Oil from coal ~ 3 years on

After 3 years' research, the pyrolysis technique under investigation by CSIRO teams in Sydney and Melbourne as a means of making oil from coal continues to look promising. *Ecos* reported the aims of the project in August 1975, shortly after the work began. This article is a progress report.

Rapid or 'flash' pyrolysis is one of three coal-to-oil approaches being researched around the world with increasing vigour as the threat of a liquid fuel drought looms larger. The others are hydrogenation, the method receiving most research attention in the United States, and the Fischer–Tropsch gas synthesis process used in South Africa's SASOL plant, the world's only commercial coal-to-oil operation.

The flash pyrolysis process, in which coal is decomposed rapidly to tar, gas, and char by heating to about 600°C in the absence of air, is a simple approach. It operates at atmospheric pressure, and no catalyst is required. Hydrogenation is plagued by engineering problems related to the need to process coal at high pressures, and the Fischer–Tropsch approach — if it is used on its own — is very inefficient in terms of liquid fuel production.

But flash pyrolysis also has disadvantages, notably a lower liquid yield per tonne of coal than hydrogenation. Balancing advantages and drawbacks, these two processes seem likely to be stronger contenders than the Fischer–Tropsch technique when and if a decision is taken to build a commercial oil-from-coal plant in Australia.

In early experiments at the Division of Process Technology at North Ryde, Sydney, Mr Ian Smith and his colleagues researching flash pyrolysis used a bench-top rig capable of processing a meagre 1 gram of coal an hour. Their second rig can deal with 100 g of coal in an hour, and a third, inaugurated in August 1976 by the Minister for Science, Senator Webster, is a two-storey-tall plant with an hourly throughput of 20 kg.

The rigs expose pulverized coal to high temperatures for less than a second. The tar produced — a viscous black material at room temperature but a freely flowing liquid at about 60°C — is the product readily converted to oil. So an important aim of the research is to find out what conditions produce the most tar from a given amount of coal.

Using coal — both black and brown — with favourable characteristics for tar production (see *Ecos* 13), the scientists have found that the bench-scale flash pyrolyser regularly converts 20–30% of the coal's mass to tar. This is much better than the yield achieved when coal is pyrolysed in the conventional retorts used for producing metallurgical coke and town gas.

With all coals tested, the temperature that produces the greatest tar yield has turned out to be about 600°C. At lower temperatures, more of the coal remains as a solid, while above 600° the tar formed tends to break down into gases.

One of the purposes of building the 20-kg-per-hour flash pyrolysis rig was to find out whether the yields achieved with the bench-top rig could be matched at this much larger scale. Results to date are encouraging.

Loy Yang brown coal from Victoria

Oil made from coal fuelled German tanks, planes, and submarines during World War II. Here a Focke Wulf fighter is refuelled.

and Millmerran black coal from Queensland have given tar yields very similar to those achieved with the small rig. When this article was written, tests were just beginning with a third coal, Liddell black from New South Wales, and early indications were again favourable.

Also, there is no sign yet of problems that the scientists thought might arise in keeping coal and the pyrolysis products flowing through the rig. Black coal tends to swell and become sticky when it is heated, causing agglomeration problems in some pyrolysis installations.

Oil from the tar

At the Division of Applied Organic Chemistry in Melbourne, Dr Peter Wailes is leading a team working on the upgrading of flash pyrolysis tar to produce synthetic crude oil capable of being fed through existing oil refineries.

The process is hydrogenation, but it is a simpler operation than hydrogenation of coal. This is partly because pyrolysis has already increased the ratio of hydrogen to carbon, one of the requirements for making oil from coal. The extra hydrogen comes from the portion of the coal that ends up as char. Also, the tar is more manageable than coal.

Hydrogenation gives a further lift to the hydrogen: carbon ratio, producing a lighter, more fluid product. It also performs the important function of removing oxygen present in the tar.

