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*Original Research*

# Modeling of the Membrane Filtration Process of Natural Waters

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## Abstract

This work presents the verification of the model of the membrane filtration process of natural waters for non-stationary filtration flux J=f(t), as well as the results of tests involving the filtration process of the said model in constant pressure conditions. Also, the characteristic time of permeate flux decay was determined, and the mechanisms reducing the efficiency of filtration process were analyzed.

**Keywords:** natural waters; membrane water treatment; ultrafiltration; microfiltration; filtration model

## Introduction

Due to the increasing pollution of natural waters the supply of water to the public is becoming a serious social problem. As specified by the regulations, potable water should comply with specific bacteriological standards and should have appropriate physicochemical properties [1]. In order to meet these requirements, it is necessary to introduce the treatment of water with disinfection being a part of it. Treatment conditions must therefore be corrected on a regular basis, depending on the quality of raw water. It must be principally ensured that the coagulants are batched in the right way and the pH of flocculation is determined. The automation of the technological process should solve that problem **[2];** however, we cannot totally eliminate the possibility that the reagents are overdosed, and hence to ensure that quality of produced water is always of the invariable high standard as desired [3]. Before the introduction to the distribution network the water is subject to disinfection to obtain the desired microbiological quality. Chlorination is the most commonly applied method of water disinfection.

Membrane techniques have not been so far applied for the treatment of drinking water, except for reverse osmosis, which was widely used for desalination of waters [4, 5]. Due to a new approach to the problem of water treatment for consumption, the said technologies are currently being taken into consideration as alternative processes for water treatment [2-3, 6-10].

The main obstacle involving the application of membranes for the treatment of potable water is the reversed and non-reversed fouling. In order to reduce it, in the system of cross-flow filtration used with ultrafiltration or microfiltration, different pretreatment techniques of water have been suggested as well as frequent back-flushing of membranes. Although the said techniques are effective in overall reduction of fouling in membranes, due to the high complexity of fouling mechanisms the comprehensive modeling of the work of membrane system with respect to water treatment is difficult to carry out. The designing and functioning of large treatment plants necessitates that the relations describing the efficiency and selectivity of membranes in such conditions were not only available but that they were relatively simple to apply and did not contain constants or empirical coefficients which are difficult to determine.

The basic problems involving the utilization of pressure membrane processes are as follows [11]:

- reduction of efficiency (flux decline) during the operation of systems, even in stationary conditions,
- efficiency of the process is becoming independent of transmembrane pressure (limiting flux), the main driving force in pressure membrane techniques,
- fouling: physical, chemical and biological events leading to irreversible changes of membrane properties.

Fouling and concentration polarization occur at the same time, leading to the reduction of efficiency of membrane processes.

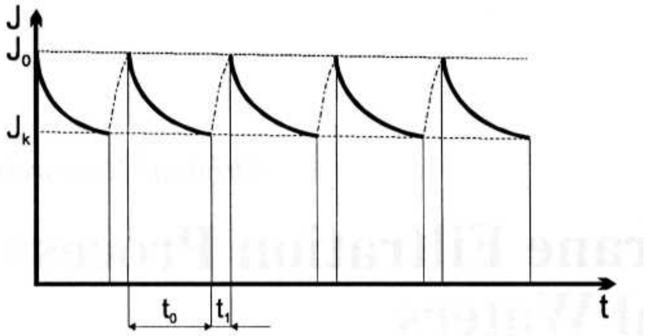

Fig. 1. Cyclic process of the filtration and regeneration of membranes; where: $J_o$ - value of opening flux, $J_k$ - value of saturation flux (closing one), $t_0$ - time of the process, $t_1$ - regeneration time.

The membranes and membrane modules used in the process of water treatment should be easy to clean with backflushing method, should be characterized by low consumption of energy and high compactness (packing density) [12].

In the tests carried out, the membranes from non-cellulose polymers were applied, that is from polyacrylonitrile, polysulphone and polyethylene. Also, non-organic, ceramic membranes having many advantages were applied[10].

With respect to the modeling process of membrane filtration of natural waters the term fouling is usually understood as the condition where the permeate flux has been lowered, which was effected by processes taking place on the surface of the membrane and in its pores.

