and enhances heat and mass transfer during chemical reaction. In many processes, the solvent shuttles hydrogen from the gas phase to the coal and is called a donor solvent. The elevated temperature cracks the coal molecules by thermally rupturing carbon-to-carbon linkages and increases the rate of the chemical reactions; high pressure keeps the solvent and products in the liquid phase at these temperatures and promotes hydrogenation by maintaining a high partial pressure of hydrogen. Catalysts are normally used to increase the rates of the desirable reactions.

I. G. Farben developed the Bergius process in Germany in the 1930s (*2*), and built 12 plants that provided liquid fuels during World War II. The English built a massive Farben plant at Billingham, run by ICI (*3*). The U.S. Bureau of Mines tested the German technology after World War II in a 200 to 300 barrels per day pilot plant (*4*). All of these efforts were technically successful, but could not compete economically with inexpensive petroleum that became available from the Middle East in the early 1950s.

## Second Generation Technology

Despite attempts to improve liquefaction technology during the 1950s and 1960s, interest in direct liquefaction remained modest until the Arab oil embargo of 1973. High oil prices and availability of federal funding for such research led to a resurgence of interest. Many processes were tested in small pilot plants; three processes were tested at large scale in the late 1970s and early 1980s: SRC-II (Solvent Refined Coal) in Tacoma, Washington; EDS (Exxon Donor Solvent) in Baytown, Texas; and H-Coal in Catlettsburg, Kentucky. These programs successfully demonstrated new technologies and laid the groundwork for future developments, but were unable to make coal liquefaction economically attractive.

The H-Coal process was developed by Hydrocarbon Research, Inc. (HRI), and was derived from their H-Oil process. The heart of both was the ebullated catalyst-bed reactor, a liquid-phase version of the familiar gas-fluidized bed processes widely used in the petroleum industry. Coal was slurried in product liquid and fed into the reactor, where it was dissolved and upgraded in a single step. The ebullated bed made it possible to use a conventional solid hydrotreating catalyst with a slurry. Hydrocyclones removed the mineral matter and undissolved coal and then the liquids were distilled. Some of the product was recycled as process solvent. Results at small scale were encouraging. The U.S. Department of Energy (DOE) and a consortium of industrial sponsors built a 250 tons per day pilot plant, or semiworks, at Catlettsburg, Kentucky. This plant ran from May 1979 through January 1983.

The H-Coal Project generally was considered to be a technical

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**Fig. 1.** A block flow diagram of the Wilsonville, Alabama, Advance Coal liquefaction R&D facility.

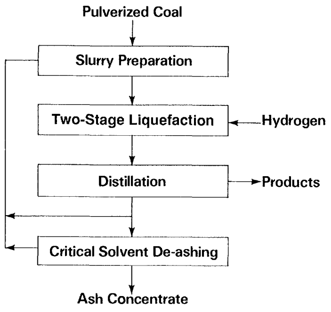

**Table 1.** Evolution of direct coal liquefaction technology. Unless otherwise indicated, units are weight percent of moisture- and ash-free coal feed.

| Plant characteristics | I. G. Farben 1935 (*2*) | Billingham 1945 (*3*) | H-Coal 1980 (*10*) | Wilsonville 1986 |
|---|---|---|---|---|
| Pressure (MPa) | 69 | 21 | 21 | 19 |
| Maximum temperature (°C) | 480 | 465 | 455 | 440 |
| Coal conversion | | 94 | 94 | 94 |
| Hydrogen consumption | 14 | 8 | 6 | 5.6 |
| Yields | | | | |
| Hydrocarbon gases | 30 | 25 | 11 | 7 |
| Distillable liquids | 54 | 54 | 51 | 65 |

success. The semiworks confirmed the yields predicted by smaller scale experiments, and a wide variety of mechanical equipment was tested and developed. Had oil prices continued to rise, a commercial project could have been the next step. However, oil prices fell slightly, and the consortium disbanded shortly after the pilot plant shut down.

Although H-Coal economics were not attractive, two developments promised reduced costs. First, a new catalyst with a bimodal pore size distribution had been developed that improved distillable liquid yields, particularly with cheaper, low-rank coals. Second, hydrogen consumption was lowered by using two reactors in series (hydrogen is one of the largest costs for converting coal into liquid fuels).

The results from the SRC-II and EDS pilot plants made these processes less likely candidates for further development. Both processes depended on a donor solvent for hydrogen during liquefaction with only the minerals in the feed coal for catalyst. (EDS did use catalyst to hydrotreat part of the recycle solvent in a second reactor, but did not use catalyst in the main liquefaction reactor.) Liquid yields were relatively low and the economics were unattractive. Two stages, at least one containing an ebullated bed of catalyst, appeared to be the most logical next development to test.

