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The use of membrane filtration for removal of aluminum from drinking water sources

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ABSTRACT

Drinking water sources contain a range of natural or anthropogenic substances, which even in relatively small concentrations need to be removed in order to complete sharpening drinking water standards. Among them, aluminum is one of the elements, which can migrate to drinking water sources by means of elution from natural rocks, as a result of discharge of wastewater containing it or as a result of aluminum-based coagulation treatment processes. In Polish regulation, the permissible amount of aluminum in drinking water is established at 0.2 mg Al/L. On the other hand, in Poland, there appear natural water sources of excellent water quality, which, however, are characterized by elevated aluminum content. In this paper, the research on the removal of aluminum by nanofiltration and ultrafiltration combined with coagulation is discussed. The obtained results suggest that either nanofiltration or ultrafiltration combined with coagulation may be used for effective removal of contaminant from drinking water, regardless of its initial concentration (up to 5 mg Al/L for nanofiltration feed, and up to 18 mg Al/L for ultrafiltration feed), the permeate parameters correspond to drinking water quality standards. The selection between suggested processes depends on the final economic effect related to membrane fouling and cleaning procedures and frequency, the investigation of which has not been the subject of the presented research.

Keywords: Drinking water; Aluminum; Nanofiltration; Ultrafiltration

1. Introduction

Aluminum (Al) is one of the most common metallic elements present in Earth crust (in up to 8%) [1]. It occurs in the environment in forms such as silicates, oxides and hydroxides, which may be combined with other elements, including sodium and fluorine, as well as in the form of complexes with organic matter [1–3]. Aluminum migrates to water during mineral weathering of feldspars, which include orthoclase, anorthite, albite, micas and bauxite, and subsequently ends up in clay minerals. A number of gemstones also contain aluminum, among which examples are ruby and sapphire [4,5].

Industrially, aluminum (metallic) is used as a structural material in the construction, automotive and aircraft branches. It is also applied in the production of metal alloys, in the electric industry, in cooking utensils and in food packaging. Aluminum compounds (inorganic and organic salts and complexes) are used as antacids, antiperspirants and food additives [6–8]. Depending on its origin (natural or anthropogenic) and form of appearance, different Al species may be identified in water, as shown in Fig. 1.

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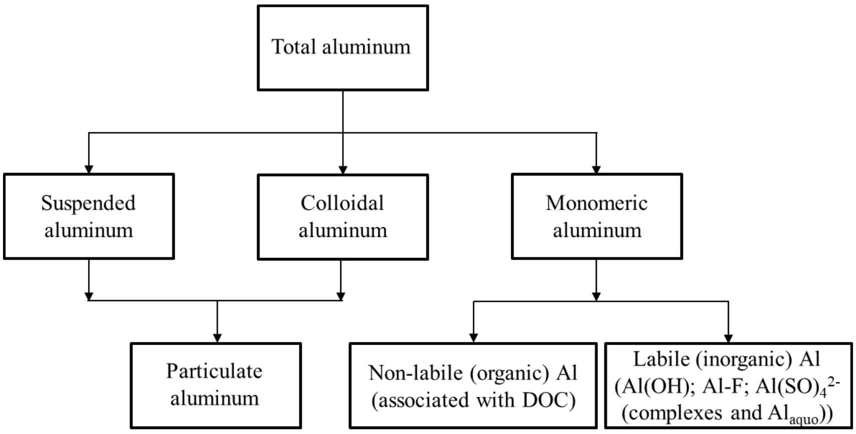

Fig. 1. Aluminum species appear in aqueous streams (natural surface and ground water or wastewater) [6].

Al-based coagulants, that is, aluminum sulfate ($Al_2(SO_4)_3$) or polyaluminum chloride (PACl) are commonly used in drinking water treatment to enhance the removal of particulate, colloidal, and dissolved substances during the coagulation process [2–5]. Literature data indicate that the use of any Al-based coagulant may either increase or decrease the Al concentration in the treated water, however, especially when aluminum sulfate is applied, the total Al concentration increases after treatment [1–3]. The presence of Al in drinking water is undesirable as it is related to possible health effects, and especially its connection with Alzheimer's disease or dialysis encephalopathy, are of the highest concern [9]. Since the 1980s it has been known that aluminum ions present in water, by the penetration of ions through the blood-brain barrier, cause malfunction of the nervous system. According to the toxicity level, aluminum aqua-complexes can be arranged in the following series: $[Al(OH)_2^+] < [Al(OH)^{2+}] < [Al(H_2O)_6]^{3+}$ [9–12].

