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COAL GASIFICATION AND LIQUEFACTION

T.D. BROWN
COAL RESOURCE AND PROCESSING LABORATORY

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by

T.D. Brown*

ABSTRACT

A review of the current status of gasification technology shows that many engineering systems are immediately available for use in the gasification of coal. Widespread international experience can be brought to bear on the atmospheric pressure gasification of all Canadian coals.

Unless a high ash content dictates the use of a fluid bed gasifier, it appears that bituminous coals can be used in fixed bed systems and that the strongly caking coals are most suited to the high pressure entrained bed systems. Existing and developing technologies in fluid bed gasification are attractive for reactive lignites and subbituminous coals.

The Fischer-Tropsch based process is the only commercial embodiment of coal liquefaction; the use of Canadian coals in this process depends only on their gasification characteristics. Current developments in hydroliquefaction are directed towards carboniferous coals of medium or high rank. It is not clear that cretaceous coals of the same rank will give similar process yields and economics.

Manager, Coal Resource and Processing Laboratory, Energy Research Laboratories, Canada Centre for Mineral and Energy Technology (CANMET), Department of Energy, Mines and Resources, Ottawa, Canada.

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INTRODUCTION

The twin coal conversion processes of gasification and liquefaction have a history which dates back beyond the current energy shortages of the western world. Indeed liquids and gas from coal were produced before this century.

Coal gas produced from a coal carbonization process was originated before 1800, and the development of gasification of non-coking coals was patented by Siemens in 1860. This patent represented the first use of air and steam together in the gas-making process. The pioneering development of coal gasification using oxygen and steam was demonstrated by Lurgi in the late 1920's. This historical sequence was all carried out at atmospheric pressure, and it was not until the immediate pre-war (1939/45) years that operation of the coal gasification process at elevated pressure became a reality.

The development of coal liquefaction processes has an analogous history to gasification but has not yet reached the stage of development where several processes can be considered to be commercially developed.

In liquefaction it is important to recognize the role of coal tar fuels which have provided a background of technology since the early 1900's. These liquids were not, however, the prime product of either the coke ovens or the towns gas industry which they supported. Many millions of tons of crude tar have been refined to produce a range of "coal tar fuels" with viscosities similar to that produced in the petroleum refining industry.

The development of the direct liquefaction process has its origins in the experimental work of Bergius (1924). This original pilot plant used a two-stage process in which the coal was mixed (and perhaps partially dissolved) in a liquid hydrocarbon prior to reaction with hydrogen at elevated temperature and pressure. The process was expanded in subsequent years until, in 1939, the German capacity was approximately 1.4 million tons of coal-derived-oil per annum.

The most recent landmark (1957) development in coal conversion is the combination of the twin technologies of gasification and liquefaction by the South African Coal, Oil and Gas Corporation (SASOL). In this process a synthesis gas ($CO + H_2$) is reacted over an iron oxide based promoter/catalyst to produce a mixture of gaseous and liquid paraffins and olefins. The process has now accumulated an operating record of 23 years and must be regarded as a commercial technology.

COAL TO GAS: THE CONVERSION ROUTES

The three process options that can be used to produce a fuel gas(1) from coal are shown in Figure 1.

1. Direct Gasification ⟶ Route 1.
2. Hydro Gasification ⟶ Route 2.
3. Pyrolysis ⟶ Route 3.

Direct gasification is based on the production of carbon monoxide and hydrogen by the reaction

$$C + H_2O \longrightarrow CO + H_2 .$$

This reaction is endothermic, and the required heat is usually supplied by the combustion reaction of the coal

$$C + O_2 \longrightarrow CO_2 .$$

The oxygen which drives the thermal reactions throughout this gasification route may be supplied as air or as elemental oxygen. The typical product from the "air-blown" gasifier contains 50% nitrogen and has a calorific value between 100 and 200 Btu/scf; the typical product from an oxygen blown gasifier has a calorific value between 300 and 450 Btu/scf.

The gaseous product can be catalytically reacted to produce a synthetic natural gas of high calorific value (1000 Btu/scf).

$$CO + 3H_2 \xrightarrow[Ni]{} H_2O + CH_4$$

Direct hydrogenation (Route 2 in Figure 1) is represented by the reaction

$$C + 2H_2 \longrightarrow CH_4$$

and is carried out at pressures above 500 psi. The advantage of this route is that it can present a single stage process giving an end product with a calorific value of about 800 Btu/scf.

The pyrolysis process (Route 3 in Figure 1) is analogous to the gas production which occurs in cokemaking. Temperatures are controlled below about 600°F to maximize the gas yield.

All three gasification routes have common gas-cleaning requirements. The hot product requires cooling with heat recovery, and all tars and liquid phase products must be removed for refining or recycling. Ammonia, sulphur gases and particulate material must be removed before the raw gas can be considered as a fuel or feedstock for a secondary process.

The potential application of coal gasification in Canada appears to be as a medium or low Btu fuel gas in specialized applications such as combined cycle power generation. The current development of increasingly stringent environmental control on sulphur emissions will encourage the use of gasification in the power industry. The raw gas must necessarily be stripped of its sulphurous impurities. The advent of turbine inlet temperatures above 1200°C postulated before 1985 allows the combined cycle to offer environmental and efficiency advantages over the conventional steam cycle equipped with flue gas desulphurization.

A second specific application may occur in the gasification of coal to produce hydrogen for the upgrading of bitumen from tar sand and heavy oils. Currently the production of hydrogen by natural gas reforming has economic advantages. However, if the oil extraction process requires a high degree of flexibility in the location of its steam or gas flooding sources, then a total utility supply by coal gasification may become attractive. This offers the ability to minimize the use of the more valuable liquid product and natural gas throughout the extraction and refining plant.

These potential uses in Canada focus on the direct gasification route using air and oxygen blown processes. The engineering systems that have been developed for these processes are discussed below:

COAL TO GAS: THE GASIFIERS

Coal gasification technology, whether under development or at the commercial scale, can be categorized according to its operating pressure, its gasifying medium or the type of process reactor. Figure 2 shows such a classification and illustrates the number of systems currently under active development.(2) Many processes can be classified under different headings. In particular those processes which developers feel have particular merit in combined cycle applications will progress through air-blown gasification to the oxygen blown system.

Figure 3 illustrates schematically three main categories of gasifiers. (1)

In a fixed bed gasifier coal is introduced to the bulk charge via a lock hopper system in a lump size commonly around 50 mm. The coal is preferably closely sized, and briquetted coal fines have been widely used in gasifiers of this type. The coal charge travels vertically downwards through the successive gasifier zones of devolatilization, endothermic gasification, exothermic combustion and dry ash discharge. These systems are characterized by large coal inventories, long coal residence times, low exit gas temperatures, high carbon conversion, and a high tar content in the product gas. This tar can be recycled into the gasifier. The carryover of particulate material is not high unless the feed coal is mechanically weak or contains excessive fines. Fixed bed gasifiers do not handle caking coals easily when special provision for pretreatment (oxidation) or mechanical charge agitation must be made. Difficulties can be experienced when using coals with low ash fusion temperatures, when excessive steam may be required to control slag formation.

These gasifiers have been used in the oxygen blown mode to give a molten slag discharge. This has the additional benefit of reducing the tar carry-over.

Fluid bed gasifiers operate with a closely sized crushed coal of 1-5 mm diameter and show the typical bed features of mixing uniformity and good gas-solid contact. They are normally operated at low temperature in a non-agglomerating mode. Higher temperature operation is possible with certain coals when controlled ash agglomeration may occur. Preheating of caking coals may be necessary with these systems to avoid defluidization. These gasifiers normally produce higher dust loadings and lower tar loadings in the product gas than do fixed bed systems.

Entrained bed gasifiers are analogous to pulverized coal fired boilers in many ways. The coal size distribution, coal preparation and injection are closely similar in the atmospheric pressure units. The furnace configuration and the overall air to fuel stoichiometry are the main differences. Coal residence time in these gasifiers is short (below 10 seconds). These units operate at much higher temperatures than fixed and fluid-bed gasifiers, and the resultant tar production is very much less. Inevitably, however, this type of gasifier gives a product with a high particulate loading.

The coal inventory of entrained bed gasifiers is small. Short term variations in coal quality and feed rate can cause major swings in gas composition. For this reason at least one process developer has chosen to use a coal-in-water slurry as the feed fluid for a pressurized, entrained bed gasifier to facilitate fuel feed control.

In molten bath gasifiers the coal (1 mm) is injected onto or below the surface of a molten pool of slag, iron or sodium carbonate along with the gasifying medium. Gasification occurs within the pool, and ash particles are not carried over in the product gas. When molten Na_2CO_3 or Fe is used, then a measure of sulphur removal occurs within the reactor.

The following sections present descriptions of representative engineering embodiments of the categories of gasifier:

THE WELLMAN-GALUSHA FIXED BED GASIFIER

This atmospheric pressure gasifier (illustrated in Figure 4) embodies some characteristic features of the fixed bed units.

A spreader-agitator is incorporated within the gasification reactor to retard blast channelling and to maintain a uniform fuel bed. The revolving grate is mounted eccentrically at the bottom of the gasifier on a post where it serves as a distributor for the air-steam or oxygen-steam blast. It also forces the ash into the ash pit.

The gasification chamber, the central distributor and the rotating grate are water cooled.

A recent study of the potential application of small gasifiers of this type to the gasification of Saskatchewan lignite has been carried out to develop costs for four qualities of gas:(3)

1. A low heating value raw gas.

   This considered an air/steam blown gasifier supplying an uncleaned gas for direct use.

2. A medium heating value raw gas.

   This considered an oxygen/steam blown gasifier supplying an uncleaned gas for direct use.

3. A low heating value clean, cold gas.

   As for (1.) above but with a clean product for transmission.

4. A medium heating value clean cold gas.

   As for (2.) above but with a clean product for transmission.

The cost performance characteristics for the Wellman-Galusha system (January 1979 dollars) are summarized in Figures 5, 6 and 7.

THE LURGI FIXED BED GASIFIER

The modern Lurgi gasifier, illustrated in Figure 8, incorporates the following features:

1. A central gasifier chamber up to 5 m in diameter.
2. Automatic coal feed at pressures up to 20 atm.

3. Continuous coal distribution and levelling at the top of the coal bed.

4. Continuous "in-bed" agitation to minimize agglomeration effects when caking coals are used.

5. Steam and water cooling on both reactor vessel walls and the rotating grate.

Gasification plants have been constructed incorporating over 60 individual gasifiers. The applications range from combined cycle power(4) production at Lunen, West Germany to the indirect coal liquefaction plant at Sasolburg, South Africa. A wide range of coals are currently being gasified using both air-steam and oxygen-steam as the gasifying medium.

A series of experiments carried out by the British Gas Corporation on a commercial Lurgi plant at Westfield have demonstrated the ability of the Lurgi system to be operated in a slagging mode.(5) Operation in this mode increases operating temperatures - and hence reaction rates. This increases throughput as well as reducing the cooling steam requirement.

The comparative gas composition from the two modes of operation are shown in Table 1.

Table 1 - Product composition from dry and slagging Lurgi gasifiers

Crude Gas Composition	Lurgi Dry	Lurgi Slagging
vol %		
CO_2	24.6	2.6
CO	24.6	60.6
H_2	39.8	27.8
CH_4	8.7	7.6
CnHm	1.1	0.4
N_2	1.2	1.0
CV (MJ/Nm^3)	12.4	14.8

THE KOPPERS-TOTZEK ENTRAINED BED GASIFIER

This atmospheric pressure gasifier is fed with a dried pulverized coal matching the specifications for a pulverized coal fired boiler (70% through 200 mesh). The gasifier has the cruciform shape illustrated in Figure 9; the coal, oxygen and steam are introduced through coaxial supply systems on the two opposed burners.

The coal is completely gasified within a one second reaction time and half of the coal ash removes by gravity as a molten slag. The gasifier temperature is controlled by the oxygen: coal ratio and the steam to coal ratio. The steam requirement is normally held down, and the gasifier can be regarded as a partial combustion system. The gaseous product corresponds closely to the equilibrium composition for the water gas equilibrium reaction.

The two-headed system illustrated can be expanded to a four headed unit when throughputs above 20,000 m^3/hr are required.

Currently over 50 units of this type are in use around the world handling fuels including naphtha, coal slurries, peat, lignite and bituminous coals. The majority (85%) of the world' coal based ammonia plants use Koppers-Totzek gasifiers.

THE SHELL KOPPERS ENTRAINED BED GASIFIER

Collaboration between Krupp-Koppers and the Shell International Petroleum Company has led to the construction and operation of a pressurized, oxygen blown (20-30 atm) version of the Koppers-Totzek System. This prototype is currently on demonstration in the Harburg (West Germany) refinery at a fuel rate of 150 tons/day.(6)

The basic configuration of the demonstration unit is similar to that of the atmospheric pressure unit. A major development program has been focussed on the coal feed and injection systems to produce a satisfactory controllable performance.

This gasifier has been developed to accommodate all solid fuels including petroleum coke and will accept coals with ash contents up to 40% and sulphur contents up to 8%. In most cases moisture levels in the feed coal are controlled below 8% to maintain gas quality and minimize oxygen consumption.

Typical performance characteristics for this demonstration gasifier are summarized in Table 2.

Table 2 - Performance of the SHELL-KOPPERS coal gasifier

	Bituminous Coals West Germany		Lignite	Brown Coal Australia
Coal				
Ash, %	9.0	27.4	6.0	1.2
Moisture, %	10.0	10.0	35.0	50.0
Carbon, %	66.5	51.4	44.6	33.0
Hydrogen, %	4.3	3.3	3.5	2.3
Sulphur, %	1.1	0.9	0.4	0.1
Oxygen	8.0	6.2	9.9	13.1
L.C.V. (k cal/kg)	6300	4860	4100	2680
Dry Product Gas Composition				
Hydrogen	31.3	30.2	30.1	28.6
Carbon Monoxide	65.6	66.5	66.1	65.8
Carbon Dioxide	1.5	1.8	2.5	4.7
Methane	0.4	0.3	0.4	0.1
Hydrogen Sulphide	0.4	0.4	0.2	0.1
Nitrogen	0.6	0.6	0.5	0.5
Argon	0.2	0.2	0.2	0.2
Relative Performance				
Coal	1	1.32	1.52	2.32
Oxygen	1	1.02	1.05	1.25
Steam	1	0.5	.20	–
Thermal Efficiency, %	77	74	77	76

THE WINKLER FLUID BED GASIFIER

This atmospheric or low pressure gasifier, shown in Figure 10, has been in common use on a commercial scale (up to 75,000 m^3/hr) since the 1920's.

A screw feed carries a crushed coal feed (<1 cm) into the bed of a system fluidized by the blast medium. The preheated blast maintains bed temperature during reaction between 1100° and 1300°K depending on the nature of the coal. As with all fluid bed systems the ash fusion characteristics of the feed coal and the active bed temperatures must be closely supervised. The high blast velocity and extensive coal surface area exposed result in a significant increase in specific gasification rate relative to fixed bed systems.

A major proportion (60%) of the fuel ash is elutriated from the bed in a semi-molten state and is quenched in a downstream radiant boiler. The remaining ash sinters and sinks through the bed onto the bed plate for removal by a skimmer into the ash removal system. In many cases a secondary over-bed blast is introduced to ensure reaction of the carbon fines elutriated from the fluid bed.

