**JOURNAL OF ENGINEERING SCIENCES**

Volume 9, Issue 1 (2022)

*Huliienko S. V., Korniyenko Y. M., Muzyka S. M., Holubka K. (2022). Simulation of reverse osmosis process: Novel approaches and development trends. Journal of Engineering Sciences, Vol. 9(2), pp. F6-F36, doi: 10.21272/jes.2022.9(2).f2*


# Simulation of Reverse Osmosis Process: Novel Approaches and Development Trends

Huliienko S. V.[1*[0000-0002-9042-870X]], Korniyenko Y. M.[1[0000-0002-3031-6212]], Muzyka S. M.[1], Holubka K.[2]

[1] National Technical University of Ukraine "Igor Sikorsky Kyiv Polytechnic Institute", 37, Peremohy Ave., 03056, Kyiv, Ukraine;
[2] University of Montpellier, 163, Auguste Broussonnet St., 34090, Montpellier, France

**Article info:**
Submitted: May 8, 2022
Accepted for publication: September 9, 2022
Available online: September 12, 2022

***Corresponding email:**
sergii.guliienko@gmail.com

**Abstract**. Reverse osmosis is an essential technological separation process that has a large number of practical applications. The mathematical simulation is significant for designing and determining the most effective modes of membrane equipment operation and for a deep understanding of the processes in membrane units. This paper is an attempt at systematization and generalizing the results of the investigations dedicated to reverse osmosis simulation, which was published from 2011 to 2020. The main approaches to simulation were analyzed, and the scope of use of each of them was delineated. It was defined that computational fluid dynamics was the most used technique for reverse osmosis simulation; the intensive increase in using of molecular dynamics methods was pointed out. Since these two approaches provide the deepest insight into processes, it is likely that they will further be widely used for reverse osmosis simulations. At the same time, for the simulation of the membrane plant, it is reasonable to use the models that required the simplest solutions methods. The solution-diffusion model appears to be the most effective and flexible for these purposes. Therefore, this model was widely used in considering the period. The practical problems solved using each of the considered approaches were reviewed. Moreover, the software used for the solution of the mathematical models was regarded.

**Keywords:** reverse osmosis, membrane, simulation, optimization, software.

## 1 Introduction

The pressure-driven membrane processes are widely used in many industries, including chemical, food, pharmaceutical, biotechnologies, water treatment, and environmental protection. The mathematical simulation of the process plays an essential role in designing and exploring such equipment since this technique defines the most rational design of apparatus and operation modes with a lower number of experimental investigations. However, there exists quite a significant number of approaches to the simulation. Therefore, the systematized information about types of mathematical models of pressure-driven membrane processes would help choose the simulation method for the particular process. In previous work [1], the attempt was to systematize the theoretical investigation of pressure driven membrane processes from 2000 to 2010. This work is an extension of the previous one, and the works published between 2011 and 2020 are considered there.

First of all, it should be noted that in work [1], in the waste majority of cases, it was considered the articles published in the leading thematic journal, namely Journal of Membrane Science and Desalination by Elsevier. In contrast, the attention to the other publication was insufficient. Also, during the considered period, new thematic journals began to be issued, particularly Membranes by MDPI. Therefore, the current work analyzed the bigger number of journals and more than 1000 publications dedicated to pressure driven membrane processes simulation. The publication distribution by years is shown in Figure 1, and the distribution among the main processes is shown in Figure 2.

It can be seen from Figure 1 that despite the drop in 2017 and 2018, the number of publications stable increased, which is evident that the actuality of such kind of investigation is increased. Moreover, the trend of the increase is more clearly seen than it was in work [1]. Figure 2, in turn, shows that the most significant number of research is dedicated to the question of simulation of reverse osmosis, which is the most widely applicated industrial pressure-driven membrane process. Also, a significant number of publications are dedicated to the simulation of nanofiltration (NF) and forward osmosis

(FO), which in work [1] was not considered. A lesser number of works are dedicated simulation of ultrafiltration (UF), microfiltration (MF), and also membrane bioreactors. The last ones were not considered in work [1] as a separate kind of process.

Taking into account a large number of publications in the considered period it was decided to make a review for each process separately. Correspondingly, the purpose of the current work is the analysis and generalization of the investigations dedicated to the simulation of reverse osmosis in the period from 2011 to 2020. The objectives of the research include: (1) the review and evaluation of areas for applications of the different approaches to RO simulations; (2) the comparison of the trends in developments of methods of RO simulation in the first two decades of the XXI century; (3) the analysis perspectives of the development of the RO simulation.

The distribution of the published works dedicated to RO simulation is shown in Figure 3. The represented data demonstrate that the research interest in this direction was steadily increasing.

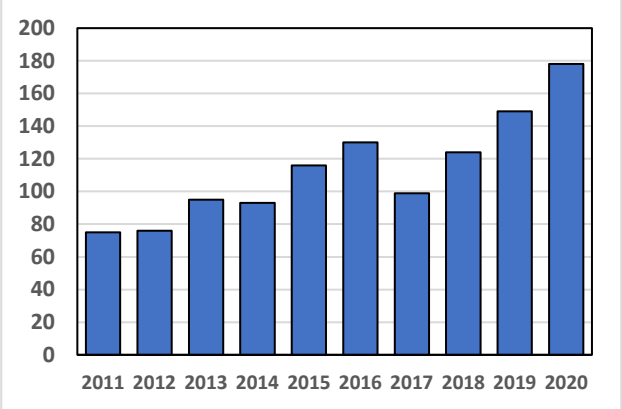


Figure 1 – The distribution of the publications selected for review by the years

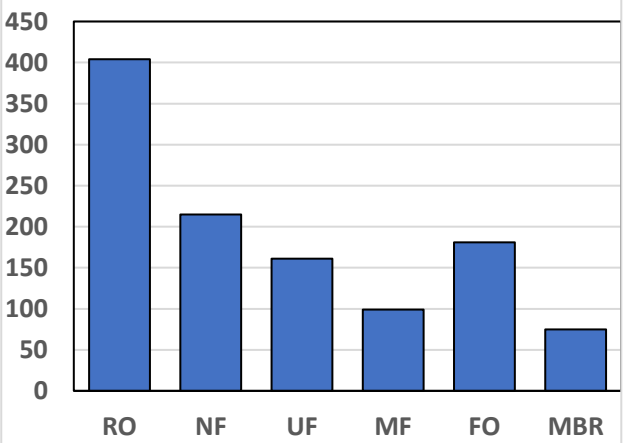


Figure 2 – The distribution the publication selected for review by the processes

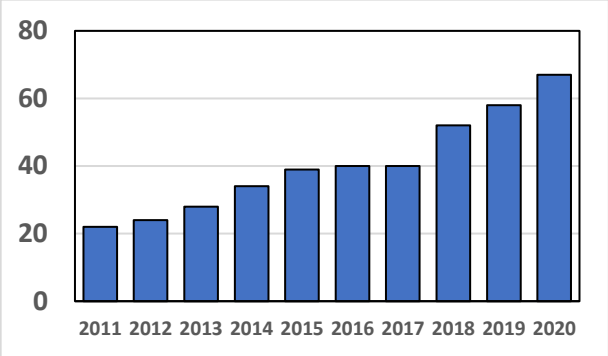


Figure 3 – The distribution of publications dedicated to the simulation of reverse osmosis by the years

As in work [1], the review does not claim comprehensiveness, however, it allows to sufficiently evaluate the main trends in the mathematical simulation of RO and the areas of application of the main approaches to the simulation.

It should be noticed that in 2011-2020 several substantial review articles were published in which the question of reverse osmosis simulation was considered. These works will be discussed below.

## 2 Research Methodology

In work [1] it was noticed that the traditional models include the following groups: irreversible thermodynamics-based models, diffusion-based models, and pore-flow-based models. The investigations with the application of computational fluid dynamics, artificial neuron networks, optimization, and economic analysis were identified as individual groups. The others approaches to the reverse osmosis simulation including semi-empirical models were also considered separately.

During 2011-2020 several reviews were published which prove the acceptability of such classifications with some corrections. In particular, in work [1] the models based on Kedem–Katchalsky equation (irreversible thermodynamics) were considered, and in work [3] detailed analysis of the irreversible thermodynamics and solution-diffusion models was made. The traditional models were also considered in the general review of RO desalination [4] and the review of the desalination processes simulation [5]. The reviewed works dedicated to the simulation with using of the computation fluids dynamics (CFD) [6-8] and molecular dynamics [9-11] were also published. Moreover, in works [12-13] it was considered both the traditional models and the molecular dynamics methods, in addition, in work [13] it was noticed that in researches until 2000 the application of the preferential sorption-capillary flow model was predominated while after 2001 the biggest progress was achieved with using of the molecular dynamic methods. The question of the RO optimization was considered in reviewed works [5, 14], and in work [15] the studies of the energy analysis of RO were revied.

In this review works the authors considered mainly modern researches, and the fraction of the works published

in the 1960s-1980s is relatively low. They significantly provide an indication of the state of art in the reverse osmosis mathematical simulation according to the individual approaches, however, no one of them did not consider all main approaches together. For example, in generalized works [12-13] the simulation using the CFD methods was not considered, and the attention to the artificial neuron networks in the reviews is almost absent. Therefore, such a review like the current work would be useful for the generalized understanding of the RO simulation and the trends of the mathematical modeling of RO.

