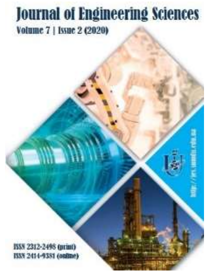
JOURNAL OF ENGINEERING SCIENCES

Volume 7, Issue 2 (2020)

Huliienko S. V., Korniienko Y. M., Metlina M. S., Tereshenko I. Y., Kaminskyi V. S. (2020). The correction of the dimensionless equation for the mass transfer coefficient estimation during the membrane modules regeneration. Journal of Engineering Sciences, Vol. 7(2), pp. F24–F29, doi: 10.21272/jes.2020.7(2).f4



The Correction of the Dimensionless Equation for the Mass Transfer Coefficient Estimation during the Membrane Modules Regeneration

Huliienko S. V.[1*][0000-0002-9042-870X], Korniienko Y. M.[1][0000-0002-3031-6212],
Metlina M. S.[1], Tereshenko I. Y.[1], Kaminskyi V. S.[2]

[1] National Technical University of Ukraine "Igor Sikorsky Kyiv Polytechnic Institute", 37, Peremohy Ave., 03056 Kyiv, Ukraine;
[2] Technical University of Kosice, 9, Letna St., 042 00 Kosice, Slovak Republic

Article info:
Paper received: September 27, 2020
The final version of the paper received: December 12, 2020
Paper accepted online: December 21, 2020

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

Abstract. The cleaning or regeneration of fouled membrane modules is an essential procedure in the membrane equipment operation. Despite the development of some successful cleaning techniques, the predictions of the membrane separation process operation parameters after regeneration is still an unsolved problem. In our previous works, the attempt to develop the methodology of estimating the membrane productivity after the regeneration of the fouled spiral wound membrane modules by cleaning the subatmospheric pressure has been made. However, this methodology requires some improvement, including the correction of the dimensionless equation to calculate the mass transfer coefficient. In this work, a set of additional experiments was carried out, and the corrections of the mass transfer correlation were done using both new and previously obtained experimental data. As a result, the improved dimensionless equation was contained as $Sh = 0.00045Re^{0.8}Sc^{0.33}(d_e/l)$. This equation is valid in the range of Reynolds number variation of 0.4–60.0 for the case of the regeneration of spiral wound modules and can be used for the prediction of the permeate flux after the regeneration procedure.

Keywords: reverse osmosis, fouling, cleaning, mass transfer correlation, diffusion coefficient, Reynolds number, Schmidt number, Sherwood number

1 Introduction

The fouling of membrane surface is one of the main problems during exploitation of the membrane equipment, including reverse osmosis [1]. Despite the development of membranes with antifouling properties [2], methods of pretreatment [3], and optimization of the operating parameters in membrane modules [4], the necessity for periodic cleaning and regeneration is still significant [5].

During the regeneration process realization, the membrane separation properties' prediction after the regeneration procedure is still important and not well understood. In our previous work [6], an attempt to develop such a prediction technique has been made. However, in that work , the dimensionless equation was used, which was obtained with some assumptions, which in many cases do not correspond to the real conditions.

Therefore, the current article aims to improve the dimensionless relationship for calculating the mass transfer coefficient during the regeneration of membrane modules. In this research, both new and previously obtained experimental data were used.

2 Literature Review

The fouling layer's composition on the reverse osmosis membranes' surface could significantly depend on operation conditions and the chemical composition of feed solution [7–10]. The most typical kinds of fouling include the precipitations of the mineral salts, primarily sodium, magnesium and calcium [7, 8], and the organic compounds [7, 9]. Considering the negative economic impact of all kinds of fouling [10] each of them's specific character, it is necessary to choose the specific strategy of the regeneration for each particular case. In the current research, the primary attention was considered on mineral scaling with is common in many practical cases.

