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MEMBRANE INSTALLATION FOR BIOGAS ENRICHMENT – FIELD TESTS AND SYSTEM SIMULATION

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The paper presents results of the field tests on membrane biogas enrichment performed with the application of mobile membrane installation (MMI) with the feed stream up to 10 Nm^3/h. The mobile installation equipped with four hollow fibre modules with polyimide type membranes was tested at four different biogas plants. Two of them were using agricultural substrates. The third one was constructed at a municipal wastewater plant and sludge was fermented in a digester and finally in the fourth case biogas was extracted from municipal waste landfill site. Differences in the concentration of bio-methane in feed in all cases were observed and trace compounds were detected as well. High selectivity polyimide membranes, in proper module arrangements, can provide a product of high methane content in all cases. The content of other trace compounds, such as hydrogen sulphide, water vapour and oxygen on the product did not exceed the values stated by standard for a biogas as a vehicle fuel. The traces of hydrogen sulphide and water vapour penetrated faster to the waste stream enriched in carbon dioxide, which could lead to further purification of the product – methane being hold in the retentate ($H_2O > H_2S > CO_2 > O_2 > CH_4 > N_2$). In the investigated cases, when concentration of N_2 was low and concentration of CH_4 higher than 50%, it was possible to upgrade methane to concentration above 90% in a two-stage cascade.

To perform simulation of CH_4 and CO_2 permeation through polyimide membrane, MATLAB was used. Simulation program has included permeation gaseous mixture with methane contents as observed at field tests in the range of 50 and 60% vol. The mass transport process was estimated for a concurrent hollow fibre membrane module for given pressure and temperature conditions and different values of stage cut. The obtained results show good agreement with the experimental data. The highest degree of methane recovery was obtained with gas concentrating in a cascade with recycling of the retentate.

Keywords: polymer membranes, biogas, methane concentration upgrading, trace biogas components, membrane mobile testing unit, multistage membrane system simulation

1. INTRODUCTION

Biomethane may be applied as a fuel for heat and electricity generation or after compression as a fuel for automobiles. The state of the art of biogas upgrading technologies with upgrading efficiency, methane loss, environmental effect, development and commercialization, and challenges in terms of energy consumption and economic assessment have been reviewed by Khan et al. (2017). However the biogas sector development has to be advanced by national laws and regulations (Stürmer, 2017). According to Miltner et al. (2017),

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generally it is expected that a substantial amount of existing biogas plants with expiring or not cost-covering green electricity feed-in tariffs will have to switch to this alternative valorisation route in the near future in order to increase the profitability of the plant investment. Many different technologies have been employed to obtain biomethane from biogas. Methods like water scrubbing or pressure swing adsorption are commonly used and can be declared as well established. Membrane gas permeation found its place among the biogas upgrading methods some years ago (Karszova et al., 2015). Membrane-based techniques are considered as capable and economic methods for methane concentration upgrading to meet requirements of further applications (energy carrier for pipeline distribution, chemical substrate, CNG, etc.). Different polymers can be applied for gas permeable membranes manufacturing for example; cellulose acetate (CA), polyamide (PI) and polyaryl-ether-ketone-ketone (PEKK). However, according to Micale (2015), membranes made of CA or PI are more efficient in comparison with PEKK for all pressures applied in this research. As a membrane material commercially available polyimide type polymer was used in this work. Current membrane carbon dioxide separation installations utilise a very limited number of materials, of which cellulose acetate (Baker and Lokhandwala, 2008), polycarbonate, polysulfone (Aroon et al., 2010) and just polyimide (Chmielewski et al., 2013; Harasimowicz et al., 2007; Polak and Chmielewski, 2010) are probably the most common. Polyimides have very good permselectivity in comparison with other polymers used in separation of CO_2. Moreover, this kind of polymer is characterised by good mechanical properties and thermal/chemical stability. They are commonly prepared by two groups of compounds: a diamine and an anhydride. A large amount of available types of both groups is contributing to a wide range of polyimides with great selective properties. This explains the fact that polyimides comprise one of the most studied types of polymers for membrane separation of CO_2/CH_4. In particular, polyimides have robust mechanical properties to withstand high-pressure process. They are characterised by long durability and high chemical stability. Polyimides are less susceptible to plasticise by carbon dioxide than CA. Therefore, they are supposed to be a suitable material for CO_2 separation. At present, polyimide membranes are commercialised by a few manufactures: Medal (Air Liquide), IMS (Praxair), and UBE Industries Ltd.

In general, permeability of a polymer for a gas mixture increases with decreasing its molecular size (Fig. 1), and increasing solubility (or condensability) of the gas. For biogas the relative permeability of its components is given in order of decreasing permeability as: $H_2 > CO_2 > O_2 > CH_4 > N_2$.

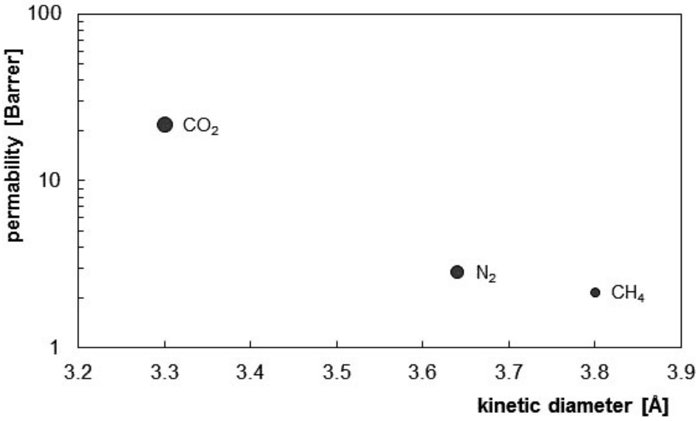

Fig. 1. Gas permeability via polyimide membrane vs molecular diameter (P = 0.6 MPa, T = 293.15 K); 1 Barrer = 10^{-10}(cm^3@STP · cm)/(cm^2· s · cm-Hg)

In particular PI and CA performance is quite similar for compression pressure of 15 and 20 bar, except for the case of 10 bar PI that turned out to be the best solution. The same conclusions were drawn in our laboratory works (Harasimowicz et al., 2007) and pilot plant used for biomethane upgrading at agriculture waste biogas plant (Chmielewski et al., 2013). Improvements in polymer synthesis, performance, membrane structure, module fabrication, and process design have contributed significantly to increase the potential range of membrane applications. Among them polyimide based membranes have great potential to be used widely in gas separation processes, especially in CO_2 separations (Favvas et al., 2017). UBE polyimide membranes in the form of hollow fibre modules were tested by Jaschik et al. (2018), for separation of $CO_2/N_2/O_2$ mixtures. The high pressure module used in these studies was designed to operate at 2.6–7.2 bar pressure range. In the case of biogas upgrading in methane single stage processes cannot provide both high product gas purity and high recovery at the same time but multistage processes can. Commonly, the design of multi stage gas permeation processes relies on heuristics and experience, so that often sub-optimal separation processes are designed (Chmielewski et al., 2013; Scholtz et al., 2015). Regarding membrane module setup, it was found that the two-stage cascade process with recycle using a polyimide membrane was not economically viable for biogas upgrading due to high recycle ratio, and thus resulting in high operating cost, whereas the three stage polyimide membrane system is quite feasible in order to obtain fuel quality of biomethane (Haider et al., 2016). However a more advanced optimisation technique such as artificial neural network that can describe a more complex phenomenon will gain momentum in the future for methane enrichment process in order to make the most effective exploitation of the biogas resource (Seman et al., 2019). New technologies may broaden the use of the biogas, for example producing renewable natural gas and compressed renewable natural gas used as vehicle fuels (Wang et al., 2011). The other possibility is application of hybrid systems using combination with absorption process (Harasimowicz et al., 2004). However, at the design stage of membrane installation, one should consider the fact that the composition of a biogas depends strongly on the organic substrate and the digestion conditions (Valenti et al., 2013). The present paper reports results of membrane installation field tests performed at biogas plants using different technologies and substrates: municipal wastewater treatment plant biogas digester using bio sludge, two agricultural biogas plants and municipal solid waste landfill site (vertical perforated piping gas collectors). There are differences in biogas production rate and its composition depending of the source as reported previously in the papers regarding biogas municipal sludge (Weiss et al., 2009), agricultural waste and silage (Luo et al., 2016), and landfill (Karapidakis, 2010). The variety of substrates used and process engineering solutions and process parameter existing in a single country, given for China, is well presented by Wang et al. (2011). Anaerobic digestions of organic solid wastes studied have shown to be a renewable energy source that can generate biogas with high methane content. Beside usage of different substrates the type of applied technological solution for anaerobic digestion processes like, conventional batch, single-stage, or two-stage may affect gas composition as well (Castellano-Hinojosa et al., 2018; Nasir et al., 2012). The research reported in this work was performed at biogas plants using different technologies. Beside testing of methane enrichment at biogas plants using fermenter tanks the test were performed for substrate received from landfill municipal waste biogas drainage collector. Comparison of the results of the field tests on the biogas enrichment in methane achieved at the same membrane system performed at biogas plants using different substrates and different technologies have not been reported until now. Therefore results of this work have great value for membrane methane enrichment technology up scaling studies and industrial stationary installations design. The process may be combined with methane storage systems (Feroldi et al., 2016).