Aluminum may be efficiently removed from water by means of cation exchange, reverse osmosis and electrodialysis. Treatment methods like aeration/stripping, chemical oxidation/disinfection and anion exchange are ineffective for Al removal. Processes such as coagulation, sedimentation and filtration (combined) as well as lime softening are moderately effective in Al removal [13–15].

In the presented research, the use of nanofiltration and ultrafiltration preceded with coagulation was investigated to remove aluminum from natural water (preliminarily alkalized). The effect of ion concentration and coagulant dose on the process's efficiency was established. The best process configuration based on aluminum removal efficiency and membrane affinity to fouling was indicated.

2. Experimental

2.1. Membrane filtration

The experiments of membrane filtration, including both nanofiltration and ultrafiltration, were carried out in the laboratory scale installation KMS Cell CF 1 (by Koch Membrane Systems) (Fig. 2), operated in a cross-flow mode. The installation was equipped with the feed tank enabling the use of feed volume of up to 0.5 L and a flat-sheet membrane cell suitable for the installation of membranes with an effective separation area of 28 cm^2. The permeate was continuously collected outside the membrane module, while the retentate was recirculated to the feed tank.

Membrane filtration experiments were performed using different types of polymeric membranes (NF and UF) supplied by Synder. Their characteristic, according to the product's data, is shown in Table 1.

In the research, NDX nanofiltration membranes were used for the direct removal of aluminum from alkalized natural water, while SM ultrafiltration membranes were used as a polishing stage after coagulation. For experimental purposes, small membranes samples (13 samples in the case of NDX (M-M13) and 4 samples in the case of SM (SM1-SM4)) were cut from one membrane sheet delivered by the supplier. NDX filtration was carried out at 2.0 MPa pressure, while SM polishing at 0.3 MPa. All processes were performed until 80% of feed (of 0.5 L volume) was recovered in the form of permeate (i.e., 0.4 L volume). The volumetric permeate flux across the membrane was calculated based on the measured volume of collected permeate according to Eq. (1):

$$J = \frac{\Delta V}{A \cdot \Delta t} \tag{1}$$

where ΔV is the permeate volume collected over Δt period, L; A is the membrane effective separation area, m^2; Δt is the time of permeation and sample collection, h.

Relative fluxes of permeates obtained during filtration of alkalized natural/coagulated water and of deionized water after the treatment process were calculated according to Eqs. (2) and (3):

$$\alpha_P = \frac{J_P}{J_0} \tag{2}$$

$$\alpha_D = \frac{J_D}{J_0} \tag{3}$$

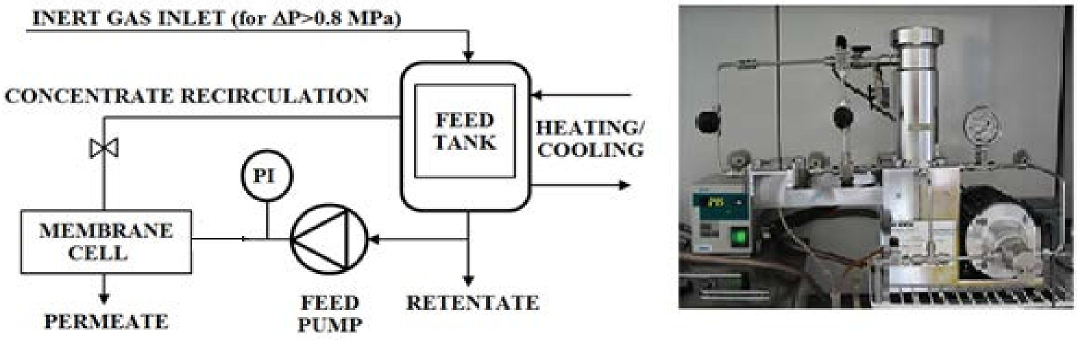

Fig. 2. KMS Cell CF1 lab-scale membrane filtration unit.

