Development of the Shell-Koppers Coal Gasification Process [and Discussion]

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Development of the Shell–Koppers coal gasification process

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The Shell–Koppers process for the gasification of coal under pressure, based on the principles of entrained-bed technology, is characterized by: practically complete gasification of virtually all solid fuels; production of a clean gas without by-products; high throughput; high thermal efficiency and efficient heat recovery; environmental acceptability.

There are numerous possible future applications for this process. The gas produced (93–98 vol. % hydrogen and carbon monoxide) is suitable for the manufacture of hydrogen or reducing gas and, with further processing, substitute natural gas (s.n.g.). Moreover, the gas can be used for the synthesis of ammonia, methanol and liquid hydrocarbons.

Another possible application of this process is as an integral part of a combined-cycle power station featuring both gas and steam turbines. The integration of a Shell–Koppers coal gasifier with a combined-cycle power station will allow of electricity generation at 42–45% efficiency for a wide range of feed coals.

The development programme includes the operation of a 150 t/day gasifier at Deutsche Shell's Harburg refinery since November 1978 and of a 6 t/day pilot plant a Royal Dutch Shell's Amsterdam laboratories from December 1976 onwards. Both facilities run very successfully. With hard coal a conversion of 99% is reached while producing a gas with only 1 vol. % CO_2.

The next step will be the construction and operation of one or two 1000 t/day prototype plants which are scheduled for commissioning in 1983–4. Towards the end of the 1980s large commercial units with a capacity of 2500 t/day are contemplated. The economy, especially of these large size units, is very competitive.

Development of the Shell–Koppers process

Shell Internationale Petroleum Maatschappij B.V., The Hague, and Krupp–Koppers GmbH (formerly Heinrich Koppers GmbH), Essen, have together been working on the development of a high-pressure coal gasification process since the beginning of 1974. A pilot plant of 6 t/day intake capacity has been in operation at Shell's Amsterdam Laboratories since December 1976. As part of this development, Deutsche Shell A.G. decided to set up an experimental plant employing this process.

The development of this large pilot plant, with a daily throughput of 150 t of coal, drew on the existing experience of the two associates in the venture. On the one hand, there was the experience of Krupp–Koppers, who have built numerous coal gasification plants employing the Koppers–Totzek process, working at atmospheric pressure, and on the other hand there was Shell's experience obtained from the Shell oil gasification process, a high-pressure process also employed in many units operating around the world.

The Shell–Koppers 150 t/day pilot plant has made a series of very successful runs since its commissioning in November 1978. The longest run to date has been 240 h.

Although the plant has been built by Krupp–Koppers in cooperation with Shell process and project engineers in The Hague and Harburg, it is the property of Deutsche Shell A.G. and is operated by that company. The capital and operating costs are expected to approach $50M.

Based on the experience with the pilot plants, designs are being made for one or two 1000 t/day prototype units which are scheduled for commissioning in 1983–4. The capacities of subsequent fully commercial units will be increased in stages to about 2500 t/day towards the end of the 1980s.

THE PROCESS

The new process is based on the principle of entrained-bed gasification at elevated pressure under slagging conditions (i.e. at temperatures sufficiently high to ensure that the ash is in the molten state). The main technical features of the process are described below.

Process features

Coal conversion

The process is suitable for the complete gasification of a wide variety of solid fuels, such as all types of coal and petroleum coke. Fuels with a high ash content (up to 40% by mass) and high sulphur content (up to 8% by mass) can be used in the Shell–Koppers gasification plant without any trouble. Even a high water content in the coal does not pose a technical problem. However, on economic grounds, particularly with a view to reducing the oxygen requirement and improving the quality of the gas, the coal is in most cases dried to a moisture content of 1–8% by mass.

Since the process requires a solid fuel to be in dust form for gasification, the entire output of a mine, including fines, is acceptable as feed. Unlike fixed-bed or fluidized-bed processes, in the Shell–Koppers process there is practically no limitation on the coal as to ash fusion behaviour or caking properties.

A new development in hand is the gasification of vacuum residues of direct coal liquefaction plants and other hydrocarbon liquids in a Shell–Koppers type reactor. An advantage of such a process over the Shell gasification process for oil is that liquids containing unconverted coal and a high percentage of ash can be gasified.

