



The influence of the moisture content in gaseous CO₂/N₂ mixture on selected parameters of CO₂ separation in a capillary polymeric membrane

Grzegorz Wiciak

Silesian University of Technology, ul. Konarskiego 18, 44-100 Gliwice, email: grzegorz.wiciak@polsl.pl

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ABSTRACT

In the paper, analysis of results of the experimental investigation on the influence of the moisture content in treated gaseous mixture on CO₂ separation process, considering CO₂/N₂ mixture separation, is presented. The paper focuses on identification of the influence and introductory analysis of selected operational parameters of the membrane on carbon dioxide separation parameters, considering CO₂/N₂ mixture, humidified in the range of 0%–100% in relative humidity. During the experimental research, the UMS A2 membrane by UBE was used. The membrane selection was supported with former investigations, performed with dry gases usage. The main aim of the investigation was to verify the influence of gas humidification on CO₂ separation parameters. The research concerned actions focusing on the determination of membranes' operational parameters and their characteristics under conditions of moisture presence in exhaust gases, considering CO₂ separation. That mixture simulated maximal CO₂ content, which might occur in exhaust gases of power production processes, especially in the case of natural gas and hard coal combustion. In the paper, characteristics and results of the comparative analysis of CO₂ separation from CO₂/N₂ mixture referring to dry and humidified gases are presented. Based on the research performed, humidification of gas unfavorably affects operational parameters of CO₂ separation process from CO₂/N₂ gas mixture.

Keywords: CO₂ capture; Membrane separation; Membrane; Gas humidity

1. Introduction

For gas separation purposes, the inorganic polymeric membranes are most commonly used [1–4,10–22]. Important factor, deciding on applicability of membrane separation processes, is the proper selection of semipermeable membrane of stated parameters and operational features.

The membrane separation consists in division of gaseous substances on membranes. That process utilizes differences in physical and chemical properties between selected ingredients of gaseous mixture and material of the membrane. The driving force for the mass transport through the membrane and the overall separation process is the difference in the chemical potential along two sides of the membrane. This may include the difference

of pressures, temperatures, concentrations, and electric potentials [1,2].

Previously performed research on capillary polymeric membranes UMS-A2 by UBE Industries Ltd., Japan, concerned only dry gases [1,2,5–10]. During experimental investigations, synthetic mixtures of gases, simulating exhaust gases from power generation processes of maximal 20% CO₂ content were utilized.

That was reasoned by fact that in power engineering technologies, where those membranes could be potentially introduced in CO₂ separation units, the use of only dry gases was compulsory due to technological limitations. Due to continuous increase in power demand, the proper and effective utilization of CO₂ reduction methods from power generation units, either in the form of capturing before combustion

* Corresponding author.

(precombustion), after that process (postcombustion) or using oxy-combustion, states the key factor. Technologies of exhaust gases treatment are defined by numerous national or international requirements on natural environment protection. Thus, an extensive scientific research in this field is performed, aiming to save the highest possible effectiveness level of power and useful heat production.

In former research aiming to identify properties of UMS-A2 membranes, used for CO₂ separation processes from power and heat units, performed in number of stages, dry gases were used [1,2,4,7,9]. Satisfactory results concerning quality of CO₂ separation process for those membranes in a laboratory scale were obtained. Those results were proven in simulations of power production systems, concerning especially the influence of the membrane capturing technology on power and heat production effectiveness, especially focusing on hard coal combustion. The investigation was performed considering especially energy consumption of CO₂ capture processes during utilization of polymeric membrane separators. Experimental results were used for performance of the analysis and computational verification of mentioned parameters [1,4,7,9,10]. During mentioned computational research, active area of single membrane equal to $A = 0.011069 \text{ m}^2$ as well as permeability coefficients $P_{\text{CO}_2} = 2.855 \text{ (m}^2\text{)/(m}^2 \text{ h bar)}$ and $P_{\text{N}_2} = 0.1 \text{ (m}^2\text{)/(m}^2 \text{ h bar)}$ were identified. In Ref. [1], results of computational simulation of operational characteristics of the membrane operation for the mixture of 20% CO₂ and 80% N₂ for a dry gas, and respective experimental data, were juxtaposed. Based on data presented in Ref. [1], considering estimated active area of the membrane, results of the experiment and simulation coincided well, showing deviations below 10%. Thus, low divergence observed (below 10%) is at acceptable level and coincidence of those results might be stated. Considering number of previously mentioned references [1,4,7,9,11–23] and other researches [23–30], the change in permeability coefficients following change in separated mixture composition might be stated. By utilization of humid gaseous mixture, vital change and fall in operational parameters of the membrane, regarding their values for the case of dry mixture, should be expected. In number of research reports [23–36], acquired data suggests vital decrease in mentioned specific parameters. Thus, in this paper, the verification of the influence of gas humidification on CO₂/N₂ separation parameters using UMS-A2 by UBE Industries Ltd. polymeric membrane, considering its utilization on exhaust gases outlets in power installations, is described.

Real exhaust gases, after completion of treatment processes, state humid gases, include some content of moisture. Steam share in exhaust gases results from the moist content in fuel and amount of water injected in exhaust gases treatment processes. On the basis of investigations [21], 20% H₂O content in exhaust gases was assumed for OP650 boiler for a fuel of 12% H₂O content and a mass flow rate of water vapor injected in exhaust gases treatment processes equaled to 20 t/h. Thus, in further investigation this factor was taken into account.

Due to initial satisfactory results, further tests aiming identification of the influence of varying moisture content in CO₂/N₂ gas on CO₂ separation processes of exhaust gases in power units and focusing on estimation of their operational

characteristics were proposed. Maximal CO₂ content in real exhaust gases from power production processes depend strongly on a fuel type [21] and for example, in the case of hard coal it equals $18.6 \div 19.2 \text{ CO}_{2\text{max}}$ for lignite $8.7 \div 19.5 \text{ CO}_{2\text{max}}$ for heavy oil $15.5\% \text{ CO}_{2\text{max}}$ for light oil $15.3 \div 15.4\% \text{ CO}_{2\text{max}}$ and for natural gas $9.5 \div 12.5(15)\% \text{ CO}_{2\text{max}}$. Thus, in the experimental research, the highest acceptable value of CO₂ was stated to be equal to 20% and two-component gaseous mixture of 20% CO₂ and 80% N₂ was used, assuming that this mixture could simulate the performance of exhaust gases good enough on this stage of the research.

On this basis, the research focused on the identification of the influence of gas humidification on CO₂ separation parameters was performed. In order to run the investigation, number of tests using gas of variable humidity in the range of 0%–100% relative humidity were done. Results of these tests were referred to former basic investigation of UMS-A2 membranes using dry gas of 20% CO₂ and 80% N₂ content.