2. EXPERIMENTS

2.1. Mobile Membrane Installation (MMI)

As a result of literature studies and many years of experience in the gas separation methods, a membrane separation technique for biogas enrichment in methane using a UBE membrane module has been selected for this study. As a basic element of the Mobile Membrane Installation (MMI) a hollow fibre module was used (Fig. 2).

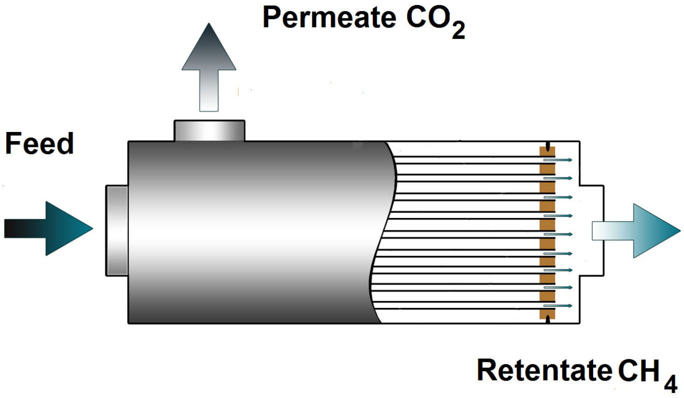

Fig. 2. Hollow-fibre module applied in the MMI construction

The enrichment ratio in such a module depends on several factors, including biogas composition, capillary length, pressure applied and ratio between permeate and retentate streams.

A scheme of the biogas two-stage membrane enrichment system is presented in Fig. 3 and its photo in Fig. 4.

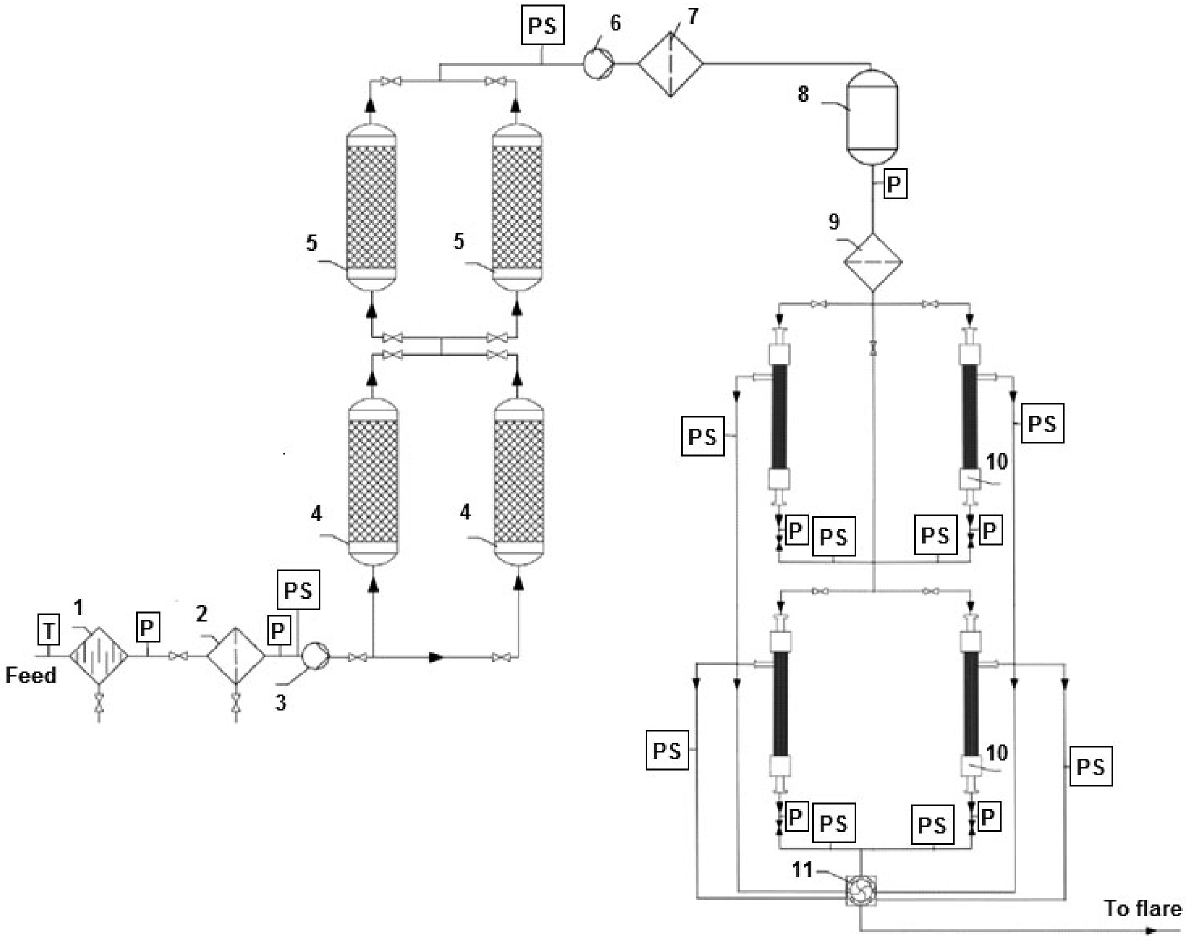

Fig. 3. Scheme of two stage membrane biogas enrichment plant: F1 – mechanical filter (Filtron AM411), 2 – dehumidifier (mechanical), 3 – fan, 4 – silica sorbents, 5 – H_2S adsorber, 6 – compressor, 7 – oil separator (mechanical), 8 – buffer tank, 9 – fine filter oil separator, 10 – hollow fibre membrane filters, 11 – processed gas stream mixer. Computerized measurement system: T – temperature, P – pressure, PS – gas composition measurements system (MSMR16)

Fig. 4. Photo of the setup at field experiment at biogas installation at municipal wastewater treatment plant. General view of MMI installed on mobile deck (left), fabric raw biogas tank, MIM and biogas flare (right). The municipal wastewater sludge biogas plant testing case

The MMI is equipped with a gas purification system, including unit section connected in the series. Firstly, solid particulate and oily aerosols are removed in a mechanical filter with FILTRON® filter cartridge. It is a typical filter material used in the automotive applications. Then, in the next step a trap is installed in which water droplets are removed from the gas stream. Final draying of the gas takes place in two vertical, cylindrical adsorption columns filled with silica gel granules (Ø grains ca. 5 mm). The moisture adsorption is up to 60% at high relative humidity (90%) of the inlet gas. Application of this stage is very important for biogas, which is usually saturated with water vapour, for further processing. The biogas dried in the adsorption column feeds a set of two desulphurisation columns in which H_2S adsorption takes place on active carbon. The other micro pollutants of biogas are adsorbed with high efficiency as well.

Then, the biogas compressed by the compressor feeds the battery section of the membrane modules. In the high-pressure part (max. 10 bar) oil aerosol separators detaining oily substances are installed. The rust and solid particles, posing a threat to the delicate surface of polymer capillaries, are stopped here too. According to the membrane manufacturer's requirements, the gas supplying the modules should be free of liquid water droplets (max. humidity 80% RH), oil aerosol above 0.01 mg/Nm^3, the hydrocarbon concentration should not exceed 0.013 mg/Nm^3. Also the number of small solid particles of diameter smaller than 0.01 µm should be limited. All mentioned contaminants limit the lifetime of the module. The membrane section contains four hollow fibre polyimide modules from UBE. The dense gas polymer separation layer is located outside the capillaries, which significantly increases the active mass transfer surface area and assures low gas pressure drop. The risk of film damage by solid particles in the module feed stream is also limited. Four modules were installed in the installation – connected in series in pairs, enabling the separation process to be carried out in proper configuration.

The mobile membrane installation has been tested at different biogas plants using different substrates as a feed. Tested arrangement of the modules is presented in Fig. 5.

The biogas enrichment tests were always preceded by a leak test of the installation. Technical nitrogen ordered from local suppliers of technical gases was used for this purpose. After verifying the tightness of the system the installation was connected to the raw biogas supplying pipe. The nitrogen installation filling was also intended to remove air from the system which creates an explosive mixture with methane.

