PERIODICA POLYTECHNICA SER. CHEM. ENG. VOL. 39, NO. 2, PP. 87–99 (1995)

DEVELOPMENT OF COAL GASIFICATION TECHNOLOGIES

Gábor SZÉCHY and Imre SZEBÉNYI

Department of Chemical Technology
Technical University of Budapest
H-1521 Budapest, Hungary

Received: November 1, 1995

Abstract

The history of development of coal gasification technologies is reviewed. The basic features of the 'first generation' technologies (Lurgi, Winkler and Koppers-Totzek), and later developments based on these are discussed. The role of coal gasification in 'clean' coal based electric power generation is discussed, as a later development to fulfil environmental requirements. Special, experimental gasification processes (gasification with nuclear heat, molten bath processes and underground gasification) are also mentioned.

Keywords: coal gasification, gasifier types, 'clean' power generation, environmental aspects.

Introduction

Coal gasification is a process where coal is converted into a combustible gas in a reaction with oxygen and steam (or air and steam). The gas produced can be combusted for power generation or heating, or can be used as feedstock for chemical syntheses. The most important components of the gas are carbon monoxide, hydrogen and methane.

The most important reactions of coal gasification are strongly endothermic, thus their equilibrium is shifted towards the products (CO and H_2) only around and above 800 °C. These reactions are accompanied also by volume increase, thus application of pressure is disadvantageous for the formation of the products.

Equilibria of methane formation reactions show a different pattern, since they are exothermic and are accompanied by volume decrease, thus methane formation is enhanced by relatively low temperature and high pressure. However, because of the quite low kinetic rate of methane formation these reactions are less significant in the determination of the primary product distribution [1].

The heat requirement of the exothermic reactions is covered in all commercial processes by the in situ combustion of coal . These types of processes are called autothermic.

There are, however, several processes in experimental or pilot plant stage, which cover the heat requirements from an external source, e.g. from the heat of a special nuclear reactor or from the exothermic heat of decomposition of certain compounds. These types of processes are called 'allothermic'. It is important with these processes that heat must be available at a temperature level of at least 800 °C, since even the most reactive coals cannot be reasonably gasified below that temperature. In principle, no oxygen is necessary in the allothermic processes, the total amount of the coal can be utilized in gasification reactions.

Industrial scale gasification of coal has a history of 100-120 years. 'Classical' gas generators (gasifiers) were run at atmospheric pressure with air and steam. Lump coal was fed from the top of the generator and contacted the air/steam mixture in countercurrent pattern. Such gasifiers were used in Hungary in quite large numbers until the end of the sixties . Hydrogen for ammonia synthesis was also produced on this basis. Around 1960, 250 such gasifiers were in use in Hungary, with a total coal gasification capacity of 1.5 million tons/year.

At present, no such gasifiers are used in Hungary, (and probably nor elsewhere in the world), this is clearly a technology of the past.

Presently, there are around 35 processes commercially available or at an advanced pilot plant stage. From these, around 10 processes were also tested on a commercial scale. All these processes belong to the autothermic type.

The development of these autothermic processes can be traced back to one of the three so-called 'first generation' processes: the fixed-bed Lurgi, the fluidized-bed Winkler and the entrained-bed Koppers-Totzek process.

First Generation Processes (Lurgi, Winkler, Koppers-Totzek)

These processes were developed in the 1920s, 30s and 40s, and they can still be found (in somewhat modified versions) in certain industrial plants of the world [1]. However, their true significance is that they provide the basis for further developments. The more recently developed processes utilize the experience gathered with these.

The basic features of the first generation processes are shown in *Table 1*: [1]. *Fig. 1* shows the generalized principles of gasification and names of processes derived from the first generation technologies.

