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Investigation of the Long-Term Performance of an Industrial-Scale Biogas Upgrading Plant with Grid Supply Applying Gas Permeation Membranes

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Within this work, the design and first experiences in long-term stability of an industrial scale biogas upgrading plant are presented. This plant supplies 100 m³ (STP)/h of biomethane to the natural gas grid and is based on the gas permeation membrane process. The investigation of the long-term performance stability is supported by means of detailed numerical process simulation. It can be shown, that the application of an adequate gas pretreatment leads to a prevention of membrane deterioration and to a significant enhancement of the overall plant durability.

1. Introduction

Within this work, the design and the operational experiences of the first industrial scale biogas upgrading plant in Austria are presented. This fully automated unit is capable of processing approximately 180m³(STP)/h of biogas and supplies 100 m³ (STP)/h of biomethane to the natural gas grid. The upgrading is based on the gas permeation membrane process and the plant has been erected in Bruck/Leitha in Lower Austria.

In the field of biogas upgrading, reliability of the process is an important feature. It influences directly the process economics. The biogas upgrading technology requires high reliability and a plant availability of 360 days a year and 24 hours a day. In this work we investigate the issue of the long term performance of the named gas permeation biogas upgrading plant. The investigation is supported by means of a detailed numerical modeling. The deterioration of the membrane selectivity and permeance together with the effects on the plant performance parameters like energy requirement and methane recovery are discussed. The methods for the membrane protection are reviewed shortly. Finally, experimental data on the long-term performance of the biogas upgrading plant in Bruck/Leitha is presented.

2. Gas Composition and Upgrading Necessities

Table 1 compares the properties of biogas and natural gas defined by the Austrian directives OEVGW G31 and OEVGW G33. It can be seen that natural gas differs from typical biogas in heating value, Wobbe Index and in its composition. Therefore,

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multiple steps are required to upgrade biogas to natural gas quality. The main process steps that have to be performed to reach the required product gas quality given in legislature are shown in Figure 1. Due to its heterogeneous origin, biogas usually contains a high number of trace components which can significantly influence the performance and long-term stability of the applied upgrading technique. Depending on the raw material of biogas production these trace components usually comprise alkanes and cycloalkanes, halogenated hydrocarbons, aromatic compounds, siloxanes, alkyl sulphides, volatile fatty acids and alcohols (Rasi et al., 2007).

Table 1 Characteristic of typical biogas and natural gas defined by the Austrian directives OEVGW G31 and G33

	Biogas	Natural Gas	Unit
Methane	45-70	-	mol %
Carbon dioxide	30-45	≤2	mol %
Ammonia	≤1 000	0	mg/m³(STP)
Hydrogen sulphide	≤2 000	≤5	mg/ m³(STP)
Oxygen	≤2	≤0.5	mol %
Nitrogen	≤8	≤5	mol %
Water (dew point)	37 at 1bar	≤-8 at 40bar	°C
Upper heating value	6.7-8.4	10.7-12.8	kWh/m³(STP)
Wobbe index	6.9-9.5	13.3-15.7	kWh/m³(STP)

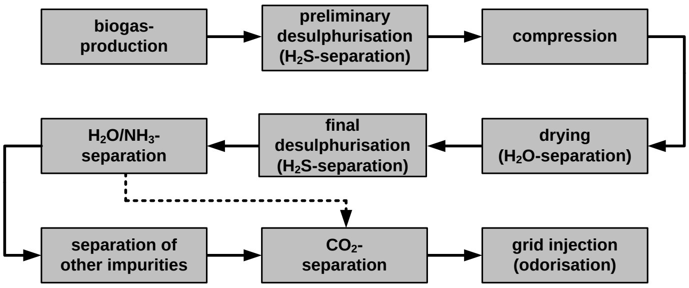

Figure 1: Main steps of the biogas upgrading process necessary to reach natural gas quality

3. Process Design Applying Gas Permeation for Biogas Upgrading

The separation of gases in membrane gas permeation occurs on the account of the solution-diffusion mechanism. Driven by the partial pressure difference between feed side and permeate side of the membrane the gas molecules first solve in the membrane, diffuse through it and finally resolve into gas domain on the other side of the membrane. Both transport phenomena solution and diffusion contribute to the membrane permeation rate of the gases. The solution rate of the gases in the membrane is mainly influenced by the condensability of the gas component, which is typically expressed in terms of critical temperature. The diffusivity rate is influenced at most by the size of the gas molecule. In glassy polymers the faster permeation of carbon dioxide in biogas upgrading allows to purify the raw biogas leaving the high-methane gas stream on the high pressure retentate side.

