Application of Low-Pressure Reverse Osmosis Membranes for Drinking Water Softening

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ABSTRACT

The processes of water purification with increasing selection of permeate were studied, considering selectivity and productivity of membranes, dynamics of changes of contents of components in the concentrate. It is shown that when chlorides and sulfates are removed from water, the increase in their content in the concentrate does not differ practically from the measured and calculated values. At the same time, the nature of dependences on the change in hardness, concentration of calcium and magnesium ions, alkalinity obtained experimentally differ significantly from the dependences obtained by theoretical calculations at permeate selection levels of > 70%. A significant difference in the determined and calculated concentrations of hardness ions in the concentrates was observed after hardness values greater than 30–40 mg-eq/dm^3. This indicates the partial removal of hardness ions and carbonates from the concentrates, which may be the reason for the formation of deposits on the membrane. Permissible values of the degree of permeate selection were determined, at which there is no intense deposition of carbonates and hydroxides of hardness ions on the membrane. With the initial water hardness > 8 mg-eq/dm^3, the degree of permeate selection could reach 60–70% without the risk of sedimentation on the membrane. Effectiveness of the low-pressure reverse osmosis membrane in the purification of mine water with an increased level of mineralization and hardness was determined. A significant difference between the determined and calculated hardness in concentration was observed already at the degree of permeate selection of 22–33%.

Keywords: mineralization, hardness, softening, desalination, reverse osmosis, ion exchange, membrane.

INTRODUCTION

The problem of ensuring the proper ecological state of the water resource potential remains relevant for many regions. As a result of the negative anthropogenic impact on water facilities, there is a problem of providing the population with high-quality drinking water. In addition, water supply sources in several regions are characterized by low water quality due to natural factors. Often, in coastal regions, underground and groundwater are characterized not only by increased mineralization, but also by increased hardness. There are regions where ground and underground waters, with satisfactory indicators for most characteristics, have hardness at the level of 8–15 mg-eq/dm^3, which significantly exceeds the permissible levels [SSNR 2.24–171–10 2010; Directive 98/83/EC 1998].

The most common method used in industry for water softening is the reactive method of softening [Seminska et al. 2017]. In this method, calcium carbonate and magnesium hydroxide are separated in the form of sediments, which are much easier to recycle compared to liquid waste. The disadvantage of the method is the relatively low water softening efficiency. Although recently a new approaches have been developed to increase the efficiency of the process through the combined use of aluminum and iron coagulants along with soda and lime [Makarenko 2014]. However, this method is hardly used for the preparation of drinking water because the water treated in this way often has a pH of > 9, which is

unacceptable for drinking water. Moreover, complex equipment is used in this process, which cannot be used in local water purification systems, and even more so in domestic conditions.

The use of ion exchange in water softening processes allows working within a wide range of pH of the solution and reaching a given depth of water softening [Lubavina et al. 2007; Shabliy et al. 2010]. The disadvantage of the method is the formation of significant volumes of liquid waste during the regeneration of cationites with a level of mineralization. Dumping such solutions into the sewer is a gross violation of environmental legislation. Today, a method of processing these solutions is known, which allows you to use the sodium chloride solution multiple times with the separation of calcium carbonate and magnesium hydroxide precipitates [Nosacheva et al. 2008]. These sediments are suitable for use in the production of building materials [Fleysher et al. 2014]. These approaches to water softening are promising for industrial installations. When used in everyday life, it is difficult to organize even a simplified version of cationite regeneration using only sodium chloride solutions.

It is obvious that a simpler and more effective method of softening underground and soil water can be membrane methods [Goncharyk et al. 2009; Gomelya et al. 2014]. Especially when the hardness of natural water does not exceed 30 mg-eq/dm³.

Of course, when the hardness salts are concentrated at high values of the permeate selection step, the hardness can reach and exceed 100 mg-eq/dm³, which will cause sedimentation on the membrane. However, at pH < 8.0, there will be mainly hydrocarbons in the solution, which will contribute to increasing the stability of the concentrates before sedimentation.

The purpose of this work was to determine the efficiency of using low-pressure reverse osmosis membranes using the example of Filmtech TW30–1812–50 for conditioning water with an increased level of hardness, and to determine the conditions for preventing sediment formation on the membrane.

To achieve the goal, the following tasks must be solved:
1. To determine the effect of initial water hardness and the degree of permeate selection on the selectivity and performance of the membrane with respect to hardness ions, alkalinity, chlorides and sulfates, and changes in the content of components in the concentrate.
2. Determine the conditions for using a low-pressure reverse osmosis membrane to soften water without the formation of carbonate deposits on the membrane.

