

Key role of membrane gas separations in the utilisation of an underground natural gas reservoir for the renewable energy storage

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Abstract

Current developments in the energy production focus increasingly on renewable resources. Especially, the number of wind and solar power facilities has grown strongly in the recent years. As a result, the worldwide installed power for these energy sources can be counted already in hundreds of gigawatts.

The growth of renewable energy production leads to difficulties in the electrical grid control and power distribution. This is because the wind and solar power is unsteady and the power delivered to the grid is determined by the weather, which is not controllable. This situation creates a background for the development of various Power-To-Gas (P2G) concepts that attempt to utilise the natural gas infrastructure for the renewable energy storage and distribution. One of these concepts is to add hydrogen to the natural gas system.

In this work we discuss a membrane gas permeation system that allows to adjust hydrogen concentration for sensitive parts of the grid or individual customers to a certain limit. Surplus Hydrogen can be utilized for electricity production or further processed for hydrogen applications. The principal setup of this gas processing technology is going to be tested in the course of the Underground Sun Storage project, where the hydrogen content of the withdrawn gas is adjusted to a limit acceptable for the grid.

Background

Driven by the climate concerns, large investments in the field of renewable energies have been made in the recent years. Besides various types of bioenergy, which in spite of many efforts remains rather marginal, the wind power and the solar power proved to be affordable renewable energy sources that in the meantime make a significant contribution to the European electrical power generation. This trend is clearly visualised in the statistical data in Figure 1. Whereas in 2000 in EU the contribution of the solar power was negligible and the wind power contribution was mere 2,4%, in 2015 wind and solar sources make already more than a quarter of the total installed generation power. In absolute numbers the contribution as for 2015 is 142 GW for the wind power and 100 GW for the solar power (understood as the sum of the photovoltaic and the concentrated solar power). According to the EWEA in 2015 the wind power covered around 11% of the total electrical energy consumed in the EU [1].

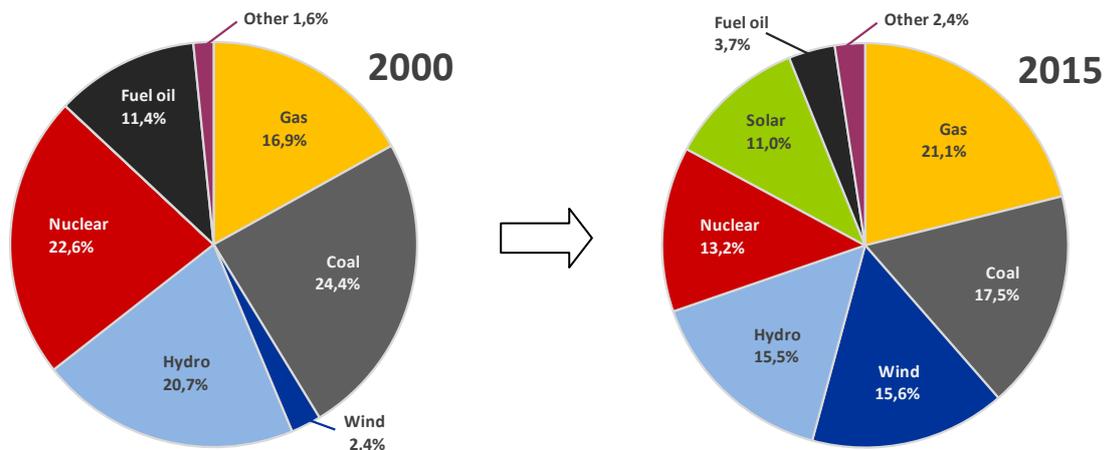


Figure 1: shares of different energy sources in the EU power mix in 2000 (left) and 2015 (right), adapted from EWEA statistics [1]

Keeping in mind the European Energy Directive for 2030 and the European Energy Roadmap for 2050 that set ambitious cuttings of greenhouse gas emission of the respective 40% and 80% in comparison to the 1990 level and also keeping in mind the EU objective to completely decarbonize the power generation sector by 2050, a strong further expansion of wind and solar energy is to be expected in the coming years. The year 2015 was marked by the installation of new wind and solar power plants with the total generation power of around 20GW, which was accompanied by numerous decommissionings of conventional fossil and nuclear fuel power plants [1].