Table 1
Characteristic of applied membranes according to producer's data [16]

		NDX	SM
Membrane type		NF	UF
Membrane polymer		PA TFC	PES
Molecular cut-off, Da		500–700	20,000
Pore size*, nm		2.5–3	100
Flux, L/m^2/h		59.5–76.5	n/d
Average retention, %	$MgSO_4$	95	
	NaCl	40	
	Lactose	90	

*Note: pore size was estimated on the basis of the dependence that 1 nm = 200 Da [17].

where J_P is the volumetric flux of natural/coagulated water; L/m^2/h; J_D is the volumetric flux of deionized water after real sample filtration, L/m^2/h; J_0 is the initial volumetric flux of deionized water, L/m^2/h, α_P is the relative permeate flux, –; α_D is the relative deionized water flux.

2.2. Coagulation

The research on coagulation was carried out using FLOKOR 1.2A commercial coagulant. It is a polyaluminum based coagulant, which contains aqueous complex solution of aluminum hydroxide chloride of chemical formula $Al_m(OH)_{3m-1}Cl_xH_2O$. The coagulant pH is ca. 3.5–4, while the content of aluminum is 10–11 wt.%. The basicity of FLOKOR 1.2 A reaches up to 80 wt.%. The coagulant was dosed to raw water in doses ranging from 10 to 40 mg Al/L (noted as FL10, FL20, FL30 and FL40). After the coagulant was dosed to the water, fast mixing (2 min, 200 rpm) followed by slow mixing (30 min, 50 rpm) and sedimentation (60 min) was applied. The supernatant was separated and directed to ultrafiltration with the use of SM membranes.

2.3. Natural (feed) water characteristic

Natural surface water contaminated with aluminum was sampled in one of the Polish cities localized in the Lower Silesia region. 10 samples of water were collected within 5 months period in order to establish the stability of water composition in time, which is also related to the impact of season condition. The characteristic of sampled water is presented in Table 2.

The analyses of natural water showed, that it characterized with relatively stable composition, very low mineralization (conductivity in the range of 30–45 µS/cm), the low summary concentration of cations <7 mg/L and anions <11 mg/L, slightly acidic pH (4.2–5) and relatively low, but exceeding the permissible level (0.2 mg Al/L) concentration of aluminum (0.283–0.716 mg Al/L, except for S5 – 0.16 mg Al/L).

2.4. Preliminary tests

S1–S3 waters were used in preliminary tests to determine the best conditions for nanofiltration (feed water pH) and coagulation (and their treatment is not discussed in this paper). In the case of NF, it was found, that the feed water pH had a negligible impact on contaminant rejection, while alkaline pH was preferable due to membrane supplier recommendations. Hence, before the nanofiltration process, feed waters were alkalized to pH in the range of 7.5–8.0 using a 0.5 M solution of NaOH (analytical grade). Additionally, two simulated waters of elevated aluminum content (2 and 5 mg Al/L) were prepared on the natural water matrix (with S10 sample) using aluminum chloride hexahydrate $AlCl_3x6H_2O$, in order to check the impact of the contaminant concentration on nanofiltration process performance. In the case of ultrafiltration, the preliminary research eliminated the possibility of its direct use for as delivered/alkalized water treatment due to the poor contaminant rejection (even the use of UF membranes of smaller cut of 1–3 kDa did not allow to obtain the desired treatment effect neither for as delivered nor alkalized water). Hence, it was decided to apply aluminum-based coagulation before ultrafiltration to enhance the treatment effect (the details of coagulation process parameters are discussed in chapter 2.2.). During the examination of coagulation process conditions, it was found that natural water samples need to be alkalized to pH ca.9.0 using 0.5 M solution of NaOH (analytical grade) prior to coagulation. The alkalization had to be performed in order to assure proper flocculation conditions (i.e., after coagulant addition, the pH of the solution was to be kept in the range of 7.5–8).

