



## Energy consumption of the hydrogen separation process from a mixture with natural gas

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### ABSTRACT

The importance of energy storage methods increases in the global energy industry. One of such methods is the production/generation of hydrogen and its further use called power-to-gas technology. Without highly efficient, reliable and cheap energy storage solutions, the energy sector, especially in Poland, may face significant problems related to climate protection and reduction of greenhouse gas emissions to the atmosphere in the nearest future. One alternative way to store and transport hydrogen is to blend small amounts of hydrogen (up to about 15%) into existing natural gas networks. When hydrogen is required, it can be separated near end-users using separation membranes. The paper presents the possibilities of using membrane technology in the process of obtaining hydrogen as fuel after its storage. The membrane separation technology can be used to separate mixtures of various gases, such as H<sub>2</sub>, O<sub>2</sub>, CO, CO<sub>2</sub>, or CH<sub>4</sub>. The paper presents selected problems of the analysis of the hydrogen separation process from a mixture with natural gas for polymeric membranes with different separation properties. The structure of the separation system has been selected in order to obtain the appropriate purity of the separated gas. The energy consumption of the process of hydrogen separation from a mixture with natural gas was determined for such a system.

*Keywords:* Hydrogen; Membrane separation; Gas purification

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### 1. Introduction

The problem of reducing greenhouse gas emissions is known to be of great economic, social and scientific importance for several decades. The main factor affecting climate change is carbon dioxide emitted into the atmosphere through various industrial processes, and primarily by the energy sector. As a result, mankind is looking for environmentally friendly alternative fuels. Among such future fuels, hydrogen seems to be applicable. It is one of the most common elements in nature. It has a higher calorific value than fossil fuels such as gas, oil, or coal. Also, burning hydrogen does not emit substances harmful to the environment [1,2]. It usually occurs in a form bound with

other elements/compounds. It can be obtained from various raw materials and in several ways. Currently, the most commonly used methods of obtaining hydrogen are steam reforming of methane, oil, and coal, as well as electrolysis processes [1,3]. It is also produced on a smaller scale using plasma technology or biological methods. Currently, the most cost-effective methods of obtaining and storing hydrogen state matter of vital investigation, which results will contribute to the widespread use of hydrogen – in everyday life and transport, power generation, and industry. The growing interest in hydrogen-based devices, technologies and systems is contributing to the global increase in demand for hydrogen, which has increased more than 2.5 times over the past 40 y [4], as it is a gas widely used by

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the petrochemical, chemical, metallurgical, pharmaceutical and textile industries. Also, the automotive industry uses hydrogen as a green fuel.

Therefore, the allocation of hydrogen in the natural gas network is indicated as an effective way of storing energy produced in the power system at times of surplus in relation to demand. This method of energy storage is supported by the possibility of using hydrogen energetically in any location where there is a natural gas network, without incurring additional capital expenditure for the development of special storage systems. Another advantage is the relatively low cost of transportation of an energy carrier such as hydrogen, for example, in comparison to the transportation of electric energy. In order to transport hydrogen to its destination, transport of this gas together with natural gas through the existing gas network may be considered. The hydrogen must then be separated from this mixture. Several different separation technologies can be used from the hydrogen/natural gas mixture, including EPH (electrochemical separation), PSA (pressure swing adsorption), or membrane technologies [1,2,5–10,22–24,29,30].

## 2. Hydrogen separation from natural gas mixtures

The technologies of hydrogen separation from other gases, including gases created on the basis of solid fuel gasification and steam reforming processes, as well as from coke oven gas, are well-recognized technologies. Gasification or reforming processes of other fuels lead to the production of hydrogen as a component of a mixture of various gases, which also quite often include a number of impurities. In such cases, independently on the required final purity of the extracted hydrogen, within the  $H_2$  separation technology, a number of unit processes can be separated; those include removal of tars, sulfur compounds, and carbon dioxide. As an example of hydrogen separation technology used on a large industrial scale, one can point to the Pressure Swing Adsorption (PSA) physical adsorption technology.

After hydrogen is stored and transported in a gas mixture, it must be separated from the gas for use. When separating hydrogen from a gas mixture, safe, automated, inexpensive, and high-purity solutions should be sought. For the separation of hydrogen from natural gas, we can use membrane separation processes, electrochemical hydrogen separation processes (EPH), or physical adsorption processes (PSA) [26,30]. For membrane separation, both polyimide membranes, dense metal membranes - especially those made from palladium and its alloys [5,6,11,12,27,30] - and carbon molecular sieves can be used.