MATERIALS AND METHODS

Membrane water softening and desalination processes were carried out using Filmtech TW30–1812–50 low-pressure reverse osmosis membrane cassettes. The medium used was: artesian water with the following characteristics: $H = 8.00–8.72$ mg-eq/dm³, $C_{Ca}^{2+} = 6.00–6.24$ mg/dm³, $A = 7.80–8.08$ mg-eq/dm³, $C_{Cl}^{-} = 11.34–15.50$ mg/dm³, $C_{SO4}^{2-} = 10.00–12.50$ mg/dm³, $pH = 7.21–7.65$; softened artesian water with characteristics: $H = 2.44$ mg-eq/dm³, $C_{Ca}^{2+} = 0.6$ mg/dm³, $C_{Mg}^{2+} = 1.84$ mg/dm³, $A = 5.20$ mg-eq/dm³; mine water (Baranovska mine) with characteristics: $H = 21.00$ mg-eq/dm³, M (mineralization) $= 2.10$ g/dm³, $A = 7.50$ mg-eq/dm³, $C_{Cl}^{-} = 1100.5$ mg/dm³, $C_{SO4}^{2-} = 250$ mg/dm³.

Using a pump, water was supplied to a reverse osmosis filter with a Filmtech TW30–1812–50 membrane cartridge. The pressure in the system is 3–5 atm. Permeate samples with a volume of 1 dm³ were taken. The initial volume of water was 10–11 dm³. The concentrate was returned to the container with the working solution. The pressure in the system was maintained by regulating the water supply by the pump and the flow rate of the concentrate. Hardness, concentration of calcium ions, alkalinity, pH of the medium, and concentration of chlorides and sulfates were determined in each permeate sample and in the concentrate after the permeate sampling. Using a stopwatch, the time required to collect 1 dm³ of permeate was determined.

Hardness and concentration of Ca^{2+} ions were determined by the trigonometric method [Nabyvanec et al. 2006], alkalinity was determined by the acid-base titration method, sulfate concentration was determined by the photometric method with barium ions, chloride concentration was determined by Mohr's argonometric method [Nabyvanec et al. 2006].

The selectivity of the membrane (R, %) was calculated according to the formula:

$$R = C_0 - C_n / C_0 \cdot 100, \% \qquad (1)$$

where: R – selectivity of the membrane, %;

C_0 та C_n – concentration of the component, respectively, in the initial solution and permeate.

Content of components Ca^{2+}, Mg^{2+}, HCO_3^-, Cl^-, SO_4^{2-} in i-th sample of concentrate (C_{ki}, mg/dm³ or mg-eq/dm³) was calculated according to the formula:

$$C_{ki} = V_0 C_0 - \sum_{i=1}^{n} (C_{pi} V_{pi}) / V_0 - \sum_{i=1}^{n} V_{ni}, \quad (2)$$
$$(mg/dm^3 \text{ or } mg\text{-}eq/dm^3)$$

where: C_{ki} – the concentration of the component in the concentrate after taking the i-th permeate sample, mg/dm³ or mg-eq/dm³;

C_0 and V_0 – concentration of the component (mg/dm³; mg-eq/dm³) in the initial solution and volume (dm³) of the initial solution;

V_{pi} – permeate sample volume (dm³);

C_{ni} – the concentration of the component in the i-th permeate sample, mg/dm³; mg-eq/dm³;

n – number of permeate samples ($n = 7$; 10).

Membrane performance (transmembrane flux rate) (j) (dm³/m²·h) was calculated according to the formula:

$$j = V_n / S \cdot \Delta\tau, \, (dm^3/m^2 \cdot h) \quad (3)$$

where: j – membrane performance, dm³/m²·h;

V_n – permeate sample volume, dm³;

S – membrane area, m²;

$\Delta\tau$ – permeate sampling time, hours.

RESULTS AND DISCUSSION

It is known that artesian waters are characterized by high quality for providing the population with drinking water. Water with increased hardness values (8–15 mg-eq/dm³) that do not meet the requirements of regulatory documents for drinking water are quite common [SSNR 2.24–171–10 2010; Directive 98/83/EC 1998]. At the same time, these waters are characterized by elevated values of alkalinity, and sometimes mineralization. The use of local reverse osmosis installations in these cases is quite promising.