The primary method of the regeneration of fouled membrane modules is cleaning, mostly with using of chemical reagents [11]. The use of cleaning without reagent is also can be applied [12]. The cleaning effect can be enhanced by using some physical processes, namely ultrasound impact [13], injection into the flow of the cleaning agent, air bubbles [14], and local boil of the cleaning agent under subatmospheric pressure [6]. The regeneration methods application can allow removing up to 91 % of the fouling layer material [13]. The presence of a large volume of experimental data and techniques can permit the optimization of the membrane separation system [13, 15]. However, in most cases, the evaluation of cleaning effectivity and the prediction of the membrane conditions after cleaning is carried out based on the empirical data, which are not systematized at the appropriate level. The physical phenomena, which take place during regeneration, including the mass transfer process between the cleaning solution and the substance in the fouling layer, are insufficiently studied.

The theory of similarity and dimension analysis methods is an effective tool for analyzing transfer phenomena under complicated conditions such as turbulent flow. These methods allow approximating empirical data in the relationships among the dimensionless parameters [16]. For the analysis of the membrane processes, that methods showed their effectiveness for the description of the mass transfer in plate-and-frame modules [16], hollow fiber modules [16, 18], and channels with spacers, which can be considered as models of plate-and-frame or spiral wound modules [19]. Dimension more negligible mass transfer correlations are also used to analyze concentration polarization intensity [17].

In previous work [6], the attempt of obtaining the mass transfer correlation for the case of spiral wound membrane module regeneration. This correlation was used for the evaluation of membrane separation process properties after regeneration. However, during the data analysis for obtaining the dimensionless equation, the diffusivities' values in solutions were calculated using the Wilke–Chang equation. The mentioned equation has overestimated the values of solute diffusivities. In work [20], it was shown that using the experimental data represented in reference literature is a more reliable way. Therefore, in the current work, the dimensionless equation's improvement for calculation of the mass transfer coefficients during membrane modules regeneration is carried out. As mentioned above, the latest data were uses as well as the previously obtained results.

3 Research Methodology

3.1 Materials

The experiments were carried out using commercially available membrane modules TFC-75 manufactured by DOW Filmtec (USA). The membrane surface area of these membranes is F = 0.46 m^2, the cross-section of the membrane channel is $S = 3.675 \cdot 10^{-4}$ m^{-2}, the spacer thickness is $\delta = 0{,}35 \cdot 10^{-3}$ m, the module length is $l = 0{,}26$ m [21].

The artificial fouling layer was deposited on the membrane surface. The sodium chloride (NaCl) was used as a fouling material. The deionized water was used as a cleaning solution.

3.2 Description of the experimental set-up

In the current research, the existing experimental set-up (Figure 1) was used. It includes the electronic scales 1, the tank with cleaning solution 2, the control valves 3 and 5, the receiver 4, the cleaning chamber 6, the membrane module 7, the vacuum pump 8, the atmospheric valve 9, the intermediate tank 10, the system for sampling 11, the personal computer 12, the electric heater 13.

In the experimental set-up, the regeneration is realized in the cleaning under subatmospheric pressure produced by vacuum pump 9. The subatmospheric pressure also provides the pressure difference required for the cleaning solution flow.

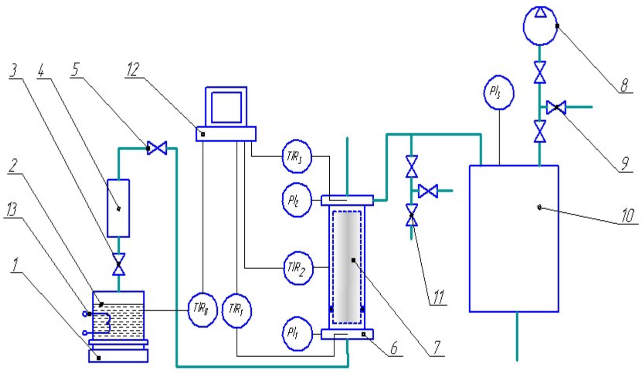


Figure 1 – The scheme of the experimental set-up

The block of chromel-copel thermocouples fits the experimental set-up. This allows to measure the temperature in the cleaning chamber 6 on the input and output of the membrane module 7 and the tank with cleaning solution 2 and in the environment. The vacuum gauges are also installed to measure the subatmospheric pressure on the input and output of membrane module 7 and in the intermediate tank 10.

For measuring the cleaning solution's concertation after passing through the membrane module 7, the system for sampling 11 is included. The concertation of NaCl in the cleaning solution is measured using the portative TDS-meter.