On an industrial scale, hydrogen gas streams are most often separated by pressure swing adsorption methods due to the maturity of this technology, as has been used for years [13,30]. The adsorption process makes it possible to obtain gases of high purity. The hydrogen obtained in this way is under high pressure - the pressure that prevailed in the adsorption column and varies from 8 to 20 bar.

An alternative way to separate hydrogen from a natural gas mixture is to use a membrane separation process. Regardless of the membrane material used, the driving force behind the separation process is the high-pressure difference on both sides of the membrane. Hydrogen is the gas that

permeates the membrane. As a result, the resulting hydrogen is under low pressure, and after the separation process, recompression is required for further use. This involves additional energy expenditure. Dense metal membranes made of palladium and its alloys (Pd and Pd-alloys) can be used for hydrogen separation [9,28,30]. They are characterized by very high selectivity and it is possible to obtain hydrogen of 99.99% purity, but due to hydrogen embrittlement, a high process temperature is required - above 573 K [14].

Further membranes applicable for hydrogen separation are carbon-based membranes. These can be divided into three groups: carbon membranes, carbon molecular sieve membranes (CMSMs), and carbon nanotubes (CNTs). Carbon membranes have higher selectivity than any other known polymer, but their strong disadvantage is brittleness and breakage. Thus, careful handling during the process is required. Selectivity and permeability of carbon molecular sieves (CMSMs) significantly vary, based on literature data [14,29]. Nonetheless, these parameters are higher than for polyimide membranes [8,15–18,29,30]. Most of the CMSMs membranes lie outside the 2008 Robeson curve shown in Fig. 1 (this curve applies only to polymeric membranes). Carbon molecular sieves were used to separate ultra-pure hydrogen from a hydrogen-enriched natural gas mixture. CMSMs membranes with selectivity above 4,000 and permeabilities of 1,000 Barrer were used (these tests were performed for conditions of 6 bar and 90°C) [14,30]. Unfortunately, the price of these membranes is unknown and CMSMs are currently not commercially available.

The paper [30] presents several membrane separation systems for CMSMs membranes, including one-stage and two-stage membranes systems containing a vacuum pump, compressor between membrane stages, and electrochemical hydrogen compressor, as well as systems with the use of sweep gas on the permeate side and hybrid systems with temperature adsorption process (TSA systems). For all the different configurations of hydrogen separation systems, energy intensity was determined to range from 3.94 kWh/kg $H_2$  to over 8.02 kWh/kg $H_2$  (and even 12.64 kWh/kg $H_2$ ).

Polymeric membranes are currently used to separate hydrogen from gaseous mixtures, including nitrogen, carbon monoxide, and hydrocarbons [8]. Polyimide membranes are characterized by high selectivity, are commercially available, economically attractive, and are close to the trade-off Robeson limit curve is shown in Fig. 1.

Another way of hydrogen separation can be electrochemical membranes, regarding fuel cell (PEM fuel cell) processes or water electrolysis processes [19,25]. Freeman and Pinnau [14], the authors reported the separation of hydrogen at 8%–100% (v/v) in a hydrogen/methane mixture on a proton exchange membrane (PEM fuel cell). Pure  $H_2$  was then obtained with a recovery rate of 80%.

Hybrid processes are also used to separate hydrogen - a combination of the membrane separation process and the adsorption process described in [15,19,20]. A schematic of such a hybrid membrane-adsorption process (HylyPure® process) [19] is shown in Fig. 2. This process allows to achieving the required hydrogen purity for different hydrogen concentrations in the feed gas (change of  $H_2$  content in the gas in the range of 1%–10% (v/v)). The first stage in the hybrid process is membrane separation,

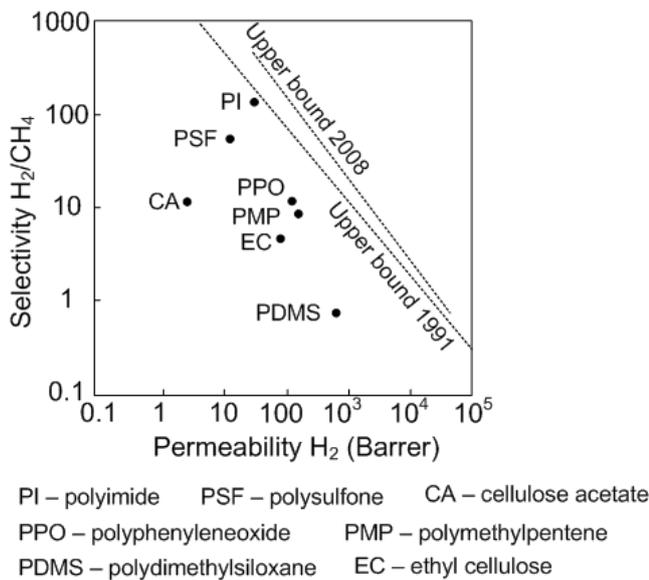


Fig. 1. Robeson curve and hydrogen permeability as a function of membrane selectivity towards methane ( $H_2/CH_4$  selectivity) [3,11,13–15].