Taking into account the works [9-11] and work [15], it was decided that except the class of models distinguished in the previous work [1] to consider individually the molecular dynamics method and energy analysis. With this, the distribution of the approaches in chosen for the review publication is shown in Figure 4.

As in the previous period, the most number of works are dedicated to the simulation with an application of CFD and optimization methods, while the number of irreversible thermodynamics-based models and pore flow-based models is still low. It was unexpected to define the relatively high number of models based on the solution-diffusion concept.

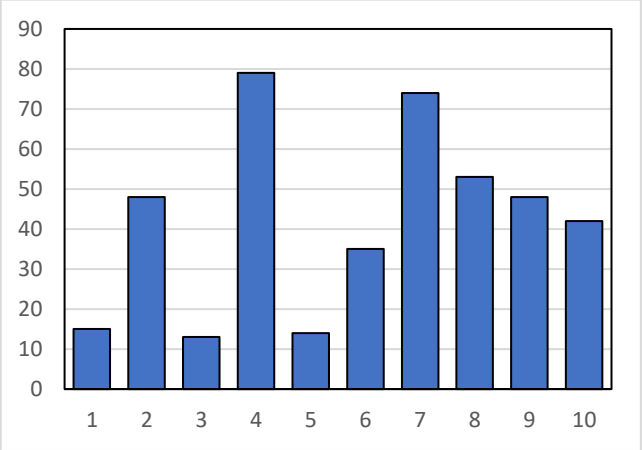


Figure 4 – The distribution of the RO models in chosen publication by classes: 1 – irreversible thermodynamics; 2 – diffusion; 3 – pore flow; 4 – computational fluid dynamics; 5 – artificial neuron networks; 6 – molecular dynamics; 7 –optimization; 8 – energy analysis; 9 – economic analysis; 10 – others models

## 3 Results

### 3.1 Irreversible thermodynamics models

The main models of this class include the Kedem–Katchalsky and the Spiegler–Kedem model, which was used in the limited extend during 2011-2020 (Figure 5). In particular. The Kedem–Katchalsky model was considered in works [2, 16-19], the works [20-27] were dedicated to the Spiegler–Kedem model, and in works [3, 28], both models were used.

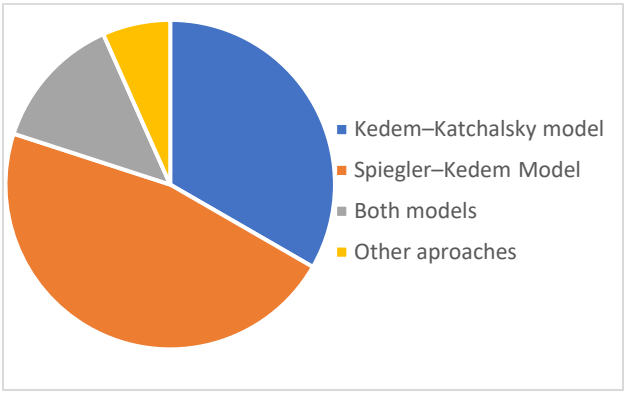


Figure 5 – The distribution of the irreversible thermodynamic based models in chosen publications

The Kedem–Katchalsky model is based on the assumption of a linear relationship between flux and potential gradient. The membrane performance, in this case, can be determined by using phenomenological coefficients [9].

The transport through the membrane of non-electrolyte binary solutions, caused by the pressure difference and also by the osmotic pressure difference, according to the Kedem–Katchalsky model can be described by the following equations [2]:

$$J_w = L_p\left(\Delta p - \sigma\cdot\Delta\pi\right) \quad (1)$$

$$J_s = \omega\cdot\Delta\pi + \left(1-\sigma\right)\cdot\overline{c}\cdot J_w \quad (2)$$

where $\Delta p$ is the applied pressure difference; $\Delta\pi$ is the osmotic pressure difference; $\overline{c}$ is the mean concentration of salt in the membrane which can be determined as mean arithmetic [2] or mean logarithm [9] value. The values $L_p$, $\sigma$ and $\omega$ are the phenomenological constants that are concentration depended.

The selectivity of process can be evaluated be using of the rejection coefficient [9]:

$$R = 1-\frac{c_3}{c_1} = 1-\frac{J_s}{J_w c_1} \quad (3)$$

where $c_1$ is the solute concentration in feed solution; $c_2$ is the solute concentration in permeate.

The osmotic pressure value can be defined by the van't Hoff equation [2, 9]:

$$\Delta\pi = RT\Delta c = RT\left(c_2 - c_3\right) \quad (4)$$

where $R$ is the universal gas constant; $T$ is the absolute temperature; $c_1$ is the solute concentration near the membrane surface.

$$\pi(i) = 1.19\left(T+273\right)\sum_1^i m(i) \quad (5)$$

where $T$ is the solution temperature; $m(i)$ – is molar concentration of ions.

On the other hand, in work [19] the osmotic pressure of glucose solution was determinated as the function of concentration:

$$\pi(c) = -RT \ln\left\{\frac{\left[\left(100 - c/M_w\right) - \left(2c/M_g\right)\right]}{\left[\left(100 - c/M_w\right) - \left(c/M_g\right)\right]}\right\} \quad (6)$$

where $R$ is the gas constant; $T$ is the solutions temperatures; $M_g$ is the glucose molar weight; $M_w$ is the water molar weight.

Also, the set of the temperature and concentration dependences of osmotic pressure is represented in the work [27].

The dependences of phenomenological constants in equations (1) and (2) from the operation condition are discussed in detail in the review work [2].

The use of the Kedem–Katchalsky model in the considered period was limited and related to the simulation of the hydrogen peroxide [16, 17], the rejection of N-nitrosamines [18] and boron [28], and also for the glucose solution concentration.

The more recent Spiegler–Kedem model describe the process of the substance transport through the membrane using the differential equations in a form [24]:

$$J_w = -A\left(\frac{dp}{dx} - \sigma\frac{d\pi}{dx}\right) \quad (7)$$

$$J_s = -B\frac{dc}{dx} + (1-\sigma)J_w\bar{c} \quad (8)$$

The rejection coefficient can be represented in a form [24]:

$$R = 1 - \frac{\left(K_{ic} - \left(D_{ip}/RT\right)V_{is}\left(8\eta/r_p^2\right)\phi\right)}{1 - \exp(-\mathrm{Pe}')\left[1 - \phi\left(K_{ic} - \left(D_{ip}/RT\right)V_{is}\left(8\eta/r_p^2\right)\right)\right]} \quad (9)$$

where $K_{ic}$ is the factor of the resistance to diffusion; $D_i$ is the diffusivity of the component $i$ in the pores; $R$ is the universal gas constant; $T$ is the absolute temperature; $V_{is}$ is the solute partial molar volume; η is the solution dynamic viscosity; $r_p$ is pore radius; ϕ is the volumetric factor; Pe' is modified Peclet number.

In work [3] the other relationship was proposed for the rejection coefficient calculation:

$$R_s = 1 - \frac{1-\sigma}{1 - \sigma\exp\left[\frac{(\sigma-1)J_w}{\omega^-}\Delta x\right]} \quad (10)$$

where σ is reflection coefficient; $\omega^-$ is the local permeation coefficient.

The osmotic pressure value is determinated in the same as in the previous case. The phenomenological coefficients (7) and (8) in contrast with the Kedem–Katchalsky model do not depend on the solvent concentration [3].

The application of the Spiegler–Kedem model also was not wide. But this model was used for the analysis of phenol rejection [22], brackish [20] and seawater [25] desalination, N-nitrosamines rejection [21, 27], boron rejection [24, 28], water purification from organic contaminant [23], and glucose concentration [24]. In works [22-23], it also noticed that the description of processes is carried out for spiral wound modules.

It also should be noticed, that the thermodynamical approach and the phenomenological equation were used in work [29]. However, the thermodynamic method were also applied for energy analysis, which will be discussed below.

However, in general, the application of the irreversible thermodynamics models, despite their relative simplicity, in considered period were limited, and the relative number substantially decreased compared with previous period, considered in work [1].

### 3.2 Diffusion based models

From Figure 4 it can be seen that in the period 2011-2020 diffusion based models were used in a quite wide range, which was unexpected, taking into account the relatively low number of publications with using this approach in the previous decade [1].

In the vast majority of cases, the solution-diffusion model was used. Its main assumption is that the skin layer is non-porous. In this case the transport of solvent and solute can be recognized as diffusion and it can be described by the equations in a form [3, 28, 30-55]:

$$J_w = A_w\left(\Delta p - \Delta\pi\right) \quad (11)$$

$$J_s = B\left(C_f - C_p\right) \quad (12)$$

where $A_w$ is the solvent penetration constant; $B$ is the solute penetration constant; $\Delta p$ is the applied pressure difference; $\Delta\pi$ is the osmotic pressure difference; $C_f$ is the solute concentration in the feed solution; $C_p$ is the solvent concentration in the permeate.

$$J_s = -D_m\frac{dC_m}{dx} = \frac{D_m}{l}\left(D_{m0} - D_{ml}\right) =$$

$$= \frac{D_m K_m}{l}\left(C_f - C_p\right) = B\left(C_f - C_p\right) \quad (13)$$

where $D_m$ is the solute diffusivity in membrane; $l$ is the active layer thickness; $C_{m0}$ is the solute concentration in membrane on the feed side; $C_{ml}$ is the solute concentration in the membrane on the permeate side; $K_m$ is the partition coefficient.

