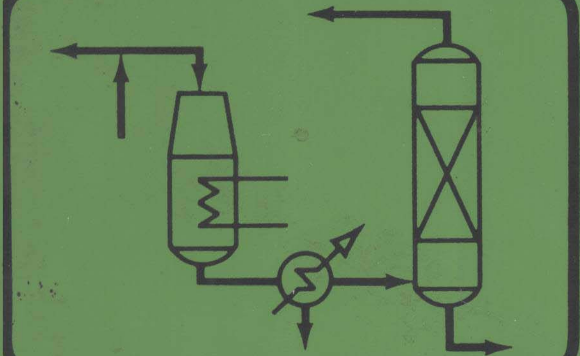
PROCESS TECHNOLOGY AND FLOWSHEETS



CHEMICAL ENGINEERING Magazine

PROCESS TECHNOLOGY AND FLOWSHEETS

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PROCESS TECHNOLOGY AND FLOWSHEETS

Introduction

The seventies have been a time of challenge for the chemical process industries. Government regulations, higher energy costs, environmental considerations, and waning hydrocarbon feedstocks have changed the ground rules of chemical production and chemical process design.

One positive aspect of this generally unpleasant situation, however, is that it has provided the world's chemical engineers with a unique opportunity for innovation. Indeed, one of the primary goals of CHEMICAL ENGINEERING'S editorial policy has been to track and report on developments of innovative technology: processes using unconventional feedstocks, techniques of turning wastes into dollars, and refinements designed to squeeze the most out of each Btu.

This book presents a selection of the most significant processes that have appeared in the pages of *CE* over the last five years. Most have been taken from the Process Technology section, while the remainder come from news and engineering feature-articles. For each article, the date of original publication is shown.

At the end of the book, we include a special section devoted to recent winners of *CE's* biennial Kirkpatrick Chemical Engineering Award. This award is given to developers of processes judged by a panel of prominent engineering educators to be the most significant additions to the body of chemical engineering technology.

Vincent Cavaseno

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Section I
COAL PROCESSING AND CONVERSION

Coal-based processes evaluated in Germany

A conference on coal conversion methods takes a look at the status of conventional technology and assesses the prospects of advanced gasification routes, including underground gasification, and two promising liquefaction processes.

☐ Coal specialists gathered in Düsseldorf, West Germany, this January under the aegis of the United Nations' Economic Commission for Europe to attend a four-day symposium aimed at reviewing the status of gasification and liquefaction processes, and evaluating coal's prospects as a rival of crude oil and natural gas.

Most experts agreed that gasification now has an edge because of the wide range of exising processes that are constantly being improved. Even discounting this technological lead, gasification is likely to dominate the picture on purely economic grounds in industrialized countries of high population density and declining natural-gas reserves. For example, in the U.S. and Western Europe, gasification will become firmly established within the next decade.

Speaking in general terms, prospects for liquefaction are not so immediately inviting, but many speakers at the symposium pinpointed this alternative as a logical complement or successor to oil refining over the medium term. In contrast, the probable role of gasification in the same period is to supplant dwindling reserves of natural gas.

Two options look especially apt for the developing countries with large coal reserves but small domestic demand: the coal can either be shipped elsewhere, or liquefied at home. The latter option, followed by hydrogenation to a relatively heavy fuel oil, seems the best way to obtain higher added value and return on resources.

Coalplexes—i.e., chemical complexes based on coal feedstock, and using both gasification and liquefaction techniques—received the endorsement of many experts in Düsseldorf. They stressed the fact that South Africa's Sasol project—a success despite much criticism from outsiders—points the way to a future coal-based chemical industry.

APPLYING PRESSURE—At Düsseldorf, conventional gasification technology was reviewed but the main emphasis was on development of second-generation processes in Europe and the U.S., using high-pressure operation to boost yields. In addition, a third generation of techniques, which use nuclear-process heat as energy input for plants, received plenty of attention.

The well-known Lurgi and Koppers-Totzek gasification methods are still undergoing further refinement. In the past decade, Lurgi has improved its pressurized units, increasing their capacities from about 15,000 to 80,000 m^3/h. The Koppers-Totzek route is being adapted from operation at "near atmospheric" pressure to functioning at 15–30 bars.