3.3 Experimental procedure and main measurements

Since this research is dedicated to defining the dimensionless equation (mass transfer correlation), as it was done in works [16–19], it is necessary to provide the measurements the parameters with are involved in the dimensionless numbers. It should be noticed that the correlation is searched in the following form:

$$\mathrm{Sh} = A\,\mathrm{Re}^m\,\mathrm{Sc}^n\left(\frac{d_e}{l}\right), \quad (1)$$

where $\mathrm{Sh} = k \cdot d_e / D$ is the Sherwood number; $\mathrm{Re} = w \cdot d_e \cdot \rho / \mu$ is the Reynolds number; $\mathrm{Sc} = \mu \cdot / (D\,\rho)$ is the

Schmidt number; $d_e = 2\delta$ is the equivalent diameter, m; k is mass transfer coefficient, m/s; D is the solute diffusivity, m^2/s; w is the cleaning solution velocity, m/s; ρ is the cleaning solution density, kg/m^3; μ is the cleaning solution dynamic viscosity, Pa·s.

In this case, it is necessary to measure the mass transfer coefficient and cleaning solution velocity. For the determination of the physical parameters, namely diffusivity, density, and dynamic viscosity, the measurements of the temperature, pressure, and solute concentration in the cleaning solution are required.

During the investigation, such parameters as pressure, temperature, and concentration NaCl in the cleaning solution were measured directly. The cleaning solution's velocity was measured by the weight method, namely the change in mass of the tank with cleaning solution 2 during the predetermined period of time was recorded. For the determination of the mass transfer coefficient, the main equation of mass transfer was used [6]:

$$M = k\left(C^* - C_1\right)F\tau\,, \tag{2}$$

M is the mass of removed fouling, kg; C* is the equilibrium concentration of solvent, kg/m3; C1 is the solvent concentration in cleaning solution, kg/m^3; τ is the process duration, s.

From equation (2), it can be obtained:

$$k = \frac{M}{\left(C^* - C_1\right)F\tau}\,, \tag{3}$$

The mass of removed fouling was defined from material balance [6]:

$$M = G\left(x_o - x_i\right)\tau\,, \tag{4}$$

G is the mass flow rate of cleaning solution, kg/s; xi and xo are the mass fractions of NaCl on the input and output of the membrane module 7.

The density and dynamic viscosity and also the equilibrium concentration of NaCl were determined by the reference data, represented in [22, 23], as it was done in work [20]. For the determination of diffusivities, the experimental data represented in [24]. The corrections of the tabulated data were carried out in the same way as in work [20].

The experimental procedure was following. The cleaning solution (the deionized water with predetermined concentration at the level of 5-15 mg/dm3) was loaded in tank 2, and using the electric heater 13, the temperature of the cleaning solution was brought to the predetermined value. Simultaneously, using the vacuum pump 8 the subatmospheric pressure was created in the intermediate tank 10. Using the control valves 3 and 5 the predetermined values of the pressure in the cleaning chamber 6 on the input and output of the membrane module 7 were set. Thereat the mass flow rate and velocity of cleaning solution and the temperature values on the input and output of the membrane module were measured, and also the sampling of cleaning solution on the output of membrane module was carried out using the system 11. The values of temperature were continuously recorded on personal computer 12 using the program unit IndexTem.

4 Results

The set of improving experiments were carried out according to the described above technique. The results obtained in previous work [6] were also used during data processing. Since in the experiments only one material, namely NaCl, used as fouling model, for providing the variation in Schmidt number, the investigation was carried out under different temperatures. In this case, the lower level corresponded to the minimum ambient temperature, which was 14°C. The upper level was chosen considering the thermal stability of the membrane. Therefore, the maximum temperature was 40°C. The cleaning chamber's subatmospheric pressure was varied in a range of 0.06–0.095 MPa, which corresponds to the absolute pressure of 0.005–0.04 MPa. The cleaning solution velocity was varied in a range of 0.0006–0.0827 m/s. These operation conditions correspond to the variation of Reynolds number in a range of 0.4-60 and the Schmidt number change in a range 568–630.