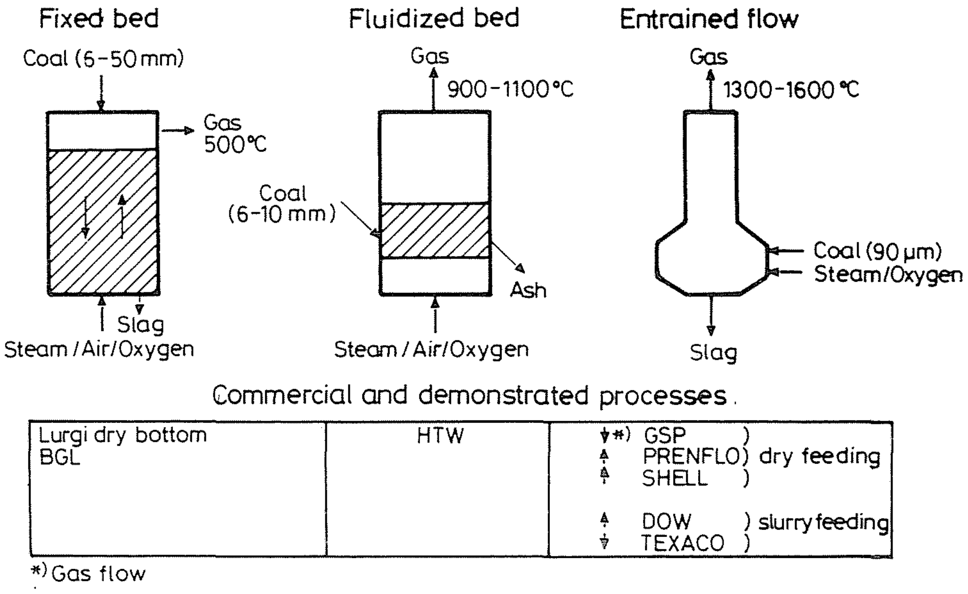

Fig. 1. Gasifier principles and commercial processes

Table 1
Characteristics of the three 'first generation' processes

Characteristics	Process		
	Lurgi	Winkler	Koppers-Totzek
Reactor (gasifier) type	Fixed bed	Fluidized	Entrained bed
Coal particle size	10–30 mm	1–10 mm	<0.1 mm
Steam/oxygen ratio, kg/STP m^3 O$_2$	9:1 - 5:1	2.5:1 - 1:1	0.5:1 - 0.02:1
Coal/gas contact pattern	countercurrent	cross-current	concurrent
Residence time of coal	60–90 min.	15–60 min	< 1 sec
Gas exit temperature	370–600 °C	800–950 °C	1400–1600 °C
Pressure, bar	20–30	1.03	1.03
Raw gas composition, vol.%			
CO+H$_2$	62	84	85
CH$_4$	12	2	0.1
Requirements towards feed coal	Should not bake, or fall apart	Must be very reactive	Ash melting temp. must be < 1450 °C
By-products	Tar, aqueous condensates	None	None

Their advantages and disadvantages are as follows:

Lurgi

advantages:
- high thermal efficiency (due to the countercurrent contact pattern)
- relatively high methane content in the raw gas
- low dust content in the gas

disadvantages:
- certain requirements towards the feed coal
- high steam consumption
- tar and aqueous condensates are also obtained

Winkler

advantages:
- small grain, high ash coals can be directly used
- no tar by-products

disadvantages:
- coal conversion is not complete
- high dust carry-out
- temperature-barrier (temperature must be definitely below the melting point of the ash, otherwise fusion of the ash particles is started, and the fluidized bed collapses.)

Koppers-Totzek

advantages:
- no tar by-products are formed
- small steam consumption
- small amounts of waste-water are formed
- disadvantages:
- large oxygen consumption
- high particle content in the raw gas, partly in form of partially melted 'sticky' ash particles.

According to a review published in 1978 [2] the worldwide number of functioning Lurgi gasifiers was 60; that of Winkler gasifiers 36; and that of Koppers-Totzek gasifiers 50. The number of Lurgi gasifiers was greatly increased further when the SASOL-II and III industrial complexes were started up in South Africa in 1980 and 1982, each having 36–36 Lurgi gasifiers. Lurgi generators were used in the early 1980s also in the then East Germany and Czechoslovakia. Koppers-Totzek gasifiers were used in Turkey, Greece and India and they were mainly applied to produce synthesis gas [1]. Generally it can be stated that synthesis gas production on coal basis is more competitive than production of heating gases, because in the latter case the coal derived heating gas must compete with natural gas itself.

This situation is true from economic point of view, but as a consequence of new environmental limitations introduced at the beginning of the 1980s concerning SO_2 and NO_x emissions from power generation, it was this sector which gave new impetus to the development and application of coal gasification. The developments and applications of the last 10 years are mainly connected to power generation.

Comparison of a Conventional Coal Fired- and an IGCC Power Plant

Coal gasification is competing not only with natural gas, but also with coal firing itself. *Tables 2* and *3* are presented [3,4] to show that coal gasification integrated into a combined cycle power station (IGCC) is the cleanest among the industrially applied coal based power generation technologies. It can be seen that IGCC technology provides the highest SO_2 retention and in respect of NO_x and dust emissions, it is at least as good as the other coal based technologies.