Table 2
Characteristics of natural water used in the research

Water sample No.	pH	Conductivity	Al^{3+}	Na^{+}	Ca^{2+}	K^{+}	Cl^{-}	SO_4^{2-}	NO_3^{-}
	–	μS/cm	mg/L						
S1	4.85	33.85	0.716	2.28	1.81	0.30	0.88	5.17	1.67
S2	4.96	40.97	0.367	3.52	1.56	0.28	1.05	7.64	1.54
S3	4.62	39.71	0.608	2.87	2.64	0.25	1.03	6.65	1.52
S4	4.69	42.02	0.324	2.76	2.97	0.24	1.18	7.88	1.67
S5	4.95	49.19	0.160	2.93	3.92	0.26	0.91	9.67	1.97
S6	4.38	44.96	0.283	2.58	2.50	0.26	1.14	7.81	1.63
S7	4.62	43.40	0.276	2.62	2.75	0.25	1.09	7.62	2.00
S8	4.28	42.40	0.345	2.55	2.72	0.24	0.89	8.11	2.26
S9	4.33	42.89	0.454	2.49	2.85	0.28	1.06	6.9	4.33
S10	4.91	43.70	0.392	1.95	2.54	0.3	1.76	7.09	2.03

2.5. Analytical methods

Feed water and permeates were characterized according to pH, conductivity and aluminum content. pH and conductivity were measured with the use of CP-505 pH-meter and CC-505 conductometer, both by Elmetron equipped with dedicated probes, that is, IJ44C pH electrode by Ionode and ECF-1 conductivity electrode by Elmetron. Aluminum concentration was established with the use of the inductively coupled plasma optical emission spectrometry (ICP-OES) method using iCAP-6500 duo by Thermo Scientific.

3. Results

3.1. Membrane characteristics

For the purpose of the research, membranes were characterized by measuring deionized water flux at pressure ranges as follows: 1.0–3.0 MPa for NDX and 0.15 to 0.6 MPa for SM membranes. The obtained results are shown in Fig. 3.

The determined characteristics indicated the typical linear behavior of all examined membrane types (the linear increase of deionized water flux with transmembrane pressure increase). The established average unit permeabilities for NDX and SM membranes were 51.7 and 931 L/m^2/h/MPa, respectively.

3.2. Natural/coagulated water filtration

The run of processes of alkalized natural water nanofiltration at TMP 2.0 MPa is shown in Fig. 4, while in Fig. 5 the run of ultrafiltration of coagulated water is presented.

The results presented in Fig. 4 indicate a slight impact of treated water on membrane performance. Periodical permeates declines could be observed, which was related to deposition of substances on membranes surface followed by their washing with feed water supplied. The observed difference in process duration was mainly the result of the use of small membrane modules (pieces of membranes cut from one membrane sheet) of small separation surface area (28 cm^2).

The results presented in Fig. 5 indicate that the ultrafiltration of supernatant after coagulation with coagulant doses 10 and 20 mg Al/L (FL10 and FL20, respectively) was very stable, that is, every next measured value of the flux was not different than the following measured value by more than 10%, while most of the time it was below 5%. In the case of FL30, the slight, proceeding increase of permeate flux was observed, which could be related either with the washing of contaminants deposited on the membrane surface by feed water supplied to the module or change of membrane surface parameters (hydrophilicity) related with contaminants deposition. The former, however, was more probable, as such the behavior was not observed in the case of other coagulant doses (FL10 and FL20). On the other hand, in the case of the highest coagulant dose, the continuous flux decline was observed what indicated an accumulation of contaminants (most probably aluminum flocks remaining in the supernatant) on the membrane surface and inside membrane pores.

In order to assess the actual impact of treated water on membranes' transportation properties (both ultra- and nanofiltration ones) relative permeate and deionized water fluxes after the process was calculated (according to Eqs. (2) and (3)), and the obtained results are discussed in chapter 3.3.

3.3. Fouling affinity

In Fig. 6 the comparison of relative permeate and relative deionized water fluxes after the process of nanofiltration of natural water is presented, while in Fig. 7 the parameters obtained for ultrafiltration of coagulated natural water are shown.