Since in the works [16, 18, 19], the exponent for Schmidt number in a wide range of operation conditions remains constant and equal n = 0.33. Current research assumed that this value of the mentioned exponent is still reasonable. In this case, obtaining the dimensionless equation is confined to the determination of exponent m for Reynolds number and factor A. This assumption's verification should be done by a similar experimental investigation in the wider range of Schmidt number variation.

During the experimental data processing, all the data set were divided on the ranges, in which the values of the Schmidt number varies down to 2 %. For each range, the determination of the exponent in relationship $Sh = A Re^m$ was carried out. That relationship is linearized in logarithm coordinates; therefore, this coordinate system was used. The graphical relationships $Sh = f(Re)$ for some rangers of Schmidt number values are represented in Figures 2–5.

In all considered ranges, the graphical relationships in logarithm coordinate systems can be approximated by linear dependence, allowing generalizing the experimental results.

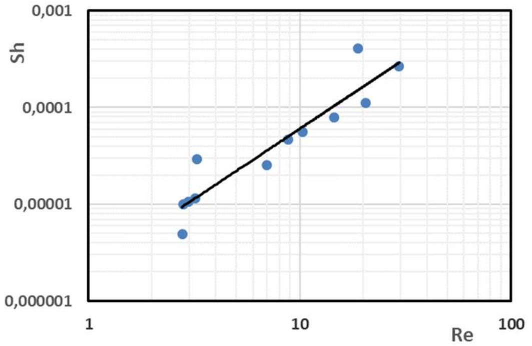


Figure 2 – The dependence of the Sherwood number from Reynolds number (Sc = 610–613)

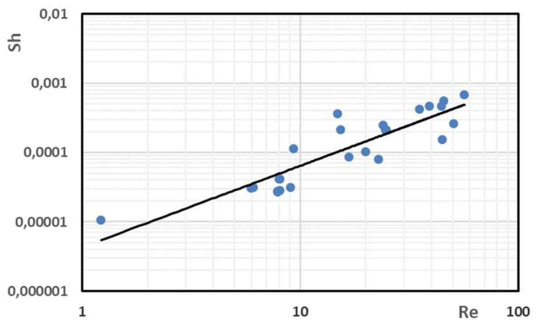


Figure 3 – The dependence of the Sherwood number from Reynolds number (Sc = 620–621)

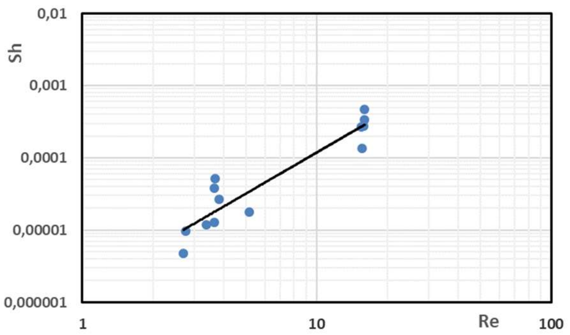


Figure 4 – The dependence of the Sherwood number from Reynolds number (Sc = 626-628)

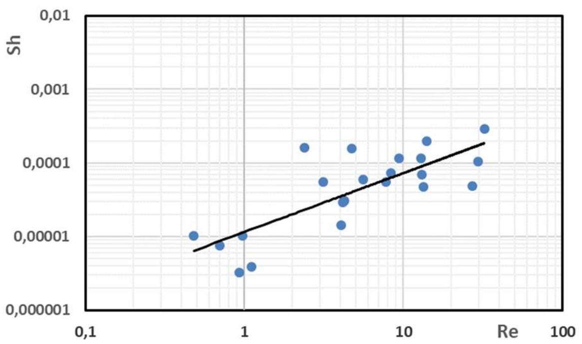


Figure 5 – The dependence of the Sherwood number from Reynolds number (Sc = 633–634)

5 Discussion

Since the graphical relationships, represented in Figures 2–5, are linear, the dependence in a form lg(Sh) = lg(Re) were linearly approximated by the mean square method, according to the technique represented in [25]. The slops of this dependences are equal to the exponents for the Reynolds number *m*. The calculated values of these parameters are shown in table 1. The mean value of the slope is equal to $m' = 1.155$, which was used for the generalization of experimental results. For this purpose, the relationship $\mathrm{Sh} = f(\mathrm{Re}^{1.155}\mathrm{Sc}^{0.33})$ was plotted in the logarithm coordinate system, as is shown in Figure 6.