Dr Wailes and his colleagues have designed and built two continuous-flow hydrogenation units, the first capable of processing about 30 g of tar per hour and the second about 100 g per hour. Late last year — after the 20-kg-per-hour flash pyrolysis unit came into service — they started receiving enough tar from the Sydney team to enable upgrading tests with it to begin. Earlier, they tested the hydrogenation process mainly with brown coal tar produced as a by-product at a Victorian plant making lump char and activated carbon.

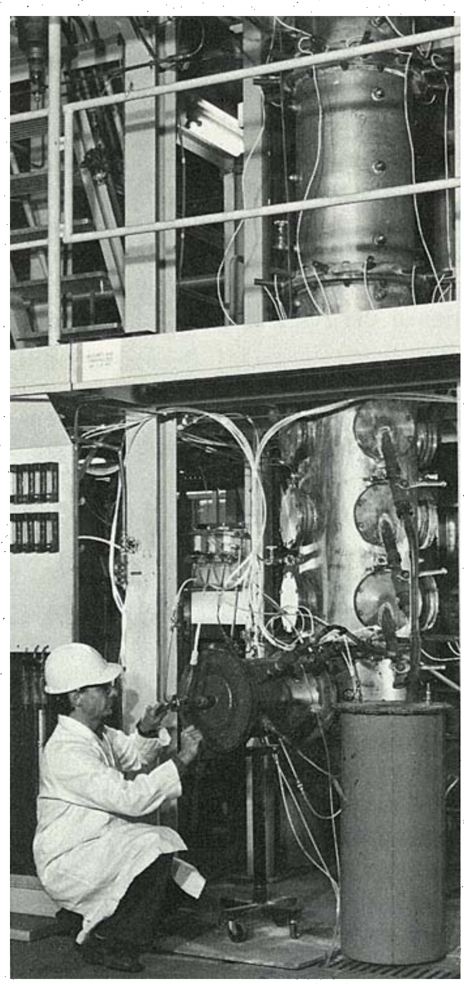
The scientists are attempting to find out what reaction conditions, and what catalysts, produce synthetic crude oil most efficiently from the tar. Hydrogen reacts with tar in the required manner at temperatures around 400°C and pressures in the vicinity of 17 000 kilopascals (about 2500 pounds per square inch). Many metals (including molybdenum, cobalt, tungsten, and nickel) can be used, singly or in combination, as catalysts.

Two other CSIRO groups are involved in the flash pyrolysis study. At North Ryde, members of the Fuel Geoscience Unit have helped select suitable coals for the study, and are now examining the behaviour of different coal types under flash heating conditions. In the Division of Chemical Engineering in Melbourne, theoretical and practical studies of reactor design and scale-up are under way.

Attached to a 1000-megawatt power station, according to the scientists' calculations, an oil plant could produce between 13 000 and 51 000 barrels of synthetic crude per day.

Using the by-products

Apart from the tar, the products of flash pyrolysis are a char, consisting mainly of carbon, and a variety of gases and vapours. These include the hydrocarbon gases methane, ethane, ethylene, propane, and propylene. Others, grouped together under the heading residual volatiles, include carbon dioxide, carbon monoxide, hydrogen, and water vapour.



This pyrolysis rig, built in 1976 at the Division of Process Technology laboratories, has a throughput of 20 kg per hour.

If flash pyrolysis is adopted as a commercial oil-producing process, these other products will also have to be put to effective use. So will the small proportion of by-products from hydrogenation of the tar.

At the Division of Process Technology, Mr Smith, Mr Jim Edwards, and Mr Ralph Tyler have drawn on data from the pyrolysis and hydrogenation research to produce performance figures for possible oil-producing plants. They assigned uses to all the pyrolysis products. The heat needed to operate the synthetic crude oil plant envisaged would come from burning pyrolysis char.

While the synthetic crude oil produced would have only 31–38% of the energy content of the original coal, the gases and remaining char would also be able to supply a lot of energy. Figures for the energy content of all products as a proportion of the coal's energy are 51% for Loy Yang brown, 72% for Millmerran black, and 75% for Great Northern black coal. The greater water and oxygen contents of the brown coal largely account for its comparatively poor performance.

