

Efficient Biomass Utilization by Polygeneration Processes- Production of Hydrogen, Electricity and Heat

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ABSTRACT

A polygeneration process is about to be implemented at the biomass gasification plant in Oberwart, Austria. Apart from conventional heat and electricity production, product gas obtained from steam gasification of wood chips is used for production of hydrogen. A membrane separation process was chosen for this application. Meeting the requirements of robustness and simplicity are benefits of this technology, however, maximizing of purity and output of hydrogen is not given highest priority. Simulation results show the gas compositions of both permeate and retentate stream as a function of different membrane stage-cuts. Basically high hydrogen content in the permeate stream can be achieved, but only with the drawback of low stage-cuts. Moreover, the trade-off between hydrogen purity and hydrogen recovery as well as the influence of the operating pressure on the purity are illustrated.

Keywords: polygeneration, biomass, gasification, membrane, hydrogen

1 INTRODUCTION

Polygeneration represents one appropriate concept towards an efficient utilization of resources for energy generation and fuels production. With regard to the limited availability of renewable resources, among them especially biomass resources, efficiency in terms of consumption and utilization becomes vital.

Polygeneration is defined as production of at least three different products, which in case of gasification typically are heat, electricity and biofuels [1].

Based on the concept of polygeneration, the fundamental idea of this project is to produce heat, electricity and hydrogen from product gas obtained from biomass steam gasification. Thereby, the overall aim is to develop economic feasible process configurations. This could be achieved on the one hand by realizing a polygeneration concept and as a consequence gaining more than one product. On the other hand process chains should be created which meet the requirements of both robustness and simplicity.

As illustrated in figure 1, the gasification process uses solid biomass as feedstock and produces gas. The produced gas undergoes gas cleaning and is processed further for production of heat and electricity. In addition, a slipstream is taken after the gas cleaning step. The slipstream passes an additional cleaning step, which is necessary for further gas processing. Afterwards, the gas is compressed and finally it is led to the separation unit. For

the separation (enrichment) of hydrogen from the product gas a membrane separation process is applied. The membrane splits the feed gas into two main gas streams, a low-pressure permeate and a high-pressure retentate stream. The permeate stream is mainly composed of hydrogen. The retentate stream (off-gas stream) contains components which remained at the feed-side of the membrane. In order to avoid any losses and following the polygeneration strategy, the off-gas is recycled and utilized for heat and electricity production.

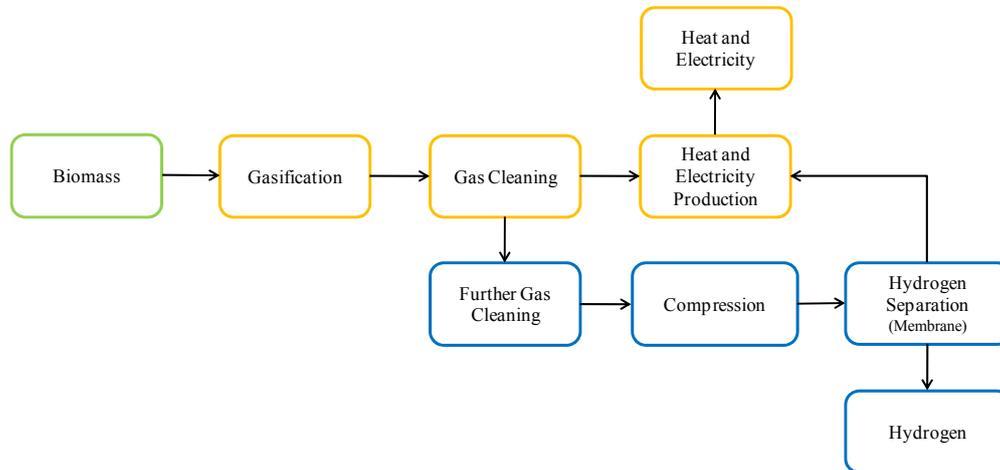


Figure 1. Flow sheet of polygeneration process

In this paper simulation results of membrane separation for hydrogen are presented which are used for designing a membrane test rig. This test rig will be operated in a slipstream of the Oberwart plant in the coming months.