A Davy Powergas development, the high-temperature Winkler (HTW) process, is being modified by Rheinische Braunkohlenwerke (Cologne) and Aachen Technical University (Aachen). Tests carried out at 1.5, 3, 6 and 9 bars show that output goes up in proportion to the square root of absolute pressure, so investigators are trying a pressurized reactor and increased reaction temperatures.

The HTW route produces gas with low CO_2 and H_2O contents, suitable for use in direct reduction of iron ores. Preheated air at 850°C, or oxygen at 300°C, complement the reaction heat supplied by combustion of part of the coal. To prevent sintering of the ash at the temperature of the bed (950–1,100°C), lime or dolomite is added; this step also provides desulfurization.

MORE ON GASIFICATION—Rheinische Braunkohlenwerke is also working on what it calls the RO process, which employs a tube-furnace to handle lignite. Raw coal is fed to a fluidized bed, then gasified in the horizontal tubes of the reactor. These tubes are indirectly heated, and temperatures can be controlled to favor the gasification reactions, which take place in zones along the pipes. At pressures of up to 40 bars, the process yields gas with a methane content of 3.5% (wet basis). A 200-kg/h pilot plant that takes part of the product gas for process heating has been used for tests, but the German firm is now building another pilot unit of twice that capacity.

Attendants at the symposium also heard of a revival of interest in the Rummel-Otto slagging gasifier (ROG), developed and commercialized in the 1950s by Dr. C. Otto & Co. (Bochum, West Germany). The ROG is a three-stage vertically mounted reactor, with a slag bath in the bottom section. In this chamber, which is about 2 m high, all the fine particles of the feed are gasified.

Powdered coal and the gasification medium—a mixture of oxygen and steam—enter the first stage

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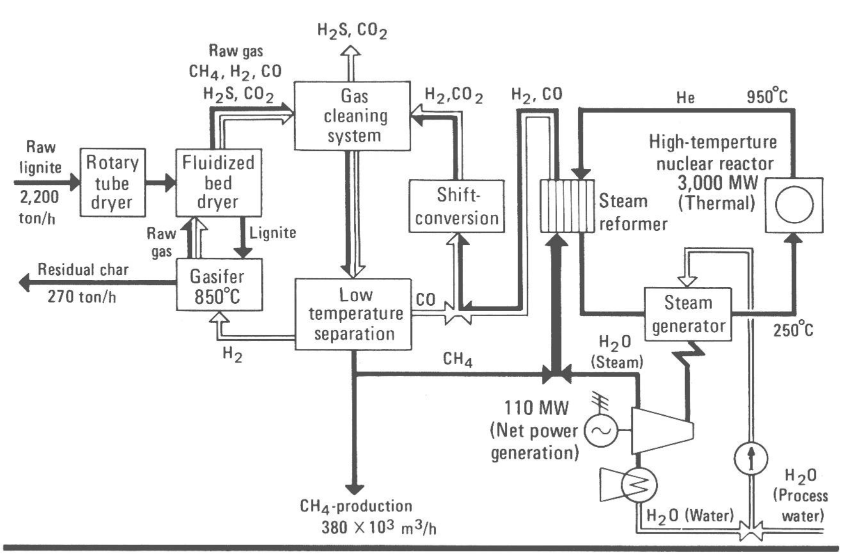
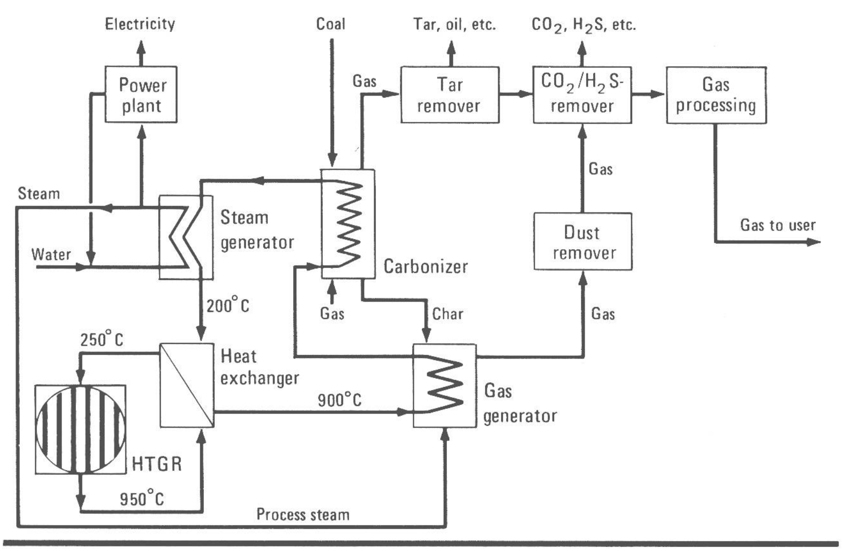