In general, in several works the relationships for determination of the constants in equation (11) and (12) are represented.

For example, in work [46] the solvent penetration constant was represented in a form:

$$A = \frac{D_i \cdot K_i \cdot c_{i0} \cdot V_i}{l \cdot R \cdot T} \quad (14)$$

where $D_i$ is the solvent diffusivity; $K_i$ is the solvent sorption coefficient; $c_{i0}$ is the solvent concentration in the feed solution; $V_i$ is the solvent molar volume; $l$ is the

membrane thickness; $T$ is the solvent temperature; $R$ is gas constant. The similar relationship was used in work [54].

The penetration constant also can be expressed by using the membrane resistance concept [30]:

$$A = \frac{1}{R_m \cdot \mu} \tag{15}$$

where $R_m$ is the membrane resistance; $\mu$ is the solvent dynamic viscosity coefficient.

Although, equation (14) directly includes the temperature, in several works for taking into account this parameter influence in the RO process the others relationships were applied. For example, in work [31] for this purpose, the change in the viscosity with temperature was used. In this case, the equations for the penetration coefficients became:

$$A(T) = \frac{A_0 \cdot \mu(T_0)}{\mu(T)} \tag{16}$$

$$B(T) = \frac{B_0 (T + 273{,}15) \cdot \mu(T_0)}{T_0 \cdot \mu(T)} \tag{17}$$

where $A_0$, $B_0$ are the coefficient values under base temperature.

In others works, the more complex relationships were used. Thus, in work [28] following equations were proposed:

$$A_w = A_0 e^{\left(-\frac{E_A}{R}\left(\frac{1}{T}-\frac{1}{298{,}15}\right)\right)} \left(\frac{10^{-3}}{24 \cdot 60 \cdot 60}\right) \tag{18}$$

$$B_s = B_0 e^{\left(-\frac{E_{Bst}}{R}\left(\frac{1}{T}-\frac{1}{298{,}15}\right)\right)} \left(\frac{10^{-3}}{24 \cdot 60 \cdot 60}\right) \tag{19}$$

where $E_A$, $E_{Bst}$ are the activation energies of the solvent and solute molecules transport through the membrane; $R$ is gas constant.

At the same time, in work [35] for water solution it was used equations in a form:

$$A_{w,T} = 9.059 \cdot 10^{-7} \left(\frac{T}{25}\right)^{0{,}62} \left(\frac{36.0 \cdot 10^5 Q_f}{400}\right)^{-0.1447} \tag{20}$$

$$B_{s,i} = B_{s.i.\mathrm{Ref}} \exp\left(0.098\left(T - T_{\mathrm{Ref}}\right)\right) \tag{21}$$

The equation (20) was also used in work [37].

In work [57] the influence of the temperature was taken into account with following relationships:

$$A_{w(T)} = A_{w(25°\mathrm{C})} \exp\left[0.0343\left(T - 25\right)\right] \text{ at } T < 25°\mathrm{C} \tag{22}$$

$$A_{w(T)} = A_{w(25°\mathrm{C})} \exp\left[0.0307\left(T - 25\right)\right] \text{ at } T > 25°\mathrm{C} \tag{23}$$

$$B_{s(T)} = B_{s(25°\mathrm{C})} \left(1 + 0.08\left(T - 25\right)\right) \text{ at } T < 25°\mathrm{C} \tag{24}$$

$$B_{s(T)} = B_{s(25°\mathrm{C})} \left(1 + 0.05\left(T - 25\right)\right) \text{ at } T > 25°\mathrm{C} \tag{25}$$

The influence of the temperature was taken into account with using of the temperature correction factor. Thus, in work [58] the following equation was used:

$$A_{w(T)} = A_{w(25°\mathrm{C})} \cdot TFC \cdot F_f \tag{26}$$

where $A_{w(T)}$ is the penetration coefficient under base temperature (25°C); $TFC$ is the temperature correction factor; $F_f$ is the fouling factor.

The analogical relationships were used in works [59-60], and in work [43] the more detailed form was applied:

$$J_w = A \cdot TFC \cdot FF \cdot \left[\left(P_F - P_P - \frac{\Delta P_f}{2}\right) - \left(\pi_w - \pi_p\right)\right] \cdot E + 6 \tag{27}$$

Also, in work [43] the relationships for the temperature correction factor were proposed:

$$TFC_P = \exp\left[0.0343\left(T - 25\right)\right] < 25°\mathrm{C} \tag{28}$$

$$TFC_P = \exp\left[0.0307\left(T - 25\right)\right] < 25°\mathrm{C} \tag{29}$$

In work [20] the long-term performance of RO plant and for the changes of the penetration coefficient in time the following relationship was proposed:

$$A_n = \delta_1 \cdot e^{-\frac{t}{\tau_1} \cdot k_{fp}} + \delta_2 \cdot e^{-\frac{t}{\tau_2} \cdot k_{fp}} \tag{30}$$

Also, in work [49] except the temperature impact the influence of pressure were taken into account with following equations:

$$A_w = A_{w_0} \exp\left(\alpha_1 \frac{T_1 - 273}{273} - \alpha_2 \left(P_f - P_d\right)\right) \tag{31}$$

$$B_S = S_{S_0} \exp\left(\beta_1 \frac{T - 273}{273}\right) \tag{32}$$

Obviously, all represented relationships have a limited range of applicability, and the condition of the validity of the equations should be checked in the corresponding publications.

The osmotic pressure values can be calculated in the same way as in the case of irreversible thermodynamics (equations (4) through (6)), and also in some works, the other methods of this parameter calculation were considered.

For example, in work [61] the van't Hoff equation were written in a form:

$$\pi = \frac{i \cdot \Re}{M \cdot 10^5} \cdot T \cdot C \tag{33}$$

At the same time, in work [48] the following relationship was proposed:

$$\Delta\pi = RT\left(\frac{J_s}{B_s}\right) \tag{34}$$

Also, the empirical temperature and concentration dependences for several substances are represented in works [35, 38, 55, 59].

For the evaluation of the selectivity both the rejection coefficient (equation (4)) and the solvent concentration in permeate were used.

For the rejection calculation were proposed following relationships:

in work [38]:

$$R_i = \left[1 + \frac{AC_{wp}}{B(\Delta p - \Delta\pi)}\right]^{-1} \quad (35)$$

where is the water concentration in permeate.

in work [47]:

$$R = \frac{A(\Delta p - \Delta\pi)}{A(\Delta p - \Delta\pi) + B} \quad (36)$$

Also, the relationship between the rejection coefficient and other parameters can be represented in a form [41]:

$$B = \frac{(1-R)\cdot J_w}{R} \quad (37)$$

or [54]:

$$\frac{1}{R} = B\frac{1}{J_w} + 1 \quad (38)$$

Also, the relationships for the solute concentration in permeate were proposed. In work [38] the following equation was used:

$$C_P = \frac{C_f B}{B + \frac{J_w}{\exp(J_w / k)}} \quad (39)$$

where $k$ is the mass transfer coefficient.

A similar form of equation was applied in works [40], [48], and [53]. At the same time, in work [59], the temperature correction factor and fouling factor and with the accounting of them, the equation for the solvent concertation in permeate calculations became:

$$C_P = B\cdot FF\cdot TCF\cdot\frac{1}{J_w}\cdot\left(\frac{C_f\cdot(1+CF)}{2}\right) \quad (40)$$

where $CF$ is the concentration factor.

For more accurate analysis of the process the solution-diffusion model was complemented. In particular, in works [33-35, 49, 53, 56] during the RO process simulation the concertation polarization was taken into account (mainly by application of the film model). Moreover, the model was complemented by the material balance [49, 53, 55, 62], the optimization methods [35, 45, 61, 63], unsteady-state conditions [32], and by the fouling impact [39]. Also, in work [64] for the description of the processes in spiral wound membrane module the cylindrical coordinate system was used.

The solution-diffusion model was primarily used for the description of the water purification [30, 33, 36, 38, 47, 50, 64, 65], including sea water [25, 40, 41, 43, 49, 52, 55, 57, 61, 66] and brackish water [20, 31, 42, 45, 58, 59, 60, 65] desalination, and also the wastewater treatment [30, 48, 51, 53-54, 56, 67]. Also, with using of this model it was described the processes of the removal of zinc [32], boron [28, 52], chlorophenol [36, 38], N-nitrosodimethylamine [38, 63], weak acids [25], and ammonia compounds [51, 54], apple juice concentration [35, 37], and also for the membrane characterization [46]. The solution-diffusion model was also widely used for the analysis of the hybrid membrane systems performance [31, 41, 42-43, 48, 55, 57, 61, 65, 69-70]. In most cases, the processes were analyzed in the spiral wound membrane modules [33, 34-35, 37, 39, 58-60, 64, 68], the hollow fiber modules [62] and laboratory cells were also considered.

For the mathematical models' solutions, the most often used software includes MATLAB [32, 36, 52, 60], ROSA [39], and the programing language C++ [36].

The other models of this class were used much less often. Thus, the solution-diffusion-imperfection model was considered in works [71, 72], and the extended solution diffusion model was applied in work [72].