The data presented in Fig. 6 indicates the low affinity of the used membrane (NDX) to fouling, as measured relative permeate and deionized water streams are always above 0.8 and vary from 0.89 to 1.07 for permeate and 0.82 to 1.12 for deionized water. Moreover, as α_P was usually larger than α_D, this may suggest that the alkaline character of the treated stream is favored by the membrane materials and it allows to improve transportation properties of the membrane. However, during nanofiltration of alkalized natural water deposition of some compounds present in the feed stream of membrane surface was observed, which is shown

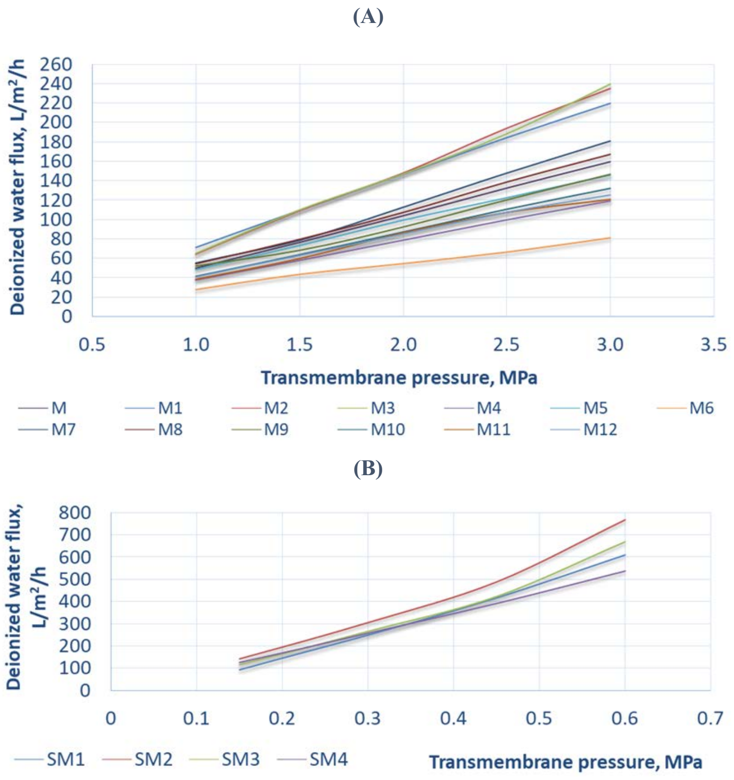

Fig. 3. The dependence of deionized water flux on transmembrane pressure determined for pristine membranes: (A) nanofiltration NDX membranes (Mno states for membrane sample no) and (B) ultrafiltration SM membranes (SMno states for membrane sample no).

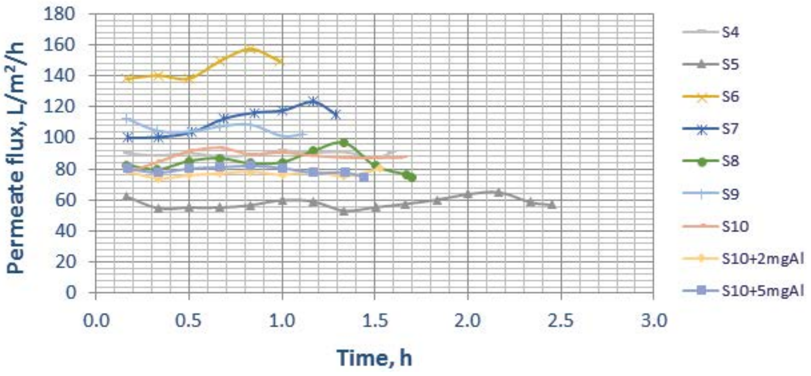

Fig. 4. Permeate flux during natural water nanofiltration at NDX membrane at 2.0 MPa pressure.

in Fig. 8. Thus, the stable, long-term operation of the system has to be confirmed during further pilot-scale research with the actual membrane module involved, and at this stage of the research, the severeness of fouling cannot be directly defined.

The data presented in Fig. 7 indicates that ultrafiltration of coagulated water treated with coagulant doses 10–30 mg Al/L slightly influenced SM membrane's transportation properties. Relative permeate fluxes established for these processes were in the range from 0.96 to 0.99, while relative deionized water fluxes varied from 0.89 to 0.98. Oppositely in the case of ultrafiltration of natural water pretreated with the highest coagulant dose, that is, 40 mg Al/L (FL40), where the relative permeate flux was only 0.8, while relative deionized water flux was 0.58. This phenomenon was most probably caused by the presence of small flocks present in supernatant after coagulation, which were not effectively separated during the sedimentation stage after coagulation. Thus, the combination of ultrafiltration with coagulation process carried out with the coagulant dose >30 mg Al/L is not recommended. On the other hand, the deposition of any colored layer was not observed for SM membranes, while the gel-like, transparent film could be noticed in the case of the membrane after treatment of FL40 supernatant.