2 DESCRIPTION OF THE POLYGENERATION PROCESS

The polygeneration concept described above is being implemented at the biomass gasification plant located in Oberwart, Austria. The gas produced in this plant was commonly used to produce heat and electricity, but in order to improve energy utilization and simultaneously meet the requirements of the polygeneration idea, a hydrogen production process is about to be put into operation.

2.1 Heat and Electricity Production

Figure 2 shows a simplified process flow sheet of the CHP plant Oberwart. Solid wet biomass (wood chips) is used as feedstock for the gasification process. Biomass is dried before it is fed into the gasifier. The gasification process is based on the dual fluidized bed technology. Accordingly, one fluidized bed is dedicated for gasification (fluidization with steam) whereas the other fluidized bed (fluidization with air) for combustion. Combustion is necessary for generating heat which is transferred through a circulating bed material to the gasification reactor in order to maintain the endothermic gasification process. Corresponding to the concept of two fluidized beds, two different gas streams are produced. In case of the gasification reactor product gas is obtained and in case of the combustion reactor it is flue gas.

Each of these two gases undergoes a certain gas treatment. The flue gas passes a gas cooler and a gas filter where particulate matter is separated. Afterwards it is led to the chimney.

Treatment of the product gas obtained from gasification also includes cooling, filtering and additional scrubbing. Within the product gas scrubber, which is operated with biodiesel (rape seed methyl ester), removal of water and tars takes place. By undergoing this treatment, the product gas gets conditioned for its further application in the gas engine.

A typical composition of the product gas at this stage of the process is given in table 1. Furthermore, the product gas contains apart from tars, trace components. Among them are nitrogen-, sulphur- and chlorine compounds.

Table 1. Typical composition of product gas from biomass steam gasification in a dual fluidized bed gasifier after gas cleaning for gas engine requirements (Data from Güssing gasifier) [2]

main components	value	unit
H ₂	35...45	vol% (dry)
CO	19...23	vol% (dry)
CO ₂	20...24	vol% (dry)
CH ₄	7...10	vol% (dry)
C _x H _y	2.5...4	vol% (dry)
N ₂	0.7...2	vol% (dry)

The product gas is used as a fuel in a gas engine for electricity and heat production. The off-gas (flue gas) from the gas engine passes a cooler and is finally led to the chimney. If necessary, an oxidation catalyst can be integrated for lowering the carbon monoxide emissions of the gas engine.

Moreover, additional electricity can be produced by an ORC (Organic Rankine Cycle), which is also implemented at the plant. Heat from both flue gas and producer gas contributes to the operation of the ORC.

In case of gas engine failure or start up the product gas can optionally be fired in a district heating boiler (Güssing plant) or in a flare (Oberwart plant).