Gas quality

The operation at very high temperatures ensures the formation of a high-quality synthesis gas essentially consisting of hydrogen and carbon monoxide (93–98 vol. % for oxygen gasification). Tars, phenols and other by-products are absent; as a rule methane concentrations in the gas do not exceed 0.2 vol. %.

Unit capacity

Both the high temperatures (above 1400 °C) and the high pressures are responsible for the high capacities attainable. Short-term targets are 50–100 t/h coal per reactor, corresponding to $2.4\text{–}4.8 \times 10^6$ m^3/day (at s.t.p.) of raw gas.

Depending on the application, the optimum pressure level can be selected. Apart from the beneficial effect of the elevated pressure on reactor capacity, there are spin-offs in terms of increased heat transfer rates in the waste heat boiler, easier gas treating and a reduction in gas compression costs.

Thermal efficiency

The chemically bound heat in the gas produced with oxygen gasification is equivalent to about 79–82% of the chemically bound heat contained in the coal feed. The recovery of the sensible heat from the hot gases leaving the reactor accounts for another 12–15 % of the heat content of the coal feed. The surplus steam produced by this cooling is generally sufficient to drive the compressors of the oxygen plant.

Heat losses to atmosphere are reduced to a minimum. The combined operation of reactor cooling system and waste heat boiler results in an efficient heat recovery by the production of superheated high-pressure steam (typically 540 °C, 10 MPa).

Environmental acceptability

A negligible environmental impact can be expected from the Shell–Koppers process. The requirements for both process and cooling water have been minimized in both the gasification and the solids removal sections. The gas produced contains no tars, phenols, condensable hydrocarbons or organic sulphur compounds. Physical and chemical means are available for removing hydrogen sulphide and traces of cyanide, carbonyl sulphide and ammonia from the synthesis gas. The only by-products from the process are elemental sulphur and the unavoidable ash from the coal as a non-leachable and inert slag. Depending on the application, the production of waste water is very low or nil.

Flow scheme of the 150 t/day *pilot plant in Harburg, Germany* (figure 1)

Coal is ground and dried to specification in a coal mill and subsequently pneumatically transported to the atmospheric cyclone hoppers (2). In the Harburg plant nitrogen from a liquid nitrogen storage vessel is used as a carrier gas for the coal. To transport the coal from the cyclone hoppers (2) to the nitrogen pressurized feed hoppers (4) a fully automated lock hopper system is used. The pneumatic transport of the coal from the feed hoppers to the burners is again accomplished with nitrogen. For good control of the process it is essential to