which provides the initial enrichment of the gas stream with hydrogen. The second separation step is the PSA system, where a pure hydrogen stream is obtained [11,15,20,21]. The remaining residual gases (PSA desorbates) are re-compressed and returned to the natural gas grid. The idea of such a system is to be economically viable, energy efficient and, environmentally friendly.

### 3. Energy consumption analysis of hydrogen membrane separation systems

This paper focuses on membrane systems for the separation of hydrogen from a natural gas mixture. The results presented are obtained from numerical studies performed using the Aspen software. A membrane separation module (membrane separator) consists of a set of individual membrane modules connected in parallel, in series, or in any combination of these connections. The performance of such membrane systems vitally depends on how the modules are interconnected with each other. The optimal configuration of the membrane separation unit due to its design remains an important issue affecting the final process parameters.

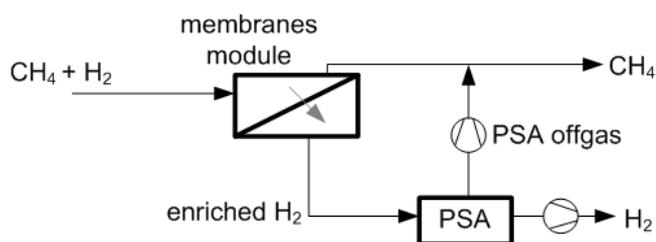


Fig. 2. Schematic of the HylyPure process concept [15,19].

The analysis of the separation process of hydrogen from a mixture with natural gas was carried out using Aspen Custom Modeler software. The separation process model was based on several assumptions. First of all, reciprocating flow on the feed side and locally undisturbed flow on the permeate side was assumed. Next, interaction between permeating components, pressure drop, and axial dispersion were neglected. Concentration polarization was assumed as negligible on both sides of the membrane. The process was set as isothermal at a constant temperature of 20°C. The gases were treated as semi-perfect. The membrane module consisted of cross-flow capillary tubes with constant values of permeability coefficients for gases being assumed. The membrane module was fed with a mixture of hydrogen and methane with proportions of:  $H_2 = 10\%$  and  $CH_4 = 90\%$ , for a nominal feed flow rate of 10 kmol/h.

The decision variables in modelling the hydrogen separation from the mixture with natural gas were:

- change in membrane module area,
- the change of the feed pressure,
- change of permeate pressure,
- change of separation system configuration,
- membranes with different separation parameters were used.

For the membrane operating conditions defined as above, the purity of the resulting hydrogen stream and its recovery rate was determined.

Two kinds of polymeric membranes were used in calculations. The first membrane (Membrane A), was assumed to be a polyimide membrane, which has a permeability coefficient for hydrogen of  $P_{H_2}^* = 28.1$  Barrer, while for methane this coefficient is  $P_{CH_4}^* = 0.25$  Barrer. This gives us an ideal selectivity coefficient for these gases of  $\alpha^* = 112.4$ . The second membrane selected for analysis (Membrane B) is a polyetherimide membrane with permeability coefficients of  $P_{H_2}^* = 1.32$  Barrer,  $P_{CH_4}^* = 0.035$  Barrer and  $\alpha^* = 222.86$  respectively. The selectivity coefficient is determined from the relationship:

$$\alpha^* = \frac{P_{H_2}^*}{P_{CH_4}^*} \quad (1)$$

where  $\alpha^*$  is the ideal selectivity coefficient;  $P^*$  is the permeability coefficient, Barrer.

