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NUMERICAL INTERPRETATION OF A MATHEMATICAL MODEL OF MEMBRANE GAS SEPARATION WITH ENERGY TRANSFER BY GAS FLOWING IN A CHANNEL TO GAS PENETRATING THIS CHANNEL FROM THE ADJACENT CHANNEL

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Comparative calculations with a mathematical model designed by the authors, which takes into consideration energy transfer from gas flowing through a given channel to gas which penetrates this channel from an adjacent channel, as well as a model which omits this phenomenon, respectively, were made for the process of separating gas mixtures carried out with an inert sweep gas in the four-end capillary membrane module. Calculations were made for the process of biogas separation using a PMSP polymer membrane, relative to helium as the sweep gas. It was demonstrated that omitting the energy transfer in the mathematical model might lead to obtaining results which indicate that the capacity of the process expressed by the value of feed flux subjected to separation is by several percent higher than in reality.

Keywords: gas separation, membrane, mathematical model

1. INTRODUCTION

Separation of gas mixture components is one of the most important tasks faced by the industrial practice. In particular, it refers to obtaining pure nitrogen and pure oxygen from air, purifying natural gas, obtaining helium from natural gas, obtaining hydrogen from the products generated in the gasification process or removing carbon dioxide from biogas. Formerly, adsorption, absorption and cryogenic methods were mainly used to complete these industrial tasks (Kerry, 2007). Over the last three decades, membrane techniques were added to the above mentioned methods of separation. With the development of polymer manufacturing technology, polymer membranes become now the main method used in the processes of gas mixture separation.

Along with the growing number of industrial processes of gas mixture separation using nonporous polymer membranes, literature started to present mathematical models of this separation process. Some of the first authors who made an attempt to model these processes were Rautenbach and Dahm (1986) as well as Pan (1986). Henceforth, the appearance of further mathematical models, which vary in terms of taken assumptions as well as inclusion or exclusion of selected effects or phenomena, may be observed in the literature. Some relevant examples are presented below.

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From the point of view of this study, which takes into account pressure drops of flowing gas streams caused by energy transfer between relevant streams, literature publications containing assumptions on pressure consistency or variability in the feed and permeate channel along the module, respectively, are significant above all. Changes in pressures in both channels are included in the models of Kovvali et al. (1992), Makaruk and Harasek (2009), Marriot and Sørensen (2003). Cocker et al. (1998), Kundu et al. (2013) and Thundyil and Koros (1997) assume changes of pressure in the permeate channel with no changes of pressure in the feed channel. In contrast, Smith et al. (1996), Tessendorf et al. (1999) as well as Morgues and Sanches (2005) assume no changes in any channels.

As regards research areas other than studies on pressure variability or consistency in the feed and permeate channels along the module, is noticeable that most authors assume no axial mixing in the feed and permeate channels. This phenomenon included in a model may be found in the study by Kovvali et al. (1992). A phenomenon of concentration polarization is examined only by few authors, for example Marriott and Sørensen (2003), Scholz et al. (2012) and Wang et al. (2002). The authors of the last mentioned study, i.e. Scholz et al. (2012), investigate also thermal effects occurring in this process, which are neglected by most other authors. In their studies, Davis (2002) as well as Makaruk and Harasek (2009) accept the variability of membrane parameters depending on the conditions in which the process is carried out.

Accepting diverse assumptions and applying different simplifications in formulating the mathematical model of membrane process by the authors of publications is also frequent in modeling membrane processes other than membrane separation of gas mixtures, such as pervaporation (Brun et al., 1985), microfiltration (Szwast et al., 2013), ultrafiltration (Tekić et al., 1996), and reverse osmosis (Kim and Hoek, 2005).

Attention should be paid to publications which include comparative results of calculations obtained by applying a model which takes into account a selected phenomenon and a model which omits this phenomenon, respectively. Based on such analysis, it is possible to specify how significant the inclusion of the selected phenomenon is for the accuracy of calculations. The above mentioned study by Kim and Hoek (2005) may serve as an example in this regard. They analyze diverse mathematical models which describe concentration polarization occurring in reverse osmosis. Results of calculations obtained from the analytical film theory model and convection-diffusion model, respectively, were compared. The first model describes one-dimensional (normal to the surface of the membrane) flow of a dissolved component through a stagnant layer of liquid near the membrane surface, neglecting the movement of the dissolved component near the membrane surface in the axial direction, whereas the second model describes two-dimensional (normal and tangent to the surface of the membrane) movement of mass, thus introducing an assumption on movement of the dissolved component along the membrane near its surface. It turned out that the results of calculations obtained by applying each of these models respectively differ inconsiderably. Therefore, introducing the additional assumption on the movement of the dissolved component along the membrane near its surface to the model did not have a quantitative impact on the obtained results. This means that the impact of the component movement in the direction along the membrane surface may be omitted in the models which do not have to be used for precise calculations. Such conclusion is obviously also valuable from the cognitive point of view.