Table 1 –The values of the exponent for Reynolds number

The value of Schmidt number (Sc)	The value of exponent *m*
580-584	2.059
596-599	1.742
600-603	1.016
604-606	1.021
610-613	1.450
614-617	1.088
620-621	1.173
622-623	1.168
624-625	1.001
626-628	1.885
629-630	0.401
631-632	0.757
633	0.795
634	0.099
The mean value	1,155

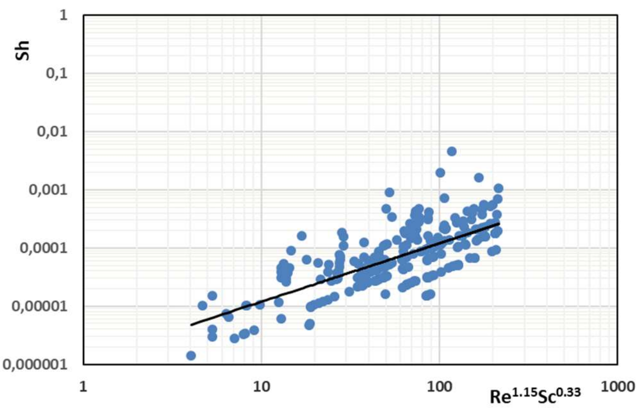


Figure 6 – The generalized dependence $\mathrm{Sh} = f(\mathrm{Re}^{1.155}\mathrm{Sc}^{0.33})$

It can be seen from Figure 6, the generalized dependence $\mathrm{Sh} = f(\mathrm{Re}^{1.155}\mathrm{Sc}^{0.33})$ can be approximated linearly in the logarithm scale. The mean squares method allows to determinate the slope, which is equal to $k = 0.69$. From this the correction of the exponent for Reynolds number can be done, namely $m = m' \cdot k = 0.8$. Keeping the value of the exponent for Schmidt number as $n = 0.33$, the final generalization was realized as the relationship in the form $\mathrm{Sh} = f(\mathrm{Re}^{0.8}\mathrm{Sc}^{0.33}(d_e/l))$, as it is shown in Figure 7.

The generalized dimensionless dependence can be approximated with the exponent (slope of the line in logarithm coordinate system) equal to 1,0023, which can be accepted as 1 with satisfying accuracy for engineering calculations. In this case, the final dimensionless relationship would be written in the following form:

$$\mathrm{Sh} = 0.00045\,\mathrm{Re}^{0.8}\,\mathrm{Sc}^{0.33}\left(\frac{d_e}{l}\right). \qquad (5)$$

The obtained dimensionless equation is valid in the range of Reynolds number variation of 0.4-60 for the regeneration of spiral wound modules.

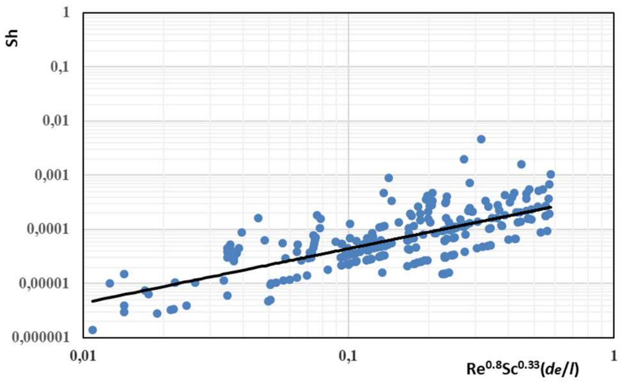


Figure 7 – The generalized dimensionless dependence

The sampling correlation coefficient of dimensionless equation (5) is equal to $r^* = 0.712$. The statistical analysis, which was carried out according to the technique represented in [25], proves the relationship's existence.