Irreversible reduction of permeate flux, which is a form of fouling, is caused principally by the adsorption of natural organic substance on the surface or inside the pores of the membrane. Such fouling may be partially removed using chemical methods, and the standard physical methods are not able to remove it. That type of fouling is very unfavorable for water treatment with the use of membranes. There are many models which describe the reasons for the reduction of permeate flux in the ultrafiltration process. However, the great majority of them had been elaborated for steady working conditions, as for example traditional model of gel polarization, or the model based on the diffusion induced by shearing force and elaborated for the ultrafiltration of colloids. Literature provides data involving fouling and the reduction of permeate flux (also for unsteady conditions). Yet the described models have been elaborated for the filtration of high-molecular, organic, solved substances of the concentration higher by several orders than the concentration of natural organic substance occurring in natural waters. Furthermore, there were investigation studies carried out in static chambers with stirring and without stirring under constant pressure. When applying the ultrafiltration process for water treatment on a big scale, it is the cross-flow ultrafiltration which is most commonly applied [4, 11].

The utilization of filtering flowing equipment in the cross-flow system can be carried out under constant pressure or with constant permeate flux.

In the tests carried out, the effectiveness of UF and MF of ground waters and surface waters was compared, with the application of membranes of various configuration (flat, tubular and capillary) and made from different materials: polymeric membranes (polysulphone, polyacrylonicrile and polyprophylene) as well as ceramic membranes.

Based on investigation studies of micro and ultrafiltration in view of the treatment and disinfection of natural waters, the following attempts have been made:

- verification of the filtration model based on the change of volumetric flux in time, with the determination of time constant characterizing the drop in efficiency of filtration process,
- verification of membrane filtration model in the condi tions of constant pressure.

## Results

Cyclic process of the filtration and regeneration of membranes can be presented graphically as in Fig. 1.

Volumetric flux J is to be defined as presented by the equation below:

$$J = \frac{1}{F} \cdot \frac{dV}{dt} \tag{1}$$

where: F - membrane surface [$m^2$],
V - filtration volume [$m^3$].

### Determination of Permeate Flux in Non-Stationary Process

Based on the equation of mass transport balance during the filtration process, the following equation can be obtained:

$$\frac{d}{dt}(J - J_\infty) + \frac{1}{t_0}(J - J_\infty) = 0 \tag{2}$$

The solution of the above equation has the following form:

$$\frac{J - J_\infty}{J_0 - J_\infty} = e^{-\frac{t}{t_0}} \quad \text{or} \quad \ln\left(\frac{J - J_\infty}{J_0 - J_\infty}\right) = -\frac{t}{t_0} \tag{3}$$

where: $J_0$ - opening flux $J|_{t=0} = J_0$
$J_\infty$ - saturation flux $J_{t\to\infty} = J_\infty$
$t_0$ - characteristic time of decay for permeate flux

Diagrams have been made for the above equation, which is presented below (for well waters and polymeric membranes PAN-15 and PSF-13) and in [13], from which the characteristic decay time of permeate flux in the process is determined:

$$t_0 = 1/a \tag{4}$$

where: a - stands for the slope of straight line, corresponding with the equation y = ax + b, which is modeling the filtration process for the investigated membrane.

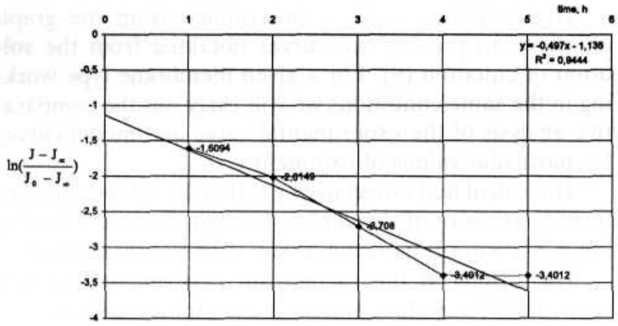

Fig. 2. Well water. Polymeric flat membrane PAN-15.

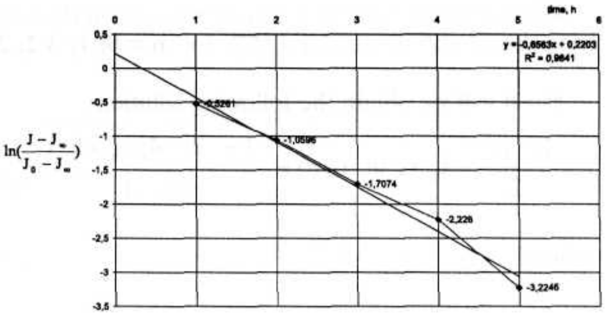

Fig. 3. Well water. Polymeric flat membrane PSF-13.