## Wilsonville

In 1972, Southern Company constructed a pilot plant in Wilsonville, Alabama, to test a process for the production of a clean boiler fuel from coal (*5*). When the Electric Power Research Institute (EPRI) was formed in 1973, it assumed a major technical and financial role in the project. The federal government became a sponsor in 1976, as did Amoco in 1983.

The original process (SRC-I) was a single-stage dissolution of coal followed by filtration and solvent recovery. The product was a solid, high-quality fuel for boilers. The facility gradually evolved into a more versatile liquefaction plant (Fig. 1), currently configured with two closely coupled ebullated bed reactors in series and a ROSE-SR (Critical Solvent De-ashing) unit to remove the minerals and unreacted coal from the liquid products. Nominal capacity at Wilsonville is 6 tons of coal per day.

Other programs have also made major contributions to recent technical progress. HRI devotes part time on their bench unit to coal liquefaction, and their participation in the development of this technology continues to be important. The pilot plant programs run by Lummus, Chevron, and Kerr-McGee advanced the technology before they shut down. Many laboratory programs, frequently DOE or EPRI sponsored, have helped develop basic chemistry and physics. However, Wilsonville has become a focus for U.S. technology development, as it is the only place to test new concepts in an integrated plant.

## Current Development Efforts Outside the United States

There is substantial coal liquefaction development in Japan, Germany, and Great Britain. Most of the funding is from the governments in these countries through cooperative programs with private industry. None of these projects has contributed significantly to current technology in the United States, partially because other governments providing the funds allow industrial participants to keep technology proprietary to a greater degree than is possible here.

The Japanese have the most ambitious program, supported through the New Energy Development Organization (NEDO), which derives its funds from a tax on imported petroleum. Low oil prices and a weakening dollar have reduced NEDO's income and delayed several projects, but they remain committed for the long term. The processes that have been tested in small integrated pilot plants and proposed for semiworks evaluation all have a thermal first stage, sometimes with an iron slurry catalyst. This is followed by solids removal and a catalytic second stage for upgrading the liquids (*6*).

The Germans had an active program until recently, with most processes similar to the original Farben process. All of these plants used a single stage with a pressure of about 31 MPa and iron-based slurry catalyst (*7*). The 200 tons per day Bottrop plant is still in operation, but is now being used to refine heavy petroleum liquids. The Pyrosol process is slightly different; it uses a milder first stage, followed by coking of the product liquids under a hydrogen atmosphere (*8*).

The British will complete a 2.5 tons per day pilot plant this year at Point of Ayr in Wales (*9*). The process uses a donor solvent under mild conditions to dissolve the coal, but no gaseous hydrogen. Ash and undissolved coal are then filtered out, and the liquid is hydrotreated conventionally. Some yield is sacrificed to the mild treatment in the first stage, but the sponsors believe that lower capital costs will compensate for the lower yield.

There are major drawbacks to the processes being developed overseas for use in the United States. First, any process must fit the feedstock available. The adjustments necessary to tailor a foreign process to U.S. coals would require a major development effort in this country, even after it is in commercial use elsewhere. Second, processes similar to those cited have been tested before in the United States and have been rejected for one reason or another.

## Process Evolution

The long-term development of direct coal liquefaction technology (Table 1) (*10*) lends perspective to the more recent advances in that technology. The data are typical for the time periods shown, rather than averages or the best. They demonstrate the broad directions the technology has taken over the years. All data are for bituminous coals. The trend is toward milder operating conditions, that is, lower pressure and temperature. The conversion of similar coals has stayed constant. However, hydrogen consumption fell because less hydrocarbon gases were produced at the increasingly mild operating conditions. Distillable liquid yield was roughly constant until recently and then began to increase. This increase in yield without major penalties in hydrogen usage or the need for more severe reaction conditions is the key to the improvement in liquefaction economics.

Figure 2 illustrates the progress in the quantity and quality of liquid products since 1983. The starting point is the H-Coal product distribution. By 1985, Wilsonville had reduced hydrogen consumption by cutting the production of gas in half, and also cracked some of the heavy, nondistillable liquid (resid) down to gas oil. The following year, all of the resid was eliminated by recycling it back through the reactors until it broke down into lighter products. This raised the distillable liquid yield to 70% of the moisture- and ash-free (MAF) coal. HRI has been able to recycle the gas oil to extinction as well (*11*), and Wilsonville is expected to confirm this improvement soon.