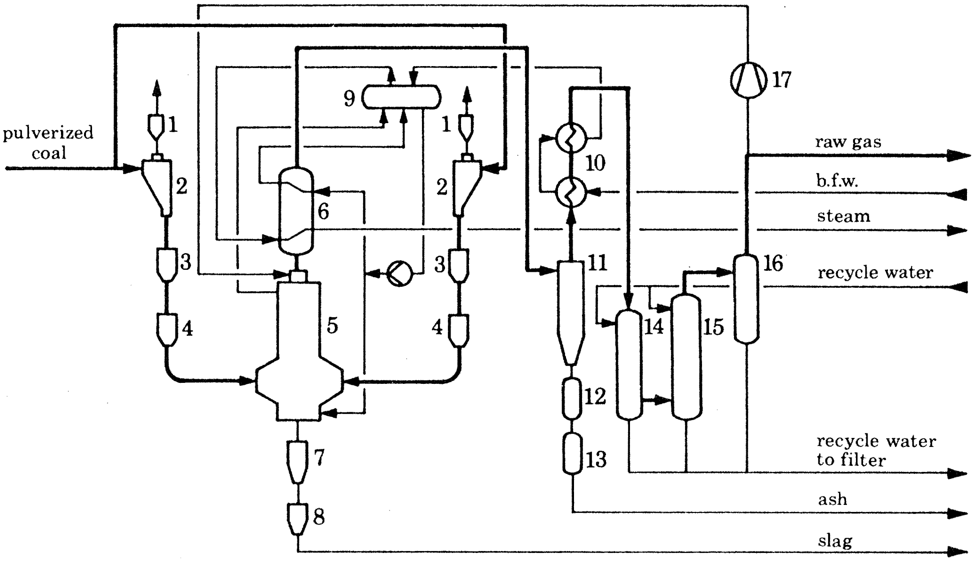

FIGURE 1. Shell–Koppers coal gasification process: flow scheme, 150 t/day Harburg pilot plant. 1, Coal filter; 2, cyclone hopper; 3, lock hopper; 4, feed hopper; 5, gasifier; 6, waste heat boiler; 7, slag breaker; 8, slag lock hopper; 9, steam drum; 10, b.f.w. preheater; 11, cyclone; 12, ash hopper; 13, ash lock hopper; 14, venturi; 15, scrubber; 16, h.p. separator; 17, recycle gas compressor.

be able to measure accurately the coal, oxygen and steam flow to the burners. The success of the pneumatic feed system has been made possible by the development of an accurate and dependable coal flow meter.

The reactor is equipped with two diametrically opposed burners and consists essentially of a pressure shell protected from the hot gases by a tube wall in which saturated steam at 5 MPa is raised. The tube wall is in turn protected by a thin layer of a refractory material. The slag, which leaves the reactor via a hole in the bottom, is quenched in water, crushed in a submerged mill and then lock-hoppered out to atmospheric pressure.

The gases leaving the reactor at about 1500 °C and 3 MPa are quenched with solid free recycled synthesis gas of 100 to 800–900 °C to solidify the entrained slag particles before they enter the waste heat boiler (6). The gases leave the waste heat boiler at a temperature of 320 °C. In the waste heat boiler superheated steam at 500 °C and 5 MPa is raised. The waste heat boiler and the reactor tube wall have a common forced circulation system. Some 90% of the solids in the gas are separated in the cyclone (11). The remainder of the solids are washed out with water in a series of scrubbers and separators (14, 15, 16) after the gas has been further cooled in two economizers (10). The gas leaving the scrubbers has a solid content of 1 mg/m^3 at s.t.p. and a temperature of 40 °C.

At present the solids in the circulating water used in the scrubbers are removed in a filter. In future they will be concentrated in a slurry, which will be reinjected upstream of the cyclone (11). The water will then evaporate and all solids leaving the waste heat boiler will eventually be separated in the cyclone, thus eliminating filtration. This system has already run very successfully in our 6 t/day pilot plant at our Amsterdam Laboratories.

Improvements

Apart from the recirculation of the solids containing water, the following improvements are currently being considered:

(1) recirculation of the solids from the cyclone back to the gasifier leading to the situation that in the end all ash entering the reactor leaves the system in the form of inert non-leaching slag;

(2) pressurizing the lock hoppers and transporting the coal with syngas instead of nitrogen, thus avoiding dilution of the gas with nitrogen;

(3) development of a non-cooled insulated brick-lined reactor;

(4) development of a water quench for applications in which a substantial part of the CO in the gas must be used to produce hydrogen by the shift reaction.

Removal of acid gases

The cooled gas, thus treated, still contains sulphur compounds, which have to be removed. The process used for their removal will depend on the particular application of the gas produced. Units for the removal of H_2S, COS and, if necessary, CO_2, employing various processes, can be hooked up to the Shell–Koppers gasification plant. Where the gas is to be used as fuel or in combined cycle electricity generation, the residual sulphur content must be brought down to whatever levels are demanded by environmental regulations for gaseous fuels.

Where the gas is to be used in gas turbines it is desirable not to remove CO_2, as its presence improves turbine power outputs.

Blast requirements, product gas and thermal efficiency

Oxygen and steam consumptions are dependent on coal feedstock quality. An oxygen demand of 0.9–1.0 t per tonne of moisture and ash-free (m.a.f.) coal is fairly typical of hard coals; for low-rank coals a figure of 0.8 t oxygen per tonne of m.a.f. coal is more representative. The steam requirement is very low and, in fact, is almost zero for some brown coals and in cases where air is used as a gasifying agent. For hard coal it is of the order of 8 % on m.a.f. coal.

The raw gas production is about 2000 m^3/t for a good-quality bituminous coal. The gas is relatively rich in carbon monoxide and the $CO:H_2$ ratio, on a volume basis, is typically between 1.8 and 2.2 for the preferred operation at minimum steam dosage and with recycle gas as a quenching medium. When water is used to quench the hot gases the $CO:H_2$ ratio of the gas will be somewhat lower.