The solution-diffusion-imperfection model takes into account the possibility of the convective transport of substances through possible pores (imperfections) in the membrane active layer. In this case, the equations for the calculation of the solvent and solute fluxes will be written in a form [71]:

$$J_w = A\cdot(\Delta p - \Delta\pi) + L\cdot\Delta p \quad (41)$$

$$J_S = B\cdot(C_f - C_P) + L\cdot\Delta p\cdot C_f \quad (42)$$

where $L$ is the leakage factor.

The rejection coefficient can be calculated by the equation [72]:

$$R = \left[1 + \left(\frac{B}{A}\right)\left(\frac{1}{\Delta p - \Delta\pi}\right) + \left(\frac{L}{A}\right)\left(\frac{\Delta p}{\Delta p - \Delta\pi}\right)\right] \quad (43)$$

The extended solution-diffusion model takes into account the influence of the applied pressure on the solute transport, which was not considered in the classical solution-diffusion model. In this case, the equations become [72]:

$$J_w = A(\Delta p - \Delta\pi) \quad (44)$$

$$J_s = B(C_f - C_p) + L_{sp}\Delta p \quad (45)$$

here $L_{sp}$ is the phenomenological coefficient.

The rejection coefficient can be calculated by the equation [72]:

$$\frac{1}{R}\left[1 - \frac{L_{sp}}{C_f A}\left(1 - \frac{\Delta\pi}{\Delta p}\right)\right] = 1 + \frac{K_s D_s}{A(\Delta p - \Delta\pi)} \quad (46)$$

The unexpected wide range of the solution diffusion models applications in the considered period is probably related to its relative simplicity, because of what there is no necessity for the application of the complex computational techniques, and also the convenience of the model for the hybrid system analysis and the model-based optimization. It should be noticed that the significant contribution in this field was done by one research group [3, 34-38, 57-58, 63]. At the same time, the solution-diffusion-imperfection model and the extended solution diffusion model do not have these advantages, therefore, as in the previous period, they were applied in a limited range.

### 3.3 Pore flow based models

As in the previous period [1], the models of this class were used rarely, yielding to the computational fluid dynamics methods. Thus, among the large number of publications dedicated to the RO simulation the models which directly considered the pore flow are mentioned in less than 5% of cases. In most cases, the preferentially sorption pore flow model (also known as Kimura–Sourirajan model) was considered. Also, the surface force-pore flow model and some other approaches were applied (Figure 6).

Unlike the solution diffusion model, the models of this class consider the active layer as porous, and the transport is carried out by both diffusion and convection [1].

The preferentially sorption pore flow model describes the solute and solvent fluxes by the equations in a form [73]:

$$J_w = A_w\left(\Delta p - \Delta\pi\right) \tag{47}$$

$$J_s = B_s\left(C_m - C_p\right) \tag{48}$$

Such equations have a similar form to solution-diffusion models, which also can be seen in works [46, 74-75]. However, as mentioned in the previous review [1], the principal difference is in the nature of the transport coefficients, which are determined by the different conceptions of the active layer structure. Unfortunately, due to the low number of publications in the considered period, the making of a detailed review of the parameters of the preferentially sorption pore flow model does not seem possible.

For the evaluation of the selectivity the concertation in permeate may be used. It can be calculated by the equation [73-75]:

$$J_s = B_s\left(C_m - C_p\right) \tag{49}$$

Also, in work [73], the recovery ratio, which was determined as the permeate and feed ratio, was calculated:

$$R = \frac{Q_P}{Q_f}\cdot 100\% \tag{49}$$

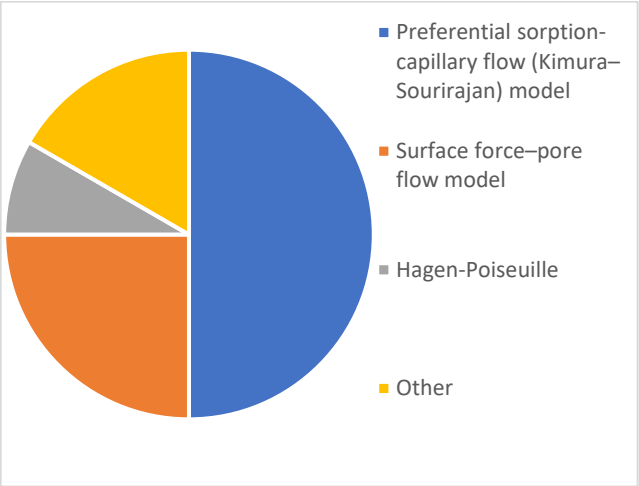


Figure 6 – The distribution of the pore flow based model in chosen publications

This model was complemented by the balance equations [73] and the optimization methods [74-75]. It was primarily used for the description of seawater desalination [74-76] and industrial effluent purification [73, 77], and also for the determinations of the characteristics and internal structure of the membrane [46]. This model was also used for the analysis of the hybrid system [76].

Also, in a few works, the surface force-pore flow model was found. This model considers the separate pore, which dimensions are characterized by the dimensionless coordinates [78-79]:

$$\rho = \frac{r}{R_w} \tag{51}$$

$$\zeta = \frac{z}{\tau} \tag{52}$$

where $r$ is the cylindrical coordinate perpendicular to the pore wall; $R_w$ is the pore radius; $z$ is the cylindrical coordinate parallel to the pore axis; $\tau$ is the average pore length.

Taking this into account, the differential equation describing the velocity profile in pores can be written in a form [78-79]:

$$\left[\frac{d^2\alpha(\rho)}{d\rho^2} + \frac{1}{\rho}\frac{d\alpha(\rho)}{d\rho}\right] + \frac{1}{\beta_1}\left[\frac{\Delta P}{\pi_2} - \frac{\pi_2\sigma_2(\rho) - \pi_3\sigma_3(\rho)}{\pi_2}\right] - \\ - \frac{1}{\beta_1}\left(1 - \frac{1}{b(\rho)}\right)\left[\alpha(\rho) + \omega(\rho)\right]\times \\ \times\left[1 + \frac{1 - \left(\frac{\pi_3}{\pi_2}\right)k\cdot\rho}{\exp\left[\alpha(\rho) - \omega(\rho)\right] - 1}\right]\exp\left(-\Phi(\rho,0)\right) = 0 \tag{53}$$

The physical meaning of the parameters in the equation (51) is described in detail in works [78-79].

The boundary conditions herewith are following [78-79]:

$$\alpha(\rho)=0 \quad \text{at} \quad \rho=1 \tag{54}$$

$$\frac{d\alpha(\rho)}{d\rho}=0 \quad \text{at} \quad \rho=0 \tag{55}$$

The solution of the equation (51) allows to represent the solvent and solute fluxes in a form [78-79]:

$$N_A=\frac{2}{X_{AB}}\left(\frac{\varepsilon}{\tau}\right)\times$$

$$\times\int_0^{1-\lambda}\frac{\left[\alpha(\rho)+\omega(\rho)\right]}{b(\rho)}\left(\pi_2+\frac{\pi_2-k\cdot\rho\cdot\pi_3}{\exp\left[\alpha(\rho)-\omega(\rho)\right]-1}\right)\times$$

$$\times\exp\left(-\Phi(\rho,0)\right)\rho d\rho \tag{56}$$

$$N_B=\frac{2}{X_{AB}}\left(\frac{\varepsilon}{\tau}\right)CRT\int_0^1\alpha(\rho)\rho d\rho \tag{57}$$

The solute concentration in permeate can be calculated by the equation [78-79]:

$$C_{A3}=C\left[1+CRT\frac{l_1}{l_3}\right]^{-1} \tag{58}$$

Moreover, in work [80] the following equation for the rejection coefficient calculation was proposed:

$$R=1-\frac{\Phi K_c}{1-(1-\Phi K_c)\exp(-\mathrm{Pe})} \tag{59}$$

where Pe is the Peclet number.

In contrast with previous models, the surface force-pore flow model is complex and requires complex solution methods without visualization, the distinctive for the CFD methods. This determined the narrow range of this model application which was only used for the description of the ground waters purification from pesticides [80].

Also, in work [81] for the description of the pressure driven membrane processes including RO the Hagen-Poiseuille equation was considered:

$$u(r)=-\frac{1}{4\mu}\frac{dp}{dx}\left(R^2-r^2\right) \tag{60}$$

However, this approach is more suitable for the simulation of the ultrafiltration and microfiltration.

Despite that, the models of this class take into account both diffusive and convective transport through the membrane, in work [82], the analytical solution-diffusion pore-flow model was presented. Also, in work [83] the similarity of the processes in the membrane with convective heat transfer was considered. However, that approaches were used rarely.

### 3.4 Computational fluid dynamics based models

As mentioned in work [1], the flow conditions have a significant influent on the operation of the processes during membrane separation, including mass transfer, concentration polarization, and fouling layers formation, the pressure drop in the membrane channels. Taking into account the critical importance of the hydrodynamic conditions not only for the membrane processes but for engineering in general, the development of the method for the mathematical description and analysis of the flow named computational fluid dynamics (CFD) is natural. The method itself is based on the mathematical description of the fluid flow, which includes the Navier-Stokes equation, the continuity equation, and the mass and energy conversation equation [84].

