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CONSIDERATIONS ON COAL GASIFICATION

J. E. Franzen

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16. Abstract

Commercial processes for the gasification of coal with oxygen are discussed. The Koppers-Totzek process for the gasification of coal dust entrained in a stream of gasifying agents is described in particular detail. The outlook for future applications of coal gasification is discussed.

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CONSIDERATIONS ON COAL GASIFICATION

J. E. Franzen
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1. The Energy Crisis and Coal Refining /83*

The spectacular events in the late fall of 1973 connected with the temporary oil embargo have refocused public attention onto an energy source and raw material, coal, which is not only fairly abundant in West Germany, but is present elsewhere on earth in quantities sufficient to meet a considerable portion of mankind's energy needs for many hundreds and perhaps thousands of years. Less publicized, however, have been the efforts made in various countries since the early 1960s, including the United States, England and West Germany, to expand the classic applications of coal in power stations and coking plants by new refining techniques, such as its conversion to fuel gas or to industrial-grade, hydrogen-rich gases for use as chemical raw materials.

Meanwhile almost all coal-producing countries have national energy programs calling for the further development of coal refining technology, especially the gasification and liquefaction of coal, through the combined mobilization of public and private resources. It is not surprising that the new interest in coal refining has brought forth many proposals which may sound ingenious but often are impractical and ignore previous technical experience and economic realities. As a result, there is a danger that the money and efforts expended in this area may not yield the desired returns. The unquestionable fact that coal has

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* Numbers in the margin indicate pagination in the foreign text.

almost limitless uses does not mean that all conceivable applications of coal are reasonable everywhere and at all times. We shall return to this point at the end of the article.

The discussions which follow are intended to give an overview of coal gasification techniques which have already been proved commercially. Special emphasis will be placed on the Koppers-Totzek process to illustrate the range of applications of coal gasification.

2. Coal Gasification

Coal can be completely combusted in coal-fired power plants or various types of coal furnaces to yield a gas which, while possessing sensible energy, no longer contains energy in chemically bound form. This form of coal utilization will not be discussed here. This article is concerned, rather, with the conversion of coal directly from its solid form into a gas which contains a maximal amount of chemically bound energy, and which can be utilized either as an energy source or as a chemical raw material or as both, depending on the country and the situation.

Of the almost incalculable number of processes proposed for the conversion of coal, those based on the use of extracting agents (coal dissolution) or the direct reaction of coal with pressurized hydrogen will not be treated here. We shall limit our discussion to processes in which the coal is gasified as completely as possible with oxygen and steam to yield a gas mixture whose composition depends on the type of coal used as well as the gasification conditions employed. Coal gasification processes which utilize the heat from a high-temperature reactor will not be discussed either, since the technology of these processes is still too far from industrial and commercial implementation.

3. Coal Gasification with Oxygen

This type of coal gasification is of special significance, inasmuch as it is employed by all coal gasification plants which have been or are presently in commercial operation.

Three basic types of gasification with oxygen are recognized, depending on the degree of dispersion of the coal and its motion during gasification:

3.1. Fixed-Bed Gasification

The coal, which is in a relatively coarse form, forms a slowly downward-moving bed in which the individual coal particles move relatively little with respect to one another, while the gasifying agents (oxygen + steam) flow upward through the bed. It is a true countercurrent process.

An example of this process, which has found the most significant development and commercial application, is Lurgi pressure gasification. The basic principle of this type of gasifier is shown in Fig. 1.

The first experimental gasifier was put into operation in 1932, and the first commercial plant in 1935/36 at the Hirschfelde lignite mine of Saechsische Werke AG. /84

Lurgi as well as British, American and East German firms and institutions are continuing developmental work on this type of gasifier.

3.2. Fluidized-Bed Gasification

This type of gasification is done with a relatively fine-grain coal, which is kept in suspension by the upward flow of gasifying agents into the loose bed of material. Since the individual coal particles can be regarded as making short-length, disordered motions about a point, the exchange of heat and material with the gasifying agents takes place under constantly changing conditions. The only commercial process of this type is the Winkler process, shown schematically in Fig. 2.

The first experimental gasifier was installed at the BASF in Ludwigshafen in 1922/23. The first commercial Winkler plant began operation in 1926 at the then-existing Merseburg ammonia works (Leunawerke).

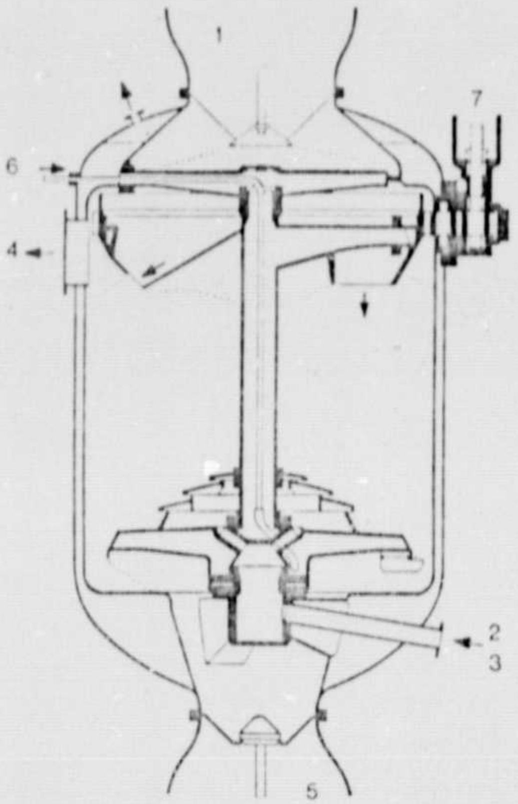


Figure 1. The Lurgi pressure gasifier.

Key: 1 - fuel inlet; 2 - oxygen; 3 - steam; 4 - raw gas; 5 - ash outlet; 6 - cooling water; 7 - drive.

There are at present numerous experimental programs for fluidized-bed gasification processes in various stages of development, including the processes designated CO_2 Acceptor, Synthane, Hygas, Hydrane, Exxon, and others.

3.3. Entrainment Gasification

In this type of gasification the finely-milled fuel is carried into and through the gasification chamber by the gasifying agents and gaseous gasification products. The relative velocity

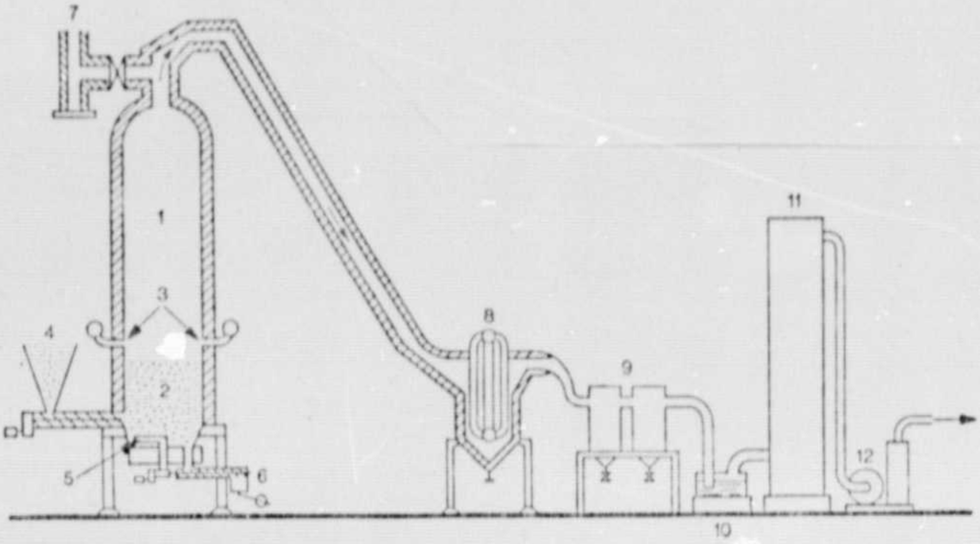


Figure 2. The Winkler gasifier.

Key: 1 - Winkler gasifier; 2 - fluidized bed; 3 - upper tuyeres; 4 - fuel inlet; 5 - agitator; 6 - ash outlet; 7 - flare; 8 - waste-heat boiler; 9 - dust collector; 10 - condensing vessel; 11 - wash tower; 12 - disintegrator.

between the cost-dust particles and the gas streams carrying them is negligible and is not a significant factor during the degasification and gasification of the solid material in the gasifier. The only commercial process of this type is the Koppers-Totzek process (Fig. 3).