The gas mixture pressure was assumed at the level of 33 bar, while the permeate pressure was varied within  $0.1 \text{ bar} \geq p_p \geq 1 \text{ bar}$ . The gas pressure at the feed corresponded to the pressure prevailing in the natural gas high-pressure network. The membrane area was treated as a decision variable and was changed in the range of  $200 \text{ m}^2 \geq A \geq 2,000 \text{ m}^2$  for polyimide membrane and was changed in the range of  $200 \text{ m}^2 \geq A \geq 1,600 \text{ m}^2$  for polyetherimide membrane. The change of membrane surface area in such a range allowed selecting process conditions of the lowest power demand of the  $H_2$  separation process keeping simultaneously possibly high purity parameters of separated hydrogen and its recovery rate. The effect of the membrane surface change and the pressure of the gas

stream received downstream the membrane (permeate) on the purity of the separated hydrogen and the hydrogen recovery rate is shown in Fig. 3.

It can be observed that the one-stage membrane separation system for both membranes is not able to provide adequate parameters for purity and hydrogen recovery, that is, to maintain parameters above 0.9 for both types of membranes. For a polyimide membrane (membrane A), in the assumed range of variation of permeate area and pressure, although a high degree of hydrogen recovery was obtained, the purity did not exceed the value of 0.85, while for a polyetherimide membrane B, the assumed hydrogen purity of 0.9 in the whole range of variation of permeate pressure was achieved, but the degree of hydrogen recovery varied from 0.6 to 0.85 at this assumed H<sub>2</sub> purity. Therefore, as an alternative to the system presented in Lamichaney et al. [20] based on the pressure swing adsorption process, it was proposed to use a two-stage membrane system in which the permeate from the first stage feeds the second membrane stage. A schematic of such a solution is shown in Fig. 4 [15].

The second stage of the membrane system contained a membrane made of the same material as that in the first separation stage. An analysis of the effect of the membrane surface and the suction pressure downstream of the membrane on the second stage was carried out. The feed stream for the second stage was obtained from the gas separation in the first stage. For the permeate obtained from the module containing a polyimide membrane (membrane A), received at an ambient pressure  $p_{p(1st)} = 1$  bar and a membrane area of  $A_{(1st)} = 1,630$  m<sup>2</sup>, the permeate gas parameters  $Y_{H_2} = 0.5545$  and  $R_{H_2(1st)} = 0.97$  were obtained. For the analysis of a two-stage system with a membrane made of polyetherimide (membrane B), the permeate flux received at ambient pressure was also chosen, for which a 97% hydrogen recovery

rate was obtained. The hydrogen purity at this point was  $Y_{H_2} = 0.6172$  for a membrane area of  $A_{(1st)} = 8,990$  m<sup>2</sup>. The results of the analysis of the use of the two-stage system on the hydrogen purity and the total hydrogen recovery rate for both membranes tested are shown in Fig. 5.

The total recovery rate was determined from the relation:

$$R_C = \frac{Y_{H_2-C} \cdot \dot{n}_{p(2st)}}{X_{H_2-F} \cdot \dot{n}_F} \quad (2)$$

where  $X_{H_2-F}$  is the hydrogen content in the feed to the membrane system;  $\dot{n}_{p(2st)}$  is the permeate stream is withdrawn from membrane system, kmol/h;  $\dot{n}_F$  is the gas stream feeding the membrane system, kmol/h.

It is noticed that for a polyimide membrane (membrane A), by using an appropriately chosen membrane surface and pressure on the permeate side, very high purity of the obtained hydrogen can be obtained – even exceeding the

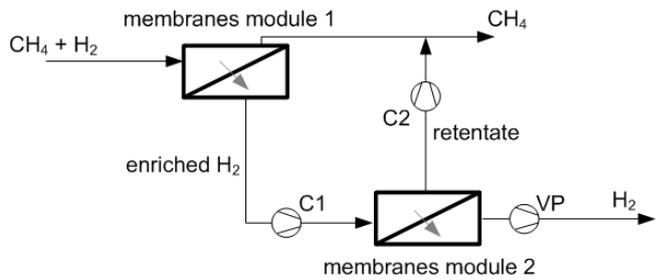


Fig. 4. Two-stage hydrogen separation system from a mixture with natural gas.