The unit has a successful record with subbituminous and lignite coals. As described here, it does not offer a totally satisfactory performance with bituminous coals and anthracites. It has, however, recently been modified to offer operation at elevated pressure.

THE CONOCO CO_2 ACCEPTOR PROCESS

This recent development of the fluid bed approach to coal gasification has been developed to the 40 ton per day level.(7) This process differs from those described previously in that the source of heat used to sustain the carbon-steam gasification reaction is

$$CaO + CO_2 \longrightarrow CaCO_3.$$

This reaction is between hot lime injected into the fluid bed and CO_2 derived from the water-gas shift reaction.

Calcium carbonate is regenerated in a separate air blown fluid bed and is recycled to the gasification reactor as shown in Figure 11.

Data derived from the pilot-scale unit indicated that the process is more efficient than the conventional Lurgi process and has a significantly lower water consumption. However, it enjoys its most efficient operation when used with lignites and subbituminous coals.

THE RUMMEL-OTTO SLAG BATH GASIFIER

In this system the endothermic gasification reactions take place in the gas space above a slag bath which provides the heat to drive the reaction.

The feed coal and gasifying media are injected through concentric nozzles inclined downwards and tangentially over a molten slag bath. Large coal particles are provided with a long residence time in the bath to ensure reaction; smaller particles react completely in the freeboard space above the bath. Excess slag is removed via a central tapping hole; Figure 12.

The gasifier shows a high particulate carry-over (10-30% of input) recessitating provision for recycling all fly-ash through the reactor. Current development calls for continued demonstration of a 250 tons/day unit.(8)

COAL REACTIVITY DURING GASIFICATION

The rate of gasification in any one of the previously described gasifiers will depend on the temperature, pressure, gas composition and the type of coal. These factors also affect the composition of the end product. The major effects in all gasifiers can be summarized:

1. High steam to carbon ratios favours increased hydrogen content in the product gas.
2. Increased temperatures reduce methane content in the product gas.
3. Increased pressures give increased methane content in the product gas.

In terms of coal reactivity, lignites and subbituminous coals show higher reaction rates than bituminous coals as is exemplified by the temperatures at which their respective gasification rates approach zero:

Lignite:	1150°F
Sub-bituminous:	1300°F
Semi Anthracite:	1400°F
Coke:	1500°F

This effect has been studied in some detail for both U.S. and Canadian coals as they might be used in a fluid bed gasifier.

After initial devolatilization, the rate of gasification of a char residue can be described by the equation:(9)

$$\frac{dX}{dt} = f_1.K.(1-X)^{\frac{2}{3}} \exp(-aX^2)$$

K is the sum of rate constants for the gasification reactions considered.

X is the fractional conversion of base carbon.

a is a kinetic factor dependent on gas composition and pressure

f_1 is a relative reactivity factor dependent on the rank of the coal.

Calculations for Canadian coals indicate that the reactivity factor can vary by a factor of 3 (between f = 0.5 and f = 1.5) as is illustrated in Fig. 13. When the overall gasification expression is considered for a range of B.C. coals the results obtained in Figures 14 and 15 are obtained.

Figure 14 shows that the rate of conversion of coal to gas reduces with increasing carbon content in the coal, and Figure 15 shows that for the case of the gasification reaction

$$C + H_2O \longrightarrow CO + H_2$$

the major gains in gasification rate due to increases in pressure are achieved by operating at pressures up to 20 atmospheres.

LIGNITE PETROGRAPHY AND GASIFICATION

It has been established in the steel industry that the relative concentration of coal macerals in bituminous coals can be used to predict the quality of coke produced during their carbonization in slot-type ovens. Similarly, recent investigations of the gasification of Saskatchewan lignites have shown that certain lignite macerals are preferentially reacted.(10)

It has been observed that the structured huminite and liptinite macerals react almost completely under most conditions representative of the Lurgi process. Inert macerals remain relatively unchanged whereas the unstructured macerals react in a manner responsive to reaction conditions. It can therefore be postulated that the gasification reactivity of lignites responds directly to the ratio:

$$\frac{\text{Structured huminite} + \text{liptimite}}{(\text{Unstructured} + \text{inert})\ \text{macerals}}$$

Current research is attempting to quantify this postulated relationship.

COAL LIQUEFACTION: THE PROCESSES

The basic coal liquefaction technologies fall into two categories - the indirect and direct routes.

In the indirect route an initial gasification step precedes the catalytic Fischer-Tropsch synthesis of a liquid product. The character of the liquid product can be varied across a specific range depending on the nature of the catalyst used and the reaction conditions in the synthesis step. This process, although thermodynamically unattractive, is the liquefaction process that is in current use on a major industrial scale at the SASOL plant in South Africa.

The two direct routes are hydroliquefaction and pyrolysis.

In hydroliquefaction the coal is reacted at elevated pressure and temperature with hydrogen. The coal preparation stage may include slurry production using a hydrogen donor as the carrier. This solvent can be a process derived liquid or a recoverable organic material. In some versions of the process the coal is dissolved directly (or perhaps under mild hydrogenation) and the liquid product upgraded in a separate, more vigorous hydrogenation process. In other versions the solution and hydrogenation occur simultaneously.

The coal pyrolysis route is one which is designed to "skim off" the volatile content of a coal to give liquid products leaving a char which may serve as a feedstock for a gasification process.

The process is currently undergoing development to incorporate extremely rapid heating of the coal in an atmosphere of hydrogen. This maximises the yield of volatile products.

The process descriptions that appear in the following sections of this paper have been selected as the four which are the closest to commercial operation rather than those which have any particular merit for use with Canadian coals.

INDIRECT LIQUEFACTION - THE SASOL PLANT

The SASOL I plant was built in the early 1950's and is based on Lurgi oxygen blown gasifiers feeding a product gas to two different Fischer-Tropsch synthesis units for liquid production.

The process route is defined in Figure 16 and the coal feed and product stream are shown in Table 3.

Two radically different synthesis procedures are used at Sasol I. The first installed unit was the Arge fixed catalyst bed reactor, Figure 17. In this vessel there are 2000 catalyst tubes filled with 14 m^3 of catalyst. The purified gas is top fed with product extraction from the reactor base, and condensed liquids are collected in a hot catch pot. With fresh catalyst, the product distribution curve peaks nearer the wax range. As the catalyst ages, the product range shifts down in molecular weight.

The essential features of the Synthol fluidized catalyst bed Fischer-Tropsch reactor is shown in Fig. 18. The main advantage of this system is the short contact time for reactant gases/liquids/catalyst in the hot reaction zone. Thus, chain growth is limited and the product spectrum is shifted to lower molecular weight compared to Arge liquids. However, the fluid bed product is much less saturated in both the diesel and gasoline fractions.

Table 3 - Characteristics of the SASOL I plant

Feed Coal	Composition wt %
Moisture (a.r.)	11
V.M. (dry)	22
Ash (dry)	36
S (dry)	0.5
N "	1
C "	51
H "	3
O "	8.5

Annual Production	Tons/year
Gasoline and Diesel Fuel	1,500,000
Ethylene	160,000
Chemicals	50,000
Tar	200,000
Ammonia	100,000
Sulphur	90,000
TOTAL	2,100,000

Comparison of Gasoline and Diesel Fractions

	Arge Fixed Bed Reactor		Synthol Fluid Bed Reactor	
	$C_5 - C_{12}$	$C_{13} - C_{18}$	$C_5 - C_{10}$	$C_{11} - C_{14}$
Paraffins	52	63	14	14
Olefins	41	27	69	61
Aromatics	0	0	5	14
Alcohols	7	7	6	5
Carbonyls	1	1	5	5
n-Paraffins	94	95	56	61

The Fischer-Tropsch reaction system is affected by poor gas quality. Hence, CO_2, H_2S and organic sulphides must be removed after gasification in the rectosol and phenosalvaine units. The CO_2 affects the reaction kinetics while H_2S and organic sulphides poison the catalysts. There have been no corrosion problems on the gas treatment stream. One waste heat boiler operated 20 years before failure.

In the Rectosol units, it helps to have sufficient basic nitrogen compounds from the coal. Otherwise the solution becomes too acidic and corrosion increases. Normally Rectosol requires maintenance every two years.

Rectosol is a cryogenic procedure using three stages of methanol extraction: (1) at -40°C, a prewash for aromatics, oils, HCN, organic sulphides and naphtha; (2) at -70°C, the main wash for CO_2 and some H_2S; and (3) at -40°C for the remaining H_2S.

For older coals, the rectosol process removes 90% of monosubstituted phenols and 10% of the less extractable polysubstituted phenols. For young coals, the figures are 100% and 95% respectively.

Unrecovered phenols pass to natural or innoculated biodegradation units.

At Sasol II a different cryogenic system will be used in order to recover primary ethylene which has a prime value.

Although this operational plant is based on Lurgi gasification, it is clear that any alternative gasification process could be used to feed the Fischer-Tropsch reactors. Selection depends only on the characteristics of the coal to be gasified.

SOLVENT-REFINED COAL - SRC-PROCESS

A schematic diagram of this process (S.R.C.-I) is shown in Figure 19. Pulverized coal is mixed with a process derived solvent in a slurry tank whence it travels via a hydrogen injector and preheater to the "dissolver" or reaction vessel. Under the reaction conditions shown in Figure 19, (450°C; 115 atm) over 90% of the carbonaceous material is taken into solution in a residence time of 30 minutes.(11)

The hydrogenation reaction in the dissolver leads to depolymerisation of the coal molecules to give a product of reduced molecular weight and higher solubility. The carrier solvent is simultaneously hydrocracked to give volatile hydrocarbons, methane and hydrogen sulphide.

After gas separation the liquid coal solution is separated from the undissolved solids in a filtration unit and the carrier solvent is thermally recovered. The solvent refined coal residue is a solid below about 175°C.

A modification of the process (SRC-II) recycles dissolver slurry product to the coal feed and increases both reactor residence time and pressure. The product is a liquid distillate fuel with a boiling range 175-450°C.

Work to date has focussed on bituminous coals with the reference coals being Illinois No. 6 and a Pittsburgh seam coal. It appears that this process will not accept high ash coals without difficulty since the filtration separation stage has a record of operational problems that have only recently been totally overcome.

THE H-COAL PROCESS

This catalytic hydroliquefaction process is illustrated schematically in Figure 20. It has been developed for conversion of high sulphur bituminous coals to provide a synthetic crude oil for further processing.(11)

Coal slurry (water free) is prepared using a coal derived solvent and premixed with hydrogen at 200 atmospheres; the product is preheated and charged to an ebulliated-bed catalytic reactor. Catalyst is periodically removed and replenished. The vapour product from the reactor is separated and/or condensed. The hydrogen rich gas product is recycled. The liquid/solid product from the reactor is flash separated and the light end distilled. The heavy bottom product may be further refined.

The operating conditions within the process can be varied to provide a degree of "tailoring" of the product. To produce a synthetic crude oil, more hydrogen is required; to produce a clean distillate fuel, the reactor temperature and pressure are lowered and the yield of residual fuel simultaneously rises. The process requires between 14,000 and 20,000 scf of hydrogen per ton of fuel processed depending on desired product slate. An adequate supply of hydrogen should preferably be generated from the process itself.

The process has been demonstrated with a range of coals including brown coals, lignites, subbituminous and bituminous coals. The hydrogen consumption with the younger (oxygenated) coals increases markedly. The experiments in the process development unit indicated a 90% solid carbon to liquid conversion.

THE EXXON DONOR SOLVENT PROCESS

This process involves the hydroliquefaction of slurried crushed coal in a tubular reactor in the presence of hydrogen and a hydrogen donor-solvent. Reaction conditions are comparable to those for the SRC and H-coal processes as can be seen in Figure 21.

The donor solvent is a catalytically hydrogenated fraction of the liquid product. Slurry leaving the reactor is separated by distillation into gas naphtha, distillates and a vacuum bottoms slurry. In this process the catalyst does not come into contact with coal minerals or high boiling liquids. The hydrogenated solvent produces a higher distillate product than does the use of an unhydrogenated solvent.

Early stages of the development of this process focussed attention on the conversion of bituminous and subbituminous coals.(11)

THE COST OF COAL LIQUEFACTION

Studies of coal conversion costs have all encountered similar problems in attempting to interrelate the estimates by the various process developers. Differences in unit size and location as well as the quality of the product have made comparisons difficult. Many studies have reported cost for liquid products solely on an energy basis ($ per million Btu). Clearly liquid fuels have different inherent worth. A million Btu of gasoline is worth more than a million Btu of boiler fuel.

A recent publication by the relatively independent Engineering Societies Commission on Energy has developed the concept of product value in making this comparison for a standard sized plant of capacity 25,000 tons/day of coal.(12) Product values were all assigned relative to premium gasoline = 1.00 and were as follows:

	Product Value
Solid Solvent Refined Coal	0.50
Char	0.47
Tar Oil	0.85
Fuel Oil	0.56
Diesel Oil	0.82
Naphtha	0.82
LPG	0.86
Gasoline	0.90
Premium Gasoline	1.00
Methanol	1.00
Methyl Fuel	0.96
Butane	1.07
Propane	1.08

The processes considered in this study were:

SRC-I : Solid Product
SRC-II : Liquid Product
H-Coal : Fuel-Oil Mode
H-Coal : Syncrude Mode
Fischer-Tropsch : Mixed Product Slate
Fischer-Tropsch : Methanol Production
Fischer-Tropsch : Methanol to Gasoline

The results, presented in Tables 4 and 5 illustrate the need to define the market into which the coal liquefaction product will be fed.

On an energy basis the H-Coal process operating in the maximum Fuel Oil Mode has an apparent advantage. On a product value basis, however, the H-Oil Syncrude Mode has an advantage.

It is interesting to note that the Fischer-Tropsch route, (gasoline via methanol), presents a significantly different picture when comparison is based on the product value concept rather than on a direct energy basis. Under these circumstances it becomes one of the most attractive options.

Table 4 - Liquefaction plant capital requirements (million $)
25,000 tons/day coal, mid 1978 dollars

	SRC-1	SRC-II	Exxon Donor Solvent	H-Oil (Syncrude)	H-Oil (Fuel Oil)	Fischer Tropsch	Methanol	Methanol to Gasoline
Coal Preparation ..	63	63	63	84	84	63	63	63
Hydrogen	152	253	190	158	138			
Gasification						228	228	228
O_2 Plant	84	129		87	67	117	175	175
Gas Shift				35	30		40	40
Acid/Sulphur Plant	60	60	60	57	57	57	57	57
Reactor	160	195	180	210	140	55	106	106
Conversion						100	75	
Gas Plant	107	30		25	30	25	10	10
Flexicoker			160					
Pollution Control .	44	44	44	40	40	40	40	40
Solvent or Catalyst Prep.			82			3		
Auxiliaries and Contingency 10% ..	422	488	491	438	369	433	500	452
TOTAL	1092	1260	1270	1134	955	1121	1294	1171

Table 5 - Product costs of liquefaction plant

	Energy Basis	Product Value Basis
	($/million Btu)	
SRC-I	3.38	6.67
SRC-II	3.62	5.59
Exxon DS	3.96	5.40
H-Coal - Fuel Oil	3.30	5.09
H-Coal - Syncrude	3.58	4.81
Fischer-Tropsch - (MPS)	4.99	5.52
Fischer-Tropsch - (Methanol)	4.37	4.54
Fischer-Tropsch - (Gasoline)	4.84	4.91

HYDROLIQUEFACTION OF CANADIAN COALS

Hydrogenation tests on Canadian coals representative of the entire A.S.T.M. coal classification spectrum were reported by CANMET in the 1930's. The test method was based on a continuous liquid phase operation similar to the hydrogenation of a coal slurry incorporated into many of the commercial schemes. The solvent vehicle for slurry preparation throughout the program was a process derived liquid from the appropriate parent coal. The principal product of the experiment was a liquid crude oil, the quantity of which can be taken as indicating the suitability of the coal to the liquefaction process.(13)

Low rank coals were prepared to give a feedstock with approximately 3% moisture and 9% ash content; higher rank coals were washed to maintain ash levels below 7%. The experiments were all carried out using a 5% catalyst addition as stannous oxide.

