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Biogas upgrading for transportation purposes – Operational Experiences with Austria's first Bio-CNG fuelling station

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Within this work, the design, build-up and operational experiences of Austria's first Bio-CNG fuelling station in Margarethen/Moos are presented. The biogas upgrading technology based on membrane separation and its performance is described, modeling and simulation tools for the plant design are presented and the integration of the upgrading plant into the existing biogas plant is outlined. It can be shown, that the application of Gas Permeation together with a local Bio-CNG fuelling station leads to an ecologically reasonable and economically feasible solution to the forthcoming needs of the transportation sector.

1. Introduction

Biogas is considered to be a renewable and sustainable energy source. It is produced in a large number of biogas plants all over Europe from a manifold of substrates like energy crops, organic wastes or agrarian residues. Besides the minor contents of malicious components like ammonia or hydrogen sulphide, this gas mainly contains methane (45 to 65 vol%) and carbon dioxide. The common technology to use this energy source nowadays is the combustion in gas-engines and generating electric power with an efficiency of 35 to 40%. Because of the rising prices of energy and resources the usage of the thereby appearing waste heat is of vital importance for an economically and ecologically efficient operation of the biogas plant.
Biogas upgrading can be an alternative solution to the production of electrical energy. Processed biogas could be used as a natural gas substitute in households or industry or as a fuel for transportation purposes (CNG engines). Depending on the local circumstances, the most suitable product gas quality and upgrading technology has to be chosen. Either, an upgrading to natural gas quality together with the supply to a nearby natural gas grid, or the upgrading to BIO-CNG quality together with a local CNG fuelling station is conceivable. Reduction of CO_2 emissions and the strong dependencies

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from foreign energy supplies is possible in both cases combined with the advantages of an increased local added value.
After extensive lab-scale examinations and after the commissioning of the industrial scale biogas upgrading plant Bruck/Leitha in Lower Austria feeding approx. 100m^3(STP)/h of bio-methane to the public natural gas grid, which has been recently reported by Makaruk et al. (2008) and Miltner et al. (2008), Austria's first Bio-CNG fuelling station has been erected in Margarethen/Moos (Lower Austria, 20km south-eastwards from Vienna), also applying the same membrane separation technique.

2. Gas Composition And Upgrading Necessities

In order to produce fuel for the Austrian market, the product has to fulfill the Austrian law "Kraftstoffverordnung" BGBL 417/04, while in Germany and many other European countries DIN 51624 is applicable. It is planned to merchandize upgraded biogas on several locations in Austria and abroad; therefore a new fuel brand called "methaPUR" has been established, unifying the numerous quality parameters in one standard. The definition of the methaPUR standard is compared to the composition of gaseous fuels (CNG – compressed natural gas) prescribed in the aforementioned laws and to the raw biogas in Table 1. It has to be mentioned, that the hydrogen sulphide content of the raw biogas is extremely low due to effective in-situ-desulphurization using commercially available liquid mixtures of metal salts and due to the favorable local substrate mixture (high content of energy crops together with liquid pig manure).
It can be easily seen that in order to upgrade biogas to accepted fuel qualities, several steps must be performed. The most important of them are the separation of malicious substances, drying and separation of carbon dioxide, nitrogen and oxygen, which results in the increase of the calorific value and the Wobbe Index.

Table 1: Composition and properties of raw biogas produced in Margarethen/Moos compared to methaPUR and CNG- quality defined by Austrian Law and DIN

Component	Raw Biogas	methaPUR standard	Austrian Law BGBL 417/04	DIN 51624	Unit
Methane	49 ±10	>95	NR	>80	mol%
Carbon dioxide	51 ±10	<5	NR	Sum <15	mol%
Nitrogen	<0,3	<1,0	NR		mol%
Oxygen	<0,1	<1,0	NR	<3,0	mol%
Hydrogen sulphide	5 (<20)	<10	NR	<5*	ppmv
Water	saturated	"dried"	NR	<40	mg/kg
Particulate matter	techn. free	techn. free	techn. free	-	mg/kg
Relative density	1,05*	0,60*	0,55-0,7	0,56-0,70*	kWh/m^3
Upper Calorific value	5,40*	>10,45	8,4-13,1	NR	kWh/m^3
Wobbe Index	5,27*	>13,6	12,8-15,7	NR	kWh/m^3