The heat content of the gas is of the order of 11.3 MJ/m^3 at s.t.p. The thermal efficiency of the gasification proper is about 82 % for oxygen–steam gasification. Apart from this percentage of the total heat in the coal that is recovered as chemical heat in the gas, about 15 % of the heat in the coal to the gasifier becomes available for steam-raising in the waste heat boiler via the sensible heat in the gas.

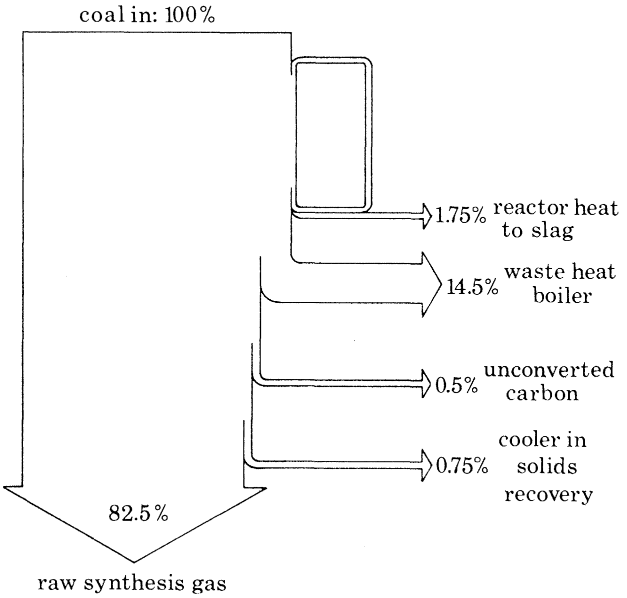

FIGURE 2. Shell–Koppers coal gasification process: typical heat balance of gasifier proper.

The total heat content of synthesis gas and steam represents approx. 94–97 % of the heat content of the coal feed (see figure 2). It is, however, more realistic to consider the overall thermal efficiency, taking into account the energy consumption of oxygen plant, coal mill and dryer, coal gasifier and auxiliary equipment. This overall thermal efficiency, calculated on a lower heating value basis, corresponds to approx. 78 % for a hard coal containing 12 % ash and 7 % moisture.

Coal quality

To demonstrate the versatility of the Shell/Koppers gasification process with regard to feed coal quality, the thermal efficiency has been calculated for a range of coals, namely: bituminous coal with moderate ash and moisture content (Illinois no. 6); sub-bituminous coal (Wyodak); coal liquefaction vacuum bottoms; brown coal, high moisture, low ash (German brown coal).

These feedstocks cover a fairly wide range of ash and moisture contents. The calorific values, correspondingly, range from 10 to 29 GJ/t as received coal (table 1).

Table 1. Shell–Koppers coal gasification: coal feed analyses

	Illinois no. 6 bituminous	Wyodak sub-bituminous	coal liquefaction vacuum bottoms	German brown coal
carbon (mass % m.a.f.)	78.1	75.6	87.1	67.5
hydrogen	5.5	6.0	5.7	5.0
oxygen	10.9	16.8	3.3	26.5
sulphur	4.3	0.9	2.4	0.5
nitrogen	1.2	0.7	1.5	0.5
ash (mass % as received)	12.0	5.9	17.6	6.4
moisture (mass % as received)	6.5	35.0	0	5.0
moisture (mass % of coal to gasifier)	2.0	2.0	0	5.0
lower heating value of coal as received/(GJ/t)	25.80	17.16	29.41	9.99

Table 2. Shell–Koppers coal gasification: coal, oxygen and steam requirements for different coals (tonnes per million cubic metres $CO + H_2$ produced (at s.t.p.))

constant plant capacity	Illinois no. 6 bituminous	Wyodak sub-bituminous	coal liquefaction vacuum bottoms	German brown coal
coal intake as received	573	810	494	1364
coal to gasifier	477	489	494	626
oxygen demand (vol. 99 % pure)	400	395	407	434
steam demand	36	12	86	20
thermal efficiency (%) (l.h.v. basis) gasifier proper	83	83	83	79
overall plant after subtraction of own consumption	78	77	77	72