In this work, when softening artesian water with a hardness of 8.72 mg-eq/dm³ using a low-pressure reverse osmosis membrane (Fig. 1), good results were obtained. Already at a pressure in the system of 3 atm, the productivity of the membrane reached ~ 20 dm³/m²·h. At low water mineralization (M < 200 mg/dm³), the osmotic pressure on the membrane was negligible. In addition, the membrane ensured high water purification efficiency. The selectivity for hardness ions exceeded 98%, for hydrocarbonates 95% with a degree of permeate selection from 10 to 90%. The efficiency of chloride removal was the lowest, $R = 70–90\%$. However, at low chloride concentrations, this result is quite satisfactory. The selectivity of the membrane is also high when removing sulfates (R = 90–98%).

However, the results were less optimistic when analyzing the composition of concentrates formed during water softening (Fig. 2, Fig. 3).

As can be seen from Fig. 2, the level of water hardness in permeate gradually increases with an increase in the degree of permeate selection.
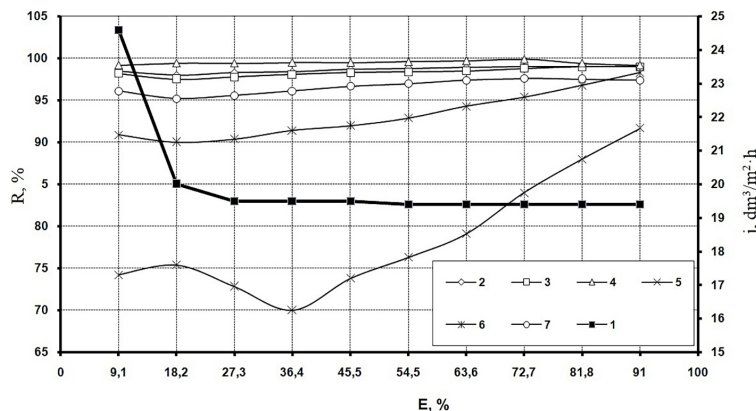

Figure 1. Change in performance (*j*) (1) and selectivity (R) (2; 3; 4; 5; 6; 7) of the low-pressure reverse osmosis membrane Filmtec TW30–1812–50 for hardness (2), calcium (3), magnesium ions (4), chlorides (5), sulfates (6), alkalinity (7) when filtering artesian water ($H = 8.72$ mg-eq/dm³, $C_{Ca}^{2+} = 6.24$ mg-eq/dm³; $A = 8.08$ mg-eq/dm³; $C_{Cl}^- = 11.34$ mg/dm³; $C_{SO4}^{2-} = 10.10$ mg/dm³; pH = 7.67) ($P_p = 3$ atm) ($V = 11$ dm³, $V_n = 1$ dm³)

However, even with the selection of 90% of the permeate, the hardness of the water did not exceed 0.34 mg-eq/dm³. A similar trend was observed when the alkalinity of permeate changed. However, when studying the dynamics of growth of hardness and alkalinity in the concentrate, a significant difference is observed between the measured and calculated values of hardness and alkalinity at permeate selection levels of > 70%. In the case of chlorides and sulfates (Fig. 3), a similar trend was not observed. Obviously, despite the high solubility of hydrocarbons at pH < 8.0, in this case, a certain precipitation of hardness ions from the concentrate was observed. If we

consider that the level of alkalinity significantly decreased in comparison with the calculated one, it is quite likely that calcium and magnesium carbonates were precipitated. This can be facilitated by a certain increase in the pH of the concentrate when filtering water through a reverse osmosis filter (Fig. 4). The increase in the pH of the concentrate and the decrease in the pH of permeate may be related to the better diffusion through the membrane of protons, compared to sodium cations and hydroxyl anions.

