PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A
MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

Coal-Based Chemical Complexes [and Discussion]

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Coal-based chemical complexes

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In addition to heat or kinetic energy, coal can be used to produce carbon (in various forms), hydrogen and chemicals containing these elements. Coal conversion developments have recently tended to be concentrated on single products to replace those in declining supply. The apparent economics depend on cost projections of other fossil fuels but the future market balance between fuels may also be important.

Proposals have been made to improve the economics of coal conversion by combining various unit processes together in 'coalplexes'. Some of these proposals are reviewed; frequently they are inspired by specific situations. Some existing coal processing plants have acquired breadth with maturity; this may become common.

Some general considerations that may be taken into account in assessing coalplexes are discussed, including capital, flexibility and conservation. The optimization of coalplexes, however, impinges on the whole structure of energy availability and use. These relations are explored and some suggestions made, including a possibly important role for synthesis gas.

Introduction

The most important use of coal traditionally has been in combustion processes for the production of heat or energy. Coal has also been used to produce carbon, in various forms, hydrogen (by reaction with water) and chemicals containing these elements. It is generally accepted that the use of coal will continue to expand, probably for several decades, and that this expansion will be accompanied by a relative change in emphasis away from direct combustion towards the production of hydrocarbon gases and liquids, for use as premium fuels and chemical feedstocks. The scale and importance of this translation is difficult to grasp; perhaps within three or four decades, several hundred thousand million dollars will have been invested in these processes on a world scale to process about 10 % of the available primary energy. It is therefore important that the new systems are as efficient as possible and compatible with a general improvement in energy application.

The driving forces for these events will be the world's growing demand for energy and the limitations on supply of natural gas and crude oil. Coal conversion process developments have tended to be concentrated on single products to replace more or less directly these other fossil energy sources; the substitute products, however, have been thought to be more expensive and therefore not ripe for commercialization. The economic justification for coal as a primary feedstock depends on projections of cost trends: basically, the cost of coal plus capital equipment compared with prices of natural gas and oil. There has been considerable speculation about the time-scale for reaching economic comparability. This was illustrated in an earlier paper (Grainger 1974), reproduced here as figure 1. However, it was also suggested in that paper that the competitiveness of coal conversion processes might be enhanced or brought forward by the adoption of the 'coalplex' concept.

Coalplexes

General concept

The concept of a coalplex is a loose one, not rigidly defined. In the words of the 1974 paper, 'The various unit processes are combined together in ways intended to reduce their overall capital costs and to increase the overall efficiency while meeting the needs of the energy and raw materials market, taking account of storage and transport costs.' Some general considerations which may be important in devising or assessing coalplexes include the following.

(*a*) Capital savings may be achieved, especially through items such as site services or coal handling, which are common to more than one of the processes.

(*b*) By-products, energy or intermediates may be transferred between processes in a way that is more efficient.

(*c*) The output of different products may be flexible, assisting a better match with markets, possibly saving on energy storage costs and the provision of marginal capacity.

(*d*) Thermal efficiency may be improved; the recovery of energy that might otherwise be wasted may be facilitated through co-generation of electricity, local distribution networks for heat and the further treatment of low-grade residues.

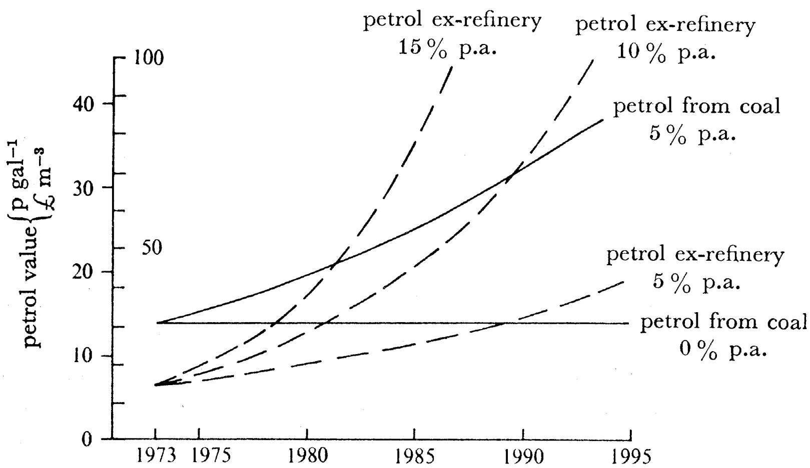

Figure 1. Petrol-from-coal processes: effect of cost annual growth rates.