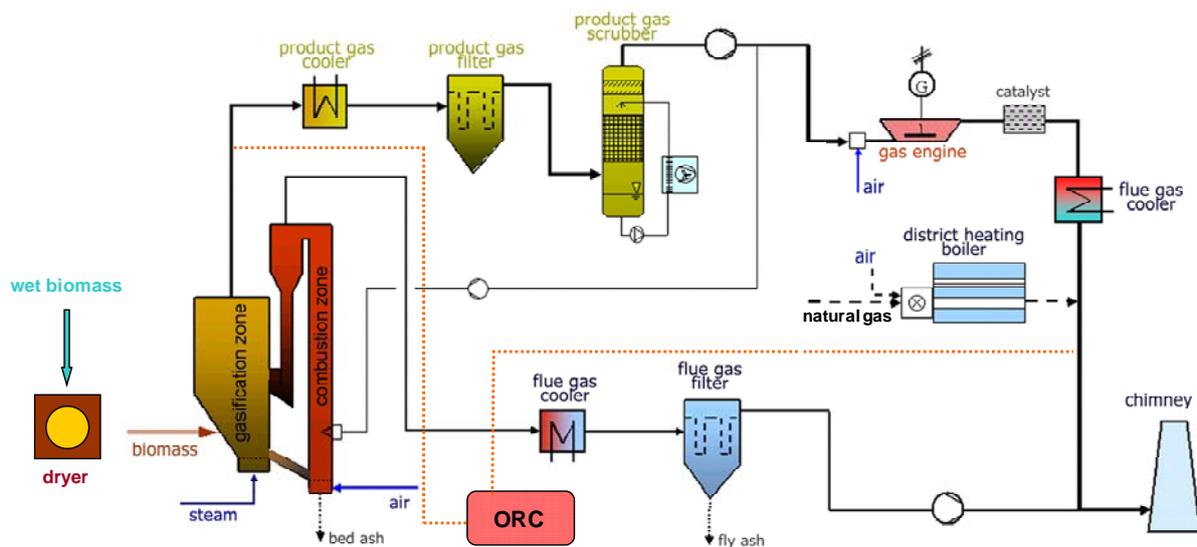


Figure 2. Simplified flow sheet of CHP plant Oberwart [3]

2.2 Hydrogen Production

For hydrogen production, a slipstream of the product gas is taken after the product gas scrubber. A membrane separation technology was chosen for separating hydrogen from the product gas. This separation technology is considered to be well suited for this application and it meets the requirements of robustness as well as simplicity.

Since the membrane has more stringent requirements with regard to gas purity compared to a gas engine, the product gas has to be further cleaned. The further cleaning consists of a second scrubber, also operated with biodiesel, but contrary to the first scrubber its operating temperature is lower. In addition an adsorptive gas cleaning is installed in order to remove much of the trace components, which otherwise could lower the performance of the membrane. After undergoing these cleaning steps the product gas is compressed to a certain pressure level and fed into the membrane.

The separation process is realized as single stage process. The feed stream is split into two streams, obtaining one permeate and one retentate stream. The permeate stream represents the product stream, containing gas components which were transported through the membrane (mainly hydrogen). On the contrary, the retentate stream represents a residual stream, consisting of gas components which remained on the feed side of the membrane.

Separation of hydrogen from the product gas by means of a membrane separation technology is in this case based on the principle of gas permeation. Gas permeation allows separation of gas mixtures without a change in the phase. Separation of different gases or gas components is achieved due to differences in molecular size and gas solubility in the membrane. [4]

The ideal selectivity $\alpha_{i/j}$ represents the most important parameter for describing membrane performance. Ideal selectivity is defined as permeability ratio of two gases or two components i and j of one gas and is given as

$$\alpha_{i/j} = \frac{P_i}{P_j} = \frac{D_i * K_i}{D_j * K_j}.$$

P ... Permeability [$m^2 / Pa s$]

D ... Diffusion coefficient [m^2 / s]

K ... Sorption coefficient [$m^3 / m^3 Pa$]

The ratio of diffusion coefficients indicates the relative motion of the molecules of the two components i and j and can therefore be considered as mobility selectivity. It is proportional to the ratio of molecular size of the two components.

The ratio of the sorption coefficients (sorption selectivity) indicates the relative concentration of the two components i and j in the membrane material and is proportional to the relative condensability of the components. [5]

The applied membrane separation process uses hollow-fiber membrane modules. More precisely, the actual membrane is constructed as a hollow-fiber and a certain number of hollow-fibers are bundled to form a module. The membrane consists of polyimide, which is a glassy polymer. Separation properties of glassy polymers heavily depend on the molecular size of the involved molecules [5]. Table 2 shows typical selectivities of the considered polyimide membrane for the mentioned components over longer alkanes (C_xH_y).

Table 2. Selectivities of the modeled polyimide membrane for main gas components of product gas

gas component	$\alpha_{\text{component/CxHy}}$
H ₂	750.0
CO ₂	83.3
CO	10.0
N ₂	4.2
CH ₄	2.5
C _x H _y	1.0

3 SIMULATION OF HYDROGEN PRODUCTION

3.1 Theoretical Background

Numerical modeling was employed in order to preliminarily investigate the applicability of the membrane separation process (gas permeation) for the separation of the hydrogen from product gas. The recently developed numerical algorithm uses a one-dimensional Gauß-Seidel finite difference method for the calculation of multicomponent transmembrane flows in membrane gas permeation systems. The algorithm was rigorously validated in a multicomponent permeation experiment and provided good agreement with the experimental results [6].