There is room to improve the yields further. Given the elemental composition of Illinois #6 coal, if (i) all of the oxygen in the coal was rejected as carbon dioxide, (ii) all of the sulfur and nitrogen were removed as hydrogen sulfide and ammonia, and (iii) enough hydrogen was added to bring the hydrogen to carbon ratio up to that in gasoline, then the yield of liquid fuel would theoretically be increased by about 25%. Although this ideal yield is unlikely to be achieved, it seems certain that significant increases toward this goal are possible with further research.

It is also important to the economics that hydrogen consumption has remained modest as yields increased. In the 1986 Wilsonville run (Fig. 2) hydrogen consumption was ~6.8% by weight of MAF coal; 20% of the hydrogen was consumed in removing heteroatoms (sulfur, nitrogen, and oxygen), 10% went into hydrocarbon gases, and fully 70% went into the liquid products. In most processes developed before 1983, only about half of the added hydrogen went into distillable liquid products (*12*).

These results are due to several improvements in the technology. Two reaction stages are now used, in contrast to the earlier trend of trying to reduce capital cost by using a single stage. The gain in control over the chemical reactions more than balances the cost for the second stage. The two reactors need to be closely coupled, again to control the chemistry. There is a greater appreciation of the importance of retrograde, or polymerization, reactions. If the free radicals formed when coal breaks down are not quickly capped with hydrogen, they react with each other to form large molecules that are much harder to break down than the original coal (*13*).

For the same reason, catalyst is used in both stages. Earlier work used a thermal first stage, followed by a catalytic second stage, because coal degradation is a thermal process, whereas hydrogenation is catalytic. However, the high reactivity of coal is now recognized, as is the need to cap radicals quickly as they form; catalyst can speed the capping reactions. Catalyst also promotes hydrocracking of coal resid to distillable liquid (*14*).

Wilsonville has shown that the ROSE-SR process improves the liquid yield by minimizing the amount of liquid rejected with the solids. This process is also more reliable and easier to operate than physical separation methods used previously.

Other changes that have no direct impact on yields have also contributed to lower costs for the liquefaction process. For example, Wilsonville has proven a low-cost water treatment process that makes the discharge of excess water environmentally benign, and a pressure let-down valve that allows substantial simplification and cost savings over the H-Coal technology.

## Cost Implications of Improvements

An important product of the large-scale pilot plants sponsored by the federal government in the 1970s and 1980s was a set of highly detailed commercial designs and cost estimates. One of these was derived by Bechtel, who designed and estimated the cost of an H-Coal plant called the Breckinridge Project (*15*). The results are in the public domain.

The Breckinridge design was very conservative, particularly in assuming extremely low construction labor productivities. The design was appropriate to the needs of the times; cost estimates for alternate fuels projects had proven to be optimistic time after time, and the sponsors felt it was critical that the costs not be underestimated. It was also assumed that the synfuels industry would be booming and cause a shortage of experienced, competent labor.

Amoco hired Bechtel in 1986 to update and revise the Breckinridge Project economics to reflect current technology and costs. The number of trains, unit capacities, and process flows within the individual units were changed, and the plant was relocated from Kentucky to the Texas Gulf Coast. Reductions occurred in major equipment and field labor costs, but bulk material costs increased. The major changes to the plant configuration included: (i) two-stage liquefaction in place of single-stage, (ii) a greatly simplified product distillation system, (iii) the ROSE-SR process for solids removal, (iv) hydrotreating-hydrocracking based on Chevron data (*16*), and (v) addition of catalytic reforming.

The basis for this design is the two-stage liquefaction data reported by HRI (Fig. 2) in which liquids boiling above 350°C are recycled to extinction. This product slate is an advantage when liquid transportation fuel is the goal; a different choice might be appropriate for producing a clean boiler or turbine fuel for a utility. The liquids boiling above 110°C are hydrocracked, and gasoline is the major product.

The resulting plant (Fig. 3) handles the same amount of MAF coal feed as the original Breckinridge plant, 15,140 tons per day. But where the Breckinridge design produced 52,350 barrels per day of liquids suitable as refinery feed, the new design generates 67,600 barrels per day of regular gasoline and 17,300 barrels per day of propane and butane. This is equivalent to 5½ barrels of liquid product for each ton of coal fed to the plant, 4½ barrels of it gasoline. The only by-products are minor amounts of phenol, ammonia, and sulfur.