Some idealized schemes

There are only a few basic components of coalplexes. At the tail-end of processes, residues containing the original ash, some carbon and possibly unseparated hydrocarbons may be combusted to provide steam and electricity, either for export or in-house use. The development of fluidized-bed combustion (f.b.c.) makes this a much more convenient disposal process; steam for gasification may be raised in this unit. Alternatively, the residues may be gasified or partly gasified (probably in an entrainment or fluidized type of gasifier) followed by combustion; the gas might be sold or used in the process, probably after shifting it first to hydrogen.

At the other end of the process, the initial attack on the coal might be intended to remove only a moderate proportion of the coal substance (perhaps 20 %) in the form of relatively small molecules containing as much of the hydrogen as possible; the residue could be a good feedstock for gasification or combustion. This 'devolatilization' might be by pyrolysis or solvent extraction; in the latter case, supercritical gas extraction is probably ideal. The demand for and value of the derivatives from the extract and the residue could determine the 'cut', which might be adjustable to some extent.

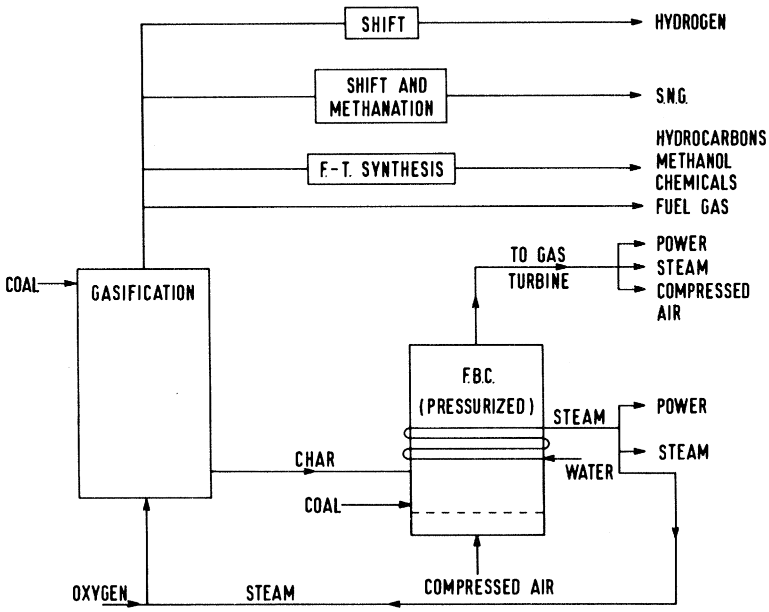

FIGURE 2. Coalplex incorporating gasification, synthesis and combustion.

Hydrogen and fuel gases could be 'balancers'. Hydrogen is a crucial raw material in coal processing and in other industries (e.g. oil refining, chemicals, ore reduction). Surpluses or deficits could be exchanged not only between coal conversion processes but with adjacent industries. Fuel gases frequently arise in coal conversion processes (e.g. Fischer–Tropsch synthesis) and recycling may be costly; an external outlet may be a more economic solution.

Carbons may also be coal conversion products; coke is an example from an old process. Chars suitable for making briquettes may be made by pyrolysis; binders may come from the volatile products of pyrolysis or from solvent extraction. Purified coal extracts can be coked to produce special electrode coke or spun into fibres.

Synthesis gas is of course a very flexible intermediate, being a possible step towards methane, methanol (and thence to gasoline), light hydrocarbons, hydrogen and ammonia.

Some simplified block diagrams may illustrate a few of the possibilities. Figure 2 is based on gasification, which might be a two-stage system, one part designed to produce mainly synthesis gas and the other aiming for a high initial methane content. The coal feed to the combustor might be simply for balancing purposes. Figure 3 is based on liquid solvent extraction; again, ancillary coal feed is provided to the gasifier. Figures 4–6 show process schemes based on initial supercritical gas extraction. The reactive properties of the char in this case make it a particularly attractive feed for hydrogasification.

In the chemicals field, coal has been considered as a feedstock, particularly for ammonia, sometimes in schemes involving other coal products. One scheme from Catalytic, Inc. (Udant 1978) is outlined in figure 7.