3.2 Boundary conditions of the simulation

In this work it was assumed that the gas transmembrane flow is ruled exclusively by the solution-diffusion mechanism. Moreover it is assumed that the membrane permeances are independent on other process parameters. Furthermore the flux-coupling effects as well as pressure losses along the membrane fibers in the feed channel and in the permeate channel were neglected.

The modeled case considered in this work is a single membrane stage operated in the counter-current configuration. It is assumed that the membrane stage is equipped with the membranes made of polyimide in the form of hollow-fibers.

The assumed feed gas composition for modeling is presented in table 3.

Table 3. Assumed feed gas composition for modeling

gas components	value	unit
H ₂	39.8	vol%
CO	22.3	vol%
CO ₂	22.4	vol%
CH ₄	10.3	vol%
C _x H _y	3.2	vol%
N ₂	2.0	vol%

3.3 Results

Figure 3 shows the hydrogen content in the permeate stream, what can be viewed as hydrogen purity, as a function of hydrogen recovery. Hydrogen recovery is defined as follows:

$$\text{hydrogen recovery}[\%] = \frac{\text{hydrogen in permeate flow}}{\text{hydrogen in feed flow}} * 100\% .$$

Furthermore, the figure also illustrates the pressure dependence of this function for pressures of 5 bar and 15 bar.

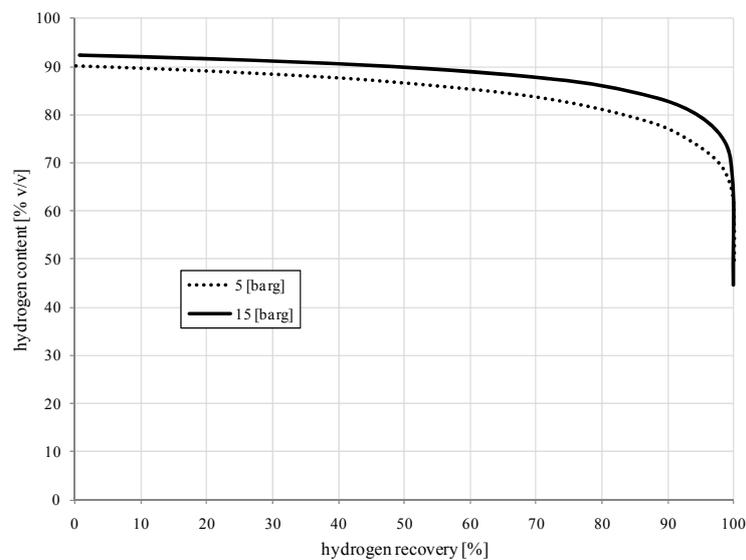


Figure 3. Hydrogen purity versus hydrogen recovery

The typical trade-off between recovery and purity can be observed. While at lower hydrogen recoveries hydrogen purities of above 90% are possible, an increase of hydrogen recovery leads to a decrease of hydrogen purity down to 80% or lower.

Referring to the pressure dependence, an increase of the operating feed gas pressure causes an improvement of the hydrogen purity. In order to give an example, at a hydrogen recovery of i.e. 60% the hydrogen purity would be about 87% at a pressure of five bar. However, at a pressure of 15 bar the hydrogen purity would increase up to a value of approximately 89%.

