

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

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The paper presents the mathematical modelling of selected isothermal separation processes of gaseous mixtures, taking place in plants using membranes, in particular nonporous polymer membranes. The modelling concerns membrane modules consisting of two channels – the feeding and the permeate channels. Different shapes of the channels cross-section were taken into account. Consideration was given to co-current and counter-current flows, for feeding and permeate streams, respectively, flowing together with the inert gas receiving permeate. In the proposed mathematical model it was considered that pressure of gas changes along the length of flow channels was the result of both – the drop of pressure connected with flow resistance, and energy transfer by molecules of gas flowing in a given channel to molecules which penetrate this channel from the adjacent channel. The literature on membrane technology takes into account only the drop of pressure connected with flow resistance. Consideration given to energy transfer by molecules of gas flowing in a given channel to molecules which penetrate this channel from the adjacent channel constitute the essential novelty in the current study. The paper also presents results of calculations obtained by means of a computer program which used equations of the derived model. Physicochemical data concerning separation of the CO<sub>2</sub>/CH<sub>4</sub> mixture with He as the sweep gas and data concerning properties of the membrane made of PDMS were assumed for calculations.

**Keywords:** membrane, membrane module, reciprocal transfer of energy between gas molecules, changes of the pressure of gas along the length of flow channel

## 1. INTRODUCTION

The research presented in this paper concerns modelling of selected membrane processes applied for separation of gaseous mixtures based on nonporous polymer membranes. The modelling involved both simple membrane installations consisting of two channels – the feeding channel and the permeate channel, respectively, as well as membrane modules.

For purposes of focusing attention, consider Fig.1 which shows a scheme of a simple installation with a single tubular membrane, though the modelling of processes taking place in this installation will be discussed later.

Two channels shown in Fig.1 with cross-sections, circular of diameter  $d$  and the ring of internal diameter  $d+2l$  and external diameter  $D$ , respectively, are separated by a thin nonporous polymer membrane of thickness  $l$ . The flow of gas streams takes place in both channels, parallel to the

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membrane surface. The first channel (feeding channel), is supplied by a high-pressure gaseous mixture composed of components  $A$  and  $B$  (in general, a multicomponent mixture) which have to be separated. The second channel (permeate channel) is supplied by an inert gas. The flow of gases in parallel to the surface of the membrane is accompanied by the flow of some molecules of these gases directed across the wall of the membrane. Feeding components,  $A$  and  $B$ , are transported from the feeding channel in which their partial pressures are high, to the permeate channel. However, molecules of the inert gas flowing in the permeate channel receiving the latter, are transported usually in small quantities in the opposite direction, i.e. to the feeding channel.

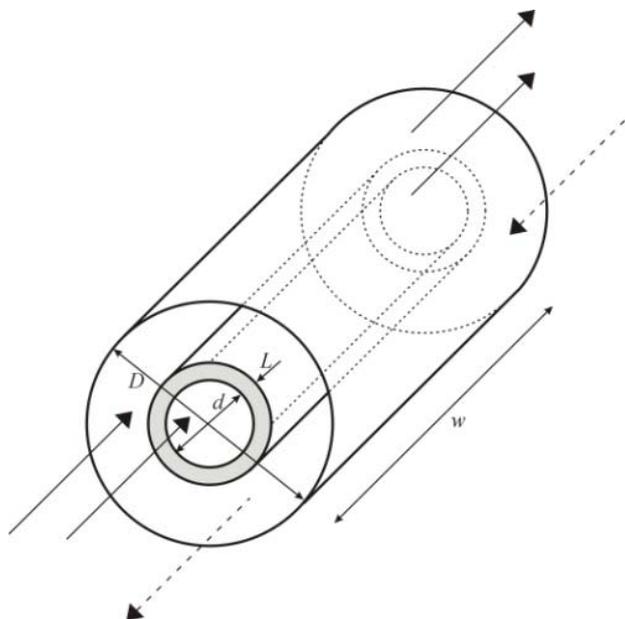


Fig. 1. Scheme of co-current and counter-current flows processes with a single tubular membrane.

The lower index 1 was assigned to symbols related to the feeding channel (feeding channels in the case of membrane modules), while the lower index 2 was related to the permeate channel. Furthermore, the lower index 1 was assigned to mass streams penetrating the feeding channel (from the permeate channel), while the lower index 2 was assigned to mass streams with opposite directions, i.e. streams passing to the permeate channel (from the feeding channel). The lower indexes 1 and 2 are generally denoted as the lower index  $j$ .

Both flows are considered – co-current and counter-current, for feeding and permeate, respectively, to flow together with the inert gas receiving permeate. Fig.1 shows the direction of the feeding flow through the internal channel of diameter  $d$  and the direction of the permeate flow through the external annular channel, respectively, for the co-current flow (continuous arrow) and for counter-current flow (dashed arrow). It was assumed that the length of channels,  $w$  was always measured from the cross-section of the feeding inlet to its channel. This assumption is valid for both feeding and permeate channels. Furthermore, as regards the permeate channel, this foundation is valid for both flows – co-current and counter-current. Isothermal processes with absolute mixing toward transverse direction to the direction of the gas flow and a lack of mixing toward the flow direction are assumed.

Different shapes of the channel cross-sections of channels have been taken into account. The hydraulic diameter of any channel  $j$  is calculated from the following general formula (Ciborowski, 1973).

$$d_j = 4r_{hj} \quad (1)$$

where hydraulic radius  $r_{hj}$  of any channel  $j$  is defined as a ratio of the cross-section of this channel  $A_j$  to its wet circumference  $O_j$ , hence:

$$d_j = 4r_{hj} = 4 \frac{A_j}{O_j} \quad (2)$$

The drop of value of the molar stream of the component  $i = A, B$  along the feeding channel, in the elementary section  $dw$  is, regarding the absolute value, equal to the molar stream of the transport of this component through the elementary area  $dF$  of the membrane and to the increase in the value of the molar stream of this component along the length of the permeate channel. These dependences are presented by Equation (3) (Yampolskii et al., 2006). Analogous dependence for the inert gas transported to the feeding channel from the permeate channel is presented by Equation (4).

$$-d\dot{N}_{1i} = P_i \frac{p_{1i} - p_{2i}}{l} dF = d\dot{N}_{2i} \text{ for } i = A, B \quad (3)$$

$$d\dot{N}_{1o} = P_o \frac{p_{2o} - p_{1o}}{l} dF = -d\dot{N}_{2o} \quad (4)$$

Creating a mathematical model of the discussed membrane process, the authors of this paper assumed that the pressure drop along the elementary length  $dw$  of the feeding channel results from flow resistances and from the delivery of a part of pressure energy (the ability of execution of technical work) by the flowing stream of gas to the elementary stream of inert gas  $d\dot{N}_{1o}$  penetrating this channel through the membrane from the permeate channel, which uses it to create its pressure energy as well and kinetic energy. In this model it is assumed that gas at the exit from the membrane has zero pressure energy and zero kinetic energy, while the energy of gas entering the membrane is dispersed and as such has been omitted in considerations of the isothermal process. Analogically, the pressure drop along the elementary length of the permeate channel overcomes of flow resistances and the delivery by flowing gas stream the part of its pressure energy to create pressure and kinetic energy of the streams  $d\dot{N}_{2A}$  and  $d\dot{N}_{2B}$  which penetrated from the feeding channel. Such an idea was presented by the authors elsewhere (Szwast and Szwast, 2013).