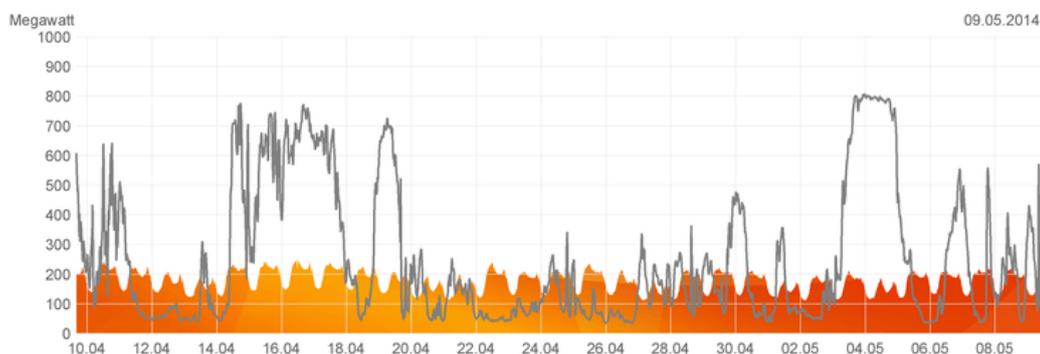


Figure 2: power consumption (orange) and power generation (dark gray) trend over a month in the wind-rich Austrian region Burgenland [2]

Although wind and solar power energy sources fulfil the climate objectives they have an undesired feature concerning the grid balancing. Since the production of the power from these renewable sources is dependent on the weather and the season, it is intermittent and cannot be controlled to follow the energy consumption. The discrepancy between the available power and the consumption is visualised on an example of a wind-rich Austrian region in Figure 2. Whereas during windy periods large surplus of energy is produced, there are other periods with the energy deficit that must be balanced. Hence, storage of large energy quantities will be indispensable to make the power generation based on renewable energy work.

Power-to-gas and the gas processing requirement

A solution for the energy storage problem associated with the renewable power generation can be provided by the utilisation and integration of the existing natural gas infrastructure. The surplus of electrical energy can be converted to synthetic natural gas, which can be stored and used later for the power generation. This is the basic idea of “Power-To-Gas” energy storage concepts that are currently discussed within the European community. According to many literature sources, storing of the electrical energy in the form of combustible gases like hydrogen and synthetic natural gas could provide very high storage capacities (in >GWh range) and high charge/discharge rates. It is one of a very few options available to manage the large surplus of renewable electrical energy that will be generated within the future years [3-6].

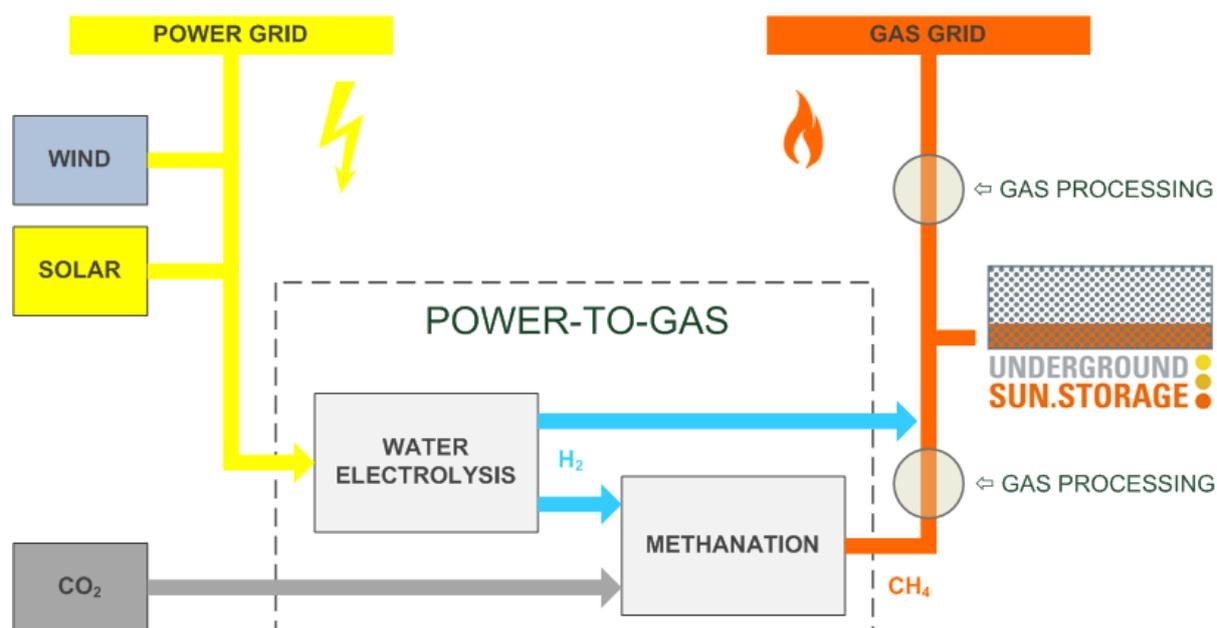


Figure 3: the basic concept of power-to-gas and possible process locations with the gas processing requirement, adapted from [4].