Hydrogasification couples nuclear heat with methane reforming **Fig. 1**



Plant combining HTGR and steam gasification of coal **Fig. 2**

through tangential tuyeres. A narrow throat connects this first stage with the second one (where entrained, fine coke particles are gasified), and as the rotating gas-stream (at 1,500–1,700°C) passes through it, large coke particles and slag droplets separate centrifugally and flow back to the slag bath.

In the third, uppermost, section, recycle gas or char is injected to quench the ascending gas stream and solidify ash droplets. Exit gas, at 800–900°C, carries 10–30% of the fresh coal-feed in the form of ash and coke, which are removed and recycled. The company plans to operate a new demonstration unit at 25 atm, producing 20,000 m^3/h of raw gas.

THIRD-GENERATION ROUTES— Hydrogasification and steam gasification, two third-generation techniques that use nuclear process heat, are also under study in West Germany. Their advantage lies in the promise of total conversion of the coal feed: Even in the advanced non-nuclear gasification plants, 30-40% of the feed is used to generate process heat, whereas the third-generation plants will pack virtually all of the coal's energy into the product gas. They depend, however, on the successful development of a high-temperature, gas-cooled nuclear reactor (HTGR).

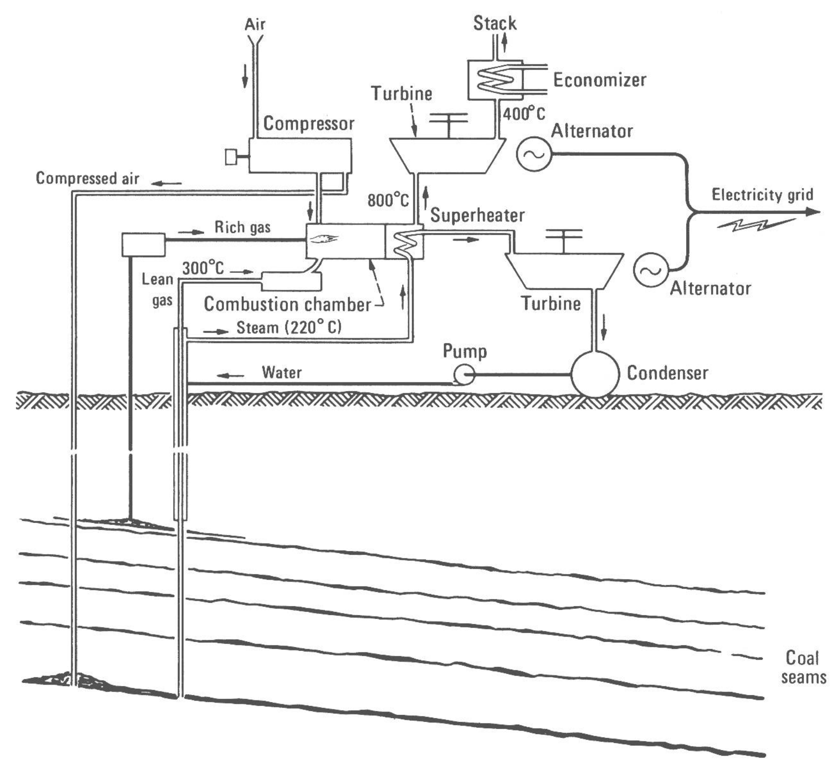
Both German projects envision a 3,000-MW (thermal) HTGR, with a high-temperature stream of helium as energy carrier. Rheinische Braunkohlenwerke is working on a hydrogasification process (Fig. 1). Predried hard coal or lignite is gasified under pressure with hydrogen in a fluidized bed to obtain a methane-rich raw gas. Unreacted hydrogen is separated and recycled; to provide the rest of the hydrogen requirements, part of the product methane goes to a steam reformer to produce CO and H_2. The HTGR's helium coolant provides the heat needed for this endothermic reaction.