Considering that Schmidt's exponent was asspumed according to the data from other researches, namely [16, 18, 19], the dimensionless equation needs further corrections, namely the verification of the value of the exponent n. This means that further investigation in this area should be dedicated to clarifying the impact of the fouling layer's chemical composition on the mass transfer intensity.

6 Conclusions

As a result of the corrective experiments, the improved dimensionless equation for calculating the mass transfer coefficient during membrane modules regeneration was obtained. The proposed equation is valid in the range of Reynolds number variation of 0.4-60 for the regeneration of spiral wound modules. The obtained equation would define the required time of regeneration according to the technique developed in work [6] with higher accuracy than the previously defined similar equation. The dependence (5) should be father improved, namely the value of exponent for Schmidt number n. This means that the impact of the fouling layer's chemical composition on the mass transfer intensity, which is the aim of further research in this area.

References

1. Tow E. W., Warsinger D. M., Trueworthy A. M., Swaminathan J., Thiel G. P., Zubair S. M., Myerson A. S., Lienhard V J. H. (2018). Comparison of fouling propensity between reverse osmosis, forward osmosis, and membrane distillation. *Journal of Membrane Science*. Vol. 556, pp. 352-364, doi: 10.1016/j.memsci.2018.03.065
2. Qi Y., Tong T., Zhao S., Zhang W., Wang Zh., Wang J. (2020). Reverse osmosis membrane with simultaneous fouling- and scaling-resistance based on multilayered metal-phytic acid assembly. *Journal of Membrane Science*. Vol. 601. 117888. doi: 10.1016/j.memsci.2020.117888
3. Shon H.K., Vigneswaran S., Cho J. (2008). Comparison of physico-chemical pretreatment methods to seawater reverse osmosis: Detailed analyses of molecular weight distribution of organic matter in initial stage. *Journal of Membrane Science*. Vol. 320, Issues 1–2, pp. 151-158, doi: 10.1016/j.memsci.2008.03.063
4. Nguyen Th.-T., Kook S., Lee Ch., Field R. W., Kim I. S. (2019). Critical flux-based membrane fouling control of forward osmosis: Behavior, sustainability, and reversibility. *Journal of Membrane Science*. Vol. 570–571, pp. 380-393, doi: 10.1016/j.memsci.2018.10.062
5. Jafari M., D'haese A., Zlopasa J., Cornelissen E.R., Vrouwenvelder J.S., Verbeken K., Verliefde A., van Loosdrecht M.C.M., Picioreanu C. (2020). A comparison between chemical cleaning efficiency in lab-scale and full-scale reverse osmosis membranes: Role of extracellular polymeric substances (EPS). *Journal of Membrane Science*. Vol. 609, 118189, doi: 10.1016/j.memsci.2020.118189
6. Korniyenko Y., Guliienko S., Lialka M. (2015). Mathematical simulation of fouled membrane modules regeneration. *Ukrainian Food Journal*. 2015. Vol. 4. Is. 3, pp. 481-493.
7. Fortunato, L., Alshahri, A. H., Farinha, A. S. F., Zakzouk, I., Jeong, S., Leiknes, T. (2020). Fouling investigation of a full-scale seawater reverse osmosis desalination (SWRO) plant on the Red Sea: Membrane autopsy and pretreatment efficiency. *Desalination*. Vol. 496, 114536, doi: 10.1016/j.desal.2020.114536.
8. Song W., Lee, L. Y., Liu E., Shi X., Ong S. L., Ng H. Y. Spatial variation of fouling behavior in high recovery nanofiltration for industrial reverse osmosis brine treatment towards zero liquid discharge. *Journal of Membrane Science*, 609, 118185, doi: 10.1016/j.memsci.2020.118185
9. Wu Y.-H., Tong X., Zhao X.-H., Bai Y., Ikuno N., Ishii K., Hu H.-Y. (2021). The molecular structures of polysaccharides affect their reverse osmosis membrane fouling behaviors. *Journal of Membrane Science*. In Press, Journal Pre-proof, 118984, doi: 10.1016/j.memsci.2020.118984
10. Jafari M., Vanoppen M., van Agtmaal M.C., Cornelissen E.R., Vrouwenvelder J. S., Verliefde A., van Loosdrecht M.C.M., Picioreanu C. (2021). Cost of fouling in full-scale reverse osmosis and nanofiltration installations in the Netherlands, *Desalination*. Vol. 500, 114865, doi: 10.1016/j.desal.2020.114865