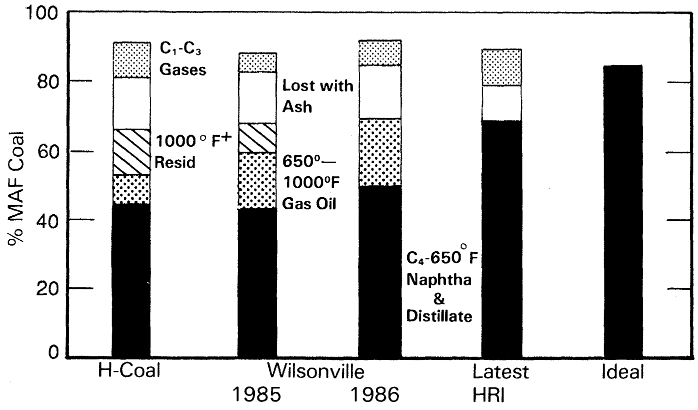

**Fig. 2.** Coal liquefaction yields and product distributions.

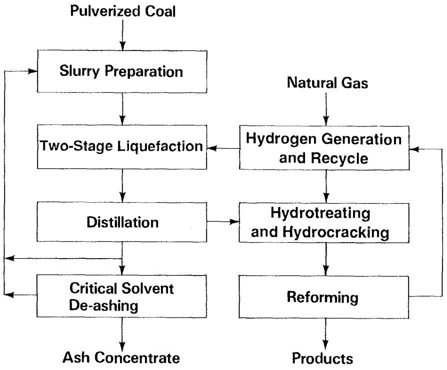

**Fig. 3.** Major units in a grass-roots commercial coal liquefaction facility.

The plant also requires a large amount of natural gas to make hydrogen, $\sim$6.4 million cubic meters per day. An alternative is to gasify the ash-reject stream and some additional coal to provide the neceesary hydrogen. This design option requires higher initial capital, but produces about the same rate of return.

The economics for the new design are quite encouraging. By assuming 100% equity financing, constant 1987 dollars, and that the ratios of the feed and product fuel costs to that of crude oil remain at current levels, the plant would produce a 10% real rate of return (after taxes and without inflation) if crude oil price jumped to $35 per barrel. On the same basis, the original Breckinridge design and cost estimate would require $93 per barrel, and a Breckinridge design with updated labor rates would require $87 per barrel to justify. So improvements in design and technology have lowered the cost for direct coal liquefaction by about 60% since the H-Coal pilot plant shutdown in 1983.

The cost is too high to make construction of a commercial facility attractive today, but the economics will probably improve as the price of crude rises. Based on the National Petroleum Council's high-crude-price scenario (*17*), a 15% escalated rate of return would be possible for a plant starting up in the year 2000; for the low-price scenario, one would need to wait until 2015. Fortunately, substantial cost reductions are probable through continued research, and earlier commercial use may be attractive.

## Future Programs

The sponsors at Wilsonville are developing a technical plan for the next several years with a target to reduce costs below $30 per barrel. The possible improvements slated for study are evolutionary rather than revolutionary; the process has evolved so rapidly since 1983 that there has been little effort spent on optimization.

Perhaps the most promising single method for reducing costs is to find a suitable cheaper coal or, alternately, a not-so-cheap coal that will give particularly high liquid yields. Development efforts at Wilsonville have concentrated on two coals, Illinois #6 bituminous and Wyodak subbituminous. The economics for the two are nearly the same; Wyodak is cheaper feedstock, but the yields are lower and the plant would be more expensive to build. This year, there are plans to test a high-carbon Pittsburgh seam bituminous and a Texas lignite. The former is frequently cited as giving very high liquefaction yields, whereas the latter is a moderate-cost coal that has given reasonably high yields in other processes (*18*).

Another important concern is to demonstrate that the high-boiling liquids can be recycled to extinction. Wilsonville yields that include gas oil, but that are otherwise similar to the HRI yields, add $2 per barrel to the needed crude oil price. The demonstration will also show if the high gas yields reported by HRI also hold at Wilsonville; in the past, less gas has been produced under similar conditions in the larger plant. Reduced gas production can lower costs by as much as $2 per barrel.

Many other modifications to equipment or operating conditions can cut the costs by lesser amounts. Enough tests have been laid out to keep the Wilsonville plant active and productive for the next several years. The goal of a 15 to 20% cost reduction can likely be achieved without a breakthrough in technology.

Beyond these evolutionary improvements, there seems to be an increasing potential for breakthroughs in the technology for direct liquefaction of coal. Progress in understanding fundamental coal structure and chemistry has been accumulating, spurred by government funding in Japan, Germany, and the United States.