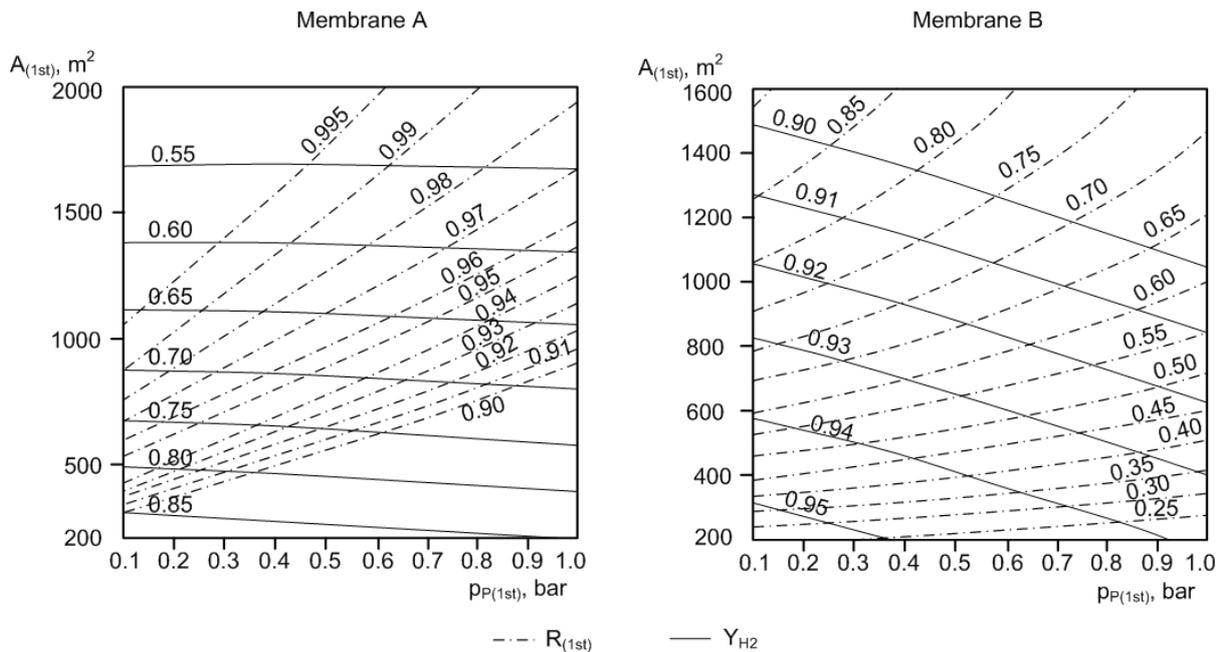


Fig. 3. Influence of membrane pressure and surface area on the purity and degree of hydrogen recovery in a single-stage system for membrane A [15] and membrane B.

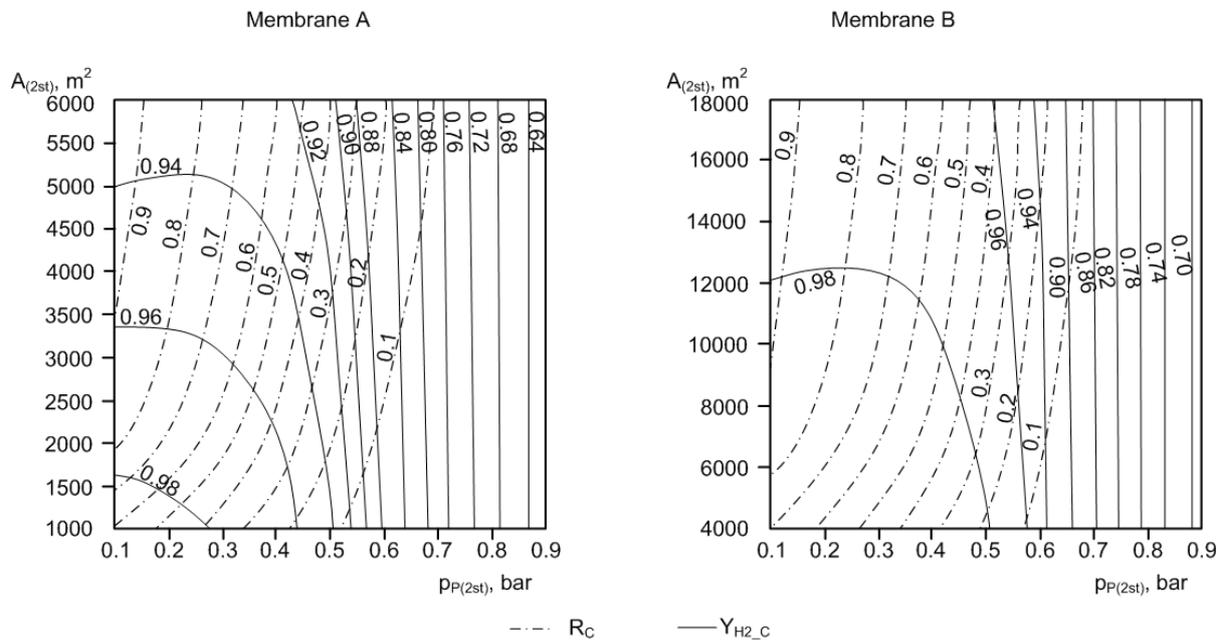


Fig. 5. Purity and hydrogen recovery as a function of change in the area at the second stage and pressure downstream of the second membrane module for the same membranes at both stages for membrane A [15] and membrane B.