Design studies

In the last few years, the coalplex concept has not received a great deal of attention, as such, interest being concentrated largely on processes intended to produce single products. In many cases, however, it is being realized that recycling by-products or intermediates to achieve the single-product aim may become increasingly expensive as this end-point is approached and

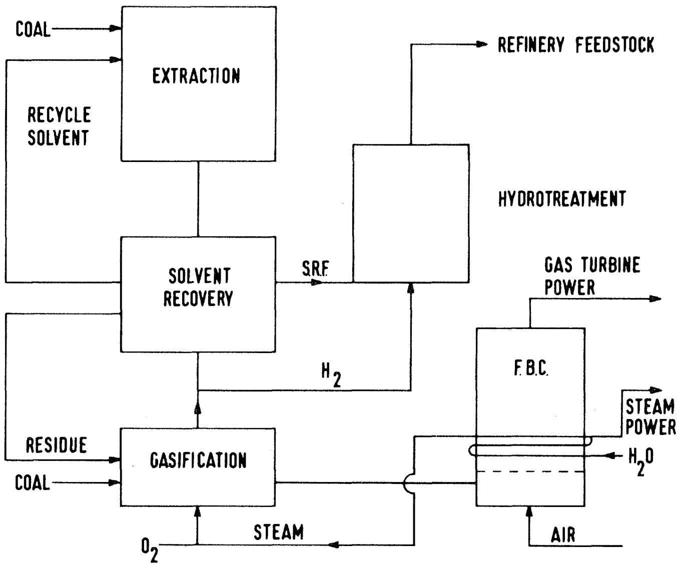

FIGURE 3. Coalplex incorporating liquefaction and fluidized combustion.

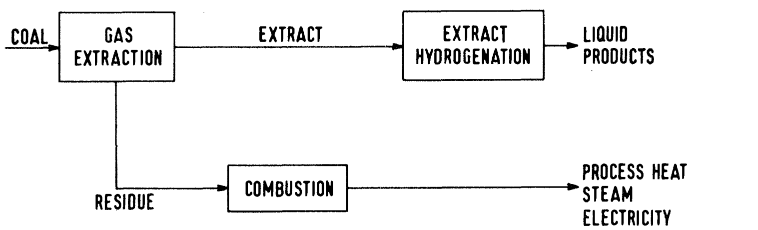

FIGURE 4. Coalplex incorporating supercritical gas extraction and combustion.

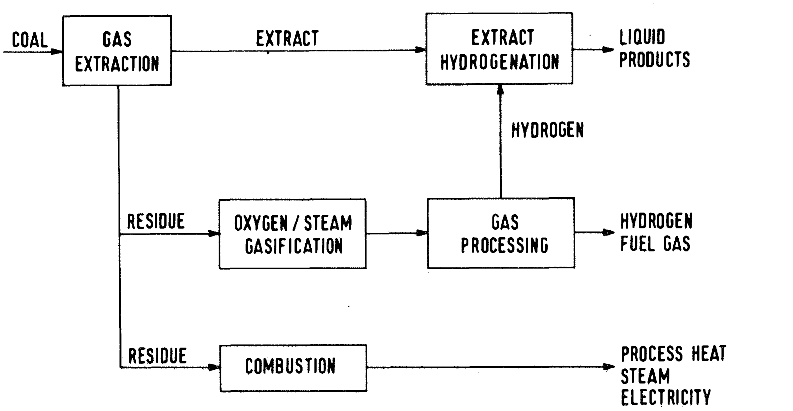

FIGURE 5. Coalplex incorporating gas extraction, combustion and gasification.

here have been studies to assess the economics where a secondary product is sold. This also applies to the treatment of residues and sometimes as a means of avoiding or mitigating a difficult step such as filtration of coal digest.

An important landmark, however, was a design study carried out for the U.S. Department of Energy by the Ralph M. Parsons Company, reporting in November 1978 (Parsons Company 1978). Conceptual designs were developed for four commercial-scale conversion complexes, which were then assessed technically and economically. A brief description of the four processes is as follows.

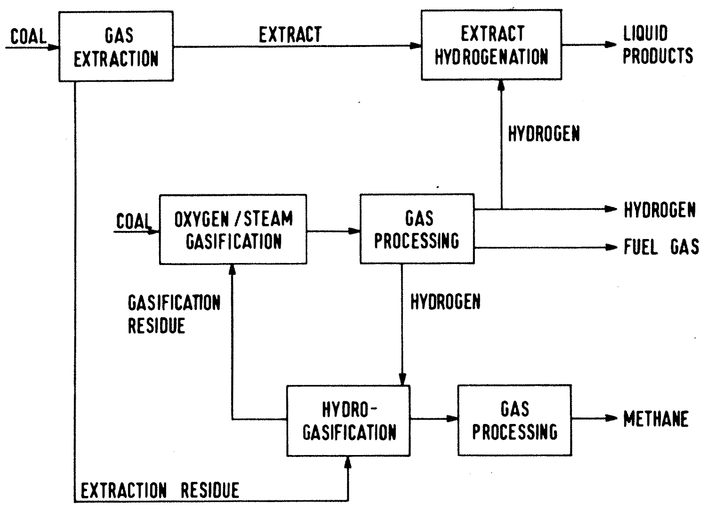

FIGURE 6. Coalplex incorporating gas extraction and hydrogasification.