The basic scheme describing the power-to-gas idea is shown in Figure 3. Here, the surplus of electrical power is converted to hydrogen by water electrolysis. This hydrogen can be injected to a natural gas grid or stored in an underground gas storage system. Alternatively, hydrogen can be used together with carbon dioxide to synthesise methane, which can be subsequently supplied to gas grids and stored. As for the methane synthesis, both the chemical methanation on nickel catalysts and the biological methanation are considered as possible options.

Hydrogen injection is likely to induce several problems related to the function and safety of natural gas grid components [7]. Hydrogen changes the natural gas combustion parameters; i.e. it reduces the heating value (volume related) and Wobbe index making the gas mixture incompatible with certain applications. Moreover other gas parameters like the gas viscosity and the heat capacity are also influenced. In general, natural gas grid components demonstrate different compatibilities with hydrogen. Certain elements like for example PVC/PE-pipings, valves and pressure regulators are compatible with relatively high hydrogen contents (30% and more). At the same time, there are other grid components like gas turbines, compressors and CNG-tanks that allow only very low hydrogen content in natural gas (<2%). Therefore it is evident, that if hydrogen injection to natural gas grids is undertaken, a flexible hydrogen separation technology for the adjustment of the hydrogen content will be required.

Moreover, on the account of the periodic hydrogen injection it is probable, that considerable hydrogen content variations will be produced within a gas grid section, a gas pipeline or a gas storage system. Thus again a hydrogen separation technology is required for the balancing of the hydrogen content and the provision of constant gas parameters to downstream gas consumers.

Hydrogen/methane separations with membranes

Among various separation processes, membrane gas permeation offers suitable features for the hydrogen separation in newly developed power-to-gas systems. In the membrane gas permeation, a gas mixture is fed under pressure into membrane modules. There, the gas mixture contacts the membrane, which is in the form of a continuous thin polymer film. Driven by the partial pressure difference, gas molecules dissolve in the polymer film, diffuse through it and desorb on the other side of the membrane. The permeation through the polymer film is ruled by the solution coefficient (S) and the diffusion coefficient (D), the product of which makes the permeability coefficient (P). The relative permeability (or permance) order for the most important gases in the permeation through a glassy polymer is shown in Figure 4.

Fast permeating gases are enriched on the low-pressure (permeate side) of the membrane and depleted on the high-pressure side (retentate). For slow-permeating components this behavior is reversed. The gas flow through thin polymer films is expressed by

$$Q = P A (p_x - p_y) / \lambda, \quad (1)$$

where A is the membrane area, p_x is the gas component partial pressure in feed/retentate, p_y is the gas component partial pressure on the permeate side and λ is the polymer film thickness. The ideal selectivity for a gas pair is then defined as

$$\alpha = P_1 / P_2. \quad (2)$$

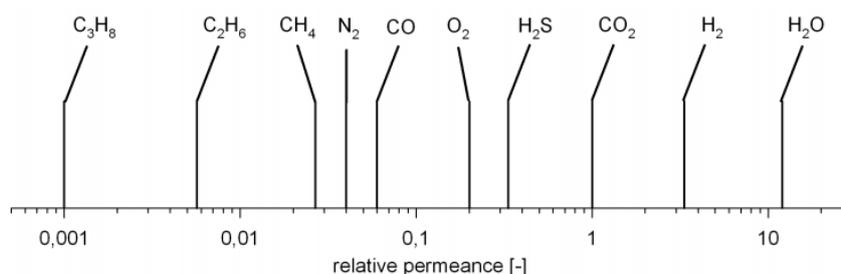


Figure 4: relative permeance order of the most important gases for a glassy polymer [8]

The ideal selectivity for a polymer material is a basic measure describing its ability to separate two gas mixture components from each other. For the relevant gases, methane and hydrogen, the ideal selectivity of available membranes is relatively high. Commercial membranes made of glassy polymers like polyimide, polyaramide and polysulphone reach ideal selectivity values for hydrogen and methane of around 100 [9]. The selectivity for hydrogen versus other aliphatics is even higher. This is an excellent selectivity value, which allows realisations of efficient gas separation processes.