2. Methodology of experimental investigation

Two main parameters, describing the effectiveness of separation process, are selectivity and permeability. The permeability determines the amount of a component transferred through the membrane, whereas the selectivity describes, which components of the feed mixture would be transferred and which would be not. The stream supplying the membrane is denoted as the feed. The feed is divided then into two substreams: a permeate (stream flowing through the membrane) and a retentate (stream retained by the membrane's). Respective streams are presented on the scheme, showing also the experimental setup (Fig. 1).

Commonly, in order to determine effectiveness of the CO₂ separation parameters, two quantities are used. One of them is a molar or a volumetric share of CO₂ in permeate (Y_{CO_2}), which identifies permeate purity; second one is carbon dioxide recovery coefficient R , denoting share of CO₂ gained from exhaust gases available in separated stream, and it is expressed by Eq. (1) as follows:

$$R = \frac{n_P(Y_{\text{CO}_2})}{n_F(X_{\text{CO}_2})} \quad (1)$$

where n – volumetric stream of P – permeate or F – feed, respectively; Y_{CO_2} – CO₂ content in permeate; and X_{CO_2} – CO₂ content in the feed.

Recovery coefficient equal to $R = 1$ denotes the case of full separation of carbon dioxide from exhaust gases. Literature [1] suggests that molar share of CO₂ in permeate (Y_{CO_2}) and recovery coefficient of carbon dioxide should not drop below 0.9 or even 0.95. Higher values of those parameters follow lower contamination of the atmosphere by carbon dioxide [1].

The permeability coefficient P_i^* of a given ingredient through the membrane is denoted as the ratio of sorption (S_i) and diffusion (D_i) coefficients, according to Eq. (2) dependence as follows:

$$D_i \times S_i = P_i^* \quad (2)$$

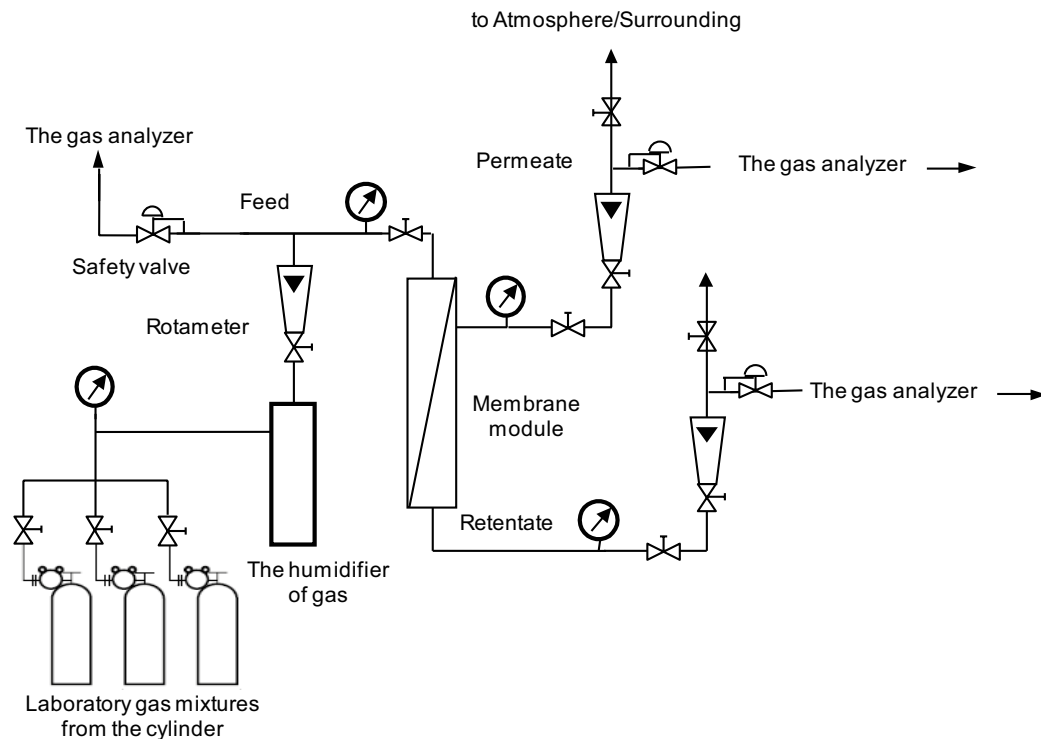


Fig. 1. The scheme of the experimental installation, involved in the research on the influence of gas humidification on CO₂ separation process parameters.

The cardinal formula, describing gas transport through the continuous membrane (the Fick's formula) may be expressed as follows:

$$J_i = -D_i \frac{dc_i}{dx} \quad (3)$$

where c_i – concentration of i th component, flowing through the membrane and x – thickness of the membrane.

Eq. (3), integrated over the thickness of the membrane and after the introduction of a number of simplifications and considerations of basic constructional features of investigated capillary membrane as well as the overall membrane module [2,10], might be finally simplified into final derivation in the following form:

$$J_i = \frac{p_i^*}{x} (p_{i,0} - p_{i,x}) \quad (4)$$

where $p_{i,0}$ – partial pressure of the i th component on the border of feed/membrane phases and $p_{i,x}$ – partial pressure of the i th component on the border of membrane/permeate phases.

On the basis of the dependence (1.4) for two-component (CO₂ and N₂) mixture, the formula describing the ratio of elementary streams flowing through the membrane may be acquired. The ideal selectivity coefficient is proportional to the ratio of values of permeability coefficients (i.e., CO₂ and N₂) $P_{CO_2}^*$ and $P_{N_2}^*$ and is expressed using Eq. (5) as follows:

$$\alpha^* = \frac{P_{CO_2}^*}{P_{N_2}^*} \quad (5)$$

The determination of elementary streams flowing through the membrane is either difficult or impossible very often. This happens usually in the case of commercially available membrane modules, of secret or inaccessible data concerning materials applied and their properties, what is observed in mentioned case as well.

