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Coal Gasification for Hydrogen Production Using Nuclear Energy

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Abstract

The most abundant fossil fuel on Earth is coal. Up-to-now coal is mainly used for electricity production and will continue to play this important role in the near and mid-term. However, coal could also substitute oil and gas by coal gasification or coal liquefaction techniques. The necessary process heat could be supplied by nuclear energy to save fossil energy resources and to lower carbon emissions to the environment. In Germany, comprehensive R&D activities were conducted in the past to investigate the utilization of nuclear energy in the heat-intensive processes of coal gasification for the large-scale production of hydrogen, synthetic natural gas and synthetic liquid fuels, respectively. Different methods of gasification for the two types of coal existing in Germany have been developed, i.e. steam gasification of hard coal and hydrogasification of lignite. Both processes were successfully demonstrated on pilot plant scale under simulated nuclear conditions.

1 Coal – More Than Just a Fuel to be Burnt

Despite of its abundant resources on earth, the conversion of coal to gaseous or liquid fuels has been commercially applied, only in a limited scale. In the 1970s, with the first oil crisis, coal resources were assumed to become a more central role and coal conversion programs were started as a contribution to an away-from-oil policy. Extensive experimental and theoretical studies included coal gasification, coal liquefaction, and advanced combustion systems aiming at improved methods for the generation of Synthetic Natural Gas (SNG), liquid hydrocarbons, and other raw materials for the chemical industries. Numerous coal gasification projects were launched to investigate various processes, on pilot plant scale [1, 2]. However, interest in coal refinement faded away again with low oil prices, since 1980s.

2 Coal Conversion Processes supported by Nuclear Energy

The conversion of coal into a gas is realized by means of a gasification agent, which reacts with the coal at temperatures > 800 °C. The gasification agent is either steam (steam coal gasification) or hydrogen (hydrogasification). Both processes have in common that high pressures are needed to achieve a high methane yield, whereas for an optimal synthesis gas production, high temperatures and low pressures are required. The gas-cooled nuclear High-

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Temperature Gas-Cooled Reactor (HTGR) is capable to provide the process heat at the required temperature level up to the range of 900-950 °C.

2.1 Steam coal gasification

Steam gasification of coal is a mature and well established technology practiced on industrial scale. In the conventional steam coal gasification processes, a part of the coal is partially oxidized to provide the necessary process heat. In the heterogeneous water gas reaction, the residual organic solids are converted to synthesis gas with 'steam' as the agent. The gasification reaction with is given by the heterogeneous water gas reaction:

$$C + H_2O \rightarrow H_2 + CO \qquad -118.5 \text{ kJ/mol C}$$

The homogeneous water gas (shift) reaction allows a further increase of the H_2 fraction. It is followed by a methanation step if the desired end product is SNG.

Gasification processes are classified according to the type of reactor. The principal lines mainly used today are those by Lurgi (since 1931), Winkler (since 1922), Koppers-Totzek (since 1941). They all were developed in Germany and exist at a large commercial scale.

1. In the Lurgi pressure gasification, a solid bed of coal moving from top to bottom is gasified by adding steam and oxygen from the bottom at a pressure of 1.5-3 MPa. Coal is inserted in smaller pieces. The counter-current flow arrangement leads to higher conversion rates and thermal efficiencies. The amount of methane generated is depending on pressure, temperature, the oxygen-to-steam ratio, reactivity of the coal, and the contents of volatile substances in the coal. The Sasol company in South Africa is the world's largest commercial applier of coal conversion technology operating gasification reactors which are able to produce 65,000 Nm^3/h of dry gas with a raw coal throughput of 54 t/h [2].
2. The High Temperature Winkler (HTW) process takes place in a fluidized bed where fine-grain coal is reacted with oxygen and steam. The fluidized bed has no reaction zones, but rather forms a homogeneous distribution of solids. The temperature must be below the ash melting point to prevent a softening and agglomeration of the ash, which would lead to a collapse of the fluidized bed. Characteristics are the simple coal pre-treatment, low oxygen consumption and good performance over a broad load range. Industrial scale is at ~60,000 Nm^3/h.
3. The Koppers-Totzek process runs in a flue stream gasifier at very high temperatures above the ash melting point. Dry coal dust is mixed with steam and oxygen/air and gasified at atmospheric pressure, in an autothermal way. The reaction zone is limited to the flame area with a co-current flow of coal and gasification agent. It has the advantage that tar formation is suppressed and other organic substances are destroyed. The conversion rate is at almost 100 %. Industrial plant capacities are in the order of 50,000 Nm^3/h.