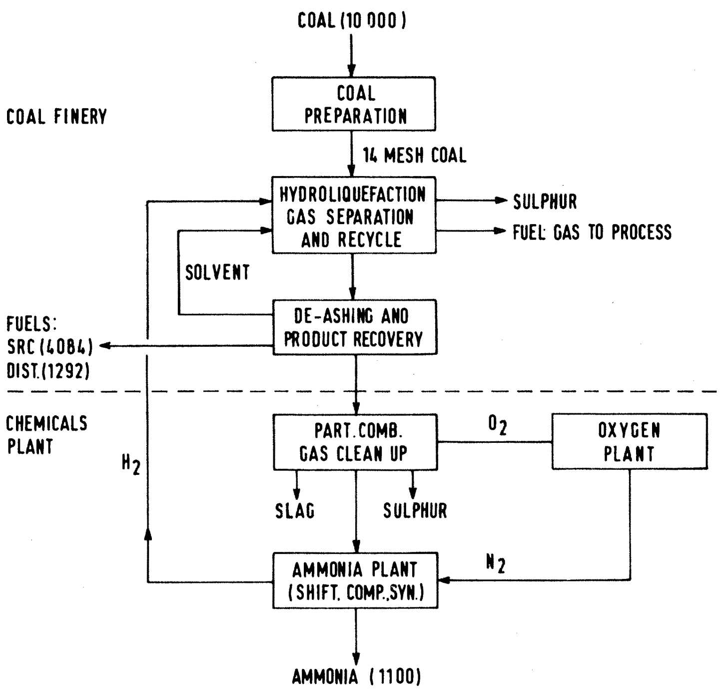

FIGURE 7. Coal refinery and chemicals plant (all figures are tonnes per day).

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(*a*) Char oil energy development (c.o.e.d.) based pyrolysis plant. Multiple fluid-bed pyrolysers produce tars, fuel gas and char. The char is gasified and some of the product is converted to hydrogen to treat the tar.

(*b*) Oil/gas. This is a version of the Solvent Refined Coal (SRC) process where the co-production of fuel gases is not discouraged.

(*c*) Fischer–Tropsch. This was a version where the heat of the reaction is recovered as high pressure steam, which is used in the process and also for generating electricity for external sale. S.n.g. is also a significant export.

(*d*) POGO (Power–oil–gas–other). This is a coal refinery that co-produces electrical power, liquid fuels, gas fuels, chemical by-products and coke. The main process units are SRC hydroliquefaction, pressurized flash pyrolysis, two-stage gasifier, combined cycle power plant and a coker. The design was capable of great variation in the balance of inputs and products. In the particular configuration chosen for comparison, about 45% of the coal went to the SRC plant, 15% to pyrolysis and 40% to gasification (some of which was used for hydrogen make-up).

Economics

The economics in the Parsons study were calculated on the basis of a captive coal mine, which makes translation to other countries, or indeed to other conversion schemes, complicated. However, the comparisons given between the different processes are interesting (see table 1).

TABLE 1

(1978 dollars. Financing 65% debt (9% interest), 35% equity (12% D.C.F.).)

	oil/gas	Fischer–Tropsch	c.o.e.d.	POGO
$\$/10^6$ Btu†	2.10	2.90	5.00	2.17
$/b.o.e.‡	2.50	16.00	30	13

† 1 Btu = 1.055 kJ. ‡ 1 b.o.e. (barrel oil equivalent) $\approx 6.1 \times 10^{18}$ J.

Unfortunately, these figures necessarily apply only to one set of relative values for products and to one balance of outputs. However, from the data provided, reoptimizations could be carried out to suit different conditions. This would be an excellent starting point for a computerized economic model. In a further description of a version of POGO, a very important claim is that the projected thermal efficiency to synfuels, by-products and gas to electricity is 75%. A combined cycle generator is proposed with an efficiency of 44%.

General studies of the economics of coalplexes have been made by the Coal Research Establishment of the National Coal Board, using published data for unit processes. These studies suggested that the capital costs of plants designed to produce two products could be significantly (up to 20%) lower, per unit of energy output, than for plants producing only one of these products. The average product cost depended on the mix of products required but there was generally a considerable range of product split over which costs were significantly lower than for either single product, usually s.n.g. or SRC. The matching of products to markets and to fluctuations in demand is, however, another, very important, matter.