The initial attempts to gasify coal dust entrained in a stream of oxygen mixed with high-temperature preheated steam (1100-1200°) were made in 1939-1944 by Friedrich Totzek and other workers with the then-existing Heinrich Koppers GmbH at German gasoline synthesis plants. In 1949/50 these experiments were continued in the U.S. in Louisiana and Missouri, where work was done on gasifying pulverized coal entrained in a stream of oxygen and steam. The pilot plant in Louisiana had been built for a coal throughput of up to 1 ton/hr by Koppers Company Inc., Pittsburgh, according to drawings and design data supplied by Heinrich Koppers GmbH. The plant, which was located on a vast /85 testing site together with other coal conversion facilities, was funded and operated by the U.S. Bureau of Mines. Engineers from both Koppers firms cooperated in the project.

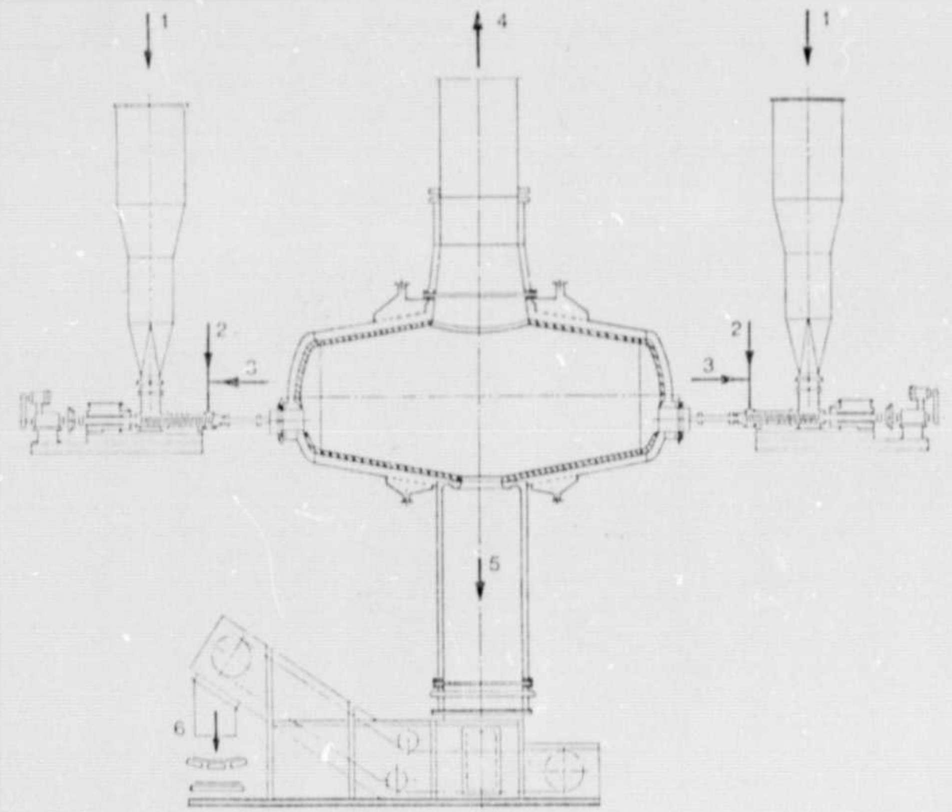


Figure 3. The Koppers-Totzek gasifier.

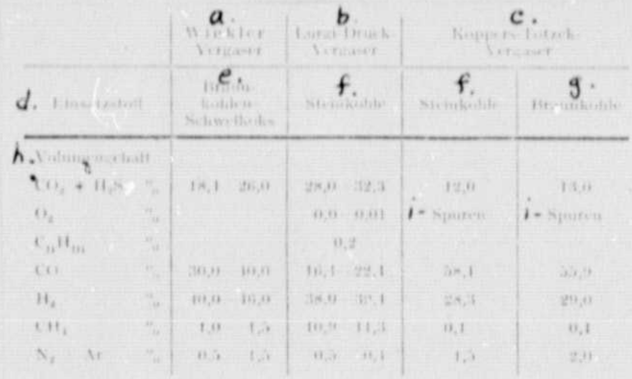
Key: 1 - coal dust; 2 - oxygen; 3 - steam; 4 - raw gas; 5 - liquid slag; 6 - granulated slag.

After completion of the experimental phase, all commercial plants were designed and built by Heinrich Koppers GmbH, and the first of these plants went operational at Typpi Oy, Finland, in 1952. The Koppers-Totzek process us thus by far the newest of the commercial gasification processes with oxygen.

There have also been parallel developments in this area, some of which date back 20 years and more. Examples are the Texaco, Rummel and Bigas processes.

Table 1 gives raw-gas analyses for the three gasification processes discussed so far.

Table 1. Raw-Gas Analysis



	a. Winkler-Vergaser	b. Lurgi-Druck-Vergaser	c. Koppers-Totzek-Vergaser	
d. Einsatzstoff	e. Braunkohlen-Schwelkoks	f. Steinkohle	f. Steinkohle	g. Braunkohle
h. Volumengehalt				
$CO_2 + H_2S$ %	18,1 26,0	28,0 32,3	12,0	13,0
O_2 %		0,0 0,01	i. Spuren	i. Spuren
C_nH_m %		0,2		
CO %	30,0 40,0	16,1 22,1	58,1	55,9
H_2 %	40,0 46,0	38,0 39,1	28,3	29,0
CH_4 %	1,0 1,5	10,9 11,3	0,1	0,1
N_2 Ar %	0,5 1,5	0,5 0,4	1,5	2,0

Key:
- a. Winkler gasifier
- b. Lurgi pressure gasifier
- c. Koppers-Totzek gasifier
- d. Feedstock
- e. Lignite low-temperature coke
- f. Anthracite
- g. Lignite
- h. Volume content
- i. Trace

Note: Commas in tabulated material are equivalent to decimal points.

In addition to these three basic types of coal gasification with oxygen, there are numerous mixed processes with special features, such as the presence of a molten phase (liquid slag, salt melt, iron melt) in the gasification chamber to intensify the exchange of heat and mass between the solid material and the gaseous reactants. So far none of these processes has reached commercial maturity, and some have not progressed beyond the pilot scale.

It is noteworthy that the three aforementioned commercial gasification processes are all of German origin, and that each was developed in pursuit of a different goal. The initial goal of the Lurgi process was to produce a heating gas (substitute town gas) from lignite. The Winkler process was originally developed to produce a high-grade activated carbon and was then directed toward making synthesis gas for the production of gasoline. Finally, the Koppers-Totzek process arose from the need to utilize the coal dust formed during the steam gasification of briquets for the production of hydrogen.

4. What Must a Gasification Process Accomplish?

The requirements placed on a technically and economically feasible coal gasification process can be summarized in five main points:

1. The properties of the coal -- "coal" referring here to any of various solid fuels such as anthracite, lignite, low-temperature coke, petroleum coke, etc. -- should have no significant influence on the practicability of the gasification process. In other words, coal properties such as bakability, content of volatiles, reactivity, ash content, ash-melting characteristics, granulation and grain distribution, mechanical strength at high and low temperatures, etc. should be no reason to exclude a certain type or grade of coal from gasification. /86

2. Ideally the gasification should yield only useful gas and ash, but no by-products which are difficult to reprocess or can be marketed only with considerable effort and expense.

3. The gasification should cause a minimal degree of environmental pollution. It should produce only minimal amounts of gaseous and liquid pollutants which can be disposed of at reasonable cost.

4. Gasification plants generally exist either to provide a public gas supply or to supply large, capital-intensive chemical complexes with coal gas. In both cases a reliable gas supply is of paramount importance; that is, there must be a minimum of production interruptions determined by the gasification itself and ancillary gas treatment facilities.

5. The costs of coal-to-gas conversion must enable the coal-derived gas to be competitive with corresponding gases produced from other energy sources.

The Winkler, Lurgi and Koppers-Totzek gasification processes do not all satisfy these requirements in the same way or to the same degree. The decision as to which process is best in a given case depends on a number of secondary questions, such as:

What is the ultimate purpose of the product gas?

Hydrogen production:	hydrogenation ammonia
Synthesis gas production:	methanol gasoline
Fuel gas production:	substitute natural gas simple fuel gas

Is it certain that the gasification plant will receive coal of a constant quality over a long period of time, or are frequent, possibly short-term variations in important coal properties likely?

Is it necessary to utilize the entire output of a coal mine for gasification, rather than certain grain sizes?

Do local conditons suggest profitable possibilities for utilizing the by-products of a certain gasification process?