The mechanism of energy transfer from the gas stream flowing through the given channel to the elementary gas stream penetrating this channel (from the flanking channel) proposed by the authors and presented above, does not take into account the energy transfer being the result of decrease in kinetic energy of the flowing gas. Such an approach can lead to some inaccuracies in results of calculations obtained from the implemented model. On the other hand, the model is relatively simple. Therefore, the model of energy transfer proposed by the authors seems to be a reasonable compromise between accuracy and simplicity of the model.

In the literature on membrane processes it is possible to find many practical formulas based on simplified mathematical models.

Modelling of the gaseous components separation process by means of membranes has been conducted in the past, when the development of polymer materials was not so advanced as currently. One of the first papers concerning this issue was the paper of Rautenbach and Dahm (1986) representing a simplified approach to the process of selective transport of components for binary gaseous mixtures. Equations presented in this work describe the isothermal separation of gaseous components and are valid in the light of the following prevailing assumptions: negligible pressure losses at the feed side, negligible axial diffusion in the feed and permeate (no backmixing), plug flow and plug-like concentration profiles at the feed and at the permeate side. After a few years, this model underwent some minor corrections as presented in the paper by Smith et al. (1996).

Similar preliminary assumptions as in the work of Rautenbach and Dahm (1986) were adopted by Coker et al. (1999). In this article considering a multicomponent system based on the example of

hollow fibre membranes, the authors resigned from the assumption that the process is isothermal. They assumed instead that the temperature of the separated gas could change resulting from gas expansion.

A similar way of approach to addressing the problem of membrane separation of gases was applied by Makaruk and Harasek (2009) and Chowdhury et al. (2005). In their papers they presented numerical models solving the discussed problem, which were obtained by replacing the mathematical model of the membrane module by a multicomponent model. Models presented in these works take into account pressure loss along the module.

The multicomponent approach to numerical resolution of the membrane separation of gases problem was applied also in the work of Kundu et al. (2013). In this work, additionally, recirculation of a part of streams was introduced within the framework of one membrane module.

The models mentioned above do not take into account non-ideal effects. Such phenomena as concentration polarization at the surface of membrane, Joule-Thompson effect or behaviour of the non-ideal gas can be found in the work of Scholz et al. (2013).

Remarks concerning numerous practical formulas emerging in the literature, based on simplified mathematical models, concern also membrane processes used for solid separation from its suspension in a liquid. In the case of liquid suspensions it is also important to mention many practical formulas which are only the result of the adjustment formulas to experimental results, and do not have theoretical background. A good example of such formulas are those quoted by Piątkiewicz (2012), concerning dependences of the permeate stream on the actual concentration of suspension in the batch type microfiltration system used for densification of suspensions. These formulas were proposed on the basis of experimental data analyses, without insight into the mechanism of these processes. The usefulness of these formulas for the description of batch type microfiltration systems used for densification of the oil-water emulsion was shown in the work of Szwaści et al. (2012). Whereas the practical application of formulas quoted in the work (Piątkiewicz, 2012) was shown by Szwaści et al. (2013) by analysing a batch type microfiltration installation used to densification of suspensions, working with the periodic impulse refreshment of the filter surface.

The above analysis of the literature indicates that in the modelling of membrane separation processes, simplifying assumptions are also adopted. In the authors' opinion of the quoted articles this does not cause, however, significant deviations of calculation results from the experimental data. It can be concluded that taking into account the simplifying assumptions mentioned above constitutes a standard procedure.

In order to emphasise the essential element of novelty emerging in the present work in comparison with the material contained in the previous literature, we refer again to the papers of Makaruk and Harasek (2009) and Chowdhury et al. (2005). Previously it was found that models presented in these works took into account losses of pressure along the length of the membrane (module). Now, we clearly emphasise that models presented there take into account only losses of pressure which result from the flow resistance. However there is no information about gas pressure changes resulting from the mutual transfer of energy among molecules of gases flowing in the given channel and molecules penetrating this channel through the membrane surface from the adjacent channel. The issue of the mutual transfer of energy among molecules of gases is taken under consideration in the present work and constitutes the essential novelty as compared to issues considered in the literature so far.

The authors expect that the readers of this article to be engineers and technologists of various specialties and chemists, so readers who are interested in specific, often only selected, aspects of membrane technology. Therefore, this paper presents both a detailed derivation of all dependencies and compact complete systems of differential equations needed to calculate the process which uses a specific membrane or module, sometimes at the cost of repeating in the text some of the equations quoted earlier.

## 2. MODELLING SELECTED PROCESSES OF MEMBRANE SEPARATION OF GASES

The present chapter is devoted to mathematical modelling of selected membrane processes used for separation of gaseous mixtures.

Firstly, processes accomplished in the installation with a single tubular membrane, shown in Fig. 1 will be modelled. In the beginning, we will discuss processes with the co-current flow of feeding and permeate streams, respectively, and then processes with the counter-current flow of these streams.

Secondly, processes accomplished in the installation with a single flat membrane will be modelled. For these processes, considerations concerning co-current and counter-current flows of feeding and permeate streams, respectively, will be conducted simultaneously, referring to detailed discussion over processes taking place in the installation with a single tubular membrane.

Finally, processes carried out in a membrane tubular module will be modelled. Similarly, considerations concerning co-current flow and counter-current flow of feeding and permeate streams, respectively, will be conducted simultaneously, referring to earlier discussion.

### 2.1. Processes in installation equipped with a single tubular membrane

The present subsection of the paper is devoted to the mathematical modelling of processes presented in Fig. 1 with a single tubular membrane.

In accordance with the general dependence (2), for processes presented in Fig.1 with a single tubular membrane, we obtain the following formulas, for the hydraulic diameter of the circular feeding channel and for the hydraulic diameter of the annular permeate channel, respectively.

$$d_1 = 4r_{h1} = 4 \frac{A_1}{O_1} = 4 \frac{\pi d^2}{\pi d} = d \quad (5)$$

$$d_2 = 4r_{h2} = 4 \frac{A_2}{O_2} = 4 \frac{\frac{\pi D^2}{4} - \frac{\pi (d+2l)^2}{4}}{\pi D + \pi (d+2l)} = D - (d+2l) \quad (6)$$

The elementary length  $dw$  of the channel corresponding to the elementary surface of the membrane is determined by the following formula which was recorded in the present work as the analogous formula for the surface of cylindrical wall in problems of heat transfer (Ciborowski, 1973)

$$dF = \frac{dF_z - dF_w}{\ln(dF_z / dF_w)} \quad (7)$$

where

$$dF_z = \pi (d+2l)dw \quad (8)$$

$$dF_w = \pi d dw \quad (9)$$

After substituting into Equation (7) the explicit dependences defining  $dF_z$  and  $dF_w$ , Equations (8) and (9) accordingly, we obtain

$$dF = \frac{\pi (d+2l)dw - \pi d dw}{\ln[\pi (d+2l)dw / \pi d dw]} = \frac{2\pi}{\ln(1+2l/d)} dw \quad (10)$$

If the membrane is very thin,  $l \rightarrow 0$ , then

$$dF \rightarrow \pi d dw \quad (11)$$

A decrease in the molar stream value of the component along the length of the feeding channel, at the elementary section  $dw$  is, regarding the absolute value, equal to the molar stream of the transport of this component across the elementary area  $dF$  of the membrane and to the increase in value of the molar stream of this component along the length of the permeate channel. After using Equations (3) and (4) and Equation (10), we can create a formula (12) valid for the installation presented in Fig.1, describing the elementary molar stream of component  $i = A, B$ , transported through the membrane and formula (13) concerning the inert gas.

$$-d\dot{N}_{1i} = P_i \frac{p_{1i} - p_{2i}}{l} \frac{2\pi d}{\ln(1+2l/d)} dw = d\dot{N}_{2i} \quad \text{for } i = A, B \quad (12)$$

$$d\dot{N}_{1o} = P_o \frac{p_{2o} - p_{1o}}{l} \frac{2\pi d}{\ln(1+2l/d)} dw = -d\dot{N}_{2o} \quad (13)$$

Below, consideration is given to co-current and counter-current flows, respectively, of gas streams in both channels of the membrane installation.

