A History of Coal Liquefaction in the United Kingdom 1967-1992

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ABSTRACT

This paper gives an overview of the coal liquefaction studies carried out by the British Coal Utilisation Research Association, (BCURA), the National Coal Board (NCB), and the British Coal Corporation (BCC), between 1967 and 1992 at their research establishments in Leatherhead, Stoke Orchard and Point of Ayr. It is based upon a recently published ~ 100 page report (Ref:1) which condensed into a single reference document the contents of several hundred internal reports not generally available outside of BCC.

INTRODUCTION

Coal liquefaction is not new. Although Berthelot observed as early as 1869 that coal could be converted to an oil-like product by chemical reduction, practical processes using hydrogenation really date from 1913, when Bergius showed that brown coal could be converted to a heavy crude oil.

The main early developments took place during the 1920s and 1930s in Germany, while in the U.K. Imperial Chemical Industries built a plant at Billingham to produce 100,000t/year of liquid fuels from bituminous coal. This operated until 1939, after which it was used instead to convert creosote oil to aviation fuel. At this time, Germany continued to depend heavily upon coal liquefaction and built several plants, in total producing 3 million t/year, with the largest complex requiring 50,000 workers to produce 600,000t/year.

The price and availability of crude oil throughout most of the world since the 1950s has meant that coal liquefaction has not been economically attractive. However, in the mid-1960s in the USA there was interest in desulphurising coal to reduce environmental problems caused by the release of large amounts of sulphur oxides during coal combustion for power generation. It was thus to de-ash (and thereby reduce sulphur content) that the Solvent-Refined Coal (SRC) direct coal liquefaction process was developed, initially by the Spencer Chemical Company with an 80kg/h continuous unit funded by the Office of the Coal Research Council.

This revival of interest stimulated the preparation of a paper describing the BCURA work on solvent extraction of coal. The paper summarised three sets of laboratory-scale experiments that were carried out during the period 1955 to 1962 to examine whether a useful balance of products could be obtained economically via the solvent-extraction of low- or medium-rank coals. There had been three periods of activity:

(1) from 1956, when the objective was to make available quantities of coal extract for assessment by industry, while removing the minimum amount of volatile material consistent with yielding a residue with smokeless fuel properties;

(2) 1961, when the purpose was to make a similar extract by the simplest method available, to satisfy further enquiries from industry; and

(3) 1962, when the objective was to test the extracts potential for conversion to jet fuel.

These experiments were conducted at atmospheric pressure, with high solvent to coal ratios (e.g. 10:1), and temperatures around 200°C. In some tests, ultrasonic energy was applied to promote extraction of the coal. The yield of the pitch-like extract was usually around 10% and never reached 20%. Though the experiments were not taken to the pilot scale, it was found that the residues obtained after this moderate degree of solvent extraction could be pressed into briquettes which withstood typical handling and weathering. These could be ignited readily, and burned without producing substantial smoke. The solid extracts, which had low ash and very low (<1ppm) boron contents, were considered to have potential application as binders or more probably as sources of carbon for graphite manufacture. It appeared that these carbon artifacts might be produced at little more than their fuel value. Using the simple hydrogenation

equipment then available (limiting conditions to a maximum of 500°C and 300bar) and stannous sulphide or stannous chloride as catalyst, 20-30% yields (based on extract) of partially-hydrogenated cyclic hydrocarbons were obtained. While these boiled in a suitable range for use as jet fuels, the calorific value (66MJ/kg) was lower than desired.

The results of an economic assessment, for an integrated plant producing annually 250,000t of smokeless fuel and a solid extract, suggested a case could be argued for investigations to be carried out at the pilot-scale level. However, in 1967 it was decided to focus on experiments to establish the feasibility of markedly increasing the yield of extract from the 20% level.