Against this background the Koppers-Totzek process has emerged as a major technique for producing ammonia from coal. The Lurgi process has become important in the production of high-heating-value gases. This does not imply, however, that the future of coal gasification is decided, either in terms of process technology or the potential applications of the product gas.

We shall cite some statistics to show the current status of the Koppers-Totzek process:

Table 2 gives a listing of all Koppers-Totzek gasification plants which have been built or are now under construction.

Table 3 lists the coal-based ammonia plants built since 1950 which employ one of the three coal gasification processes for the production of synthesis hydrogen.

As the Tables show, the Koppers-Totzek process has been employed exclusively for gas production in coal-based ammonia plants located, understandably enough, in countries where coal is available at low cost. This is due primarily to previous price conditions in the energy sector, as well as political and monetary factors. However, the strong price shifts during recent years for the various energy sources, whether solid, liquid or gaseous, and the general price trend for large-scale chemical projects make the Koppers-Totzek process attractive for many other applications as well (see Fig. 4).

Table 2. Plants for the Gasification of Various Fuels by the Koppers-Totzek Process

	a. Vergaser ausgelegt für	b. Anzahl der Vergasereinheiten	c. für (CO + H_2) im Normalzustand m^3/d	d. Verwendungszweck des Synthesegases	e. Auftragsjahr
Charbonnages de France, Mazingarbe, Frankreich – f	q. Steinkohlenstaub, Koksofengas, Restgas	1	75 150 000	z. Methanol- und Ammoniaksynthese	1949
Typpi Oy, Oulu, Finnland – g	r. Steinkohlenstaub, Öl	3	140 000	aa. Ammoniaksynthese	1950
Nihon Suiso Kogyo Kaisha, Ltd., Tokyo, Japan	s. Steinkohlenstaub	1	210 000	Ammoniaksynthese	1954
Empresa Nacional Calvo Sotelo Puentes, Spanien – h	t. Lignitstaub	3	212 000	Ammoniaksynthese	1954
Typpi Oy, Oulu, Finnland – g	r. Steinkohlenstaub, Öl	2	140 000	Ammoniaksynthese	1955
S. A. Union Chimique Belge, Zandvoorde, Belgien – i	u. Bunker-C-Öl, Anlage ergänzungsfähig für Kohlenstaub-Vergasung	2	176 000	Ammoniaksynthese	1955
Amoniaco Português S.A.R.L., Estarreja, Portugal	v. Schwerbenzin, Anlage ergänzungsfähig für Lignit- und Anthrazitstaub-Vergasung	1	169 000	Ammoniaksynthese	1956
j. Griechische Regierung Ptolemais, Griechenland	w. Lignitstaub, Bunker-C-Öl	4	629 000	Ammoniaksynthese	1959
Société el Nasr d'Engrais et d'Industries Chimiques, [illegible], Ägypten – k	x. Raffinerierestgase, Flüssiggas, Leichtbenzin	3	778 000	Ammoniaksynthese	1963
Chemical Fertilizer Company Ltd. Mae Moh, [illegible], Thailand	t. Lignitstaub	1	217 000	Ammoniaksynthese	1963
Azot Sanayii T.A.S., Ankara, Kutahya, Türkei – l	t. Lignitstaub	1	175 000	Ammoniaksynthese	1965
Chemieanlagen Export-Import GmbH Werk VEB Zeitz, DDR – m	y. Vakuumrückstand Heizöl	2	300 000	bb. Rohgas für Hydrierwasserstoff	1966
Kobe Steel Ltd., für Industrial Development Corp. of Zambia Kafue bei Lusaka, Zambia	s. Steinkohlenstaub	1	211 320	aa. Ammoniaksynthese	1967
Nitrogenous Fertilizers Industry S.A. Ptolemais, Griechenland – n	t. Lignitstaub	1	135 000	Ammoniaksynthese	1969
The Fertilizer Corporation of India Ltd., Ramagundam, Indien – o	s. Steinkohlenstaub	3	2 000 000	Ammoniaksynthese	1969
The Fertilizer Corp. of India Ltd., Talcher, Indien – o	s. Steinkohlenstaub	3	2 000 000	Ammoniaksynthese	1970
Nitrogenous Fertilizers Industry S.A. Ptolemais, Griechenland – n	t. Lignitstaub	1	242 000	Ammoniaksynthese	1970
The Fertilizer Corporation of India Ltd., Korba Indien – o	s. Steinkohlenstaub	3	2 000 000	Ammoniaksynthese	1972
AE & CI Limited Modderfontein, Südafrika – p	s. Steinkohlenstaub	6	2 150 000	Ammoniaksynthese	1972
Indeco Chemicals Ltd., Kafue, Zambia	s. Steinkohlenstaub	2	220 000	Ammoniaksynthese	1974

Key:

a. Gasifier designed for
b. Number of gasifiers
c. m^3/day for ($CO+H_2$) under normal conditions
d. Application of synthesis gas
e. Year of commission
f. France
g. Finland
h. Spain
i. Belgium
j. Greek government Ptolemais, Greece
k. Egypt

q. Anthracite dust, coke oven gas, residual gas
r. Anthracite dust, oil
s. Anthracite dust
t. Lignite dust
u. Bunker-C oil, plant can be modified for coal-dust gasification
v. Heavy gasoline, plant can be modified for lignite- and anthracite-dust gasification
w. Lignite dust, bunker-C oil
x. Residual refinery gases, liquid gas, light gasoline
y. Vacuum residue, fuel oil

[Key continued next page]

[Key to Tab. 2, continued]

l. Turkey
m. East Germany
n. Greece
o. India
p. South Africa

z. Methanol and ammonia synthesis
aa. Ammonia synthesis
bb. Raw gas for hydrogenation-process hydrogen