Numerous modified process variants have been developed aiming at an adjustment of the feedstock quality, an optimization of the product gas composition and, of course, an efficiency improvement. Variants differ by temperature and pressure range, grain size of the coal, and residence time. Coal conversion is estimated to be around 95 % and the total efficiency (based on higher heating value) to be ~ 70 %. Main issues of coal gasification are the handling of solid material streams and the large amounts of CO_2, SO_2, and ash requiring a complex cleaning system [1]. Emissions of carbon dioxide can be suppressed, if a CO_2-emission-free, external heat source is applied (allothermal gasification). For example, with an HTGR, the heat provided by the hot helium coolant can be introduced indirectly into the gas generator, another part being used for the steam production, and the rest still usable for electricity production.

2.2 Hydrogasification

In the hydrogasification process of coal, hydrogen is added to convert the coal into a methane-rich raw gas, in an exothermic reaction. This is ideal for the production of substitute natural gas (SNG). The hydrogen can be provided either by taking the coke left from the hydrogasification and convert it with oxygen and steam in an HTW process, or by taking a part of the produced methane for steam reforming. Both processes need high temperatures, which could be provided by nuclear energy, in an allothermal process. The gasification reaction with the agent 'hydrogen' and the main product methane is:

$$C + 2\,H_2 \rightarrow CH_4 \qquad +87.5\ \text{kJ/mol C}$$

Kinetics of the process are more complex compared to steam gasification. The advantage of hydrogasification compared with steam coal gasification is its 200 K lower pre-heating temperature which reduces potential corrosive attack. A major drawback, however, is the low conversion rate, i.e., the large amount of residual coke of up to 40 %. In contrast to steam gasification, the hydrogasification process still needs to be demonstrated at a larger commercial scale.

3 Nuclear Process Heat for Coal Gasification

3.1 Drivers for nuclear coal refinement

There are several drivers for nuclear energy to be introduced as a primary heat source into the coal gasification process [3]:

- In the conventional gasification process, a significant additional amount of feedstock is necessary to provide process heat at the required temperature level. Substitution of the process heat by nuclear energy would allow resource savings of up to 40 %.
  A respective reduction in CO_2 and other, coal-specific emissions will be achieved, at the same time.
- The conversion to liquid hydrocarbons will reduce dependency on oil imports and diversify energy supply.
- If cost of nuclear heat is sufficiently low, it may help to stabilise fluctuating energy cost.

3.2 The Prototype Nuclear Process Heat (PNP) Project

In Germany, the concept of the pebble-bed HTGR has been developed and became subject of various projects. Motivation for the PNP project was to take advantage of the large resources of both energy carriers coal and uranium and to create an additional use on the process heat market, for the coal. HTGR are characterized, among other features, by their operation at an average coolant exit temperature of up to 950 °C [4], due to the fully ceramic core and fuel elements. The maximum operational temperature of HTGR is mainly determined by the capabilities of metallic materials for heat exchangers. As most chemical processes are performed at lower pressures some adaptation of the reactor design and of the chemical process has been necessary, because HTGR normally operate at pressures of 4-9 MPa. Heat transfer under varying operational load conditions, hot gas mixing in the core bottom, or the lifetime of hot gas thermal insulation have been comprehensively investigated in extensive experiments.

The project started with an integrated arrangement of the core and the heat exchangers in a Prestressed-Concrete Reactor Pressure Vessel (PCRV), with a maximum power of 3000 MWth per unit. This concept was afterwards changed towards modular HTGR with a power size of 170-200 MWth per unit to allow for a passive decay heat removal in case of an accident. A particular safety significance has connection between the vessels for the nuclear heat source and for the steam generator or heat exchanger. This connection has to be designed as pressure vessel according to the leak-before-break principle, where the early detection of a leakage would allow the immediate plant shutdown. An underground placement of the nuclear unit might be recommended to ensure an enhanced protection against fire, aircraft crash or atmospheric explosions of gas clouds in the adjacent coal conversion facilities.