In a higher-efficiency variant of the process, hydrogen for the methane-forming reaction is produced in the gasification reactor by injecting a preheated, steam-containing gasification stream. The residual, low-sulfur char that is left over can go to power stations.

Bergbau-Forschung GmbH, at Essen, is studying steam-based gasification. Ground coal is precarbonized and devolatilized (Fig. 2), and the resulting low-temperature char is gasified, using superheated steam. Nuclear process heat supplies the needed energy. The HTGR's primary helium loop is heat-exchanged with a secondary helium cycle, heating the latter to 900°C. This stream then feeds through indirect heater tubes in the gasifier, and through heater tubes in the pretreater. Whatever heat is left over in the secondary helium stream is used to raise steam.

LIQUEFACTION TECHNOLOGY— Two interesting possibilities were described by the research arm of Britain's National Coal Board (Cheltenham, U.K.). Although both have similarities to known solvent-refined coal (SRC) processes, the British entries differ in that they maintain separate extraction and hydrogenation stages, thereby producing a range of "cuts."

One technique employs anthracene oil as solvent in a continuous recycle loop. Crushed dried coal is slurried with the oil, then pumped to



INIEX scheme for high-pressure underground gasification Fig. 3

a digester operating at 370-450°C. Up to 85% of the coal dissolves; once filtered and solvent-stripped, the resulting liquid is fed into a catalytic hydrogenation reactor.

This unit operates at hydrogen partial pressures of 200-340 bars, and temperatures of 400-480°C. Gaseous and liquid phases separate at high temperatures; the gaseous phase is cooled, scrubbed at high pressure, and recycled with addition of makeup hydrogen. Light oils recovered from the aqueous liquor go to the fractionation stage along with the liquid fraction.

Fractionation yields light- and middle-oil products. Some of the middle and heavy oils are recycled as solvent.

Experts say this process may prove to be competitive with crude-oil refining for production of aromatic feedstocks, but there is still a great deal of development work to be done, especially on hydrogenation catalysts. This technique is also under study in other countries, including the Soviet Union and Poland.

The British coal board is looking at a liquefaction route that employs compressed gases at near-critical temperatures to obtain a coal extract. Coal-tar- or petroleum-naphtha-derived gases at about 100 atm and 300-400°C could typically extract up to one-third by weight of the coal; the low-melting-point product is recovered by reducing the exit gas stream's pressure.

An economic study by U.S. company Air Products & Chemicals—based on NCB data—shows that manufacturing costs for a 3-million-metric-ton/yr plant are comparable with those published for existing liquefaction processes. Capital costs, at 1974 prices, are estimated at $140 million. Solid residue from the process is said to be ideally suited for fluidized-bed combustion in power generation facilities

IN-SITU GASIFICATION—Among the most interesting fringe ideas reviewed in Düsseldorf was *in-situ* gasification of coal. P. Ledent, a director of Belgium's Institut National des Industries Extractives (INIEX), stated that it holds great promise, at least for deep coal-deposits. Although a number of low-pressure processes have been exploited, the high-pressure routes needed for deep deposits have yet to be proven.

Soviet technology is currently operational at several sites—one of them Angren, in Uzbekistan, 120 km southeast of Tashkent. The U.S. firm Texas Utilities Services, Inc. (Dallas), has already licensed the Soviet knowhow and will use it to gasify a deep lignite deposit.

Still, and despite ambitious plans in the late 1950s, Soviet underground gasification facilities produce energy equivalent to only 200,000 metric tons/yr of coal. Costs are high, so the gas must be consumed locally; prices are not competitive with either natural gas or conventionally mined lignite.

Two new, more-promising schemes originate in the U.S. One of them is under development at Morgantown, W.Va., by the U.S. Bureau of Mines. It uses new directional drilling techniques to exploit thin seams that contain low-volatile matter. The drilling method, which involves boring parallel holes across the seams, will serve to produce a longwall generator.

Deep deposits are also the target of a second U.S. project, under study by the Lawrence Livermore Laboratory (Livermore, Calif.). Explosives introduced through drill holes will hopefully fragment about 600 tons of coal per ton of explosive. After access holes fitted with casings are drilled at the top and bottom of the fragmented seam, a fire will be started at the top of the formation.

Gasification will proceed in a downward direction, aided by injection of a mixture of water and oxygen at 35-70 bars. The high pressure favors formation of CO_2 and methane; after removal of water vapor and CO_2, a high-quality pipeline gas results. U.S. deposits eligible for this type of treatment could supply 30 × 10^{13} m^3 of gas—about 300 times annual U.S. consumption.