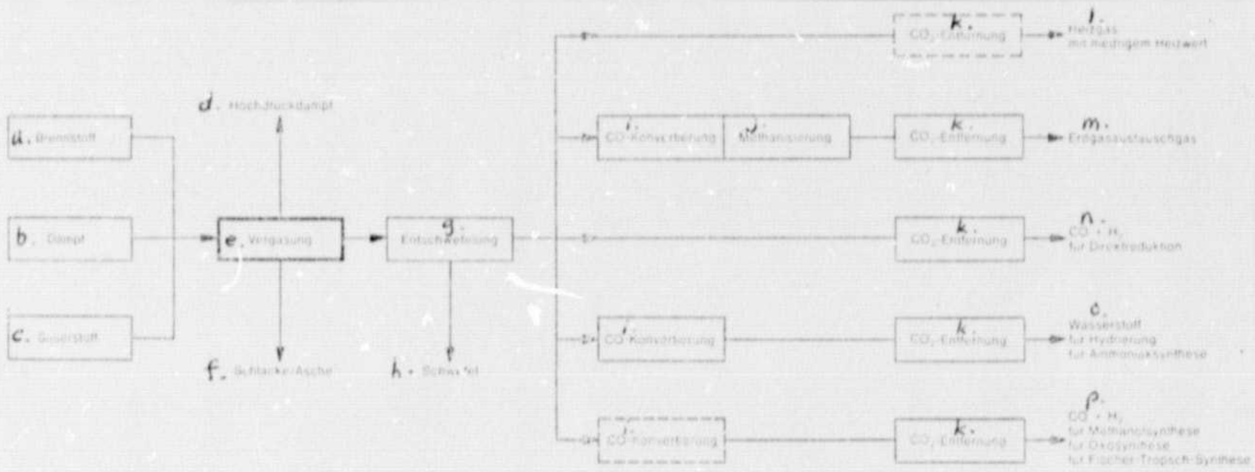


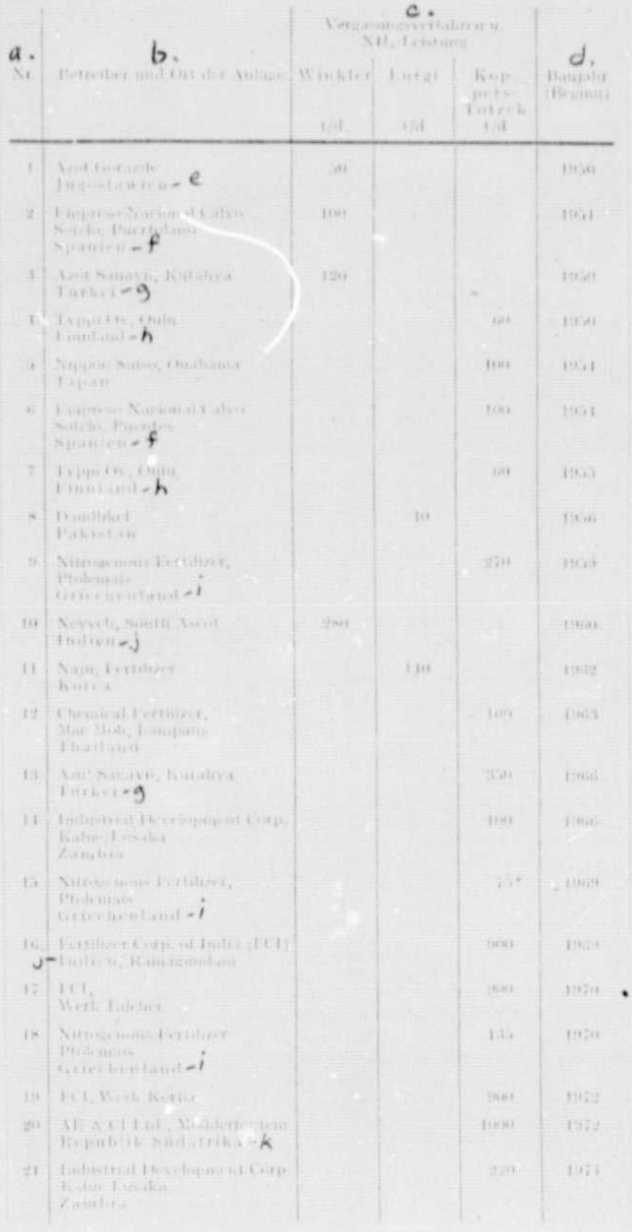
Figure 4. Possible applications of the Koppers-Totzek process.

Key:

a. Fuel
b. Steam
c. Oxygen
d. High-pressure steam
e. Gasification
f. Slag/ash
g. Desulfurization
h. Sulfur
i. CO conversion

j. Methanation
k. CO_2 removal
l. Low-heating-value fuel gas
m. Substitute natural gas
n. CO + H_2 for direct reduction
o. Hydrogen for hydrogenation, for ammonia synthesis
p. CO + H_2 for methanol synthesis, for oxosynthesis, for Fischer-Tropsch synthesis

Table 3. Coal-Based Synthetic Ammonia Plants
(as of January 3, 1976)



a. Nr.	b. Betreiber und Ort der Anlage	c. Vergasungsverfahren u. NH_3-Leistung Winkler t/d	Lurgi t/d	Koppers-Totzek t/d	d. Baujahr (Beginn)
1	Azot Gorazde Jugoslawien -e	50			1950
2	Empresa Nacional Calvo Sotelo, Puertollano Spanien -f	100			1951
3	Azot Sanayii, Kütahya Türkei -g	120			1950
4	Typpi Oy, Oulu Finnland -h			60	1950
5	Nippon Suiso, Onahama Japan			100	1954
6	Empresa Nacional Calvo Sotelo, Puentes Spanien -f			100	1954
7	Typpi Oy, Oulu Finnland -h			60	1955
8	Daudkhel Pakistan		10		1956
9	Nitrogenous Fertilizer, Ptolemais Griechenland -i			270	1959
10	Neyveli, South Arcot Indien -j	280			1960
11	Naju, Fertilizer Korea		110		1962
12	Chemical Fertilizer, Mae Moh, Lampang Thailand			100	1963
13	Azot Sanayii, Kütahya Türkei -g			350	1966
14	Industrial Development Corp., Kafue, Lusaka Zambia			100	1966
15	Nitrogenous Fertilizer, Ptolemais Griechenland -i			75*	1969
16	Fertilizer Corp. of India (FCI) j- Indien, Ramagundam			900	1969
17	FCI, Werk Talcher			900	1970
18	Nitrogenous Fertilizer Ptolemais Griechenland -i			135	1970
19	FCI, Werk Korba			900	1972
20	AE & CI Ltd., Modderfontein Republik Südafrika -k			1000	1972
21	Industrial Development Corp. Kafue Lusaka Zambia			220	1974

Key: a. No.
b. Operating concern and location of plant
c. Gasification process and NH_3 output
d. Year construction began
e. Yugoslavia
f. Spain
g. Turkey
h. Finland
i. Greece
j. India
k. Republic of South Africa
*) The ammonia production given equals the design output of synthesis gas.

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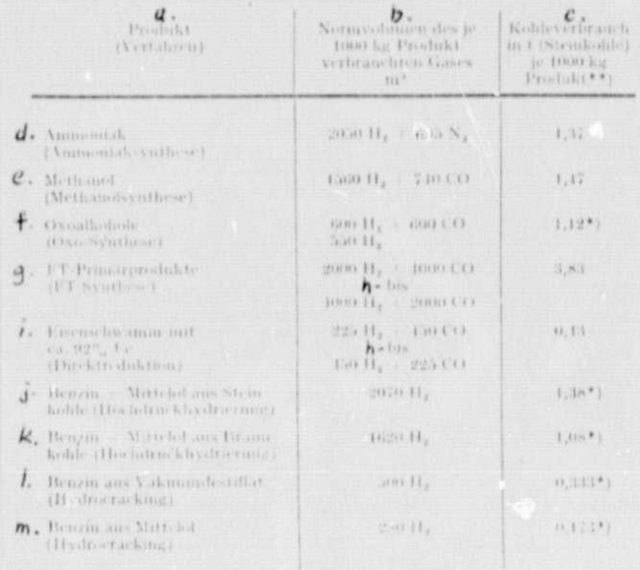
The specific quantities of coal required for the various applications shown in Fig. 4 are listed in Table 4. /87

Table 4. Specific Energy Consumption for Various Hydrogenation Processes



a. Produkt (Verfahren)	b. Normvolumen des je 1000 kg Produkt verbrauchten Gases m³	c. Kohleverbrauch in t (Steinkohle) je 1000 kg Produkt**)
d. Ammoniak (Ammoniaksynthese)	2050 H_2 + 685 N_2	1,37
e. Methanol (Methanolsynthese)	1500 H_2 + 740 CO	1,47
f. Oxoalkohole (Oxo-Synthese)	600 H_2 + 600 CO 550 H_2	1,12*)
g. FT-Primärprodukte (FT-Synthese)	2000 H_2 + 1000 CO h- bis 1000 H_2 + 2000 CO	3,83
i. Eisenschwamm mit ca. 92% Fe (Direktreduktion)	225 H_2 + 150 CO h- bis 150 H_2 + 225 CO	0,43
j. Benzin + Mittelöl aus Steinkohle (Hochdruckhydrierung)	2070 H_2	1,38*)
k. Benzin + Mittelöl aus Braunkohle (Hochdruckhydrierung)	1620 H_2	1,08*)
l. Benzin aus Vakuumdestillat (Hydrocracking)	500 H_2	0,333*)
m. Benzin aus Mittelöl (Hydrocracking)	260 H_2	0,173*)

Key:

a. Product (process)

b. Normal volume of gas consumed per 1000 kg product, in m^3

c. Coal consumption in tons (anthracite) per 1000 kg product**)

d. Ammonia (ammonia synthesis)

e. Methanol (methanol synthesis)

f. Oxoalcohols (oxosynthesis)

g. FT primary products (FT synthesis)

h. to

i. Iron sponge with approx. 92% Fe (direct reduction)

j. Gasoline + medium oil from anthracite (high-pressure hydrogenation)

k. Gasoline + medium oil from lignite (high-pressure hydrogenation)

l. Gasoline from vacuum distillate (hydrocracking)

m. Gasoline from medium oil (hydrocracking)

*) Here only the coal used for the production of synthesis gas or hydrogenation-process hydrogen is taken into account.

**) Assuming a coal heating value H_u = 77.92 kWh/kg (6.7 Mcal/kg).

In Table 4 the specific gas or coal consumption indicated pertains only to the actual chemical reaction involved. It does not include the additional energy requirements for other plant facilities.

It can be seen from Table 4 that Fischer-Tropsch synthesis has a very high specific coal consumption. Hence the cost of the coal is a decisive factor. Fischer-Tropsch plants are not economically feasible, based on current European anthracite prices, and this situation would not change significantly even if the cost of crude oil were to double with no increase in the cost of anthracite.

We shall now describe three of the applications of the Koppers-Totzek process given in Fig. 4 in somewhat greater detail: the production of a medium-heating-value fuel gas, the synthesis of ammonia, and the direct gaseous reduction of iron ore.