When analyzing the literature models of the membrane gas separation process, the authors of this study did not encounter models which take into account, at least apparently, the energy required for the transport of mass fluxes through the membrane. Therefore, they suggested their own mathematical model (Szwast and Szwast, 2015) which, in their view, fills the gap. The authors (Szwast and Szwast, 2015) assumed that the pressure drop on the elementary length dw of the feed channel results from the flow resistance and the transfer of some pressure energy by the flowing gas stream (ability to execute technical work) to give pressure energy and kinetic energy to the elementary sweep gas stream which penetrated from the permeate channel, through the membrane, into that channel. This model assumes that gas at the exit of the membrane has zero pressure energy and zero kinetic energy, whereas the energy of gas entering the mem-

brane is dispersed and as such it is omitted in the discussions on the isothermal process. Similarly, pressure drop on the elementary length of the permeate channel results from the flow resistance and the transfer of some pressure energy through the flowing gas stream to give pressure energy and kinetic energy to the separated gas streams which penetrated from the feed channel. Additionally, the following assumptions are adopted: flows of gases in the feed and permeate channels are plug flows, gas is perfectly mixed in the cross-section, the process is isothermal, gas is described by the equation of ideal gas state.

Authors in the previous study (Szwast and Szwast, 2015) presented the formulation of relevant equations for diverse geometries of the membrane module as well as diverse methods of carrying out the process. This study aims at presenting numerical interpretation of the model proposed therein and comparing the results of calculations obtained by applying this model with the results of calculations obtained by applying the model which does not take into account energy transfer. The considerations taken here are limited to the selected module, namely the capillary module.

2. MATHEMATICAL MODEL OF THE PROCESS IN THE CAPILLARY MEMBRANE MODULE

This study focuses on the capillary membrane module. Figure 1 presents a fragment of the module longitudinal section made for the elementary length dw of the module, related to a single capillary membrane and its immediate surroundings. The entrance fluxes of the feed and the sweep gas, respectively, may be co-current (all fluxes are then marked with a continuous line) or counter-current (fluxes of sweep gas and permeate are then marked with a dotted line).

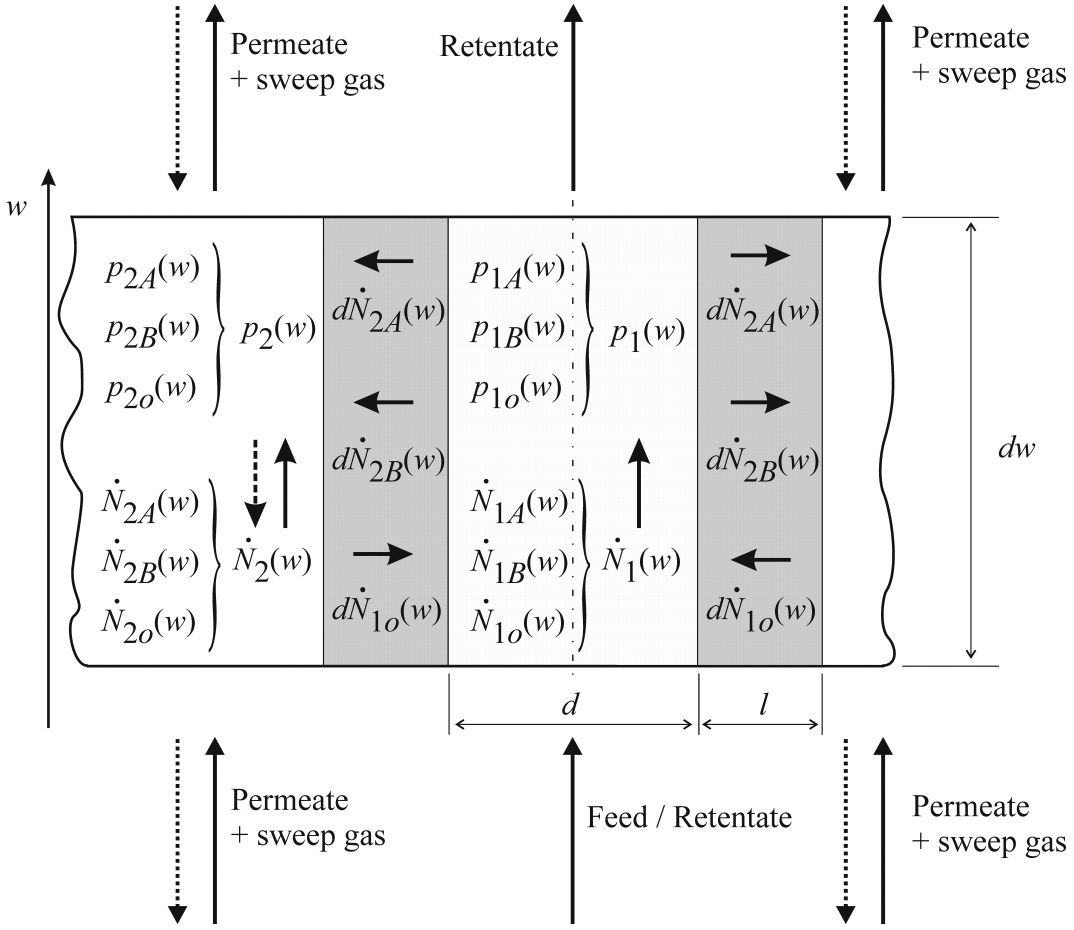

Fig. 1. Fragment of the capillary module longitudinal section related to a single capillary membrane and its immediate surroundings made for the elementary length dw of the module

The feed channel is the sum of inner volumes of n tube membranes placed in the capillary module, whereas the permeate channel is the outer-tubular space. Each tubular membrane is of inside diameter d and thickness l and the inside diameter of the capillary membrane module D which limits the permeate channel. It is assumed that the length of the channels w is always measured from the section of the feed entrance of its channel. It applies to the feed channel as well as the permeate channel, but as for the permeate channel, the assumption applies both to co-current and counter-current flows.