### 2.1.1. Co-current flow through the installation with a single tubular membrane

The pressure drop along the length  $dw$  of the feeding channel results from flow resistances and delivery of the part of the pressure energy by the flowing gas stream to create pressure and kinetic energy of the stream  $d\dot{N}_{1o}$  which leave the membrane. Below every item of energy balance sheet is described.

The pressure drop along the length  $dw$  of the feeding channel resulting from flow resistance can be presented using the following equation (the Darcy-Weissbach equation):

$$(dp_1)' = -\lambda_1 \frac{dw}{d_1} \frac{\bar{u}_1^2}{2} \rho_1 \quad (14)$$

The pressure drop along the length  $dw$  of the feeding channel resulting from transferring of a part of pressure energy by the flowing stream of gas to create the pressure energy of stream  $d\dot{N}_{1o}$  results from the following equation

$$-(dp_1)'' (\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}) = p_1 d\dot{N}_{1o} \quad (15)$$

and can be presented in the following form:

$$(dp_1)'' = -p_1 \frac{d\dot{N}_{1o}}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} \quad (16)$$

The pressure drop along the length  $dw$  of the feeding channel resulting from the transferring of a part of pressure energy by the flowing stream of gas to create the kinetic energy of stream  $d\dot{N}_{1o}$  results from the following equation

$$-(dp_1)''' (\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}) = \frac{\rho_1 \bar{u}_1^2}{2} d\dot{N}_{1o} \quad (17)$$

and can be presented in the following form:

$$(dp_1)''' = -\frac{\rho_1 \bar{u}_1^2}{2} \frac{d\dot{N}_{1o}}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} \quad (18)$$

The total decrease in pressure along the length  $dw$  of the feeding channel is a sum of pressure decreases mentioned before, thus:

$$dp_1 = (dp_1)' + (dp_1)'' + (dp_1)''' \quad (19)$$

After using Equations (14), (16) and (18), Equation (19), describing the total pressure drop along the length  $dw$  of the feeding channel, assumes the following form:

$$dp_1 = -\lambda_1 \frac{dw \bar{u}_1^2}{d_1} \rho_1 - p_1 \frac{d\dot{N}_{1o}}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} - \frac{\rho_1 \bar{u}_1^2}{2} \frac{d\dot{N}_{1o}}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} \quad (20)$$

Analogically, the pressure drop along the length  $dw$  of the permeate channel results from flow resistance and from transferring a part of pressure energy of the flowing gas stream to create pressure and kinetic energies of streams  $d\dot{N}_{2A}$  and  $d\dot{N}_{2B}$  which penetrated membrane, (it should be emphasised that the analogy between Equations (20) and (21) concerning signs before expressions describing individual types of the energy results from the fact that co-current flow of feeding and permeate is discussed now).

$$dp_2 = -\lambda_2 \frac{dw \bar{u}_2^2}{d_2} \rho_2 - p_2 \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} - \frac{\rho_2 \bar{u}_2^2}{2} \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \quad (21)$$

Having transformed Equations (12) and (13), Equations (20) and (21) take up accordingly the following forms:

$$\frac{dp_1}{dw} = -\lambda_1 \frac{\bar{u}_1^2 \rho_1}{2d_1} - \frac{p_1}{l} \frac{2\pi l}{\ln(1+2l/d)} \frac{P_o(p_{2o} - p_{1o})}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} - \frac{\rho_1 \bar{u}_1^2}{2l} \frac{2\pi l}{\ln(1+2l/d)} \frac{P_o(p_{2o} - p_{1o})}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} \quad (22)$$

$$\begin{aligned} \frac{dp_2}{dw} = & -\lambda_2 \frac{\bar{u}_2^2 \rho_2}{2d_2} - \frac{p_2}{l} \frac{2\pi l}{\ln(1+2l/d)} \frac{P_A(p_{1A} - p_{2A}) + P_B(p_{1B} - p_{2B})}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} + \\ & - \frac{\rho_2 \bar{u}_2^2}{2l} \frac{2\pi l}{\ln(1+2l/d)} \frac{P_A(p_{1A} - p_{2A}) + P_B(p_{1B} - p_{2B})}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \end{aligned} \quad (23)$$

In order to obtain a complete system of equations necessary to solve the problem, it is necessary to add Equations (24) to (29) obtained after conversion of Equations (12) and (13), to Equations (22) and (23):

$$\frac{d\dot{N}_{1A}}{dw} = -P_A \frac{p_{1A} - p_{2A}}{l} \frac{2\pi l}{\ln(1+2l/d)} \quad (24)$$

$$\frac{d\dot{N}_{1B}}{dw} = -P_B \frac{p_{1B} - p_{2B}}{l} \frac{2\pi l}{\ln(1+2l/d)} \quad (25)$$

$$\frac{d\dot{N}_{1o}}{dw} = P_o \frac{p_{2o} - p_{1o}}{l} \frac{2\pi l}{\ln(1+2l/d)} \quad (26)$$

$$\frac{d\dot{N}_{2A}}{dw} = P_A \frac{p_{1A} - p_{2A}}{l} \frac{2\pi l}{\ln(1+2l/d)} \quad (27)$$

$$\frac{d\dot{N}_{2B}}{dw} = P_B \frac{p_{1B} - p_{2B}}{l} \frac{2\pi l}{\ln(1+2l/d)} \quad (28)$$

$$\frac{d\dot{N}_{2o}}{dw} = -P_o \frac{P_{2p} - P_{1o}}{l} \frac{2\pi l}{\ln(1 + 2l/d)} \quad (29)$$

Partial pressures are calculated according to the following formula:

$$p_{ji}(w) = p_j(w) \frac{\dot{N}_{ji}(w)}{\dot{N}_{jA}(w) + \dot{N}_{jB}(w) + \dot{N}_{jo}(w)} \quad (30)$$

The mean linear velocity of a gas mixture is calculated on the basis of the following formula derived from the Clapeyron equation:

$$\bar{u}_j(w) = \frac{[\dot{N}_{jA}(w) + \dot{N}_{jB}(w) + \dot{N}_{jo}(w)]RT_j}{p_j(w)A_j} \quad (31)$$

The mean density of a gas mixture is calculated on the basis of the modified Clapeyron equation:

$$\bar{\rho}_j(w) = \frac{p_j(w)}{RT_j} \bar{M}_j(w) = \frac{p_j(w)}{RT_j} \frac{\dot{N}_{jA}(w)M_A + \dot{N}_{jB}(w)M_B + \dot{N}_{jo}(w)M_o}{\dot{N}_{jA}(w) + \dot{N}_{jB}(w) + \dot{N}_{jo}(w)} \quad (32)$$

Equations (30) to (32) are valid for both channels of the installation,  $j = 1, 2$ , and for all components of gaseous mixtures,  $i = A, B, o$ .