ELECTRODE COKE VIA THE SOLVENT-EXTRACTION PROCESS, 1967-1979

BCURA as part of a collaborative project with NCB, initially concentrated its investigations into the solvent extraction of coal, upon the influence of operating conditions on extraction yields and product distributions. The range of variables studied included temperature, pressure, gaseous atmosphere, residence time, coal feed size, coal rank, and solvent type, and explored their impact upon engineering problems of solids separation and distillation. This information was to be obtained in the first place from batch equipment, although it was appreciated that the product yields and operating conditions would not be identical to those pertaining to a continuous-extraction process of the type which would be needed for any large-scale exploitation of the solvent-extraction process. The experiments showed that UK coals could be solvent-extracted but when a USA economic study of the SRC process was converted to the UK situation, it was evident that the process was not at that time economic in the UK; the relative cost of coal and oil in the USA was much more favourable for liquefaction than in the UK. Other factors that favoured the economic viability of the process in the US in comparison to the UK included the relatively high sulphur content of US coals, combined with the incentives to reduce atmospheric emissions - in the UK at that time it was argued that abatement of SO_2 emissions was unnecessary, in part as a consequence of climatic and geographic conditions that led to dispersion of chimney plumes. In the US a positive value was assigned to the high sulphur ash residues for landfill applications.

During the experimental work at BCURA, however, quantities of coal extracts were prepared under different conditions to those used in the SRC process. UK outlets for this re-constituted, de-ashed coal were sought, e.g. as potentially high value feedstocks for the manufacture of electrode binders and carbon fibres. Particularly encouraging was the discovery, during testing of the coal extracts at BCURA, that the properties of coke made from the coal extract compared favourably with premium-grade petroleum coke used in the manufacture of electrodes. At that time, such a grade of coke had about three times the value of coal, thus providing the basis for a potential economic process. The electrode-coke process which did not require the use of expensive hydrogen and the associated high-pressure equipment was thus developed.

Following a series of tests, culminating in a nine-cycle demonstration run, based around a 2litre autoclave reactor, the decision was taken by the NCB-led research committee to build a 0.5t/day plant at BCURA. Construction of this was scheduled to begin in February 1971 but in the event BCURA'S status as a Research Association ceased in the week before the contractors were due to start site operations at Leatherhead, and it was decided to transfer the whole project to the Coal Research Establishment (CRE) at Stoke Orchard, near Cheltenham. Virtually all of the equipment was moved and reinstalled within a few months. At the same time, W.C. Holmes and Co Ltd began construction at Stoke Orchard of the larger rig, now termed the 'extract plant'. Erection was completed during the latter half of 1971 and instrumentation and ancillary engineering aspects, carried out by NCB staff, were completed shortly afterwards. Commissioning was completed in February 1972 and coal first fed in March 1972.

During the following six years nearly 90 runs were carried out. By far the most significant was Run 45 which started on 13th January 1975 and continued for 168 days, during which time over 30t of coke were produced. In general, this was a period when the project size increased considerably, mainly due to the increased manpower needed to operate the extract plant; the team of 6 at BCURA increased in about a year to 24 at CRE.

Assessment of coke quality in terms of its suitability for electrode manufacture was carried out initially by Anglo Great Lakes (AGL) using 2kg samples and later on 250kg batches from the extract plant. AGL's long experience was extremely useful but much of the assessment was empirical and based solely upon experience with petroleum cokes and, thus, possibly not applicable to coal-extract cokes.

During 1973, the Atomic Energy Research Establishment (AERE), Harwell was contracted to fabricate some 25mm- and 75mm-diameter graphite electrodes which were subsequently submitted directly to British Steel Corporation (BSC) for testing in arc-steel furnaces. These electrodes performed much better in BSC's tests than would have been predicted by AGL (or any other graphite manufacturer) in that they had improved resistance to thermal shock compared to electrodes made with premium-grade petroleum coke. Although having a relatively high coefficient of thermal expansion, the cracks formed in the extract-coke electrodes propagated only slowly, reducing the rate of electrode degradation.