Subscript 1 is assigned to symbols related to the feed channel and subscript 2 is assigned to symbols related to the permeate channel. Additionally, subscript 1 is assigned to the mass flows which penetrated the feed channel (from the permeate channel) and subscript 2 is assigned to the mass flows penetrating in the opposite direction, i.e. to the permeate channel (from the feed channel). Subscripts 1 and 2 are generally represented by subscript j.

The feed is the mixture of A and B components which are to be separated, while the retentate is the part of the feed which did not penetrate the membrane to the permeate channel and usually takes a small amount of sweep gas which penetrated the membrane from the permeate channel, while the permeate is the part of the feed which penetrated the membrane to the permeate channel and is carried by the sweep gas. Inlet of the pure sweep gas is located at the entrance of the permeate channel.

Below there are eleven equations – Eqs. (1)–(11) – which constitute a mathematical model of the two-component gas mixture separation process carried out with the inert sweep gas in the four-end capillary membrane module. These equations were derived in the previous study of the authors (Szwast and Szwast, 2015).

Drop in the molar flux value of the component $i = A, B$ along the feed channel – Eqs. (1) and (2) – on the elementary length dw equals, in relation to the absolute value, the molar flux of this component transport through the elementary surface dF of the membrane and increase in the value of the molar flux of this component along the length of the permeate channel (Yampolskii et al., 2006). By contrast, increase in value of the molar flux of sweep gas $i = o$ along the feed channel – Eq. (3) – on the elementary length dw equals, in relation to the absolute value, the molar flux of this component transport through the elementary surface dF of the membrane and drop in the value of the molar flux of this gas along the length of the permeate channel. Similar relationships are presented in relation to the permeate channel – Eqs. (4)–(6)

$$\frac{d\dot{N}_{1A}}{dw} = -P_A \frac{p_{1A}(w) - p_{2A}(w)}{l} \frac{2\pi l \cdot n}{\ln\left(1 + \frac{2l}{d}\right)} \tag{1}$$

$$\frac{d\dot{N}_{1B}}{dw} = -P_B \frac{p_{1B}(w) - p_{2B}(w)}{l} \frac{2\pi l \cdot n}{\ln\left(1 + \frac{2l}{d}\right)} \tag{2}$$

$$\frac{d\dot{N}_{1o}}{dw} = P_o \frac{p_{2o}(w) - p_{1o}(w)}{l} \frac{2\pi l \cdot n}{\ln\left(1 + \frac{2l}{d}\right)} \tag{3}$$

$$\frac{d\dot{N}_{2A}}{dw} = \pm P_A \frac{p_{1A}(w) - p_{2A}(w)}{l} \frac{2\pi l \cdot n}{\ln\left(1 + \frac{2l}{d}\right)} \tag{4}$$

$$\frac{d\dot{N}_{2B}}{dw} = \pm P_B \frac{p_{1B}(w) - p_{2B}(w)}{l} \frac{2\pi l \cdot n}{\ln\left(1 + \frac{2l}{d}\right)} \tag{5}$$

$$\frac{d\dot{N}_{2o}}{dw} = \mp P_o \frac{p_{2o}(w) - p_{1o}(w)}{l} \frac{2\pi l \cdot n}{\ln\left(1 + \frac{2l}{d}\right)} \tag{6}$$

where, if a double sign $\pm$ or $\mp$ occurs, the upper character refers to co-current flows and the lower character refers to counter-current flows. The second fraction on the right side of Eqs. (1)–(6) defines the effective perimeter of the tubular membrane calculated as the logarithmic mean of the inner and outer perimeters. Multiplication of the perimeter by dw defines the effective elementary surface area of the membrane, whereas the partial pressure is calculated according to Equation (7) applicable in the same way to all module tubes.

$$p_{ji}(w) = p_j(w)\frac{\dot{N}_{ji}(w)}{\dot{N}_{jA}(w)+\dot{N}_{jB}(w)+\dot{N}_{jo}(w)} \qquad \text{for } j=1,2 \text{ and } i=A,B,o \tag{7}$$

The essence of the model proposed by the authors (Szwast and Szwast, 2015), which takes into account energy transfer from gas flowing through a given channel to gas which penetrates this channel from an adjacent channel, is contained in Eqs. (8) and (9) that describe the elementary drop in the total pressure dp_1 on the elementary length dw of the feed channel as well as elementary drop in the total pressure dp_2 in the permeate channel, respectively. The essence of this model contained in Eqs. (8) and (9) will be discussed below Eq. (11).