Table 3. Shell–Koppers coal gasification: wet synthesis gas composition (percentages by volume)

component	Illinois no. 6 bituminous	Wyodak sub-bituminous	coal liquefactions vacuum bottoms	German brown coal
H_2O	1.5	2.6	2.1	11.3
H_2	31.6	32.5	33.6	26.9
CO	64.0	62.8	61.8	55.0
CO_2	0.8	1.3	1.0	6.1
CH_4	—	—	0.1	—
$H_2S + COS$	1.4	0.3	0.7	0.2
N_2	0.5	0.3	0.5	0.3
Ar	0.2	0.2	0.2	0.2

The data presented in table 2 are indicative of the oxygen and steam consumptions, and of the overall thermal efficiencies for the coal feedstocks specified. Assuming a constant production of synthesis gas ($CO + H_2$), it is evident that the amount of coal to be processed increases as the rank, and correspondingly the heating value, of the coal goes down and as moisture and ash contents go up. Variations in oxygen demand are generally well below 10% for the various feedstocks.

Some details of the gas composition are given in table 3. Hydrogen to carbon monoxide ratios are not materially affected by a change in coal feed composition. At CO_2 concentrations not exceeding 5 vol. %, as shown in table 3, the selective removal of H_2S does not normally present a problem. Typical CO_2 concentrations are below 2 vol. %.

Results of experiments in the 150 t/day pilot plant

The plant is at present in the trial phase in which it is aimed to demonstrate the feasibility of the various systems. So far the unit has run for a total of 300 h at pressures of 1.8–2.0 MPa. Difficulties have been experienced with the lock hopper system, but these have now been overcome. Further, there have been problems with fouling of the waste heat boiler. Operation of the coal transport and flow measuring system, the burners, the reactor and the quench have been very satisfactory.

Options for process utilization

It is a matter of process optimization to assess for each project the most economic combination of heat recovery, power generation and treating temperatures in accordance with the economic factors prevailing. An important alternative to the base case is air-blown gasification. This version is primarily of interest for use in situations where coal gasification is integrated with combined-cycle power generation. It could also be considered as a means of producing low-energy fuel gas for industrial applications. Some examples of Shell–Koppers process utilization are briefly reviewed below.

Production of CO-*rich gas for fuel gas, chemical synthesis and direct ore reduction*

Oxygen-blown gasification with the use of a gas quench (see figure 3 for flow scheme), coupled to a process for the removal of sulphur compounds and carbon dioxide, produces a dry gas typically consisting of some 30–34 vol. % H_2, 62–64 vol. % CO and a small volume percentage of inerts. The gas as such is very suitable for industrial heating. The use of coke as a feedstock and the use of CO_2 as a moderator instead of steam will result in gases with a CO content of 80 vol. % and higher.

Production of hydrogen or hydrogen-rich gas for ammonia, methanol and Fischer–Tropsch type synthesis

As shown in figure 4, the most common option is to use a water quench and to apply a CO-shift conversion with a sulphur-tolerant catalyst followed by selective H_2S and CO_2 removal.

Production of methane (*s.n.g.*)

The raw gas can be converted into methane by the same process route as described above for hydrogen-rich gas followed by methanation and drying. The diseconomy of the absence of

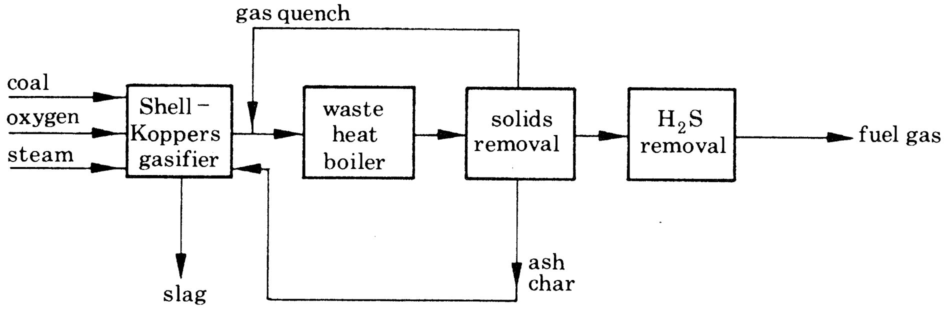

FIGURE 3. Shell–Koppers coal gasification process: typical block scheme for fuel gas production.