The knowledge of streams of individual gases in their mixtures explicitly determines the composition of these mixtures, which simplifies finding their average viscosity. Equations permitting to find the average viscosity of the mixture of gases are available for example in handbooks on the thermodynamics of processes.

Knowing the supplementary diameter of a channel and the average linear velocity, the average density, and the average viscosity of the mixture of gases, we can calculate the Reynolds number and then the resistance of flow. In order to calculate the coefficients of resistance  $\lambda_j(\text{Re}(w))$ , we ought to use correlations readily available in the literature.

### 2.1.2. Counter-current flow through the installation with a single tubular membrane

Similarly to the installation with the co-current feeding and permeate flow, in the installation with the counter-current flow, the return of the feeding stream conforms with increasing values of the linear dimension of channel length  $w$  (Fig.1). Therefore, equations describing the pressure drop in the feeding channel for a counter-current flow installation are the same as those for the co-current installation. This means that Equations (20) and (22) are true both for the installation with co-current flow and counter-current flow of feeding and permeate streams. However, the return of permeate stream is inconsistent with increasing values of the linear dimension of channel length  $w$ . Therefore, the pressure drop of this stream should be referred to the elementary length of the channel  $-dw$ . An alternative approach to the problem is taking into consideration the positive direction of the measurement of channel length and the positive elementary length of the channel  $dw$ ; then one ought to state the increase in value of individual components of the pressure that concerns the positive differentials of the pressure. Only this alternative approach is applied in the present work.

The change of pressure value along the length  $dw$  of the permeate channel resulting from flow resistance can be expressed with the following equation (Darcy-Weissbach equation):

$$(dp_2)' = \lambda_2 \frac{dw}{d_2} \frac{\bar{u}_2^2}{2} \rho_2 \quad (33)$$

The change of pressure along the length  $dw$  of the permeate channel resulting from the delivery of a part of pressure energy by the flowing stream of gas and creation of the pressure energy of streams  $d\dot{N}_{2A}$  and  $d\dot{N}_{2B}$ , results from the following equation

$$(dp_2)''(\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}) = p_2 d\dot{N}_{2A} + p_2 d\dot{N}_{2B} = p_2(d\dot{N}_{2A} + d\dot{N}_{2B}) \quad (34)$$

and can be presented in the following form:

$$(dp_2)'' = p_2 \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \quad (35)$$

The change of pressure along the length  $dw$  of the permeate channel resulting from the delivery of a part of pressure energy by the flowing stream of gas and creation of the kinetic energy of streams  $d\dot{N}_{2A}$  and  $d\dot{N}_{2B}$ , results from the following equation

$$(dp_2)'''(\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}) = \frac{\rho_2 \bar{u}_2^2}{2} (d\dot{N}_{2A} + d\dot{N}_{2B}) \quad (36)$$

and can be presented in the following form:

$$(dp_2)''' = \frac{\rho_2 \bar{u}_2^2}{2} \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \quad (37)$$

The total change of pressure along length  $dw$  along the permeate channel is a sum of pressure changes mentioned before, thus:

$$dp_2 = (dp_2)' + (dp_2)'' + (dp_2)''' \quad (38)$$

After using Equations (33), (35) and (37), Equation (38) describing the total change of pressure along length  $dw$  of permeate channel assumes the following form:

$$dp_2 = \lambda_2 \frac{dw}{d_2} \frac{\bar{u}_2^2}{2} \rho_2 + p_2 \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} + \frac{\rho_2 \bar{u}_2^2}{2} \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \quad (39)$$

After using Equation (12), it is possible to eliminate differentials  $d\dot{N}_{2A}$  and  $d\dot{N}_{2B}$  and Equation (39) assumes the form of Equation (41). To obtain a compact form of the mathematical model for the counter-current flow installation, Equation (41), valid for the permeate stream in the counter-current flow installation, was preceded with Equation (40) valid for the feeding stream, the same as Equation (22) for the feeding stream in the co-current flow installation (compare the content of the paragraph over the paragraph containing the Equation (33)).

$$dp_2 = \lambda_2 \frac{dw}{d_2} \frac{\bar{u}_2^2}{2} \rho_2 + p_2 \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} + \frac{\rho_2 \bar{u}_2^2}{2} \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \quad (39)$$

After using Equation (12), it is possible to eliminate differentials  $d\dot{N}_{2A}$  and  $d\dot{N}_{2B}$  and Equation (39) assumes the form of Equation (41). To obtain a compact form of the mathematical model for the counter-current flow installation, Equation (41), valid for the permeate stream in the counter-current flow installation, was preceded with Equation (40) valid for the feeding stream, the same as Equation (22) for the feeding stream in the co-current flow installation (compare the content of the paragraph over the paragraph containing the Equation (33)).

$$\frac{dp_1}{dw} = -\lambda_1 \frac{\bar{u}_1^2 \rho_1}{2d_1} - \frac{p_1}{l} \frac{2\pi l}{\ln(1+2l/d)} \frac{P_o(p_{2o} - p_{1o})}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} - \frac{\rho_1 \bar{u}_1^2}{2l} \frac{2\pi l}{\ln(1+2l/d)} \frac{P_o(p_{2o} - p_{1o})}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} \quad (40)$$

$$\begin{aligned} \frac{dp_2}{dw} = & \lambda_2 \frac{\bar{u}_2^2 \rho_2}{2d_2} + \frac{p_2}{l} \frac{2\pi l}{\ln(1+2l/d)} \frac{P_A(p_{1A} - p_{2A}) + P_B(p_{1B} - p_{2B})}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} + \\ & + \frac{\rho_2 \bar{u}_2^2}{2l} \frac{2\pi l}{\ln(1+2l/d)} \frac{P_A(p_{1A} - p_{2A}) + P_B(p_{1B} - p_{2B})}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \end{aligned} \quad (41)$$

In order to obtain a complete system of equations valid for the counter-current flow installation, we should add Equations (42) to (47) to Equations (40) and (41). Equations (42) to (47) are obtained after conversions of Equations (12) and (13). Equations (42) to (47) valid for the feeding stream are identical with Equations (24) to (26), while Equations (45) to (47) take into account the fact that they were derived in relation to the permeate stream flowing into the opposite direction than the feeding stream.

$$\frac{d\dot{N}_{1A}}{dw} = -P_A \frac{p_{1A} - p_{2A}}{l} \frac{2\pi l}{\ln(1+2l/d)} \quad (42)$$

$$\frac{d\dot{N}_{1B}}{dw} = -P_B \frac{p_{1B} - p_{2B}}{l} \frac{2\pi l}{\ln(1+2l/d)} \quad (43)$$

$$\frac{d\dot{N}_{1o}}{dw} = P_o \frac{p_{2o} - p_{1o}}{l} \frac{2\pi l}{\ln(1+2l/d)} \quad (44)$$

$$\frac{d\dot{N}_{2A}}{dw} = -P_A \frac{p_{1A} - p_{2A}}{l} \frac{2\pi l}{\ln(1+2l/d)} \quad (45)$$

$$\frac{d\dot{N}_{2B}}{dw} = -P_B \frac{p_{1B} - p_{2B}}{l} \frac{2\pi l}{\ln(1+2l/d)} \quad (46)$$

$$\frac{d\dot{N}_{2o}}{dw} = P_o \frac{p_{2p} - p_{1o}}{l} \frac{2\pi l}{\ln(1+2l/d)} \quad (47)$$

Partial pressures are calculated from formula (30), the mean linear velocity of a gaseous mixture from formula (31), and the mean density of a gaseous mixture from formula (32).

