



Evaluation of the effect of body fluid analogs on the parameters of nanofiltration during the purification of swimming pool water

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Abstract

This study presents the possible application of membrane nanofiltration for purifying swimming pool water by removing compounds from human body secretions. The effect of the matrix (swimming pool, demineralized, tap water) on the analyzed pollutants was evaluated. We used asymmetric flat membranes (polyamide-thin film composite) in a dead-end filtration system. The effect of the matrix on the transport properties of the membranes was noticeable. A maximum reduction in value of permeate flux was obtained for 15.6 μL fluid analog solution/L of pool water. For all the analyzed matrices, a distinct reduction in the value of relative permeate flux was obtained along with an increase in the content of the investigated compounds in the feed. There was a marked increase in the separation properties along with an increase in the pollutants. The average value of the retention coefficient for total organic carbon was 87.21% and 79.67% for the concentrations of 3.90 and 15.60 μL fluid analog solution/L of pool water, respectively. This study provides evidence that further research into highly effective solutions for purifying pool water would be beneficial.

Keywords Water treatment · Closed circuit · Total organic carbon · Pressure-driven membrane processes · Secretions of the human body

1 Introduction

Every public swimming pool facility must have its own water treatment circuit to maintain the required physico-chemical and microbiological standards of water [1–4]. Such circuits are an example of closed-loop circuits where water is treated and disinfected continuously during the operation of the facility. Disinfection of water is very important in the prevention of growth of pathogens. The most common source of water supply to the swimming pools is water mains, and rarely, it is deep well intakes. The supplied water contains organic and inorganic compounds, which are naturally present, in addition to the disinfection by-products (DBPs) that are generated during the course of its treatment [5]. Additional pollutants may enter the water mains during the course of its distribution to various pool facilities [6]. However, the primary factors

that significantly reduce the quality of water in basins are organic micropollutants—introduced by the users of the pool (e.g., residues of cosmetic products)—and products of human metabolism (e.g., saliva, urine, sweat, and epidermis) [7, 8]. More than 600 different compounds have been identified which belong to the class of DBPs, which cause harmful effects on the human body [9, 10].

Filtration on filtration beds is a common technique of water treatment across various swimming pool facilities. It is usually accompanied by the process of contact coagulation. However, the effectiveness of coagulation in the removal of micropollutants (especially the small-particle micropollutants) is low, on a level of 10–20% [11, 12]. In addition, water treatment systems in many pool facilities still use single-layer sand filtration beds that do not ensure complete elimination of pathogens [13]. Improvement of the effectiveness of the filtration beds may be achieved

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by using an additional sorption layer of activated carbon. Depending on the type of micropollutant, the removal efficiency can be at a level of 20–85% [13, 14].

The water quality in a swimming pool is specifically affected by various factors, including the quality of the water supplied to the circuit, the load of the facility (the number of swimmers), the treatment technology (including the type of the filtration bed), and the type of facility (indoor or outdoor pool). To maintain the required microbiological and physicochemical standards, it is necessary not only to use an appropriate water treatment technology but also to disinfect it. Chemical disinfecting agents reduce the growth of pathogens, such as *Escherichia coli* and *Legionella sp.*, in swimming pool water (POOLW) [15, 16], and they also react with various pollutants present in the water. The presence of DBPs in POOLW has become a more common problem that has been broadly studied [7, 17, 18]. Due to the progress in instrumentation technology in the last few decades, several DBPs in POOLW have been identified [19, 20]. The common presence of trihalogenomethanes (THMs), halogen-acetic acids (HAAs), halogen-acetonitrile (HAN), chloramines (CAM), and many other chemical compounds demonstrates that a long and/or systematic contact with water and air in POOLW may result in serious consequences to the health of the users of such facilities [10, 20, 21].

Mains water in swimming pool circuits contains not only natural organic matter (NOM) but also chlorides, bromates, and DBPs, including chloramines and bromides [5, 7]. It is also used to make up for losses due to splashing and absorption of water by the swimwear. A common practice among pool staff is to dilute water in the circuit if the concentration of combined chlorine or chloroform exceeds the standard value.

However, natural products of human metabolism are known to be the largest contributors of DBPs. It is estimated that a single swimmer during an hour of physical activity excretes 50 mL of urine and 200 mL of sweat resulting in an increase in the content of nitrogen-based organic compounds. These compounds react easily with chlorine-based disinfectants [22–25]. The concentration and composition of DBP precursors classified as body fluid analogs (BFAs) are strongly correlated with the number of persons using the pool (this value is usually given as the number of persons per hour or per surface (in meter square), their hygiene habits, and the intended use of the pool (swimming, children's recreational, jacuzzi, etc.)). However, previous studies have confirmed the presence of urea, ammonia, uric acid, creatinine, creatine, lactic acid, citric acid, hippuric acid, uracil, ornithine, chlorides, sulfates, cations (K^+ , Na^+ , Ca^{2+} , Mg^{2+} , and Zn^{2+}), and amino acids (histidine, glycine, cysteine, asparagine, lysine, arginine, and guanine) in POOLW [7, 23]. It has been confirmed that the reactivity