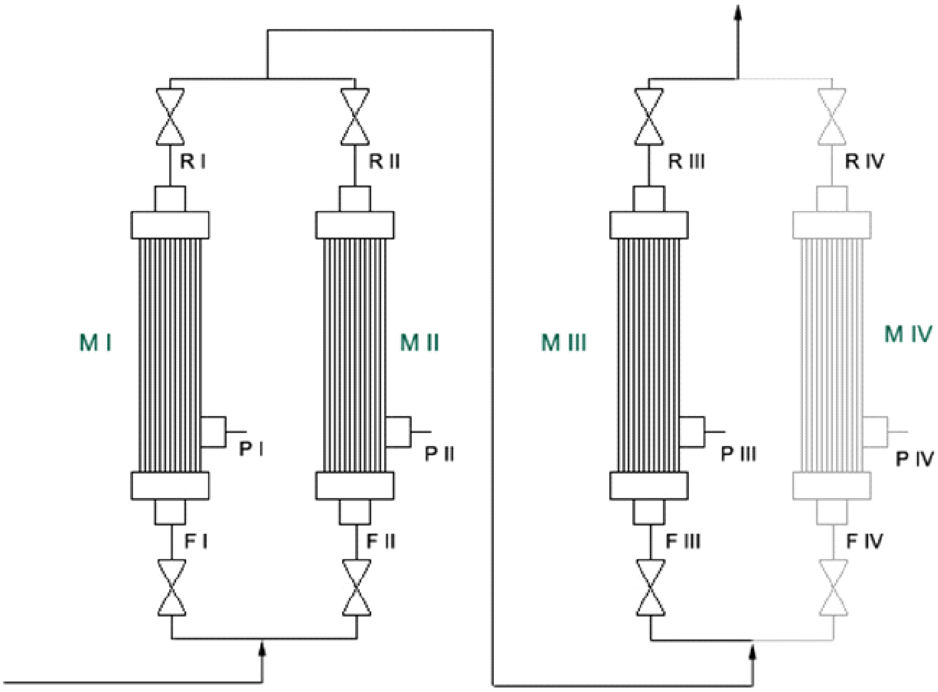

Fig. 5. Scheme of the testing rig composed of two stages; two modules in the first stage and one in the second; symbols M I – M IV mean hollow fibre membrane modules

2.2. Analytical methods

The system was equipped with thermometers, flow meters and pressure gauges. Concentration of gases in the inlet and outlet streams was measured using DP 27 Bio Nanonsens analysers (CH_4, CO_2, O_2, H_2S) (accuracy CH_4 ± 1%; CO_2 ± 1%; O_2 ± 2% in the range; H_2S ±1%) and Gas Hunter Alter analyser and GA45 Plus Geotechnical Instruments (CH_4 and CO_2 accuracy ±0.5%; RH ±0.2%; t ±0.15 °C; p ±0.5%) were used additionally to measure humidity, pressure and gas temperature. Selected gas samples were also analysed by chromatographic methods. Sampling and measurements of biogas composition were performed by certified laboratory according to EN ISO 10715:2005 by means of a glass pipette sampling and sorbents.

The trace components in the biogas were measured as well. Sampling and measurements of biogas composition were performed by certified laboratory of Oil and Gas Institute (Cracow, Poland) according to EN ISO 10715:2005 using a glass pipette and sorbents while mercury sampling according to EN ISO 6978-2:2007. Other analytical methods are described by EN ISO 6974-5:2006. The methods cover GC – TCD/FID/FPD. IC. Single-Purpose Atomic Absorption Spectrometer AMA-25 (mercury), thermo-hygrometry (water).

2.3. Agricultural biogas plant

Biogas from two different plants using agro products has been tested. Both of them are localized in East Poland. The first one is based on NAWARO$^{©}$ (NAWARO 2019) technology and has nominal capacity 1.27 MW_e. The substrates used are maize, whey and grass silage, distillery whole silage and potato pulp. The second one uses original two stage Polish technology (Kryłowicz et al. 2008) of 1.2 MW_e nominal power. This solution is employing hydraulic mixing (Chmielewski et al. 2012). Maize silage was a main substrate used during biogas composition testing.

2.4. Wastewater treatment plant biogas installation

The municipal wastewater plant is localized in Central Poland. Nominal flow of treated wastewater is equal to 13.500 m^3 per day. Anaerobic digestion of sewage sludge has long been used for solids reduction by wastewater treatment facilities, but has gained recognition as a form of energy production. Biogas is formed as a by-product of anaerobic digestion in separate fermenters.

2.5. Municipal landfill biogas recovery installation

Municipal waste (mainly organic) landfill field is localized in the North-Eastern Poland. Landfill gas consists of methane as a main component (ca. 50%). 82 biogas recovery wells were constructed at this field, 38 m high and occupying territory of ca. 14 ha. 1997 was the beginning of the landfill gas deposit extraction and the nominal working parameters were reached in 2001. The recovery of the gas is equal to 460 Nm^3/h and generators of nominal power 1.24 kW_e each are supplying 1.25 MW_e and 990 kW_t.

3. RESULTS OF FIELD TESTS AND THEIR DICUSSION

The average composition of the raw biogas (CO_2, CH_4, N_2, O_2, H_2S – before desulfurization, water content) measured during MIM operation at tested sites is given in Table 1. Caloric value of raw biogas was calculated as well for all cases.

Table 1. The average gas composition at the investigated site during the MMI testing

Compound	Unit	Municipal sludge	Agricultural Plant 1	Agricultural Plant 2	Landfill
CO_2	% mol/mol	30.93	44.03	47.45	31.87
CH_4	% mol/mol	65.82	50.18	52.19	50.55
N_2	% mol/mol	2.56	5.49	0.28	16.72
O_2	% mol/mol	0.69	0.29	0.08	0.86
H_2S before desulphurisation	mg/m^3	> 560	381.4	470.5	n.t.
Water	g/m^3	10.1	27.3	12.5	9.7
Density	kg/m^3	1.12	1.30	1.32	1.21
Caloric value	MJ/m^3	23.65	18.04	18.77	18.16

n.t. – not tested

To measured content of micro gas components, analysis was made using grab sampling method in accordance to a procedure described earlier and samples collected during the tests were tested by accredited laboratory. The results of analysis are given in Table 2.

MIM biogas experiments lasting a week were performed at each site defined above. From gases listed in Table 1 the highest boiling point has water, therefore in front of membrane unit a dryer has to be applied. A simple condensation unit in the first stage and additional silica gel column were used in our case. This arrangement was described at the section related to the MIM construction. The results of biogas enrichment in methane, based on line gas composition monitoring, are presented in Table 3.

Table 2. Trace compounds in biogas from different sources/substrates (normal gas conditions)

Compound	Unit	Municipal sludge	Agricultural Plant 1	Agricultural Plant 2	Landfill
CO_2	%mol/mol	30.9314	44.0325	47.45	31.865
CH_4	%mol/mol	65.8248	50.1805	52.1892	50.5546
N_2	%mol/mol	2.5564	5.4961	0.2795	16.7216
O_2	%mol/mol	0.6874	0.2908	0.0813	0.8582
CO	%mol/mol	n.d.	n.d.	n.d.	n.d.
H_2	%mol/mol	n.d.	n.d.	n.d.	n.d.
Benzene	mg/m^3	0.07	0.03	0.95	0.09
Toluene	mg/m^3	3.69	0.65	0.46	3.72
Ethylbenzene+xylenes	mg/m^3	26.26	4.96	7.39	76.88
H_2S before desulphurisation	mg/m^3	> 560	381.4	470.5	n.t.
H_2S after desulphurisation	mg/m^3	123	6.5	n.t.	42.3
COS	mg/m^3	n.d.	n.d.	n.d.	n.d.
Mercaptans	mg/m^3	n.d.	n.d.	n.d.	n.d.
CS_2	mg/m^3	n.d.	n.d.	n.d.	n.d.
Total Cl	mg/m^3	6.54	7.29	7.63	8.06
Total F	mg/m^3	0.2	0.07	0.19	0.39
Hg	ng/m^3	8.1	9.24	5.82	6.46
Anthracene	µg/m^3	n.d.	n.d.	n.d.	n.d.
Fluorene	µg/m^3	n.d.	n.d.	n.d.	n.d.
Naphtalene	µg/m^3	3.1	0.6	7.4	9.7
Hexamethyldisiloxane	mg/m^3	n.d.	n.d.	n.d.	n.d.
Hexamethylcyklotrisiloxane	mg/m^3	2.34.	n.d.	0.76	4.04
Oktamethyltrisiloxane	mg/m^3	0.08	n.d.	n.d.	n.d.
Oktamethylcyklotetrasiloxane	mg/m^3	4.11	0.94	1.76	17.48
Decamethylcyklopentasiloxane	mg/m^3	2.02	0.04	0.22	1.61
Si total	mg/m^3	3.28	0.37	1.04	8.96
Sum of siloxanes	mg/m^3	8.68	0.98	2.74	23.71
Water	g/m^3	10.1	27.3	12.5	9.7
Heat of combustion	MJ/m^3	26.25	20.02	20.82	20.15
Caloric value	MJ/m^3	23.65	18.04	18.77	18.16
Wobbe Index	MJ/m^3	28.15	19.95	20.65	20.81
Density	kg/m^3	1.12	1.3016	1.3154	1.2124
Relative density	[–]	0.8694	1.0067	1.0174	0.9377

n.d. – not detected. n.t. – not tested

Table 3. The methane upgrading ratio at the investigated site during the MMI testing

FEED	Unit	Municipal sludge	Agricultural Plant 1	Agricultural Plant 2	Landfill
CO_2	% mol/mol	30.93	44.03	47.45	31.87
CH_4	% mol/mol	65.82	50.18	52.19	50.55
N_2	% mol/mol	2.56	5.49	0.28	16.72
O_2	% mol/mol	0.69	0.29	0.08	0.86
H_2S before desulphurisation	mg/m^3	> 560	381.4	470.5	n.t.
PRODUCT					
CO_2	% mol/mol	0.58	0.00	4.43	4.13
CH_4	% mol/mol	93.74	74.00	95.16	73.92
N_2	% mol/mol	5.62	25.77	0.07	21.76
O_2	% mol/mol	–	–	–	–
H_2S final	mg/m^3	21.03	78.43	118.07	63.65

n.t. – not tested

Remarkable differences regarding concentration of different compounds in biogas generated in the plants using different substrates and being operated in different conditions are observed (Table 1). The highest concentration of biomethane was found in the case in which as a substrate sludge from municipal waste water plant was applied (above 65% v/v). In three other cases the recorded concentration of CH_4 was in the range of 50–52% v/v. High nitrogen concentration has been measured for gas from municipal landfill recovery (over 16% v/v). Gaseous product recovered from landfill wells contain beside high concentration of nitrogen, BTEX compounds (although benzene concentration is not too high in this and three other cases) and siloxanes (Table 2).