Thus, an important aspect during investigation states the determination of real selectivity coefficient, expressed as the ratio of shares of subsequent ingredients in a permeate to their shares in a feed [1]. The determination of that coefficient was part of this investigation as well, and it was performed using Eq. (6) due to the lack of full characteristics of membrane parameters, especially concerning the influence of humidity on its operational parameters [1]. Comparing parameters of the membrane fed with dry, two-component gaseous mixture (CO₂ and N₂) and further experiment with humid mixture, only selectivity coefficients α linking carbon dioxide and nitrogen were used. Such selection was introduced due to the necessity of the identification of the influence of third component of the mixture (water vapor, distorting the separation process) on the selectivity coefficient α .

$$\alpha = \frac{\frac{Y_{CO_2}}{Y_{N_2}}}{\frac{X_{CO_2}}{X_{N_2}}} \quad (6)$$

During the research, number of parameters describing separation process, for example: the purity of the product – share of carbon dioxide in permeate (Y_{CO_2}), the

recovery coefficient (R), and the real coefficient of selectivity (α), were identified. Considering carbon dioxide, the latter parameters were referred only to nitrogen in order to enable the comparison of experimental results of the research on dry and wet mixtures. The recovery coefficient R for carbon dioxide was calculated based on Eq. (1).

During the research, following methodology, aiming the comparison of results for dry and wet gases investigation, was assumed. Values of process parameters were acquired on the basis of direct measurements of: temperatures, stream flow rates, and concentration (volumetric shares) of gases inflowing to and outflowing from the membrane. In this research, in order to identify mentioned parameters for dry gases, the two-ingredient mixture of 20% CO_2 and 80% N_2 was used. The feed of gaseous nitrogen–carbon dioxide mixture was humidified using dedicated moisturizer, aiming to get variable relative humidity of gas mixture. The first task during measurements was to stabilize membrane feed with stream flow rate in the range of $50 \div 525$ L/h in number of constant (considering further comparison of mentioned analyses) points of operational characteristics. These points were controlled using high-pressure gas reducer and valves located at the outlet pipelines of the feed and the retentate (as shown in Fig. 1), which were used for precise regulation of stream flow rate values. After the stabilization was reached, the reading of operational parameters was performed. During the experimental investigation, minimal instability of the flow rate at the level of $\pm 1\%$ caused mainly by limited accuracy of used instruments and a simple regulation method, were observed. At the end of measurements for the dry gas, dedicated humidifier was implemented to the system, and measurements were performed once again at the same characteristic points, as in the previous series. Membrane was fed at the constant pressure of 5 bar. The moistured feed supplied to

the membrane caused deviations of total feed flow rate in the limited range. This resulted from a simple regulation method (manual control of flow rates by feed and retentate valves), as well as assumed method of regulation of humidity supply (sealing of the system) and necessary for keeping other vital parameters constant.

The measurement begun with feeding of the membrane with a dry gas, with further introduction of moisture up to the level of water vapor content in the mixture roughly equal to 23% in order to stabilize the humidification process. Next, the moisture content in the gas was consequently raised, while registering further process points and keeping other operational parameters – feed pressure, feed flow rate – constant, with subsequent observations of changes in the gas concentration as the separation process proceeded. The executed process was linked with certain dynamics. Stable parameters of the separation process of humid gas are presented in the results section (Section 3). At the end of the research for both systems, data collected for both parts of the experiment, concerning dry and moisturized mixture, were compared. During all tests included in the research, constant pressure of 5 bar and invariable temperature of gases equal to 22°C were kept. Total time of a single test was roughly equal to 6 h. Registered flow rates instabilities, oscillating at the level of $\pm 1\%$ did not essentially influence on obtained parameters values. Thus, the process was deemed as the stable one.

3. Analysis of the comparative research results

Fig. 2 presents characteristics of the recovery coefficient R as function of feed flow rate Q_f (at constant relative pressure equal to 5 bar), gained on the basis of the influence of gas humidity on CO_2 separation parameters of capillary

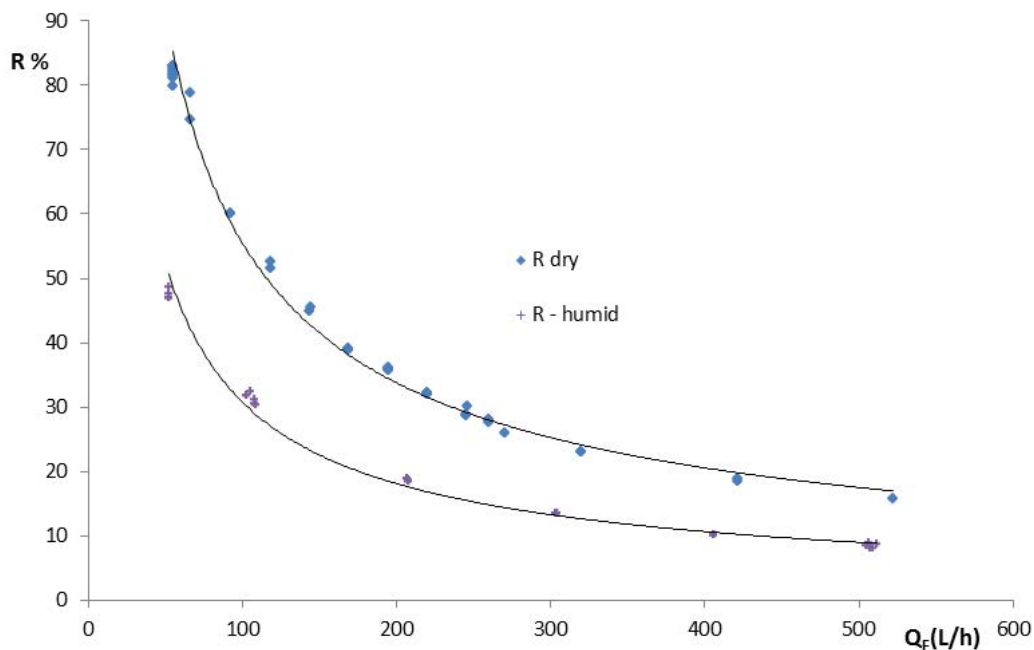


Fig. 2. Comparative chart of characteristics of recovery coefficient for dry and humid (100% relative humidity) gases in reference to the feed flow.

polymeric membrane UMS-A2. Two subcharacteristics are presented, where one refers to the dry gas case and the second refers to the moist gas of 100% relative humidity. The essential influence of gas humidification on values of the recovery coefficient R is visible. Significant drop in the R parameter for humid gas in the whole feed range flow rate from 50 to 525 L/h is visible. The drop in this parameter equaled around 30% in the range from 50 to 100 L/h of stream flow. The recovery coefficient for dry gas was in the range of $R = 55\%$ – 85% , whereas for humid gas $R = 30\%$ – 50% . The rise in feed flow rates resulted in both cases in proportional fall in the recovery parameter to below $R = 10\%$ for the moist gas case. The increase in the flow rate followed further decrease in R parameter values for both characteristics (dry and humid mixtures), which corresponded to former investigations of the membrane used [1].