The results of this work are presented in Figure 22 where a current series of Australian data is added for comparison. On the basis of this data it appears that the maximum yield is offered by carboniferous high volatile bituminous A coal. The yields from lower rank coals decrease progressively from this level of 77% and are apparently lower with cretaceous coals of the same rank.

This early data can be interpreted in the light of current ideas about the development of plasticity in coals and the relationship between natural plasticity and the role of hydrogen donors (solvents) in the liquefaction process.

It has been postulated that an essential precursor of hydrogen transfer to components in the coal structure is solution and/or physical melting of the coal by rupture of weak structural elements in the coal.

This is a natural process in the heating of coals which develop a plastic phase when it gives rise to a decrease in the viscosity of the coal. In this case the natural bitumen may act as a hydrogen donor until the hydrogen inventory is consumed. Repolymerisation follows, and a semi-coke is produced.

The removal of the carrier fluid (vehicle) appears to be deleterious to the liquefaction process as well as to the development of plasticity.

Although no clear relationship between liquefaction and thermal plasticity has been developed, the hydrogen transfer mechanisms in the two processes are similar. This implies that the ratio between inert macerals and reactive (plastic) macerals can play an important role in determining the recycle ratios of donor solvents that will be required in the process route.

It also suggests that, where coals show a high plasticity, the natural hydrogen donation makes the hydroliquefaction process more attractive than it will be for the inert or low plasticity coal.

This argument does not mean that low plasticity coals cannot be liquefied by direct hydrogenation but rather that a different process optimisation must be used. Higher recycle ratios will, in general, mean more expensive process equipment. Low plasticity is usually associated with low-rank oxygenated coals which give high gas yields and hydrogen consumption and thus further increase process cost.

No specific predictions can be made about the applicability of individual processes to the liquefaction of Western Canadian coals beyond the general level developed in this discussion. Conclusive yield data can only be developed after process optimization experiments have been carried out using specific coal deposits as the feedstock for each process.

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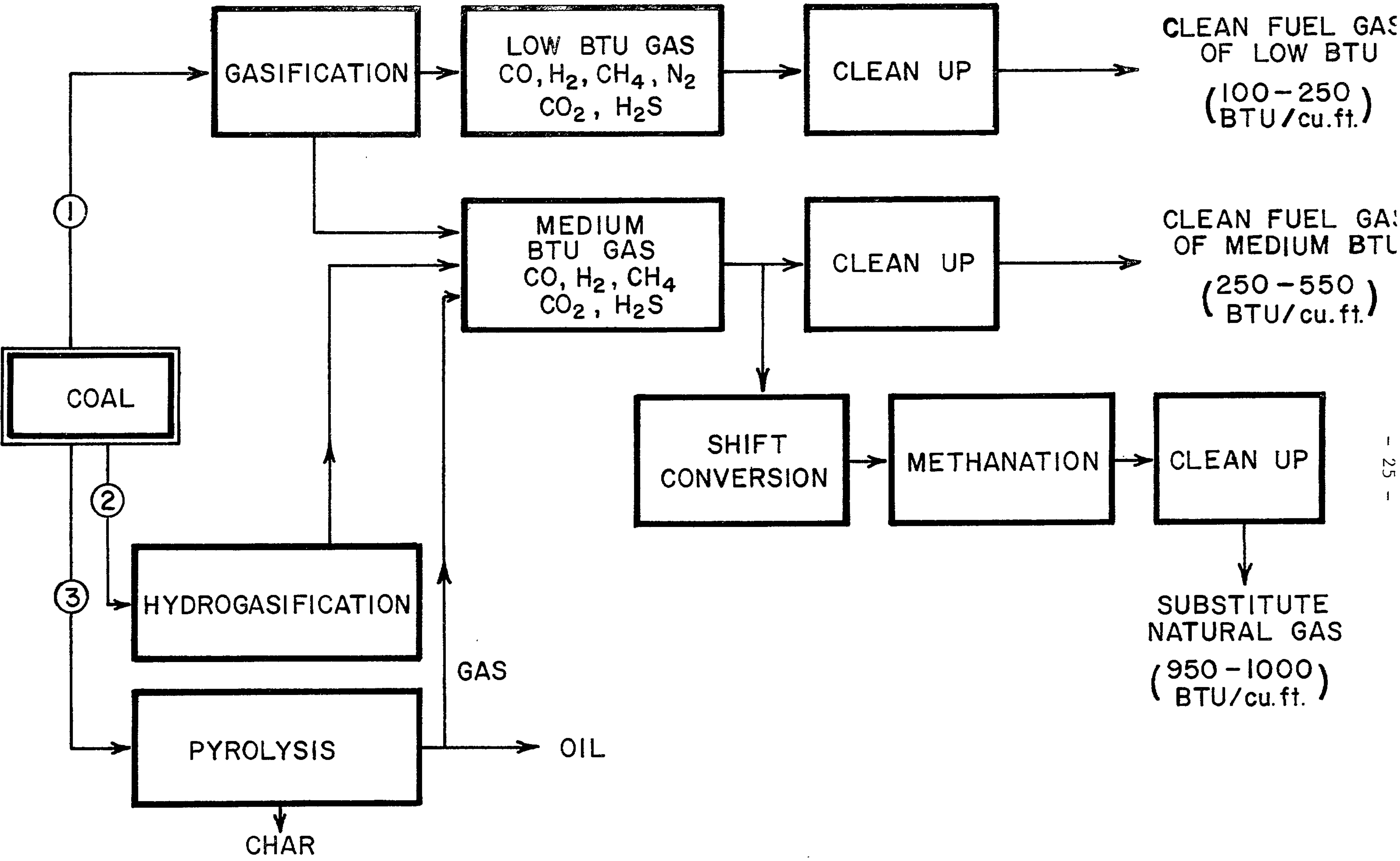

Fig. 1 - Coal gasification process routes.

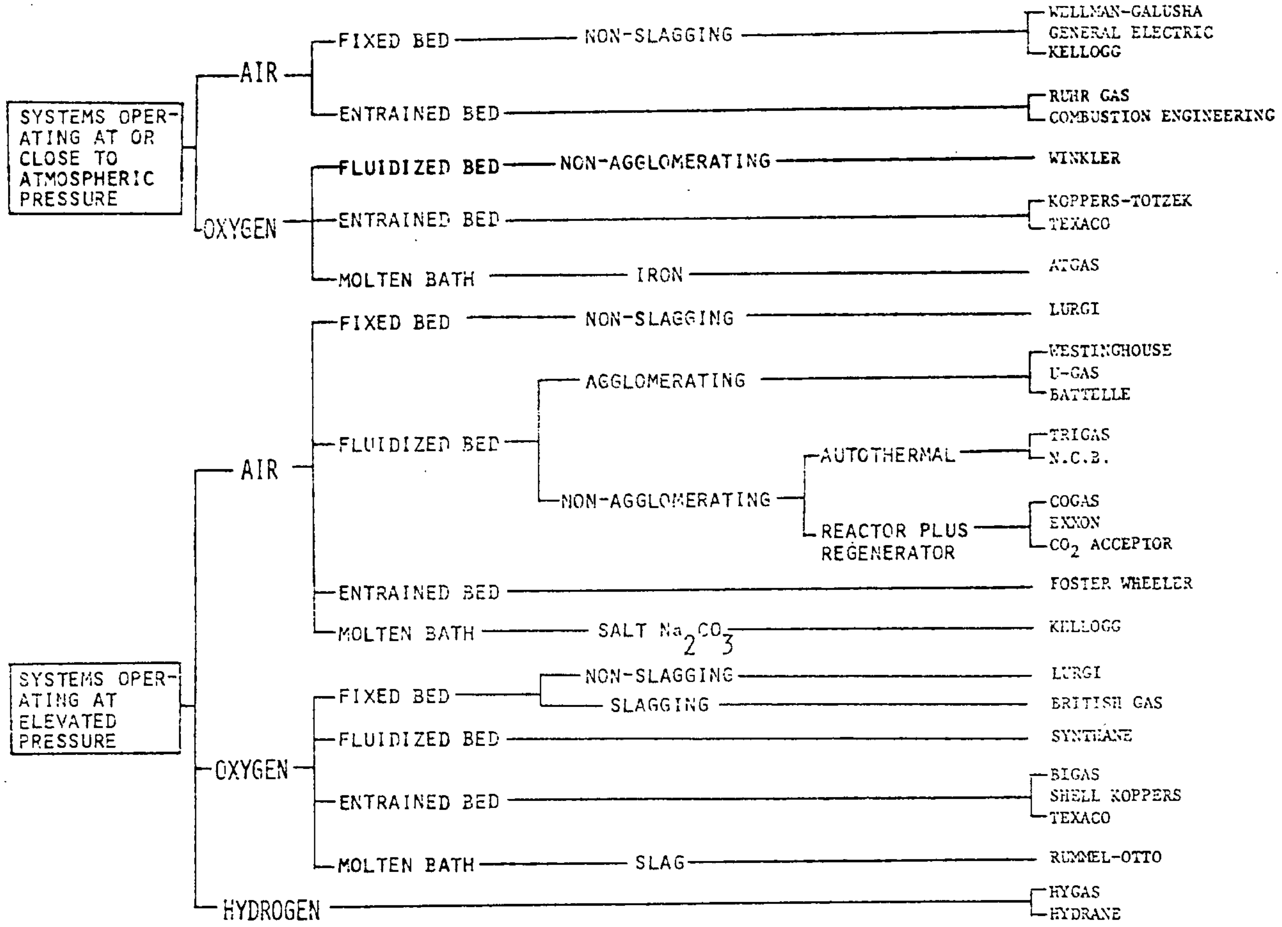

Fig. 2 - Coal gasification processes.
(after Cooke and Robson, 1978)

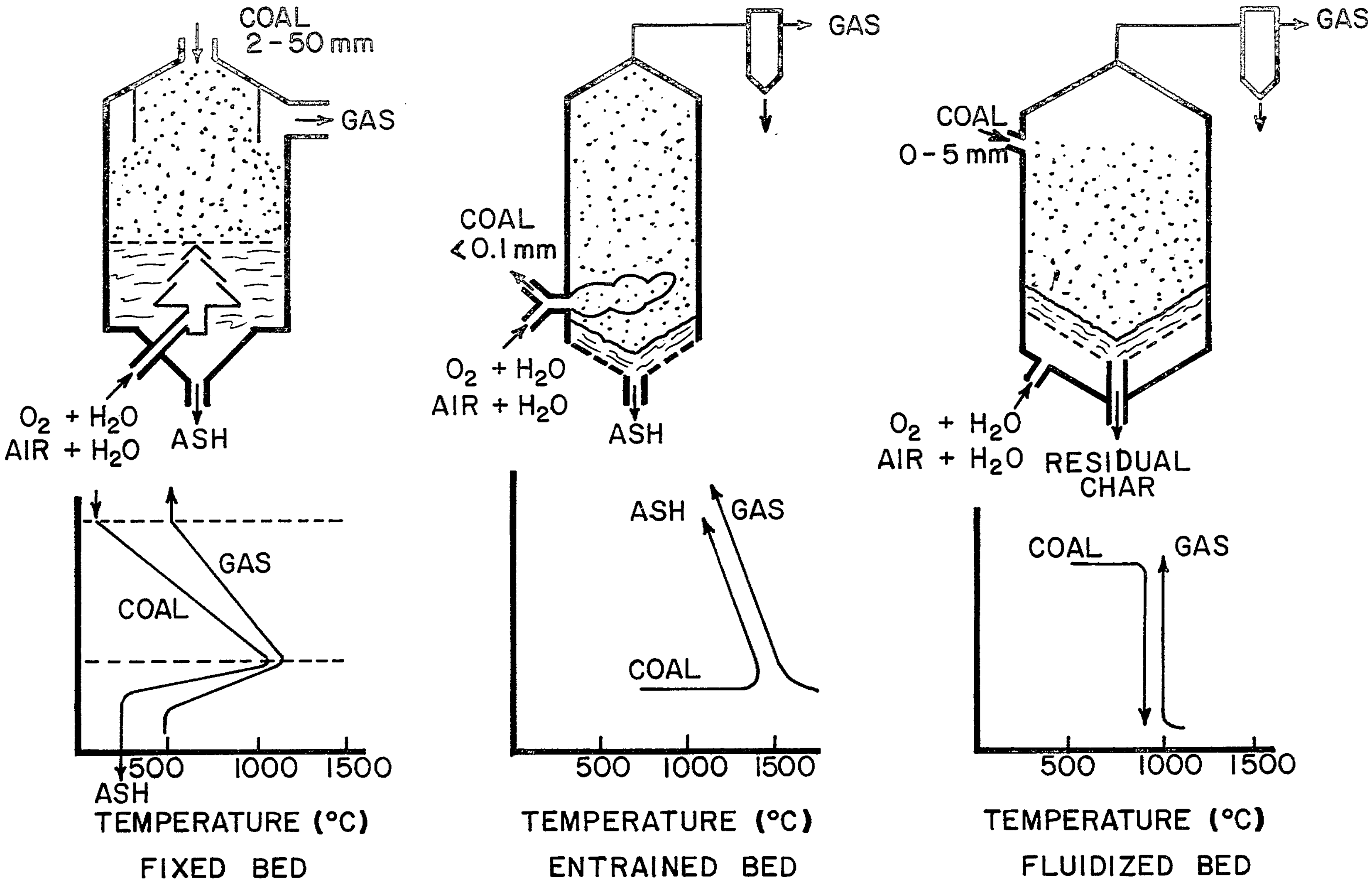

Fig. 3 - Three categories of gasifiers.