There is a need for improved economic modelling that could take account of all cost factors in the production of coal conversion products and relate these to the whole structure of energy availability and use. The demand for energy, including that possibly derived from coal, varies widely and in a complex fashion: diurnally, seasonally and in sympathy with the level of economic activity; there are real resource costs, not coordinated among energy suppliers, in matching demand with supply. The optimum pattern will change with time and there will be political factors as well as pure economic ones. Models to take account of these factors would be complex but are feasible, and are necessary to the full understanding of coalplexes and the way that inbuilt flexibility of output, requiring extra capital, might be recouped. Any departure from the optimum overall energy system represents a real waste of energy. The relation with non-fossil energy sources must also be considered, but the main effect of such an analysis might be to emphasize the flexibility of coal and the need to provide facilities to exploit it.

The future

Whatever the potential advantages of coalplexes may be, it seems unlikely that a complete large-scale complex of the type discussed in the Parsons report will be ordered as a single entity in the near future. Finding the right financial and institutional framework for a 'straightforward' s.n.g. or SRC plant seems difficult enough. How then may coalplexes evolve? And if this is a gradual process, can a visualization of the long-term outlook help the short term?

It is possible that there is an inherent tendency for coal processes to become diversified with time. The coking industry may be a case in point; originally, coke was virtually the only product but subsequently the recovery and exploitation of by-products became quite complex and financially attractive. Gasoline was the primary objective of Sasol 1 but other products are also made, and substantial expenditure has been incurred on a pipeline system to sell gas. It will be of interest to see whether Sasol 2 and 3 diversify, although the initial policy seems to be against this. There does appear to be a general tendency in the development of the main gasification and liquefaction processes to seek opportunities for profitable exploitation of by-products. The increasing prices of primary energy will provide an incentive for maximum recovery of energy values.

Co-processing of coal and oil

Oil refineries are to some extent a model for the development of coalplexes, although perhaps historically unduly self-sufficient. Coalplexes might develop by coal conversion's being 'grafted-on' to oil refineries. A situation could be envisaged in which coal might be converted into SRC as a discrete stage and then processed in a manner analogous to oil, probably in existing refinery plant. This might not, however, be ideal if the opportunities for maximizing the value of all potential coal liquefaction products, and not merely the SRC, were missed. In any case, this would ensure that the coal process could not be economic until very large changes in relative prices of oil and coal occurred.

A more transitional procedure would certainly be better. Initially, coal could be used to provide process steam and power and, a little later, hydrogen (the use of which is growing in refineries). At present, low-value products such as heavy oils and off-gases are used for these purposes; by replacing them with coal, they could be used in more valuable ways, increasing refinery output from a given oil input by 5–15%.

Subsequent to the peripheral use of coal in oil refinery practice, various possibilities for intermingling of process streams exist. SRC-type liquids might be added to oil but since coal derivatives tend to be rich in aromatics, some adjustments to the refinery might be necessary. Alternatively, coal liquids derived by pyrolysis or gas extraction might be added to refinery streams; in those cases, the high proportion of active char would be a very suitable material for producing hydrogen, steam and power, thus enhancing integration.

Petroleum products are not normally good solvents for coal but some success has been claimed, notably in Russia, probably because oil, rich in aromatics, may have been available. However, some refinery streams, especially those associated with delayed coking, tend to be higher in aromatics than normal and are useful coal solvents; additional heavy fractions made in this way could then be added to the feedstock. The addition of powdered coal to petroleum streams at various stages has been studied and has been claimed sometimes to give a higher yield or value (or to reduce the feedstock cost) in hydrogenation, cracking or distillation stages. In these cases, the mechanism may be that the coal contributes extra liquids by pyrolysis or solvent extraction; coal particles may also provide catalytic or nucleating effects.

Thus, the prospects are excellent for integration of coal into many aspects of the oil refinery process, thereby approaching in this way a coalplex configuration. This would be enhanced if oil–coal refineries developed closer external links through the export of heat, electricity and fuel gases.