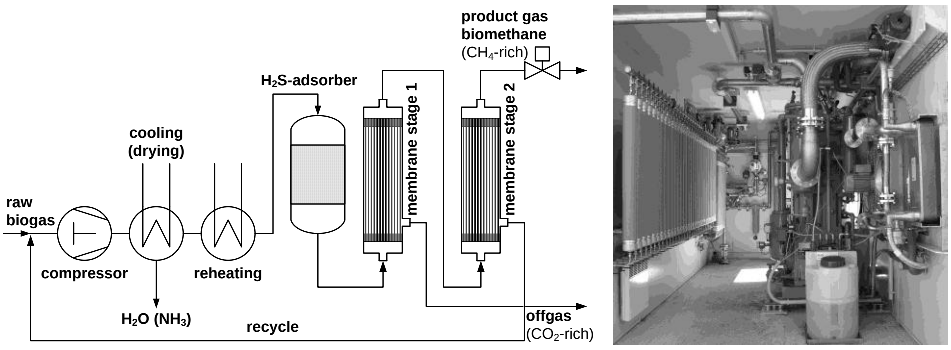

Figure 2: Flowsheet and photo of the interior of the biogas upgrading plant in Bruck/Leitha

A great variety of membrane cascades are available for the process design of biogas upgrading. The two stage cascade with recycle of the second permeate and one compressor was suggested in several works to be the most favorable configuration in biogas upgrading (Schell and Houston, 1983; Rautenbach and Welsch, 1993; Makaruk et al., 2010). Thus, this layout has been chosen to be implemented in the biogas upgrading plant Bruck/Leitha which supplies approximately 100 m^3 (STP)/h of biomethane to the natural gas grid since its commissioning in 2007. The flowsheet of this plant as well as a photo of the interior are given in Figure 2.

As reported by several studies some of the trace compounds can have adverse effects on the membrane performance in the gas permeation (White et al., 1995; Al-Juaied and Koros, 2006). The permeation of heavy hydrocarbons can promote temporary and permanent polymer changes like plasticization, swelling and compaction of the membrane material. Therefore the reduction of the concentration of these components is a crucial step in biogas upgrading. Adequate gas pretreatment must be applied if high process performance needs to be preserved for longer operational times. In Bruck/Leitha this separation is mainly done by cooling to low gas temperatures (freeze drying), filtering and adsorption.

4. Process Simulation of Biogas Upgrading

In order to investigate the influence of the lowering of membrane performance on the process behavior extensive simulation work has been performed. Simulations have been done using the recently developed algorithm for modeling of membrane gas permeation systems (Makaruk and Harasek, 2009). The method uses the Gauß-Seidel method to solve the discretized one-dimensional conservation equations for each of the gas components. The method uses a typical solution-diffusion model to calculate the transmembrane flow. Within this model the pressure loss, concentration polarization and species diffusion are neglected. The modeling approach had been validated experimentally and good agreement of the modeling results with experimental findings was determined during the validation. More information on the modeling approach can be found in Makaruk and Harasek (2009).

Figure 3 presents the results for the variation of the selectivity in a biogas upgrading process using a two-stage membrane gas permeation plant with recycle. The selectivity reduction is simulated through the increase of the methane permeance while the permeance of the carbon dioxide is kept constant. In the left chart it can be noticed that the selectivity deterioration results in a reduction of methane recovery and in a slight increase of the specific isentropic compression power. This takes place if only the plant operating pressure is adjusted to the changing selectivity in order to preserve constant methane content in the produced gas (dashed lines). If both upgrading parameters, methane content and methane recovery, are to be invariable to the selectivity reduction, the compressor gas volume flow needs to be increased in addition to the pressure adjustment (see right chart in Figure 3). The required increase of compressor revolutions results in a considerable increase of the specific isentropic compression power (continuous lines in left chart of Figure 3). As a consequence the operating costs of the biogas upgrading increase as well.

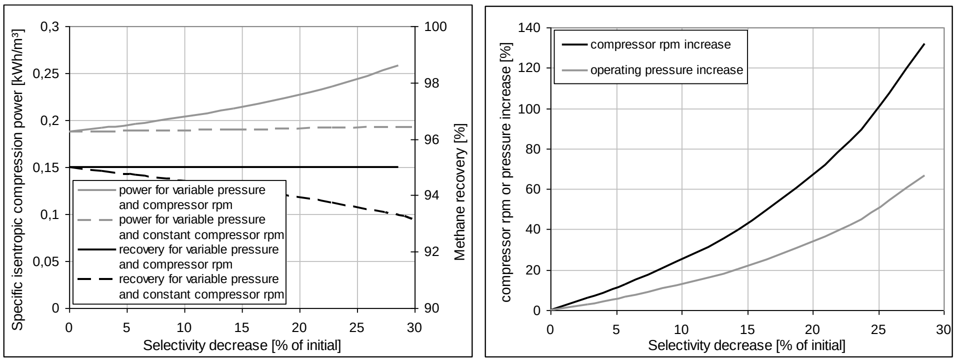

Figure 3: Simulation results for the variation of selectivity during biogas upgrading