Much encouraged by these results and the economics based upon a capital costing carried out under contract by Kelloggs, plans were made to make enough coke to fabricate 300mm-diameter electrodes that could be tested by BSC on production arc-steel furnaces. This required the extract plant to run continuously for six months. To improve the project's ability to assess the coal-extract coke, facilities were set up at Stoke Orchard to produce and test small-diameter graphite rods. Samples of coal-extract cokes were assessed under a collaborative agreement with SIGRI (Germany) and a new agreement was negotiated with AGL. AGL used 20t of calcined extract coke supplied by NCB to fabricate 300mm-diameter graphite electrodes. The NCB graphite electrodes were vigorously tested at the Craigneuk works of BSC and normal production levels were maintained throughout the test period. It was concluded that coal-extract coke should be suitable for manufacture of electrodes up to and including 610mm-diameter, the largest in use at the time.

New designs and costings of larger plants, based upon specifications produced by the project, were obtained in 1977 from Catalytic Inc. However, the market for electrode coke was shrinking due to the severe contraction of the steel industry, particularly in Europe. Furthermore Conoco, at Immingham, were producing 200,000t/year of coke and Phillips, at Moerdyk in the Netherlands, were about to bring on-stream another premium-grade coke plant using a new feedstock, ethylene cracker tar. This new product also had problems getting market acceptance.

Much had been learnt about the various unit operations that made up the coal-to-electrode coke process. This was invaluable in the development of a process to produce transport fuels from coal when interest in this re-emerged because of the world crises in crude oil supply in the mid-1970s.

TRANSPORT FUELS FROM COAL - THE LIQUID SOLVENT EXTRACTION (LSE) PROCESS 1973-1986.

The first coal hydrogenation studies in the UK since the 1960s at BCURA began in June 1973 using a 2litre autoclave. At this time, a number of factors were threatening world crude oil supplies and the possibility of meeting at least a portion of the UK demand by synthesising oil from UK coal was considered strategically desirable. The extract plant sited at CRE was capable of supplying various streams which could have been hydrogenated (or pyrolysed) to liquids of which coal solution (the filtrate from the coal digest) and coal extract (evaporated filtrate, a solid at room temperature) were favoured technically because:

- they contain little inorganic material to contaminate catalysts and cause blockages in the plant
- the relatively unreactive inertinite portion of the coal has been removed
- they can be made fluid easily e.g., by the application of heat.

Coal extracts contain higher proportions of coal-derived material than coal solutions, so that changes in the coal material can be detected more readily. However, the higher softening points of coal extracts make them more difficult to pump.

The primary aim of the preliminary investigation was to hydro-treat coal extracts using the autoclave in order to explore possible process configurations and identify those with potential for development. However, the work had the additional aim of detecting possible problems at the more extreme conditions likely to be encountered during scale-up. An extra function was the

production of liquid samples for evaluation as chemical feedstocks by the Dutch State Mines. This work was carried out in the period June to December 1973 and was the precursor to a substantial programme of research supported and partially funded by European Economic Community (EEC) between 1974 and 1986.

Autoclaves were the only equipment available in the first few years but by 1976 coal-extract solutions were provided to British Petroleum (BP) for tests in their continuous-hydrocracking unit (CHU). Although originally it was hoped to continue this arrangement, separation of the extraction and hydrocracking stages between two sites hindered detailed recycle experiments. Consequently, additional funding was obtained and BP built a new CHU, installed it at CRE and provided training for operators and maintenance engineers. This unit, together with associated facilities built at various times, e.g. extract production using a dedicated integrated solvent-extraction plant (ISEP), was the key experimental facility for the research programme. Many aspects of liquefaction, eg catalyst selection, were first studied or screened on smaller equipment but the aim was always to confirm potential process improvements in recycle runs on the CHU-ISEP equipment [or the Integrated Liquefaction Plan (ILP) as it was renamed in 1983].

The main criteria by which success during repeated recycling was gauged were that 100% recovery of solvent was achieved, that the power of the solvent to dissolve the coal remained adequate and that pitch-plus-filter cake represented no more than 40wt% (dmmf) of the coal. Solvent quality during repeated recycling demanded much attention; obtaining the balance between hydrocracking (of the extract) and hydrogenation (of the solvent) proved to be particularly difficult. However, solutions to all the problem areas were found and in 1984 what was deemed a totally successful run was carried out.