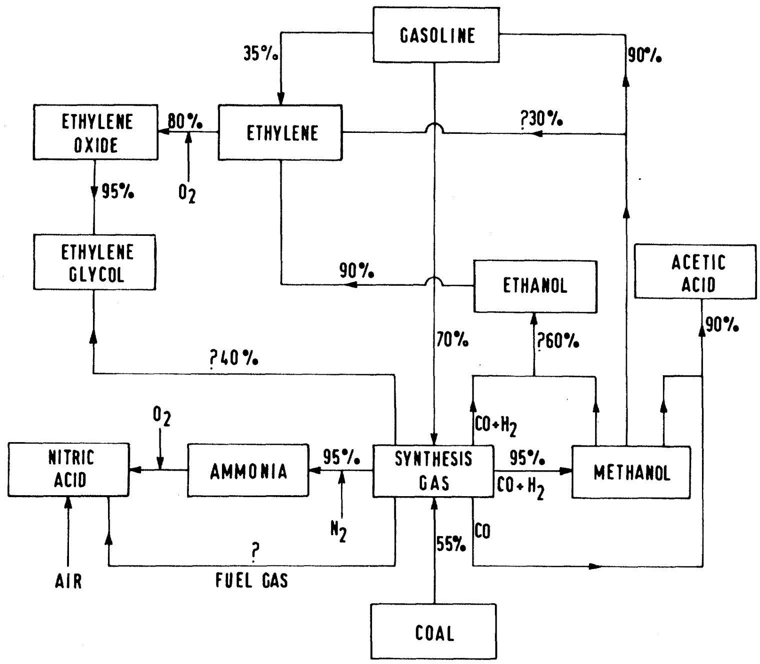

Figure 8. Petrochemicals from synthesis gas (efficiencies of steps are shown).

Synthesis gas

An alternative plan for the evolutionary development of coalplexes might be through the establishment of centres for the production of synthesis (or medium calorific value) gas, on sites capable of expansion and reasonably convenient for coal supplies and markets, for chemical feedstock, fuel gas, heat and electricity. The manufacture of such gas is already economic or nearly so in many parts of the world, based on straightforward competitive economics. Considered more broadly as a method of increasing the value of coal and of conserving supplies of natural gas and light oils, the case for such plants seems compelling.

Once such plants are established, with multiple markets, there would be excellent opportunities for further integration, both upstream and downstream. Upstream, instead of using

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raw coal for gasification, there might be a preliminary separation of coal liquids by pyrolysis or gas extraction; the gasifiers could provide heat and hydrogen. Downstream, chemical synthesis might be developed, plus methanol or gasoline; these possibilities are well illustrated in figure 8, due to Andrew (1977). Alternatively, if the hydrogenation routes to coal liquids were preferred, the gasifiers could again provide hydrogen and heat. The sizes of additional units could be determined in relation to the supply–demand model to optimize overall energy availability with minimum reserve.

If cheap heat or hydrogen became available through non-fossil sources, such as nuclear power, these could be integrated.

Conclusions

Coalplexes have potential for increasing the efficiency of the much greater contribution from coal in the future; the current development of a wide range of unit processes emphasizes this opportunity. The analytical tools probably need considerable development, however, if the best choices are to be made and developed.

Major process developments, nominally for a single product, may evolve in the direction of greater diversity and overall energy economy. Two other evolutionary ways of approaching efficient coalplexes are either (*a*) through supplementing petroleum feedstocks in refineries or (*b*) by starting with synthesis gas manufacture and integrating such plants with upstream and downstream processes. In both cases, maximum links with external energy flows should be sought.

An appreciation of the potential of coalplexes could influence the way in which early coal conversion processes are established, with long-term benefits.

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Discussion

L. Petrakis (*Gulf Research and Development Co., Pittsburgh, Pennsylvania* 15230, *U.S.A.*). The first U.S. field test of underground coal gasification (u.c.g.) for steeply dipping coal beds (s.d.b.) has recently been completed.† The test was performed by Gulf Research and Development Company and T.R.W. under a cost-shared agreement with the U.S. Department of Energy (D.o.E.). This field test was part of a wider effort currently funded by the U.S. D.o.E. to evaluate the technical and economic feasibility of u.c.g. The project utilized (*a*) the technology base developed by the U.S. D.o.E. in other field tests and (*b*) translations of the considerable Russian experiences.

The site (near Rawlins, Wyoming) for the field test was selected carefully to meet a variety of geological and environmental criteria and to establish a meaningful data base for estimating costs of commercial facilities. The test burn was completed in early December 1979, on a tract on the eastern rim of the Continental Divide Basin, in a 7 m thick coal bed dipping at 63°. The

† Project managed by Mr R. H. Graham and Dr A. H. Singleton.

coal was ignited at a vertical depth of 400 ft (122 m), with the use of directionally drilled wells. Table 2 summarizes the data obtained from the test including production rate and quality of the product. Figure 9 illustrates the procedure developed to gasify s.d.bs, a significant and otherwise inaccessible resource. Some of our project objectives were to investigate the effects of water–air injection, steam–air injection, and steam–oxygen injection.