## 2.2. Processes in the installation equipped with a flat membrane

The present subsection of the work is devoted to the modelling of processes presented in Fig.2, taking place in the installation with a flat membrane.

In accordance with dependence (2), for processes with a flat membrane, we obtain the following formula describing supplementary diameters of rectangular feeding and permeate channels, respectively, each of which has a cross-section  $A_j$ , common width  $s$ , and individual height  $A_j/s$  for  $j = 1, 2$ :

$$d_j = 4r_{hj} = 4 \frac{A_j}{O_j} = 4 \frac{A_j}{2(s + \frac{A_j}{s})} = \frac{2A_j s}{s^2 + A_j} \text{ for } i = A, B \quad (48)$$

The elementary surface of a membrane, defined by the following formula, corresponds to the elementary length  $dw$  of the channel:

$$dF = sdw \quad (49)$$

The decrease in the value of the molar stream of the component  $i = A, B$  along the length of the feeding channel in the elementary section  $dw$ , is equal, regarding its absolute value to the molar stream of transport of this component through the elementary surface  $dF$  of the membrane and is equal to the increase of the molar stream of this component along the length of the permeate channel.

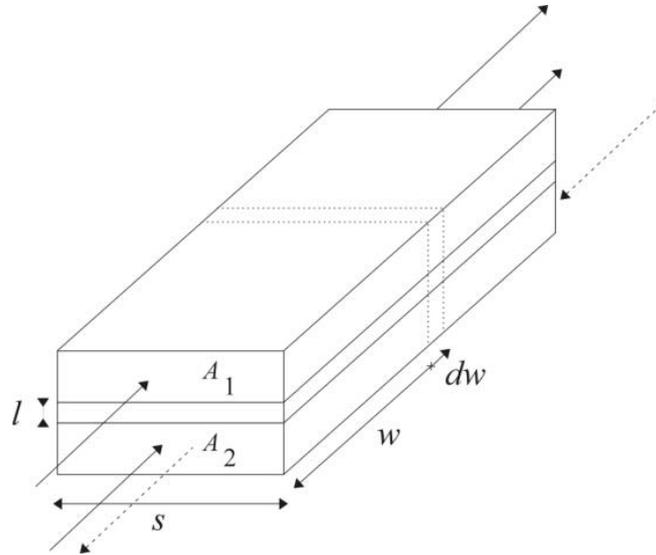


Fig. 2. Scheme of co-current flow and counter-current flow processes with a flat membrane

After using formulas (3), (4) and (49), it is possible to derive the formula (50) describing the elementary molar stream of the component  $i = A, B$  transported by the membrane and the formula (51) concerning the inert gas.

$$-d\dot{N}_{1i} = P_i \frac{p_{1i} - p_{2i}}{l} sdw = d\dot{N}_{2i} \quad \text{for } i = A, B \quad (50)$$

$$d\dot{N}_{1o} = P_o \frac{p_{2o} - p_{1o}}{l} sdw = -d\dot{N}_{2o} \quad (51)$$

Below, there are considerations of the mathematical model of processes in an installation with a flat membrane, valid for both – co-current flow and counter-current flow streams of gases in both channels of considered membrane installation.

The pressure drop along the length  $dw$  of the feeding channel results from flow resistances and delivery of a part of pressure energy by the flowing gas stream to create energy of the pressure and the kinetic energy of the stream which penetrated through the membrane from the permeate channel. This pressure drop is described by Equation (52) identical with Equation (20), valid for the feeding channel in the installation with a single tubular membrane and binding for both – co-current and counter-current flow of feeding and permeate streams (compare content of the section over the paragraph containing Equation (33)).

$$dp_1 = -\lambda_1 \frac{dw \bar{u}_1^2}{d_1} \rho_1 - p_1 \frac{d\dot{N}_{1o}}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} - \frac{\rho_1 \bar{u}_1^2}{2} \frac{d\dot{N}_{1o}}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} \quad (52)$$

As regards the permeate channel (where the energy of pressure of the flowing stream of gas is transmitted to streams  $d\dot{N}_{2A}$  and  $d\dot{N}_{2B}$  to create their energy of pressure and kinetic energy), the equation valid for the co-current flow is identical with Equation (21) derived for the co-current installation with a single tubular membrane, while the equation valid for the counter-current flow is

identical with Equation (39) derived for the counter-current installation with a single tubular membrane. Equations (21) and (39) can be presented together in the form of Equation (53) describing pressure changes in the permeate channel in the installation with a flat membrane. Equation (53) presented with the upper sign of the double symbol of the mathematical operation concerns the co-current flow of feeding and permeate, while the equation presented with the lower sign concerns the counter-current flow, respectively.

$$dp_2 = \mp \lambda_2 \frac{dw}{d_2} \frac{\bar{u}_2^2}{2} \rho_2 \mp p_2 \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \mp \frac{\rho_2 \bar{u}_2^2}{2} \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \quad (53)$$

After using Equations (50) and (51), Equations (52) and (53) take respectively the following forms:

$$\frac{dp_1}{dw} = -\lambda_1 \frac{\bar{u}_1^2 \rho_1}{2d_1} - \frac{p_1 s}{l} \frac{P_o(p_{2o} - p_{1o})}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} - \frac{\rho_1 s \bar{u}_1^2}{2l} \frac{P_o(p_{2o} - p_{1o})}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} \quad (54)$$

$$\begin{aligned} \frac{dp_2}{dw} = & \mp \lambda_2 \frac{\bar{u}_2^2 \rho_2}{2d_2} \mp \frac{p_2 s}{l} \frac{P_A(p_{1A} - p_{2A}) + P_B(p_{1B} - p_{2B})}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} + \\ & \mp \frac{\rho_2 s \bar{u}_2^2}{2l} \frac{P_A(p_{1A} - p_{2A}) + P_B(p_{1B} - p_{2B})}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \end{aligned} \quad (55)$$

In order to obtain a complete system of equations valid for the installation with the flat membrane, both for the co-current flow and counter-current flow of gas streams, we should add Equations (56) to (61), obtained after conversions of Equations (50) and (51), to Equations (54) and (55). It is important to notice that Equations (56) to (58) concern the feeding channel and therefore their right sides begin with the same signs of mathematical operations as Equations (24) to (26) and (42) to (44), valid for the feeding channel of the installation with the tubular membrane. However, Equations (59) to (61) concern the permeate channel and therefore their right sides must begin with same signs of mathematical operations as Equations (27) to (29), if Equations (59) to (61) should concern the co-current flow of feeding and permeate streams. They should begin with the same signs of mathematical operations as Equations (45) to (47), if Equations (59) to (61) should concern the counter-current flow of feeding and permeate streams. Summing up, Equations (59) to (61) presented with the upper signs of double symbols of mathematical operations concern the co-current flow of feeding and permeate, while equations presented with the lower signs concern the counter-current flow.

$$\frac{d\dot{N}_{1A}}{dw} = -P_A \frac{p_{1A} - p_{2A}}{l} s \quad (56)$$

$$\frac{d\dot{N}_{1B}}{dw} = -P_B \frac{p_{1B} - p_{2B}}{l} s \quad (57)$$

$$\frac{d\dot{N}_{1o}}{dw} = P_o \frac{p_{2o} - p_{1o}}{l} s \quad (58)$$

$$\frac{d\dot{N}_{2A}}{dw} = \pm P_A \frac{p_{1A} - p_{2A}}{l} s \quad (59)$$

$$\frac{d\dot{N}_{2B}}{dw} = \pm P_B \frac{p_{1B} - p_{2B}}{l} s \quad (60)$$

$$\frac{d\dot{N}_{2o}}{dw} = \mp P_o \frac{p_{2o} - p_{1o}}{l} s \quad (61)$$

Partial pressures are calculated from formula (30), the mean linear velocity of a gaseous mixture from formula (31), and the mean density of a gaseous mixture from formula (32).