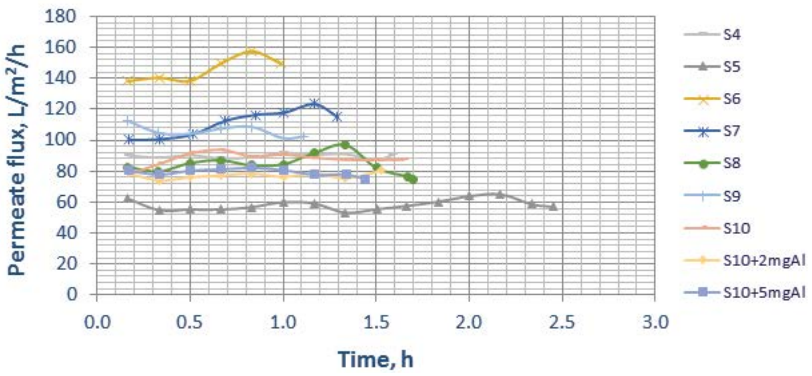

Fig. 5. Permeate flux during coagulated natural water ultrafiltration at SM membrane at 0.3 MPa pressure.

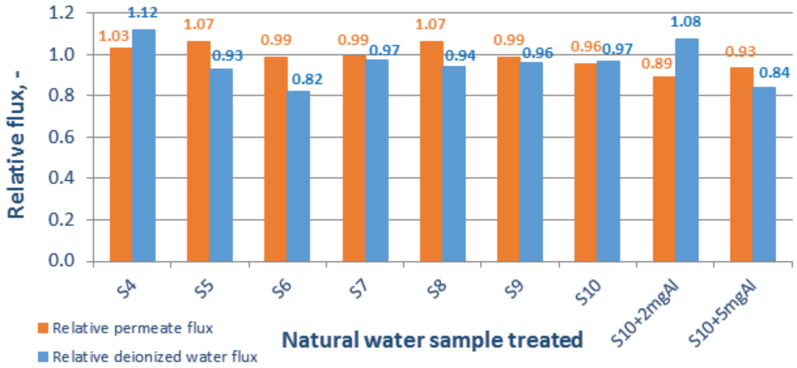

Fig. 6. The comparison of relative permeate and relative deionized water fluxes after the process of nanofiltration of alkalized natural water (for pristine membrane $\alpha_{P,D} = 1$).

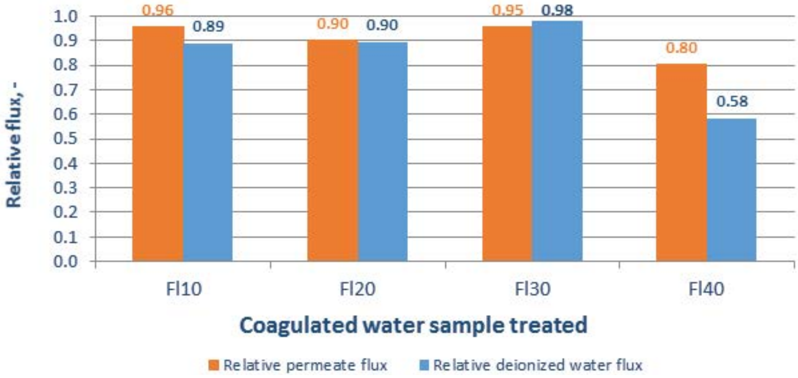

Fig. 7. The comparison of relative permeate and relative deionized water fluxes after the process of ultrafiltration of coagulated natural water.

3.4. Quality of permeate

The efficiency of both treatment processes was evaluated in regard to the removal of the target contaminant, that is, aluminum. In Tables 3 and 4, the concentration of aluminum in feed waters and permeates obtained during nanofiltration and ultrafiltration processes, respectively, together with retention rates, are presented.