5. Fuel Gas Production

The raw gas from a Koppers-Totzek coal gasification plant normally has a heating value from 3.024 to 3.256 kWh/m^3 (2.6 to 2.8 Mcal/m^3), which is between the heating value of furnace gas or coke-generator gas and the once-popular town gas.

As a result of the high mean gasification temperature of about 1500° C, the raw gas contains no condensable components other than water vapor. More than 85% of the gas volume consists of carbon monoxide and hydrogen; the rest is comprised of carbon dioxide, nitrogen, argon and sulfur compounds, mainly hydrogen sulfide along with a small amount of carbon oxysulfide. After the gas has left the gasifier at an average temperature of about 1500° C and has released its sensible heat in a high-pressure boiler (up to 100 bar steam pressure), it is cleaned of /88 dust, which consists of ash and some coal and coke dust, in a wet washing system. The purity achieved by this wet cleaning process (normally below about 20 mg dust/m^3) is generally sufficient to enable the gas to be used as a fuel gas. The gas is then compressed to the working pressure of several bars required in the gas handling and distribution system. Next the sulfur is removed from the gas by any one of numerous physical, chemical or combined desulfurization processes. At this point the gas is in compliance with air purity standards.

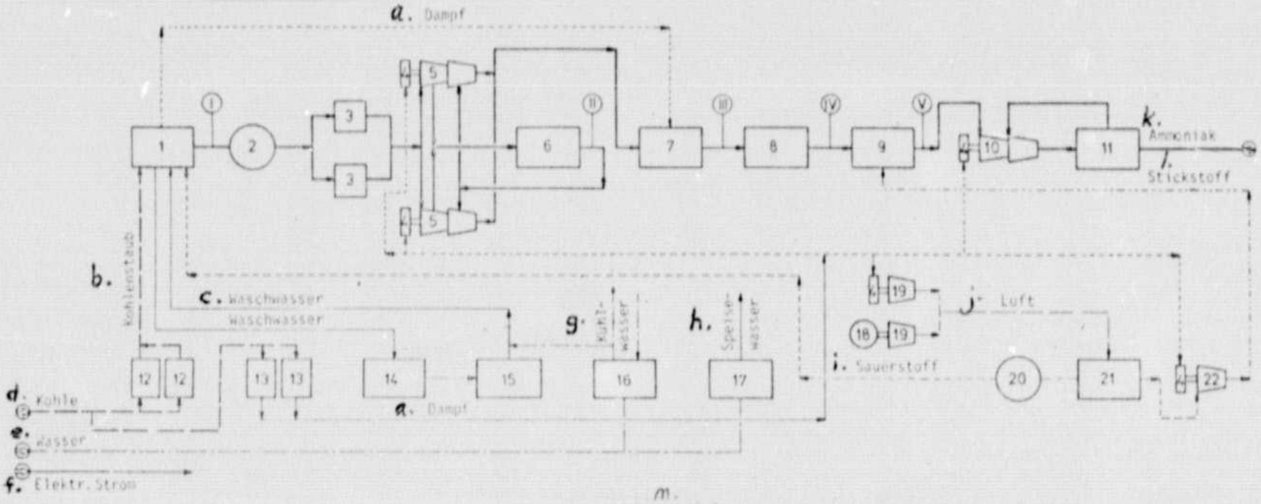
The gas can be utilized in almost all industrial situations which previously required the use of coke-oven or natural gas. In most cases only a modification of the burners is necessary. The conversion from natural-gas-fired boilers to Koppers-Totzek gas offers still another energy advantage: the thermal efficiency of gas-fired boilers attains a strong maximum at fuel-gas heating values of about 3.140 kWh/m^3 (2.7 Mcal/m^3), falling off sharply at lower values and somewhat more gradually at higher values.

The extent to which natural gas can or should be replaced by Koppers-Totzek gas as an industrial fuel depends primarily on the impact of future natural gas shortages (already acute in the U.S.) and associated production losses on the profitability of the affected manufacturing plants, and whether the cost of the desulfurized Koppers-Totzek gas can maintain the profitability of these plants, at least in part. One must not only compare the cost of the thermal unit of Koppers-Totzek gas with that of the natural-gas thermal unit, but also take into account the adverse effect of production capacities left unutilized due to gas shortages. It would be inadvisable in any case to convert Koppers-Totzek gas completely into synthetic natural gas (SNG) just so it can be burned in existing ovens, etc. in places where natural gas is scarce.

6. Ammonia from Coal

Ammonia is synthesized from hydrogen and nitrogen in a volume ratio of 3:1. The Koppers-Totzek gas, which contains up to 60% CO by volume (depending on the initial fuel) in accordance with the homogenous water gas equilibrium at the temperature achieved at the end of gasification, must therefore undergo extensive processing to obtain pure hydrogen from the complex gas mixture. Nitrogen is obtained from the air separation unit, which supplies the oxygen for gasification.

Fig. 5 shows the block diagram of an ammonia plant which produces 1000 tons of water-free ammonia per day based on the Koppers-Totzek gasification of anthracite.



m. Gasanalyse

n. Volumgehalt		I	II	III	IV	V
CO_2	%	13,37	11,27	11,29		
$H_2S + COS$	%	0,59				
CO	%	54,45	56,02	3,00	5,02	
H_2	%	30,00	31,06	54,63	93,14	75,0
N_2	%	0,95	0,98	0,64	1,12	25,0
Ar	%	0,54	0,57	0,37	0,61	
CH_4	%	0,10	0,10	0,07	0,11	
o. Druck	p. bar	0,86	28,9	49,2	47,1	45,1
q. Temperatur	°C	32	28	35	56	23

Figure 5. Modderfontein ammonia plant (South Africa).
[Key next page]

Key to Fig. 5: a - steam; b - coal dust; c - wash water; d - coal; e - water; f - electric current; g - cooling water; h - feedwater; i - oxygen; j - air; k - ammonia; l - nitrogen; m - gas analysis; n - volume content; o - pressure; p - bars; q - temperature. 1 - coal dust gasification; 2 - raw gas container; 3 - electrostatic filter; 4 - turbine; 5 - raw gas compressor; 6 - sulfur removal; 7 - carbon monoxide conversion; 8 - carbon dioxide wash; 9 - liquid nitrogen wash; 10 - synthesis gas compressor; 11 - ammonia synthesis; 12 - coal preparation; 13 - steam boiler; 14 - settling tank; 15 - wash water system; 16 - cooling water system; 17 - feedwater treatment; 18 - motor; 19 - air compressor; 20 - oxygen container; 21 - air separation; 22 - nitrogen compressor.

The plant consumes about 2160 tons of anthracite per day at full-capacity operation. This includes the quantity of fuel consumed in a steam generator for producing the driving steam for the turbines of the large compressors. Besides coal, the plant is fed with: water, electric energy, chemicals and catalysts. /89 Besides the useful products (ammonia and sulfur), the plant produces as waste materials ash, slag and, of course, large amounts of carbon dioxide.

The gasification section consists of six two-headed gasifiers whose feed coal is processed by a milling and drying unit and whose output is treated by washing facilities for the removal of finely-divided contaminants. Electrostatic filters normally reduce the dust content of the gases to less than 0.5 mg per cubic meter. The waste-heat boilers, which are coupled directly to the gasifiers in the gas stream, produce a saturated steam at 55 bar. Steam at 2.5 bar is produced in the double cooling jacket of each gasifier. One of the gasifiers, still without its insulation prior to installation, is shown in Fig. 6.