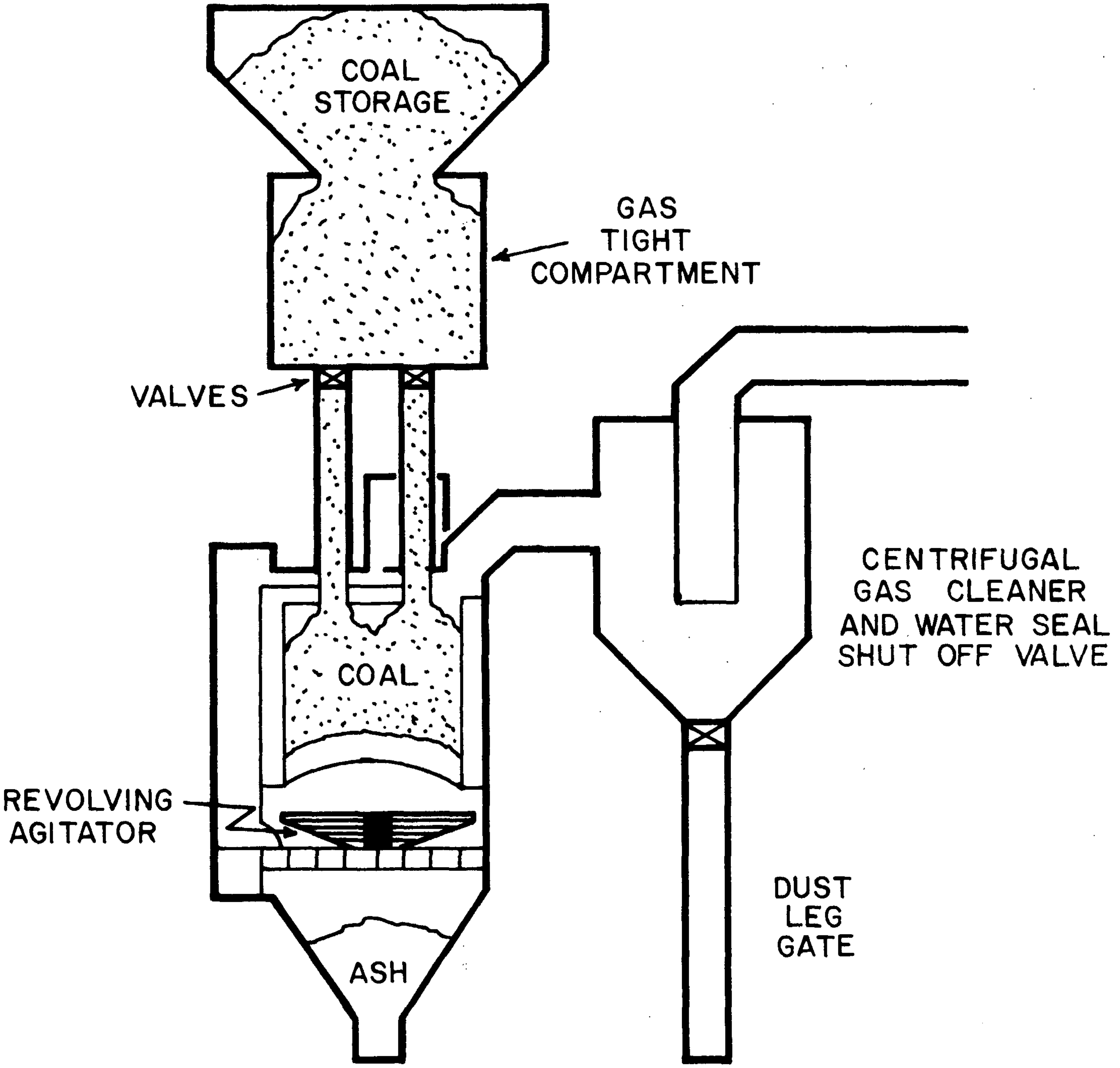

Fig. 4 - The Wellman-Calusha fixed bed gasifier.

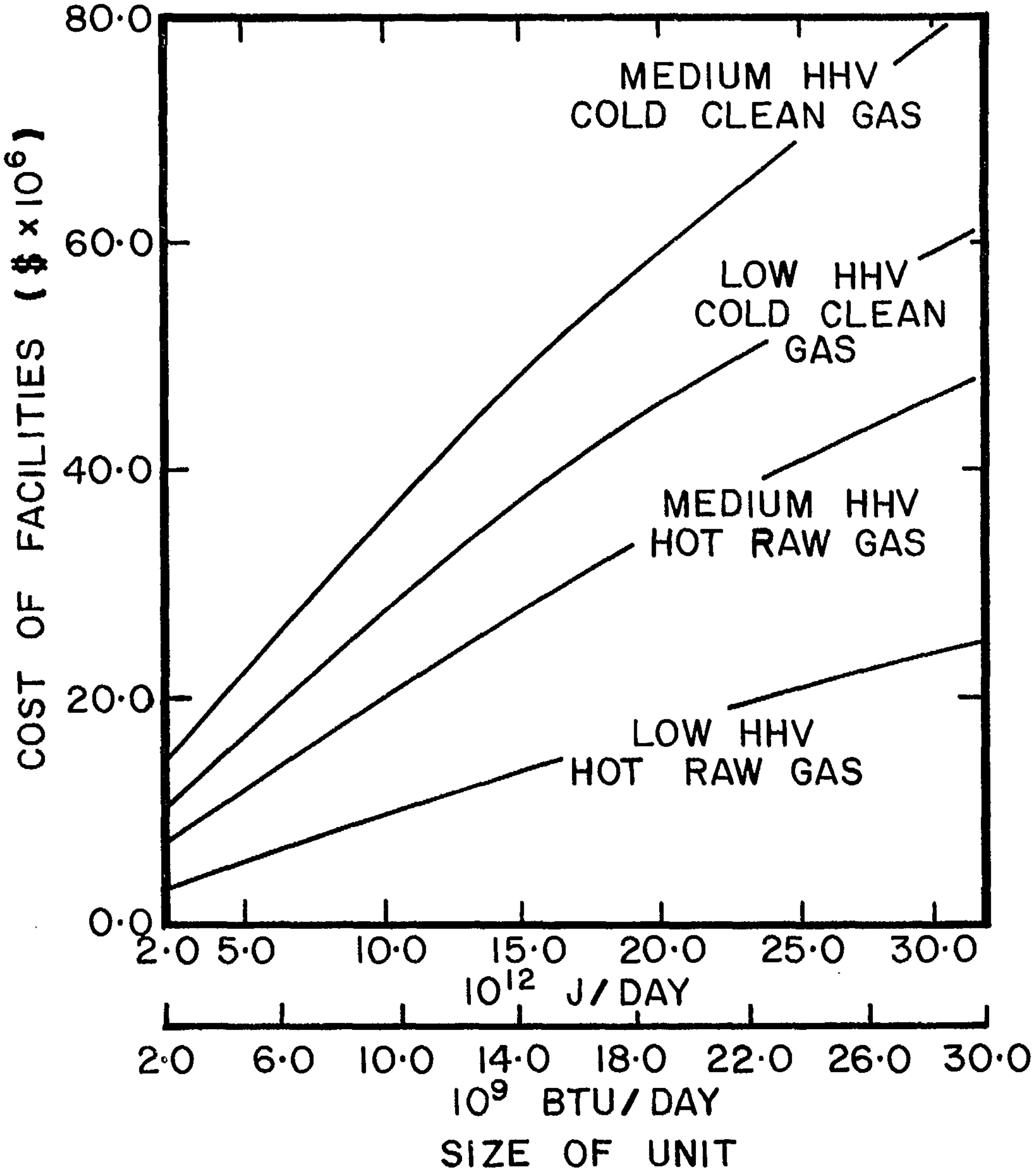

Fig. 5 - The cost of facilities: Wellman-Galusha gasification.

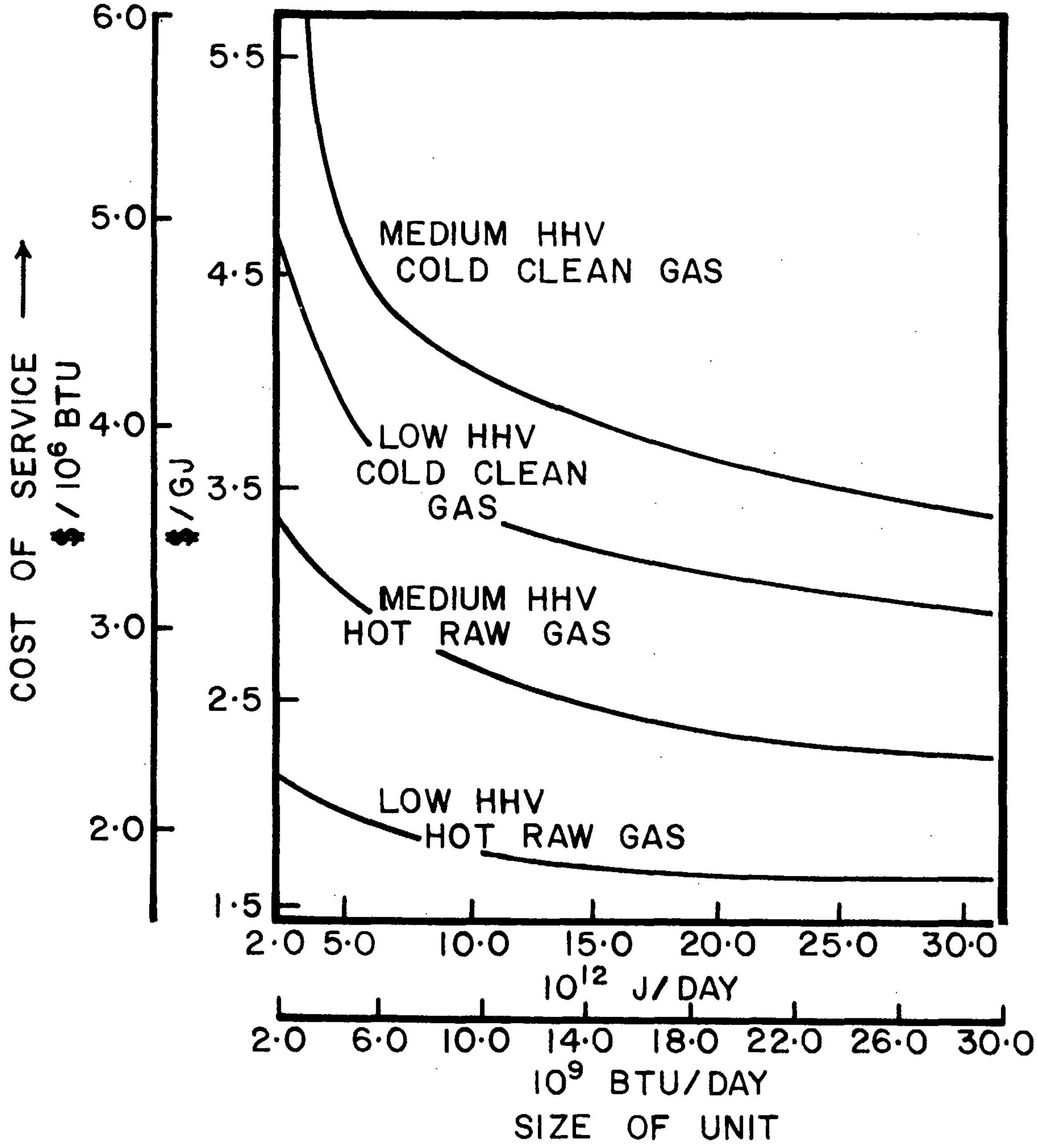

Fig. 6 - The cost of service: Wellman-Galusha gasification.

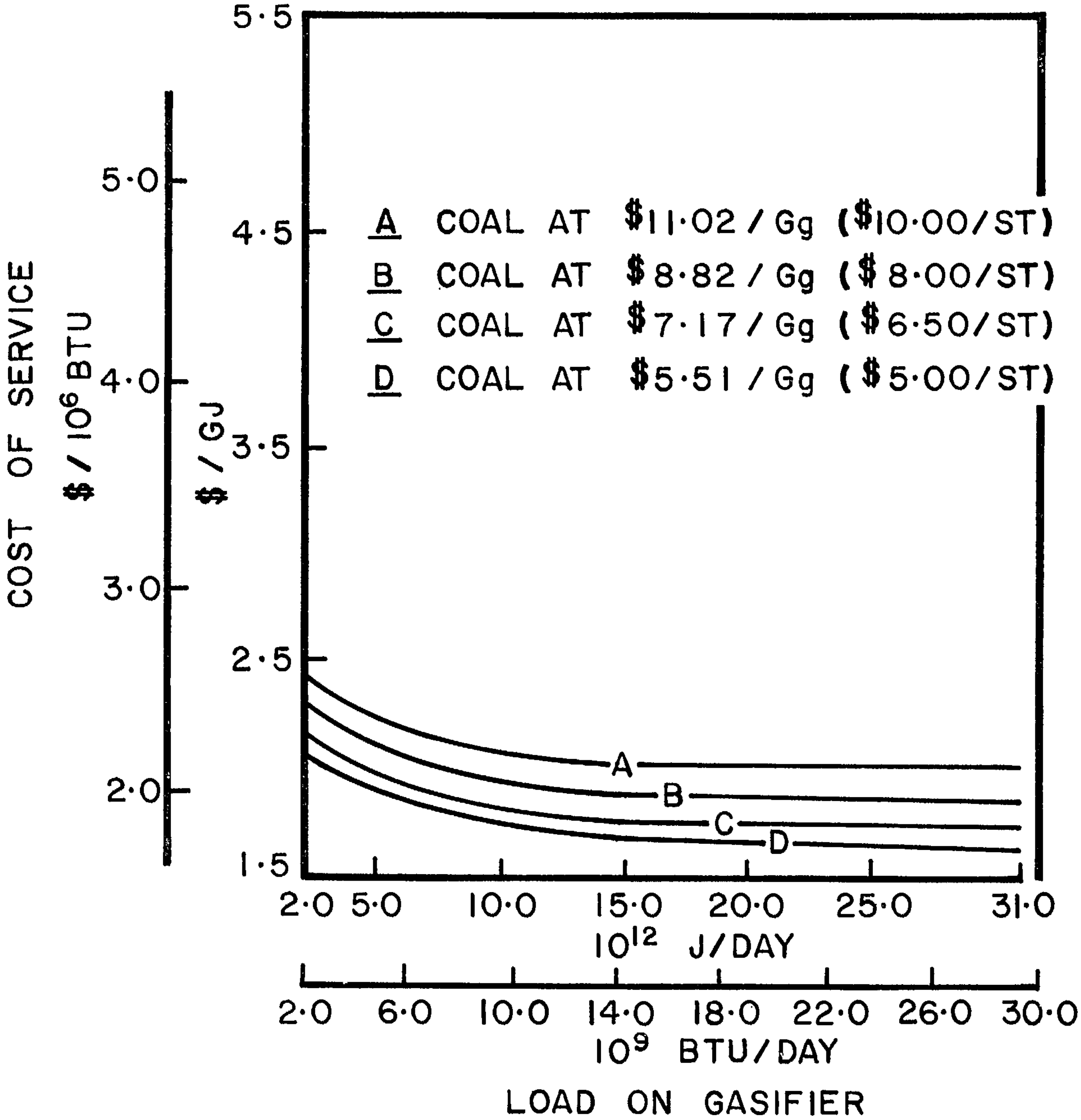

Fig. 7 - The effect of coal price on the cost of service: Wellman-Galusha gasification.