$$\frac{dp_1}{dw} = -\lambda_1\frac{\overline{u_1}^2\overline{\rho_1}}{2d_1} - \frac{p_1}{l}\frac{2\pi l\cdot n}{\ln\left(1+\dfrac{2l}{d_w}\right)}\frac{P_o\left[p_{2o}(w)-p_{1o}(w)\right]}{\dot{N}_{1A}+\dot{N}_{1B}+\dot{N}_{1o}} + \\ -\frac{\overline{\rho_1 u_1}^2}{2l}\frac{2\pi l\cdot n}{\ln\left(1+\dfrac{2l}{d_w}\right)}\frac{P_o\left[p_{2o}(w)-p_{1o}(w)\right]}{\dot{N}_{1A}+\dot{N}_{1B}+\dot{N}_{1o}} \tag{8}$$

$$\frac{dp_2}{dw} = \mp\lambda_2\frac{\overline{u_2}^2\overline{\rho_2}}{2d_2} \mp \frac{p_2}{l}\frac{2\pi l\cdot n}{\ln\left(1+\dfrac{2l}{d_w}\right)}\frac{P_A\left[p_{1A}(w)-p_{2A}(w)\right]+P_B\left[p_{1B}(w)-p_{2B}(w)\right]}{\dot{N}_{2A}+\dot{N}_{2B}+\dot{N}_{2o}} + \\ \mp\frac{\overline{\rho_2 u_2}^2}{2l}\frac{2\pi l\cdot n}{\ln\left(1+\dfrac{2l}{d_w}\right)}\frac{P_A\left[p_{1A}(w)-p_{2A}(w)\right]+P_B\left[p_{1B}(w)-p_{2B}(w)\right]}{\dot{N}_{2A}+\dot{N}_{2B}+\dot{N}_{2o}} \tag{9}$$

If a double sign $\mp$ occurs, the upper character refers to co-current flows and the lower character refers to counter-current flows. The average linear velocity of gas mixture is calculated with Equation (10) and the average density of gas mixture with Equation (11).

$$\overline{u_j}(w) = \frac{\left[\dot{N}_{jA}(w)+\dot{N}_{jB}(w)+\dot{N}_{jo}(w)\right]RT_j}{p_j(w)A_j} \tag{10}$$

$$\overline{\rho_j}(w) = \frac{p_j(w)}{RT_j}\overline{M_j}(w) = \frac{p_j(w)}{RT_j}\frac{\dot{N}_{jA}(w)M_A+\dot{N}_{jB}(w)M_B+\dot{N}_{jo}(w)M_o}{\dot{N}_{jA}(w)+\dot{N}_{jB}(w)+\dot{N}_{jo}(w)} \tag{11}$$

When getting back to the essence of the authors' model (Szwast and Szwast, 2015), which takes into account the phenomenon of energy transfer from gas flowing through a given channel to gas which penetrates this channel from an adjacent channel, three expressions occur on the right sides of Eqs. (8) and (9), respectively. They are discussed below.

The first expressions on the right sides of Eqs. (8) and (9) describe, in relation to the elementary length of the channel dw, elementary drops $-(dp_j)'$ in the pressure of gas flowing in a given channel caused by flow resistance. They are described by the Darcy–Weissbach's equation.

The second expressions on the right sides of Eqs. (8) and (9) describe elementary drops $-(dp_1)''$ in the pressure of gas flowing in the channel j caused by the fact that this gas gives back part of its pressure

energy to the gas which penetrates this channel to give pressure to the last mentioned one in a given section of the discussed channel. With reference to the second expression on the right side of Eq. (8), the flux $\dot{N}_{1A}+\dot{N}_{1B}+\dot{N}_{1o}$ flowing in the feed channel suffers pressure drop $-(dp_1)''$ owing to which the sweep gas flux penetrating the feed channel $d\dot{N}_{1o}$ obtains pressure p_1 within the discussed section of this channel (as emphasized earlier, the assumption for this model is that gas at the exit of the membrane has zero pressure energy and zero kinetic energy). It is described by Eq. (12)

$$-(dp_1)''\left(\dot{N}_{1A}+\dot{N}_{1B}+\dot{N}_{1o}\right)=p_1 d\dot{N}_{1o} \tag{12}$$

which, after considering Eq. (3) leads to an elementary description of pressure drop $-(dp_j)''$ on the elementary length dw of the feed channel, $-(dp_1)''/dw$, is the same expression as the second expression on the right side of Eq. (8). The second expression on the right side of Eq. (9) is formed similarly. The permeate channel is penetrated by components A and B simultaneously and that the right sides of Eqs. (4) and (5) should be considered with a sign "+" or "−" for co-current flows and counter-current flows, respectively.

The third expressions on the right sides of Eqs. (8) and (9) describe elementary drops dp_j caused by the fact that gas flowing in the channel j gives back some of its pressure energy to gas which penetrates this channel to give the latter one linear velocity which the gas running in the channel has in a given section of the discussed channel. Deliberations on the forms of the third expressions on the right sides of Eqs. (8) and (9) should be conducted similarly to deliberations on the second expressions, but they should be started from Eq. (13) rather than Eq. (12).

$$-(dp_1)'''\left(\dot{N}_{1A}+\dot{N}_{1B}+\dot{N}_{1o}\right)=\frac{\rho_1\overline{u_1}^2}{2}d\dot{N}_{1o} \tag{13}$$

Placing the second and third expressions on the right sides of Eqs. (8) and (9) is a novelty compared to the previous mathematical models described in the literature on the subject.

The model being the reference in comparisons presented in this study, that is the model which omits energy transfer between the stream of gas flowing in a given channel and the stream which penetrates this channel through the membrane, is formulated in a similar way. However, Eqs. (8) and (9), which describe pressure drops along the channel length, contain only the first component out of three on the right side of these equations.