The parameters of permeates obtained during nanofiltration of alkalized natural waters indicated on very high removal efficiency of aluminum (retention rate in the range from 91.3% to 99.6%) regardless of its initial concentration in the feed water (from 0.16 to 5.24 mg Al/L). The final concentration of the contaminant was always below the established quality standard for potable water, that is, 0.2 mg Al/L. Considering membrane capacity, fouling affinity and permeates quality it can be concluded that nanofiltration accompanied with pH adjustment may be the single-stage operation used to remove trace aluminum from natural waters.

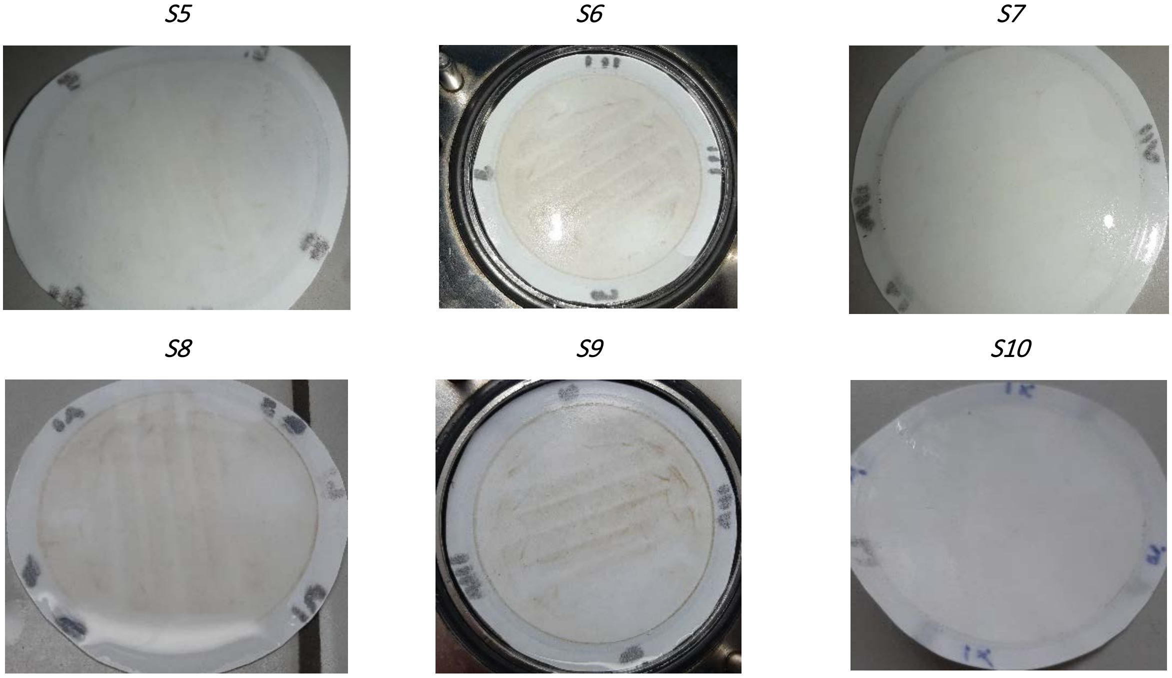
S5 S6 S7
S8 S9 S10

Fig. 8. Photos of deposits present on nanofiltration membranes surface after natural water treatment.

Table 3
Concentration of aluminum in feed waters and permeates obtained during nanofiltration of alkalized natural waters

Natural water sample	Al concentration, mg/L		Al retention, %
	Feed	Permeate	
S4	0.324	0.019	94.1
S5	0.160	0.014	91.3
S6	0.283	<0.005*	98.2
S7	0.276	<0.005*	98.2
S8	0.345	<0.005*	98.6
S9	0.454	<0.005*	98.9
S10	0.392	0.0059	98.5
S10 + 2 mgAl	2.390	0.020	99.2
S10 + 5 mgAl	5.240	0.023	99.6

*For retention rate calculation, the value of lowest detectable concentration (Limit of detection (LOD) – 0.005 mg Al/L) was taken.