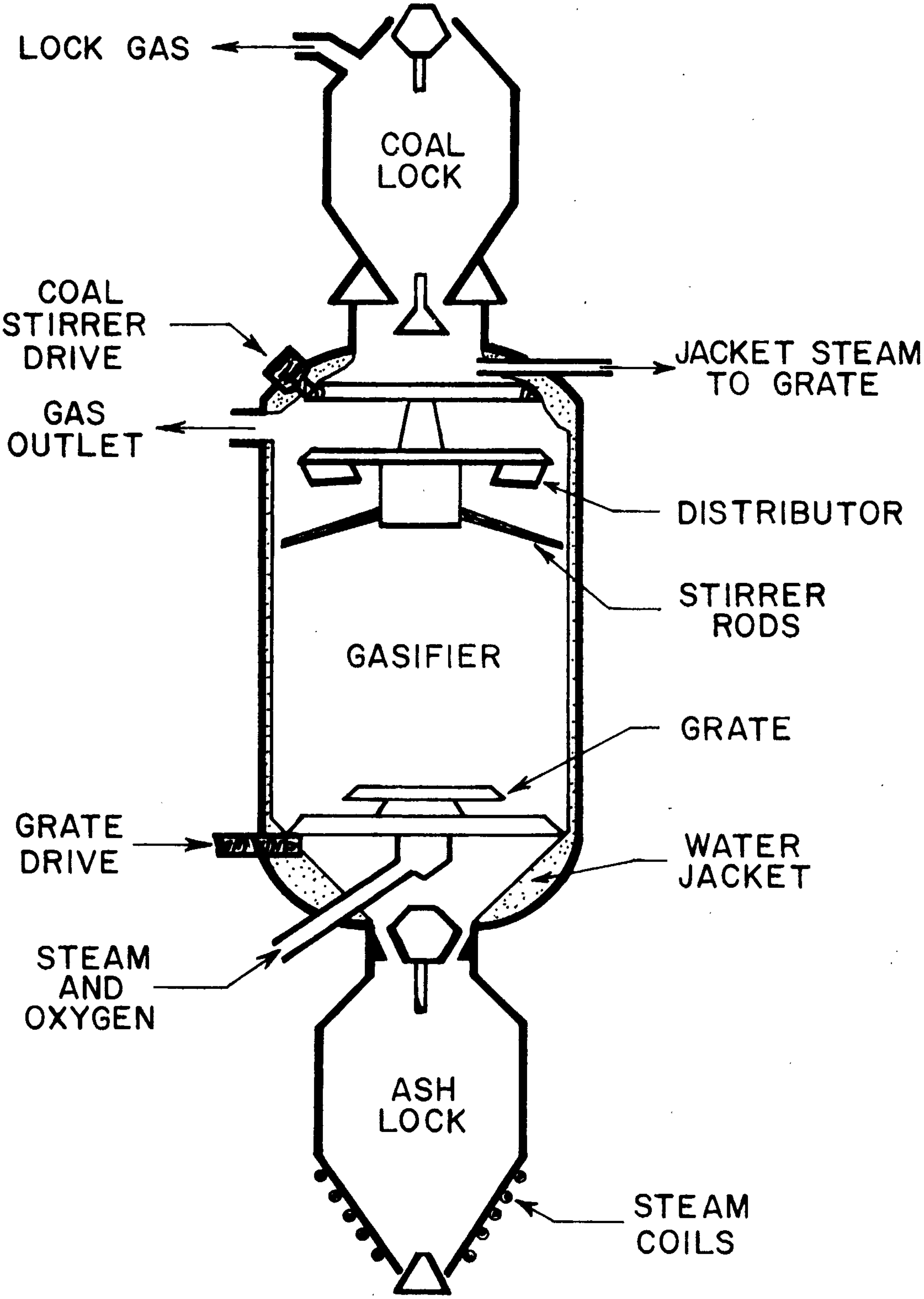

Fig. 8 - The Lurgi fixed bed gasifier.

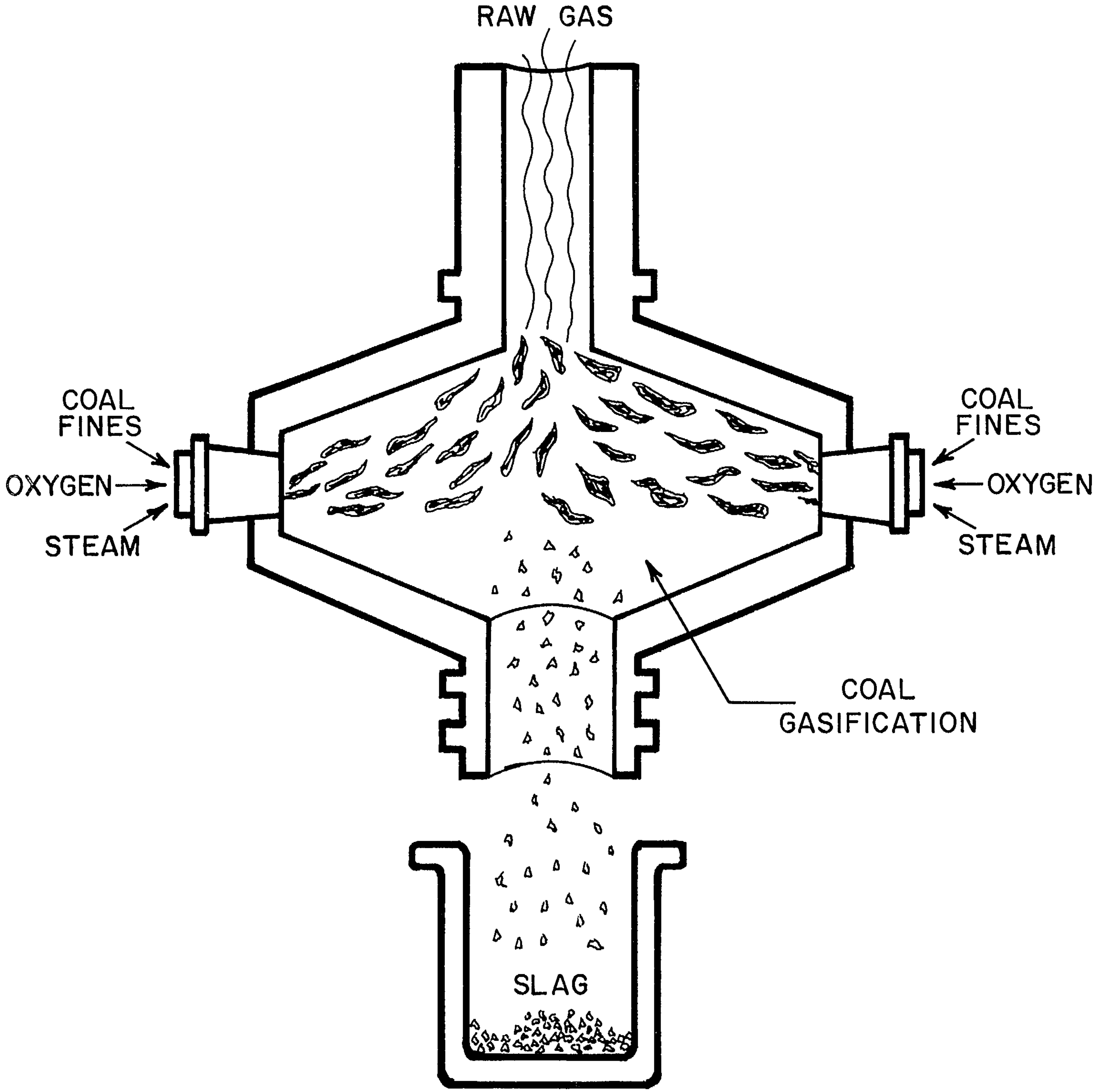

Fig. 9 - The Koppers-Totzek entrained bed gasifier.

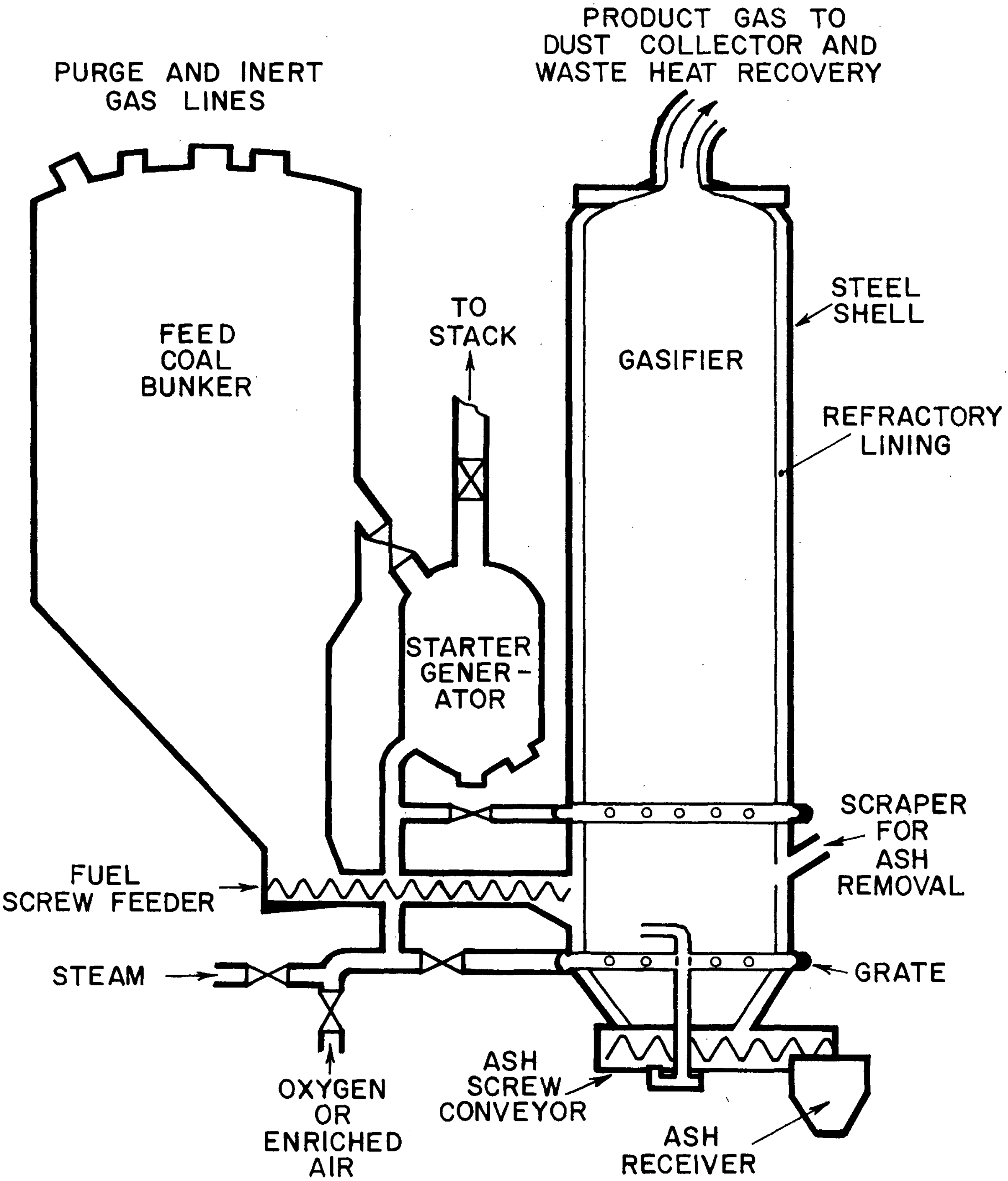

Fig. 10 - The Winkler fluid bed gasifier.

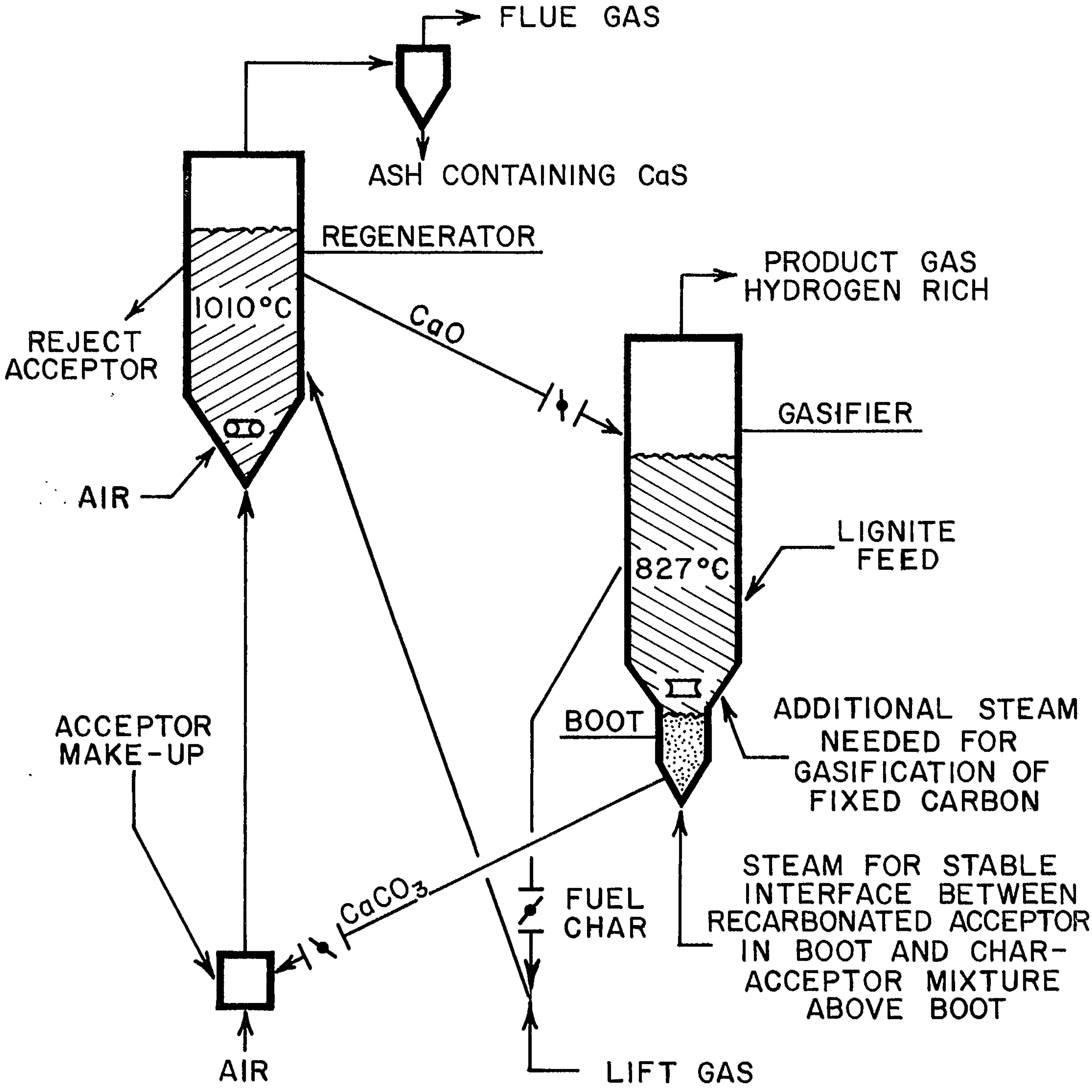

Fig. 11 - The Conoco CO_2 acceptor process.