During the period 1977-1986, over 30,000h running of the ILP were achieved with nearly 6,000h in 1984. Such long periods of operation were essential to prove catalyst life. Mass balances were carried out for virtually every day of operation and closures of over 95% became standard. The complexity of the plant, plus the amount of handling necessary to take the many samples that were needed to assist interpretation, meant that these good balances were only achieved by continued vigilance by operators and supervisors. The success of the ILP programme owed much to the teamwork of the more than 30 people involved. In addition to the studies supporting the ILP programme, much work was carried out during this period on the various unit operations, particularly on filtration using both small- and large-scale equipment (e.g. the extract plant in which other filters were installed). Secondary refining facilities were constructed with which finished gasolines and diesels were prepared, tested and also demonstrated in various vehicles (a lawnmower driven by Sir Derek Ezra, then Chairman of the NCB, a dumper truck by John Moore, Energy Minister, Automobile Association vans in the Lord Mayor of London's procession, as well as tests to measure fuel consumption, emissions, etc).

Proposals to build demonstration plants were made in the late 1970s and subsequently the siting of such a plant at Point of Ayr, North Wales, was agreed upon at an early stage. Consequently, Point of Ayr coal was chosen as the standard coal for use in experiments from 1981. Many other coals from around the world were tested on the small-scale and several in the ILP, including Illinois No. 6 coal which proved extremely easy to process. One of the major advantages of the LSE process that emerged was its ability to handle virtually any coal regardless of rank or ash content.

The design specification for the Point of Ayr plant was based upon information obtained up to 1983. Many configurations were considered and four incorporated into the design, i.e.

- single-stage hydrocracking
- two-stage parallel hydrotreatment
- two-stage series hydrotreatment pitch recycle to hydrocracking

LSE PROCESS DEVELOPMENT PILOT PLANT, POINT OF AYR, 1980-1992.

During the mid-1970s there were proposals to build larger electrode-coke plants, and these were considered in the Coal Industry Examination carried out in 1974. Its terms of reference were: "To consider and advise on the contribution which coal can best make to the Country's energy requirements and the steps needed to secure that contribution".

With regard to the LSE project at the time, to quote the report: "The NCB have pointed out that the prime objective of the project is the development of a liquid fuel from coal process, and that whilst the economics of electrode-coke production look attractive, the market is limited and a commercial venture could not carry the cost of developing the whole process. The extract hydrogenation stage provides the bridge to lighter hydrocarbon liquids, both fuels and chemical feedstocks, but this is still in the laboratory stage".

The report concluded: "We consider that the combination of NCB expertise, relevance to the UK economy and commercial prospects, justifies concentration on three areas, fluidised bed combustion, coal liquefaction by solvent extraction and pyrolysis".

Support came relatively quickly for the fluidised bed combustion project (at Grimethorpe, UK) but it was some time before an acceptable pilot plant project for the LSE process was formulated. However, in 1980 Matthew Hall Ortech (MHO) were chosen as contractors to perform design and costings of a 25t/day pilot plant comprising LSE and supercritical-gas extraction (SGE) front-ends with a common hydrocracking facility. SGE was abandoned soon after in 1982, partly as a result of MHO's reservations about further scale-up of this process.

However, the LSE 25t/day plant plan did not receive UK Government support. In late 1982, ICI were called in by the Chief Scientist at the Department of Energy as consultant to advise on the scale of the development proposed by the NCB. 1CI in their 'audit' proposed that engineering information could be obtained at a scale of 1t/day; NCB continued to consider that 25t/day was necessary on the basis of the process information that had been produced at CRE. However, in order to proceed, a compromise was reached in which a throughput of 2.5t/day was agreed (the base case in the specification, therefore, was 100kg/h of dry, mineral matter-free, coal).