3. COMPUTATIONAL METHODS

A numerical comparative analysis was carried out for the four-end module which contains $n = 20$ capillary tubes with the active length of membranes $w_k = 1.2$ m, their inner diameter $d = 1$ mm and thickness adopted for the calculations $l = 1$ μm. It was also assumed that these capillary tubes are placed in a vessel in the form of a pipe with inner diameter $D = 20$ mm. The four-end module is adapted to carry out the process with the sweep gas. The feed, in the discussed case, is put into the capillary tubes and the sweep gas washes the membranes on their outer sides (bore side feed). The authors use such module geometry, created in accordance with their own technology (Szwast et al., 2008, Szwast et al., 2017), in their experimental research.

Calculations were made using data related to the process of biogas component separation with the initial composition 60%$_{wt}$ CH_4 and 40%$_{wt}$ CO_2, which, when rounded up, translates into the molar ratio 4:1. Helium parameters were adopted as the parameters of sweep gas and parameters of PMSP (poly[l-(trimethylsilyl)-l-propyne]), of which permeability for individual gas is: $P_{CH_4} = 4190$ barrer, $P_{CO_2} = 18000$ barrer, $P_{He} = 4100$ barrer (Takada et al., 1985), were assumed as the parameters of the membrane. This membrane, if commercially available, could be successfully used to separate biogas components. Higher

permeance of CO_2, compared with CH_4, which as an unwanted component may be easily removed from the biogas stream, supports this fact.

Calculations were made for the operation of the module in the steady state with boundary conditions set partially at the beginning and partially at the end of the module. It was assumed that the feed is fed under absolute pressure 1.5×10^6 Pa (15 bar_{abs}) and throttled with a valve at the exit of the retentate to the value of 1.2×10^6 Pa (12 bar_{abs}). With reference to the sweep gas, it was also assumed that it is put into the permeate channel with a fixed pressure (fixed at a little higher level than the atmospheric pressure which is discussed further in the text) and put out to the atmosphere as permeate, where the pressure is 0.1×10^6 Pa (1 bar_{abs}). Pressures in all four endings are then fixed, that is Eqs. (8) and (9) have boundary conditions set on both sides. It should be strongly emphasized that for the specific module and specific gas mixture, i.e. the defined permeability features of the membrane and for the defined pressures on all four endings of the module the capacity of this module, expressed by the feed flux, results from the above adopted assumptions and is not a value imposed by a designer or an operator of equipment. The feed stream entering the module is therefore a free quantity, while this freedom is limited only to one of two feed components. Let us assume that the freedom concerns the component A. Then $\dot{N}_{1A}(0)$ is a free quantity. The value of $\dot{N}_{1B}(0)$ is calculated from the composition of feed. Since there is no sweep gas component in the feed, $\dot{N}_{1o}(0) = 0$. The same applies to the sweep gas flux led to the permeate channel, which means that the value of this flux results from the adopted assumptions rather than an imposed value. The quantity $\dot{N}_{2o}(0)$ is free, and the absence of components A and B in the sweep gas causes $\dot{N}_{2A}(0) = \dot{N}_{2B}(0) = 0$. In turn, all fluxes at outlets of both module channels are free quantities.

In order to make calculations, software in Delphi was prepared solving the set of eight differential equations, Eqs. (1)–(6) and (8)–(9) as well as three accompanied algebraic, Eqs. (7) and (10)–(11). Numerical integration of differential equations takes place according to the 4th order Runge–Kutta method. This method of integrating is commonly accepted for solving problems related to the membrane gas separation (Hwang and Thorman, 1980; Li et al, 1990; Tsuru and Hwang, 1995).

Due to the fact that some of the set boundary conditions refer to the beginning and some to the end of the process, calculations are made with the iteration method, which means that values of appropriate quantity are assumed and then verified. Below a calculation procedure will be presented for separation of mixture with set composition, with fixed pressure at the entrance of the feed and at the entrance of the sweep gas as well as fixed pressure at the exit of these streams.

To begin the calculations related to co-current flows, two values should be assumed, i.e. the value of the flux of one component from the two-component feed with the set composition at the entrance of the feed channel and the value of the sweep gas flux at the entrance of the permeate channel. As the composition of two-component feed is known, such assumption of the value of one of the feed flux components results in the flux of the feed being fully described, obviously for the time of specific iteration. When the calculation algorithm is used for such assumed values of the discussed quantities, calculation values of the pressure of retentate and the pressure of permeate leaving the module as well as calculation values of molar fluxes of all three components of retentate and permeate which leave the module, are obtained. Obtained calculation values are obviously functionally dependent on the assumed value of flux of one component from the two-component feed with the set composition at the entrance of the feed channel as well as the value of the sweep gas flux at the entrance of the permeate channel. Then calculation values of the pressure of retentate and the pressure of permeate leaving the module should be compared with the fixed values for these pressures. The analysis of such comparisons makes it possible to assume appropriate values for the next iteration, that is to assume a new value of the flux of one component from the two-component feed with the set composition at the entrance of the feed channel as well as the value of the sweep gas flux at the entrance of the permeate channel. The calculation procedure is completed when calculation consistencies and fixed pressures at the retentate and permeate exit are obtained, respectively.