For the steady state laminar flow of the Newtonian fluid these equations can be written in a form:

- the continuity equation [84]:

$$\frac{\partial u}{\partial x}+\frac{\partial v}{\partial y}+\frac{\partial w}{\partial z}=0 \tag{61}$$

- the Navier-Stokes equation [84]:

$$u\frac{\partial u}{\partial x}+v\frac{\partial u}{\partial x}+w\frac{\partial u}{\partial z}=-\frac{1}{\rho}\frac{\partial P}{\partial x}+\frac{\mu}{\rho}\left[\frac{\partial^2 u}{\partial x^2}+\frac{\partial^2 u}{\partial y^2}+\frac{\partial^2 u}{\partial z}\right] \tag{62}$$

$$u\frac{\partial v}{\partial x}+v\frac{\partial v}{\partial x}+w\frac{\partial v}{\partial z}=-\frac{1}{\rho}\frac{\partial P}{\partial x}+\frac{\mu}{\rho}\left[\frac{\partial^2 v}{\partial x^2}+\frac{\partial^2 v}{\partial y^2}+\frac{\partial^2 v}{\partial z}\right] \tag{63}$$

$$u\frac{\partial w}{\partial x}+v\frac{\partial w}{\partial x}+w\frac{\partial w}{\partial z}=-\frac{1}{\rho}\frac{\partial P}{\partial x}+\frac{\mu}{\rho}\left[\frac{\partial^2 w}{\partial x^2}+\frac{\partial^2 w}{\partial y^2}+\frac{\partial^2 w}{\partial z}\right] \tag{64}$$

- mass conversation equation [84]:

$$u\frac{\partial C}{\partial x}+v\frac{\partial C}{\partial y}+w\frac{\partial C}{\partial z}=D\left[\frac{\partial^2 C}{\partial x^2}+\frac{\partial^2 C}{\partial y^2}+\frac{\partial^2 C}{\partial z^2}\right] \tag{65}$$

In these equations: *u*, *v*, *w* are the velocity projections on the coordinate axes; *P* is the pressure; *C* is the solute concentration; ρ is the feed solution density; μ is the feed solution viscosity; *D* is the diffusivity.

The equations are also represented in a similar way in works [85-97], including written for the two-dimension simplification [89, 91, 96], in the cylindrical coordinates [90], and matrix form [93]. Moreover, in works [93-94], the unsteady-state conditions were taken into account.

However, the equations (61) – (65) due to their awkwardness are often written in an operator form with using of the differential operators, including the full derivative operator, the Hamilton operator, the Laplace operator etc., and also the rules of the vector and tensor analysis. In this case, the equations of the mathematical model of the fluid flow can be written in a form:

- the Navier-Stokes equation [98]:

$$\rho(\bar{u}\cdot\nabla)\bar{u}+\nabla P-\mu\nabla^2\bar{u}=0 \tag{66}$$

- the continuity equation [98]:

$$\nabla \cdot \bar{u} = 0 \quad (67)$$

- mass conversation equation [98]:

$$D\nabla^2 C = \bar{u} \cdot \nabla C \quad (68)$$

The equations are written in a similar way, for example, in works [6, 99–126].

For accounting of the unsteady-state conditions these equations can be rewritten in a following way [127]:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u) = 0 \quad (69)$$

$$\frac{\partial (\rho u)}{\partial t} + (u \cdot \nabla) \rho u = -\nabla p + \nabla \cdot (\mu \nabla u) \quad (70)$$

$$\frac{\partial C}{\partial t} + (u \cdot \nabla) C = \nabla \cdot (D \nabla C) \quad (71)$$

The similar equations are in works [128-129].

Also, the equations are represented in the reviews [6-8].

As it was mentioned above, equations (60)-(64) are suitable for the laminar flow. However, in the channels of the membrane apparatuses, there exist favorable conditions for turbulence development. Therefore, the models of flow are often supplemented by turbulence models. The k-ω and k-ε models are most widely used. The distribution between them is represented in Figure 7.

The k-ε turbulence model operate by the terms of the specific turbulent kinetic energy k and the dissipation rate ε. The equations of this model are following [138]:

$$\frac{\partial (\rho k)}{\partial t} + \nabla \rho \bar{u} k = \nabla \left[ \left( \mu_k + \frac{\mu_{eff}}{\sigma_k} \right) \nabla k \right] + \mu_{eff} S^2 - \rho \varepsilon \quad (72)$$

$$\frac{\partial (\rho \varepsilon)}{\partial t} + \nabla \rho \bar{u} \varepsilon = \nabla \left[ \left( \mu_k + \frac{\mu_{eff}}{\sigma_k} \right) \nabla \varepsilon \right] +$$

$$+ C_1 S \rho \varepsilon - C_2 \frac{\rho \varepsilon^2}{k + \sqrt{\nu \varepsilon}} \quad (73)$$

The physical meanings of the parameters in equations (72) and (73) are described in work [138].

This model was also used, for example, in works [91, 137, 139].

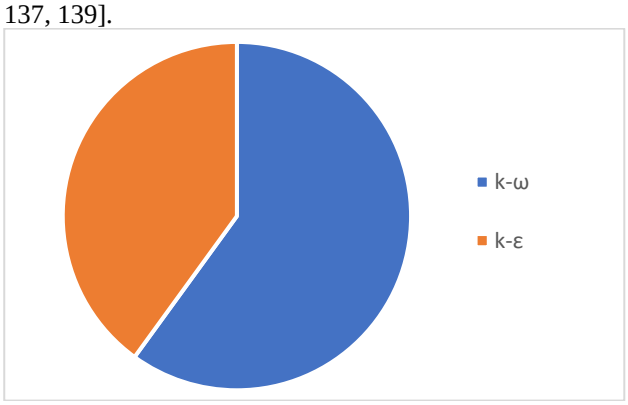


Figure 7 – The distributions of the main turbulence models in the chosen articles

In contrast, the other model uses the value of the specific dissipation rate ω instead of ε. This model can be represented in a form [97]:

$$u_j \frac{\partial u_i}{\partial x_j} = -\frac{1}{\rho} \frac{\partial P}{\partial x_i} + \frac{1}{\rho} \frac{\partial}{\partial x_j} \left( (\mu + \mu_t) \frac{\partial u_i}{\partial x_j} \right) \quad (74)$$

where the turbulent viscosity can be calculated by following equation [97]:

$$\mu_t = \rho \frac{a_1 k}{\max (a_1 \omega; \Omega F_2)} \quad (75)$$

The equations for calculations of the k and ω values according this model are following [97]:

$$u_j \frac{\partial (\rho k)}{\partial x_j} = \tau_{ij} - \beta^* \rho \omega k + \frac{\partial}{\partial x_j} \left[ (\mu + \sigma_{k1} \mu_t) \frac{\partial k}{\partial x_j} \right] \quad (76)$$

$$u_j \frac{\partial (\rho k)}{\partial x_j} = \frac{\gamma}{\nu_t} \tau_{ij} \frac{\partial u_i}{\partial x_j} - \beta \rho \omega^2 + \frac{\partial}{\partial x_j} \left[ \left( \mu + \sigma_\omega \mu_t \frac{\partial \omega}{\partial x_j} \right) \right] +$$

$$+ 2\rho (1 - F_1) \sigma_{\omega 2} \frac{1}{\omega} \frac{\partial k}{\partial x_j} \frac{\partial \omega}{\partial x_j} \quad (77)$$

This model was also used, for example, in works [85-88, 97, 126].

Since the equations of models are differential, the starting and boundary conditions are needed for the obtain the result. These conditions strongly depend from the geometrical and physical conditions of the considered process, for example, for the equations (61) – (65) for the case of the fluid flow in the spiral wound membrane channel with the spacer in work [84] the following boundary conditions were applied:

- inlet boundary conditions:

$$C = C_0; u = \frac{m}{\rho A_1}; v = w = 0 \quad (78)$$

- membrane boundary conditions:

$$C = C_m; u = v = w = 0 \quad (79)$$

- filament boundary conditions:

$$\frac{\partial C}{\partial n} = 0; u = v = w = 0 \quad (80)$$

- symmetry face boundary conditions:

$$\frac{\partial C}{\partial z} = 0; \frac{\partial u}{\partial z} = \frac{\partial v}{\partial z} = \frac{\partial w}{\partial z} = 0 \quad (81)$$

- outlet boundary condition:

$$P = P_0 \quad (82)$$

Also, the boundary conditions are described in details, for example, in works 89-90, 92, 109, 117, 127-132, 134, 140], and also in the review work [8].

Due to the complexity of the equations described above, the mathematical models should be solved by numerical methods, mainly by using specialized software. In work [1], it was noticed, that for the case of RO, the most widely used commercial software included ANSYS FLUENT and ANSYS CFX, however, in the considered period, the range of applied software was wider. The distribution of them in chosen publications is shown in figure 8. Since the ANSYS software is still the most widely used, it is reasonable to consider the ratio between the main algorithms (FLUENT and CFX) that are shown in figure 9. Thus, the ANSYS FLUENT was most widely used, in particular, in works [90-91, 95-96, 107, 110, 116, 118, 124, 126-127. 133, 137-139, 141-145]. The ANSYS CFX was applied to a lesser extent, for example in works [85-88, 93, 113, 128-131, 134, 146-147, 121]. Moreover, the ANSYS was used without pointing out the algorithm in works [84, 99, 121, 123]. Among the other software un should be noticed the wide range of applications of COMSOL Multiphysics, for example, in works [98-99, 101-106, 111-112, 119, 122-123, 148], and open-source product OpenFOAM [92, 94, 114, 132, 135, 137].