Several processes that use somewhat different chemistry are already under development. One type uses carbon monoxide (or, alternatively, synthesis gas) and water in place of hydrogen to liquefy low-rank coals, and is purported to offer both higher yields and faster reaction kinetics compared to hydrogen (*19*).

An even more attractive way to transfer hydrogen to coal would be directly from methane. Methane chemistry is receiving much attention today by petroleum companies, as evidenced by the growing number of papers being presented on such research (*20*). A major emphasis in this research is converting methane to higher molecular weight hydrocarbons without reducing it first to synthesis gas. Such technology might be applied to induce methane to react directly with coal.

Different kinds of catalysts also offer some interesting possibilities. Although iron is a mediocre hydrogenation catalyst, it seems to inhibit retrograde reactions, particularly with low-rank coals. Supported iron catalysts might offer advantages. Slurry catalysts other than iron oxide or sulfide might also be useful. Veba Oel uses a lignite char in the hydroprocessing of petroleum resid, although they consider it an additive rather than a catalyst (*21*). Dow developed a process that used a colloidal suspension of molybdenum as a catalyst (*22*), and Exxon has a petroleum-resid upgrading process called M-Coke that is also based on micrometer-sized molybdenum catalyst (*23*).

Several techniques are available for gently dismantling the coal structure with a minimum of damage and under mild conditions. Although such methods are used only to study the structure of coals, and are generally unsuitable for commercial processes, they do suggest that coal can be broken down easily by careful control of the chemistry. Examples include the use of potassium crown ethers (*24*), reductive alkylation with sodium hydroxide and ethanol (*25*), and transalkylation techniques that use superacids (*26*). Biological degradation of coal has begun to receive serious consideration, and has had a surprising degree of success in a short time (*27*). For example, enzymes from *Polyporus versicolor* that can solubilize low-rank coals have been isolated and characterized.

## Outlook

The development of new technology to reduce the cost of direct liquefaction of coal has been successful since 1983. Costs have been reduced by 60%, and improvements likely to be tested in the next several years can lower the costs another 20%. Opportunities for new technologies that offer low-cost routes to making transportation fuels from coal seem better than ever.

One school of thought in the United States says that it makes sense to put current synfuels technology aside and wait until oil prices rise to make the technology economically attractive. This course of action has not worked in the past and is no more promising for the future. But recent history has proven that direct coal liquefaction can be improved, and costs lowered significantly through a steady research effort.

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# Cooling, Stopping, and Trapping Atoms

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**Significant advances have been made in the ability to control the motion of neutral atoms. Cooling and trapping atoms present new possibilities for studies of ultracold atoms and atomic interactions. The techniques of laser cooling and deceleration of atomic beams, magnetic and laser trapping of neutral atoms, and a number of recent advances in the use of radiative forces to manipulate atoms are reviewed.**

THIS ARTICLE REVIEWS THE USE OF ELECTROMAGNETIC forces, particularly radiative forces, to influence atomic motion. Earlier reviews (*1*) have given good accounts of both experimental and theoretical work in this field. We concentrate on advances in the manipulation of neutral atoms, especially laser cooling of atomic beams and electromagnetic trapping of atoms.

Thermal motion of atoms is the bane of many measurements in atomic physics. The precision of ultrahigh-resolution atomic spectroscopy is invariably limited by the motion of the atoms being observed. Doppler effects both shift and spread the frequencies associated with transitions between atomic energy levels. A host of techniques that are based on the use of nonlinear laser spectroscopy, as well as spectroscopy with laser beams perpendicular to a well-collimated atomic beam, have provided the means of observing optical spectra that are nearly free of the first-order Doppler effect. Unfortunately, the second-order Doppler effect, associated with the relativistic time dilation, is unaffected and continues to plague the spectroscopist. The finite observation times available when one looks at rapidly moving atoms also limit the precision with which measurements can be made. As a result, the best spectroscopic measurements, from radio to optical frequencies, are limited by motional effects (*2, 3*).

Detailed studies of collision phenomena, which require precise knowledge of the initial velocities of the collision partners, are similarly hampered by the randomness of thermal motion. Neither the direction nor the magnitude of the relative velocity of two colliding atoms is well defined when velocities are distributed thermally. The use of thermal atomic beams, for which the direction of each atom's velocity is well defined, does not completely solve the problem. Velocity selection of an atomic beam leads to a well-defined velocity, but selection is inefficient in that most of the atoms are not used. Velocity compression by means of supersonic expansions can yield high beam flux with relatively well-defined velocity,

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