It should be noted that during reverse osmotic water desalination, a decrease in the determined content in the concentrate was noted in
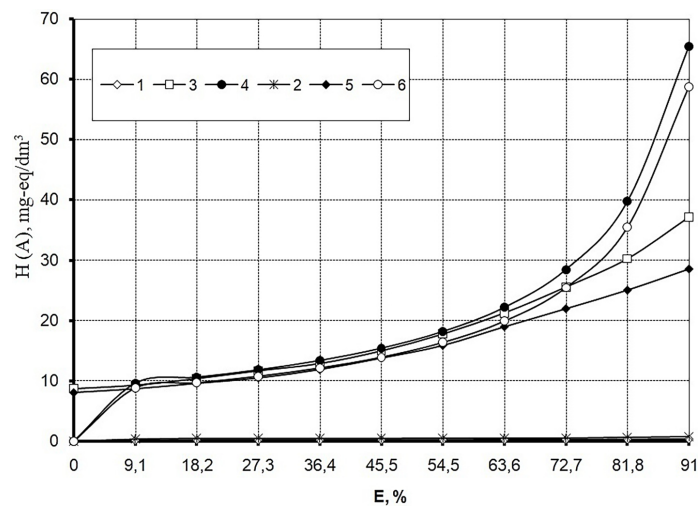
Figure 2. Dependence of hardness (1) and alkalinity (2) of the permeate, hardness measured (3) and calculated (4), alkalinity determined (5) and calculated (6) in the concentrate during filtration of artesian water through a low-pressure reverse osmosis membrane (P_p = 3 atm) (V = 11 dm³, V_n = 1 dm³)
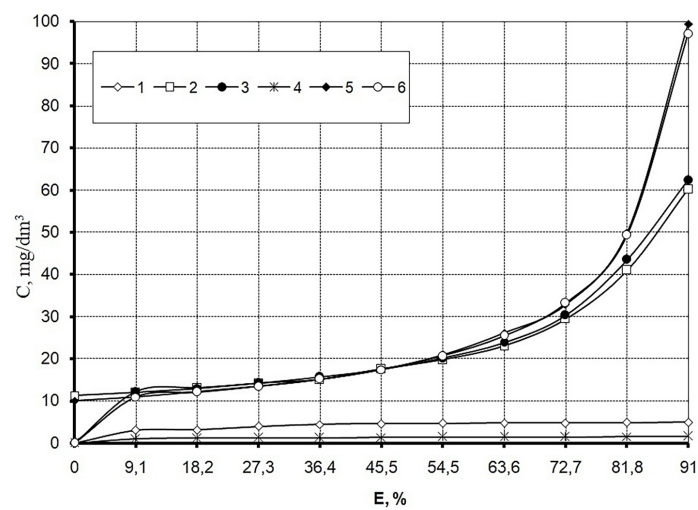
Figure 3. Change in the concentration of chlorides (1; 2; 3) in permeate (1), concentrate (2) calculated in concentrate (3), sulfates (4; 5; 6) in permeate (4), concentrate (5) calculated in concentrates (6) with an increase in the degree of permeate selection when filtering artesian water (H = 8.72 mg-eq/dm³, C_{Ca}^{2+} = 6.24 mg-eq/dm³; A = 8.08 mg-eq/dm³; pH = 7.61; C_{Cl}^{-} = 11.34 mg/dm³; $C_{SO_4}^{2-}$ = 10.10 mg/dm³) through a low pressure reverse osmosis membrane (water volume V = 11 dm³, sample volume V_n = 1 dm³), P_p = 3 atm

comparison with the calculated values not only of calcium ions, but also of magnesium ions (Fig. 5). Obviously, in this case, the co-precipitation mechanism of calcium and magnesium carbonates is implemented.

It can be seen from the above results that a significant difference in the determined and calculated concentrations of hardness ions in the concentrates is observed only after hardness values greater than 30–40 mg-eq/dm³ are reached at a degree of permeate selection of 70–90%. Obviously, when using water with reduced hardness, the probability of sediment formation will be lower.

Thus, when softened artesian water (Fig. 6, Fig. 7) was used, effective desalination of water

was observed (Fig. 6) with high values of selectivity and productivity of the membrane with a low probability of sediment formation on its surface (Fig. 7). In this case, the determined and calculated hardness and alkalinity values of the concentrate did not differ practically.

It is clear that the use of reagent softening before reverse osmosis is appropriate only in industrial installations of large capacity. It is simpler to use ion exchange to stabilize water treatment before reverse osmosis [Makarenko et al. 2013; Gomelya et al. 2014]. This approach is appropriate only when the water is characterized by high mineralization. In other cases, you can limit yourself only to ion-exchange or baromembrane water
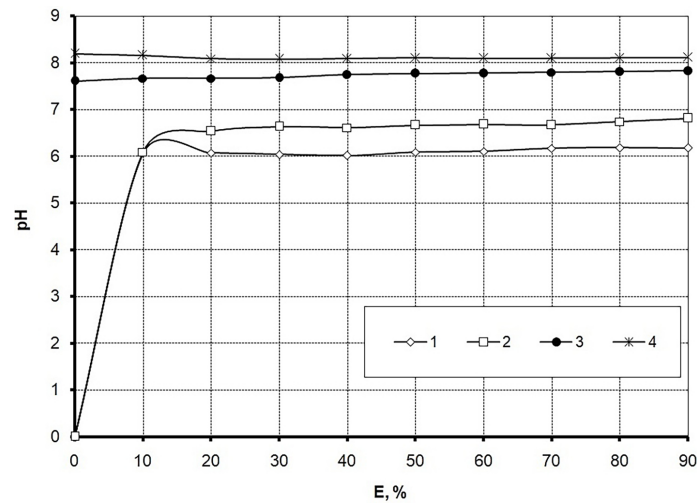