The other software, including MATLAB [102, 104, 117], ROSA [149-151], NUMECA [152], MUSUBI [153], was used rarely. Also, in work [154], it was carried out the analysis of the possibility of applying the RO seawater desalination by the commercial systems CORMIX, VISUAL PLUMES, and VISJET. Meanwhile, the popular engineering system SolidWorks was not used almost. Only in work [121], the geometry was created in this system, whereas the calculations were carried out in ANSYS.

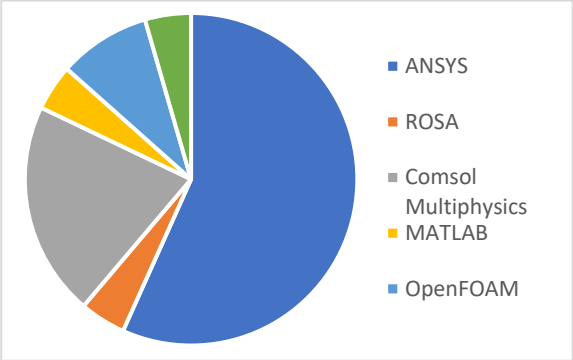


Figure 8 – The application of the software for the RO simulations

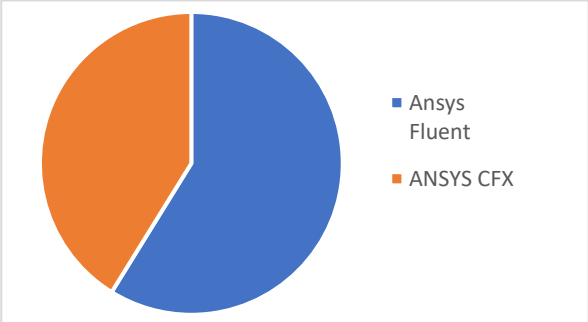


Figure 9 – The application of the algorithms of the ANSYS software for RO simulation

In some works, namely [23, 89, 97, 100, 108-109, 140, 155-157], it was not pointed out the applied software or mentioned that the algorithm of the mathematical model solution was designed independently.

Taking into account the importance of the geometry of the channels for the simulation by the CFD method, great attention was dedicated to the module design. As in others approaches, the greatest number of researches were carried out for the spiral wound membrane modules, in particular in works [6, 23, 84, 88, 98, 101, 104, 106, 110, 112-114, 117, 121, 141, 149, 151, 155-157]. In this case, it should be noticed, that in work [149] it was declared that the influence of the channel curvature is significant and the neglect of this factor can lead to a significant mistake. At the same time, the channels of the spiral wound membrane modules were conditionally rolled out into the plane, in particular in works [6, 88-89, 104]. Also, in a significant number of works the hollow fiber modules were considered [86, 106, 116, 121, 125-126, 137, 139], whereas the plate and frame modules were considered in a lesser number of the researches [91, 108-109]. The same situation is observed for the tubular modules [127, 137], and also for the multichannel ceramic modules [124] and similar designs [152]. Also, in some works the original designs were considered including rotating [90, 142] and test and lab cells [98, 135, 143]. The works, in which the patterned [111] and corrugated [97] membrane surface, should be noticed individually.

Taking into account the importance of the spiral wound modules and the correspondingly large number of studies, it was expected, that a large number of the investigations by CFD methods were dedicated to the influence of spacers on the process performance. Thus, in work [147], the important parameters, which impact the process, were pointed out. They include the angle between the spacer filament, the spacer thickness, and the filament placement in the channel. In some works, for example [92, 141, 144, 155] the commercial spacers designs were considered. Also, it should be noticed, that in works [87, 92, 95] the spacer filament was considered cylindrical, and in work [144] it was considered with the elliptical cross-section. Therefore, along with this, it should be pointed out the works [104[ and [105], in which the real geometry of the spacer was determined by the microscope measurements and X-ray computer tomography and reproduced in the CFD programs. Moreover, in the result of the comparison with the ideal geometry, in work [104] it was mentioned, that the cylindrical form of the spacer filament is a good approximation, whereas in work [105] it was pointed out the possible inaccuracies (in particular the overestimation of the pressure drop), which can be obtained without taking into account the real geometry of spacer filament. The comparison of the wound and non-wound spacers was another direction of the investigations, as was shown in works [84, 103, 153]. In this case, in work [103], it was pointed out, that the wound geometry has advantages, while in work [153] it was shown the bigger pressure drop in the channel with the wound spacers. Great attention was dedicated to the spacer filament placement by the channel width, in particular in works [96, 100, 114, 119, 129, 134].

At that, in most cases, it was pointed out, that the pest results were obtained for the zigzag placement of the filament. The influence of the angle between spacers was investigated in works [87-88, 106, 145, 155-157]. In work [1] it was pointed out, that the determination of the optimal angles is still debatable. The result of the consideration of the mentioned publication confirms this statement. In contrast with the previous period, in 2011-2020, it was proposed a large number of novel spacer designs, in particular in many studies the design with the different thicknesses of the filament between the spacer nodes, in particular in works [99, 101, 102, 112, 113, 118]. Also, it was proposed the sinusoidal spacer design [123] and the design with the screw thread on the filament [107]. Despite the benefits of the proposed designs, there are some doubts about the possibility of realizing these geometrical forms for the manufacturing of the real spacers taking into account their real thickness. The same doubts are also related to the original design of the hollow fibers with the specific cross-section, with was described in the work [116]. These doubts are appearing from the accounting of the results of the comparison of the real and ideal geometry, carried out in works [104-105]. Moreover, in work [148] it was also pointed out the significant influence of the deviations from ideal geometry for the multibore polymer membrane modules. It also should be noticed the application of the multilayer spacers [121, 130]. The novel designs of the spacers were also considered in the review [8].

In works using the CFD method, the separation process for which the simulation is carried out is not often pointed out, however, in most cases, this is about the desalination processes [85-87, 101, 108-109, 152].

In general, the mathematical simulation of the RO using CFD methods, as was predicted in work [1], is intensively developing and, likely, in the near future, will remain the most effective and widely used.

### 3.5 Artificial neural networks based models

In work [1], among the novel approaches except the CFD methods, the artificial neuron networks (ANN) were considered. Such systems are built by analogy with the architecture of the biological nervous systems, which mainly consist of simple nerve cells or neurons, which work in parallel for the simplification of quick decisions. Similarly, the neuron networks are made from a large number of primitive processing elements, which are organized in a massive parallel set. Then, in the neuron networks the artificial connections (synapses), connect these elements. They are characterized by the set of the weight coefficients, which could be updated in the learning proves [79, 158-160]. Thus, the artificial neuron networks are the computational method for comparing the input and output data from the process be the nonlinear regression model. The ANN method has the ability to analyze the relationship between input and output data based on the functions of the biological neuron networks, as mentioned above. The simple architecture of the ANN includes three-layer namely, input, hidden, and output. The input signal received from an external source is multiplied by the mentioned above weight coefficients. When the result of the multiplication exceeds the threshold, the signal would be released and sent to the output depending on the ANN activation function. In this respect, three stages, namely training, testing, and validating, with several computational operations are applied to achieve the desired goal through ANN [161].

The advantages of the simulations using the ANN include [160]: (1) the ability of the simulation of complex relationships, which is impossible for the models based on conventional mathematical methods; (2) in many cases the ANN has higher accuracy than conventional mathematical models; (3) the ANN can be adapted with new data and modernized with them; (4) the best combination of the design parameters in the ANN can be obtained by the trial and error method.

In each stage of the ANN, the main computational operation has the following form [161]:

The equations of the output signal for any hidden or output layer are determinated by the following equations [161]:

$$a_1 = \sum_{i=1}^{i=n} \left( W_i X_i + b1 \right) \tag{83}$$

$$a_2 = \sum_{j=1}^{j=k} \left( W_j a_1 + b2 \right) \tag{84}$$

where $a_1$, $a_2$ are the output signals of the first and second layer correspondingly; $X$ is input parameter; $W$ and $b$ are the weight coefficients and bias, correspondingly [161].

The activation function is used for the data normalization in range [0 -1] and [-1 1], which is discrete or continuous in a form of sigmoid [161]:

$$f\left(s\right) = \frac{1}{1+e^{-s}} \tag{85}$$

or in a form of the hyperbolic tangent:

$$f\left(s\right) = \left(e^{s} - e^{-s}\right) / \left(e^{s} + e^{-s}\right) \tag{86}$$

$s$ is the value of the input parameter. The network may ne linear on nonlinear in the activation function [161]:

$$f = \begin{cases} 1 \text{ if } s > 0 \\ -1 \text{ otherwise} \end{cases} \tag{87}$$

The normalization equation has following form [161]:

$$s_i = \frac{0.8}{d_{\max} - d_{\min}} \left( d_i - d_{\min} \right) + 0.1 \tag{88}$$

$d_{\min}$, $d_{\max}$ and $d_i$ are minimum, maximum and $i$ value of the input/output data correspondingly. The predicted or target equation can be represented in a form [161]:

$$Y = \sum_{j=1}^{j=k} f_2 \left( W_j f_1 \left( \sum_{i=1}^{i=n} W_i X_i \right) \right) \tag{89}$$

$Y$ is predicted output value. The weight matrices are calculated by applying an error back-propagation method.