Due to highest methane concentration the biogas from municipal wastewater plant has the highest heat of combustion value above 23 MJ/m^3 while in all other cases this number is close to 18 MJ/m^3. In the first case the concentration of both, BTEX and siloxanes is much lower than in the case of landfill gas, but still above the amount observed in the case of agricultural plants, which is logical. It seems that this kind of fuel after primary purification can be used for heating purposes and after siloxanes concentration control for electricity generation as well, since cogeneration systems are the most economical. Organic silicon compounds are listed as the most unwanted compounds affecting energy utilization in biogas produced from waste materials, due to the fact that silica particles are formed in combustion process and deposited on mechanical internal parts of generator my cause wearing of movable elements. These are semi-volatile organosilicon compounds, that are used in a number of industrial applications and in consumer products such as shampoo, cosmetics and lubricants. Therefore they may be found in municipal waste landfills and sewage from wastewater plants, while they volatilise into landfill gas or digester gas in the second case. Interestingly, in all investigated cases similar trace concentrations of mercury are observed. The air pollution from coal fired power plants and domestic furnaces/boilers may be expected as the source.

The tests of methane concentration upgrading performed with use of MIM have demonstrated feasibility of membrane process application in all investigated cases. The required enrichment of gas in the methane can be achieved by membrane technique with application of a two-stage process which was demonstrated during the performed field tests if the feed meets requirements of the process. Methane concentration in biogas from Wastewater Plant was upgraded from 65.8% v/v up to 93.7% v/v and similar results were obtained for Agricultural Plant 2 where this concentration increased from 52.2% v/v up to 95.2% v/v. The lower enrichment ratio was obtained in the case of Agricultural Plant 2 (from 50.2% v/v up to 74.0% v/v) and Landfill (from 50.5% v/v up to 73.9% v/v). In both these cases high concentration of nitrogen in the feed was observed and the presence of the gas hinders methane enrichment efficiency due to the fact that permeability of this gas is the lowest ($CO_2 > CH_4 > N_2$) when taking into account other components of the mixture. Therefore nitrogen mostly retains with methane in the retentate. It is important to point out that the traces of hydrogen sulphide and water vapour penetrate faster to the waste stream enriched in carbon dioxide, which can lead to further purification of the product – methane being held in the retentate ($H_2O > H_2S > CO_2 > O_2 > CH_4 > N_2$). However, the preliminary desulphurisation and water (condensate and vapour) have to be applied in membrane technology utilization. Moreover, even the required methane enrichment ratio was obtained for good quality biogas feed. A three-stage process is recommended to avoid too high methane losses in the permeate being the waste stream. Several operating parameters affect gas separation by membrane: these include feed-gas flow and composition, pressure differential across the membrane, gas temperature, online membrane area, and the required product gas specification. Process optimisation methodology was elaborated by Valenti et al. (2013) and may be applied for full scale membrane industrial plant design.

The biogas from the plants using pure agricultural substrates is the best candidate for further methane concentration enrichment via different techniques, including tested membrane process. Some amount of siloxanes is observed (0.98 and 2.74 mg/m^3). However their concentrations in the case plants using agricultural substrates are very low in comparison with observed municipal sludge plant 8.68 mg/m^3 and landfill gas 23.71 mg/m^3) and this gas fuel is a good candidate for its upgrading to meet vehicle fuel requirements. Some differences for both plants using agricultural substrates illustrate necessity of substrate purity control in such case and avoiding admixture of waste with biomass.

4. COMPUTER METHANE ENRICHEMENT SYSTEM ARRAGEMENTS SIMULATION

Modelling membrane permeation of the gas mixture leads to better understanding of the process, verification of the results, and is also an important step in the design of industrial plants. Computer simulations accelerate scaling up of work, and by identifying the optimal configuration of modules in a multi-stage system it is possible to skip the experimental intermediate scale. Modelling using the results obtained in a lab-scale and MMI pilot tests may provide valuable information regarding the use of the same type membrane at different process conditions. By determining the area needed to obtain the desired product purity, and the achievable efficiency of the module, it is possible to evaluate investment and operating costs. There is some theoretical studies describing permeation running in the one module without recirculation of exhaust recycle streams (Al-Juaied and Koros, 2005; Wang et al., 2002), but in practice a high purity of the product or very high retention degree of important compound requires recirculation of one gaseous stream: permeate or retentate – depending on the purpose of the process. The very important factor related to the methane loses in permeate is configuration of the membrane system. Therefore, in relation to the experimental results, a computer simulation for a single and a multiple stage system has been performed. An elaborated method can be applied for industrial system design.

A few mathematical procedures describe gas permeation and optimise single or multistage membrane systems. Mathematical modelling of gas separation process was introduced by a few groups of researches. First simulation was carried out by Stern and Wang (1978). The authors used ordinary differential equations for binary mixture in one module. Simulation of membrane separation of multicomponent mixture passing through a simple module was described by Pan (1986). This procedure could be solved by a trial-and-error method and iterative system. A related method was presented by Bhide and Stern (1993). Thundyil and

Koros (1997) used a model called 'succession of states' for a finite of elements. The authors proposed this method to solve transport mechanism for co-current, counter-current and cross–flow configurations. Some part of research on gas permeation applied single differential equations of Runge–Kutta algorithms (Li et al., 1990; Tsuru et al., 1995). Other procedures for the calculation of gas separation in a single and multistage installation are based on the Gaussian-Seidl finite difference method (Makaruk and Harasek, 2009).

Many studies have been related to study membrane configuration for the natural gas purification, but similar results are applicable for biogas separation as well. The main objective of the computer simulation based on experimental input was an optimisation of the installation structure to reduce cost and increase efficiency of membrane plant performance.

4.1. Flow simulation for single module

Modelling of membrane gas separation requires an examination of the properties of gases and it also takes into account the characteristics of the membrane. Numerical program for calculating of the transport of carbon dioxide through a polyimide membrane has been elaborated by the authors. To simulate the permeation process we used Matlab 7.12. Model of concentrations and pressures distributions along the length of the membrane was determined by the method of Runge-Kutta 4th order. The calculations use permeability coefficients for different gases, determined experimentally.

In the work the following assumptions have been made:

- the pressure drop across the membrane is described by the Hagen–Poiseuille equation (Coker et al., 1998; Kaldis et al., 2000; Kim et al., 2012; Wang et al., 2002; Thundyil and Koros, 1997).
- the process takes place under isothermal conditions,
- lack of concentration polarization,
- no dispersion along the axis,
- deviation from the ideal gas disregarded in calculating properties of the gas, i.e. density and viscosity,
- resistance carrier porous layer is not included; mass transport occurs only in the active layer (Taveira et al., 2001),
- no deformation of the fibres by the action of pressure (Tsuru and Hwang, 1995),
- the co-current flow of streams occurs in the membrane.

The following equations of balance were adopted:

- In case of the co-current flow of flux passing through the membrane the flux of the flow can be represented by Eq. (1)

$$J_i = P_i(p_{i,F} - p_{i.P}) = P_i(P_F x_{i,F} - P_P x_{i.P}) \quad i = 1, 2, \ldots, n \tag{1}$$

- For a binary mixture containing CO_2 and CH_4 expressions describing the various streams of gases take the form of:

$$J_{CO_2} = P_{CO_2}(P_F x_{CO_2,F} - P_P x_{CO_2.P}) \tag{2}$$

$$J_{CH_4} = P_{CH_4}(P_F x_{CH_4,F} - P_P x_{CH_4.P}) \tag{3}$$

- The mass flow through the membrane can be expressed by using the volumetric flow rates. For the retentate stream passing through the capillary differential equation becomes:

$$\frac{dQ_R}{dz} = -\pi \cdot D_o \sum_{i=1}^{n} J_i \tag{4}$$

- For a permeate stream:

$$\frac{dQ_P}{dz} = \pi \cdot D_o \sum_{i=1}^{n} J_i \tag{5}$$

- The distribution of concentrations of the individual components is described by the following equations:

$$Q_R \frac{dx_{i,F}}{dz} = -\pi \cdot D_o \left(J_i - x \sum_{j=1}^{n} J_j \right) \quad i = 1, 2, \ldots, n-1 \tag{6}$$

$$Q_P \frac{dx_{i,P}}{dz} = \pi \cdot D_o \left(J_i - x \sum_{j=1}^{n} J_j \right) \quad i = 1, 2, \ldots, n-1 \tag{7}$$

- The following boundary conditions have been adopted:

$$z = 0, \quad Q_R = Q_F, \quad Q_P = 0, \quad x_{i,R} = x_{i,F}, \quad x_{i,P} = x_i^*, \quad P = P_F \tag{8}$$

  In this equation:

$$x_{i,}^* = \frac{P_i \cdot x_{i,F}}{\sum\limits_{j=1}^{n} P_j \cdot x_j}, \quad i = 1, 2, \ldots, n-1 \tag{9}$$

- For CH_4 concentration changes above equation can be described as follows:

$$x_{CH_4}^* = \frac{P_{CH_4} \cdot x_{CH_4,F}}{P_{CH_4} \cdot x_{CH_4,F} + P_{CO_2} \cdot x_{CO_2,F}} \tag{10}$$

- For a binary mixture of CO_2-CH_4 equations take the following form:

$$\frac{dQ_R}{dz} = -\pi \cdot D_o \cdot (J_{CH_4} + J_{CO_2}) \tag{11}$$

$$\frac{dQ_P}{dz} = \pi \cdot D_o \cdot (J_{CH_4} + J_{CO_2}) \tag{12}$$

- The gradient of the concentration of CH_4 in the retentate stream can be described by the following equation:

$$\frac{dx_{R,CH_4}}{dz} = -\frac{\pi \cdot D_o}{Q_R} \left(J_{CH_4} - x_{R,CH_4} \cdot (J_{CH_4} + J_{CO_2}) \right) \tag{13}$$

- Equation describing gradient of CH_4 concentration in the permeate stream takes the following form:

$$\frac{dx_{P,CH_4}}{dz} = \frac{\pi \cdot D_o}{Q_P} \left(J_{CH_4} - x_{P,CH_4} \cdot (J_{CH_4} + J_{CO_2}) \right) \tag{14}$$

The above equations allow determining the concentration gradients in permeate and retentate streams along the membrane. Thereby it is possible to calculate the degree of retention of the desired product as well. Based on the above equations, the required membrane area to achieve a specified concentration of methane in the retentate was also calculated. In the balance equations experimentally determined permeation coefficients were employed.