Figure 4. Change in pH (1; 2; 3; 4) in permeate (1; 2) and concentrate (3; 4) with increasing degree of artesian water permeate selection (1; 3) and softened artesian water (2; 4)
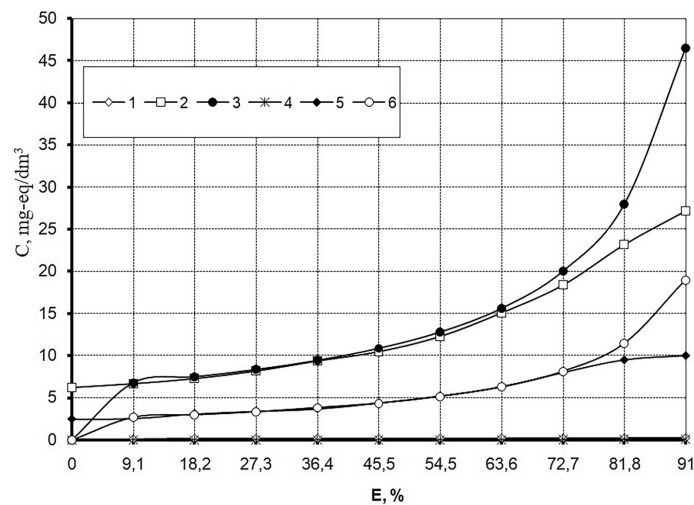

Figure 5. Changes in the concentration of calcium (1; 2; 3) magnesium (4; 5; 6) in the permeate (1; 4), the measured concentration (2; 5) and the calculated concentration (3; 6) with an increase in the degree of permeate selection during artesian filtration water through a low-pressure reverse osmosis filter (P_p = 3 atm, V = 11 dm³, V_n = 1 dm³)
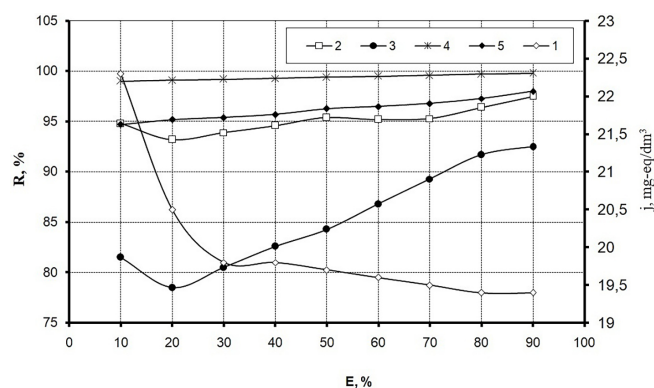
Figure 6. Change in productivity (1) and selectivity (2; 3; 4; 5) of a low-pressure reverse osmosis membrane in terms of hardness (2), calcium (3), magnesium (4) and alkalinity (5) of softened artesian water depending on the degree of permeate selection ($V = 10$ dm³, $V_n = 1$ dm³, $P_p = 3$ atm)

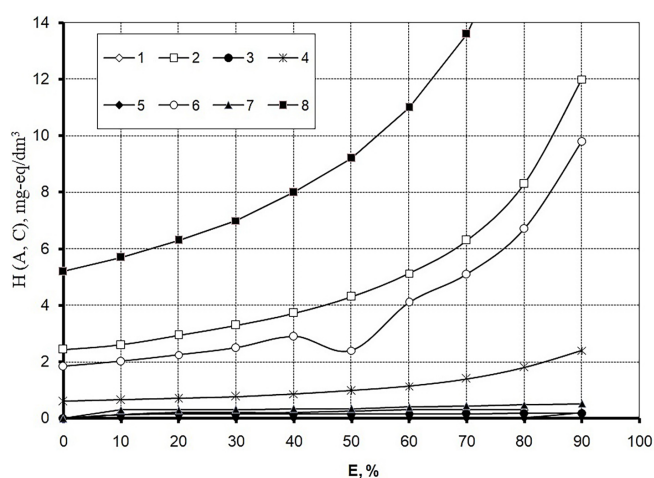**Figure 7.** Change in hardness (1; 2) of softened artesian water ($H = 2.44$ mg-eq/dm³, C_{Ca}^{2+} = 0.6 mg-eq/dm³; $C_{Mg}^{2+} = 1.84$ mg-eq/dm³, $A = 5.2$ mg-eq/dm³; pH = 8.162) calcium (3; 4), magnesium (5; 6) and alkalinity (7; 8) concentrations in permeate (1; 3; 5; 7) and concentrate (2; 4; 6; 8) when filtered through a low-pressure membrane ($V = 10$ dm³, $V_n = 1$ dm³) (calculated values in the concentrate, mg-eq/dm³: $H = 12.37$; $A = 26.0$; $C_{Ca}^{2+} = 2.6$; $C_{Mg}^{2+} = 10.03$)