In addition to very good separation capabilities, the membrane gas permeation has a series of further advantages relevant to power-to-gas systems, which are:

- 1) Compactness and good scale-down
- 2) Continuous operation and easy control
- 3) Operation without chemicals
- 4) Use of the available pipeline pressure for the separation resulting in a low energy consumption
- 5) Along with hydrogen, separation of other undesired components like carbon dioxide and water vapour within one process step.

Hydrogen separation in Underground Sun Storage

Underground Sun Storage is the acronym of a cooperative research project in Austria under the lead of RAG. The project focuses on the storage of renewably generated hydrogen in underground porous structures. Besides several work packages focusing on different technical, social and economic aspects of hydrogen storage and use, the main objective of the project is the experimental investigation and demonstration of the hydrogen storage process at an industrial scale. To achieve this, a pilot plant for the hydrogen generation and storage at a gas storage facility of RAG in Upper Austria has been erected and commissioned. In the first phase, the mixture of natural gas and hydrogen will be injected into the reservoir. For the tests, the planned storage input gas flow equals ca. 700 Nm³/h and the stored hydrogen content equals 10% (v/v).

Along with the hydrogen storage tests, a pilot membrane unit for the hydrogen separation has been constructed (Figure 5) and will be operated during the gas withdrawal phase of the stored hydrogen-natural gas mixture. The general aim here is to investigate the membrane process in the hydrogen separation and the gas conditioning to reduce the 10% (v/v) hydrogen content down to the regulated 4% (v/v) (Austrian ÖVGW G31). The natural gas mixture with the 4% hydrogen content can be subsequently supplied to the gas grid. The permeate flow with enriched hydrogen can be used for the power generation at the discharge phase or can be recycled to the gas reservoir depending on the energy demand (Figure 6).



Figure 5: hydrogen separation unit using based on a one-stage membrane gas permeation system constructed within the Underground Sun Storage project

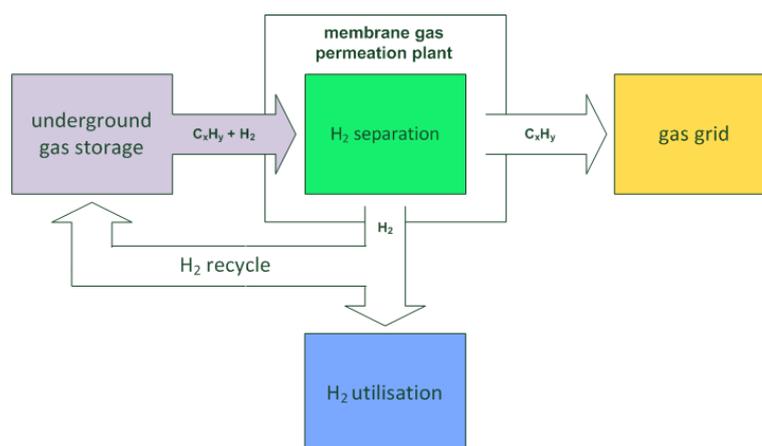


Figure 6: simplified scheme for the integration of a membrane hydrogen separation unit at an underground gas storage facility

Modelling results of the hydrogen separation for the membrane unit are shown in Figure 7 and Figure 8. The presented case embraces a constant withdrawal rate of 400 Nm³/h of a mixture of natural gas and 10% (v/v) hydrogen, which is directly fed to the membrane unit. At the withdrawal, the feed pressure sinks gradually from the maximal storage pressure of 68 bar down to the bottom storage pressure of 28 bar. The membrane unit uses directly this pressure for the gas separation and is able to reduce the hydrogen content below 4% (v/v) in the retentate. As for the permeate stream, the hydrogen is enriched to 40 - 60 % (v/v) depending on the feed pressure (Figure 8). The second most abundant component in the permeate is methane, while the content of other components is below 0,3% (v/v).