## Filtration Model in Constant Pressure Conditions

The generalized filtration model for constant pressure conditions was presented by Hermia in the form of a generalized equation [14]:

$$\frac{d^2t}{dV^2} = k\left(\frac{dt}{dV}\right)^n \tag{5}$$

where:
V - volumetric efficiency of filtration
t - filtration time
k,n - coefficients characterizing the mechanisms of filtration process.

The results of the analyses [15-22] were used for the description of filtration in constant pressure conditions.

The equation (5) is complemented with the following conditions:

$$V(t)\,|_{t=0} = 0 \qquad \text{and} \qquad \frac{dV}{dt}\,|_{t=0} = F_0 \cdot J_0 \tag{6}$$

or

$$V(t)\,|_{t=0} = 0 \qquad \text{and} \qquad V(t)\,|_{t=t_0} = V_0 \tag{7}$$

The value of coefficient k occurring in the equation (5) can change depending on the exponent n, which slightly impedes the analysis of filtration process.

In order to introduce the dimentionless value of coefficient $k_n$ (from equation 5), we introduce new quantities referring to the dimentionless volume (u) and dimentionless time (τ):

$$\tau = \frac{t}{t_0} \qquad \text{and} \qquad \upsilon = \frac{V}{V_0} \tag{8}$$

Hence, after the transformation we obtain:

$$\frac{d^2\tau}{d\upsilon^2} = \left(k \cdot \frac{t_0^{n-1}}{V_0^{n-2}}\right) \cdot \left(\frac{d\tau}{d\upsilon}\right)^n \quad \text{or} \quad \frac{d^2\tau}{d\upsilon^2} = k_n \cdot \left(\frac{d\tau}{d\upsilon}\right)^n \tag{9}$$

Equations (5) and (9) contain apparently two parameters k and n or ($k_n$ and n). Transforming respectively the conditions (6) for the equation (9) we obtain:

$$\upsilon(\tau)|_{\tau=0} = 0 \quad \text{and} \quad \frac{d\upsilon}{d\tau}|_{\tau=0} = j_0 \tag{10}$$

where:

$$J_s = \frac{1}{F_0} \cdot \frac{V_0}{t_0}; \qquad j_0 = \frac{J_0}{J_s}$$

$j_0$ - dimentionless opening flux of the filtration,

$F_0$ - opening surface of the filtration,

$J_s$ - average filtration flux with respect to opening surface,

$V_0$ - filtrate volume after time $t_0$

Now, referring to the equation (1) we can write as follows:

$$J_0 = \left(\frac{1}{F} \cdot \frac{dV}{dt}\right)|_{t=0} \quad \text{therefore} \quad j_0 = \frac{\left(\frac{dV}{dt}\right)|_{t=0}}{\left(\frac{V_0}{t_0}\right)} \tag{11}$$

Then, $j_0$ has the following graphical representation (Fig. 4):

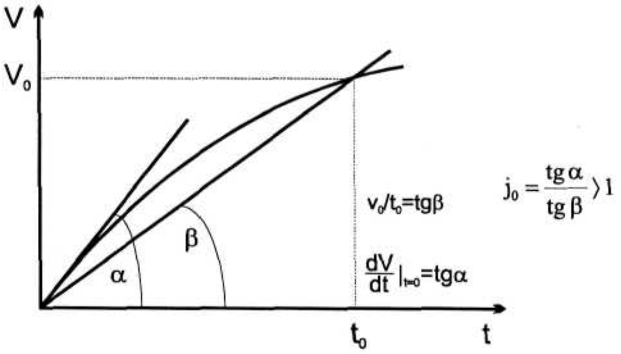

Fig. 4. Graphical interpretation of the dimentionless flux $j_0$

Conditions presented by the equation (7) assume the following form:

$$\upsilon(\tau)\,|_{\tau=0} = 0 \qquad \text{and} \qquad \upsilon(\tau)\,|_{\tau=1} = 1 \tag{12}$$

We determine now the solution of equation (9), satisfying the conditions (10) and (12), for: n = 0; 1; 3/2; 2.