Table 2
Emissions from coal-based power generation technologies* [4]

Emissions, tons/year; for one MW electricity producing capacity			
	SO_2	NO_x	Solid wastes**
Powdered coal firing + flue-gas desulphurization (PCF+FGD)	13	7	680
Atmospheric fluidized bed combustion (AFBC)	6	4	1090
Integrated gasification combined cycle (IGCC)	4	3	270

*Based on Illionis coal, 3.5% sulphur
**Dry

The advantages are also shown in *Fig. 2* where IGCC is compared with a traditional coal-powder fired power station equipped with flue-gas desulphurization. It can be seen that in addition to smaller SO_2 and NO_x emissions, a further advantage is that readily saleable elemental sulphur is produced instead of gypsum.

These advantages are following from the fact that the raw gases of coal gasification can be better purified and they represent much smaller volumes than the flue gases of coal combustion. The sulphur content of coals is converted mainly to H_2S in the gasification process and this can

Table 3
Environmental impact of coal-based power generation technologies [3]

Technology	CO_2 emission kg/kWh	SO_2 retention %	NO_x conc,* mg/m^3	Particulates* mg/m^3
Powdered coal firing + flue gas desulphurization	0.87	90	500–650	50**
Circulated fluidized bed combustion (CFBC)	0.86	90	100–300	≈ 30***
Pressurized fluidized bed combustion (PFBC)	0.82	90	100–300	≈ 10****
Integrated gasification combined cycle (IGCC)	0.78	99	120–300	negligible emission

*NO_x concentration in the flue gas, at 6 vol.% 0_2 content of the flue gas
**Predicted performance
***Filter bag house
****Ceramic filters

be removed almost qualitatively from the gas, and subsequent can be converted into elemental sulphur by the Claus process successfully practised in the petroleum industry for a long time.

Recent Coal Gasification Processes Realized on the Industrial Scale

The main development trends are summarized in *Fig. 3*, which does not include all development projects. As a general tendency, the increase of gasification pressure and temperature can be observed.

Increased gasification pressure is justified not only by an increased gasifier output, but also by the fact that it is advantageous if the gas product is available at 20–40 bar, no matter whether it is used for combustion or as synthesis gas. Let us review now the development of the three 'first generation' processes.

It can be seen from *Fig. 3* that the fixed-bed Lurgi process was developed in two directions. From 1979 to 1983, the 'Ruhr 100' project was realized in West-Germany, where a pilot plant was built to carry out fixed-bed gasification at pressures up to 100 bar. Thus, by increasing the pressure of gasification from 25 to 95 bar, the methane content of the raw gas has increased from 9 to 17 vol.%, and the thermal efficiency of the process increased from 80 to 85%, while the amount of converted coal (that is, unit throughput) also roughly doubled [5].

Another development of the Lurgi process is the British Gas–Lurgi slagging gasifier technology. A demonstration plant has been working in