Table 2. Rawlins no. 1 field test summary

	air injection (30 days total)	oxygen injection (5 days O_2)	total test (35 days)
product heat value/(Btu/ft^3)†	165	250	181
av. prod. wet gas/(10^6 ft^3/day)	4.32	4.10	4.34
energy production/10^6 Btu			
daily	613	740	614
total	18400	3100	21500
dry gas composition‡ (mol%)			
N_2	52.5	1.6	—
H_2	12.6	31.4	—
CO	15.8	34.0	—
CO_2	14.4	29.9	—
CH_4	4.5	2.8	—
C_2^+	0.11	0.17	—
average amount coal consumed per day/ton	34.0	39.1	34.7

† Includes some tars.
‡ Approximately 12% water was present in the wet product gas during the air injection phase, approximately 17% water during the oxygen injection.

According to plan, approximately 1200 tons† of coal were used during the test. The heating value of the product gas initially climbed to approximately 180 Btu/ft^3‡ and, as expected, gradually declined to the 120–130 Btu/ft^3 range. Air injection rates of 1600–2000 ft^3/min§ were used, and product gas rates between 3000 and 4500 ft^3/min were obtained. The water–air blown phase lasted for 21 days and on the average produced the desired target values of 5.8×10^6 ft^3/day of product gas quality. After the 21 day air-blown test, air injection was run at a reduced rate for about 8 days to conserve coal for the steam–oxygen test (table 2). A 5-day steam–oxygen blown experiment was conducted after the steam–air phase. As expected, steam–oxygen injection easily doubled the product gas heating value to the 240–260 Btu/ft^3 range. Oxygen flows were generally kept between 300–1000 ft^3/min, producing between 1500 and 4000 ft^3/min. Steam/oxygen ratios were varied between 0.2 and 2.7.

The process was terminated by shutting off all injection flows and by shutting in the injection wellhead. A day later the product wellhead was shut in. Reactor pressures generally remained below 20 lbf/in^2 (*ca.* 138 kPa) and reactor temperatures began a steady decline. This method of terminating the process appears effective in this test and no doubt its effectiveness depends on the predetermined freedom from fractures and discontinuities in the coal seam.

A total of 28000 ft^3 (790 m^3) of coal had been used by the end of the test. The probable dimensions of the burnt-out zone are in the order of 50 ft (15 m) long, 23 ft (7 m) thick, and 25–30 ft (8–9 m) wide. The reactor shape is indefinite owing to the small number of instrumentation wells emplaced on site; further reactor definition will be provided by post-burn measurements to be performed in the near future.

† 1 U.S. ton = 0.9072 t.
‡ 1 Btu/ft^3 (at s.t.p.) = 37.3 kJ/m^3.
§ 1 ft^3 (at s.t.p.) = 0.0283 m^3.

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The environmental studies concentrated on three major aspects: groundwater quality, air quality and subsidence. One year of baseline groundwater and air quality monitoring was conducted before the test. Samples of groundwater immediately before, during and after the test were taken from three wells located as closely as possible to the proposed burn zone. No change in water quality was seen in these wells after 10 days into the test. Further analyses are still forthcoming. Air quality was monitored from an environmental trailer located downwind