softening. When using reverse osmosis water desalination to prevent deposits on the membrane, it is advisable to reduce the degree of permeate selection. Thus, when the degree of permeate selection was reduced to 60–70% (Fig. 8, Fig. 9, Fig. 10), effective water softening was achieved. At the same time, the content of hardness ions and alkalinity in the concentrate was close to the calculated values. Obviously, precipitation of hardness ions in this case practically did not occur.

Interesting results were obtained during low-pressure reverse osmosis membrane filtration of mine water with hardness of 21 mg-eq/dm³, mineralization of 2.1 mg/dm³, chloride content ~ 1100 mg/dm³, sulfates 250 mg/dm³.

The results are shown in Fig. 11. In this case, the performance of the membrane at a pressure of 5 atm decreased to 12 dm³/m²·h.

When the degree of permeate selection was reached at the level of 55%, as a result of the growth, the productivity of the membrane decreased to 8.6–4.6 dm³/m²·h. The selectivity for all components, except for chlorides and hydrocarbons, was quite high – at the level of 98–99%. The selectivity for hydrocarbons varied from 90 to 98%, for chlorides – from 89 to 95%. But 91% of 110 mg/dm³ reach 121 mg/dm³. And when the chloride content in the concentrate is constantly increasing, even with a relatively high selectivity of the membrane, the concentration of chlorides in permeate will be quite high (table 1). As can be seen from the table, according to ions of hardness, alkalinity, sulfate concentration, mineralization, cleaning efficiency was satisfactory up to the degree of permeate selection of 98%. According to chlorides – up to the degree of selection
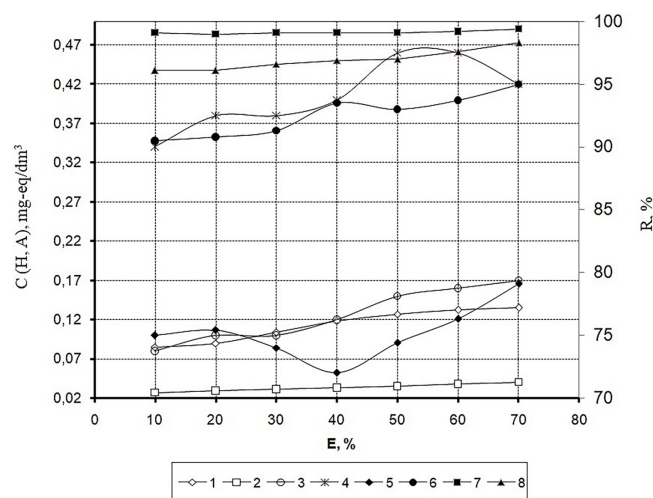
Figure 8. Dependence of the concentration of chlorides (1), sulfates (2), hardness (3), alkalinity (4) of the permeate, the selectivity of the reverse osmosis membrane for chlorides (5), sulfates (6), hardness ions (7) and alkalinity (8) on the degree of selection of permeate when filtering artesian water
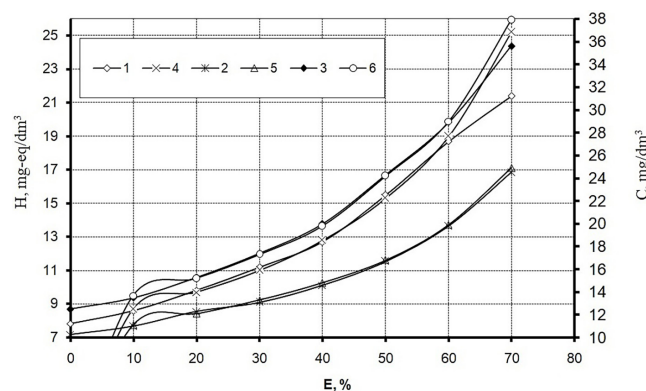

Figure 9. Changes in the determined (1; 2; 3) and calculated (4; 5; 6) values of alkalinity (1; 4) of the concentration of chlorides (2; 5) and sulfates (3; 6) in the concentrate with an increase in the degree of permeate selection during reverse osmosis purification artesian water ($V_p = 7$ dm^3, $V_n = 1$ dm^3, $P = 3$ atm)

of permeate 78%. This is a fairly high efficiency of water purification and desalination, if we take into account the small operating pressure and the satisfactory performance of the membrane.