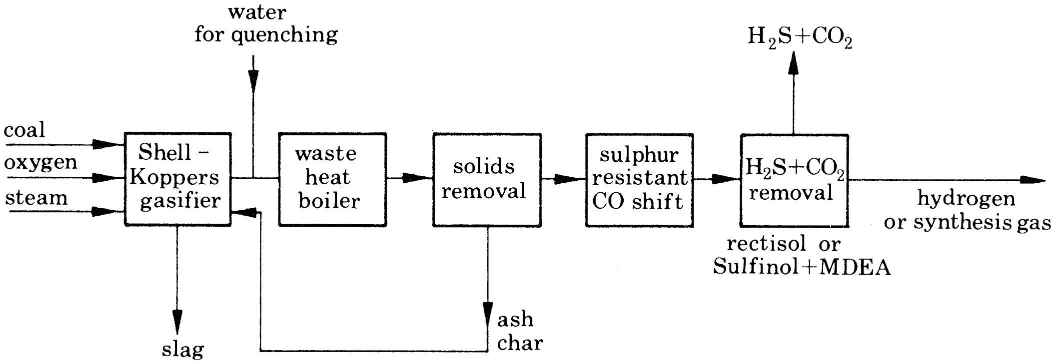

FIGURE 4. Shell–Koppers coal gasification process: typical block scheme for hydrogen or synthesis gas production.

methane in the raw gas, an ostensible drawback of the Shell–Koppers process in the manufacture of s.n.g., is largely outweighed by the relative simplicity of the processing of the gas.

Integration of the Shell–Koppers process with combined-cycle power generation

The integration of the Shell–Koppers coal gasification process and electricity generation in a gas-turbine–steam-turbine cycle offers an efficient and environmentally acceptable means of meeting future energy requirements.

The process lends itself to the full utilization of the high efficiency of gas turbines with high inlet temperatures, owing to the high proportion of electricity generated in the gas turbine, namely up to 60% in an advanced scheme (see, for example, figure 5). At the same time an efficient steam cycle can be maintained by the production of high-quality steam in the waste heat boiler of the gasifier.

The particulate-free pressurized fuel gas produced in the gasifier is a low–medium-energy fuel gas which gives a low production of NO_x in the gas turbine combustor.

Both the air-blown and the oxygen-blown versions of the Shell–Koppers process can be used at approximately the same station efficiency of 42–45% for gas turbines with inlet temperatures of about 1200 °C, which are expected to be available in the mid-1980s.

ECONOMICS

The investment for a gas plant to synthesize 36×10^6 m^3/day (at s.t.p.), based on U.S. location in mid-1979, is estimated at $793M. This figure applies to the processing of a hard