of human body fluids with chlorine is greater than that of NOM with chlorine. A mixture of BFA forms more chloroacetic acid (e.g., HAA) than trichloromethanes (e.g., THM). Citric acid increases the formation of THM and HAA [7, 17]. Another DBP precursor is creatinine, which forms stable chloramines identified as dichloramines [24]. The presence of amino acids contributes to the formation of not only chloramines but also cyanogen chlorides. Trichloramines are also formed in the presence of creatinine, urea, histidine, and arginine [7]. Urine also contains active components and metabolic products of various medications such as painkillers and antibiotics, which along with the residues of cosmetics might enter the water [20, 23].

Even though many DBPs are present at concentrations below 1 mg/L in POOLW [8], their presence is still harmful to humans. It has been confirmed that micropollutants in the form of DBPs produce various enzymes that cause oxidative stress in the cells of living organisms [9]. Previous studies have confirmed the genotoxic, cytotoxic, and mutagenic properties of DBPs present in POOLW [8–10, 26]. High rates of genotoxicity have been associated with DBPs with low molar mass [10]. A majority of DBPs has a molar mass below 1000 g/mol and as much as 30% of them are those with a molar mass below 200 g/mol [25, 26]. DBPs cause irritation of the mucous membrane when the person is in the POOLW and prolonged contact with DBPs may cause various health issues such as asthma, bladder cancer, and atopic dermatitis [20, 26, 27]. Thus, the benefits of swimming can be achieved only when the negative effect of toxic substances has been reduced. Therefore, it is necessary to look for effective ways to treat POOLW, which will make it possible to eliminate both precursors and the products of their reactions with DBPs.

Continuous monitoring of water quality is immensely important to the operators of the pool facility because they are aware of their responsibility for human health and of the legal consequences that they will face if they fail to perform their duties. In addition to an extended physicochemical (total concentration of trihalomethanes, chloroform, nitrates, and oxidizability) and microbiological analysis, continuous measurements, which are usually performed on a daily basis, include measurements of pH, redox potential, free chlorine, bound chlorine, and turbidity. This analysis is necessary to evaluate the quality of POOLW; however, they do not present a complete picture of the presence of DBPs. Among the parameters that describe anthropogenic pollution, usually the most important are believed to be the concentration of total organic carbon (TOC), total nitrogen (TN), ultraviolet absorbance at a wavelength of 254 nm (UV_{254}), and turbidity [10]. A particularly important parameter that indicates the presence of body fluids in POOLW is the concentration of dissolved organic carbon (DOC) [5]. The concentration of DOC

is related to the number of people bathing and the formation of DBP; consequently, monitoring and reducing the concentration of DOC are very important to maintain the quality of POOLW [5].

Along with the quality of POOLW, it is important to analyze the correlation between the selected parameters of quality. For example, the presence of trihalomethanes and heterocyclic aromatic amines is strongly correlated with the content of TOC in POOLW but is only weakly correlated to free chlorine [28–30]. Moreover, in the case of analysis of chemicals related to body fluids, measurement of TOC is particularly important because it indicates the potential for the formation of trihalomethanes. Although, as a standard, UV_{254} is a frequently analyzed parameter and the quality of disinfected water is assessed based on this value, till date, no correlation has been identified between the concentration of DBPs and the values of UV_{254} .

UV_{254} is a measure of aromaticity and may be an indicator between the potential of formation of THM and HAA; however, it is not a direct determinant of the concentration of DBPs [28, 29]. In addition to the commonly used indicators of water quality, it is also recommended to measure the content of total inorganic salts (TDS), inorganic nitrogen (ammonium), and organic nitrogen [30].

Despite very detailed descriptions of the problem of pollution with human body fluids in swimming pool water given in multiple publications worldwide, the water treatment technologies continue to concentrate on bed filtration (filtration with contact coagulation). Departure from use of single-layer sand beds to use of multilayer beds or zeolite fillings in swimming pool facilities has certainly significantly improved the quality of water in the circuits. However, upgrade processes are performed most of all in newly built facilities. On the other hand, the technological progress achieved in water filtration still does not solve the problem of fine-particle pollution in the circuits (water is changed once a year), which are not removed in the course of filtration processes [11–14]. Another issue that, in the opinion of the applicant, should be brought attention to, in the context of a change of swimming pool water treatment technology, is the high water consumption (and wastewater production) in swimming facilities. Because in the filtration process pollutants are gathered in filtration beds, which leads to gradual pressure loss, in order to meet the physicochemical and sanitary requirements concerning water quality, the filtration bed flushing process must be performed once every 2–3 days. In order to perform the filtration bed flushing process, 4–6 m³ of water (taken from an overflow tank) is required for every m² of the filtration bed [4]. This process generates a large volume of wastewater, which is drained to the sewer system.