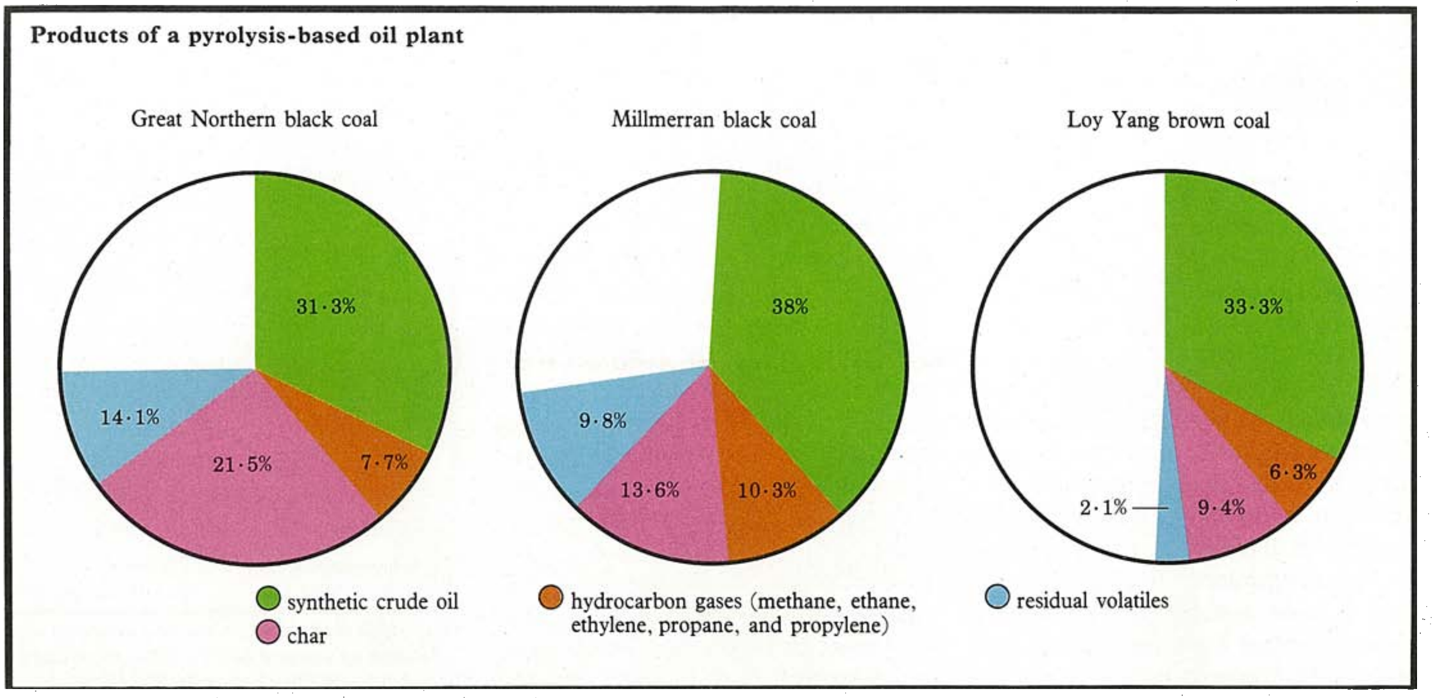
. . . to make electricity

One way to utilize all the pyrolysis products would be to link the oil-from-coal plant with an electricity-generating station. The char and gas would fuel the power station boilers while the tar was converted to oil.

Attached to a 1000-megawatt power station, according to the scientists' calculations, an oil plant could produce between 13 000 and 51 000 barrels of synthetic crude per day. The estimated production rate varies with the type of coal and the power station's 'load factor', which is the proportion of its potential electrical output actually achieved.

The lower oil-production figure, which is equivalent to 2·2% of the crude oil used in Australia in 1974–75, assumes a 50% load factor for a power station burning Great Northern black coal. Loy Yang brown coal and a load factor of 75% give the higher figure, equivalent to 8·5% of that year's oil use.