The accuracy of the weight coefficient depends on the minimal error ($E$) of the output $\{z(k);\ 1 \le k \le K\}$ on the learning stage and calculated using the sum of the square error [161]:

$$E = \sum_{k=1}^{K} \left[ e(k) \right]^2 = \sum_{k=1}^{K} \left[ d(k) - z(k) \right]^2 = \\ = \sum_{k=1}^{K} \left[ d(k) - f\left( W_x(k) \right) \right]^2 \quad (90)$$

where $[W_x]$ is the weight matrices; $[x]$ is input vector; and $d$ is desired target value.

In general, there are three criteria used for the evaluation of the ANN performance, namely coefficient of correlation (r), mean square error, and mean absolute error [162].

The ANN are especially effective when it is difficult to develop mechanistic models [161].

In some works, the ANN were used together with other methods, in particular with the solution-diffusion model [158, 163], the surface force-pore flow model [79], the CFD methods [164], and also with the surface response methodology [166]. Moreover, in work [79], it was noticed, that the ANN predictions were more accurate than the surface force-pore flow model ones. Also. The ANN were applied for consideration of the systems with a high level of fouling [167-168], moreover, in work [168], it was mentioned that mechanistic models oversimplify the fouling phenomena.

In most cases, the ANN methods of simulation were used for the analysis of the desalination processes [158, 160, 162, 166, 168-169] and the water and wastewater treatment [161, 163, 165, 167].

In general, during 2011-2020, ANN were used for the RO process simulation in a limited range. The likely reason for this is the high enough level of the development of mechanistic models, in particular, the suitability of the CFD methods for these purposes. The ANN and be used the most effectively in systems with a high level of fouling [167-168] or systems with nonconventional energy sources [163, 169].

### 3.6 Molecular dynamics based models

In addition to the CFD and ANN methods, the molecular dynamics (MD) methods are a relatively new approach to the RO process. As in the case of CFD, this method is based on the numerical solution of the Newton equations of motion, but on the molecular level [10].

The concept of classical molecular dynamics is based on Newtonian mechanics. According to the second Newton law, the relationship among the mass, acceleration, and force excreted to the particle i is described by the equations [10]:

$$F_i = m_i a_i = m_i \frac{d^2 r_i}{dt^2} \quad (91)$$

where $m_i$ is the particle mass; $a_i$ is the acceleration; $F_i$ is the force excreted to the particle; $r_i$ is the distance between particles; $t$ is the time.

The Newtonian force also can be expressed in a form of the gradient of the potential energy [10]:

$$F_i = -\nabla_i U(r_i) = -\frac{\partial U(r^N)}{\partial r_i} \quad (92)$$

Using the relationships, represented by equations (90) and (91), it is possible to obtain the trajectories for all atoms, which are described by the successive positions, velocities, and momenta. These trajectories become the initial data for the prediction of the bulk properties of the systems, with further conations to the physical phenomena [10].

Insight the determined system, the pair of atoms are in interaction, irrespective of that they are bounded or separated by the distance. These interactions formed the basis of the simulation by the MD method. From this force, the equation for the potential energy can be obtained for the description of the stretching, vibrations, and rotation of the particles around the bounds as a result of the intermolecular force. This equation can be represented in the following way [10]:

$$U_{total} = U_{bond} + U_{angle} + U_{dihedral} + \\ + U_{VdW} + U_{Coulomb} + U_{external} \quad (93)$$

The values $U_{bond}$, $U_{angle}$ and $U_{dihedral}$ take into account the stretching, bending, and torsion, which take place in atoms, correspondingly. On the other hand, the values $U_{VdW}$ and $U_{Coulomb}$ describe the non-bounded interactions. Moreover, the van der Waal's forces have origin in the weak force existing for the non-bounded atoms, whereas the $U_{Coulomb}$ is used for the description of the electrostatic interactions, which are caused by electrostatic interactions. The last term of the total potential energy $U_{external}$ takes into account the external forces, applied to the system. In the investigation of the RO process, the main external force is the applied pressure [10].

The protentional energy of the intermolecular interaction also can be described by the following equation [171, 172]:

$$U_{(r_{ij})} = 4\varepsilon_{ij} \left[ \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{6} \right] + \frac{q_i \cdot q_j}{4\pi\varepsilon_0 r_{ij}} \quad (94)$$

where $r_{ij}$ is the distance between particles $i$ and $j$; $q_i$ and $q_j$ are the patiale charges of the $i$ and $j$; $\varepsilon_{ij}$ and $\sigma_{ij}$ are empirical Lennard-Jones parameters.

The MD method has an advantage, which consists of the ability to achieve the temporal and spatial solutions which are difficultly available by experimental investigations. For example, the solutions are possible for the dimension less than Angstrom and the periods of time less than a femtosecond [173].

Taking into account that the considered approach requires large volumes of calculations, the necessity of the

special software application is evident. For the MD methods realization of the many software packages, algorithms, and codes. In particular, for the RO simulations the following software was used: Visual Molecular Dynamics (VMD) [171-172, 174-178], Nanoscale Molecular Dynamics (NAMD) [171-172, 174-175, 179-180], Groningen Molecular Simulation package (GROMACS) [176-178, 181-183], Chemistry at Harvard Molecular Mechanics (CHARMM) [172, 175-176, 184], Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [185-187], Materials Studio [181, 188-191], especially Amorphous Cell package of Material Studio software [192-193], Assisted Model Building and Energy Refinement (AMBER) [180, 192-195], Optimized Potential for Liquid Simulations (OPLS-AA) [181-182], Condensed-phased-optimized Molecular Potential for Atomistic Simulation Studies (COMPASS) [188, 190], DL_POLY [194-195]. The data about such software developers and more detailed pieces of information about its application are represented in the review work [11].

During the simulation by MD methods, statistical techniques were used, including the Monte-Carlo method [174, 192-194] and surface response methodology [189]. A more detailed description of the MD methods is represented in the review work [10].

Taking into account the features of the method, during its application special attention was dedicated to the membrane material. In many works, conventional materials, such as polyamides [11, 175, 179, 184, 187, 190, 192-195], sulfonated diamine [189], and polyether sulphones [181], were considered. At the same time, a significant number of publications were dedicated to the use of membrane manufacturing such novel materials as graphene [9, 182, 187, 196] and graphene oxide [178, 197]. Also, the considered method was used for the analysis of the performance of the membrane made using the carbon [172, 176, 196, 198] and aluminosilicate [185], nanoporous carbon [199], boron nitride [171, 200], fullerite [188], and MoSe2 [177]. It should be noticed that in work [188] it was claimed that the fullerite membranes have the higher flux than the graphene and conventional membranes, and in work [177], it was declared that the flux of the productivity of the proposed membranes is who orders of magnitude higher than the commercially available membrane one.

In most cases, the investigation was dedicated the water treatment [175-176, 180, 190-191, 194-195, 199, 201], including desalination [11, 177, 182-185, 187-188, 192-193, 196-197, 200] and the heavy metal ions removal [171-172, 178].

### 3.7 Optimization and process control

In work [1], it was noticed that the determination of the optimal condition and processes control are the important practical application of the mathematical simulation of the pressure driven membrane processes, including RO. The large number of publications dedicated to these questions in the period from 2011 to 2020 completely confirm this statement.

Also, in work [1] it was noticed, that the optimization problem consists in finding the most beneficial values of the operation parameters. The important factor of the successive optimization procedure includes the choice of the target function and the optimization criterion. As in 20200-2010, in the considered period in most works authors used the economical optimization criteria, primarily the minimization of costs [49, 61, 74-75, 202-217], and also the minimization of the energy consumption, which is the significant part of the operational cost [27, 38, 49, 66, 73, 204-205, 219-228]. Also, the economic criteria included the profit maximization [229-230], and the minimization of the cost factor, which in work [231] was determined as the ratio of the water price and energy price in a hybrid system with reverse osmosis and pressure retarded osmosis. The main technological parameters for optimization included maximization of the removal of the component from solution 27, 36, 38, 63, 200, 221, 232-233], minimization of the concertation [234], maximizing of the permeate concertation [35], maximizing of the recovery ratio [235], maximizing of the productivity [236-237], and minimization of the applied pressure [36]. The other direction of the optimization involved the minimization of the fouling level [238] and the minimization of the environmental impact [230]. In the set of works, multiobjective optimization was applied [239-240].

In many works, the optimization was carried out based on the conventional mathematical models described in previous chapters. In particular, the solution-diffusion model was the most widely used [35-36, 38, 61, 66, 75, 204, 209, 214, 218, 221, 241]. Also, it was applied the Kedem-Katchalsky model [205, 219], Spiegler-Kedem model [27], Kimura-Surirajan [73], the surface force-pore flow model [78], and also the molecular dynamics models [200]. Moreover, the factor experiment method and other regression and statistical methods were applied [212, 228, 232, 235, 243-244]. The questions of the optimal control and processes control were considered in the works [220, 236-237, 245-248].

For the optimization problem solution, a large number of methods and algorithms can be used. In the considered researches the most widely used methods included the nonlinear programming [35, 49, 61, 74-75, 204-211, 218, 222, 229-230], the generic algorithm [28, 38, 237, 241-242], the particle swarm method [202, 247, 250], the sequential quadratic programming [219, 233], the surface response methodology [232], the pattern search algorithm [251], the harmony search algorithm [66], and the bees algorithm [213]. The work [216] should be pointed out, since in it the nineteen optimization algorithms were considered and compared.