*… calculated
NR… not regulated

3. Process Design, Process Simulation And Plant Realization

Within this work, a novel membrane separation technique named Gas Permeation has been applied for the main biogas upgrading steps. The main advantages of the process are a stable and continuous operation and excellent control behavior. Furthermore, no expensive regeneration or chemicals are needed. The whole process is very simple, straight-forward and compact. The separation technique uses a dense polyimide-membrane with different solubilities and diffusivities for the various gas species in the raw biogas feed. Using this membrane material, most unwanted gas species are quantitatively removed from the feed stream and transported through the membrane to the permeate stream. Nitrogen shows similar behavior as methane and therefore cannot be removed by this technique and remains in the product gas stream, the so-called retentate. The membranes are in form of hollow fibers with the high pressure feed/retentate stream on the inner side of the tube and the low pressure (almost atmospheric) permeate on the outside of the tube. Many of these fibres are collected to form a membrane module that is fed with pressurized biogas. A simplified scheme showing the principles of the Gas Permeation separation technique together with a microscopic view of one membrane hollow fiber is presented in Figure 1.

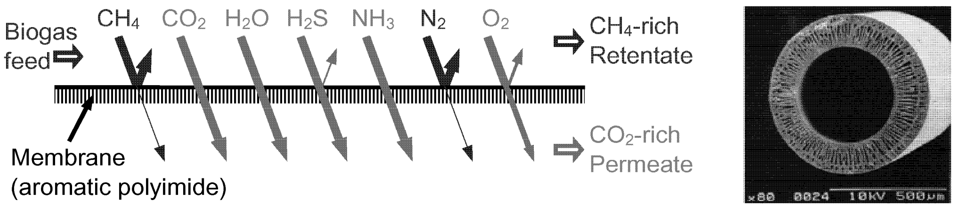

Figure 1: Principle of gas separation using Gas Permeation (left) and SEM-view (scanning electron microscope) of the hollow fiber membrane (right)

Since the mixture of NH_3, H_2S and humid gas can jeopardize the membrane material, some gas preprocessing before the gas permeation is necessary. These pretreatment steps include drying of the gas by a two-step cooling (cooling water and refrigerant) and a final desulphurization by means of adsorption on iron oxide. These steps are operated at a pressure of about 9bar(g) downstream the biogas compressor. After the cooling step the biogas is reheated with the compressor waste heat to provide an optimum temperature for the subsequent adsorption and membrane separation process steps. The methane content of the produced gas is controlled via the retentate gas pressure by a proportional valve that is located at the retentate outlet of the membrane modules. Using this control strategy a gas with various methane contents can be produced. Additionally, the volume flow of the produced bio-methane can easily be adjusted with an enhanced PID controller manipulating the rotating speed of the compressor using a frequency converter. Like any other separation technique, Gas Permeation cannot transfer all of the methane in the raw biogas feed to the produced bio-methane. As a result, the carbon dioxide-rich permeate (so called “offgas”) still contains little amounts of methane and other separated substances. In order to achieve a zero-emission strategy regarding methane the upgrading plant is perfectly integrated into the existing biogas plant and the Offgas is delivered back to the existing gas engine (CHP with raw biogas). Thus, the remaining methane is not emitted to the atmosphere, but is burned and its chemical energy is used to produce heat and power. The quality of the produced bio-methane stream (methaPUR) is permanently monitored; only if the quality constraints are met, the gas is delivered to the high pressure compressor and subsequently to the high pressure storage of the Bio-CNG fuelling station. A flowsheet of the upgrading plant and the fuelling station is given in Figure 2.