Examples of computer simulations are presented in graphical form. Model predictions confirm the actual course of methane enrichment using the module with polyimide hollow fibre membrane very well. Compatibility of modelled permeation profiles with experimental data was determined by designating the correlation coefficients.

Figures 6–8 demonstrate a permeation process of a mixture model composed of 50 ± 2% vol. CH_4 and 50 ± 2% vol. CO_2. The process was conducted under isothermal conditions at a pressure in the range from 0.4 to 0.6 MPa.

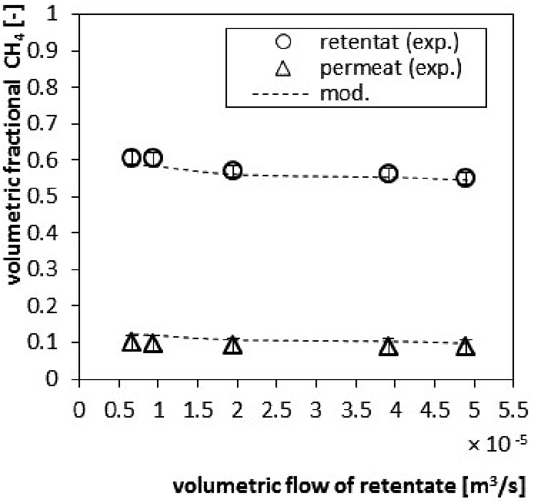

Fig. 6. Methane content in both streams as a function of retentate volumetric flow [P = 0.4 MPa, T = 298 K]

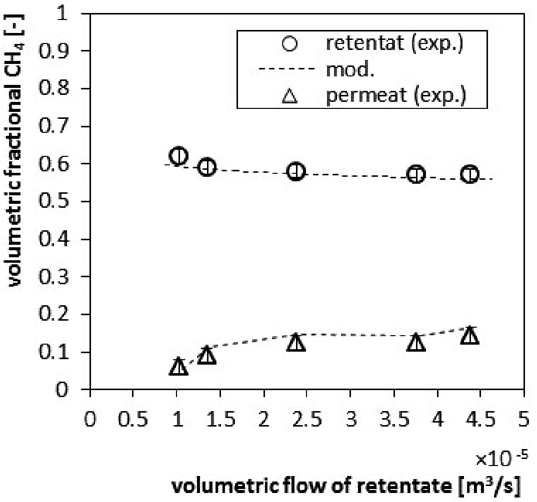

Fig. 7. Methane content in both streams as a function of retentate volumetric flow [P = 0.5 MPa, T = 298 K]

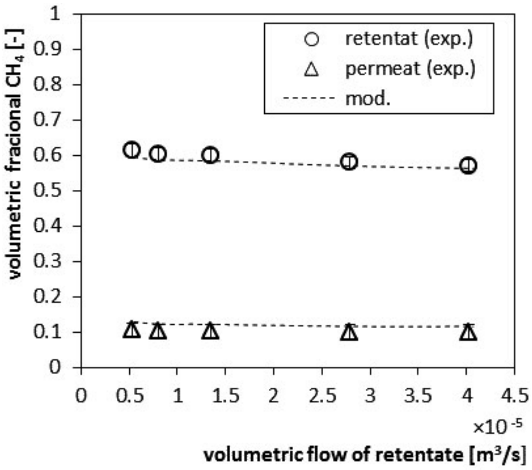

Fig. 8. Methane content in both streams as a function of retentate volumetric flow [P = 0.6 MPa, T = 298 K]

Analogous simulations were performed for a mixture containing 60 ± 2% vol. CH_4 and 40 ± 2% vol. CO_2. Figs. 8–11 show the concentration of methane in the two streams leaving the unit as a function of the volumetric flow of the retentate. Permeation process was carried out at constant temperature, at the pressure range from 0.4 MPa to 0.6 MPa.

Good convergence of simulation results with experimental data has been obtained, although better fitting was achieved for the retentate stream. An indicator of the convergence of model predictions and experimental data are correlation coefficients determined based on the concentration of methane in the exhaust stream. The values of the correlation coefficients for each simulation are shown in Table 4.

Table 4. Correlation coefficient estimates for methane concentration in the retentate stream

Process pressure [MPa]	0.4	0.5	0.6
		Correlation coefficients	
Mixture of 50% CH_4 + 50% CO_2	0.9995	0.9995	0.9997
Mixture of 60% CH_4 + 40% CO_2	0.9985	0.9844	0.9411

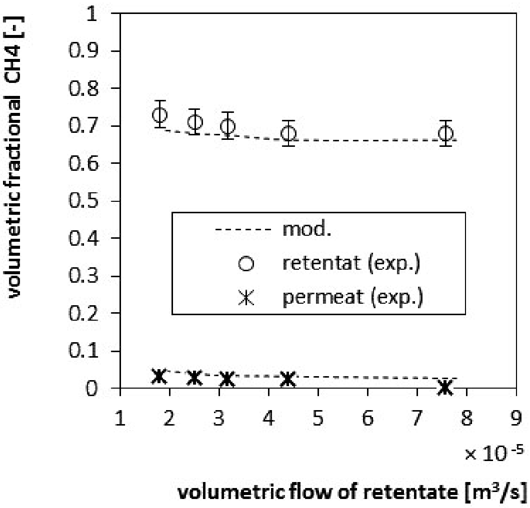

Fig. 9. Methane content in both streams as a function of retentate volumetric flow [P = 0.4 MPa, T = 298 K]

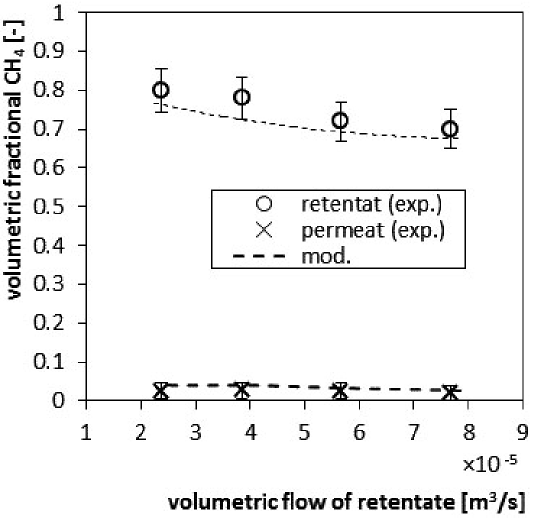

Fig. 10. Methane content in both streams as a function of retentate volumetric flow [P = 0.5 MPa, T = 298 K]

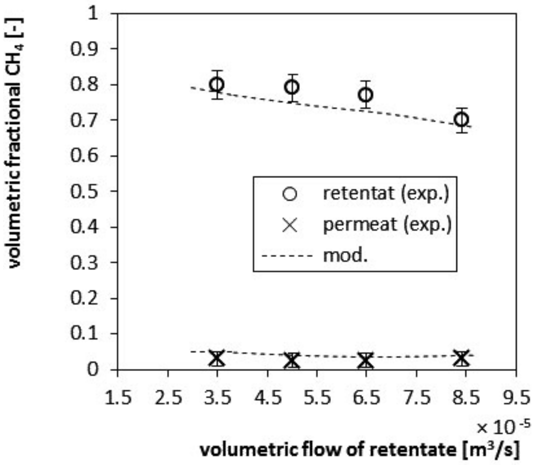

Fig. 11. Methane content in both streams as a function of retentate volumetric flow [P = 0.6 MPa, T = 298 K]

Model equations were used to verify the results obtained at the laboratory stand for CO_2/CH_4 mixtures with different configurations. The program was also used to the simulations and calculations cascades multistage modelling systems.

4.2. Simulation for multistage process

In order to obtain high product purity, it is often necessary to use multi-stage installation. The total number of modules to be applied in the system is dependent on the flow of the biogas and the performance of a single module. Each separator is characterised by its own specific range of optimal performance. Operation below the lower limits of feed flow or below the respective pressures significantly reduces the effects of separation. However, exceeding the upper limits of pressure may cause damage of the module. Parallel connection of the modules allows obtaining the required volumetric flow and there is no risk to exceed the optimum operation of a single module. Targeting a summary of all retentate streams from the first stage to the next allows reaching the required purity of the product. The use of identical modules reduces the number of modules at the following stages, thereby creating so-called Christmas tree's structure which comes from separation cascade theory.