Figure 4 depicts the content of the gas components in the permeate stream versus the membrane stage-cut at an operating feed gas pressure of 15 bar. The membrane stage-cut is defined as follows:

$$\text{membrane stage - cut} = \frac{\text{permeate flow}}{\text{feed flow}} .$$

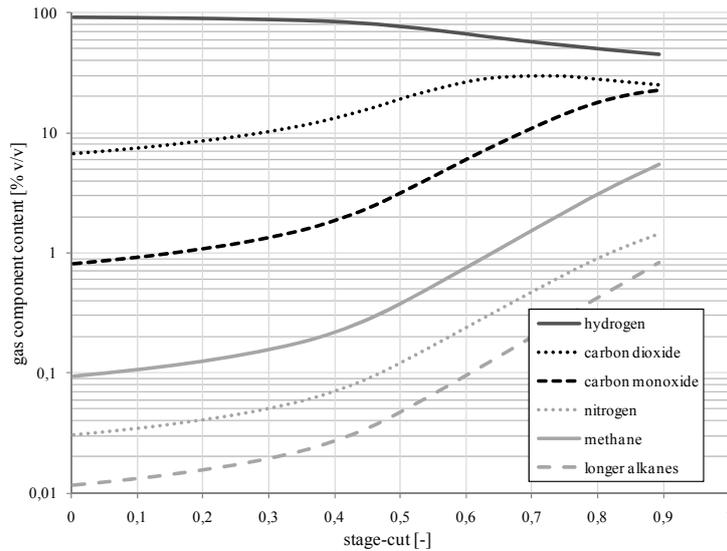


Figure 4. Gas composition of the permeate stream versus membrane stage-cut

As can be seen in this figure, the produced permeate stream is mainly composed of hydrogen. Apart from hydrogen, the gas also contains other gas components which also permeated at lower rate through the membrane. The amount of these gas components in the hydrogen-rich gas depends on the membrane stage-cut. In the range of lower stage-cuts (~ 0.15) a content of approximately 8 vol% of carbon dioxide can be expected in the produced gas (permeate stream). The next prevalent gas component is carbon monoxide with a content of approximately 1 vol%. The contents of other gases such as nitrogen, methane and lower alkanes are expected to be below 2000 ppmv.

In general, an increase of membrane stage-cut results in a decrease of the hydrogen content and in an increase of the amounts of other gases.

Figure 5 illustrates the content of the gas components in the retentate stream versus the membrane stage-cut at an operating feed gas pressure of 15 bar. This figure is a supplement to figure 4, since figure 4 shows the composition of the permeate stream under equal process conditions.

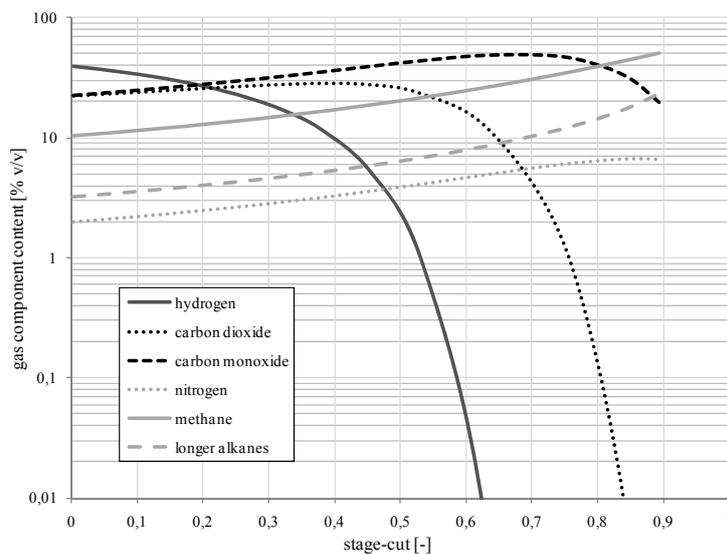


Figure 5. Gas composition of the retentate stream versus membrane stage-cut

In the range of average to higher membrane stage-cuts the predominate gas components are carbon monoxide and methane with contents from 20 vol% to 50 vol%. The contents of longer alkanes and nitrogen are expected to be 3 vol% and 5 vol% respectively at membrane stage-cuts of 0.5 and rising to 6 vol% and 14 vol% respectively at membrane stage-cuts of 0.8.