For the stream flow rates exceeding 100 L/h, a difference between R values for both wet and dry gases decreased; its values changed in range of 15%–25% up to 200 L/h. The increase in the feed stream flow rate above 200 L/h resulted in further drop in difference in R parameters, keeping finally constant value close to 10%. Almost parallel shift of R characteristics for the moist gas, in reference to the dry gas case, might have proven disadvantageous influence of gas humidification. The reason of that fact may lay in the vital difference in size of H_2O particles comparing with CO_2 and N_2 , and better permeability of water through this particular polymer and additional load of the membrane, caused by water transport through the membrane.

Thus, the performance of gas (especially carbon dioxide) separation processes from mixture, contaminated with steam, was unbeneficial for that type of the membrane and it consumed more energy, considering keeping constant

R values on the desired level of $R = 80\%$ – 95% for CO_2 separation from exhaust gases.

Data presented in Fig. 3 show the comparison of characteristics of the permeate purity Y_{CO_2} , which was observed during supplying membrane with moist (of relative humidity equal to 100%) and dry gases. Research was performed at changeable stream flow rate in the range of 50–525 L/h, as previously, keeping relative pressure of 5 bar constant. By analogy to the previously acquired results (Fig. 2), vital drop in separation operational parameters occurred. Crucial drop of permeate purity Y_{CO_2} acquired, referring to the dry gas case, was observed. The action of the membrane at the feed flow rate in the range of $Q_F = 50$ – 100 L/h should be emphasized: differences in Y_{CO_2} between dry and moist gases case were negligible, and kept in that range up to $Q_F = 200$ L/h – according to characteristics, shown in Fig. 2. The increase of the feed flow rate above 200 L/h resulted in the drop of the permeate purity Y_{CO_2} and further decrease in its values, comparing to the dry gas case, in the range of 200–525 L/h. Furthermore, at the highest feed stream flow rate of $Q_F = 525$ L/h sudden drop in permeate purity equal to 8% points was visible.

Concluding, in the range of feed flow rate below 200 L/h significant drop in permeate purity Y_{CO_2} was not observed; values of that parameter were in the range of 65%–75% for both dry and moist gases. The rise of the feed flow rate above 200 L/h resulted in stable trend, when considering Y_{CO_2} parameter for moist gas, its values kept constant at around 75% level, while for dry gas case its values exceeded 75% and acquired its maximum at around 80%. Differences in Y_{CO_2} purity for both gases increased up to 2%–8% point for the maximal values of the feed stream flow rate. Thus, in the range of the low feed flow rate, the humidification of the feeding gas did not affect the Y_{CO_2} parameter significantly,

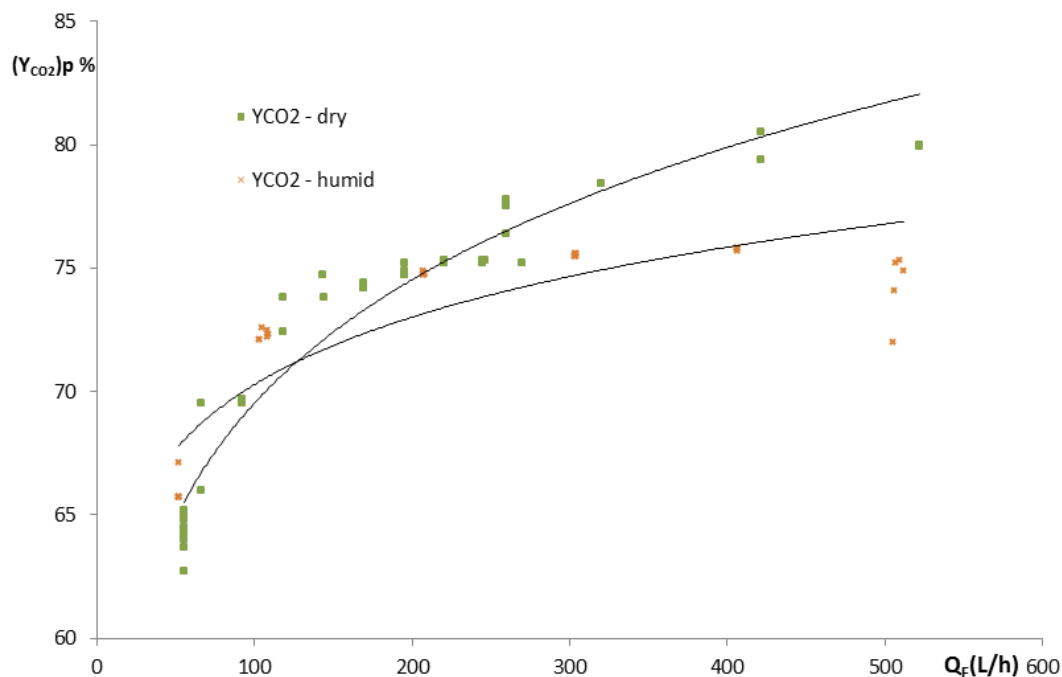


Fig. 3. Comparative chart of characteristics of permeate purity Y_{CO_2} for dry and humid (100% relative humidity) gases in reference to the feed flow.

however, further increase in the feed flow rate caused degradation of the permeate purity parameter. Moreover, for maximal feed flow rates sudden, sharp drop in Y_{CO_2} was observed.

Characteristics of the influence of the gas humidification on the real selectivity coefficient α is presented in Fig. 4. The membrane was investigated under variable feed flow rate in the range of 50–525 L/h, at constant working gas pressure equal to 5 bar. Referring to the previous research, vital similarity to characteristics shown in Fig. 3 was stated. The similarity consisted in the fact, that values of this parameter for dry and moist gases supply were indicated by analogical trend lines in the feed flow rate range below 200 L/h. In Fig. 4, insignificant differences of selectivity coefficient α , equal to 1%–2% points, between dry and moist gases are visible. Nevertheless, the increase of the feed flow rate above 200 L/h caused the increase in that difference of α coefficient values up to 4% points around maximum feed flow rate. Similarly to data shown in Fig. 3, sudden step decrease in the selectivity for maximal value of feed flow rate $Q_F = 525$ L/h was observed. Characteristics of a dry gas indicated on a continuous trend of rising values of α , whereas moisture content in gas supplied to the membrane resulted in the decrease in selectivity coefficient values and further lowering trend for feed flow rates exceeding 200 L/h. Up to that value of the feed flow rate, no vital differences, comparing dry and moist gas cases, were stated.