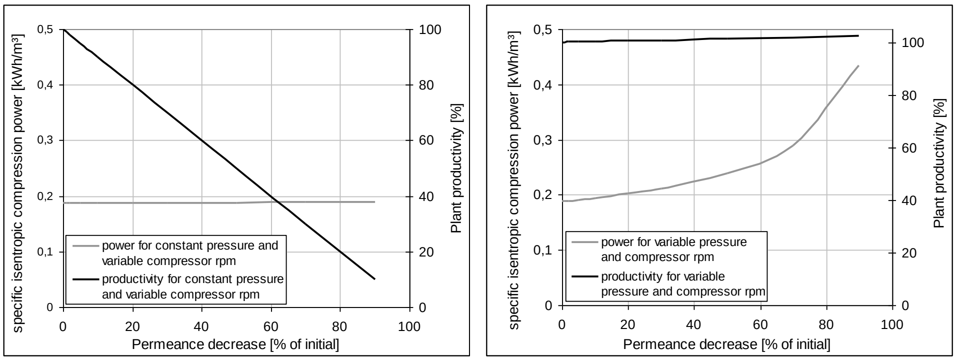

Figure 4: Simulation results for the variation of permeance during biogas upgrading

Figure 4 shows the dependence of the plant productivity on the reduction of the membrane gas permeance for constant operating pressure. It can be seen that in order to keep the high methane content of the produced gas the compressor volume flow needs to be reduced which results in a decrease of the plant productivity (see left chart). In this case the productivity of the plant depends linearly on the membrane permeance. If a constant productivity is to be maintained, the operating pressure needs to be increased to compensate for the decreasing permeance. In this case the compressor volume flow can remain constant. The right chart in Figure 4 presents the dependence of the plant productivity and specific compression power on the permeance decrease. As expected, the specific compression power increases owing to the operating pressure increase. The operating pressure increase leads also to a small increase of the plant productivity.

5. Operational Experiences and Performance Evaluation

As discussed in the previous section the reduction of membrane parameters like membrane selectivity and membrane permeance lead to a deterioration of the plant performance. This results mainly in the decrease of the energy efficiency of the upgrading process. In addition to the polymer changes caused by the presence of heavy hydrocarbons, the membrane performance can be affected by dust that is present in the gas and by condensing water. Therefore, gas pretreatment before the gas is contacted with the membrane is a crucial point in biogas upgrading.

A large number of processes are available for the removal of volatile components from gas streams (Khan and Ghoshal, 2000) which can be adapted to biogas upgrading. These processes include processes like thermal and catalytic oxidation, condensation, bio-filtration and absorption. Although these processes provide high removal efficiencies they result in relatively high investment and operating costs. On the other hand, adsorption on activated carbon provides an alternative that allows for an uncomplicated and inexpensive gas pretreatment process removing a large variety of trace organic components from gas streams (Shin et al., 2001).

The gas pretreatment of the membrane biogas upgrading plant in Bruck/Leitha consists of several steps like gas filtration, gas drying and adsorption on activated carbon. The steps are introduced into the process in order to prolong the lifetime of the membrane.

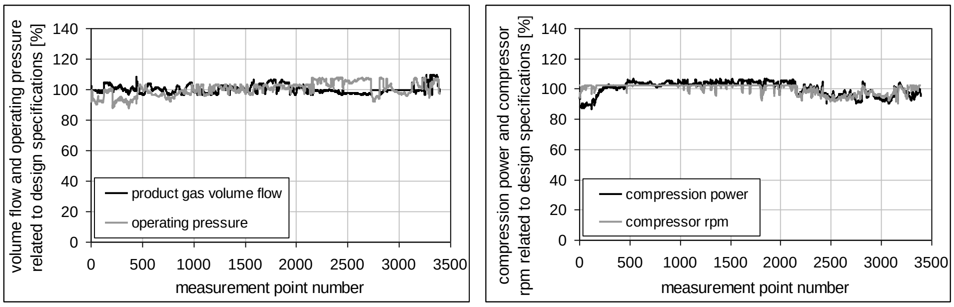

Figure 5: Long-term operational data related to design specifications of the biogas upgrading plant Bruck/Leitha ranging from January 2008 to December 2009

Figure 5 presents the most important performance parameters of the plant within a time frame of 24 months. It can be noted that the performance of the plant is not reduced within this monitoring period, which can be attributed to a suitable gas pretreatment.

6. Summary

Membrane gas permeation is a viable process for biogas upgrading provided that an adequate gas pretreatment is applied. This has also been reported by Miltner et al. (2009), Rautenbach and Welsch (1993) and Schell and Houston (1983). The 24-month operation of a biogas upgrading plant equipped with gas pretreatment proved that the application of gas pretreatment results in reasonable membrane lifetimes and higher plant durability.

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