Calculations were made both for the co-current flows and counter-current flows. However, due to similarities between the conclusions resulting from the analysis of the results obtained for both systems, the authors decided not to discuss the results related to the counter-current flows and thus not to discuss how they are obtained.

The exemplary solution to the problem with two-point boundary conditions in another process of chemical engineering, i.e. in the process of absorption with chemical reaction is described in the study of Zarzycki and Chacuk (1993).

4. RESULTS AND DISCUSSION

This chapter is dedicated to the analysis of calculation results for co-current flows obtained for the model derived by the authors which takes into consideration energy transfer between appropriate mass streams and the reference model which omits such energy transfer, respectively.

The calculation procedure discussed above requires starting the calculations upon setting values of all boundary pressures. The set values of feed pressure at the entrance of its channel and retentate pressure at the exit of this channel as well as the values of permeate pressure at the exit of its channel are presented in the previous chapter. Thus, all that remains is the issue of setting values for sweep gas pressure at the entrance of the permeate channel.

Putting sweep gas into the membrane module aims at decreasing partial pressure of the separated components of the feed in the permeate channel which penetrated through the membrane from the feed channel to the permeate channel (Franz et al., 2013). This may take place only by washing out these components from the permeate channel by sweep gas. Intuitively, it does not require using high overpressures of sweep gas at the entrance of the permeate channel, compared to the atmospheric pressure which the permeate leaving the channel has. Therefore, several series of calculations were made. Increasingly higher pressure of sweep gas at the entrance of the permeate channel was assumed in each subsequent series. The lowest pressure taken for calculations had the value of 102 000 Pa (1.02 bar_{abs}), and each subsequent pressure was higher by 2000 Pa. Calculation results obtained using the model which did not take account of energy transfer had a physical sense for all the analyzed pressures of sweep gas at the entrance of the permeate channel. In contrast, calculation results obtained using the author's model, i.e. the model which take account of energy transfer between appropriate gas streams, had a physical sense only when the pressure of sweep gas at the entrance of the permeate channel was not lower than 114 000 Pa (1.14 bar_{abs}). The interpretation of this fact seems obvious. The model derived by the authors takes into consideration transferring some of the energy of the gas flowing in the channel to the gas which penetrates this channel to obtain kinetic energy by the latter and, at the same time, washing out the gas which penetrates the channel, and finally, completing the task set for the sweep gas.

Below there is a graphical analysis of calculation results made using each of the discussed models for the value of the pressure of sweep gas at the entrance of the permeate channel equal to 114 000 Pa (1.14 bar_{abs}), that is the value for which calculation results made using each model have a physical sense. The subsequent graphs dedicated to the analysis of calculation results present results obtained for the author's model marked by continuous lines and results obtained for the reference model marked by dotted lines.

The analysis of total pressure variability calculated with both models compared, respectively, shows no essential differences between the trajectories of the total pressure variability in the feed channel as well as an insignificant, yet noticeable, difference in trajectories of the total pressure variability in the permeate

channel (Fig. 2a). Such conclusion was expected due to the boundary conditions set on both sides for both channels of the module, which were the same for calculations using each of the discussed models.