Characteristics indicated in Fig. 5 show results of experimental research on variable humidification of 20% CO_2 and 80% N_2 gaseous mixture (in the range of 0 ÷ 100% water content in a gas) on selectivity coefficients defined by

Eqs. (5) and (6). The investigation indicates on the way of degradation of the selectivity coefficient, depending on how the water steam gets into the exhaust gases. The research was performed in order to enable the saturation of the exhaust gases with the moisture during the gas flow.

In Fig. 5, the phase of gas humidification from 0% to 20% with subsequent degradation in the selectivity coefficient and varying feed flow rate (caused by reaction of the membrane on unstable moisture content) is visible. The selectivity coefficient α for 0% water content kept constant around $\alpha = 17$, degraded to 13 for relative humidity equal to 20% H_2O with subsequent drop in the feed stream flow rate from 518 to 514 L/h. Acquiring constant process parameters and humidification level of 23% H_2O , the rise in feed flow rate from 514 to 518 L/h occurred, keeping the value of the selectivity coefficient in unchanged range of $\alpha = 12$ –13.

Any further change in moisture content in the gaseous mixture – 58%, 73%, 80%, and 100% – resulted in the drop in values of the α coefficients relatively to the moisture content, however, certain unsteadiness of the flow rate in the range of $\Delta Q_F = 1 \div 3$ L/h appeared for humidities equal to 58% and 73%, respectively. Gas humidification exceeding 80% of relative humidity resulted in a change of the feed flow rate from 517 to 513 L/h at 100% humidity content, which might have proven the maximal load of the membrane, primarily devoted to water vapor separation.

Thus, any water content in the gaseous mixture during stabilization of the separation process results in unsteadiness of the feed flow rate. Greatest values of that variability occur at 80 ÷ 100% H_2O content, whereas for humidity in the range of 58 ÷ 73% H_2O these are the lowest.

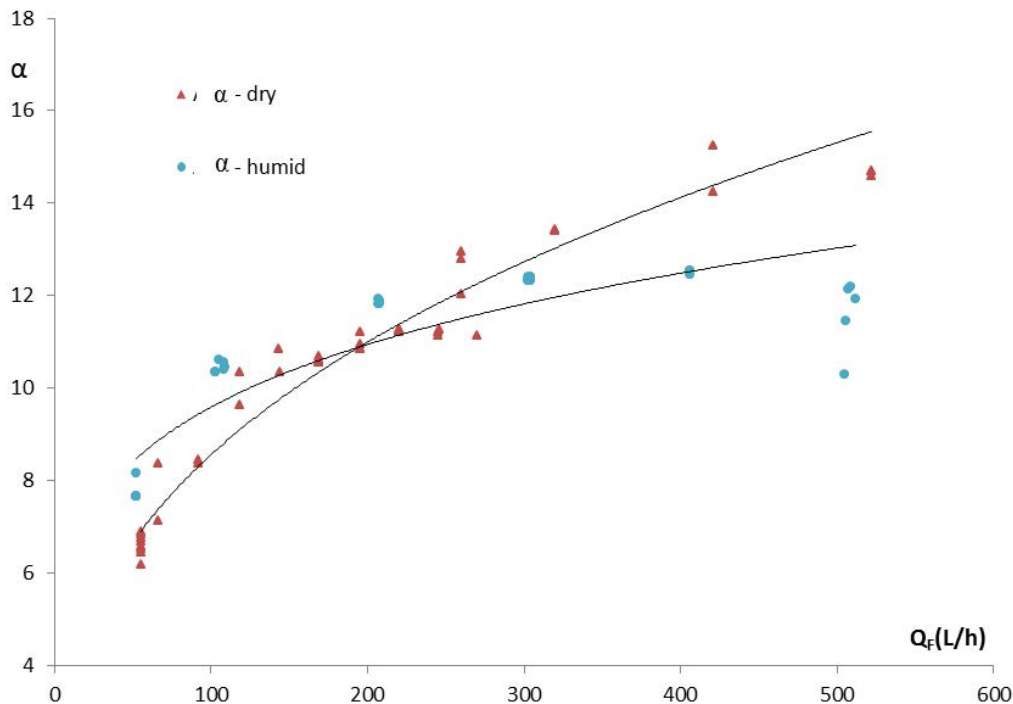


Fig. 4. Comparative chart of characteristics of real selectivity coefficient α for dry and humid (100% relative humidity) gases in reference to the feed flow.

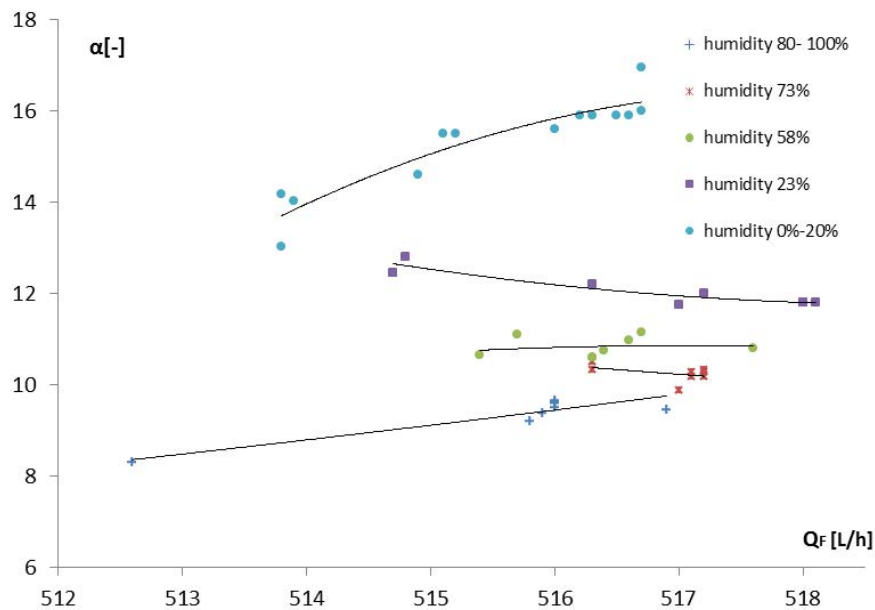


Fig. 5. Characteristics of degradation and influence of variable humidity in the range of 0%–100% H₂O on real selectivity coefficient in reference to the feed flow rate.