If the membrane selectivity is not good enough or if the required process efficiency cannot be achieved, cascade installation has to be used. Single stage system with recirculation of one of the streams fits cascade theory as well. Single stage layout plants are characterised by simplicity, low power consumption and low demand area, but relatively low enrichment is reason why obtained low caloric product cannot be applied as a substitute of natural gas. Moreover, if the used module is characterised by unsatisfactory selectivity for gas components carbon dioxide and methane. The loss of valuable component in the permeate stream may be significant. The solution may be attained by recycling of the permeate stream (Fig. 12), but it will increase the cost of higher energy requirement to run a system.

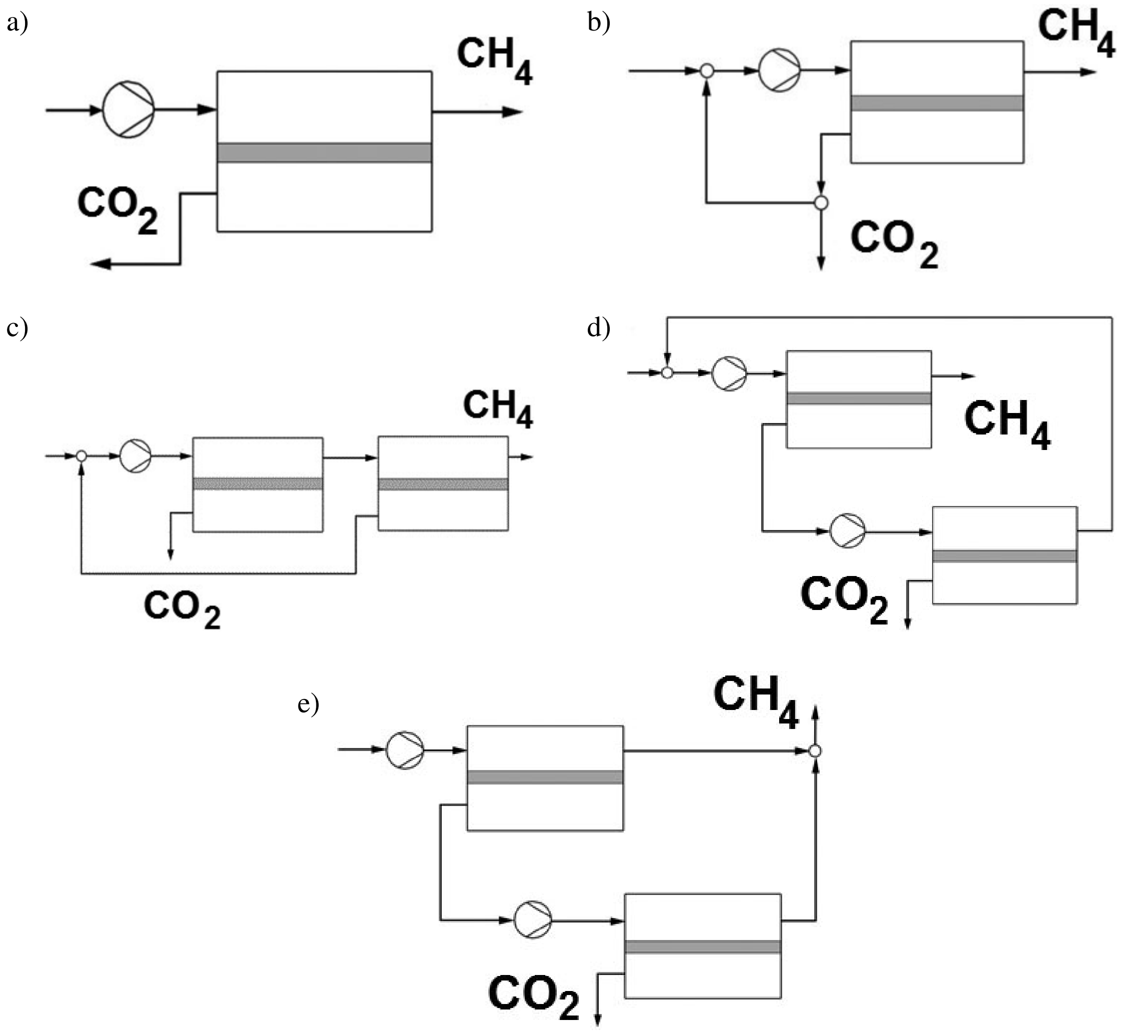

Fig. 12. Different arrangement of membrane cascade installation for separation of CO_2 from feed gas

Methane enrichment in the next stage retentate stream is achieved although at the same time additional stream of permeate containing some methane is produced, what leads to this combustible component losses. The problem of loss reduction can be solved using the recirculation of the permeate stream of the second stage to the first module (Fig. 12c). When first module permeate has high enough concentration of methane, it is proposed to introduce it to the next step. This solution is used in situations where the retentate of the first stage is characterised by high enrichment in methane and there is no need for its further enrichment.

The retentate of the second stage may be also mixed with the raw biogas which feeds the first stage (Fig. 12d) or can be mixed with the first block's retentate (Fig. 12e). Examples of computer simulations of the installation can be found in the literature (Lababidi et al., 1996; Pettersen and Lien, 1995; Qiu et al., 1989; Watanabe, 1999). Summary of significant parameters characterising single and two stage systems for a given product purity of 98% and under conditions of constant flow is presented in Table 5.

Table 5. The comparison of four commonly used types of biogas enrichment installation (Deng and Hägg, 2010)

	a	b	c	d
Feed flow [Nm^3/h]	1000	1000	1000	1000
Pressure [MPa]	20	20	20	20
Enriched biogas flow [Nm^3/h]	566	638	769	745
Purity of CH_4 [% vol.]	98	98	98	98
Purity CO_2 [% vol.]	78	92.5	98.1	92.2
Methane recovery [% obj.]	85.5	92.7	99.7	97.3
Total membrane area [m^2]	956	1226	1297	1167
Investment cost [M$]	1.86	2.34	3.06	2.99
Operational cost [$/$Nm^3$ enriched biogas]	0.062	0.084	0.088	0.089

Three-stage configurations are the subject of very few theoretical works on modelling separation processes (Datta and Sen, 2006; Kim et al., 2012; Makaruk and Harasek, 2009; Qi and Henson, 1998).

In order to model multi-stage systems a program has been written. It allows modelling of carbon dioxide and methane permeation process through the polyimide membrane placed in the hollow fibre module.

The Runge–Kutta method of 4^{th} order applied in Matlab 7.12 enables to determine the distribution of concentrations and pressures as a function of the length of the module. Permeability coefficients for the test gases designated previously in experiment were used during calculations. Model simulation allows determining the effect of the flow streams dividing ratio (stage cut) on the composition of the outlet streams.

Assumptions made are as follows:

- process pressure is fixed at 0.6 MPa,
- the process runs under isothermal conditions (294.15 K),
- co-current flow of streams in the module,
- all membranes are made from the same type of polymer,
- set up the stage cut are constant for all modules in the system,
- the composition of the gas in the feed system is unchanging ($x_{CH_4} = 0.5$ and $x_{CO_2} = 0.5$),
- flow of the feed is fixed ($Q = 0.17$ m^3STP/h),
- permeation of gas components is described by the earlier discussed equations.

4.3. Results of process computer simulation and their discussion

Assessment of suitability of individual configuration is possible by comparing all systems common used in gas separation yield. Modelling permeation systems was made for both single and two stages. The starting point for the calculation was the experimental studies using a single module. Next, a simulations for one stage system with recycling of the permeate flux (Fig. 13) were made. Modelling was performed for three of the recirculation of the permeate stream (0.25, 0.5, 0.75).

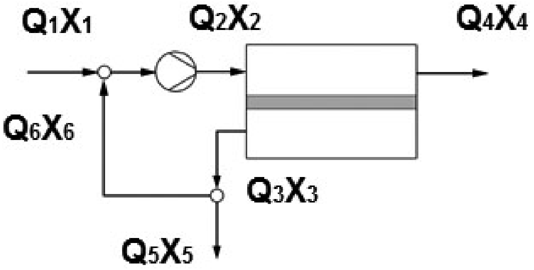

Fig. 13. One-step system with a recirculation of permeate stream

The increase of permeate recycle causes a decrease of methane loss in the process. There is also reduced volume of the permeate stream withdrawn from the system, so that the methane recovery rate of the total stream mixture increases. The disadvantage of a high degree of recirculation of the permeate flux is progressively dilution of the feed system (Fig. 14).