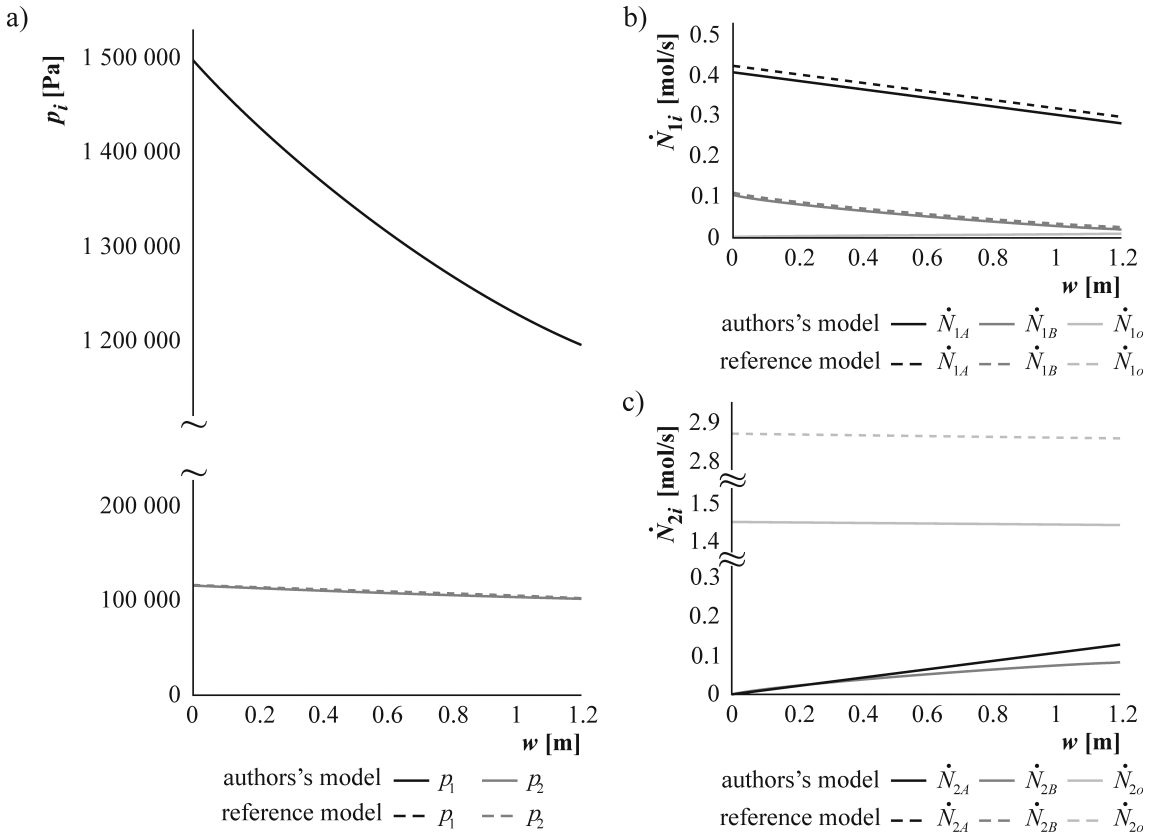

Fig. 2. Trajectories of variability along the membrane module: a) of pressures in the feed and permeate channels, b) of molar fluxes of individual components in the feed channel, c) of molar fluxes of individual components in the permeate channel

Figures 2b and 2c, respectively, present trajectories of molar flux variability of individual gases, *A*, *B* and *o*, in the feed and permeate channels. In the feed channel – Fig. 2b – flux values of mixture components subjected to separation, *A* and *B*, calculated with each compared models, differ between each other considerably, with higher values of these fluxes (dotted line) being obtained for the reference model. Values of sweep gas fluxes, *o*, in the permeate channel – Fig. 2c – show a considerable difference, with a considerably higher value of this flux (dotted line) being obtained for the reference model, i.e. the same as in relation to gas fluxes *A* and *B* in the feed channel. The observed differences in flux values referred to above should be explained in the following way. With fixed pressures on both ends of each module channel the total pressure loss is determined along the channel, which is the same for both models. However, in the case of the authors' model, the total pressure loss results both from the flow resistance and energy transfer between the streams (see Eq. 8 or Eq. 9), while in the case of the reference model, the same total pressure loss results only from the flow resistance (see Eq. 8 or Eq. 9 with the last two components on the right side of equation omitted). Pressure losses related to flow resistance on the entire length of the channels must then be lower, in relation to the authors' model, than the respective losses related to the reference model. This is possible when calculations using the authors' model refer to lower molar fluxes which supply the appropriate module channels than the appropriate supplying fluxes in relation to the reference model.

To present the precision of the results obtained during iteration calculations as well as the exact differences between the results of calculations made respectively with each of the discussed models, three tables containing data related to both ends of each membrane module channel have been drawn up. Each table refers to different pressure of sweep gas at the entrance of the permeate channel. Table 1, refers to pressure

of 114 000 Pa, that is pressure for which the calculation results are presented in Fig. 2. The other two tables refer to higher sweep gas pressures at the entrance of the permeate channel, i.e. 118 000 Pa and 120 000 Pa, respectively.

The iteration method used in the presented problem aims at selecting the value of flux of one component from the two-component feed with a set composition at the entrance of the feed channel as well as the value of sweep gas flux at the entrance of the permeate channel as to obtain calculation values of pressures at the ends of both channels equal to the value set for these pressures, here it is 1 200 000 Pa and 100 000 Pa, respectively. This objective was achieved with the accuracy for all decimal digits.

The data contained in Table 1 allow to determine absolute and relative differences between the values of quantities which are essential for the process, i.e. values of the feed fluxes and sweep gas at the entrance of the appropriate module channels.

Table 1. Comparison of the calculation results for the sweep gas pressure of 114 000 Pa

	Authors' model		Reference model	
	Inlet $w = 0$	Outlet $w = w_k$	Inlet $w = 0$	Outlet $w = w_k$
p_1 [Pa]	1 500 000	1 200 000	1 500 000	1 200 000
p_2 [Pa]	114 000	100 000	114 000	100 000
$\dot{N}_{1A}$ [mol/s]	0.3963	0.2748	0.4130	0.2916
$\dot{N}_{1B}$ [mol/s]	0.0991	0.0211	0.1033	0.0233
$\dot{N}_{1o}$ [mol/s]	0.0000	0.0086	0.0000	0.0091
$\dot{N}_{2A}$ [mol/s]	0.0000	0.1215	0.0000	0.1214
$\dot{N}_{2B}$ [mol/s]	0.0000	0.0780	0.0000	0.0799
$\dot{N}_{2o}$ [mol/s]	1.4500	1.4414	2.8707	2.8616

The flux of the feed at the entrance of its channel calculated with the use of the authors' model is $(0.3963 + 0.0991)$ mol/s $= 0.4954$ mol/s, while the flux calculated with the reference model is $(0.4130 + 0.1033)$ mol/s $= 0.5163$ mol/s. Thus, the absolute difference is $(0.5163 - 0.4954)$ mol/s $= 0.0209$ mol/s and the relative difference is equal to $(0.0209 \text{ mol/s} \times 100\%)/(0.4954 \text{ mol/s}) = 4.23\%$. As regards sweep gas at the entrance of the permeate channel, the corresponding absolute difference is $(2.8707 - 1.4500)$ mol/s $= 1.4207$ mol/s and the relative difference is $(1.4207 \text{ mol/s} \times 100\%)/(1.4500 \text{ mol/s}) = 97.98\%$.