value of 0.98. At the same time, the total hydrogen recovery in such a process will not exceed 70%. The total hydrogen recovery rate increases with a rising area on the second stage of the membrane module. It is possible to recover 90% of the hydrogen contained in the stream supplied to the separation system while maintaining the purity of the  $H_2$  stream at less than 0.96. For a polyetherimide membrane (membrane B), the purity of the permeate is as high as 0.99, while the total degree of recovery does not exceed 80%.

Therefore, a modification of the system  $x$ , where different membranes were used on both stages, was postponed. The membranes were selected to obtain both a high degree of hydrogen recovery and high purity of the hydrogen. A more detailed analysis of the influence of the polyetherimide membrane surface (membrane B) on the obtained hydrogen recovery rate and purity was carried out for the assumed feed pressure of the membrane module amounting to  $p_F = 33$  bar. The results of this analysis are shown in Fig. 6. It can be observed that for a properly sized membrane in the membrane module, a very high hydrogen recovery rate can be achieved, while the proportion of hydrogen in the gas mixture increases from 0.1 to about 0.5. However, the second gas stream obtained (retentate stream – gas retained on the membrane) consists mainly of methane. Fig. 7 shows methane purity and methane recovery ratio in the retentate stream.

The permeate obtained from the first stage with a polyetherimide membrane (membrane B) was directed to the second stage of the module, where a polyimide membrane (membrane A) was located in order to maintain a high level of hydrogen recovery and to obtain a purity of at least 80% of the obtained hydrogen. An analysis of the effect of membrane surface area and suction pressure downstream in the second stage of the module was carried

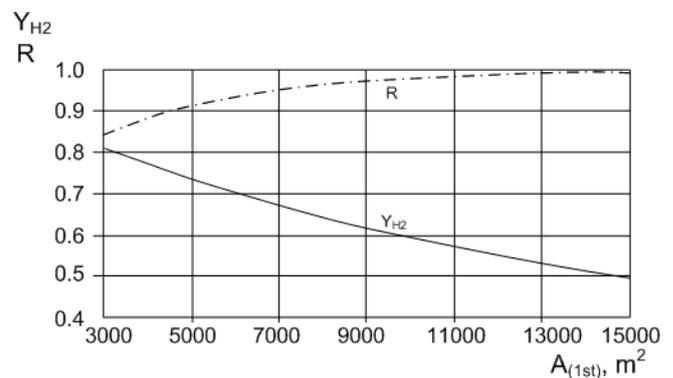


Fig. 6. Purity and hydrogen recovery as a function of the change in surface area of a polyetherimide membrane (membrane B) for a feed pressure of  $p_F = 33$  bar and permeate pressure  $p_P = 1$  bar.

out. The permeate obtained from the first membrane stage, received at an ambient pressure  $p_{P(1st)} = 1$  bar, was selected for analysis on the second membrane stage, for which the recovery degree  $R_{H_2(1st)} = 0.99$ . This recovery degree was obtained for a membrane area of  $A_{(1st)} = 13,590$   $m^2$ , while the purity of the gas permeating the membrane was then equal to  $Y_{H_2} = 0.5207$ . The results of the effect of changing the permeate pressure and membrane area on the second stage of the system are shown in Fig. 8.

Considering the characteristics shown in Fig. 8, it can be seen that for an appropriately selected membrane area and pressure prevailing on the permeate side, very high purity of hydrogen can be obtained, with purity values corresponding to the order of 0.96 with a total hydrogen recovery rate ranging from 0.45 to 0.85. For a hydrogen

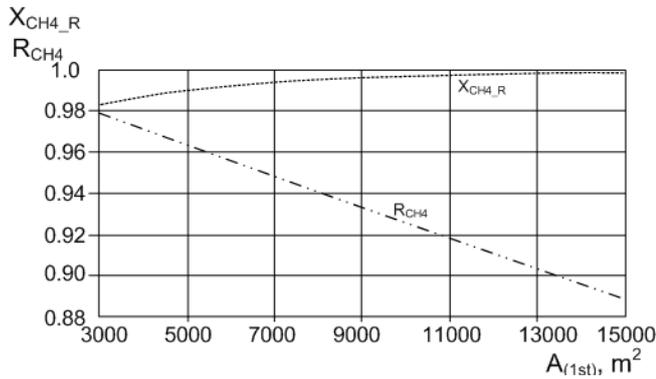


Fig. 7. Purity and recovery rate of methane in the retentate stream as a function of the change in surface area of the polyetherimide membrane (membrane B) for a feed pressure of  $p_f = 33$  bar and permeate pressure  $p_p = 1$  bar.

