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Processing of coal mine gas with low methane concentrations for use in high temperature fuel cells

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1. Introduction

Coal mines are emitting off-gases containing methane. The methane concentrations are ranging from 20 Vol.-% for coal seam methane (CSM) from active mines to 80Vol.-% for coal mine methane (CMM) from closed-down mines. As such it can be considered as an energy rich resource and is collected by suction systems to be fed into pipelines for domestic and industrial consumption as well as to decentralised power generation units as gas engines or high temperature fuel cells. Within Germany an additional benefit is that methane emitted from coal mines counts as a renewable energy and hence falls under the renewable energy legislation with the associated economical benefits. Another important reason for drawing off the emitted methane is its ecological impact when vented to the atmosphere: methane is 20 times more harmful a greenhouse gas compared to carbon dioxide. However, methane contents below 35 Vol.-% cannot be used in gas engines and fuel cells. An additional aspect is safety: depending on methane and oxygen concentrations, the coal mine gas might form an explosive mixture. In these cases suction and/or compression of the gas is prohibited by safety regulations and the gas is vented to the atmosphere for operating mines whilst the recovery is simply being stopped for closed down mines. One possibility to increase the methane content and hence prevent venting of the gas is to apply a gas permeation process using methane selective membranes. STEAG Saar Energie, an operator of coal mine gas pipeline networks and power plants in the German Federal State Saarland, the engineering consultancy OTS and the GKSS Research Centre Geesthacht GmbH formed a consortium to investigate this technology. In a second stage of the project, the E&C company Borsig Membrane Technology is also involved. The project is funded by the German Ministry of Economics and Technology.

2. Process description and process design

Feeding the gas into a pipeline at a pressure of 9 bar is conducted in a two stage process. In a first stage gas is drawn from the coal mines by blowers where the pressure is increased from 700 mbar to 1.3 bar. Subsequently compressors provide the pipeline pressure. As long as the oxygen content of the gas is below 6 Vol.-%, the system can be operated as described for all methane concentrations. For oxygen contents in excess of 6 Vol.-% the suction and compression stages can be operated for methane contents above 25 Vol.-% only. The operation of blowers only is permitted for methane contents between 22 Vol.-% and 25 Vol.-%. In this case the drawn off gas is vented to the atmosphere for operating mines whilst the suction from closed down mines is suspended. If the methane content falls below 22 Vol.-%, no drawing off of gas at all is allowed.

Hence there are two possible options for integrating a membrane unit into the process, i.e. using the gas downstream of the blowers or downstream of the compressors as the feed gas. It was decided to design a pilot process possessing the flexibility to be employed for both pressure levels. Furthermore, two membrane modules can be installed so that two-stage operation is possible. Figure 1 shows a simplified flowsheet of the pilot plant.