EUROPEAN CONTRIBUTION—German researchers at Rheinische Braunkohlenwerke and Aachen Technical University have also studied high-pressure underground gasification. Their process involves periodic variation of gas pressure in the underground gas-generation zone, aimed at broadening the reaction area and promoting gas-solid reactions. Conditions (40-60 bars, 800°C) favor direct hydrogenation of carbon to methane; some of the methane product will be hydrated to generate hydrogen needed for the underground reaction.

INIEX has its own *in-situ* technique, which can also collect mine gas (mostly methane) released from upper seams during gasification of lower ones (Fig. 3). A network of holes is bored down to the lowest seam of a deposit, and then successive seams are worked in an upward direction. Gas recovery is aided by injecting air at high pressure, with a backpressure of 15–25 bars maintained at the gas-outlet bores.

INIEX envisages either a constant-pressure operation, or cycling between 30–50 bars and 15–25 bars. A power plant sited at the surface would be able to use lean gas and steam produced by cooling the exit bore-holes, as well as high-Btu gas (methane) to offset changes in the calorific value of the lean gas.

Ledent stresses the advantages of such high-pressure, deep-deposit ventures—e.g., seams at depths of 600–700 m can be exploited, and the hydrostatic pressure of overlying strata contains the gas and eliminates pollution risks. One major hurdle is the high cost of drilling, but this can be outweighed by the favorable factors.

In contrast, low-pressure *in-situ* gasification is useful only in thick, shallow seams that can be easily exploited by conventional mining. There are also environmental problems—up to 30% of the gas may leak, and underground water-sources may be contaminated.—PETER SAVAGE, *European Editor, London.*

Coal research: momentum builds in Germany, U.K.

Development of coal gasification and liquefaction processes is moving ahead rapidly in those countries. Several technologies will be demonstrated within five years.

☐ Efforts to develop new coal-utilization technology are hitting high gear in West Germany and the U.K., with about a dozen projects entering the pilot-plant or demonstration-plant stage. Benefits for the chemical process industries are promised by coal gasification and liquefaction, as these technologies should ease shortfalls of petroleum and natural gas that are likely to occur in the late 1990s.

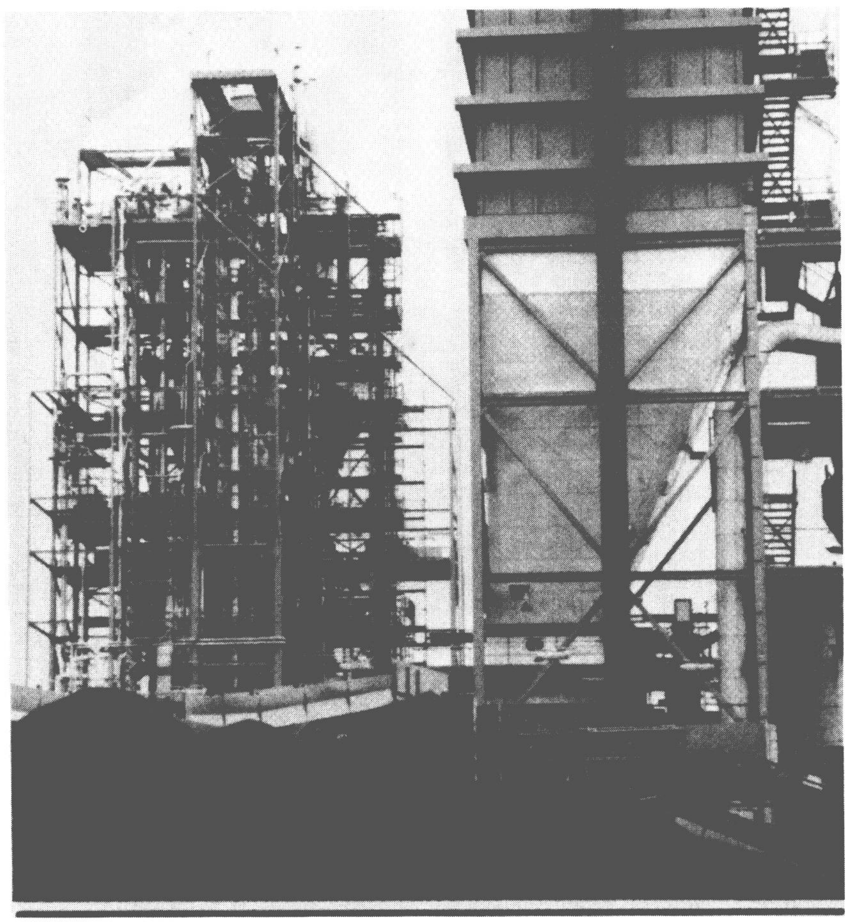
Because of the longterm nature of the development programs—most will not prove their worth before the mid-1980s—engineering contractors cannot expect an immediate bonanza. And experts throughout Europe are cautious about the prospects for individual processes, aware that in the U.S., at least, many processes have already fallen by the wayside.