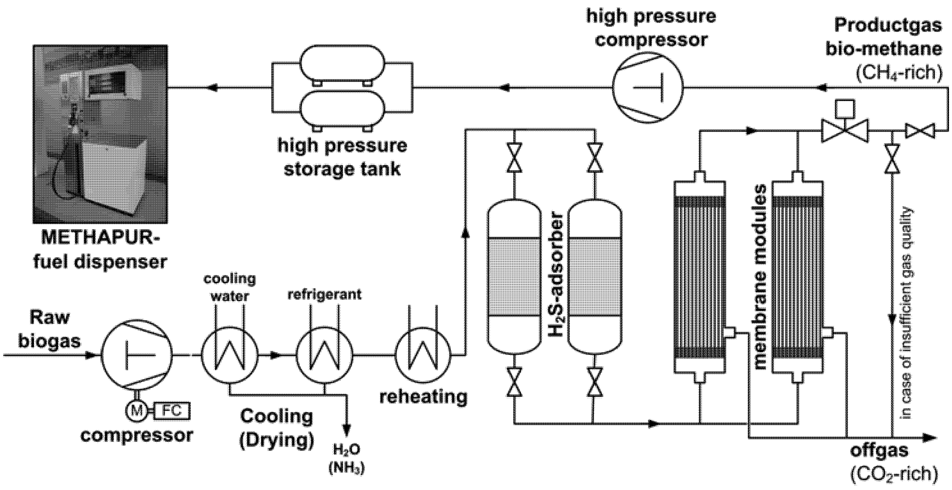

Figure 2: Process concept scheme for biogas upgrading plant and Bio-CNG fuelling station

The tool for the dynamic simulation of gas permeation systems that was developed previously (Makaruk et al., 2008) was used to aid the design of the biogas upgrading plant. The tool is specialized to model the dynamics of gas permeation systems, but it can be applied for the static modelling as well.

A series of simulations was run to find an optimal process configuration. Fixed boundary conditions: feed volume flow of 80 m^3(STP)/h and methane content in the feed gas of 50% and in the product gas of 95 vol% were applied for each simulation run. The membrane area was varied resulting in different feed pressures that were required to reach the Bio-CNG quality specification. The results of the modelling are presented in Figure 3. It can be seen that for low membrane area the required feed pressure is high but decreases significantly when increasing the membrane area. As a result, the operational costs are reduced as well. However, certain trade-offs exist, since the addition of membrane area leads to higher investment costs and to some reduction of the methane recovery and the Bio-CNG volume flow. The optimal process configuration for the biogas upgrading is identified as shown in Figure 3. It results in acceptable operational and investment costs as well as in relatively high methane recovery and product gas volume flow. If should be noticed, that the methane recovery can be significantly enhanced by the application of a two-stage permeator configuration.

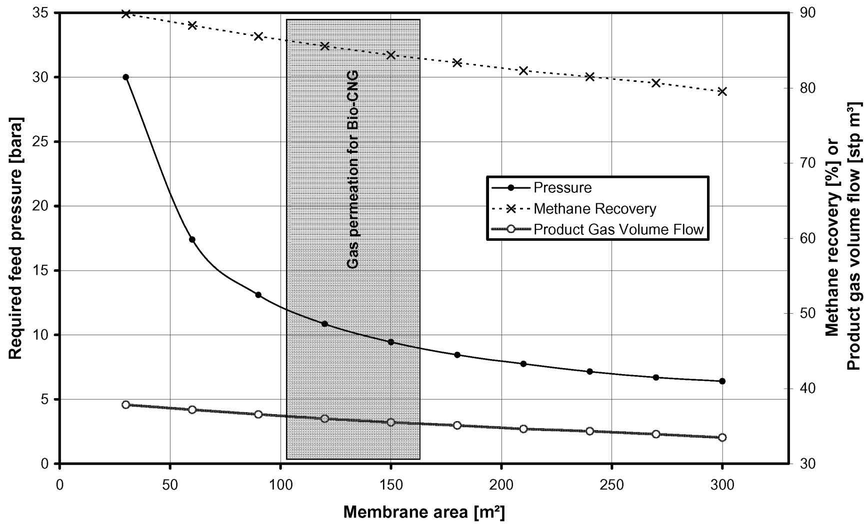

Figure 3: Modelling results for the variation of membrane area for design purposes

The biogas upgrading plant and the fuelling station have been built at the existing biogas plant Margarethen/Moos in Lower Austria; the opening has been celebrated at the end of August 2008. The realized biogas upgrading plant has a capacity of 25kg/h (corresponding to 33m³STP/h) bio-methane (methaPUR) and is operated in parallel to the existing 500kW CHP-gas engine. The projected annual amount of 150.000kg gas supplies up to 200 CNG cars (annual road performance of 15.000km each) with a 100% renewable fuel. The upgrading plant and the fuelling station and its components are designed to be operated completely without any personnel (except maintenance).