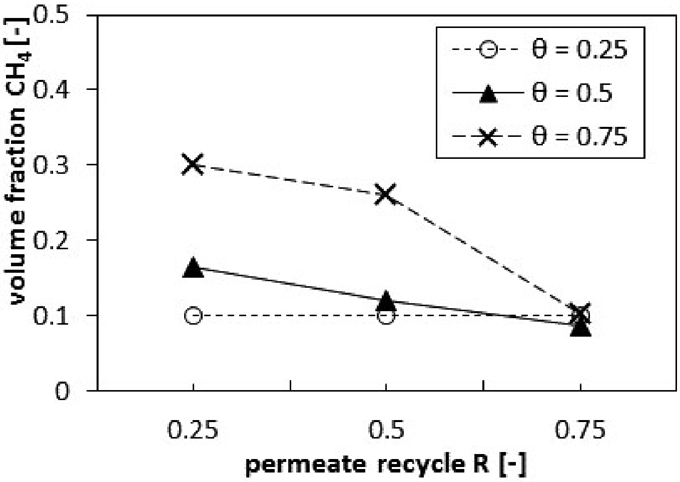

Fig. 14. Influence of permeate recycle on methane content in permeate stream as a function of stage cut parameter

The volumetric flow of the retentate stream increases, while concentration of methane contained therein is decreasing, which leads to a reduction of the final product quality and decrease the degree of retention of CH_4. Methane retention is defined as:

$$R = 1 - \frac{w_{i,P}}{w_{i,F}} \tag{15}$$

In the calculation of the size of the stage cut was changed. This parameter greatly influences the degree of methane enrichment of product, which has previously been demonstrated experimentally. At the same time, stage cut exerts an impact on the process economy. With the increased stage cut, the concentration of methane retained on the high pressure is also increasing, and is undoubtedly an advantage of the process (Fig. 15).

Computer simulations were conducted to compare the process of permeation in the system with recycling of the permeate stream with the installation without them but with the same membrane area. Calculations were made for the three values of recycle permeate flux as well as for the three sizes of stage cut parameter. Total recovery of methane has been determined. The results are presented in Tables 6–8.

Regardless of the stage cut, higher concentrations of methane were obtained for one modular system without recirculation. Experimentally determined effect of methane enrichment on size of stage cut has been confirmed. Increase in the degree of methane content in the retentate stream is associated with

Table 6. A comparison of modelling results for one step system with and without recirculation of permeate stream [Q_F = 0.17 Nm^3/h, P = 0.6 MPa, T = 294.15 K, θ = 0.25]

	Recirculation [%]	Stage cut = 0.25				
		Retentate [volume fraction]		Permeate [volume fraction]		Recovery CH_4 [%]
		CH_4	CO_2	CH_4	CO_2	
One stage system with recirculation of permeate stream	25	0.6	0.4	0.1	0.9	96
	50	0.57	0.43	0.1	0.9	97.1
	75	0.53	0.47	0.1	0.9	98.4
One stage system without recirculation of permeate stream	25	0.623	0.373	0.13	0.87	93.4
	50	0.624	0.376.	0.13	0.87	93.4
	75	0.63	0.37	0.13	0.87	93.9

Table 7. A comparison of modelling results for one step system with and without recirculation of permeate stream [Q_F = 0.17 Nm^3/h, P = 0.6 MPa, T = 294.15 K, θ = 0.5]

	Recirculation [%]	Stage cut = 0.5				
		Retentate [volume fraction]		Permeate [volume fraction]		Recovery CH_4 [%]
		CH_4	CO_2	CH_4	CO_2	
One stage system with recirculation of permeate stream	25	0.75	0.25	0.163	0.837	86
	50	0.69	0.31	0.12	0.88	92
	75	0.6	0.4	0.087	0.913	96.5
One stage system without recirculation of permeate stream	25	0.789	0.211	0.20	0.8	80
	50	0.785	0.215	0.199	0.891	80.5
	75	0.785	0.215	0199	0.891	80.5

Table 8. A comparison of modelling results for one step system with and without recirculation of permeate stream [Q_F = 0.17 Nm^3/h, P = 0.6 MPa, T = 294.15 K, θ = 0.75]

	Recirculation [%]	Stage cut = 0.77				
		Retentate [volume fraction]		Permeate [volume fraction]		Recovery CH_4 [%]
		CH_4	CO_2	CH_4	CO_2	
One stage system with recirculation of permeate stream	25	0.86	0.14	0.34	0.66	52.9
	50	0.86	0.14	0.26	0.74	68.8
	75	0.79	0.21	0.1	0.9	90.3
One stage system without recirculation of permeate stream	25	0.88	0.12	0.40	0.6	35.1
	50	0.89	0.11	0.42	0.58	29
	75	0.88	0.12	0.38	0.62	42.7

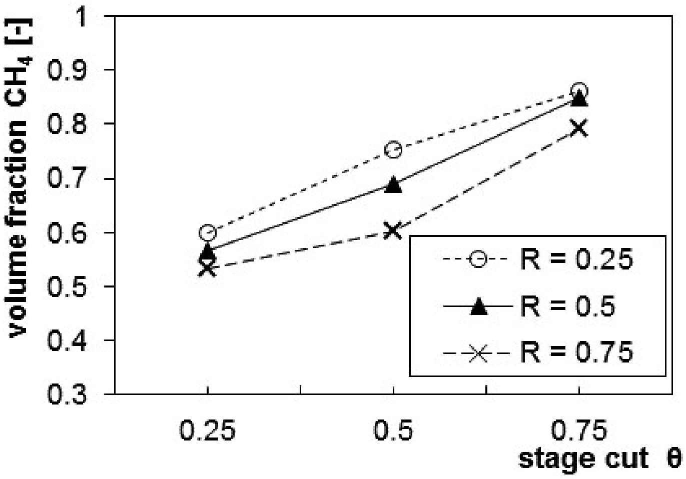

Fig. 15. Influence the degree of methane retention and stage cut parameter on concentration of methane in the retentate stream

a reduction of its flow. Hence, methane recovery is reduced. In case where the aim of the process is obtaining the purest product, it is proposed to apply a single-stage system without recycle permeate stream with a stage cut equalled to 0.75. The highest methane recovery takes place in the system with 75% recirculation of permeate stream and a stage cut of 0.25.

If one stage system does not assure the desired purity of the product the possibility of a multi-stage system application should be considered. In the stripping cascade the first stage retentate stream feeds further module, while the second stage permeate is recycled in its entirety at the beginning of the installation, where it connects with the feed stream system – as shown in Fig. 16 (Rautenbach, 1996).

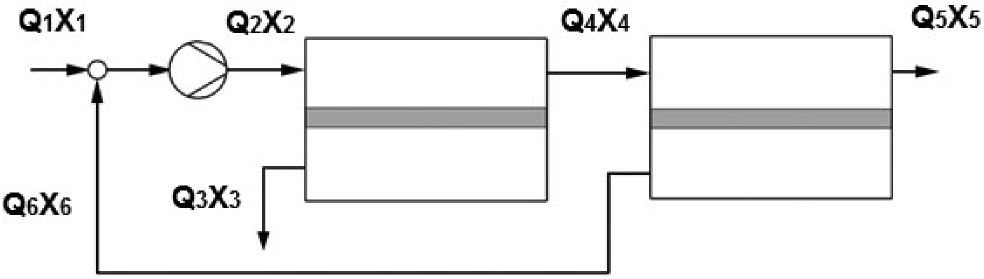

Fig. 16. Two stage system with recirculation of the permeate (stripping)

With the increasing stage cut the methane enrichment degree of the retentate stream also increases (Fig. 17). For the stage cut of $\theta = 0.75$ a product with 97% methane content was obtained. The high value of the stage cut is also associated with insufficient recovery of methane, which is caused by the fact that the increasing permeate stream is enriched in the desired component and considerably increases the amount of wasted methane.

The high stage cut causes the need to use membranes with larger surface area, increasing the unit investment cost (Fig. 18).

The comparison of the results of modelling process in a cascade with permeate feedback with a process running in a single stage and having the same area has been shown in Table 9. For stage cut of less than 0.5 it is preferable to perform a single-stage permeation system, whereby the methane concentration reaches 83% vol. In order to obtain a greater enrichment it is advisable to use a two-stage system, which allows obtaining a product with methane content higher than 97% CH_4 vol.

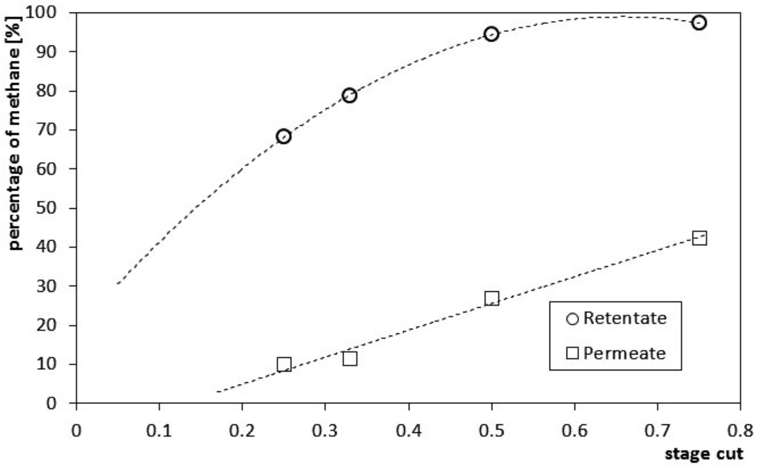

Fig. 17. Effect of stage cut on the concentration of methane in the streams leaving the stripping cascade