Brown coal produces more oil than black in this situation. This is because its comparatively low output of surplus char, after some has been used to supply the energy needed for coal drying and pyrolysis, means that much more brown



The charts show the scientists' estimates of the proportions of the coal's energy content that would end up in the products of a flash pyrolysis plant coupled with a tar-hydrogenator.

coal is needed to produce a given amount of electricity.

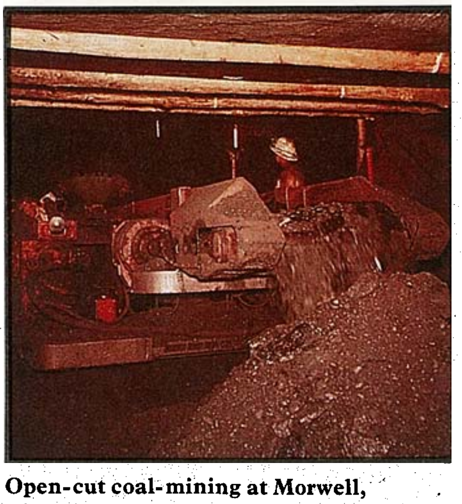
The scientists calculate that the combined oil and electricity plant would use four times as much Loy Yang coal as the power station on its own, compared with only about twice as much Great Northern. More than 100 000 tonnes of Loy Yang coal would have to be fed into the combined plant every day if the power station's capacity was 1000 megawatts and the load factor was 75%. The equivalent figure for Great Northern coal is less than 15 000 tonnes.

. . . or gas, or motor fuel

Another way to utilize the char and gas would be to produce a substitute for natural gas (mainly methane) from them. Technology exists to convert char first into a mixture of carbon monoxide and hydrogen and then, in the presence of a catalyst, to combine the gases to produce the natural-gas substitute. The methane component of the gas produced during pyrolysis would also be available.

According to the calculations, a gas plant attached to a pyrolysis operation producing 15% of Australia's 1974–75 oil consumption could produce 1326 million cubic metres of natural-gas substitute per year if Loy Yang brown coal was used. If the feedstock was Great Northern black coal, the gas output would be 2772 million cubic metres, which is equivalent to 59% of Australia's natural gas consumption in 1974.

The third suggested use for the char and gas is conversion to motor fuels (petrol and diesel) by the Fischer–Tropsch gas-synthesis process used commercially to produce oil from coal in South Africa. Carbon monoxide and hydrogen produced from the char, together with most of the hydrocarbon gases produced during pyrolysis, would be the process ingredients. The remaining gases would provide the necessary heat.



Open-cut coal-mining at Morwell, Victoria
. . . and underground mining at Coalcliff, N.S.W.
Any commercial oil-from-coal plant would require vast amounts of coal.

The scientists calculate that, using Loy Yang brown coal, oil made from the pyrolysis tar together with motor fuel produced from the char and gas would have 39% of the energy content of the original coal. The equivalent figure for Great Northern black coal is 44%, and for Millmerran black coal, 48%. These figures compare most favourably with the 20% efficiency for liquid fuel production that the South Africans expect to achieve with their new SASOL II oil-from-coal plant, which will use black coal similar to Great Northern.

The CSIRO flash pyrolysis team plans by the end of this year to attach a hydrogenating unit to the 20-kg-per-hour pyrolysis rig at North Ryde. The combined plant will produce synthetic crude oil from pulverized coal, and enable the scientists to investigate ways of efficiently integrating all steps in the process. The plant is intended to be self-sustaining, with char from the pyrolysis stage providing all the heat needed to run it.

Results achieved with this integrated plant — yields and running performance — will play a large part in determining whether and how soon the project is further scaled up. The next step would be a pilot plant, probably with a throughput of some 25 tonnes of coal a day. A further step, a 'commercial-demonstration' plant treating coal in quantities approaching those required in a full-scale operation, would probably be required before a commitment was made to build a fully commercial installation. But that is very much in the future.