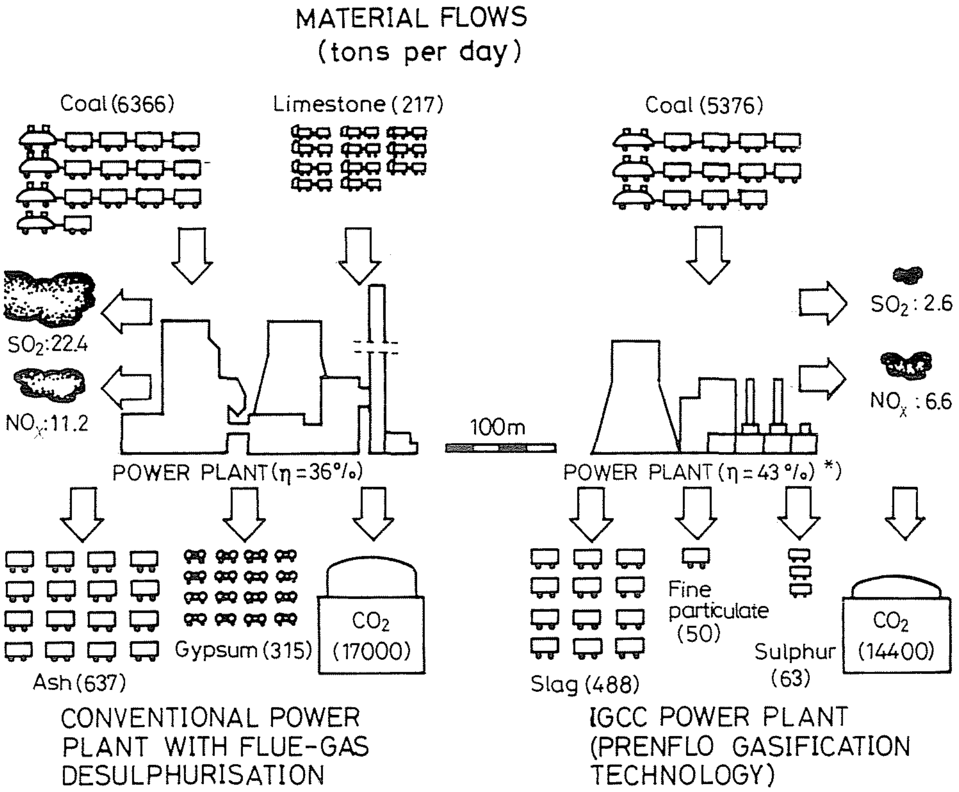

Fig. 2. Comparison of a conventional coal fired- and an IGCC power plant
Basis: Power generation capacity: 700 MWe
Feed coal (dry basis):
Lower heating value: 26.5 MJ/kg
Ash: 10%
Sulphur: 1.2%
Carbon: 73%

Scotland for more than 20 years, based on that technology. The temperature at the bottom of the gasifier is approximately 2000 °C, thus slag is removed in molten state. Steam consumption can be greatly reduced since a considerable portion of steam in the original Lurgi technology was used as a cooling agent. A further advantage is that the material obtained by cooling the molten slag immobilizes heavy metals and other pollutants in its matrix, thus its disposal is less problematic than that of the original Lurgi ash [1,3].

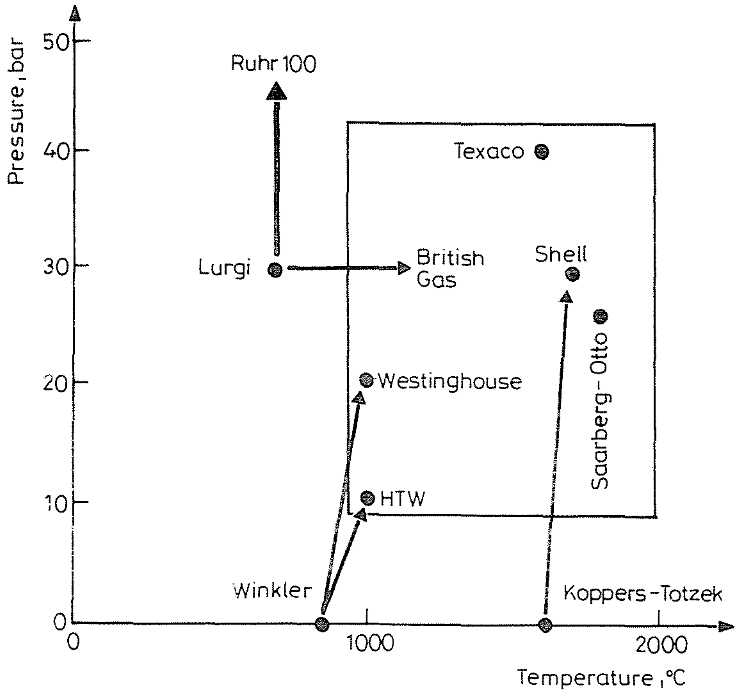

Fig. 3. Main development trends of gasification processes

The fluidized-bed Winkler process was further developed in an experimental plant in West Germany between 1979 and 1984. Gasification of brown coal was carried out at 10 bar, and gasification temperatures were also elevated up to 1100°C with the help of additives increasing the melting point of the ash (HTW process) [5]. Since that time, several commercial plants have been built, based on the HTW process (see *Table 5*). Among them is the IGCC power-plant project 'Kobra', near Cologne, where gasification is carried out already at 25 bar.

As it can be seen from *Fig. 1*, most of the later developments are based on the entrained-bed gasifier, that is on the Koppers-Totzek type of process.