4. Operational Experiences

The monitored operation of the upgrading plant over a period of about half a year showed, that the desired product gas quality had been guaranteed by the plant's control system at any time and even under varying raw biogas compositions (methane 40 to 52vol%, hydrogen sulphide 1 to 19 ppmv). This result has been attested by the plant's online-gas analyzers as well as by an independent laboratory (NUA, www.nua.co.at).
Figure 4 shows some operational parameters during a typical biogas upgrading and high pressure storage tank filling procedure. Whenever the pressure level in the high pressure storage falls under a defined threshold a START request is sent to the upgrading plant, which is always kept in standby-mode. After this request the upgrading plant's compressor and the fully automated biogas upgrading start. During the startup, the product gas quality usually does not comply with the specifications and thus the produced gas is delivered back to the CHP-gas engine (feedback operation). After about 5 to 10 minutes all quality specifications are met and the produced gas is fed to the high pressure compressor and fills up the high pressure storage tank (feeding operation). As shown in Figure 4 the upgrading plant operates about 3 hours until a STOP-request is generated. This request is triggered by exceeding the upper pressure threshold in the

storage tank (depending on storage volume and contemporaneous fuelling operation of the bio-CNG fuelling station). Thus, the upgrading plant’s compressor is stopped and is set back into standby-mode. During the whole upgrading operation the combined quality/quantity-control systems assure adequate product gas quality according to the specifications and the achievement of the preset product gas volume flow.

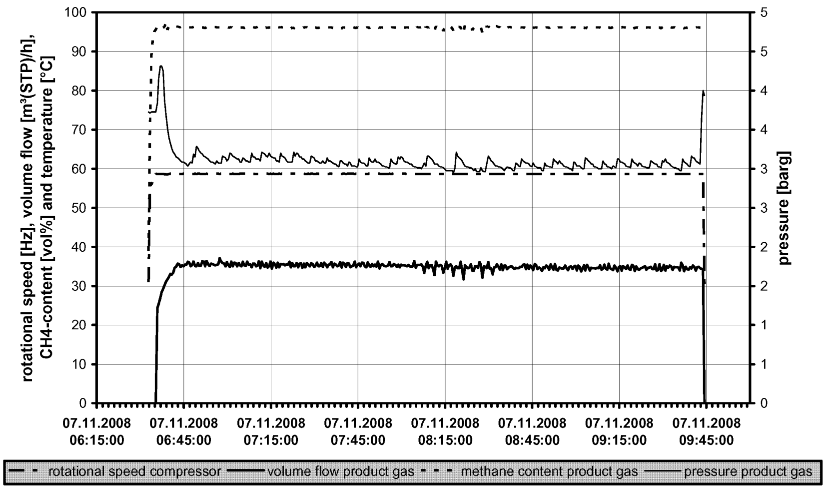

Figure 4: Some important operational parameters during biogas upgrading and filling of the high pressure storage tank

The observed excellent suitability of the membrane separation based biogas upgrading technique for fast start/stop operation is quite unique compared to other upgrading technologies (e.g. PSA, TSA, water scrubbing, amine scrubbing). As a result, this technology is favourable for applications demanding this feature like the considered small, decentralized fuelling station with strongly fluctuating demand over time. Additionally, the overall energetic efficiency of the upgrading plant is outstanding. Related to the energy content of the produced gas (Lower calorific value) the total demand of electrical power for the upgrading plant (not including the high pressure compressor) has been determined to be 3,0% throughout the monitored operational period, which is a very low value compared to other upgrading technologies.

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