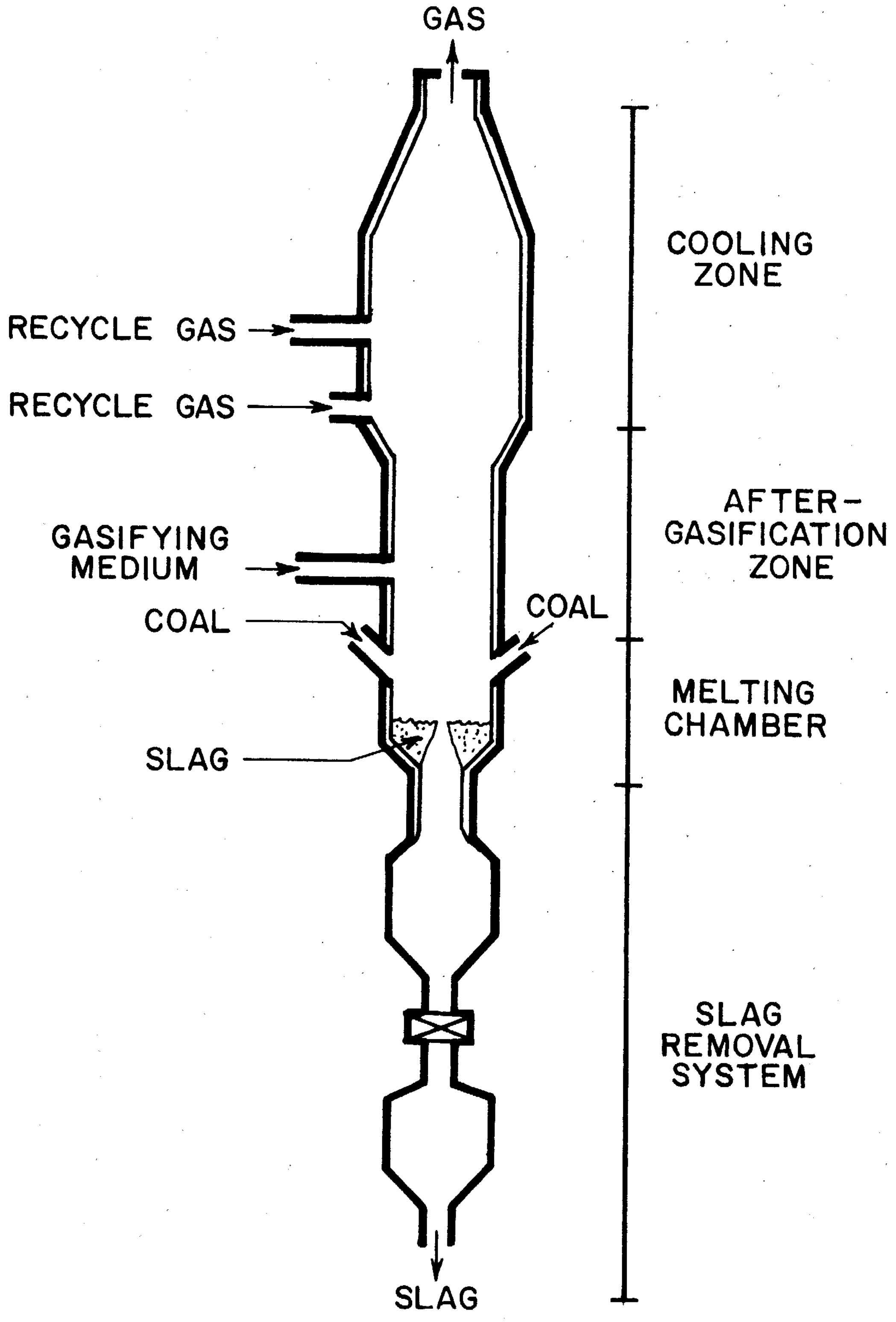

Fig. 12 - The Rummel-Otto slag bath gasifier.

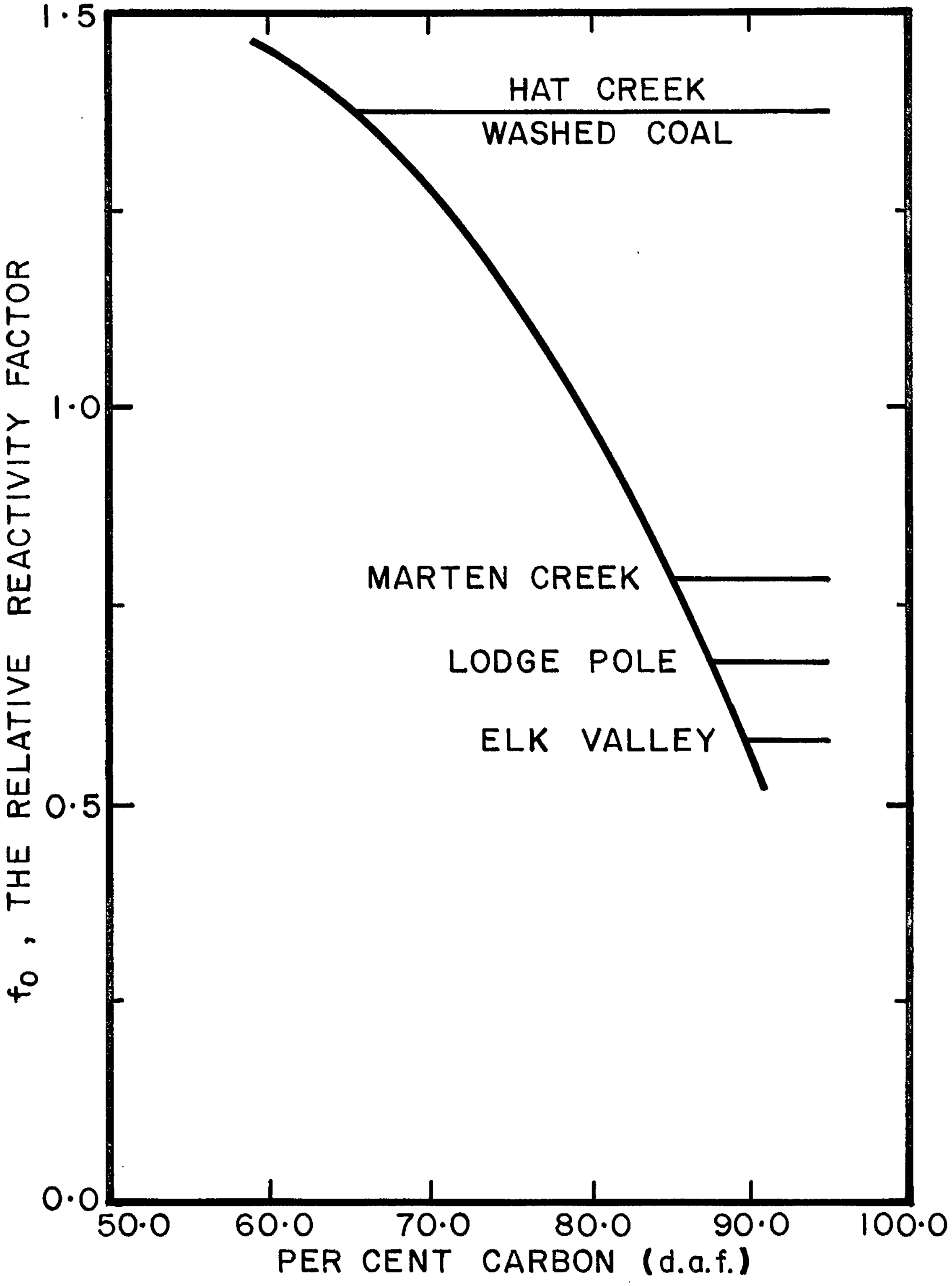

Fig. 13 - The relative reactivity factor for Canadian coals.

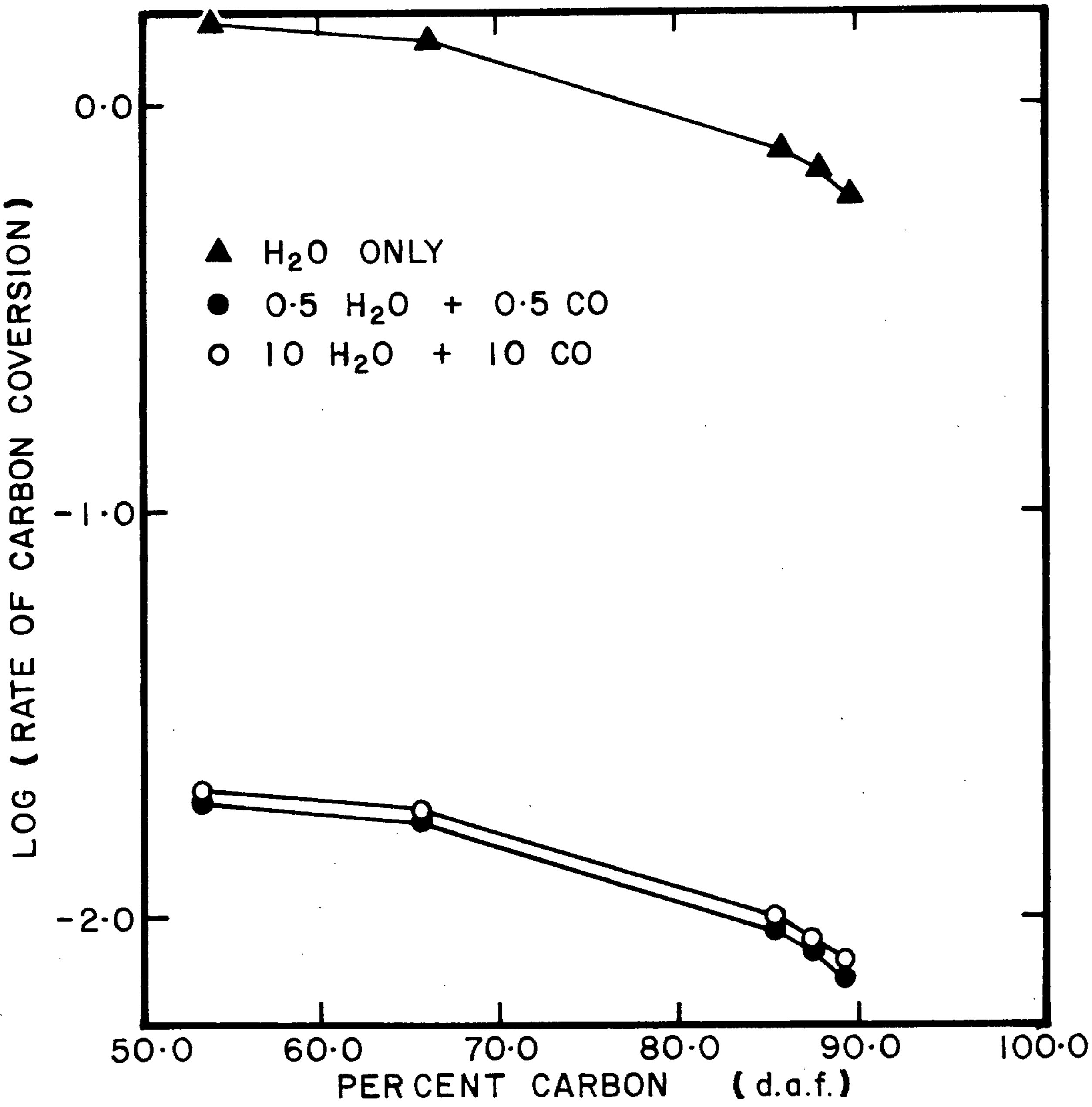

Fig. 14 - The rate of gasification of Canadian coals.

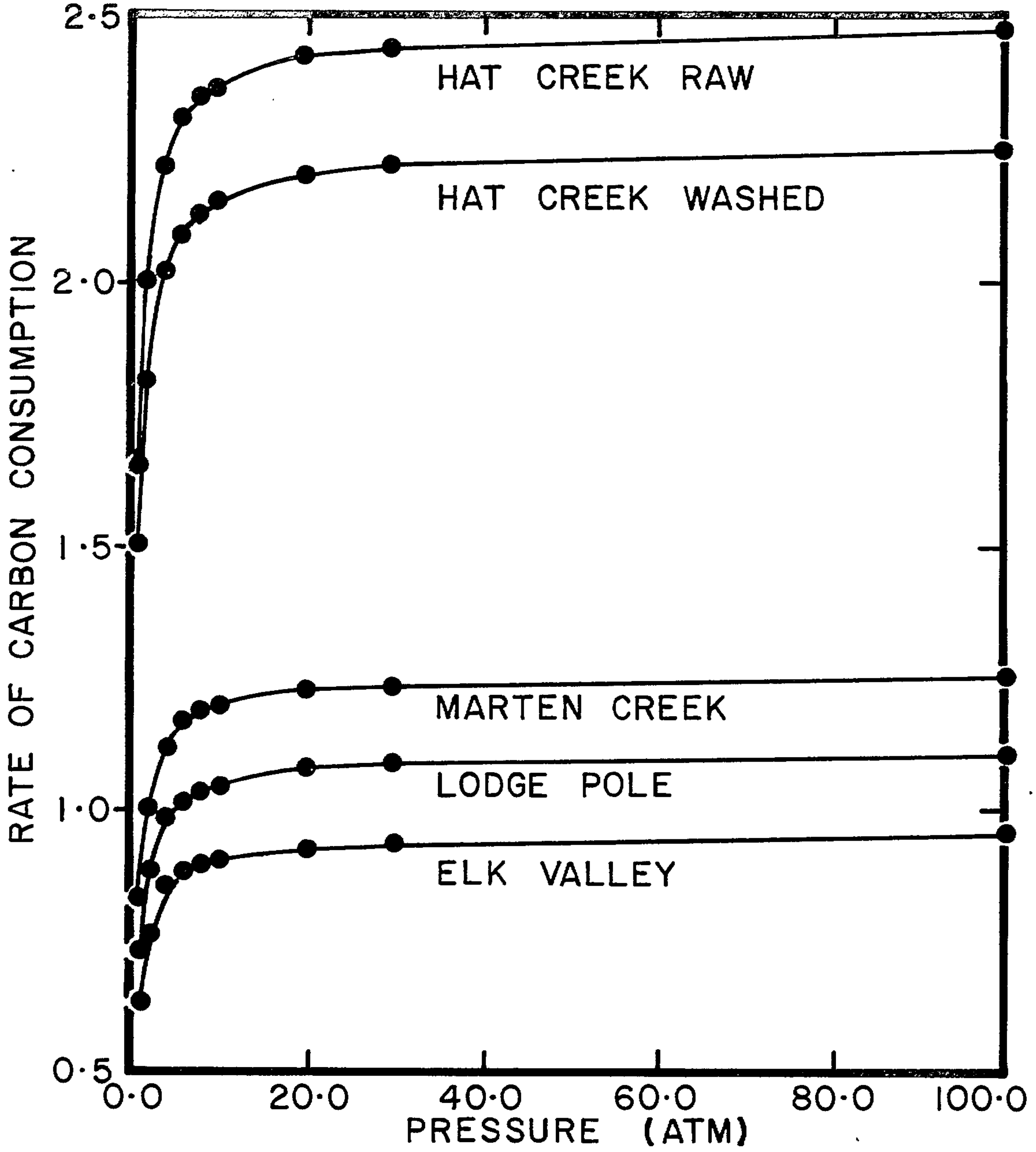

Fig. 15 - The rate of gasification of Canadian coals; the effect of pressure.