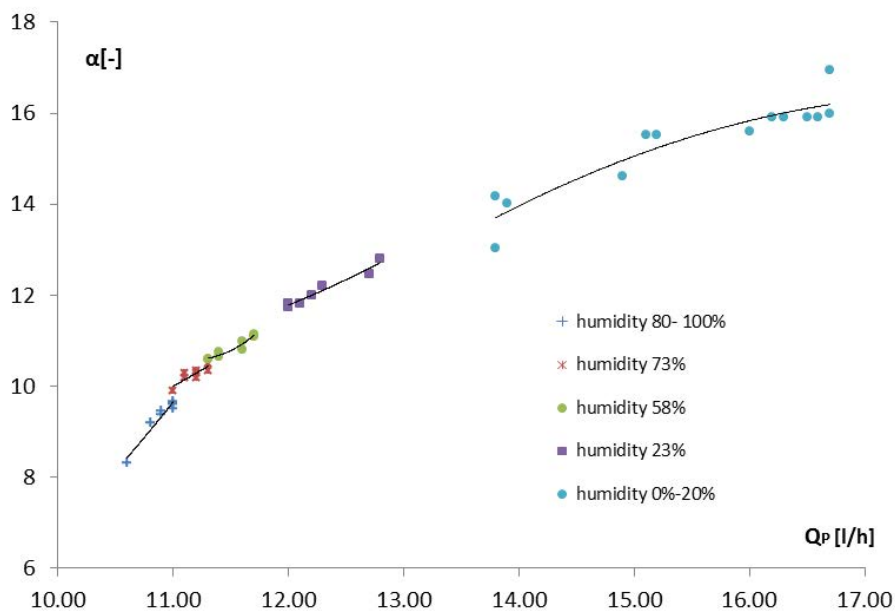


Fig. 6. Characteristics of degradation and influence of variable humidity (0%–100% H₂O) of gas on real selectivity coefficient α in reference to the permeate stream flow rate Q_p .

In Fig. 6, the change in the selectivity coefficient α as function of the permeate flow rate Q_p is presented. That characteristics presents degradation of the selectivity coefficient resulting from the increase in the moisture content in the gaseous mixture (20% CO₂ and 80% N₂). Furthermore, the drop in permeate flow rate is also visible. That characteristics shows how the rise in water content results in drop of CO₂ separation process parameters of the membrane. Based on that characteristics, the action of retaking its primary function of water separation from gas being supplied is introduced. The

same characteristics shows the drop in the selectivity coefficient from $\alpha = 17$ to $\alpha = 8$ following the rise in the moisture content with the subsequent decrease in the permeate flow rate from ≈ 20.8 L/h to roughly $Q_p = 15.4$ L/h.

Characteristics shown in Figs. 5 and 6 indicate on the degradation of the selectivity coefficient as a function of the gas stream flow rate, depending on H₂O increasing content in the gaseous mixture used. Thus, the destructive influence of the moisture content on CO₂ separation from exhaust gases process may be stated.

According to the combustion, where moisture content of 20% of relative humidity in exhaust gases appears, the visibility of that influence on separation process should be stated. Moreover, this level of gas humidification results in the decrease in CO₂ separation of around 17 ÷ 18%, in comparison with dry exhaust gases. The drop in the permeate flow rate equals 18%, in comparison with the dry gas case. The variability of the feed flow rate change in the range of

1% and it is negligible in this case. Concluding, the feed gas humidification results in overall drop in carbon dioxide separation parameters (CO₂, α , Q_p) at the level of 20%.

Characteristics shown in Figs. 7 and 8 indicate on the influence of the moisture content on Y_{CO_2} (Fig. 7) and R , α (Fig. 8) coefficients. The investigation shown was performed for constant parameters of the feed: Q_f (L/h) and p (bar). These characteristics indicate on the degradation of

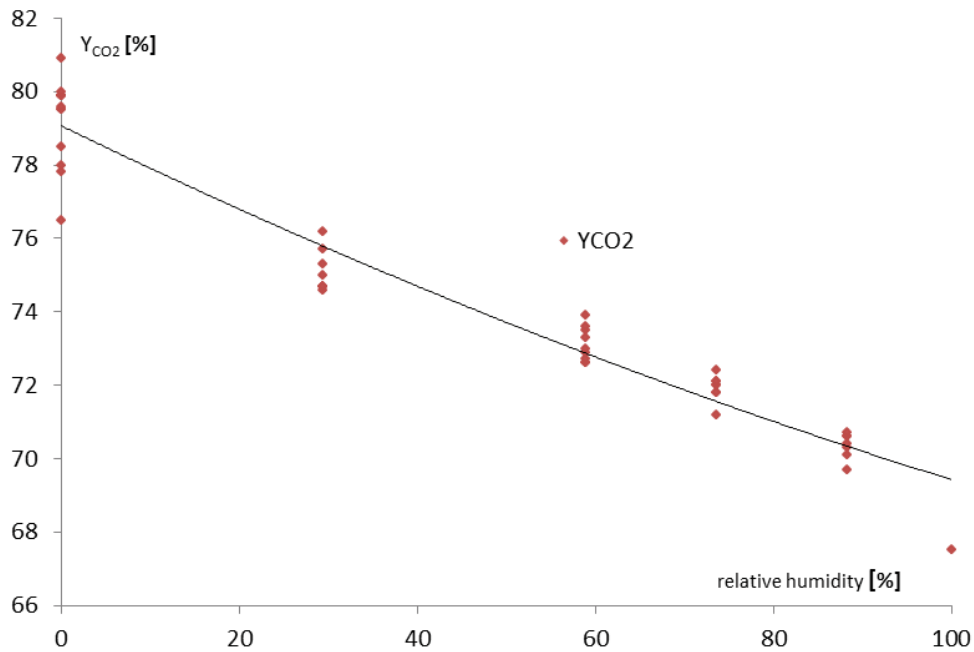


Fig. 7. Characteristics of changes in permeate purity as a function of changes in relative humidity 0%–100% H₂O.

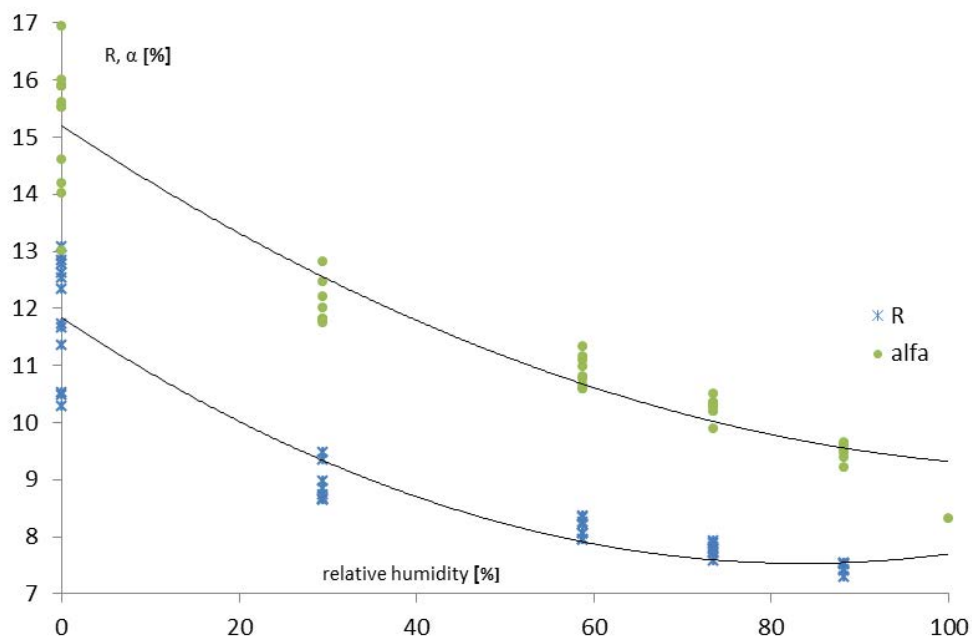


Fig. 8. Characteristics of changes in R and α in dependence of the change in relative humidity 0%–100% H₂O.