Not surprisingly, most of the effort in development is centered in Western Europe's two largest coal-producing nations—the U.K. and West Germany. Research funds now come predominantly from two sources: national governments and the Paris-based International Energy Agency. But later this year, researchers may get additional substantial backing from the European Commission, which is planning a $100-million subsidy scheme for promising projects. (A final decision on the list of beneficiaries is not expected before December.)

GERMAN GASIFIERS—Nine weighty projects are underway in Germany, which originally developed a variety of coal gasification and synthesis techniques in the 1930s.

Government funding of the new German processes is relatively generous. The German Ministry for Research and Technology will put up 81% of the projected $62.5-million outlays for liquefaction and 71% of the planned $126 million to be spent on gasification between 1977 and 1980. The remainder will come from the International Energy Agency, the companies themselves, and possibly the European Commission.

The current German processes in the works range from straightforward improvements of established Lurgi-type gasifiers to sophisticated new



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systems that use nuclear process heat for gasifying coal (for more details of some of these coal-conversion processes, see Coal-Based Processes Evaluated in Germany, pp. 3–6). They include:

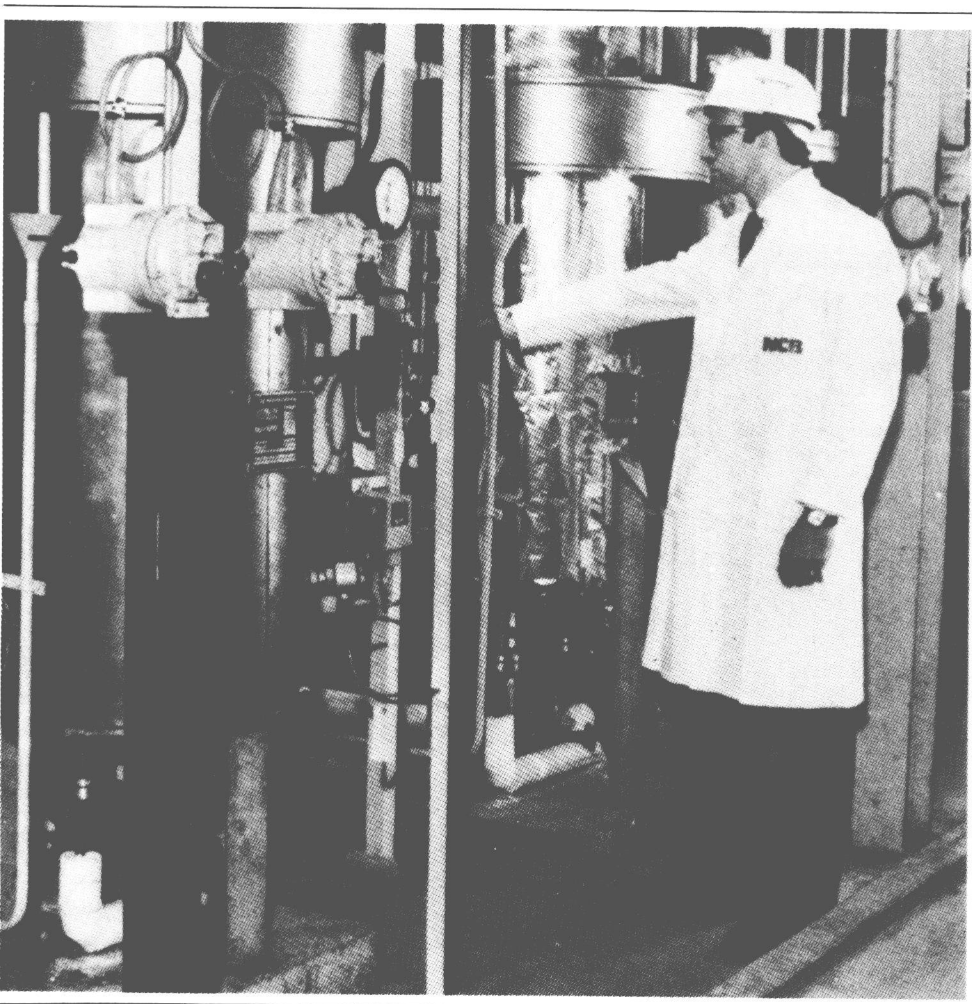
■ *A pressurized, high-temperature Winkler gasifier.* This fluidized-bed system, which operates at 1,100°C and 10 atm, has been onstream since early this year at a 1-mt/h pilot plant. The developer, Rheinische Braunkohlenwerke AG, now plans to spend the equivalent of $73.5 million on a 6-yr project to build a demonstration plant that will gasify 15 to 20 mt/h of coal. Construction will begin in 1980.