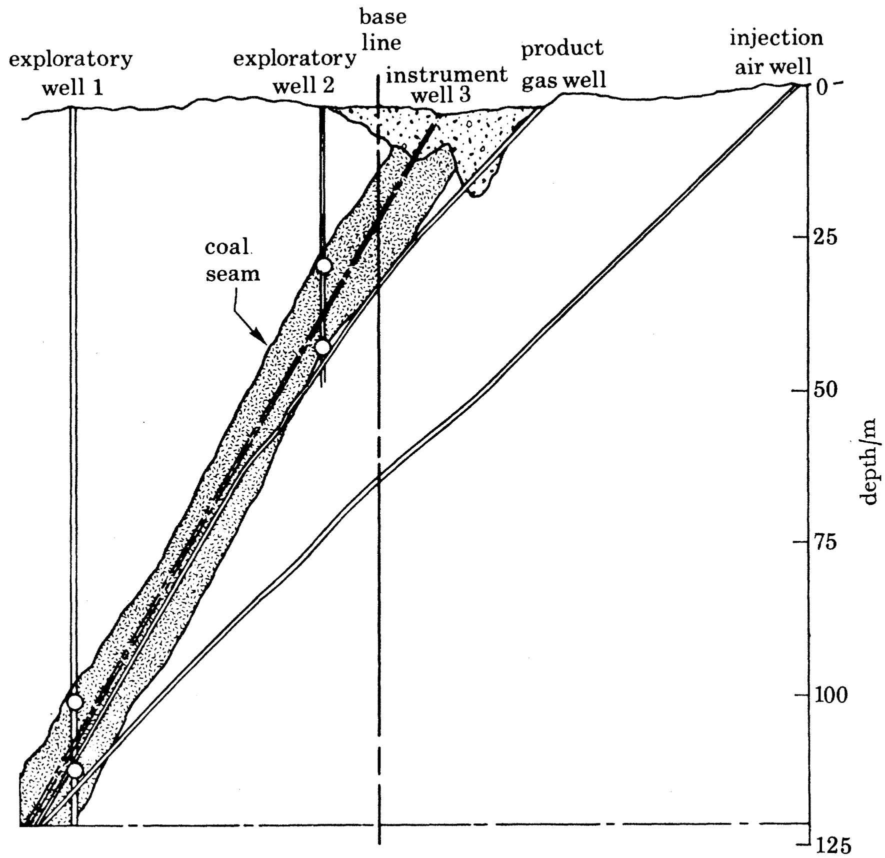

FIGURE 9. Underground coal gasification for steeply dipping coal beds: module 1 well cross section.

of the flare. A PT MAX plume dispersion model was used to locate the trailer in an area where maximum pollutant concentration would be expected. A preliminary analysis of the data indicates that air quality levels during the first 3 weeks showed little variation from levels collected during the same quarter of the baseline period. None of the standards for gaseous pollutants were violated for the same period. Before the test, a series of 10 subsidence monuments were placed on the most likely areas where subsidence was likely to occur. Surveys taken at various times during the test indicated that there was no movement of the surface.

L. GRAINGER. This is an important report of what appears to have been a highly successful test. The gas during the air injection period was an excellent fuel gas. During the oxygen–steam injection period, the gas could be used as a synthesis gas, possibly as part of a coalplex. The possibility of having only some wells on oxygen blowing, with the rest providing heat and power for producing and using synthesis gas, might be considered, as might the possibility of combining underground coal gasification (u.c.g.) with coal produced conventionally nearby.

The economics would require careful analysis and this method might prove temporarily more expensive than some alternatives. However, Dr Petrakis rightly refers to steeply dipping beds as 'a significant and otherwise inaccessible resource'. This applies also to deep seams. When energy prices generally increase sufficiently, u.c.g. will almost certainly come into its own, not as a competitor for conventional mining but as an adjunct. At that time, u.c.g. processes that produce synthesis-type gases may well be most desirable.

G. J. Lawson (*Department of Minerals Engineering, University of Birmingham, U.K.*). Many ways of converting coal have been described at this meeting, and their integration into a form of 'coalplex'. As far as liquid fuel manufacture is concerned, methods involve either coal hydrogenation or the use of synthesis gas mixtures.

However, it appears to be assumed that sufficient coal will be available for such processes, which are aimed to take over economically as oil becomes scarce; Professor Kölling pointed out that coal supplies only one-third of the F.R.G.'s energy needs, and that three times the current production would be needed if oil were not available. Although coal reserves are plentiful, is it possible that not enough manpower and machinery will be available to deliver the required coal to the surface? The National Coal Board has extensively mechanized its mining operations, but output is only half the peak value reached in the 1950s. Could the Board double its output?

Alternatively, should greater attention be paid to the possibilities of underground gasification, and to subsequent 'synthesis gas' processing routes?

L. Grainger. Dr Lawson may note the reply to Dr Petrakis. U.c.g. may be best applied to 'difficult' coal deposits and may be relatively long term. In the medium term, indigenous coal resources, especially in the industrialized countries, should be exploited in an optimum fashion. This means that maximum thermal efficiencies should be sought at each conversion and utilization stage, on the lines described in my paper.

Nonetheless, some industrialized areas, especially western Europe and Japan, will need to import large quantities of coal, and arrangements for this should be made as soon as possible; this should include a review of the whole processes of getting, moving, converting and using coal.

Any requirement for imports of coal into the United Kingdom will probably be much later than in Western Europe generally, owing both to the U.K.'s reserves of oil and gas and also to the potential for continuous though gradual expansion of coal production. An increase to, say, 170 Mt per year by 2000 has been mentioned and is technically feasible. Manpower and machinery could be available, provided energy and investment policies give adequate priority to coal. The implications of such policies are very significant for the U.K., which underlines again the need to get maximum efficiency out of the coal used.