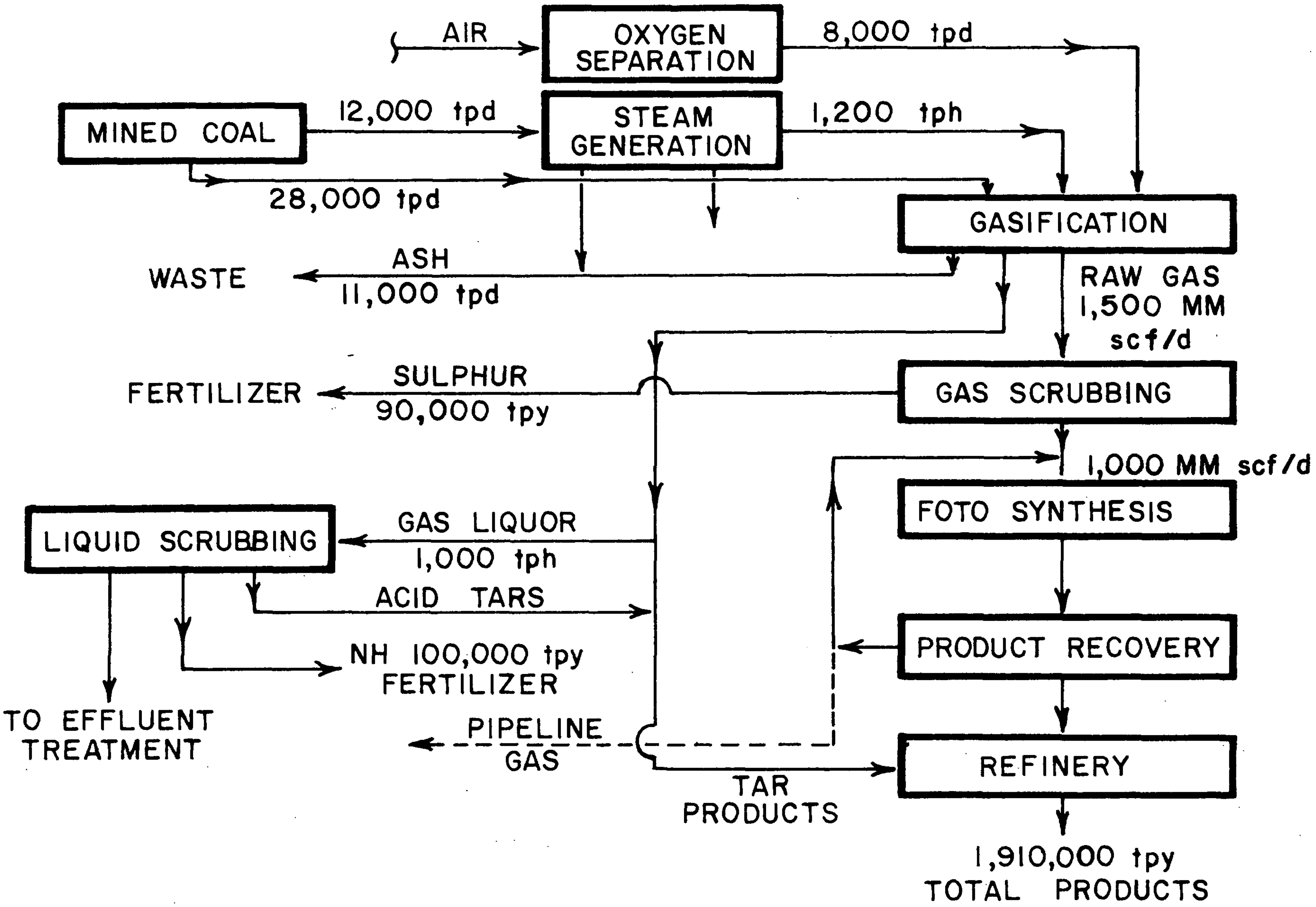

Fig. 16 - The process route: SASOL I.

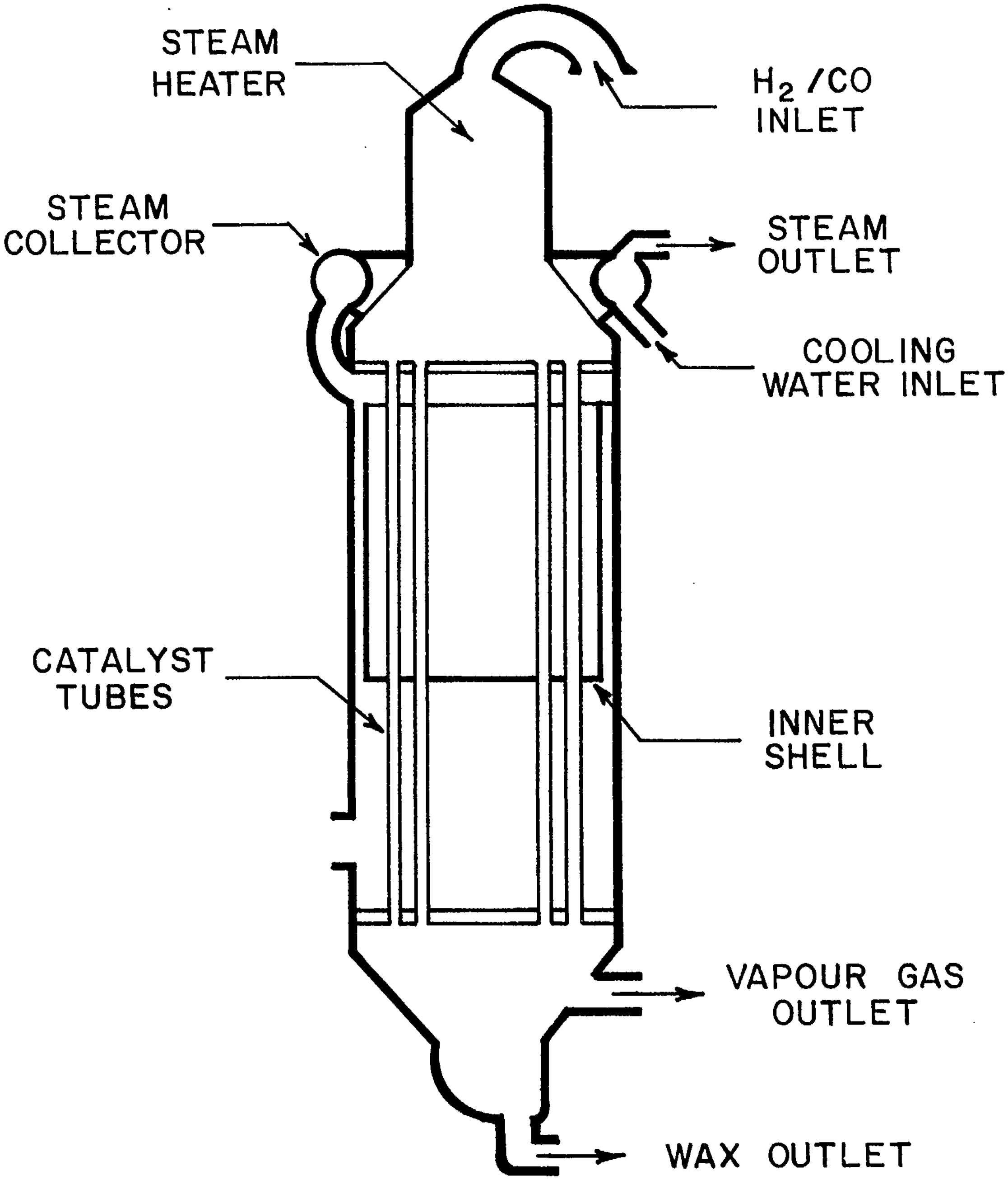

Fig. 17 - The Arge fixed bed reactor: SASOL I.

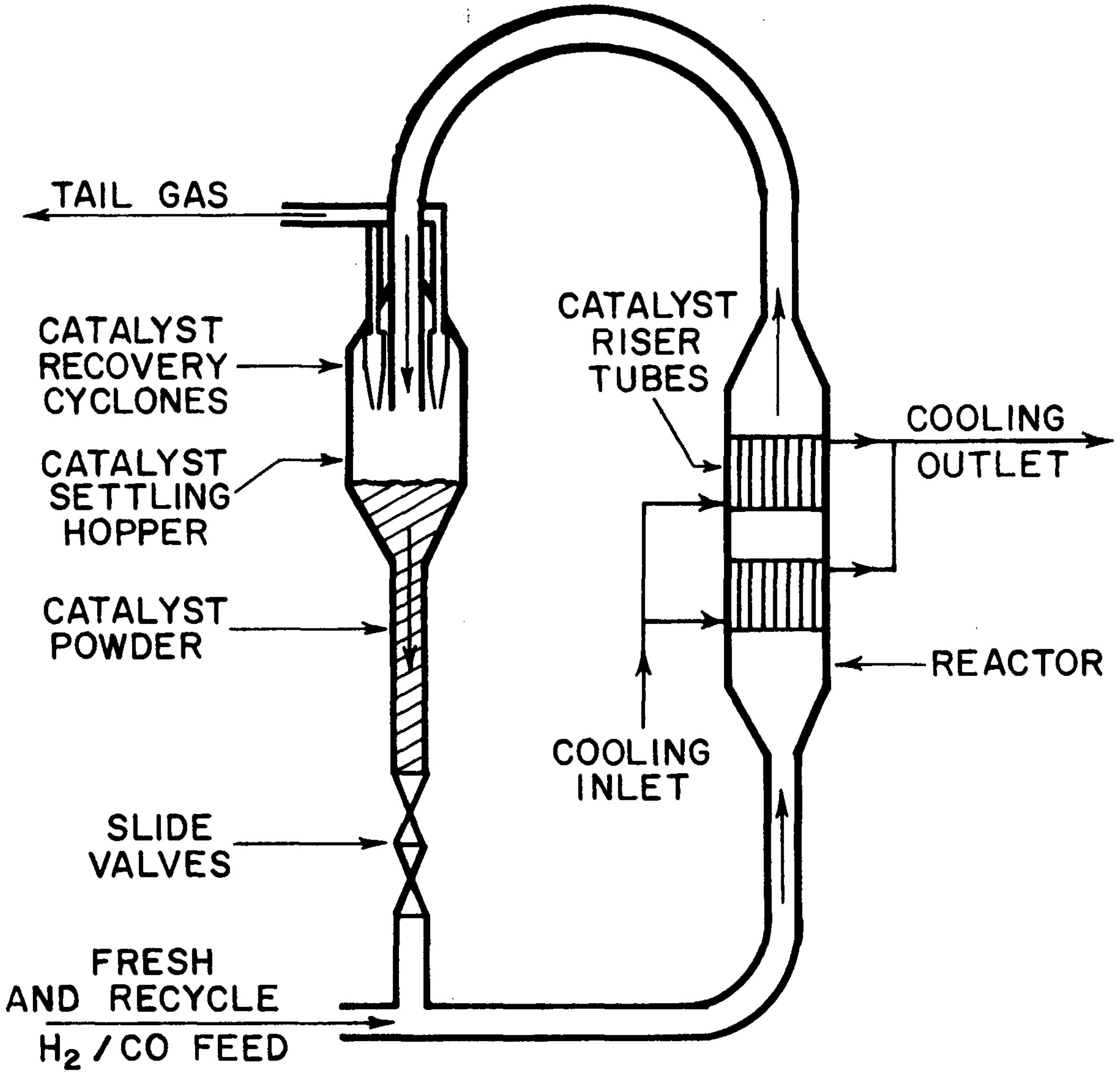

Fig. 18 - The Synthol fluidized reactor: SASOL I.

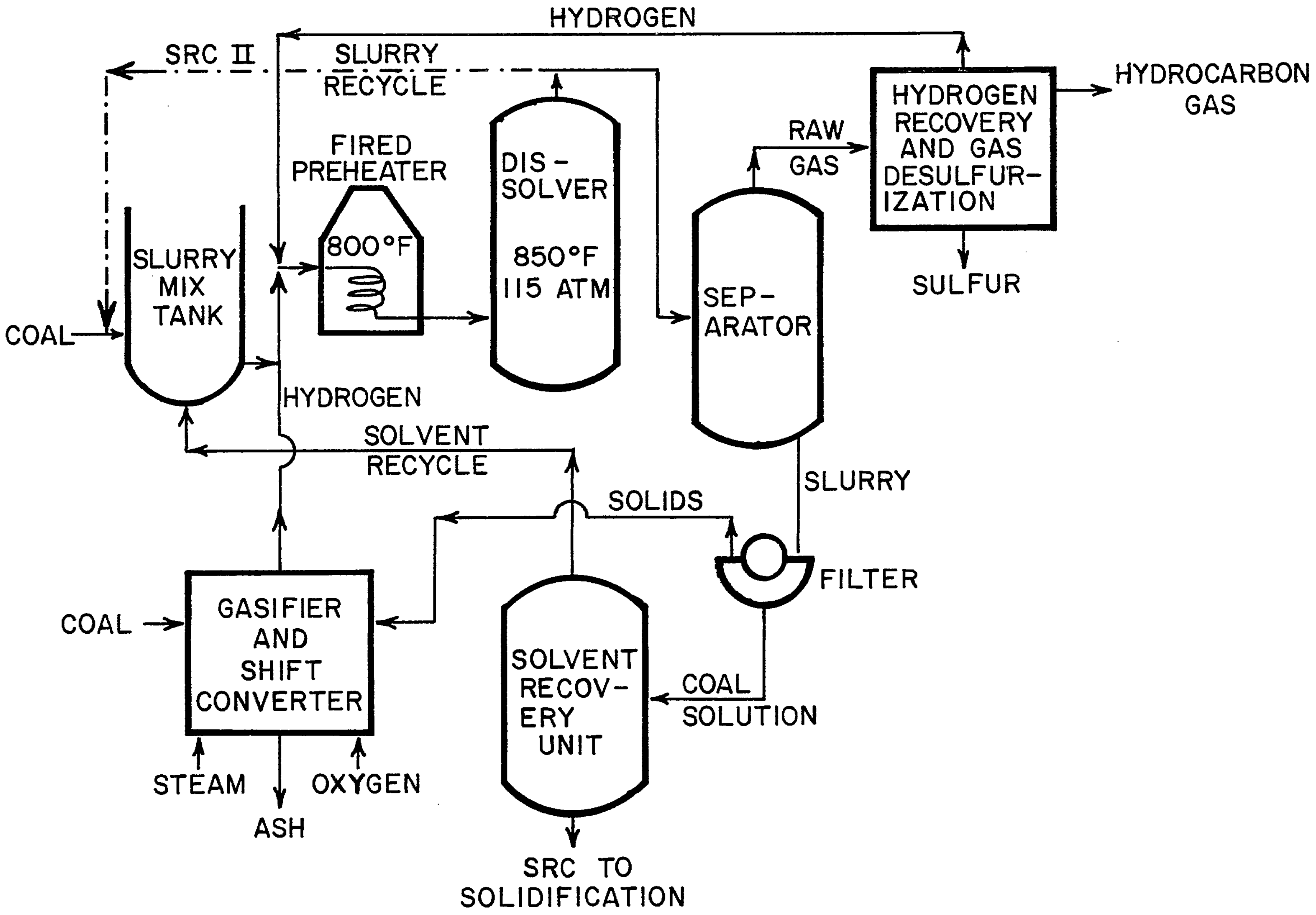

Fig. 19. The S.R.C. process.