It should be noted that during the desalination of mine water, a significant difference between the determined and calculated hardness in concentration was observed already at the degree of permeate selection of 22–33%. At the degree of permeate selection ~ 88%, a decrease in the content of sulfates in the concentrate was also observed, in comparison with the calculated one. This can be explained by the fact that at a sulfate concentration of 46 mg-eq/dm^3 and a calcium concentration of ~ 100 mg-eq/dm^3, precipitation of calcium sulfate is possible. In this case, it is advisable to apply preliminary softening of water before ion exchange [Makarenko et al. 2013; Gomelya et al. 2014]. It is possible to use antiscalants.

Undoubtedly, under these conditions, it is necessary to carry out periodic acid washing of the membrane. In domestic conditions, it is advisable to use 1–3% solutions of citric acid.

CONCLUSIONS

The processes of water softening when using low-pressure membranes were studied. It is shown that these membranes are characterized by high efficiency in the purification of artesian water pipes of hardness ions and sulfates, as well as efficiency in the removal of chlorides and hydrocarbons from water. It was established that when water hardness values in the concentrate are higher than 40 mg-eq/dm^3 with high hydrocarbonate content in treated water the deposition of hardness ions on the membrane can be observed.
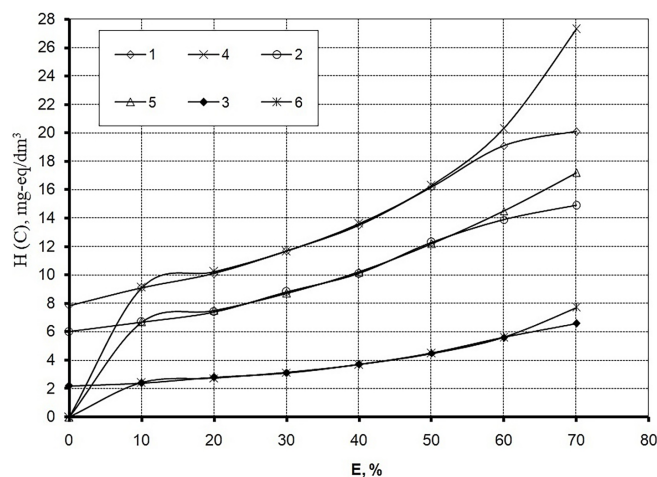
Figure 10. Change in the determined (1; 2; 3) and calculated (4; 5; 6) hardness values (1; 4), calcium (2; 5) and magnesium (3; 6) concentration in the concentrate with an increase in the degree of permeate selection during filtration artesian water through a low-pressure reverse osmosis filter ($V = 7$ dm^3, $V_n = 1$ dm^3, $P = 3$ atm)
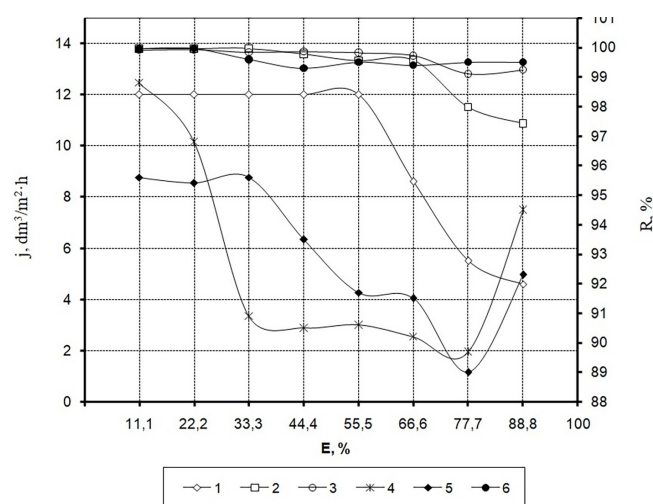