For n = 0 we obtain the following solution:

$$\tau = \frac{k_0 \cdot \upsilon^2}{2} + j_0 \cdot \upsilon \quad \text{or} \quad \upsilon(\tau) = \frac{-1 + \sqrt{1 + 4j_0|j_0 - 1| \cdot \tau}}{2|j_0 - 1|} \tag{13}$$

$$\text{where:} \qquad k_0 = 2\frac{|j_0 - 1|}{j_0} \tag{14}$$

In the analogous way for n = 1 the solution is expressed with the following equation;

$$\upsilon(\tau) = \frac{1}{k_1} \cdot \ln(1 + j_0 \cdot k_1 \cdot \tau)$$

$$\text{or} \tag{15}$$

$$\upsilon(\tau) = \frac{1}{2|j_0 - 1|} \cdot \ln\left(1 + \left(e^{2|j_0 - 1|} - 1\right) \cdot \tau\right)$$

$$\text{where:} \quad k_1 = 2|j_0 - 1| \tag{16}$$

For **n = 3/2** the solution has the following form:

$$\upsilon(\tau) = \frac{1}{k_{3/2}} \cdot \left(2\sqrt{j_0} - \frac{4}{\frac{2}{\sqrt{j_0}} - k_{3/2} \cdot \tau}\right)$$

$$\text{or} \tag{17}$$

$$\upsilon(\tau) = \frac{j_0}{|j_0 - 1|} \cdot \left(1 - \frac{1}{|j_0 - 1| \cdot \tau + 1}\right)$$

$$\text{where:} \qquad k_{3/2} = \frac{2|j_0 - 1|}{\sqrt{j_0}} \tag{18}$$

In the same way, for **n = 2**, the solution has the following form:

$$\upsilon(\tau) = \frac{j_0}{k_2} \cdot \left(1 - e^{-k_2 \cdot \tau}\right) \quad \text{or} \quad \upsilon(\tau) = \frac{1 - e^{-k_2 \cdot \tau}}{1 - e^{-k_2}} \tag{19}$$

$$\text{where:} \qquad k_2 = 2 \cdot \frac{|j_0 - 1|}{j_0} \tag{20}$$

Based on the results obtained from the experimental tests we make the dependence graphs of dimentionless volume as the function of dimentionless time x (Fig. 5).

Subjecting the above graphs to linear approximation we obtain the following;

$$\upsilon_{exp}(\tau) = c \cdot \tau \tag{21}$$

where c stands for average increase rate of dimentionless volume as the function of dimentionless time.

The numerical value c determined from the graph allows us to present the curves obtained from the solution of equation (9). For a given membrane type working in the same conditions we will carry out the comparative analysis of the experimental curve and model curves for particular values of exponent n.

The calculated correlation coefficient will be the conformity measure of the curves. Its highest value indicates the best conformity of the model with the experiment.

The results of the carried out experiments for well waters and for surface waters are presented in Table 1.

The majority of membrane separation processes involve the micro- and macromolecular mixtures, except for microfiltration and, to a certain degree, the ultrafiltration process of colloidal suspension, which are connected with the separation of the components of heterogenic mixtures. In the ultrafiltration process the character of the mixture can change due to the change of environmental conditions such as temperature, reaction, concentration, etc. The formation of aggregates or their disintegration is possible during the ultrafiltration of macromolecules and is connected with the distribution of electrical charges of the molecules. The selection of an appropriate model in such a case is difficult.

The generalized model describing major filtration cases of suspensions in constant pressure conditions was presented by Hermia [14], Fig. 6. The model in question describes the change of filtration process efficiency using a simple two-parameter equation (5), where V stands for filtrate volume and t for filtration time. The constants in this equation are characteristic for different mechanisms