The gas is next compressed to about 30 bar in the first /90 stages of the raw-gas compressors and is then reacted with methanol at low temperature to reduce the sulfur content to 2 ppm. After desulfurization the gas is further compressed to 52 bar by

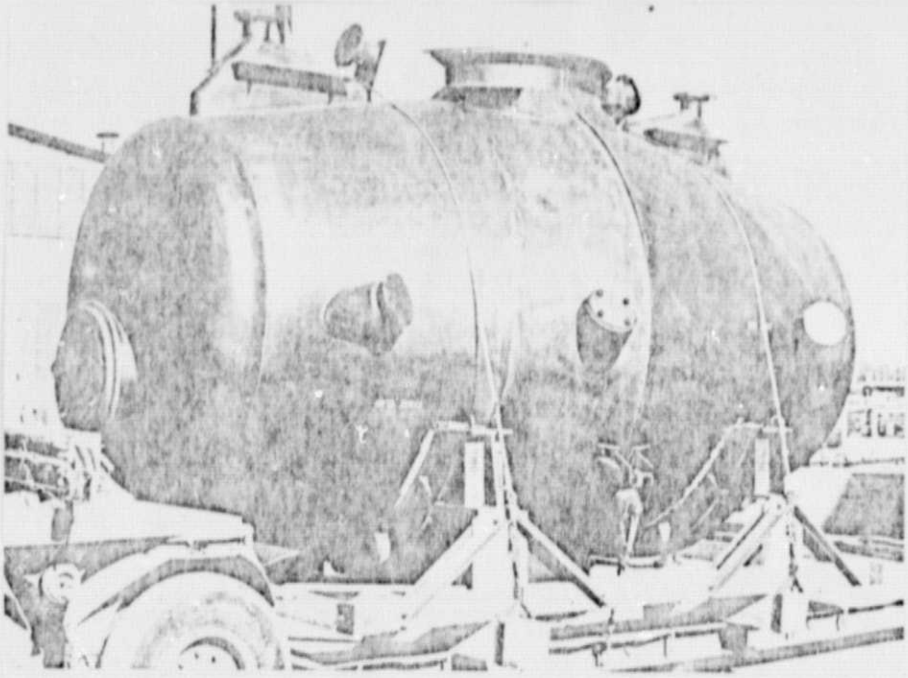


Figure 6. Two-headed gasifier prior to installation, without insulation.

another stage of the raw-gas compressors. This is followed by the conversion step in which the carbon monoxide is largely converted into corresponding quantities of hydrogen and carbon dioxide by a catalytic reaction with water vapor. The carbon dioxide is then separated by cold washing with methanol. At this point the gas is more than 93% hydrogen by volume, with carbon monoxide and methane as the only remaining impurities. These are removed by washing with liquid nitrogen. The 3:1 hydrogen-to-nitrogen ratio necessary for ammonia synthesis is also obtained in this step. Next the synthesis gas mixture is compressed to 220 bar; this step is followed by catalytic ammonia synthesis. A perspective drawing of the ammonia plant described is shown in Fig. 7.

Conventional two-headed gasifiers are capable of producing up to about 22,000 m^3 raw gas per hour. This corresponds to a

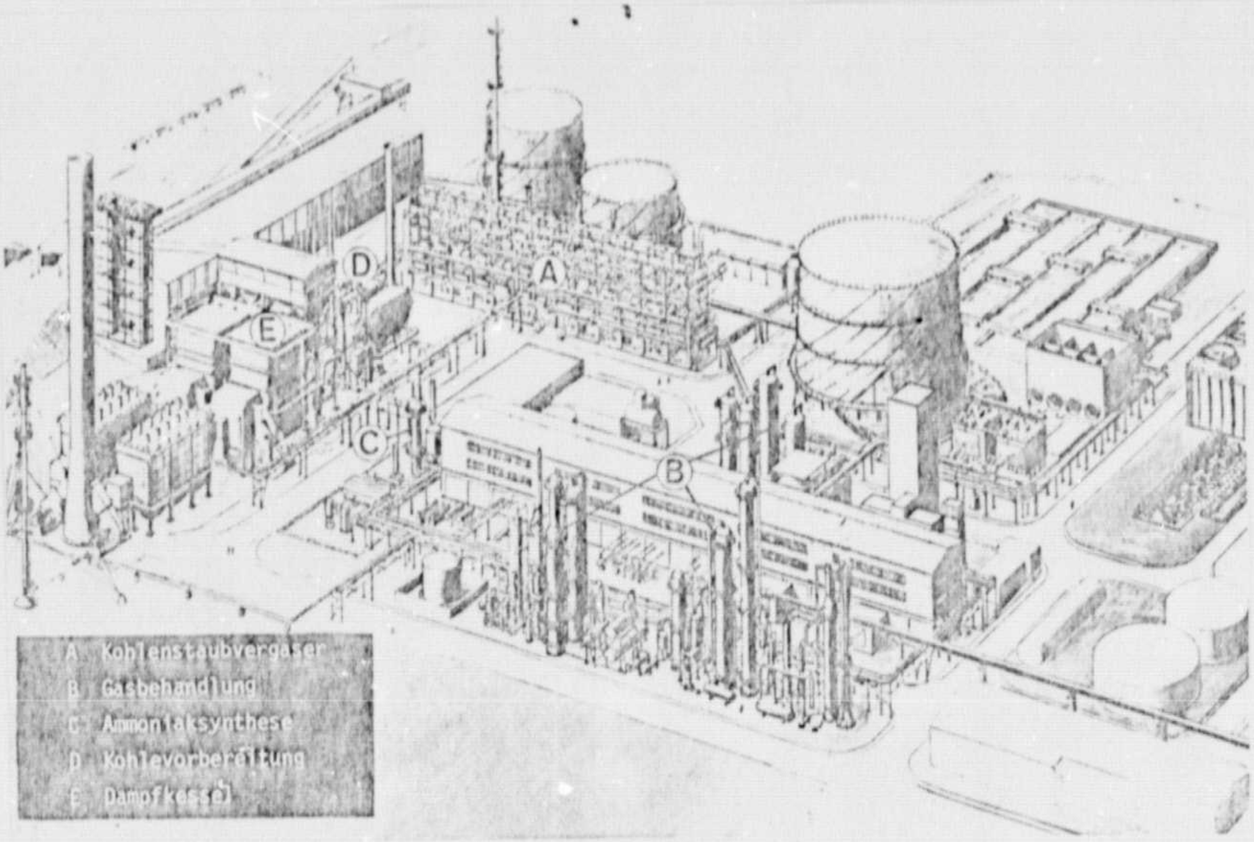


Figure 7. Perspective drawing of an ammonia plant.

Key: A. Coal-dust gasifier
B. Gas treatment
C. Ammonia synthesis
D. Coal preparation
E. Steam boiler

coal throughput (anthracite with about 2% lignite and 8% moisture) of about 12 tons/hour, distributed between two burner heads. In terms of design, there are no basic obstacles to increasing the number of burner heads per gasifier to a multiple of two, although a limit will be set by the sizing of the high-pressure boiler installed immediately after the gasifier. Gasifiers with four burner heads each are already under construction at ammonia plants in India. The first of these plants will soon begin operation at the Talcher plant in Orissa. Fig. 8 shows a four-headed gasifier during installation.

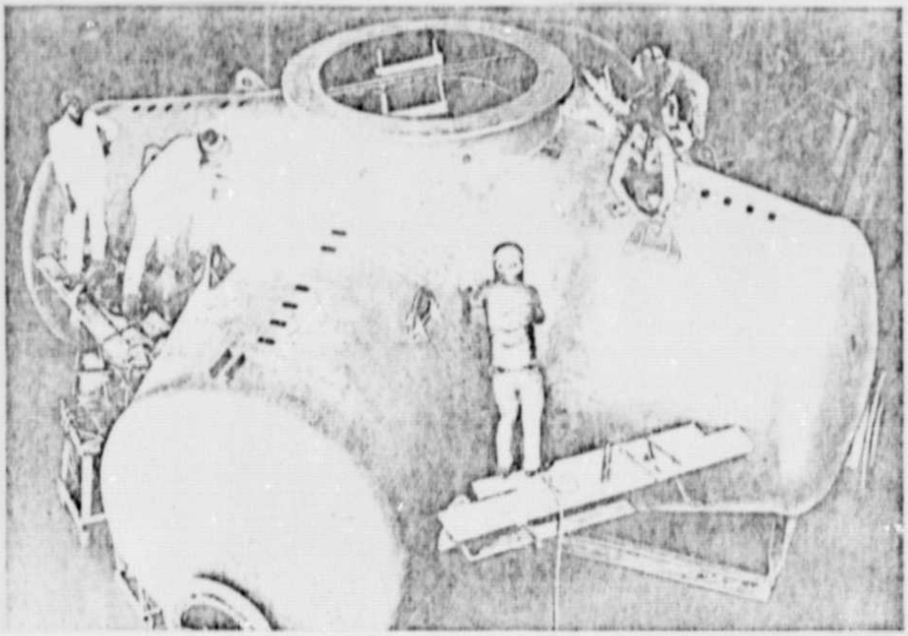


Figure 8. Four-headed gasifier prior to installation.