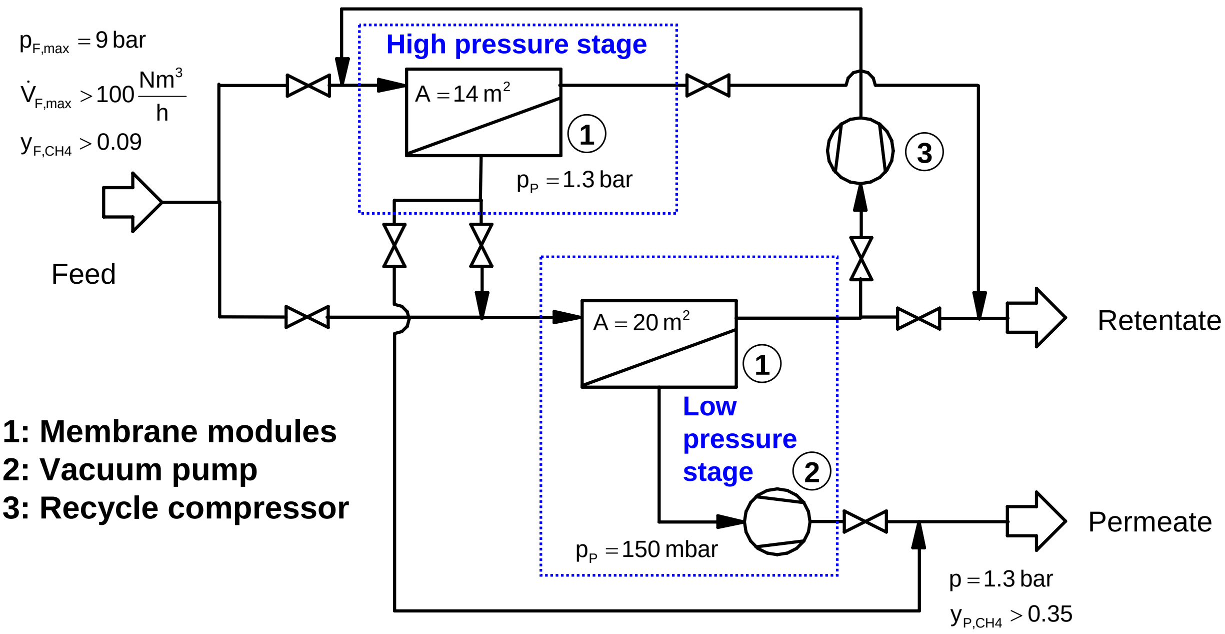

Figure 1: Simplified flowsheet of two stage gas permeation process

The feed gas can be directed to either of the two membrane modules by means of valves. The driving force for the high pressure stage is generated by the compressors. The permeate is at a pressure of 1.3 bar and either forms the methane enriched product gas or, in case the required methane content cannot be achieved by one-stage operation, is fed to a second low pressure stage. For this stage a vacuum pump operating at a pressure of 150 mbar supplies the driving force. A recycle compressor can be employed to feed the retentate of the low pressure stage back to the feed side of the high pressure stage. If only operation of the blowers is allowed, the low pressure stage on its own can be employed to upgrade the coal mine gas.

The membranes employed are silicone based high flux membranes. The methane/nitrogen selectivity of this material is limited, but still allows for an increase in methane concentration to the required level of 35 Vol.-% in the permeate, provided the methane content in the feed is high enough. For the process design, application of GKSS envelope type membrane modules [1] was assumed. However, in later stages of the project the use of spiral wound membrane modules is also planned.

In order to predict the operating behaviour of the unit, it was modelled using the equation oriented process simulator Aspen Custom Modeler®. The model employed for simulating the membrane modules accounted for real gas behaviour and concentration dependent permeation as described in reference [2]. Figure 2 shows the simulated performance of the low pressure stage. The operating conditions are given in the figure. It is apparent that a methane content exceeding 35 Vol.-% in the permeate can only be achieved if the methane concentration in the feed is higher than 23 Vol.-%, when carbon dioxide is present in the feed gas. In case no carbon dioxide is present only 21 Vol.-% of methane are required in the feed. The carbon dioxide content of the feed gas influences the performance of the gas permeation unit since the permeance of carbon dioxide is considerably higher than that of methane for silicone based membrane materials. Furthermore does carbon dioxide induce swelling of the membrane and hence influences the permeation rates of the other components present. The carbon dioxide content has also an impact on the recovery. With no carbon dioxide present, methane, nitrogen and oxygen are permeating independently. If carbon dioxide is present in the feed the predicted results are different: the membrane is plasticized and additional permeation pathways are being formed. These allow increased amounts of methane pass through the membrane and thus increase recovery, albeit on the expense of a reduced permeate purity.

For a two stage process as indicated in Figure 1 operated with a feed pressure of 9 bar and a feed flowrate of 200 Nm^3/h, a feed methane concentration of 16.5 Vol.-% is required to achieve 35 Vol.-% of methane in the product at maximum carbon dioxide concentration. Furthermore is the methane recovery positively affected. This performance increase is however at the expense of additional investment and operating costs due to the more complex plant layout and the energy consumption of the recycle compressor.