The value of the relative difference for sweep gas is considerable higher than that of the feed, which results from the fact that – compared to the feed channel – a larger flux of gas, to which gas flowing in the channel gives back some of its energy, penetrates through the membrane to the permeate channel. This, according to the above described mechanism for the authors' model, translates into a considerably higher reduction in the value of the sweep gas flux which supplies the permeate channel than the reduction of the feed flux.

The results of calculations made with data in Table 2 and Table 3 lead to the conclusion that with the increase of the sweep gas pressure at the entrance of the permeate channel, the relative differences for the feed fluxes are higher, while in the case of the sweep gas fluxes they are lower. If the sweep gas pressure equals 118 000 Pa, the relative difference for the feed fluxes is 4.47% and for the sweep gas fluxes it is 23.82%. If the sweep gas pressure is 120 000 Pa, the relative difference for the feed fluxes is 4.54% and for the sweep gas fluxes it is 18.47%.

Table 2. Comparison of the calculation results for the sweep gas pressure of 118 000 Pa

	Authors' model		Reference model	
	Inlet $w = 0$	Outlet $w = w_k$	Inlet $w = 0$	Outlet $w = w_k$
p_1 [Pa]	1 500 000	1 200 000	1 500 000	1 200 000
p_2 [Pa]	118 000	100 000	118 000	100 000
$\dot{N}_{1A}$ [mol/s]	0.3954	0.27370	0.4130	0.2916
$\dot{N}_{1B}$ [mol/s]	0.0988	0.0206	0.1033	0.0233
$\dot{N}_{1o}$ [mol/s]	0.0000	0.0091	0.0000	0.0094
$\dot{N}_{2A}$ [mol/s]	0.0000	0.1217	0.0000	0.1214
$\dot{N}_{2B}$ [mol/s]	0.0000	0.0783	0.0000	0.0799
$\dot{N}_{2o}$ [mol/s]	2.7015	2.6924	3.3449	3.3355

Table 3. Comparison of the calculation results for the sweep gas pressure of 120 000 Pa

	Authors' model		Reference model	
	Inlet $w = 0$	Outlet $w = w_k$	Inlet $w = 0$	Outlet $w = w_k$
p_1 [Pa]	1 500 000	1 200 000	1 500 000	1 200 000
p_2 [Pa]	120 000	100 000	120 000	100 000
$\dot{N}_{1A}$ [mol/s]	0.3951	0.2734	0.4130	0.2916
$\dot{N}_{1B}$ [mol/s]	0.0988	0.0205	0.1033	0.0233
$\dot{N}_{1o}$ [mol/s]	0.0000	0.0093	0.0000	0.0095
$\dot{N}_{2A}$ [mol/s]	0.0000	0.1217	0.0000	0.1214
$\dot{N}_{2B}$ [mol/s]	0.0000	0.0783	0.0000	0.0799
$\dot{N}_{2o}$ [mol/s]	3.0105	3.0012	3.5665	3.5570

5. CONCLUSIONS

Comparative calculations with a mathematical model designed by the authors, which takes into consideration energy transfer from gas flowing through a given channel to gas which penetrates this channel from an adjacent channel, as well as a model which omits this phenomenon, respectively, were made for the process of separating gas mixtures carried out with an inert sweep gas in the four-end capillary membrane module.

It was demonstrated that omitting the process of energy transfer in the mathematical model might lead to obtaining results which indicate that the capacity of the process expressed by the value of feed flux subjected to separation is by several percent higher than in reality.

The numerical interpretation of the authors' model carried out in this study shows that it is necessary to include energy transfer between respective gas streams in the membrane module. It is the authors' model which makes it possible to describe the process of washing out permeate from its channel, i.e. the process to give kinetic energy to the gas flux which penetrates a given channel of the membrane module.

More significant differences occur in the values of the respective fluxes which supply the channels of the membrane module obtained for the concerned models, when the membrane used for the process is more permeable for separated components.

SYMBOLS

A_j cross section area of channel j, m
d capillary inner diameter, m
d_j equivalent diameter of channel j, m
D membrane module outer diameter, m
F membrane surface area, m^2
l membrane thickness, m
M_i molecular mass of component i, $kg \cdot mol^{-1}$
$\overline{M_j}$ average molecular mass of gas in channel j, $kg \cdot mol^{-1}$
n quantity of membranes in module, –
$\dot{N}_{ji}$ molar flow rate of component i in channel j, $mol \cdot s^{-1}$
p_j total pressure in channel j, Pa
p_{ji} component i partial pressure in channel j, Pa
P_i membrane permeability for component i, $mol \cdot m \cdot s^{-1} \cdot m^{-2} \cdot Pa^{-1}$, barrer
R gas constant, $J \cdot mol^{-1} \cdot K^{-1}$
T_j temperature in channel j, K
$\overline{u_j}$ average linear gas velocity in channel j, $m \cdot s^{-1}$
w distance of a given module section to the feed inlet section, m

Greek symbols

λ_i friction factor in channel j, –
$\overline{\rho_j}$ average gas density in channel j, $kg \cdot m^{-3}$

Subscripts

1, 2 feed channel and permeate channel, respectively
A, *B* components of feed mixture
o sweep gas

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