### 2.3. Processes taking place in membrane tubular module

The present subsection of the work is devoted to the modelling of processes in the tubular membrane module shown in Fig.3. The permeate stream marked with the continuous line concerns co-current flow processes, while the dashed line represents counter-current flow processes.

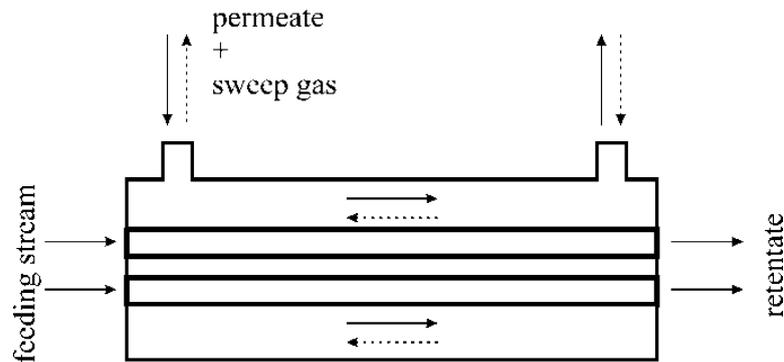


Fig. 3. Scheme of co-current flow and counter-current flow membrane tubular module

A feeding stream flows through  $n$  parallel, independent, tubular membranes. Every tube has a supplementary diameter equal to its natural geometric inside diameter, and identical to Equation (62) with Equation (5). Index "1" is assigned to values related to the feeding flow, thus:

$$d_1 = d \quad (62)$$

However, the permeate flows through the channel (intertubular space), parallel to tubes. Index „2” is assigned to this channel, and its supplementary diameter is calculated from Equation (63) resulting from the general Equation (2).

$$d_2 = 4 \frac{\frac{\pi D^2}{4} - \frac{n\pi(d+2l)^2}{4}}{\pi D + n\pi(d+2l)} = \frac{D^2 - n(d+2l)^2}{D + n(d+2l)} \quad (63)$$

If a membrane is very thin -  $l \rightarrow 0$ , then:

$$d_2 = \frac{D^2 - nd^2}{D + nd} \quad (64)$$

The field of the cross-sectional area of the total feeding channel is equal to the product of the number of tubular membranes in the module and the cross-sectional area of each of the independent tubes, that is the area of a circle with diameter  $d$

$$A_1 = n \frac{\pi d^2}{4} \quad (65)$$

while the field of the right sectional area of the permeate channel is calculated according to the formula:

$$A_2 = \frac{\pi}{4} [D^2 - n(d+2l)^2] \quad (66)$$

If a membrane is very thin,  $l \rightarrow 0$ , then:

$$A_2 = \frac{\pi}{4}(D^2 - nd^2) \quad (67)$$

It is natural to assume that in any cross-section of the module, the status of all tubular membranes is the same. Therefore, just like for single membranes, partial pressures are calculated according to formula (30), the mean linear velocity of the mixture of gases according to formula (31), while the average density of the mixture of gases is calculated from the formula (32). Apparently, it would be necessary to use formulas (65) and (66) or (67), respectively, defining adequate cross-section areas of the module.

In order to modify Equations (12) and (13), describing the transport of individual gases through the area of a single tubular membrane, representing the elementary length of the channel  $dw$ , to a useful form for the description of this transport in the membrane module consisting of  $n$  tubes, account should be taken of the fact that currently the mass transfer area is  $n$  times greater, thus:

$$-d\dot{N}_{1i} = P_i \frac{p_{1i} - p_{2i}}{l} \frac{2\pi l n}{\ln(1+2l/d)} dw = d\dot{N}_{2i} \quad \text{for } i = A, B \quad (68)$$

$$d\dot{N}_{1o} = P_o \frac{p_{2o} - p_{1o}}{l} \frac{2\pi l n}{\ln(1+2l/d)} dw = -d\dot{N}_{2o} \quad (69)$$

The decrease in pressures along elementary lengths of feeding and permeate channels in a tubular module, respectively, can be described with the same equations as those for an installation with the single tubular membrane, i.e. by Equation (20) valid for the feeding stream, both in the case of co-current flow and counter-current flow and by Equation (21) valid for permeate in the case of the co-current flow and by Equation (39) valid for permeate in the case of the counter-current flow. These equations can be presented together in the form of Equations (70) and (71) in the identical form as Equations (52) and (53).

$$dp_1 = -\lambda_1 \frac{dw \bar{u}_1^2}{d_1} \rho_1 - p_1 \frac{d\dot{N}_{1o}}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} - \frac{\rho_1 \bar{u}_1^2}{2} \frac{d\dot{N}_{1o}}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} \quad (70)$$

$$dp_2 = \mp \lambda_2 \frac{dw \bar{u}_2^2}{d_2} \rho_2 \mp p_2 \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \mp \frac{\rho_2 \bar{u}_2^2}{2} \frac{d\dot{N}_{2A} + d\dot{N}_{2B}}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} \quad (71)$$

Equation (70) is valid for a feeding stream, both in the case of co-current flow and counter-current flow, while Equation (71) is valid for permeate; Equations presented with the upper sign of the double symbol of the mathematical operations concern the co-current flow of feeding and permeate streams, while equations presented with the lower sign concern the counter-current flow.

After using Equations (68) and (69), Equations (70) and (71) assume, respectively, the forms:

$$\frac{dp_1}{dw} = -\lambda_1 \frac{\bar{u}_1^2 \rho_1}{2d_1} - \frac{p_1}{l} \frac{2\pi l n}{\ln(1+2l/d)} \frac{P_o(p_{2o} - p_{1o})}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} - \frac{\rho_1 \bar{u}_1^2}{2l} \frac{2\pi l n}{\ln(1+2l/d)} \frac{P_o(p_{2o} - p_{1o})}{\dot{N}_{1A} + \dot{N}_{1B} + \dot{N}_{1o}} \quad (72)$$

$$\begin{aligned} \frac{dp_2}{dw} = & \pm \lambda_2 \frac{\bar{u}_2^2 \rho_2}{2d_2} \pm \frac{p_2}{l} \frac{2\pi l n}{\ln(1+2l/d)} \frac{P_A(p_{1A} - p_{2A}) + P_B(p_{1B} - p_{2B})}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} + \\ & \pm \frac{\rho_2 \bar{u}_2^2}{2l} \frac{2\pi l n}{\ln(1+2l/d)} \frac{P_A(p_{1A} - p_{2A}) + P_B(p_{1B} - p_{2B})}{\dot{N}_{2A} + \dot{N}_{2B} + \dot{N}_{2o}} + \end{aligned} \quad (73)$$

In order to obtain a complete system of equations valid for the tubular module, we should add Equations (74) to (79) to Equations (72) and (73). Equations (74) to (76) are valid for feeding, both for

co-current flow and counter-current flow, while Equations (77) to (79) are valid for permeate; the ones with upper signs of double symbols of the mathematical operations concern the co-current flow of feeding and permeate streams, while equations with lower signs concern the counter-current flow.