Figure 11. Dependence of the productivity (1) of the reverse osmosis membrane Filmtech TW30–1812–50, its selectivity for hardness ions (2), calcium (3), alkalinity (4), chlorides (5), and sulfates (6) on the degree of permeate selection during filtration water from the mine "Tarnovska"

Table 1. Dependence of permeate characteristics and process parameters on the selected volume of permeate when cleaning 9 dm^3 of water from the "Tarnovska" mine ($H = 21$ mg-eq/dm^3, $M = 2.1$ g/dm^3, $A = 7.5$ mg-eq/dm^3, $C_{Ca}^{2+} = 12.5$ mg-eq/dm^3; $C_{Mg}^{2+} = 8.5$ mg-eq/dm^3; $C_{Cl}^{-} = 1700$ mg/dm^3; $C_{SO4}^{2-} = 250$ mg/dm^3; pH = 7.922) ($P = 5$ atm) on a low pressure reverse osmosis membrane

Vn, dm^3	E, %	H, mg-eq/dm^3	C_{Ca}^{2+}, mg-eq/dm^3	C_{Mg}^{2+}, mg-eq/dm^3	A, mg-eq/dm^3	C_{Cl}^{-}, mg/dm^3	C_{SO4}^{2-}, mg/dm^3	M, g/dm^3	pH	Sampling time, min
0	0	21.0	12.5	8.5	7.5	1100	250	2.1	7.922	-
1	11.1	0.01	0.01	0	0.1	53.3	0.1	0.178	6.078	25
2	22.2	0.01	0.01	0	0.3	63.9	0.1	0.214	6.397	25
3	33.3	0.05	0.03	0.02	1.0	71.0	1.3	0.238	6.342	25
4	44.4	0.08	0.03	0.05	1.5	124.3	2.9	0.305	6.418	25
5	55.6	0.15	0.05	0.10	1.5	195.3	3.1	0.371	6.533	25
6	66.7	0.30	0.10	0.20	2.0	230.8	4.4	0.506	6.805	35
7	77.8	2.0	0.50	1.50	3.0	479.3	5.9	0.827	7.011	55
8	88.9	2.3	0.8	1.50	3.0	621.3	10.0	1.000	7.136	65

Table 2. Changes in the characteristics of the concentrate, measured (I) and calculated (II), process parameters from the selected volume of permeate during the purification of 9 dm³ of water from the "Tarnovska" mine when using a cartridge with a low-pressure reverse osmosis membrane ($P = 5$ atm)

Vn, dm³ (E, %)	H, mg-eq/dm³		$C_{Ca^{2+}}$, mg-eq/dm³		$C_{Mg^{2+}}$, mg-eq/dm³		A, mg-eq/dm³		M, g/dm³		pH	C_{Cl^-}, mg/dm³		$C_{SO_4^{2-}}$, mg/dm³	
	I	II	I	II	I	II	I	II	I	II	I	I	II	I	II
0	21.0	-	12.5	-	8.5	-	7.5	-	2.1	-	7.922	1100	-	250	-
1 (11.1)	22.0	23.63	13.0	14.0	9.0	9.5	8.0	8.4	2.2	2.3	7.630	1210	1231	270	281
2 (22.2)	23.0	27.0	15.0	16.0	10.0	10.9	10.0	9.6	2.4	2.6	7.604	1400	1398	315	321
3 (33.3)	24.0	31.5	19.0	18.8	13.0	12.75	12.0	11.0	2.6	3.0	7.621	1610	1619	380	375
4 (44.4)	25.0	37.8	20.1	22.5	14.0	15.3	13.0	13.0	3.1	3.6	7.599	1920	1918	445	449
5 (55.5)	37.0	47.25	22.3	28.1	17.0	19.1	16.0	15.8	3.8	4.5	7.705	2355	2349	565	561
6 (66.6)	40.0	63.0	25.0	37.4	18.2	25.5	22.0	20.4	4.5	5.9	7.736	3060	3055	751	746
7 (77.7)	74.0	94.50	48.0	55.5	19.5	37.9	25.0	29.1	5.3	8.4	7.728	4340	4342	1020	1116
8 (88.8)	90.0	189.0	60.0	109.4	40.0	80.0	25.0	55.2	6.5	17.24	7.774	8075	8064	1454	2222

This tendency increases when using water with increased hardness, alkalinity and mineralization.

It is shown that with preliminary softening of artesian water or a reduction of permeate selection degree, it is possible to prevent sedimentation on the membranes. With a water hardness of ~ 8 mg-eq/dm³, the degree of permeate selection can reach 60–70% without the risk of sedimentation. In other cases, preliminary water softening is advisable.

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