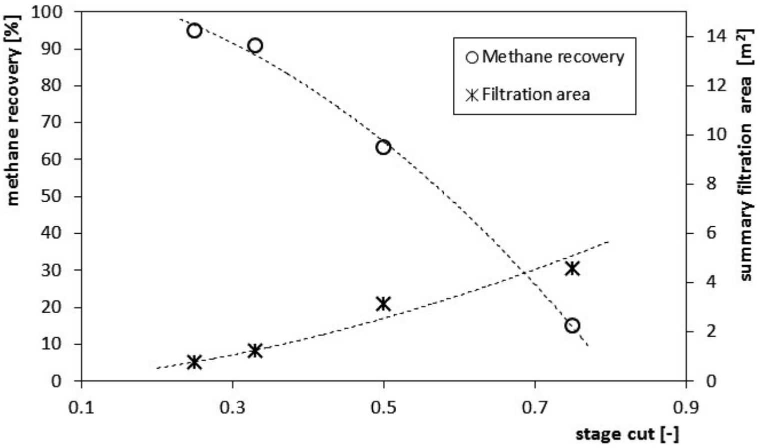

Fig. 18. The influence of the stage cut on methane recovery and the summary filtration area required in the two-stage system with recirculation

Table 9. The comparison of cascade with permeate recirculation and one stage system with the same area filtration

	Stage cut	Retentate [volume fraction]		Permeate [volume fraction]		Recovery CH_4 [%]
		CH_4	CO_2	CH_4	CO_2	
Two stage system	0.25	0.68	0.32	0.099	0.901	94.3
	0.33	0.788	0.212	0.115	0.885	90
	0.5	0.945	0.055	0.27	0.73	62.9
	0.75	0.974	0.026	0.424	0.57	14.8
One stage system	0.25	0.749	0.251	0.178	0.828	84.1
	0.33	0.83	0.17	0.241	0.759	72.88
	0.5	0.899	0.101	0.464	0.536	14.72
	0.75	0.9	0.1	0.5	0.5	2.1

Computer simulations have also been used to describe a two-stage concentrating cascade, in which the first stage permeate stream is recompressed and purified in a separate module. Retentate stream coming from the second module can be re-purified in the first module, or received together with the retentate stream from the first stage (Fig. 19).

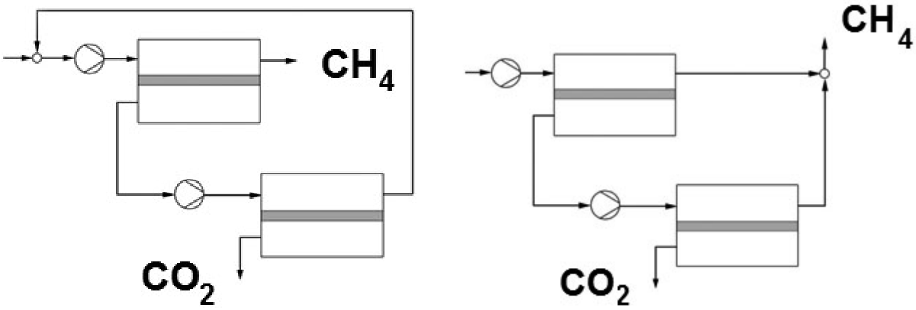

Fig. 19. Two types of concentrating methane cascade for biogas enrichment

The most important advantage of this kind of cascade is to obtain high purity permeate stream. In both cases the methane content in permeate was less than 5% vol. CH_4. The degree of retention amounted to 96.9% for the first case, and 94.7% for the system without recycle of the retentate stream. The disadvantage of the concentrating cascades is to produce relatively low calorific value product. The concentrations of methane in the retentate streams are 72% vol. and 65.7% vol., respectively. The comparison of the results of modelling process runs in cascade concentrating with the retentate back and one stage system of the same area is shown in Table 10.

Table 10. The comparison of concentrating cascade with retentate recirculation and one stage system with the same membrane area

	Retentate [volume fraction]		Permeate [volume fraction]		Recovery CH_4 [%]
	CH_4	CO_2	CH_4	CO_2	
Cascade system with recirculation retentate stream	0.72	0.28	0.022	0.978	95.9
One stage system	0.83	0.18	0.245	0.755	72.1

High recovery of methane and the production of trace amounts of methane in the permeate stream take place in the two-stage system. Choosing the right solution depends on the objectives the process. For obtaining highly enriched in methane retentate it is recommended to use a single-stage system, which allows getting a higher enrichment of the product, and the recovery of methane is bound 96%. In order to obtain high purity carbon dioxide it is proposed to use a concentrating cascade.

For simulations of CO_2/CH_4 permeation in cascade system without retentate recirculation analogously high methane recovery was achieved. Permeate stream was obtained with a very high content of carbon dioxide. The high-pressure retentate was characterised by a methane concentration of 72% vol. In comparison with the same filtration area in one stage system the highest recovery methane was obtained in cascade without retentate recirculation. On the other hand a more purified product was obtained in one module installation (Table 11). Table 12 shows a comparison of four configurations assuming the flow dividing ratio of $\theta = 0.5$.

Table 11. The comparison of concentrating cascade without retentate recirculation and one stage system with the same membrane area

	Retentate [volume fraction]		Permeate [volume fraction]		Recovery CH_4 [%]
	CH_4	CO_2	CH_4	CO_2	
Cascade without recirculation of retentate	0.657	0.343	0.038	0.962	97.9
One stage system	0.826	0.273	0.237	0.763	73.7

Table 12. The comparison of results modeling for all type of installation biogas enrichment

	Retentate		Permeate		Methane retention [%]	Filtration area [m^2]
	Flow [m^3(STP)/h]	CH_4 [vol. fraction]	Flow [m^3(STP)/h]	CH_4 [vol. fraction]		
Permeate recirculation	0.097	0.752	0.073	0.164	75	0.927
Stripping cascade	0.057	0.945	0.114	0.27	85.6	3.1
Concentrating cascade with retentate recirculation	0.1133	0.72	0.057	0.022	96.9	1.23
Concentrating cascade without retentate recirculation	0.127	0.657	0.043	0.038	94.7	1.17

From the assessment of the suitability of each configuration for biogas enrichment the most preferably represents a stripping cascade. The product has a high concentration of methane, which allows the use it as a fuel. On the other hand, this solution suffers from low productivity and a high loss of methane in the permeate stream. Moreover, demand for surface in this type of system is the greatest of all solutions. The highest degree of retention of methane is obtained by its concentrating in cascade with recycling of the retentate.

5. CONCLUSIONS

Constructed and tested in the field Mobile Membrane Installation (MMI) is a good tool for testing modules offered by the manufacturers at the specific field installations and providing design data for biogas plant operators planning to construct a plant for upgrading their product to be supplied to the grid or compressed for automotive applications (CNG). After the testing MMI in all these sites (all longer than one month of continuous operation) no remarkable changes in module permeability fluxes, pressure drop on the flow elements, etc. have been observed. Moreover, taking into account the fact that over 700 membrane installations for natural gas upgrading by CO_2 removal are working all over the world, it can be stated that this technology has future for biomethane upgrading as well. The eight year operation of such a plant (natural gas upgrading) is reported by Abu El Ela Mohamed and Nabawi (2008). The adjustment in system operating conditions due to natural membrane aging and other requirements to fulfil targets of maximising system hydrocarbon recovery and meeting acceptable CO_2 sales gas specification were introduced. However, the system was operational over the whole period. Then modules were replaced by new ones. These all experiences from oil and gas industry may be useful for methane enrichment plants and helpful in the development of this important sector of renewable energy generation based on biomass. The applied simulation computer program is found to be an important method to describe permeation of CO_2 and CH_4 mixture through polyimide membrane and to design multistage systems where pressure product is desired. In general, this study illustrated importance of the system configuration on the process efficiency and energy requirements related to the enrichment of feed stream in the methane. Modelling methods presented in the paper allow to predicts efficiency of membrane separation process and specify amount of methane possible final recovery and its concentration and volume flow rate at the individual stages in multistage systems. Reliable simulations allow minimizing the loss of valuable methane, which is very important especially when biogas is considered as fuel. The highest degree of retention of methane is obtained by gas concentrating in cascade with recycling of the retentate.

SYMBOLS

d_p	Pore size, m
d_k	The kinetic diameter of gas molecule, Å
l	Capillary length, m
n	Number of fibbers, –
p	Partial pressure, cmHg. Pa
w	Mass fraction, –
x	Volumetric fraction, mol mol^{-1}
A	Filtration area, m^2
D_i	Diffusion coefficient of component i, m^{2}s^{-1}
D	Diameter, m
J	Flux, m^3(STP)· m^{-2}s^{-1}
M	Molecular weight, g mol^{-}1
Q	Flow, m^3(STP) h^{-}1
P	Pressure, cmHg . Pa
P	Permeance, GPU = 10^{-6} cm^3 cm^{-2}s^{-1}cmHg^{-1}
P	Permeability, Barrer = 10^{-10} cm^3 cm cm^{-2}s^{-1}cmHg^{-1}
R	Gas constant, J mol^{-1}K^{-1}
R_r	Retention rate, –
S	Sorption coefficient, mol cm^{-3}
T	Temperature, K

Greek symbols

α	Membrane selectivity, –
α^*	Ideal membrane selectivity, –
γ	Activity coefficient of pressure, –
δ	Thickness of active layer, m
ε	porosity, m^{3}m^{-3}
θ	Stage cut, –
λ	The mean free path of molecule, m
μ	Viscosity of gas, Pa s
ν	Velocity, m s^{-1}
ρ	Density, g cm^{-3}
ϕ	Pressure ratio, Pa Pa^{-1}

Index

i, *j*	Components
B	Biogas
F	Feed
P	Permeate
R	Retentate

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