7. Iron Sponge Production (Direct Reduction)

It has been long known that iron ore can be converted by reducing gases into a largely metallized product without smelting. However, we believe that Friedrich Totzek was the first to suggest, in a 1949 patent [15], that the CO- and H_2-rich gas produced by coal dust gasification under pressure could be fed up into a pressurized shaft furnace to react with the descending iron ore in a countercurrent operation. The patent pertained to:

"A process for the production of iron sponge by the reduction of iron ore by means of gases containing carbon monoxide and hydrogen at temperatures below the melting point of iron and iron ore, whereby a pulverized fuel is first gasified with oxygen in a non-ore-reducing apparatus to yield a reducing gas with a carbon monoxide and hydrogen content exceeding 70%, and preferably 80%, and whose temperature corresponds to that to be employed during reduction of the ore to iron sponge, preferably about 1000° C; /91 the reducing gas is then fed directly from the gasifying apparatus into the reducing apparatus, where it is brought into contact with the ore."

Aside from special cases, direct reduction in general and direct reduction with gas in particular have not met with wide approvel. Blast furnace processes were so greatly improved after the last war (larger throughputs, improvement of load through use of sinter and pellets, increase of wind temperature, counterpressure at the throat, reduction of specific coke consumption by oil injection) that it appeared less attractive to reduce iron ore by any means other than the blast furnace. However, two factors have changed this situation in recent years:

1. Good coking coal is absent or scarce in many countries with iron ore reserves.

2. For economic reasons the development and expansion of an iron industry in developing nations or smaller industrial nations could be best accomplished on the basis of reducing units which are themselves economical (producing 300,000 to 500,000 tons of iron sponge per year) and can be enlarged in stages, rather than large blast furnaces which smelt 5000 tons or more raw iron per day.

While all gas reduction plants to date have employed a reducing gas manufactured from hydrocarbons (natural gas, liquid gas, petroleum), reducing gas produced from coal by the Totzek process has recently become an attractive alternative in cases where hydrocarbons are unavailable and coal is plentiful, or if coal is available domestically at an attractive per-thermal-unit cost due to recent shifts in energy prices.

A modern direct-reduction plant based on a reducing gas produced by the Koppers-Totzek process is shown schematically in Fig. 9.

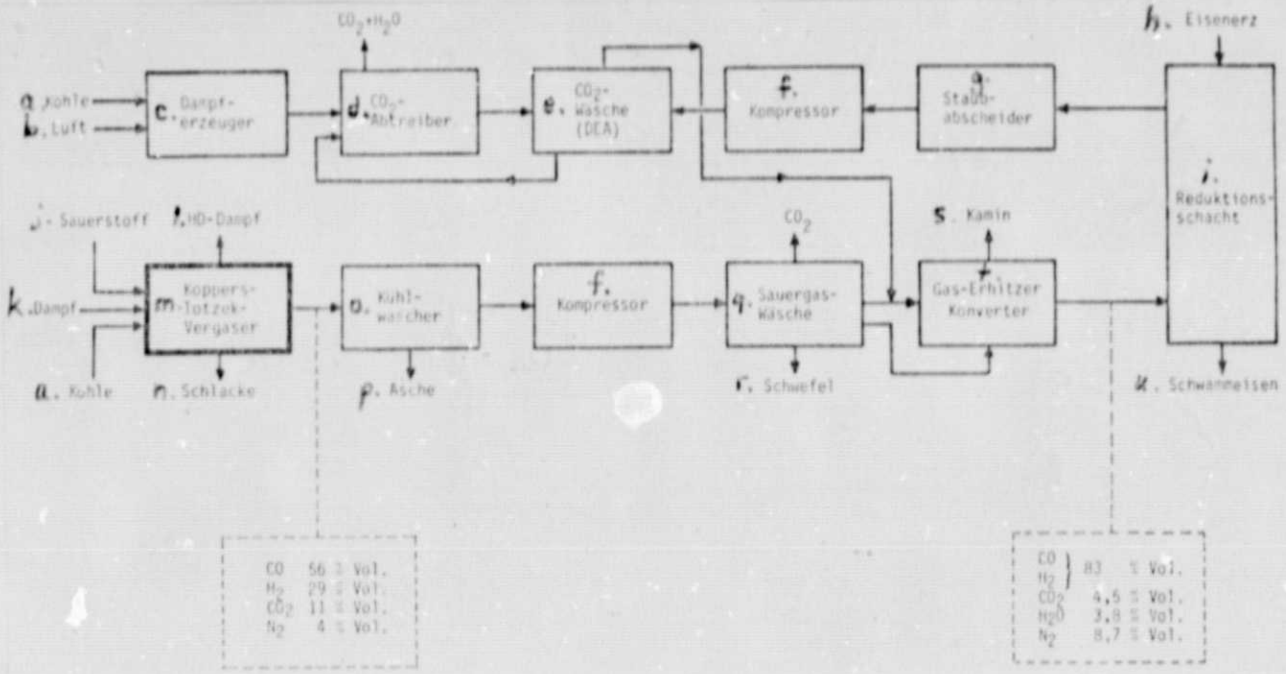


Figure 9. Direct reduction of iron ore by means of Koppers-Totzek gas.

Key:
a. Coal
b. Air
c. Steam generator
d. CO_2 distilling unit
e. CO_2 scrubber (DEA)
f. Compressor
g. Dust collector
h. Iron ore
i. Reduction shaft
j. Oxygen
k. Steam
l. High-pressure steam
m. Koppers-Totzek gasifier
n. Slag
o. Cold scrubber
p. Ash
q. Acid gas scrubber
r. Sulfur
s. Shaft
t. Gas heater, converter
u. Iron sponge

8. Entrainment Gasification under Pressure

The Koppers-Totzek process is carried out at a pressure that deviates only slightly from atmospheric. This has several advantages:

1. The apparatus for feeding the coal dust and removing the solid or liquid gasification residue is relatively simple.
2. The gasification zone can easily be monitored optically during operation, enabling appropriate measures to be taken rapidly and without system shutdown in trouble situations.
3. Repair work is generally faster and simpler in unpressurized gasifiers than in pressure gasifiers.

Opposing these advantages is the fact that the entire product gas must be subsequently brought to a pressure suitable for further processing (generally greater than 15 bar). Thus, it was proposed quite early at Heinrich Koppers GmbH that entrainment gasification be carried out under pressure. As early as February, 1949, Joseph Daniels [16] suggested specifically how coal dust could be fed dry by mechanical means into a pressurized chamber for gasification with oxygen. A major problem which arose during later testing of this feed mechanism was how to prevent oxygen from escaping from the pressurized chamber into the coal-dust-filled charging bins of the low-pressure zone.

Pneumatic methods for feeding the coal against the gasifier pressure were also examined in detail. /92

When it became clear in the early 1950s that the commercial outlook of coal gasification was poor in the industrialized countries due to the abundance of inexpensive petroleum and petroleum products, but promising for the production of ammonia in developing nations where coal was inexpensive, efforts were made to simplify the gasification process in order to help solve the shortage of technical specialists in the developing nations. Thus, a significant portion of the personnel at the Koppers-Totzek gasification plants in Ptolemais, Greece, consisted at first of Macedonian tobacco farmers who tended their small fields near the plant in their free time, or to be more precise, who worked in the Ptolemais nitrogen plant when their farming chores permitted.

Still, since 1952 no fewer than eight ammonia plants have been equipped with Koppers-Totzek systems for coal gasification at normal pressure.

The situation has changed since the 1973 oil-price crisis, which will one day develop into an oil-supply crisis. The industrial countries have shown renewed interest in coal as a chemical raw material, and the pressure version of the Koppers-Totzek process has been re-examined. Pressure gasification would bring the following benefits:

- reduction of compression costs;
- higher degree of coal gasification, especially for coals with low reactivity;
- reduced heat losses;
- simplified gas purification;
- larger gas output per gasifier.

A price must be paid for these benefits. This consists chiefly in the development and testing of new instrumentation for the movement of solid materials (coal, ash) under pressure and for the monitoring and control of the gasification process. This instrumentation will surely lead to new designs for large-capacity pressure gasification plants.

To develop the pressure gasification process, the Heinrich Koppers GmbH and Shell International Petroleum in Maatachappij agreed in 1974 to cooperate in the joint construction of a demonstration plant. It is expected that this plant will yield the first results for a pressure version of the Koppers-Totzek process with a throughput of 6 ton/hour coal sometime in 1977. This demonstration plant is currently being constructed at the Shell petroleum refinery in Hamburg by the Krupp-Koppers GmbH.