The process involves adding lime or dolomite to the reactor bed to prevent sintering of the ash at the high temperatures used. Besides, the additives desulfurize the reaction gases.

■ *A 100-atm version of a Lurgi gasifier.* A joint effort of Ruhrgas AG, Ruhrkohle AG and STEAG, an electric utility, this fixed-bed gasifier will be demonstrated in a 70- to 150-mt/d plant due onstream at Dorsten (in the Ruhr) next spring. Investment costs will exceed $50 million.

■ *An entrained-phase gasifier.* Ruhrkohle and Ruhrchemie AG are involved in a project to demonstrate Texaco's entrained-phase gasification process. A 6-mt/h plant will be started up next April, operating at 1,300 to 1,500°C and 70 atm.

The process uses a wide range of coals, and produces a synthesis gas that contains 30 to 40% hydrogen, 50 to 60% carbon monoxide, and less than 1% methane. Of the $15 million investment, the German government is contributing $9 million, Ruhrkohle about $4 million, and Ruhrchemie approximately $2 million.



British development uses supercritical gas extraction of coal

■ *A liquid-slag gasifier.* This method, which uses a liquid-slag bath in the base of the gasification vessel, will be tried out in a demonstration plant feeding on 10 mt/h of coal to turn out 20,000 m^3/h of gas. Cost of building the plant and for one year's operation is put at $25 million, 75% of which will be government-financed. At presstime, it was due onstream shortly at Voelklingen, where it will be run by process developers Saarbergwerke AG and Dr. C. Otto GmbH.

■ *An improved I.G. Farben process.* Saarbergwerke is also planning a pilot plant for coal hydrogenation, taking off from old I.G. Farben liquefaction technology. A 6-mt/h unit, scheduled to be commissioned at Voelklingen in late 1979, will produce a coal oil for further processing into chemical feedstocks and fuels. The pilot unit will cost $8.5 million to build, and $5 million to run in its first two years of operation.

Saarbergwerke is aiming to streamline the I.G. Farben process, which saw its best days during World War II, when it produced all of Germany's aviation and diesel fuel, and 50% of its vehicle gasoline. Saarbergwerke hopes to upgrade the technology by cutting the operating pressure to 300 atm and replacing a centrifuge step with vacuum distillation.

■ *A catalytic hydrogenation process.* Bergbau-Forschung GmbH (Essen) has built a 20-kg/h pilot plant for catalytic hydrogenation of coal, also based on established I.G. Farben technology. The unit operates at 300 atm and 480°C. Together with chemical firm Veba-Chemie AG, Bergbau-Forschung plans to have a $70-million, 200-mt/d demonstration plant onstream at Bottrop in 1980.

Both Bergbau-Forschung and Saarbergwerke admit that there is some overlap in their work. Saarbergwerke, in fact, is part owner of Bergbau-Forschung. The reason that both groups are pressing on with their similar developments is that they have different patrons: the State of North Rhine Westphalia is funding Bergbau-Forschung's venture and the Bonn government, 75% of Saarbergwerke's.

■ *An advanced Koppers-Totzek gasifier.* Developers Deutsche Shell AG and Krupp-Koppers GmbH will bring onstream a 6-mt/h demonstration plant at Hamburg this autumn