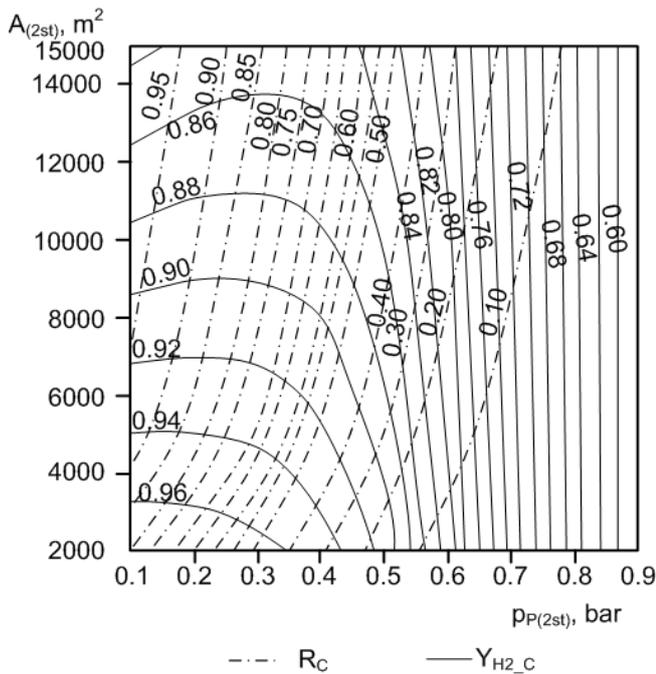


Fig. 8. Purity and total hydrogen recovery as a function of vacuum and area for a system with membrane B on the first separation stage and a polyimide membrane (membrane A) on the second stage of the membrane module.

purity of 0.9, the recovery rate ranges from 0.15 to 0.95 depending on the pressure and membrane area.

To determine the energy intensity of such a process, the power demand was determined for a separation system consisting of two membrane stages, for which the driving force of the process was the overpressure of the gas stream feeding the membrane module of 33 bar and the suction pressure induced by a vacuum pump located on the permeate side downstream of the second membrane stage.

When determining the energy consumption of the hydrogen separation process, the power demand of the compressor was neglected, as the hydrogen was recovered

from the gas network where the gas transfer pressure of 33 bar was ensured. An internal vacuum pump efficiency of 0.85 and a mechanical efficiency of 0.98 was assumed for the calculations, which were determined from the equation:

$$E_{sepH_2} = \frac{N_{elVP}}{\dot{m}_{PH_2}}, \text{ kJ/kg}_{H_2} \quad (3)$$

where  $N_{elVP}$  is the vacuum pump power requirement, kW;  $\dot{m}_{PH_2}$  is the hydrogen stream in the permeate, kg/s.

The results of the vacuum pump power demand are shown in Fig. 9, while Fig. 10 illustrates the energy consumption of such a process for 1 kg of hydrogen separated, for two selected membrane surfaces on the second stage.

As shown in Figs. 9 and 10, both the electricity demand to run the vacuum pump and the energy consumption of the process increase with a change of pressure on the permeate side. For different membrane surfaces, the power requirement curves for a low vacuum (up to about 0.7 bar) have a very close approximation of about 20 W, but beyond this vacuum value, the curves have a close approximation and define a certain power requirement area for the separation process. This power varies from a value of about 20 W to a value of about 1 kW. However, for the energy consumption of the process, the vacuum limit is a pressure of 0.5 bar, where the amount of energy needed to separate 1 kg of hydrogen is not significantly affected by the membrane surface area. The energy intensity of the separation process varies from 180 kJ/kg<sub>H<sub>2</sub></sub> to about 1,900 kJ/kg<sub>H<sub>2</sub></sub> for the maximum suction pressure induced by the vacuum pump.