9. The Future of Coal Gasification

In attempting to make a short-term and perhaps even a long-term prediction for the applications of coal gasification, one must take the following facts into consideration:

1. It is unlikely that known processes will be supplanted by new gasification technologies, since all future developments in coal refining must also be based on the natural laws governing the reaction of carbon with oxidizing or hydrogenating gaseous reactants. The commercial practice of existing coal gasification plants, regardless of their design details, has already shown what is possible and what is not. Thus, the basic principles have already been largely established in terms of technical development.

2. Petroleum and petroleum products as well as natural gas /93

> will meet more than half the energy demand of the industrialized nations for another 25 years and probably longer. This means that a coal-derived energy source or chemical raw material will have to compete with these natural hydrocarbon sources for some time to come. This is especially true if the enormous reserves of oil shale and tar sand can one day be converted into competetive oil sources.

It thus appears that for the next 20 years or so, economic considerations will generally be an overriding factor in the selection of a gasification process. Of course other considerations such as foreign exchange problems, the achievement of autarky, military needs, etc. may be more important in certain special projects, but even then the most economical alternative will be chosen, based in part on a comparison of the specific features of the various coal gasification processes.

Thus, before a coal gasification project is begun, the process supplier must be asked about its economic prospects so that substantial expenditures will not be made on a project that is without a real chance for commercial implementation.

If we apply the preceding considerations to future coal conversion projects, we may, with some caution, make the following predictions:

<u>Ammonia from Coal</u>

There are good prospects for this application of coal gasification in all countries where coal is inexpensive and petroleum and natural gas are, for whatever reason, unavailable or costly. The fact that the Koppers-Totzek process can gasify virtually any solid fuel, privided it can be finely milled, makes this process especially promising for the production of ammonia on a broad base of solid feed materials.

Fuel Gas for Power Plants and Industrial Furnaces

Calculations of power plant thermodynamics have shown that the economy of power generation from coal can be improved if the low-heating-value combustion gas produced by coal gasification is desulfurized and then burned in a gas turbine which feeds directly into a classic steam boiler (combined cycle). The pressure version of the Koppers-Totzek process now under development shows good medium-term prospects in this area due to its broad fuel base, especially if a way is found to carry out the pressure gasification with air without sacrificing gasification efficiency.

However, even the conventional form of the Koppers-Totzek process which has been commercially tested in more than 40 gasifiers could be employed for the production of a low-Btu fuel gas if it becomes necessary to compensate for an expected shortage of natural gas in the industrial sector. This possibility is receiving serious consideration in tne United States.

Reducing Gas for Iron Sponge Production

Up to now all gas reducing plants for the production of iron sponge have been based upon a reducing gas obtained by the cracking of natural gas or liquified gas or the oxygen gasification of heavy oil. As long as these hydrocarbons are readily available locally, there is little economic prospect for the use of coal to produce a reducing gas. However, there are many countries which have substantial reserves of non-coking coal along with large iron ore reserves, but possess no other natural fossil fuels. In these countries coal gasification will play a role in the relatively near future as a means of manufacturing reducing gas.

Gasification of Char and Petroleum Coke

In various coal conversion processes which are not yet utilized commercially (solvent refined coal, hydrogenating and non-hydrogenating carbonization), a residue is formed which contains more than 50% carbon and consists partly of extremely fine grains. Moreover, the delayed and fluid cokers used in oil refineries produce a low-ash coke which, due to its high sulfur content, will not be usable as a boiler fuel in the future. This suggests still another application for the Koppers-Totzek process, which yields a gas that can be utilized for all purposes which require either hydrogen or a mixture of carbon monoxide and hydrogen. A large-scale experiment done a few months ago with a Koppers-Totzek gasifier at an ammonia plant in Spain has shown that the char from the carbonization of coal as well as the petroleum coke from delayed and fluid cokers can be readily converted into raw synthesis gas.

Production of Synthetic Natural Gas (SNG)

If the desire or need arises to maintain a supply of the very convenient fuel natural gas to private consumers, and there are no economically- or technically-feasible alternatives to the use of coal, the thermal-unit cost of such a substitute natural gas will be quite high.

In the United States, for example, where the process will be based chiefly on anthracite, it is predicted that the cost of a thermal unit from coal-derived SNG will be from three to four times the cost for Texas natural gas.

In West Germany, with its higher initial level of all energy costs, it is predicted that synthetic gas from solid fuels will cost two to three times more than natural gas, depending on the cost of the coal-derived thermal unit. Due to this very high cost of SNG, combined with the contractually-ensured supply of natural gas to West Germany for the next 15 to 20 years, it appears that the conversion of coal to natural gas will be of secondary importance in the FRG during this period. Outside the FRG, for example in the United States, natural gas will probably be produced from coal on an expanded scale within the next ten years. /94

Methanol from Coal

At present, methanol synthesis gas is obtained by the steam reforming of the lighter hydrocarbons (methane, liquid gas, naphtha) and by the partial oxidation of heavy hydrocarbons. As long as these raw materials are available and the metahnol market is not greatly expanded by the use of methanol as an engine fuel, coal will not be seriously considered for the production of methanol synthesis gas. It is hardly likely, therefore, that this application of coal will achieve importance in the near future.

Liquefaction of Coal by the Fischer-Tropsch Process

As Table 4 shows, the specific consumption of coal per unit primary product is so large that nearly four tons of coal are required for one ton of primary product. The manufacture of engine fuels from coal by the Fischer-Tropsch process fails in the industrialized coal-producing countries simply by virtue of excessive coal costs.

If the Fischer-Tropsch plant is so designed that olefins and oxygen-bearing products (especially higher alcohols) are produced along with saturated hydrocarbons, plant operation may become economical if coal prices are low enough. This is apparently the reasoning behind plans to expand the Sasol plant, the only Fischer-Tropsch plant in the western world.

REFERENCES

1. Totzek, F., "On a universal process for the gasification of coal dust," Paper presented to the Energy Committee of the Commission for the Four Year Plan, Berlin, June 12, 1942 (typescript).

2. Totzek, F., "Coal dust gasification by the Koppers-Totzek process," Paper presented at the Haus der Technik, Essen, June 2, 1947; summary in Stahl und Eisen 66/67, 363 (1947).

3. Newman, L. L., "Oxygen in the production of hydrogen or synthesis gas," Ind. and Engn. Chem. 40, 559-582 (1948).

4. Krönig, W., Die katalytische Druckhydrierung von Kohlen, Teeren und Mineralölen [The Catalytic Pressure Hydrogenation of Coals, Tars and Mineral Oils], Berlin, Göttingen, Heidelberg, 1950.

5. Portatius, B. v., "Recent developments in the Winkler gasification process," In: Kolloq. Gasanwendung am 8. Nov. 1956 in Leipzig [Gas Applications Symposium of Nov. 8, 1956, in Leipzig], Berlin, 1957; Freiberg. Forschungsheft, Paper 69, pp. 5-25.

6. Schuster, F., "On calculating the equilibrium composition of gasification gases, Part 1," Gaswärme 7, 125-129 (1958).

7. Hubmann, O., "The gasification of solid fuels under pressure," Brennstoff-Chem. 40, 65-71 (1959).

8. Schellberg, W., "Coal-based ammonia plants," Paper presented at ICI Operating Sympos. in Amsterdam, April 4, 1974, "Ampo" Publ., Imp. Chem. Ind. Ltd., Paper 21.

9. Frendberg, A., "Performance characteristics of existing utility boilers when fired with low-Btu gas," Paper presented to Power Res. Inst. Conf. on Power Generation - Clean Fuels Today, April 8-10, 1974.

10. Goeke, E. K., "Status of coal dust gasification technology," Paper presented at Seminar by Fertilizer Assoc. of India, New Delhi, April 29-30, 1974, Paper II-2.

11. Kamody, J. F. and Farnsworth, J. F., "Gas from the Koppers-Totzek process for steam and power generation," Paper presented at the Ind. Fuel Conf., Purdue Univ., West Lafayette, Indiana, Oct. 3, 1971 (typescript copy).

12. Franzen, J. E. and Goeke, E. K., "SNG production based on Koppers-Totzek coal gasification," In: Proc. of the 6th

13. Staege, H., "The production of hydrogen and CO + H_2-containing gas from coal by the Koppers-Totzek process," Gaswärme Int. 25, 287-292 (1976).

14. Carinci, G. G. and Meisner, D. C., "Applications of coal gasification and the Midrex process of direct reduction," In: Usos des Carbon en Siderurgia: Abastecimiento y Tecnologias, Congr. del Inst. Lat. Amer. del Fierro y el Acero, Mexico, July 25-30, 1976, pp. 305-312.

15. DBP 872 952, 1949.

16. DBP 972 895, 1949.