3.3 Coupling between nuclear and chemical plant

For the nuclear steam coal gasification process, it was foreseen that the heat from the reactor coolant be transferred to an additional intermediate circuit via a helium-helium intermediate heat exchanger (He-He IHX). Two different He-He IHX components were constructed by German companies, one with a helical tube bundle and the other one with U-tubes, designed for a power level (~125 MW) representative for large and medium-sized plants. Both components were tested with 950 °C helium on the primary side [5], at a 10 MW scale. For the PNP nuclear hydrogasification of coal, it was foreseen to place the steam-methane reformer directly into the primary circuit of the HTGR. The nuclear steam reformer component was tested as part of the EVA-ADAM system at FZJ, an energy transportation system based on nuclear steam reforming and methanation processes.

3.4 Nuclear steam gasification of coal

A new component for nuclear steam coal gasification was the gas generator with allothermal heating. Tests were conducted with a gas generator on semi-technical scale with a fluidized bed of about 1 m^2 base area and a height of up to 4 m, laid out for a coal throughput of ~200 kg/h. Different from the conventional concept, coal was gasified indirectly by means of a tube-type immersion heat exchanger which was placed into the fluidized bed to transfer heat from a separate helium circuit to simulate nuclear conditions [6].

The semi-technical plant was used for testing components, feeding devices, insulation, investigating broad ranges of operating conditions, and applying different types of coal. Since the temperature provided by the helium is limited, catalytic coal gasification was also investigated in the semi-technical plant and enhanced reaction rates were observed. The catalyst, however, was found to be not effective until a certain threshold value, also corrosion effects were enhanced. The semi-technical plant was in hot operation for approx. 26,600 hours with more than 13,600 hours under gasification conditions (750-850 °C, 2-4 MPa). Maximum capacity was 0.5 t/h of coal, the total quantity of gasified coal was 2400 t [6, 7].

The commercial-size gas generator was foreseen to have a thermal power of 340 MW. It was designed, unlike the semi-technical plant, as a horizontal pressure vessel (Figure) to contain a fluidized bed with the shape of a long-stretched channel to allow for long residence times. It consisted of four parts (modules). Coal is introduced through several inlets in the first module; the gasification zone spreads over the other three modules. In the fourth module, the remaining ash is cooled and removed. Each module contains steam inlets in the bottom section and an immersion heat exchanger bundle, through which heat is transferred from the hot helium to the fluidized bed.

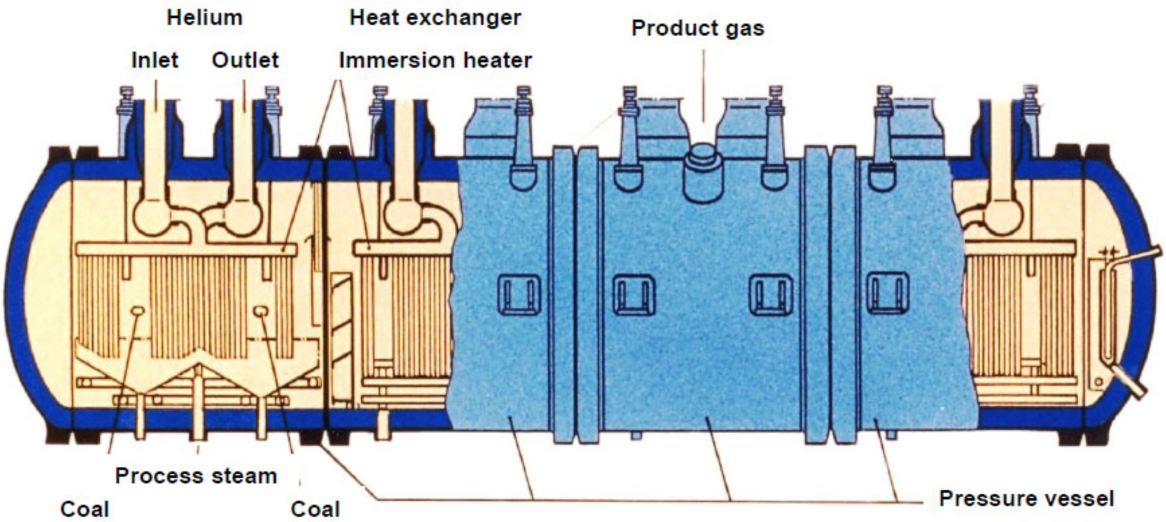

Figure: Schematic of an industrial-scale gas generator for nuclear steam coal gasification.