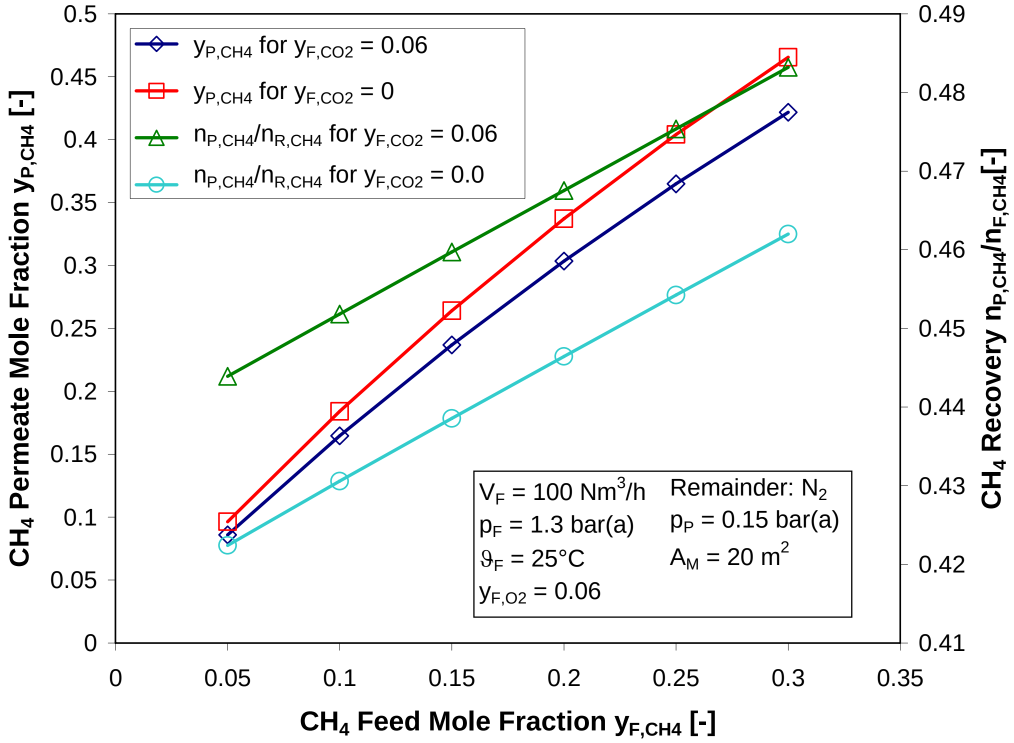

Figure 2: Simulated performance of low pressure stage

The pilot plant is currently in the commissioning phase at a STEAG site. First experimental results obtained from the low pressure stage indicate that the methane content can be enhanced at a reasonable recovery. The high pressure stage has been delivered to the site and will be tied into the process. Figure 3 shows a photograph of this stage.

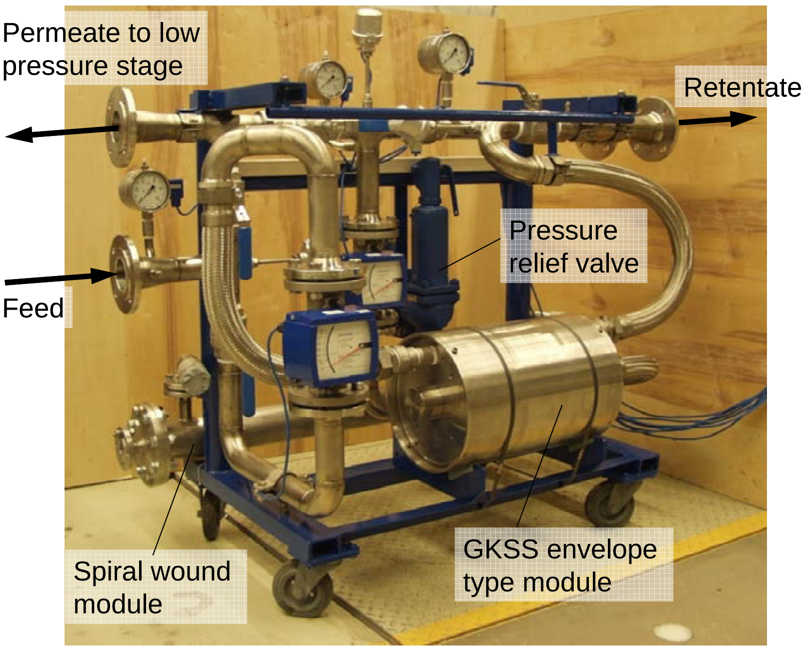

Figure 3: High pressure stage

3. Conclusions and future work

The theoretical studies conducted so far indicate that gas permeation processes can be employed to increase the methane content of coal mine gas so that it can be employed as a feed stock for decentralised power generation units. However, various process parameters as well as overall performance have to be evaluated by means of pilot plant operation. Aspects to be investigated include:

- use of different membrane module types, i.e. envelope type and spiral wound,
- validation of simulation tools by pilot plant data,
- influence of the carbon dioxide content on the performance,
- control of the unit with respect to safety-relevant changes in methane and oxygen concentrations in the feed and the resulting influence on the quality of the product (permeate) gas,
- long term stability of the membrane process with respect to “real world” operation, i.e. assessment of the influence of changing compositions, possible condensation and entrainment of dust or compressor oil on the operating performance and
- economical evaluation of the process.

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