separation parameters as a function of moisture content in exhaust gases (simulated by 20% CO₂ and 80% N₂ gaseous mixture).

Those investigations were performed according to the value of the feed flow rate, at which the highest values of selectivity α and acceptable permeate purity Y_{CO_2} were acquired considering dry gas. In former research, the influence of the feed stream flow rate and the feed pressure on CO₂/N₂ separation process parameters was investigated. In those experiments, the recovery coefficient R for the lower range $Q_F = 50\text{--}100$ L/h exceeded 95%, whereas maximal values of selectivity α were observed for $Q_F = 400\text{--}500$ L/h. Therefore, the decision of performance of the research of gas humidification influence on the separation process parameters (α , Q_F , Y_{CO_2}) for environmental point of view should be based on highest values of α , taking into account relatively low values of R and Y_{CO_2} for that range of Q_F and pressure p . In majority, the low values of those parameters result from the low volume of gas transport inside the UMS-A2 membrane module and relatively low membrane's area, equal to $A = 0.011069$ m².

4. Summary

Based on the research performed, the statement of the deterioration in carbon dioxide separation parameters, especially in recovery coefficient R , permeate purity Y_{CO_2} and real selectivity coefficient α , resulted from the rise in moisture content, may be introduced. The increase of the moisture content in a gas results in degradation of all separation parameters. Membrane feed parameters assumed in the paper were forced by the necessity of referring the results to the former investigations, where dry gas of the same partial concentration of components (20% CO₂ and 80% N₂) was used.

The increase in the moisture content in a gas up to 20% caused the drop in selectivity coefficient α by 17%–18%, in comparison with the dry gas [1]. Next, the drop in permeate stream flow rate of around 18% occurred. Maximal moisture content by 100% relative humidity resulted in the degradation of selectivity coefficient by 52.9%. Thus, the moisture content in the range of 0%–100% H₂O in synthetic exhaust gases (20% CO₂ and 80% N₂) caused the instability of the membrane operation, emerging as variations in a feed flow rate in the range below 1%.

On the basis of the performed research, the maximal acceptable level of the relative humidity of the exhaust gases in the feed for this type of the membrane equals 20%. Thus, this value refers to the maximal water vapor content in exhaust gases. In that range of water vapor content, the membrane reveals relatively insignificant degradation of operational parameters, in comparison with higher values of relative humidity. Therefore, the operation of membrane is acceptable in the range up to 20% of water in exhaust gases. The investigation was performed for temperature equal to 22°C in all tests. The supply pressure of 5 bar and feed flow of $Q_F = 518$ L/h, with deviation oscillating at the level of $\pm 1\%$ resulting from presence of water and prime action of the membrane used, was kept.

Thus, the optimal operational environment for the investigated membrane is probably its feeding with either a dry gas or a gas of a trace moisture content.

Symbols

Y_{CO_2}	—	Purity of the permeate for CO ₂ , %
Y_{O_2}	—	Purity of the permeate for O ₂ , %
X_{CO_2}	—	Purity of the feed for CO ₂ , %
$n_{p'}Q_p$	—	Flow rate of permeate, L/h
$n_{f'}Q_f$	—	Flow rate of feed, L/h
p	—	Feed pressure, bar
R	—	Recovery rate, %
α	—	Real selectivity factor
P_i^*	—	Permeability of an i th component, m ³ /(m h bar)
S_i	—	Sorption coefficient of an i th component, m ³ /m ³ bar
D_i	—	Diffusivity coefficient, m ² /s

References

- [1] J. Kotowicz, K. Janusz-Szymańska, G. Wiciak, Membrane technologies for the capture of carbon dioxide from exhaust gases for the supercritical carbon power plants Wydawnictwo Politechniki Śląskiej, Monografia – Politechnika Śląska, nr. 551, Gliwice, 2015 (in Polish).
- [2] M. Bodzek, J. Bohdziewicz, K. Konieczny, Membrane techniques in environmental protection, Wydawnictwo Politechniki Śląskiej, Gliwice, 1997 (in Polish).
- [3] G. Wiciak, K. Janusz-Szymańska, J. Kotowicz, Experimental and numerical studies of CO₂ separation of polymer membranes using a gaseous reference mixture, Rynek Energii, 2 (2014) 98–103 (in Polish).
- [4] K. Janusz-Szymańska, G. Wiciak, Comparative analysis of the results of the numerical and measurement experiment in the context of research of parameter of CO₂ separation using the membrane method, Energetyka, 11 (2013) 795–799 (in Polish).
- [5] L. Remiorz, Numerical and experimental study of acoustic CO₂ separation, Wydawnictwo, Politechniki Śląskiej, Monografia – Politechnika Śląska nr 562, Gliwice, 2015 (in Polish).
- [6] G. Wiciak, K. Janusz-Szymańska, L. Remiorz, The Impact of CO₂ Concentration on the Properties of a Polymer Membrane Separator Intended for the CCS Technology, R. Zevenhoven, Ed., 27th International Conference on Efficiency, Cost, Optimization, Simulation and Environmental Impact of Energy Systems (ECOS 2014), Turku, Finland, 15–19 June 2014, Vol. 2, Red Hook, Curran, 2014, pp. 1316–1330.
- [7] G. Wiciak, L. Remiorz, J. Kotowicz, Instalacja laboratoryjna do synchronicznych badań separacji ditlenku węgla metodami membranową i akustyczną. Analiza systemów energetycznych, Praca zbiorowa, Pod red. B. Węglowskiego, P. Dudy. Kraków: Wydaw. Politechniki Krakowskiej, 2013, pp. 331–349.
- [8] A. Janusz-Cygan, The Use of Solid Membranes for the Separation of Carbon Monoxide from Exhaust Gases, Instytut Inżynierii Chemicznej Polskiej Akademii Nauk – Praca doktorska, Gliwice, 2016 (in Polish).
- [9] K. Warmuziński, A. Janusz-Cygan, M. Jaschik, M. Tańczyk, A hybrid separation process for the recovery of carbon dioxide from flue gases, Energy Procedia, 37 (2013) 2154–2163.
- [10] E. Powell, G. Qiao Greg, Polymeric CO₂/N₂ gas separation membranes for the capture of carbon dioxide from power plant flue gases, J. Membr. Sci., 279 (2006) 1–49.
- [11] E. Biernacka, T. Suchecka, Membrane Technologies in the Environmental Protection, Wydawnictwo SGGW, Warszawa, 2004 (in Polish).
- [12] S. Dushyant, D.R. Luebke, H.W. Pennline, A Review of Carbon Dioxide Selective Membranes. A Topical Report, National Energy Technology Laboratory, United States Department of Energy, 2003.
- [13] J. Marano, J. Ciferino, Integration of gas separation membranes with IGCC identifying the right membrane for the right Job, Energy Procedia, 1 (2008) 361–368.
- [14] S. Yan, M. Fang, W. Zhang, W. Zhong, Z. Luo, K. Cen, Comparative analysis of CO₂ separation from flue gas by membrane gas absorption technology and chemical absorption technology in China, Energy Convers. Manage., 49 (2008) 3188–3197.