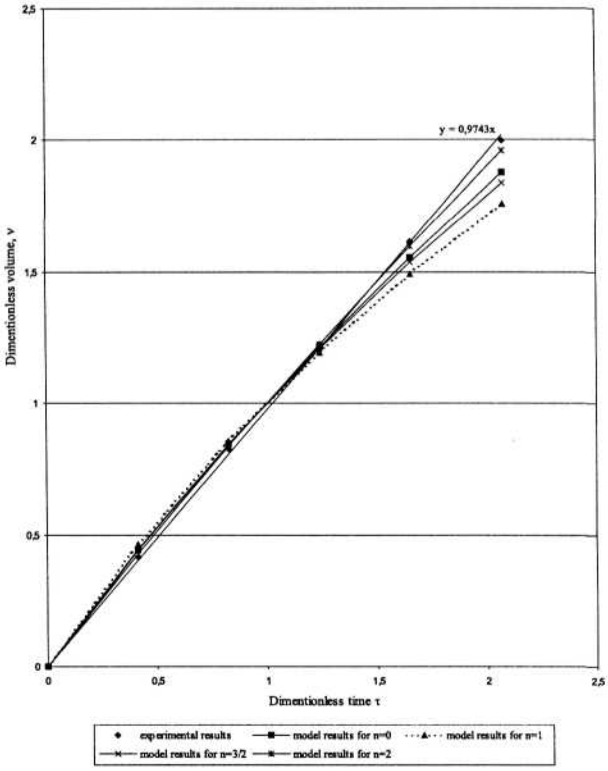

Fig. 5. Dependence graphs of dimentionless volume u as the function of dimentionless time τ for well water and membrane PAN-13

Table 1. Correlation coefficient involving the fitting of experimental curve and model curves depending on the value n for the filtration process using different membranes.

| Membrane type | Correlation coefficient | | | | |
|---|---|---|---|---|---|
| | n = 0 | n = 1 | n = 3/2 | n = 2 | Value of correlation coefficient max. |
| Well waters | | | | | |
| PAN-13 | 0.999078 | 0.996374 | 0.998418 | 0.999847 | 0.999847 |
| PAN-15 | 0.999167 | 0.997983 | 0.998817 | 0.999969 | 0.999969 |
| PSF-13 | 0.999622 | 0.981059 | 0.995442 | 0.996626 | 0.999622 |
| PSF-15 | 0.998210 | 0.989558 | 0.995040 | 0.997200 | 0.998210 |
| PAN/PSF-15 | 0.999706 | 0.999262 | 0.999653 | 0.999995 | 0.999995 |
| UF 15 kD | 0.996184 | 0.993517 | 0.994127 | 0.999103 | 0.999103 |
| UF 300 kD | 0.997359 | 0.995756 | 0.996382 | 0.999514 | 0.999514 |
| MF 0.1 μm | 0.996405 | 0.993854 | 0.994291 | 0.999378 | 0.999378 |
| $PP_{capillary}$ | 0.999907 | 0.997307 | 0.999791 | 0.999166 | 0.999907 |
| $PSF_{capillary}$ | 0.999587 | 0.998210 | 0.999431 | 0.999946 | 0.999946 |
| Surface waters | | | | | |
| PAN-13 | 0.999782 | 0.997285 | 0.999620 | 0.998975 | 0.999782 |
| PAN-15 | 0.999983 | 0.998284 | 0.999955 | 0.999935 | 0.999983 |
| PSF-13 | 0.999893 | 0.993002 | 0.999723 | 0.999272 | 0.999893 |
| PSF-15 | 0.999995 | 0.999954 | 0.999997 | 0.999981 | 0.999997 |
| PAN/PSF-15 | 0.999704 | 0.994078 | 0.999965 | 0.999522 | 0.999965 |
| UF 300 kD | 0.999509 | 0.997679 | 0.999265 | 0.999899 | 0.999899 |
| MF 0.1 μm | 0.999584 | 0.999058 | 0.999589 | 0.999662 | 0.999662 |
| MF 0.2 μm | 0.999454 | 0.997426 | 0.999181 | 0.999835 | 0.999835 |
| MF 0.45 μm | 0.996123 | 0.987892 | 0.998386 | 0.846029 | 0.998386 |
| $PP_{capillary}$ | 0.999480 | 0.998652 | 0.999275 | 0.999993 | 0.999993 |
| $PSF_{capillary}$ | 0.999992 | 0.999976 | 0.999992 | 0.999994 | 0.999994 |

reducing the efficiency during the constant-pressure filtration. They can therefore be applied as convenient criteria to identify these mechanisms after the experiment has been carried out.