$$\frac{d\dot{N}_{1A}}{dw} = -P_A \frac{p_{1A} - p_{2A}}{l} \frac{2\pi l n}{\ln(1 + 2l/d)} \quad (74)$$

$$\frac{d\dot{N}_{1B}}{dw} = -P_B \frac{p_{1B} - p_{2B}}{l} \frac{2\pi l n}{\ln(1 + 2l/d)} \quad (75)$$

$$\frac{d\dot{N}_{1o}}{dw} = P_o \frac{p_{2o} - p_{1o}}{l} \frac{2\pi l n}{\ln(1 + 2l/d)} \quad (76)$$

$$\frac{d\dot{N}_{2A}}{dw} = \pm P_A \frac{p_{1A} - p_{2A}}{l} \frac{2\pi l n}{\ln(1 + 2l/d)} \quad (77)$$

$$\frac{d\dot{N}_{2B}}{dw} = \pm P_B \frac{p_{1B} - p_{2B}}{l} \frac{2\pi l n}{\ln(1 + 2l/d)} \quad (78)$$

$$\frac{d\dot{N}_{2o}}{dw} = \mp P_o \frac{p_{2p} - p_{1o}}{l} \frac{2\pi l n}{\ln(1 + 2l/d)} \quad (79)$$

As was mentioned above, partial pressures was calculated from the formula (30), the mean linear velocity of the mixture of gases from formula (31), while the mean density of the mixture of gases was calculated according to formula (32), whereas in the formula (31) we should use dependences (65) and (66) or (67), respectively.

### 3. SAMPLE RESULTS OF COMPUTER CALCULATIONS

The derived mathematical models of chosen processes of the membrane separation of gaseous mixtures with the participation of nonporous polymer membranes were used to create a program for computer calculations.

Sample results of calculations were obtained for separation of the mixture CO<sub>2</sub> (component *A*) and CH<sub>4</sub> (component *B*) and for He as the inert gas in the installation with the single flat membrane, for which properties of the membrane made of PDMS were used, and its thickness  $l = 2 \mu\text{m}$  was assumed. For the gases migrating through the membrane, at preset temperature  $T = 293\text{K}$ , permeabilities for individual gases amounted to:  $P_{\text{CO}_2} = 4500$  barrer,  $P_{\text{CH}_4} = 1350$  barrer and  $P_{\text{He}} = 560$  barrer, respectively, whereas  $1 \text{ barrer} = 10^{-10} \text{ N} \cdot \text{cm}^3 \cdot \text{cm} / (\text{cm}^2 \cdot \text{s} \cdot \text{cmHg}) = 3,39 \cdot 10^{-16} \text{ mole} / (\text{m s Pa})$ .

#### 3.1. Co-current flow processes in the installation with a single flat membrane

Results of calculations were obtained for the following initial conditions:  $p_1(0) = 1500000 \text{ Pa}$  and  $p_2(w_k) = 300000 \text{ Pa}$  and  $\dot{N}_{1A}(0) = 6 \text{ mole/s}$ ,  $\dot{N}_{1B}(0) = 24 \text{ mole/s}$ ,  $\dot{N}_{1o}(0) = 0 \text{ mole/s}$ ,  $\dot{N}_{2A}(0) = 0$ ,  $\dot{N}_{2B}(0) = 0$ ,  $\dot{N}_{2o}(0) = 10 \text{ mole/s}$ .

Figure 4 presents changes of total pressures of gases along the length of the feeding and permeate channels respectively, while Fig.5 presents the variability of the value of streams of separated gases *A* and *B* along the feeding channel.

To show how slow the discussed processes conducted in installations with single membranes are, the results of calculations were referred to very long channels,  $w_k = 15\text{m}$ . The results clearly indicate the need for conducting these processes in membrane modules which in comparison with installations with single membranes, have, on the unit of length of the installation, much larger surfaces of membranes through which separated components of the gaseous mixture are transported. In practice, the cascades composed of modules with a convenient length are used, for example each module being 1.5 m long (Szwast, 2012).

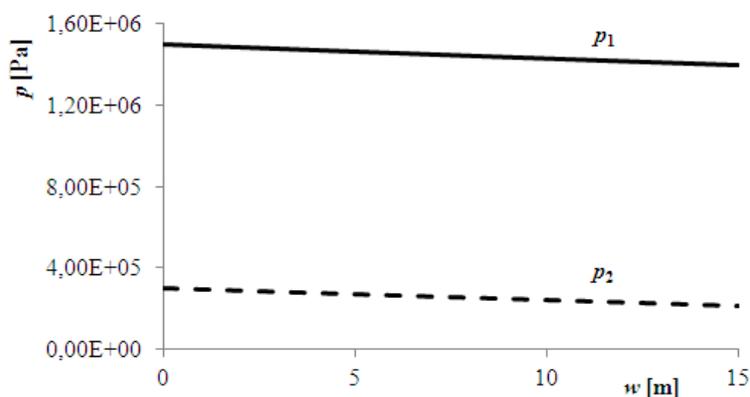


Fig. 4. Variability of total pressures along the length of channels

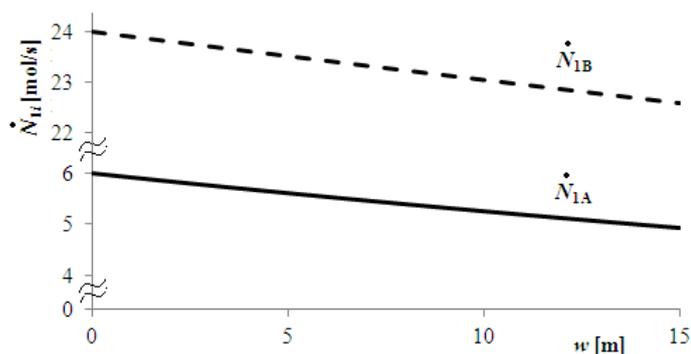


Fig.5. Variability of streams of separated gases *A* and *B* along the length of feeding channel

### 3.2. Counter-current flow processes in the installation with a single flat membrane

Sample results of calculations were obtained for the separation of mixture  $\text{CO}_2$  (component *A*) and  $\text{CH}_4$  (component *B*) and for He as the inert gas, i.e. for the same system which was taken into consideration during calculations concerning the co-current flow system. Similarly to the co-current flow system, we also used the properties of the membrane made of PDMS and its thicknesses of  $l = 2 \mu\text{m}$ .

It should be emphasised that for counter-current flow installations, two-point initial conditions exits. For the specific problem the values of pressures  $p_1(0)$  and  $p_2(w_k)$  as well as the values of streams  $\dot{N}_{1A}(0)$ ,  $\dot{N}_{1B}(0)$  and  $\dot{N}_{2o}(w_k)$  in the face of  $\dot{N}_{1o}(0)=0$ ,  $\dot{N}_{2A}(w_k)=0$  and  $\dot{N}_{2B}(w_k)=0$  are known. Prior to calculations we ought to assume a value  $p_2(0)$  and the values of  $\dot{N}_{2A}(0)$ ,  $\dot{N}_{2B}(0)$  as well as  $\dot{N}_{2o}(0)$ . Then the knowledge (from the conditions of a process or from assumptions, respectively) of the value  $p_1(0)$  and  $p_2(0)$  as well as  $\dot{N}_{1A}(0)$ ,  $\dot{N}_{1B}(0)$ ,  $\dot{N}_{1o}(0)=0$  and  $\dot{N}_{2A}(0)$ ,  $\dot{N}_{2B}(0)$ ,  $\dot{N}_{2o}(0)$  enables integration of the system of differential equations describing the counter-current flow process, starting from  $w = 0$ . The correctness of the assumption of the value  $p_2(0)$ ,  $\dot{N}_{2A}(0)$ ,  $\dot{N}_{2B}(0)$  and

$\dot{N}_{2o}(0)$  verifies the conformity of the values  $p_2(w_k)$ ,  $\dot{N}_{2A}(w_k)$ ,  $\dot{N}_{2B}(w_k)$  and  $\dot{N}_{2o}(w_k)$  calculated during the integration with values assigned for these physical quantities. In the present work we assumed the following figures:  $p_1(0) = 1500000$  Pa and  $p_2(w_k) = 300000$  Pa, and  $\dot{N}_{1A}(0) = 6$  mole/s,  $\dot{N}_{1B}(0) = 24$  mole/s,  $\dot{N}_{1o}(0) = 0$  and  $\dot{N}_{2A}(w_k) = 0$ ,  $\dot{N}_{2B}(w_k) = 0$ ,  $\dot{N}_{2o}(w_k) = 10$  mole/s.