3.5 Nuclear hydrogasification of coal

The hydrogasification process can also be combined with a nuclear heat source. But unlike the steam coal gasification, the nuclear heat is not coupled directly into the gasification reactor [8]. One variant is steam-methane reforming, where a part of the product gas methane is drawn off and the high temperature nuclear heat is used for the endothermic reforming reaction. In a second variant, the nuclear heat is taken to pre-heat the hydrogen produced during the gasification process itself (water gas shift reaction) to 800-950 °C. Nuclear heat is also used for steam production and the residual power for electricity generation. Compared to the first variant, it has a simpler process scheme. A drawback is the fact that high gasification pressures (8 MPa) are needed.

Between 1976 and 1982, the Rheinische Braunkohlenwerke, Wesseling, investigated the hydrogasification process in a 1.5 MW semi-technical test facility with both lignite and hard coal [8, 9, 10]. The reactor of 8 m height contained a fluidized bed with 0.2 m diameter where hydrogen was injected as gasification agent. The hydrogen was electrically pre-heated to 750 °C and could, if necessary, be further heated to 1000 °C by partial combustion. Operational parameters could be varied in a broader range. A part of the hydrogen was used as a carrier medium for the coal input.

The test facility was operated for about 27,000 h with more than 12,000 h under gasification conditions. The throughput was 320 kg/h of lignite or 160 kg/h of hard coal, the total quantity gasified was 1800 t.

From 1983 to 1985, a follow-up pilot plant was operated over 8300 h, with half of the time under coal gasification conditions with high availability. It included, unlike the semi-technical plant, all post-processing components of gas treatment up to the stage of SNG production. The plant had a throughput of 9.6 t/h corresponding to a total power of 50 MW. Gasification of more than 17,000 t of brown coal was made to yield a total of 11 million Nm^3 of SNG, whose fraction in the raw gas was between 22 and 36 %. The SNG production was at a rate of up to 6400 Nm^3/h.

4 Summary and Conclusions

Within the frame of the PNP project, comprehensive R&D activities on nuclear coal gasification were conducted in Germany in a cooperation between the Research Center Jülich and partners from the coal and nuclear industries. The results can be summarized as follows:

- The use of nuclear primary energy promises a saving of ~40 % of the coal resources, increase the specific SNG output, and reduce specific emissions of carbon and other pollutants to the atmosphere, respectively. As a transition step, process steam could be supplied from a nuclear heat source for use in conventional steam coal gasification.
- Special importance for process heat projects had the 45 MWt AVR test reactor in Jülich, which was operated between 1967 and 1988. It became the world's first pebble-bed reactor to successfully achieving a coolant outlet temperature of 950 °C proving the feasibility of the pebble-bed HTGR concept under high temperature process heat conditions with a high availability. (The same helium outlet temperatures are currently demonstrated for the Japanese block reactor HTTR as well.)
- Nuclear coal gasification is considered to be a complementary option in the energy supply strategy and may help to significantly reduce the dependency on oil and natural gas imports.

- A wide variety of coal gasification processes has been developed and demonstrated in the past, which are differently appropriate depending on user requirements, plant size, and coal characteristics.
- Allothermal steam coal gasification with both main types of coal in Germany, hard coal and lignite, was demonstrated in numerous experiments. The new major components that needed to be developed were the gas generator and the steam methane reformer.
- The operation of the semi-technical plants confirmed the technical feasibility of allothermal, continuous coal gasification under the nuclear conditions of a process heat HTGR.
- By the manufacture and successful operation of high temperature heat-exchanging and heat-transporting components on the 10 MW power level under simulated nuclear conditions in KVK and other related test facilities, valuable practical experience with high-temperature helium plants on a representative scale was acquired. Both IHX types were considered appropriate to be designed for a power of 170 MW and an operation time of 140,000 h at 950 °C.
- A key problem remained the selection of appropriate high temperature materials for the heat exchanging components such as steam reformer and He-He intermediate heat exchanger, but also for the gas generator in the case of catalytic coal gasification.

The idea of constructing a PNP prototype plant was abandoned at the beginning of the 1990s, when economic analyses had shown that competitiveness of nuclear SNG from expensive German coal with the cheap oil and gas available on the world markets was not given. Therefore, further funding and developments for both lines, steam gasification and hydrogasification were terminated.

Future activities could take benefit from a re-evaluation of the studies conducted in the past on HTGR process heat applications by comparing against current technologies and market conditions. The goal should be to select promising applications under the current industrial practice within existing and evolving markets. Superior safety features and high reliability are considered to be prerequisites for the introduction of nuclear process heat and nuclear combined heat and power generation.

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