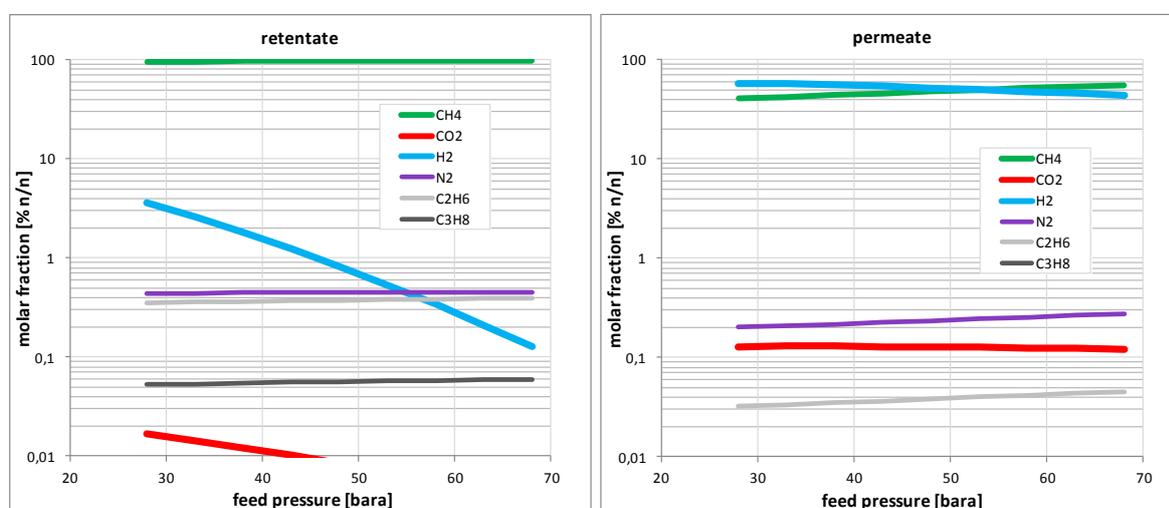


Figure 7: modeling of the gas composition in the retentate and permeate in the hydrogen separation of a natural gas - hydrogen mixture stream, gas flow 400 Nm³/h, hydrogen content in the feed gas 10% (v/v), pressure ramp 68 bar down to 28 bar.

Modelled gas recoveries for the aforementioned separation process are shown in Figure 8. The single stage membrane is able to achieve hydrogen recoveries between 70% and 99% in the permeate, while the methane recovery in the retentate is in the range of 86% to 95%, depending on the feed pressure. It is visible, that gas parameters achieved in the separations for the feed gas pressure ramp are variable. In the test phase an optimized control procedure using control valves in the feed, the permeate and the retentate will be applied to level out variations and provide the constant separation parameters.

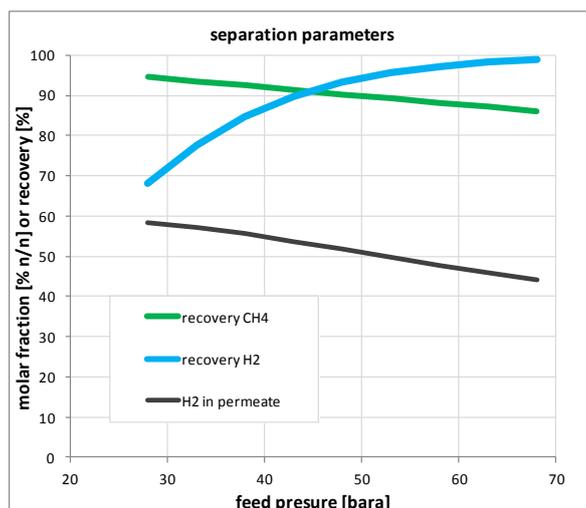


Figure 8: modeling of hydrogen recovery in permeate and methane recovery in retentate in the hydrogen separation of a natural gas – hydrogen mixture stream, gas flow 400 Nm³, hydrogen content in the feed gas 10% (v/v), pressure ramp 68 bar down to 28 bar.

Outlook

Next years will bring further extensive investments in the wind and solar power. Power-to-gas systems will have to be implemented in order to cover the acute need for the electrical energy storage. Typically, power-to-gas involves injection of hydrogen to natural gas grids and it will be desired to adjust the hydrogen content for the gas grid balancing as also to separate hydrogen from gas mixtures in order to use it for the power generation during periods with the electrical energy deficit.

Membrane gas permeation is a compact and flexible process that can be installed at different gas grid localisations and provide a technical solution for the hydrogen separation problem. The advantages of membranes are the high separation efficiency, low energy consumption and simple operation.

More information on the progress in the Underground Sun Storage project and the hydrogen separation unit can be found at <http://www.underground-sun-storage.at>.

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