Figure 6 presents changes of total pressures of gases along the length of the feeding and permeate channels, respectively, while Fig.7 presents the variability of the value of streams of separated gases *A* and *B* along the feeding channel.

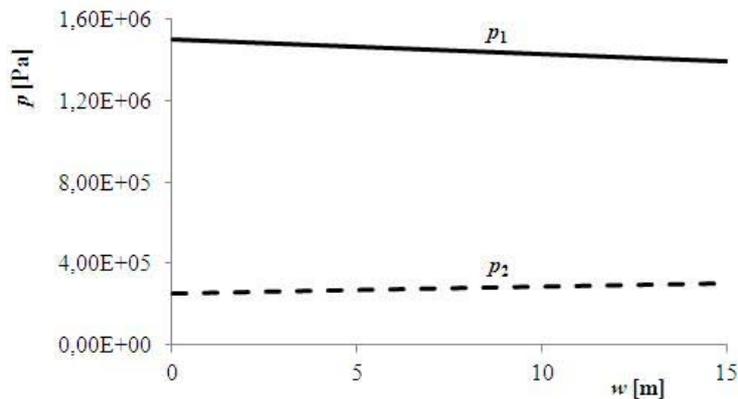


Fig.6. Variability of total pressures along the length of channels

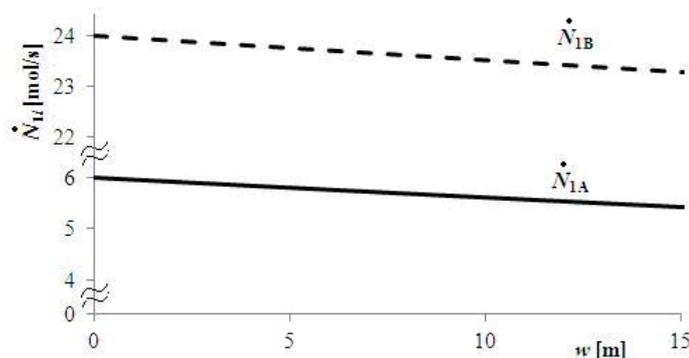


Fig.7. Variability of streams of separated gases *A* and *B* along the feeding channel

The computer program appeared to be very effective. The analysis of an extensive set of computer-calculated data and the attempts to optimum structures composed of membrane modules will become a subject of the following publication.

#### 4. SUMMARY

The paper presents the mathematical modelling of isothermal processes of gas separation taking place in installations with nonporous polymer membranes. The modelling concerns both simple membrane installations consisting of feeding and permeate channels – and membrane modules. Account was taken of different shapes of the cross-section in the channels. Particularly, modelling was conducted for processes proceeding in an installation with a single tubular membrane, processes proceeding in an installation with a single flat membrane and processes proceeding in a membrane tubular module.

Consideration was given to co-current flows and counter-current flows, for feeding and permeate streams, respectively, flowing together with the inert gas receiving the permeate.

In the proposed mathematical model account was taken of the fact that gas pressure changes along the length of channels were the result of both, the pressure drop connected with the resistance of flow, and transfers of energy by molecules of gas flowing in a given channel to molecules which penetrated this channel from the flanking channel. The authors assumed that the flowing stream of gas transfers a part of its pressure energy (the ability to do the technical work) to the elementary gas stream which penetrated this channel, through the membrane from the flanking channel for creation of the energy of pressure and the kinetic energy of this elementary gas stream. In this model it was assumed that gas at the exit of the membrane had a zero pressure and kinetic energy, while the energy of gas entering the membrane was dispersed and as such was omitted in considerations of the isothermal process.

The literature on membrane technique takes into account only the pressure drop connected with flow resistance. The important issue of the mutual transfer of energy among gas molecules, taken into account in the present work, constitutes the essential novelty in comparison with issues considered in the literature so far.

On the basis of calculation results one emphasised that the discussed processes proceeding in installations with single membranes were very slow and required very long channels. Consequently, one emphasised the need for conducting these processes in membrane modules which, in comparison with installations with single membranes, have on the unit of length of the installation much greater surfaces of membranes through which separated components of the gaseous mixture are transported.

The developed computer program calculations created after using mathematical models derived in the work, concerning selected processes of the membrane separation of gaseous mixtures with the participation of nonporous polymer membranes appeared to be very effective.

The analysis of the significant number of results of computer calculations which allows to determine the influence of pressure drop connected with energy transfer between molecules on the exactness of final results and to search of optimum structures composed of membrane modules will become a subject of the following publication.

## SYMBOLS

$A_j$	cross-section of channel $j$ , $m^2$
$D$	internal diameter of permeate channel for tubular membrane, m
$d$	internal diameter of feeding channel for tubular membrane, m
$d_j$	hydraulic diameter of channel $j$ , m
$F$	area of membrane side, $m^2$
$F_w$	area of internal side of cylindrical membrane, $m^2$
$F_z$	area of external side of cylindrical membrane, $m^2$
$l$	thickness of the membrane, m
$M_i$	molar mass of component of the gas $i$ , kg/mole
$\dot{N}_{ji}$	$\dot{N}_{ji}(w)$ , molar stream of gas $i$ in channel $j$ , (for membrane module and feeding stream, total (summary) stream in all individual membrane tubes), mole/s
$n$	number of individual tubular membranes in membrane tubular module
$P_i$	$P_i(T(w))$ permeability for gas $i$ , mole/(m s Pa)
$p_j$	$p_j(w)$ , pressure of gaseous mixture in channel $j$ , Pa
$p_{ij}$	$p_{ij}(w)$ , partial pressure of gas $i$ in channel $j$ , Pa
$R$	gas constant, J/(mole K)
$r_h$	hydraulic radius of channel, m
$s$	width of rectangular channel, m

$T_j$	$T_j(w)$ , temperature of gases in channel $j$ , K
$\bar{u}_j$	$\bar{u}_j(w)$ , mean velocity of gas stream in channel $j$ , m/s
$w$	$0 \leq w \leq w_k$ , linear dimension of the length of channel, m
$\bar{\rho}_j$	$\bar{\rho}_j(w)$ , mean density of gases in channel $j$ , kg/m <sup>3</sup>
$\lambda_j$	$\lambda_j(w)$ , coefficient of the flow resistance in channel $j$

#### Subscripts

$1, 2$	gaseous phase for feeding and permeate, respectively
$A, B$	separated gaseous components
$o$	inert gas component

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