#### 4. Conclusion

This paper shows the possibility of using membrane systems to separate hydrogen from its mixture with natural gas. Such systems could be used to recover hydrogen after its transport through natural gas networks. Investigated systems are characterized by a large specific membrane area with a relatively small module size. In addition, such systems are simple to operate, characterized by uncomplicated service, and are less energy consuming than other methods of separating hydrogen from gas mixtures, such as PSA technology. This is due to the fact that the driving force of the separation process is the overpressure of the gas entering the separation system after its transport and the lack of phase change of the agent – the agent remains in the gas phase from the beginning of the process. Properly selected configuration of the membrane system and membranes will allow achieving a high degree of purity of the separated medium and a high degree of hydrogen recovery.

For a polyimide polymeric membrane with an ideal selectivity coefficient of hydrogen to methane of 112.4 for a single-stage system, it is possible to recover more than 90% of the hydrogen present in the stream feeding the membrane module with a purity not exceeding 0.85. For the polyetherimide B membrane with an ideal selectivity coefficient of 222.86, we can achieve a hydrogen purity of 0.9 over the entire range of permeate pressure variation, while the hydrogen recovery ranges from 0.6 to 0.85 at this assumed H<sub>2</sub> purity.

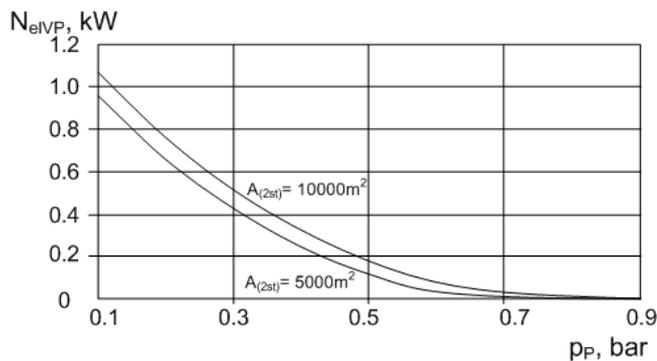


Fig. 9. Vacuum pump power demand at the second separation stage for two selected polyimide membrane surfaces area at the second stage.

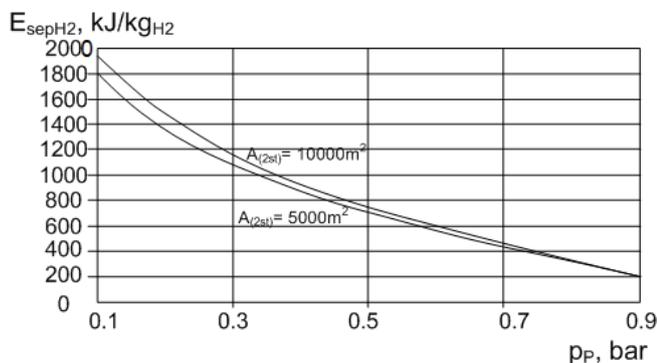


Fig. 10. Energy consumption of the separation process in relation to 1 kg of hydrogen separated for two selected polyimide membrane surfaces area at the second stage.

For two-stage systems consisting of polyimide membranes only, for a properly chosen membrane surface and permeate pressure, a hydrogen purity of less than 0.96 can be obtained with a total hydrogen recovery rate of 90%. With a polyetherimide membrane, the permeate purity can be as high as 0.99 at a total hydrogen recovery rate of less than 80%.

For the two-stage system, in which the permeate obtained from the first stage with a polyetherimide membrane was directed to the second stage of the module, where the polyimide membrane was located, the hydrogen purity of 0.96 was obtained at a total hydrogen recovery rate ranging from 0.45 to 0.85. For a hydrogen purity of 0.9, the recovery rate ranges from 0.15 to 0.95 depending on the pressure and membrane surface area.

The energy consumption of the hydrogen separation process varies from 180 kJ to about 1,900 kJ/kg of hydrogen separated for the maximum suction pressure induced by the vacuum pump.

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### Symbols

$Y_i$	—	Purity of the permeate for $i$ -th component
$X_i$	—	Purity of the feed for $i$ -th component
$n_p$	—	Permeate flow, kmol/h
$n_F$	—	Feed flow, kmol/h
$p_F$	—	Feed pressure, bar
$p_P$	—	Permeate pressure, bar
$R$	—	Recovery ratio
$P_i^*$	—	Permeability of an $i$ -th component, Barrer, (m <sup>3</sup> (STP)/(m h bar)
$\alpha$	—	Real selectivity factor
$\alpha^*$	—	Ideal selectivity factor

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