Gasification temperatures are very high (1300–1500 °C) with these types of processes, thus no tar or aqueous condensates are obtained, and quality characteristics of the feed coal are of minor importance. Feeding of the pulverised coal can be accomplished in dry form, or in form of an aqueous slurry, in a cocurrent pattern. Among these processes, the greatest

Table 4

Performance of different coal gasification processes into combined cycle power plants [3]

	Gasification process integrated into the power plant				
Description	British Gas-Lurgi (BGL)	DOW	PRENFLO	SHELL	TEXACO
Temp. of gas from reactor* (°C)	500	1430	1500	1450	1300-1500
Gasification efficiency (%)	92.2	74.3	80.6	80.9	75.2
Gross capacity					
– P steam turbine (MWe)	83.0	129.6	120.7	112.6	130.7
– P gas turbine (MWe)	144.5	144.5	144.5	144.5	144.5
– P gas expander (MWe)	–	–	–	–	5.3
– P total (MWe)	227.5	271.3	265.2	257.1	280.4
Energy consumption (MWe)	12.7	26.7	27.5	22.1	29.3
Net capacity (MWe)	214.8	244.6	237.7	234.9	251.1
Net efficiency** (%)	41.0	38.5	40.9	40.2	39.5
Degree of desulphurization (%)	95	99.5	99.5	95	99.5

*Based on licensors' statements

**Calculated at high heat value (HHV); for a more advanced gas turbine available, net efficiency would increase by about 0.7%.

amount of experience has been accumulated with the Texaco process, which was originally developed for the gasification of heavy crude oil residues.

A few data of four processes using entrained bed gasifiers in IGCC power generation are shown in *Table 4*, where the performance of the fixed bed British Gas–Lurgi (BGL) process is also shown as a comparison [3].

It can be seen that the BGL process has the highest gasification efficiency, while entrained-bed processes offer a higher SO_2 retention.

In *Table 5*, an attempt was made to summarize the basic features of the commercial scale coal gasification plants started up in 1980 or later. As it was mentioned earlier, it can also be observed here that while production of synthesis gas was the more common purpose of coal gasification in the early and mid 80s, power plant applications started to be more characteristic in the second half of the 80s.

A recent IGCC power plant was built in the Netherlands, in Buggenum. Gasification is carried out here by the Shell–process. The calorific value of the feed coal is equal to 585 MW at full load, while the produced gas represents a calorific value of 460 MW, – that is, the thermal efficiency of the gasification is 78.6%. The lower heating value of the raw gas is 11 MJ/kg, and it consists of 65 vol.% CO and 30 vol.% H_2. The adi-

Table 5

Coal gasification plants in operation or under construction

Project or company name	Location	Gasification capacity tons of coal/day	Gasification process	Remarks (year of startup, goal of gasification)
SASOL II and III	Secunda, Republic of South Africa	2×40000	Lurgi	1980,1982 synthesis gas
Dakota Gasif.	Beulah, USA		Lurgi	1985, heating gas
Tennesse Eastman	Kingsport, USA	820	Texaco	1983, acetic anhydride
Coolwater	Barstow, USA	900	Texaco	1984, 1989, IGCC 120 MW electricity
Ube Ind.	Japan	1600	Texaco	$H_2 \rightarrow NH_3$
Ruhrkohle	Oberhausen, Germany	800	Texaco	synthesis-gas for oxo-synthesis
Plaquemine	Plaquemine USA	2400	DOW	1987, 160 MW IGCC
Rheinbraun	Berrenrath, Germany	730	HTW	synthesis-gas for MEOH 1988
Kemira	Oulu Finland	960	HTW	synthesis-gas for NH_3 (from peat) 1988
RWE-Kobra	Köln Germany	2880	HTW	320 MW IGCC 1994-95
SEP-Holland	Buggenum, Netherlands	2000	Shell	250 MW IGCC, 1994
Thermie	Puertollano Spain	2600	Prenflo	320 MW IGCC, 1996
NEX	Nynäshamn, Sweden	5300	Texaco	2×365 MW IGCC, 1994

abatic flame temperature of this gas is very high (≈ 2400 °C), thus NO_x formation is also high. To reduce the flame temperature (and NO_x formation), the gas is diluted with nitrogen and saturated with steam thus its lower heating value is decreased to 4.3 MJ/kg (4.4 MJ/m^3) and its CO content to 25 vol.%, H_2 content to 12 vol.% [6].