- [15] L. Zhao, R. Menzer, E. Riensche, L. Blum, D. Stolten, Concepts and investment cost analyses of multi-stage membrane systems used in post-combustion processes, *Energy Procedia*, 1 (2009) 269–278.
- [16] M. Harasimowicz, P. Orluk, G. Zakrzewska-Trznadel, A.G. Chmielewski, Applications of polyimide membranes for biogas purification and enrichment, *J. Hazard. Mater.*, 144 (2007) 698–702.
- [17] J. Davidson, K. Thambimuthu, Technologies for Capture of Carbon Dioxide, Proceedings of the Seventh Greenhouse Gas Technology Conference, Vancouver, Canada, International Energy Association (IEA), Greenhouse Gas R&D Programme, 2004.
- [18] K. Janusz-Szymańska, A. Dryjańska, Possibilities for improving the thermodynamic and economic characteristics of an oxy-type power plant with a cryogenic air separation unit, *Energy*, 85 (2015) 45–61.
- [19] R. Abdulrahman, I. Sebastine, Natural gas dehydration process simulation and optimization: a case study of Khurmala Field in Iraqi Kurdistan Region, *World Academy of Science, Eng. Technol.*, 78 (2013) 469–472.
- [20] M. Pronobis, Modernization of Power Boilers, WNT, Warszawa, 2002 (in Polish).
- [21] R.W. Baker, K. Lokhandwala, Natural gas processing with membranes: an overview, *Ind. Eng. Chem. Res.*, 47 (2008) 2109–2121.
- [22] T.S. Chung, L. Ying Jiang, Y. Lia, S. Kulprathipanja, Mixed matrix membranes (MMMs) comprising organic polymers with dispersed inorganic fillers for gas separation, *Prog. Polym. Sci.*, 32 (2007) 483–507.
- [23] H. Feng, H. Zhang, L. Xu, Polymeric membranes for natural gas conditioning, *Energy Sources Part A*, 29 (2007) 1269–1278.
- [24] C.V. Funk, D.R. Liyod, Zeolite-filled microporous mixed matrix membranes: prediction of gas separation performance, *J. Membr. Sci.*, 313 (2008) 224–231.
- [25] M. Netusil, P. Ditzl, Comparison of methods for dehydration of natural gas stored in underground gas storages, *Inz. Ap. Chem.*, 49 (2010) 87–88.
- [26] H. Sijbesema, K. Nymeyer, R. Marwijk, R. Heijboer, J. Potreck, M. Wessling, Flue gas dehydration using polymer membranes, *J. Membr. Sci.*, 313 (2008) 263–276.
- [27] D. Bergmair, S.J. Metz, H.C. de Lange, A.A. van Steenhoven, System analysis of membrane facilitated water generation from air humidity, *Desalination*, 339 (2014) 26–33.
- [28] M. Kasperkowiak, J. Kołodziejek, B. Strzemieska, A. Voelkel, Effect of impregnating agent and relative humidity on surface characteristics of sorbents determined by inverse gas chromatography, *J. Chromatogr. A*, 1288 (2013) 101–104.
- [29] K.-J. Huang, S.-J. Hwang, W.-H. Lai, The influence of humidification and temperature differences between inlet gases on water transport through the membrane of a proton exchange membrane fuel cell, *J. Power Sources*, 284 (2015) 77–85.
- [30] Q.-f. Jian, G.-q. Ma, X.-l. Qiu, Influences of gas relative humidity on the temperature of membrane in PEMFC with interdigitated flow field, *Renewable Energy*, 62 (2014) 129–136.
- [31] M. Giacinti Baschetti, M. Minelli, J. Catalano, G.C. Sarti, Gas permeation in perfluorosulfonated membranes: influence of temperature and relative humidity, *Int. J. Hydrogen Energy*, 38 (2013) 11973–11982.
- [32] M.A. Díaz, A. Iranzo, F. Rosa, F. Isorna, E. Lopez, J.P. Bolivar, Effect of carbon dioxide on the contamination of low temperature and high temperature PEM (polymer electrolyte membrane) fuel cells. Influence of temperature, relative humidity and analysis of regeneration processes, *Energy*, 90 (2015) 299–309.
- [33] J.R. Pauls, D. Fritsch, T. Klassen, K.-V. Peinemann, Gas permeation measurement under defined humidity via constant volume/variable pressure method, *J. Membr. Sci.*, 389 (2012) 343–348.
- [34] S. Naudy, F. Collette, F. Thominet, G. Gebel, E. Espuche, Influence of hygrothermal aging on the gas and water transport properties of Nafion® membranes, *J. Membr. Sci.*, 451 (2014) 293–304.
- [35] L. Ansaloni, M. Minelli, M. Giacinti Baschetti, G.C. Sarti, Effect of relative humidity and temperature on gas transport in Matrimid®: experimental study and modeling, *J. Membr. Sci.*, 471 (2014) 392–401.
- [36] S. Vengatesan, K. Panha, M.W. Fowler, X.-Z. Yuan, H. Wang, Membrane electrode assembly degradation under idle conditions via unsymmetrical reactant relative humidity cycling, *J. Power Sources*, 207 (2012) 101–110.