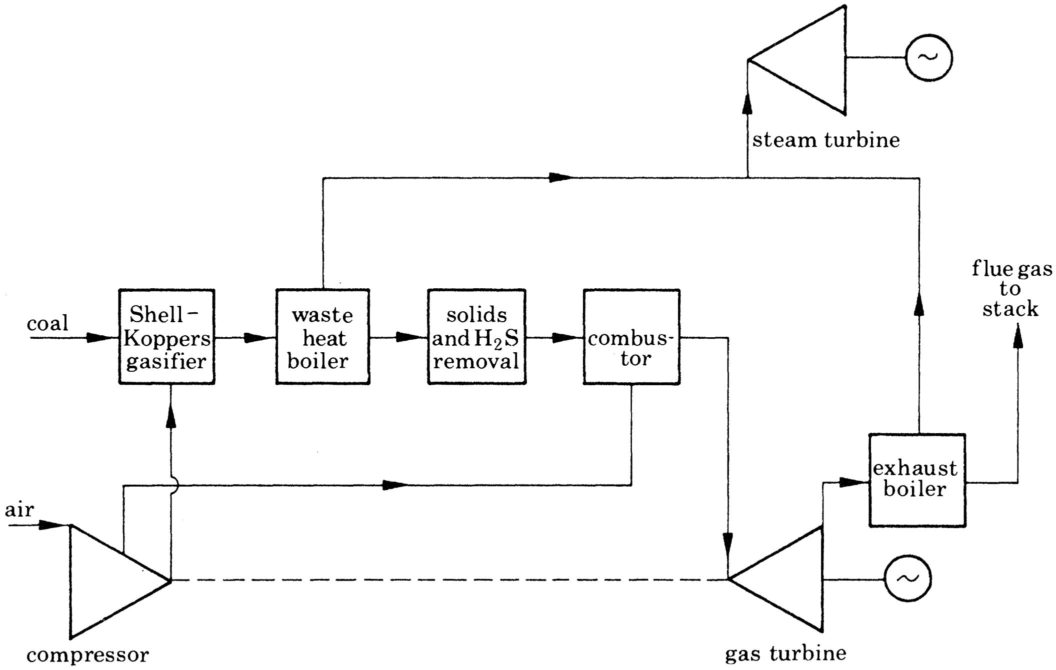

FIGURE 5. Shell-Koppers coal gasification process plant; combined cycle power.

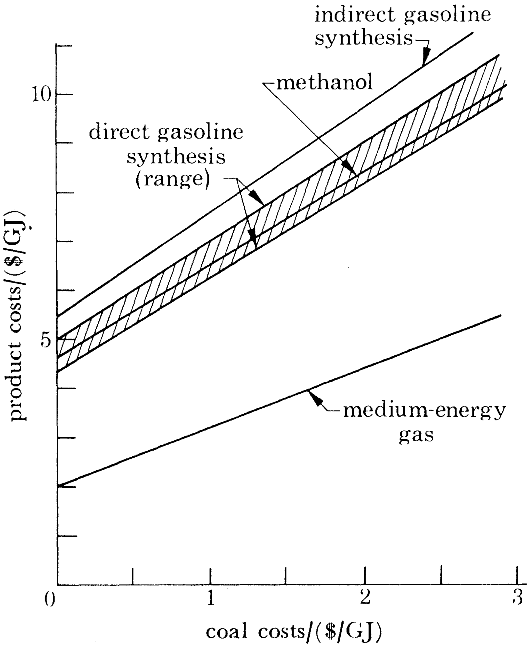

FIGURE 6. Shell-Koppers coal gasification: coal conversion costs; economics for various applications. Basis: Illinois, U.S.A.; dollar values at mid-1979.

coal containing 10% ash and 10% moisture, and includes the investment of coal handling and storage, coal mill and dryer, oxygen plant, water treatment, ash disposal facilities and off-sites. Excluded is the investment in plants for the further processing of the dry, particulate and sulphur-free synthesis gas. The corresponding cost of the synthesis gas before and after conversion into methyl fuel, and subsequently to gasoline on a heating value basis, is shown as a function of the coal feed unit cost in figure 6. For a low-cost coal feed, as would be available from open-cast mining of large coal reserves, the synthesis gas cost would amount to some \$2.8/GJ. For deep-mined coal, as available in Europe, the cost could be as high as \$4.7/GJ. The corresponding figures for methyl fuel are \$6.2 and \$9.0/GJ and for gasoline \$5.7–7.6 and \$8.5–10.4/GJ respectively. The economics of Shell–Koppers coal gasification are only marginally affected by variations in coal rank, ash content or moisture content. Only the costs of the coal mill and dryer and of the ash disposal facilities are significantly influenced, but these have a minor impact on the overall economics.

Discussion

J. H. Chesters (*Chairman of the Watt Commission on Energy*). The enthusiasm and clarity with which the last three papers have been presented makes the production of oil from coal seem rather easy, and free of serious development problems apart from scaling-up.

I know, however, that certain plants have experienced trouble owing to the abrasion of refractories by high-velocity dust-laden gases – sufficient indeed, to put certain sections out of commission in days rather than years. As the refractories were of the type successfully employed in catalytic crackers and were installed with great care, I should like to ask whether any of the speakers have experienced such troubles, and if so how they overcame them.

J. C. Hoogendoorn. In the Fischer–Tropsch process for production of liquid fuels from synthesis gas made by Lurgi pressure gasification, only minor use is made of refractories in some of the process steps (furnaces, etc.) but these are normal standard applications that pose no specific problems.

M. J. van der Burgt. At Shell, in the 150 t/day pilot plant in Harburg, Germany, no abrasion problems were experienced during 750 h of operation.