For the solution of the considered optimization problems the wide range of the software and programing languages was applied including Matlab [73, 203, 250, 252], gPROMS [63, 218, 235], GAMS [49, 209, 211, 246, 254], Design–Expert [232], ASPEN [214], ChemCAD [215], ROSA [206-207], Modelica [219], C++ [36, 38], R [244].

As in the most considered RO simulation methods, the optimization was applied mainly for the analysis of the water treatment processes, primarily the seawater and brackish water desalination [49, 61, 66, 73-75, 200, 202-203, 206-210, 212-217, 219-227, 229, 234-235, 237-240, 242, 244-246, 248-250, 251-255], the potable water production from ground waters [256-257], the wastewater treatment [230-231, 238, 258], and recycling [211], and also the ultrapure water production [259]. In some works, it was considered the special pollutant removal including chlorophenol [36, 204, 233, 243], N-nitrosamine [27, 63], arsenic [205], boron [218] and bentazon [232]. Except for the water treatment, the concentration of the apple juice [35] and the maple syrup [228] was also considered.

The optimization methods appear to be effective for the analysis of the hybrid systems performance, in which the RO was combined with thermal distillation [214-215, 241], forward osmosis [238], pressure retarded osmosis [219, 229, 231], and forward osmosis and electrodialysis [224]. The other important direction was the optimization of the system with renewable energy sources [216, 260], including solar [202, 216, 253] and wind [216] energy.

It should be noticed, that the application of the optimization methods can give a significant economic effect, for example, it was noticed the achievement of decreasing of energy consumption by 16% in work [227], the decreasing operational cost by 26% in work [209], and the economy of the 27 % of the annual costs of the obtained product in work [211].

### 3.8 Energy analysis

It was mentioned above that energy consumption is a significant part of the operative costs during RO exploration. In the period from 2010 to 2020, a large number of articles dedicated to calculations and analysis of the energy parameters of RO were published, therefore, it is reasonable to consider this direction in general terms.

Mainly, the energy analysis of the RO systems was based on the mass and energy balances [43, 228, 261-266], theoretical calculations of the specific energy consumption [43, 228, 261-266], the exergy analysis [15, 43, 241-242, 263, 265, 275, 286-290], the pinch-analysis [292], and the calculations of the thermodynamical effectivity [293-294].

It should be noticed that, in many cases, the energy and exergy balances were written for the full set-up rather than RO apparatuses. At that the RO can be combined with energy recovery systems (including pressure exchangers and turbines) [276, 288-289, 290], Rankin cycle [296], internal combustion engine [269], electrodialysis [268, 270], distillation [262-263, 282, 290-291], humidification-dehumidification [43], membrane distillation [278, 298], forward and pressure retarded osmosis [278, 299], photovoltage system [264], and capacitive deionization [284].

As in the case of the optimization, the grate attention was dedicated to renewable energy sources including solar energy [262-264, 272-273, 275, 289, 293, 297, 301-303], wind energy [267, 287, 262-263, 301-302, 304], and also tidal energy [266, 272].

As in all previous cases, the high effectiveness of the analysis was achieved via the application of specialized software and programming languages. For the energy analysis such means as ROSA [150, 265, 272, 295, 300], Matlab [261, 295], ASPEN [205], Visual Design Software [288], WAWE [305], EES [289], and R [244] were widely used.

In most cases, the desalination processes were considered. Also, in many works, the significant economical effect was pointed out. For example, in work [287] on the results of the exergo-economic analysis, the authors managed to find a way of decreasing energy consumption by 24%. In works [288] a similar analysis shows that the application of the turbine allows to decrease the energy consumption by 49%, and the application of the pressure exchanger – by 77. In work [289]. It was shown that application of the Pelton turbine allows for recovery of 24% of the consumed energy.

### 3.9 Economic analysis

During consideration of most approaches, especially optimization and energy analysis, it was pointed out that the economic parameters have significant importance for the performance analysis of the RO systems. Therefore, it is reasonable also to consider in general terms the economic methods of the analysis of the considered process.

Except optimization [202, 204-205, 231, 241] and energoeconomic analysis [205, 230, 241, 266-270, 280, 285-287, 306], the economical models included the determination of the relationship for calculation of the capital cost (CAPEX) [55, 204-205, 234, 230, 266, 268, 270, 280, 286, 297, 300, 307-317], operating costs (OPEX) [55, 204-205, 209, 230, 234, 266, 280, 300, 307, 309-318], and product price [268, 274, 280, 297-298, 302, 306-307, 314, 319-324]. Among other parameters the profit [325], cost saving coefficient [319], cost factor [231], and water price index [326] can be pointed out. In certain works, the simplified economic balance [327] and socio-economic evaluation [328] were considered.

The other joint characteristic of the investigation based on economic analysis with ones based on optimization and energy analysis is the significant number of works dedicated to hybrid systems and systems with renewable energy sources. In particular, the hybrid systems RO-distillation were considered in works [55, 230, 241, 307-308, 314-315], and the works [321, 241, 300, 309, 311] were dedicated to the systems RO-FO and RO-PRO. The other hybrid systems including electrodialysis [270, 310], crystallization [268, 270], and power plant [55, 274]. Among the alternative energy sources, the most attention was dedicated to solar energy [202, 297, 302, 308, 320-321, 328]. Wind energy [302, 306], tidal energy [266], and geothermal energy [314] were also considered.

Among the software, in works dedicated to the economic analysis of the RO systems, it was mentioned ROSA [231, 266, 324], COMFAR III [323], and ESS [324].

As in all other approaches to the RO simulation, in most cases, the application of RO for the seawater and brackish

waters desalination [55, 202, 209, 234, 241, 266-270, 274, 280, 287, 297, 302, 306-309, 311-312, 314, 317-324] was considered. In addition, the questions about wastewater treatment [298, 300, 310, 315-316, 327], including the removal of certain pollutants such as chlorophenol [204] and arsenic [205], were regarded.

## 4 Discussion

In certain works, the other approaches, which cannot be related to the described above groups, were applied. Those models can be based on the empirical data (both with and without combination with conventional models) [329-336], the mass and energy balances [337-341], the Monte-Carlo method [342-344], the concentration polarization models [345-347] and osmotic pressure [348], the boundary layer theory [349], the dynamic modeling [350], the materials flow analysis [351], the risk and fault analysis [352-353], the numerical methods [354], the chemical kinetics and equilibrium [355], or its combinations [356]. However, such investigations were low numerous and it does not seem possible to define some of them as perspectives.

It should also be noted, that in some works for the RO description the extended Nernst-Plank equation [358-359] and the Donnan equilibrium [359] were used, however, such approaches are conventionally used for the NF, as was shown in work [1], Moreover, the special software (such as ROSA or Q+), which was widely used for the analysis of the economical and energy paraments of RO systems, can be also used for the simulation of the technological parameters and system design, as it was realized, for example, in works [340, 360-365].

## 5 Conclusions

The technological, energetic, and economic advantages of the reverse osmosis process in conjunction with the benefits of the mathematical simulation determined the considerable spread of this type of investigation. Therefore, it is natural, that the stable trend increasing in the number of publications dedicated to the reverse osmosis simulation is observed. The analysis of publications about this question in the period from 2011 to 2020 allows also traces the main direction and the advantages and disadvantages of the main approaches to the RO simulations.

Thus, the number of publications dedicated to the application of the CFD methods for the mathematical simulation of RO appears to be the largest. The increasing computational power of computer equipment and the development of specialized software, including the appearance the open-source programs, are the evident reasons for this. It is logical to assume, that the same factors promoted the significant increase in the application of the molecular dynamics methods in comparison with the previous decade. Such a trend allows us to conclude, that these two approaches will remain the most widely used in the near future. The main advantages of this method consist in the possibility to obtain a deep understanding of the processes which take place in the membrane apparatus (CFD) or in the membrane itself and on its surface (MD). They also allow us to determine the influence of the large number of factors affecting the process and, therefore, the productivity and effectiveness of separation. However, the simulation using these methods requires the application of more complex and cumbersome calculation methods, and also a high level of skills of operation in corresponding software.

On the other hand, the published results indicate that the interest of the researchers and engineers in hybrid systems, in which the RO is combined with other processes, mostly with distillation, forward osmosis, and renewable energy sources increased significantly. For the simulations of such systems, it is reasonable to apply the simulation techniques which require simpler calculation methods. It was quite unexpected that, for these purposes, the solution-diffusion model was used in the widest range. In the considered period it was used both for the direct RO simulation and for the optimization, energy and economic analysis, and even as boundary conditions in CFD investigation.

At the same time, the application of the irreversible thermodynamics methods, pore flow based models, and other diffusion based models was limited and, likely, further these models will be used mainly for the specific processes analysis. Also, the application of artificial neuron networks was limited, which probably is due to the fact that the possibility of the mechanistic model construction is relatively high for the case of RO. In these conditions, the ANN will be the most effective in systems that are significantly complicated by concentration polarization and fouling.

Therefore, based on the current review it is possible to choose the strategy for the simulation of the RO system. It also should be noticed that represented in the current review the analysis of the software applied for the reverse osmosis simulation, would be useful for educational purposes.

## 6 Acknowledgments

Authors would like to thank the Armed Forces of Ukraine, National Guard, Territorial Defense, Volunteers for providing security to perform this work. This work has become possible only because resilience and courage of the Ukrainian Army.

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