Table 4
Concentration of aluminum in feed waters and permeates obtained during ultrafiltrations of coagulated natural waters

Natural water sample	Al concentration, mg/L		Al retention, %
	Feed	Permeate	
FL10	1.81	<0.005*	99.7
FL20	1.62	0.041	97.5
FL30	2.10	0.021	99.0
FL40	17.6	0.018	99.9

*For retention rate calculation, the value of the lowest detectable concentration (Limit of Detection (LOD) – 0.005 mg Al/L) was taken.

The results presented in Table 4 indicate the possible use of alkalization with NaOH and coagulation/sedimentation followed by ultrafiltration for removal of aluminum from water. Due to coagulation, aluminum flocks of a size suitable to be rejected by applied UF membranes by means of sieving effect were formed and sufficient Al retention was observed. The final concentration of the contaminant was always below the permissible value established in the Regulation of the Ministry of Health on quality of water dedicated to potable purposes (0.2 mg Al/L). Such the treatment configuration also allowed for the use of the lowest, among examined, coagulant dose, that is, 10 mg Al/L (while research with lower coagulant doses is to be continued). However, the impact of a trace of coagulation flocks on membrane transportation properties during a long-term process carried out with the use of the final membrane module has to be investigated, in order to confirm the final usability of the process.

4. Conclusions

Two types of membrane processes, that is, nanofiltration using Synder NDX membrane and ultrafiltration combined with coagulation using Synder SM 20 kDa membrane were used to treat natural water contaminated with aluminum. The obtained results allow us to conclude that:

- The usability of nanofiltration to efficient aluminum removal was confirmed and the final content of the contaminant in produced permeates was always below the quality standard established in national regulation on potable water quality (i.e., <0.2 mg/L). The performance of the process at TMP = 2.0 MPa allowed for high process capacity and the average permeate flux was 88 L/m^2/h. It was also observed, that the initial concentration of Al in natural water (ranging from 0.16 to 5.24 mg Al/L) had a negligible impact on both, retention rate and membrane capacity. Nevertheless, the deposition of contaminants layer on the membrane surface was observed after the process, but the influence of this layer on membranes transportation properties was insignificant. However, the impact of this phenomenon on final nanofiltration process performance has to be confirmed during long-term research with the target membrane module.

- The use of alkalization with NaOH, coagulation followed by sedimentation and ultrafiltration also may be considered as an efficient method of removal of aluminum from natural water. Due to membrane capacity, the process should be carried out with a coagulant dose below 30 mg Al/L (in the case examined within the research Flokor 1.2 A coagulant). It was shown, that coagulant dose 10 mg Al/L was sufficient enough to obtain the desired treatment effect, that is, to obtain filtrate of aluminum concentration corresponding to potable water standard. The advantage of such the configuration of the process is high capacity at low TMP, which at this research was kept on the level of 0.3 MPa. However, the impact of a trace of coagulation flocks on membrane transportation properties during a long-term process carried out with the use of the final membrane module has to be investigated, in order to confirm the final usability of the process. Moreover, the research with lower coagulant doses is to be continued.

To sum up, both investigated techniques/systems allow to efficiently remove aluminum from natural water. The choice between direct nanofiltration or coagulation combined with ultrafiltration should be made on the basis of raw water parameters and the appearance of accompanying pollutants, which were not present in water tested within this research (e.g., natural organic matter, NOM). In such a case, the combined system seems to be a better solution. On the other hand, treatment of water of good quality, as one investigated in this case, can be efficiently performed by means of direct nanofiltration. The process characterized with lower permeate flux at higher TMP, while, in opposite to combined system (CS), its performance was related to the formation of only one waste stream (i.e., retentate, while in the case of CS, the concentrated streams after sedimentation and ultrafiltration were formed), which additionally, due to the lack of use of external chemicals could be directly discharged to the environment (whereas post-CS streams need to be deposited either to the sewage system or treated prior their discharge to the environment).

Symbols

ΔV — Permeate volume collected over Δt period, L
A — Membrane effective separation area, m^2
Δt — Time of permeation and sample collection, h
J_P — Volumetric permeate flux, L/m^2/h
J_D — Volumetric flux of deionized water after real sample filtration, L/m^2/h
J_0 — Initial volumetric flux of deionized water, L/m^2/h
CS — Combined system
TMP — Transmembrane pressure, MPa
α_P — Relative permeate flux
α_D — Relative deionized water flux

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