Hydrogenation

One factor sure to play a big role in determining whether flash pyrolysis becomes a commercial oil-making process is the success of research on the most promising alternative, hydrogenation of coal. Various approaches to this are being tried around the world. The broad aim is to achieve maximum conversion of coal to liquid fuel with minimum hydrogen consumption at the lowest possible pressure.

Methods of producing a variety of fuels are being examined. For example, the aim of some research is to produce a clean boiler fuel from high-sulphur coal, while other experiments are directed towards the production of synthetic crude oil that can then be refined into liquid fuels ranging from gasoline to fuel oil.

All hydrogenation methods involve the transfer, under pressure and with the aid of a slurrying oil, of hydrogen to coal. Some require the assistance of a catalyst. Of the broad approaches, solvent-refined coal processes do not require a catalyst, and donor-solvent-type processes aim at avoiding the need for one. The third approach, the Bergius method pioneered in Germany before World War II, requires a catalyst.

In parallel with its flash pyrolysis studies, CSIRO is examining hydrogenation of Australian coals. Scientists at the Division of Applied Organic Chemistry are looking at the prospects of developing catalysts that are more efficient, durable, and economic than those now available. They are particularly interested in throw-away catalysts such as red mud, a waste product from alumina refineries, which was used extensively by the Germans during World War II.

In the Fuel Geoscience Unit and the Division of Process Technology, the fundamental physical and chemical processes involved in coal hydrogenation, and the effects of coal properties on liquid fuel production, are under examination. As well as increasing knowledge of the processes involved in hydrogenation, these studies should help provide a basis for selecting the best coals for particular processes or the best processes for particular coals.

Research on coal hydrogenation (both catalytic and non-catalytic) is also going on at other centres in Australia, including the Australian Coal Industry Research Laboratory (ACIRL) in Sydney and BHP's Melbourne Research Laboratory.

Last year, the Federal, Queensland, New South Wales, and Victorian governments agreed to finance, with the West German government, a $3 million feasibility study for an oil-from-coal plant in Australia producing 60 000 barrels per day. The technology envisaged by the West German proponents involves preliminary hydrogenation of the coal, gasification of the unconverted coal residues to carbon monoxide and hydrogen, and Fischer–Tropsch synthesis of these gases to liquid fuels.

Clearly there are many ways to make oil from coal, and many refinements are sure to flow from the world-wide research. Working out which method would be most economic with particular Australian coals will be no easy task, and will require all the expertise in coal conversion that is developing as the research proceeds.

Assessing and controlling the environmental impact of any coal-to-oil plant will also be difficult. Huge amounts of coal will have to be mined to supply the plant, and very large quantities of water will be required to keep the process running. Coal ash, dirty water, and waste gases will be some of the pollutants requiring treatment and disposal.

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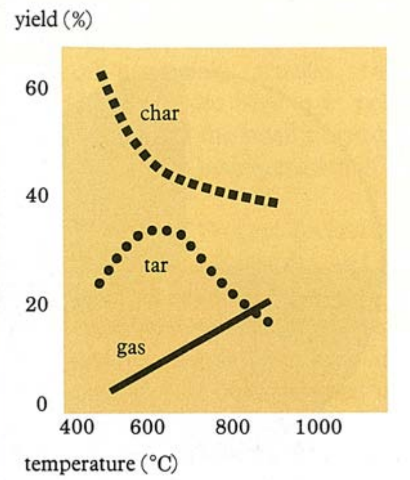
How pyrolysis temperature affects tar, char, and gas yield



Tar yield from the three coals tested is greatest at around 600°C. The graph shows results for Millmerran black coal.

The bench-top pyrolysis rig that can process 1 g of coal per hour.

More about the topic

Coal to oil by flash pyrolysis. J. H. Edwards, I. W. Smith, and R. J. Tyler. *Proceedings of the Fifth Australian Conference on Chemical Engineering, Chemeca 77, Canberra, September 1977*, 79–82.

Coal conversion research in Australia. R. A. Durie. *Proceedings of the Third International Conference on Coal Research, Sydney, October 1976*, 1977.