The EEC had always been highly supportive and initial approval to provide significant funding for the 25t/day plant had been granted but had to be cancelled. The UK Government stipulated that to receive their support for the 2.5t/day plant, EEC funding had also to be obtained, project management should be strengthened and at least one private company should participate. Oil company support was not forthcoming for various reasons. However, this impasse was finally resolved by an agreement between Ian McGregor (Chairman of NCB) and Peter Walker (Secretary of State for Energy) in which funding from the Department of Energy and the EEC was released on the understanding that the NCB would make every endeavour to find a commercial partner. (This was eventually achieved when Ruhrkohle Oel und Gas GmbH and Amoco Corporation joined the Project in 1987 and 1991, respectively).

A steering committee was responsible for the project and it set the following objectives:

“The next stage in the development of the NCB's Liquid Solvent Extraction (LSE) process is to design, build and operate a 2.5t/day Process Development Pilot Plant (PDPP).

The primary objective is to demonstrate that the LSE process can be operated continuously using solvent derived from the feed coal; this requires that solvent quality and quantity be satisfactorily maintained and controlled and satisfactory yield pattern of distillates obtained”.

The project was nominally separated into four phases, i.e. specification, construction, commissioning and operation, and each phase was contracted separately. During the first phase, funded by the Department of Energy and BCC (the successor to NCB), the overall project costs and scheduling were ascertained. Following this, the complete project was approved in October 1985 by BCC's Board, with agreements reached with the Department of Energy and the EEC.

There were contractual and other problems during the construction phase which delayed commissioning and the planned recruitment, and the full project operating time was re-phased accordingly. However, the delay at Point of Ayr provided the opportunity to review again all aspects of the process and as a result additional experimental projects were proposed with

separate funding arranged with the European Coal and Steel Community (ECSC) and BCC. This made particularly good use of CRE staff who were to form the core of the Point of Ayr staff and significant advances in certain areas were made.

In addition to the CRE experimental work, studies had also been made on economic aspects. This included a commercial plant design and costing performed by Costain Ltd. The design reflected the evolved LSE process and the costing procedure enabled variations in the process (e.g. when using different coals) to be estimated with some confidence. The objective of this particular study was two-fold. In addition to giving up-to-date economic figures, it was intended to guide the Point of Ayr plant's operating programme such that the variables studied were those that could have the biggest economic impact rather than those which were academically perhaps more interesting.

Another study commissioned in 1991 was to consider how coal liquefaction might be introduced into the European refinery scene. This required the contractor, Trichem Consultants Ltd, first to predict the future European oil refinery situation well into the next century and then to estimate the value to refiners of coal-derived liquids.

A way of expressing the process economics is to calculate what the price of crude oil has to be for a coal liquefaction plant to give a nominal internal rate of return, e.g. 10%. This 'breakeven oil price' was around $35/bbl (for coal priced at $60/t) although this could be reduced in several ways. For example, if the coal price was reduced to $20/t, the break-even oil price could be as low as $25/bbl. Clearly, a liquefaction plant located near a cheap source of coal would seem to be a good solution. Unfortunately, most locations of cheap coal are such that the plant construction costs would be much higher (than for the base-case coastal location) and no net benefit would accrue. Consequently, the search for the ideal combination of cheap coal and cheap plant construction costs continued.

CONCLUSIONS

Direct liquefaction technology in the UK has advanced considerably from the stage in 1967, when little was known of the way in which UK coals behaved. Work on the electrode coke process not only extended knowledge of liquid solvent-extraction of coals but also showed conclusively that coal-based carbon materials were technically feasible and could be produced commercially. The LSE process extended the first development to show that petrol and diesel fuels could be produced with high efficiency, not only from UK coals but also from essentially any coal worldwide.

Knowledge accumulated from developing and proving these processes has been achieved through the efforts of many individuals and organisations, including those who ensured the availability of funds, those who were creative in overcoming problems and those who engineered the solutions. The key benefit of all these contributions and developments is that UK liquefaction technology has been advanced to the point that when, as it eventually will, the price of oil rises, action can be taken to enable coal to provide a realistic, alternative source for transport fuels.

REFERENCE

(1) G.M. Kimber, "A History of U.K. Coal Liquefaction" COAL, R078, ETSU, Harwell, U.K., Dec. 1996.