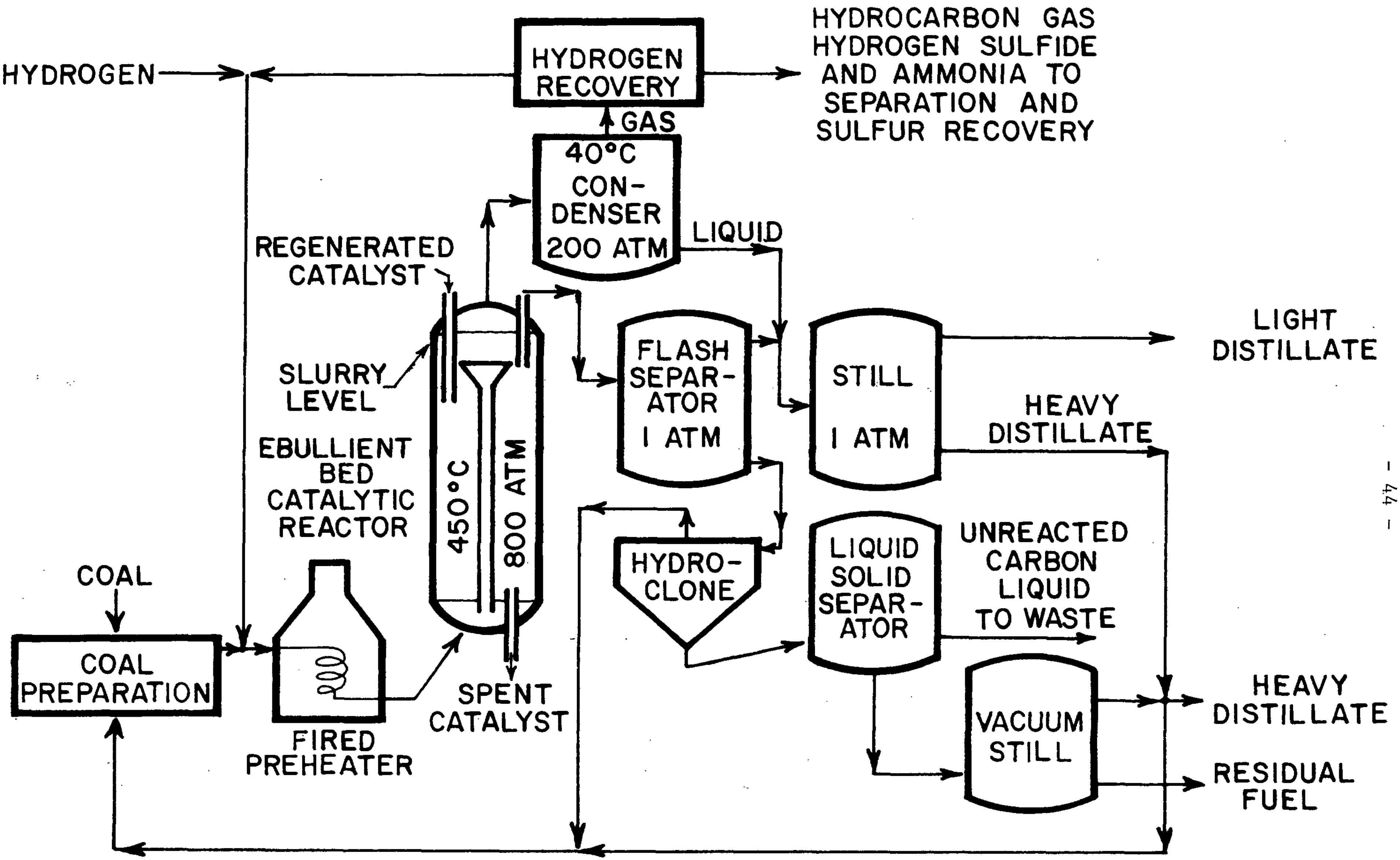

Fig. 20 - The H-Coal process.

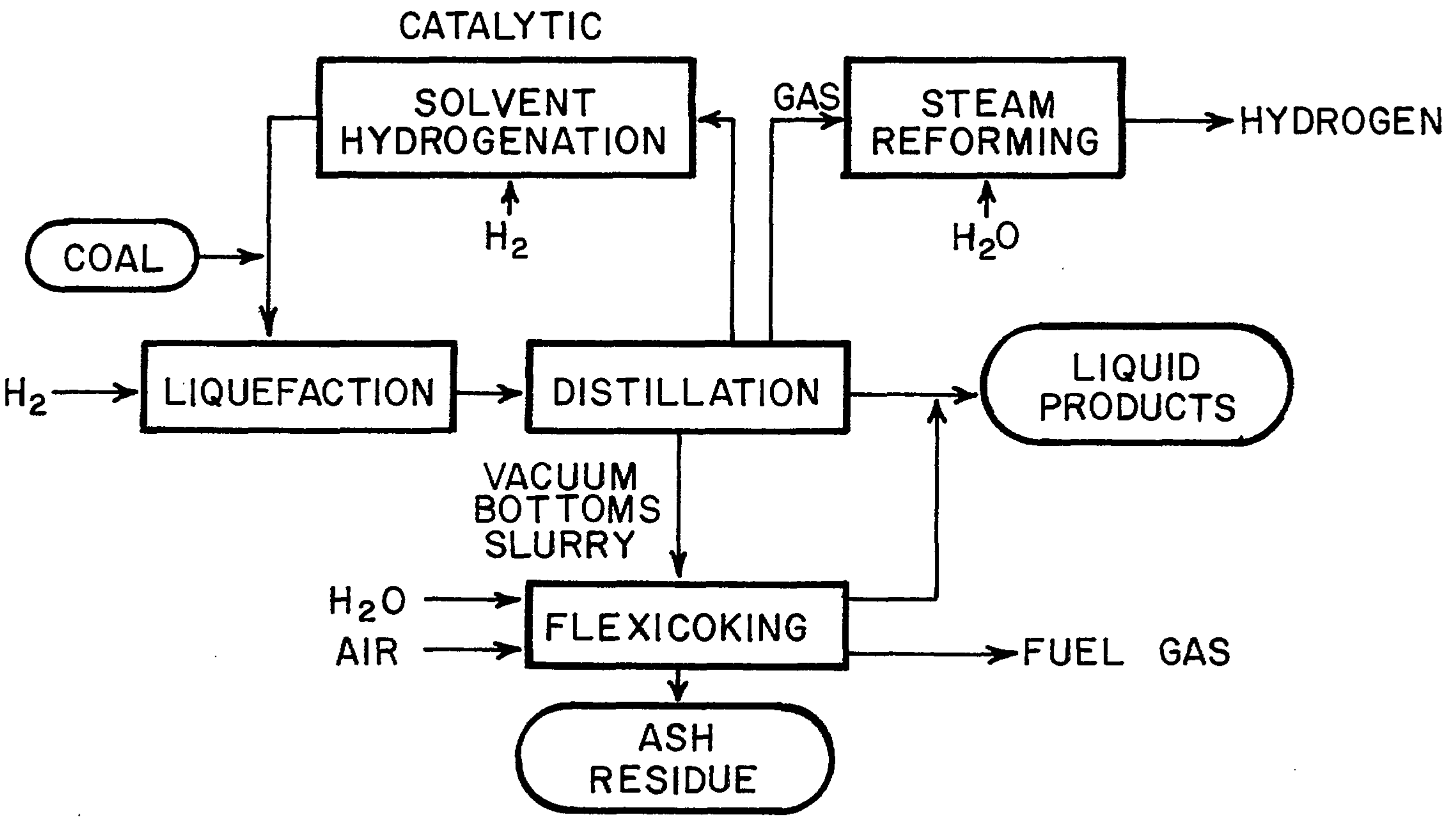

Fig. 21 - The Exxon donor solvent process.

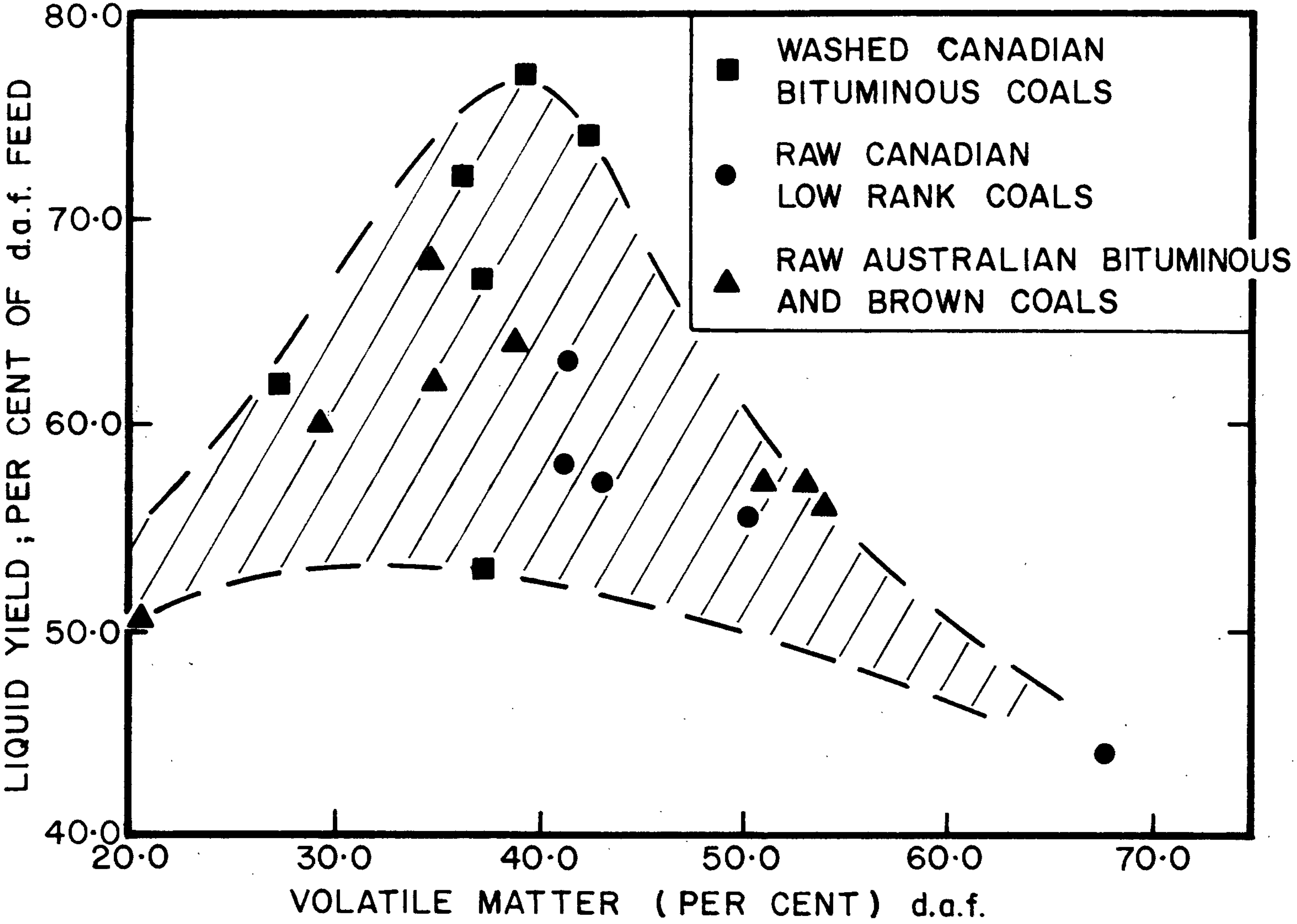

Fig. 22 - The liquid yield from catalytic hydrogenation of a series of Canadian coals.

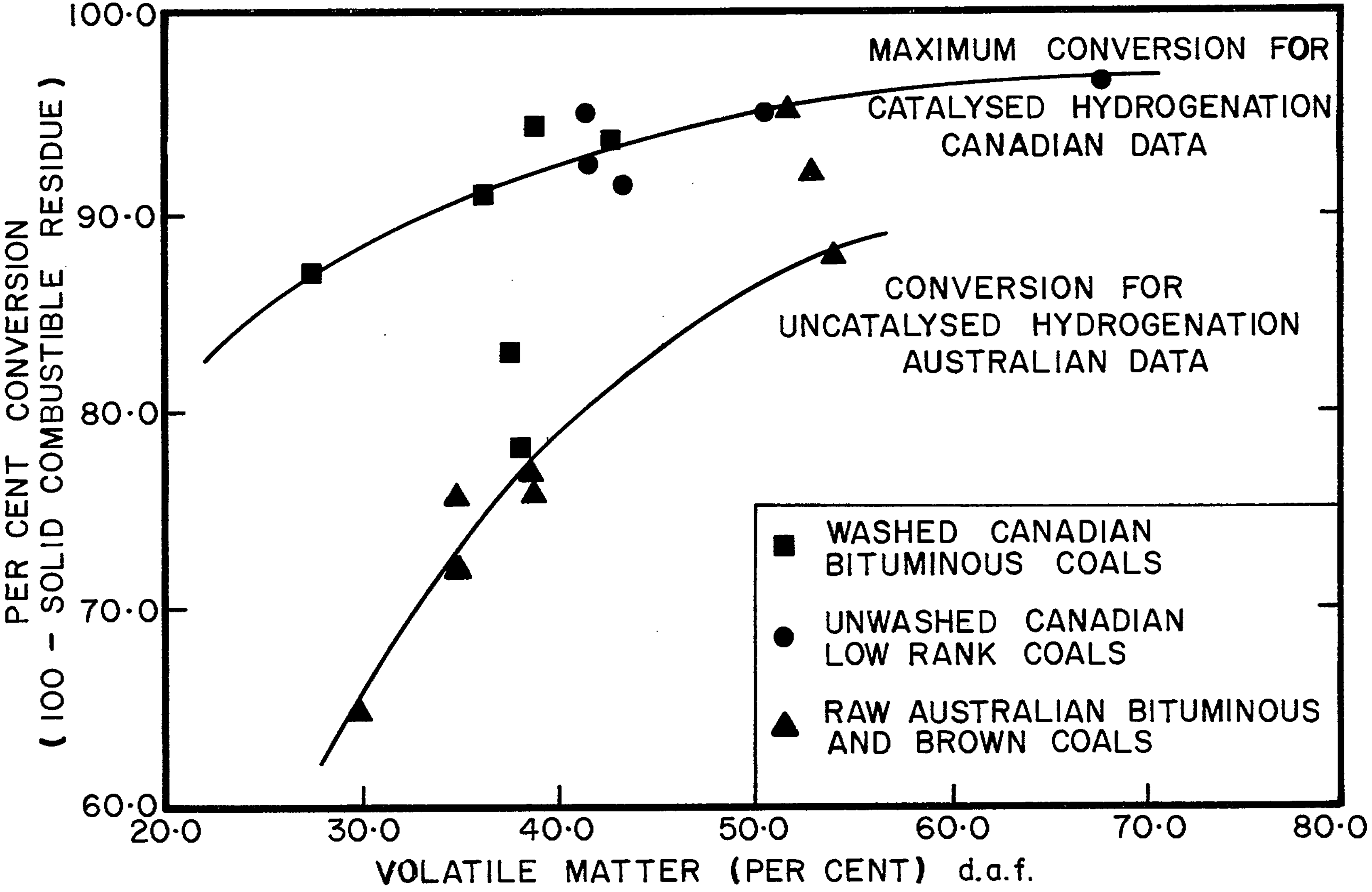

Fig. 23 - The total conversion from catalytic hydrogenation of a series of Canadian coals.