Hydrogen and carbon dioxide show similar behaviour regarding their content in the retentate stream at increasing stage-cuts. The content of both hydrogen and carbon dioxide sharply decreases at a certain stage-cut. The reason is that at this certain stage-cuts the whole amount of hydrogen and carbon dioxide contained in the feed gas has permeated through the membrane and therefore nothing remains at the retentate side of the membrane.

Corresponding to figures 4 and 5, figure 6 shows the expected gas composition of both permeate and retentate stream for an operating point with stage-cut of 0.14 an operating feed gas pressure of 15 bar.

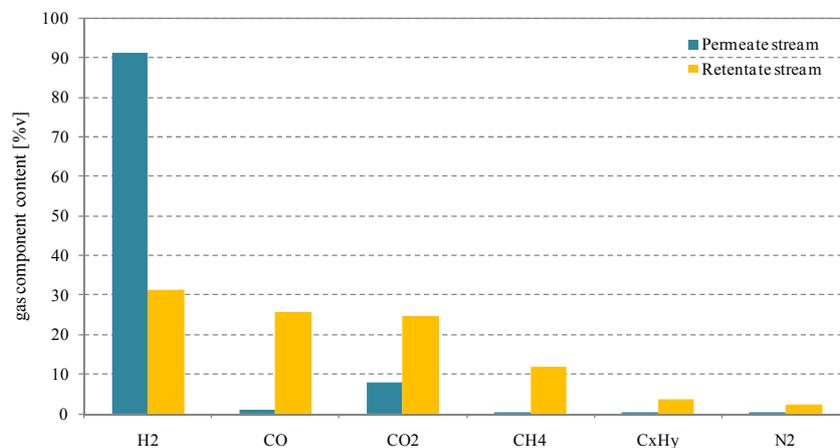


Figure 6. Gas composition of the permeate and retentate stream (stage-cut 0.14, 15 bar operating pressure)

Among the gas components of the permeate stream the most prevalent hydrogen, followed by carbon dioxide and carbon monoxide. The content of methane, longer alkanes and nitrogen is expected to be at a relatively low level.

On the contrary, the retentate stream is mainly composed of quite equal amounts of hydrogen, carbon monoxide and carbon dioxide. Apart from these three components, also methane, longer alkanes as well as nitrogen are present but to a lower extent.

4 CONCLUSION & OUTLOOK

This paper presents the polygeneration process which is going to be implemented at the biomass gasification plant in Oberwart, Austria. In addition to heat and electricity production, a hydrogen production process is going to be realized using a membrane separation process. A simulation of this process was carried out and results show that separation of hydrogen from product gas is basically possible. The content of hydrogen depends on several parameters such as the desired hydrogen recovery, the operating pressure and the stage-cut.

The hydrogen production process will be put into operation at the plant within the next few weeks. Results of the first operational experiments will give insight into the feasibility of this process and the obtained product gas qualities. Moreover, more information about how trace

components contained in the product gas of the gasification process affect the membrane will be gained.

If the obtained retentate stream composition possibly fits other process requirements, next steps could be evaluating more sophisticated applications of this gas instead of heat and electricity production only.

5 ACKNOWLEDGEMENT

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6 REFERENCES

- [1] Hofbauer, H.; “Gas production for Polygeneration Plants”, International Conference on Polygeneration Strategies 2009, Vienna (2009)
- [2] Potetz, A., Rauch, R. and Hofbauer, H.; „OptiBtLGas – Steam Reforming of Product Gas from Biomass Steam Gasification”, International Conference on Polygeneration Strategies 2009, Vienna (2009)
- [3] Kotik, J.; Flow sheet Oberwart, Personal communication, Vienna (2010)
- [4] Scott, K.; “Handbook of Industrial Membranes”, Elsevier (1995)
- [5] Baker R.W. and Lokhandwala K.; “Natural Gas Processing with Membranes: An Overview”, Ind. Eng. Chem. Res. 47 (2008), p. 2109 – 2121
- [6] Makaruk, A. and Harasek, M.; “Numerical algorithm for modeling multicomponent multipermeator systems”, Journal of Membrane Science 344 (2009), p. 258 – 265