- According to the mechanism of complete blocking of pores it is assumed that all molecules of the suspension take part in the blocking of pores. Each molecule which comes to a hole in the filtering surface effects its blocking. The molecules do not subside on each other and do not exert any other influence on each other. When such assumptions are made, we have proportional dependence between the filtrate volume and the blocked surface of the membrane. It is expressed by proportion constant. Respective constants in the general equation can be expressed in ihe following way:

$$n = 2, \qquad k = \sigma u_0$$

where: $u_0$ stands for linear velocity of filtrate.

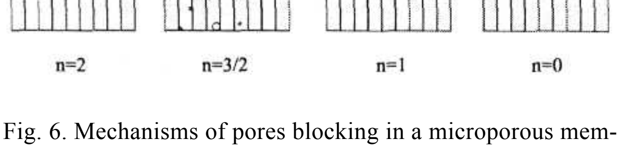

Fig. 6. Mechanisms of pores blocking in a microporous membrane.

- The mechanism of standard blocking of pores assumes that the blocking is taking place inside the pores whose volume is getting smaller proportionally to the filtrate volume. It is assumed that all pores have the same diameter and length at the initial instant. The parameters of the equation (5) have in such a case the following form:

$$n = \frac{3}{2}, \qquad k = 2\frac{C}{l}\sqrt{\frac{J_0}{F_0}}$$

where: l is the length of pores, $F_0$ is the surface of pores at the initial instant, $J_0$ is the permeate flux at the initial instant and C is the volume ratio between the stopped suspension particles and volume of permeate which passed through the membrane during that time period.

- Transitory mechanism of pores blocking assumes that not all particles reach the membrane pores, but some of them subside on particles that have already settled on the membrane. The probability that the particles will reach the pores which have not been yet blocked is getting smaller and smaller as the surface of these pores is getting smaller. The constants in the equation (5) can be expresses as follows:

$$n = 1, \qquad k = \frac{\sigma}{F_0}$$

- According to the cake mechanism of pores blocking, the flow of filtrate is getting smaller due to the increase of resistance in the layer by the wall of the membrane caused by the accumulation of the mass of components retained on the membrane. The total resistance is in such a case understood as the total of cake resistance values and the resistance of the membrane itself. It is also assumed that the resistance of cake is proportional to the mass of the settled substance:

$$R(t) = R_0 + \frac{\alpha \cdot \rho \cdot \phi \cdot V(t)}{(1 - m \cdot \phi) \cdot F}$$

where: $\alpha$ - i s the specific resistance of filtration cake, $\rho$ - filtrate density, $\varphi$ - share of solid body in the filtrated suspension, m - ration between the mass of humid cake and dry cake. The values of parameters in the general equation are as follows:

$$n = 0, \qquad k = \frac{\alpha \cdot \rho \cdot \phi}{F^2 \cdot R_0 \cdot J_0 \cdot (1 - m \cdot \phi)}$$

It should be emphasized that parameter n may assume only discrete values: 2, 3/2, 1 and 0, whereas constants k have different dimensions for each case, in order to preserve the physical interpretation of the events, according to the model.

The model can be applied as the convenient criterion to identify these mechanisms after the experiment has been completed, by recording the volumetric permeate flux in time, in constant pressure conditions.

The mechanism of pores blocking in microporous membranes for the investigated cases corresponds with the value n = 0 for the following polymeric membranes: PAN 13, PAN 15, PSF 13, PSF 15, $PP_{cap}$, and the value n = 2 for ceramic membranes and the capillary membrane PSF, since the correlation coefficient has the highest value. However, it is difficult to assess unmistakably that the blocking mechanism of pores is carried out with a 100% correspondence with the assumption on complete blocking of pores, i.e. when n = 2, since when analyzing the values of correlation coefficient for the other calculated values n = 3/2,1 and 0 it was found that they do not differ considerably from each other. We must make a supposition that the process is carried out according to the mixed mechanism, that is, when a part of the particles effect complete blocking and the remaining ones settle down, and the cake blocking mechanism of permeate flow is observed.

## Conclusions

- It has been found that the mechanism of pores blocking in membranes is most often carried out according to the cake blocking mechanism of pores, and in some types of membranes it corresponds with the mechanism of complete blocking of pores.
- Characteristic time of filtration process decay was determined for particular membranes and investigated waters. Good correlation was obtained between the experimental and theoretical results following from the averaged mass balance equation with respect to space coordinate.

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