11. Madaeni S.S., Samieirad S. (2010). Chemical cleaning of reverse osmosis membrane fouled by wastewater, *Desalination*. Vol. 257, pp. 80-86, doi: 10.1016/j.desal.2010.03.002
12. Ibrahim S., Nagasawa H., Kanezashi M., Tsuru T. (2020). Chemical-free cleaning of fouled reverse osmosis (RO) membranes derived from bis(triethoxysilyl)ethane (BTESE), *Journal of Membrane Science*, 601, 117919, doi: 10.1016/j.memsci.2020.117919
13. Li Y.-Sh., Shi L.-C., Gao X.-F., Huang J.-G. (2016). Cleaning effects of oxalic acid under ultrasound to the used reverse osmosis membranes with an online cleaning and monitoring system, *Desalination*. Vol. 390, pp. 62-71, doi: 10.1016/j.desal.2016.04.008
14. Dayarathne H.N.P., Choi J., Jang A. (2017). Enhancement of cleaning-in-place (CIP) of a reverse osmosis desalination process with air micro-nano bubbles, Desalination. Vol. 422, pp. 1-4, doi: 10.1016/j.desal.2017.08.002
15. Lu Y., Hu Y., Xu D., Wu L. (2006). Optimum design of reverse osmosis seawater desalination system considering membrane cleaning and replacing, *Journal of Membrane Science*, Vol. 282, pp. 7–13, doi: 10.1016/j.memsci.2006.04.019
16. Zhang L.-Zh., Li Zh.-X. (2013). Convective mass transfer and pressure drop correlations for cross-flow structured hollow fiber membrane bundles under low Reynolds numbers but with turbulent flow behaviors, *Journal of Membrane Science*, Vol. 434, pp. 65–73, doi: 10.1016/j.memsci.2013.01.058
17. Cavaco Morão A. I., Brites Alves A., Geraldes V. (2008). Concentration polarization in a reverse osmosis/nanofiltration plate-and-frame membrane module, *Journal of Membrane Science*, Vol. 325, pp. 580–591, doi: 10.1016/j.memsci.2008.08.030
18. Ren Zh., Yang Y., Zhang W., Liu J., Wang H. (2013). Modeling study on the mass transfer of hollow fiber renewal liquid membrane: Effect of the hollow fiber module scale, *Journal of Membrane Science*, Vol. 434, pp. 65–73, doi: 10.1016/j.memsci.2013.03.030
19. Koutsoua C.P., Yiantsios S.G., Karabelas A.J. (2009). A numerical and experimental study of mass transfer in spacer-filled channels: Effects of spacer geometrical characteristics and Schmidt number, *Journal of Membrane Science*, Vol. 326, pp. 234–251, doi: 10.1016/j.memsci.2008.10.007
20. Huliienko S. V., Protsiuk O. O., Gatilov K. O., Kaminskyi V. S. (2019). The estimation of feed solution composition influence on concentration polarization layer resistance during reverse osmosis. *Journal of Engineering Sciences*, Vol. 6(2), pp. F24-F29, doi: 10.21272/jes.2019.6(2).f4.
21. Huliienko S., Leshchenko O. (2020). Influence of operating pressure on concentration polarization layer resistance in revers osmosis, *Ukrainian Food Journal*, Vol. 8 (1), pp. 119-132, doi: 10.24263/2304- 974X-2019-8-1-13
22. Nikolskij, B. (2013). *Chemist's Handbook, 3 – Chemical Equilibrium and Kinetics. Properties*. Demand Ltd.
23. Yaws, C. (1999), *Chemical Properties Handbook: Physical, Thermodynamic, Environmental, Transport, Safety and Heals Related Properties for Organic and Inorganic Chemicals*. New York, McGraw-Hill.
24. Dytnerskii, Iu. I. (1986). *Baromembrane Processes. Theory and Calculation*. Moscow, Chemistry.
25. Perry R. (1997). *Perry's chemical engineering handbook* (7th ed). New York, McGraw-Hill.