Investment Costs

Coal gasification integrated into a combined cycle power plant is an expensive technology. According to an International Energy Agency report published in 1993 [7], the investment cost of a 260 MW power plant including a 'typical' entrained-bed gasifier with 'wet' feeding (an aqueous coal suspension is fed to the gasifier) is 1913 USD/kW, which is 25% higher than the investment costs of a coal-based conventional power station of the same output (equipped, of course, with flue-gas cleaning). Investment costs of the IGCC plants are much more sensitive to the plant size (nominal capacity) than those of the conventional coal-powder fired power stations. Thus, if the output of the power plant will be reduced to 150 MW, the investment costs per kW will increase by 19.3% [7].

It should be noted that the choice of the particular gasification process to be integrated into the power plant is important, but not decisive from the point of view of investment cost, since a considerable portion of the investment consists of plant sections like coal preparation, oxygen plant, Claus plant which are similar or even the same for all the different, presently practised processes. This can be seen also in *Table 6*, where the distribution of the investment costs is shown for an IGCC and a conventional coal-powder fired power plant of the same size.

Table 6
Comparison of the distribution of investment costs for a conventional and an IGCC power plant

Conventional power plant		IGCC power plant	
Power plant equipment	75%	Combined cycle power plant equipment	45%
Flue gas desulphurization	13%	Coal preparation	8%
Flue gas NO_x reduction	6%	Oxygen plant	14%
Electrostatic particle emission reduction	6%	Gasifiers	9%
		Boiler for heat utilization	11%
		Gas desulphurization	7%
		Wet particle filters	6%
Total	100%	Total	100%

Table 6 shows that 55% of the investment of the IGCC plant is falling on gas production and purification, and only 45% is the share of the costs of the actual power generation section. Gasifiers themselves make only 9% of the total investment.

A Few Experimental Technologies or Technology Groups

At the end of this review, three gasification routes should be mentioned, which cannot be classified as commercially available, but are quite remarkable because of their completely different nature.

The first of these routes is gasification with heat from a nuclear power station. This is an allothermic process where heat must be available at a minimum temperature level of 750–800 °C. Thus, conventional pressurized-water nuclear power plants are not suitable for this purpose, but a special high-temperature nuclear power plant must be constructed which provides heat at 900–950 °C. Such a power plant was run in Jülich, West Germany, and the applicability of the concept was experimentally demonstrated. A great advantage of this concept is that no oxygen is necessary for the gasification, consequently the costs of an oxygen plant can be omitted [1].

Another different concept provides the basis for a group of processes, where the heat for the gasification is supplied by molten iron or molten salts. Molten iron is applied in the Humboldt, Klöckner and the Sumitumo processes where coal gasification is carried out similarly to (or even in combination with) steel making. In the Kellogg process, a sodium carbonate melt is used as heat source. Here, gasification can be carried out at 930–1030 °C (instead of the 1400–1450 °C of the molten iron processes), because of the catalytic effect of the sodium carbonate melt. A plant for the demonstration of the Humboldt process was in operation in West Germany from 1985 to 1987, where sulphur content of the raw gas could be reduced to 10-20 ppm by addition of limestone and appropriate slag withdrawal [1,5].

The third concept, or group of processes, is the underground (in situ) gasification of coal. This is not a new concept, since its appeal was recognised 90–100 years ago. Its successful realization, however, requires the solution of many problems, and because of environmental risks (ground water pollution, formation of depressions and underground deformations), the commercial application of these kinds of processes is not very likely in densely populated areas [1].

Underground gasification of coal requires more mining and geological knowledge and skills than chemical experience. The main problem here is to increase the gas permeability of the coal seams. This can be achieved by the formation of channels in the seam, by means of boreholes, fracturing, controlled combustion, or by combination of these methods [1].

In the then Soviet-Union, several projects and at least three commercial power plants were based on the underground gasification of coal. West Germany and Belgium carried out a joint experimental project lasting several years to study the gasification of a deep-lying (800–900 m) coal seam. In the United States of America between 1973 and 1983 thirteen experimental underground gasifications were carried out in Wyoming. The experimental gasifications usually lasted for 30–50 days and produced gases with lower heating values of 4–6 $\mathrm{MJ/m^3}$ [1].

Conclusion

In conclusion, it can be stated that for the gasification of coal several modern technologies are available, which were thoroughly studied and some are also commercially proven. These technologies offer a possibility to meet the present environmental regulations in power generation and to utilize coal in 'clean technologies'.

These technologies, however, are not really competitive at the presently prevailing coal/petroleum and coal/natural gas price ratios, and require large investments.

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