An alternative to classical treatment systems is pressure membrane systems that are highly effective in the

removal of suspended solids, microorganisms, DBPs, and micropollutants [12, 31, 32]. Pressure membrane systems must be optimized for the operating parameters of the process if it has to be used for the treatment of POOLW, in addition to the observation and description of the phenomena that hinder their practical use in actual systems.

Thus, the reason for the experimental and computational works conducted is an evident lack of studies, that would describe the properties related to the transport and separation properties of substances associated with components of body fluids in unit nanofiltration (NF) processes and integrated systems, with the use of polymer membranes. Individual studies on the use of pressure membrane processes have focused on the total stream of pollutants [31–39] and have not analyzed a specific problem that may constitute an obstacle to the use of membrane systems in the future.

Pressure membrane systems have an advantage of the high degree of freedom when it comes to selecting the nature of the process and the operating parameters. Micro- and ultrafiltration processes focus on microparticles and macroparticles, which are usually inorganic particles, organic colloids, microbes, and dissolved organic substances [36–41]. Just the inclusion of pressure membrane processes in hybrid or integrated systems guarantees a high physicochemical and microbiological quality of the permeate, which enables its reuse [34, 35]. However, NF results in a high rate of removal of NOM and THM precursors [39]. The effectiveness of the removal of TOC and THM is strongly dependent on the membrane used [34].

Effective removal of TOC and DOC is by membranes with a low limit of cutoff, not exceeding 1000 Da [21]. Moreover, NF membranes are more resistant to chlorine than that of reverse osmosis membranes and are useful in the removal of fine particles [39]. Removal of DBPs can be monitored by measuring the concentration of adsorbable organic halides (AOX), THM, and DOC [39, 40]. However, what makes their use difficult is a quick reduction in the transport properties of NF membranes due to the blockage of pores by organic compounds. Studies using model substances concern mostly wastewater that contributes to fouling (blocking of membrane pores as a result of reversible or irreversible sorption processes). The substances that have been frequently reported to cause fouling are humic acid, sodium alginate, and bovine serum albumin [42, 43]. It must be emphasized that the hydrophilic protein fraction also constitutes an important factor that blocks membranes [42]. An increase in the concentration of bivalent cations, a reduction in the pH of the solution, and an increase in the concentration of electrolytes significantly contribute to the contamination of membranes as a result of changes in the intermolecular electrostatic actions [44,

45]. Given the presence of cations in BFA solutions, one can expect a reduction in the efficiency of membranes.

The efficiency (i.e., transport properties) of membranes is defined by the volume flux of permeate, which indicates the presence of feed material flowing through a unit of surface area of the membrane in a unit of time [46]. However, the separation properties are defined by the retention coefficient, i.e., the extent to which the substance present in the feed solution is stopped [47]. The inseparable element in the observation of membranes structures are nanoscopic analysis paths of their degradation and blocking effects. Ex situ methods such as scanning electron microscopy (SEM), atomic force microscopy (AFM), or spectroscopic ellipsometry allow for the in-depth supplementing knowledge on membrane-forming properties of the materials [48–52]. Information on the microstructure of the examined membranes, the possibility to create elemental maps, and pores' distribution topography are some of the selected applications of the modern imaging methods of fouling.

The consequences of fouling result in higher operating costs due to a reduction in the efficiency of membranes. Moreover, frequent cleaning of membranes reduces their separation properties and leads to the shorter service life of the epidermal layer, which is very important in the process of NF performed by using asymmetric membranes [42]. Identification of the polluting substances makes it possible to reduce or avoid the fouling of membranes. Therefore, the matrix in which those pollutants exist (e.g., deionized water, mains water, or POOLW) must also be analyzed to prevent fouling. Worldwide and in Poland, the effect of matrix on the transport and separation properties of pressure membranes has been studied [53–56]. However, to the best of our knowledge, no authors have analyzed specific compounds from the BFA class in different matrices with reference to the properties of membrane materials. Polymer membranes demonstrate, among others, sensitivity to chlorine [50, 51]. As a result, it is important to conduct a comparative analysis of the effect of chlorine and other oxidants on the membrane-forming material and its susceptibility to biological decomposition and hydrolysis [57, 58]. This will make it possible to determine the chemical cleaning (e.g., with sodium hypochlorite) of the studied material, e.g., with sodium hypochlorite [28]. Given the susceptibility of nitrogen compounds in the formation of DBPs, membranes can be cleaned with citric acid, which is a compound that does not contain such class of chemicals.

Therefore, in this study, we aimed to analyze the transport and separation properties of the commercial NF membrane. In addition, we tested the possibility of using this process to remove impurities from the group of substances analogous to the secretions of the human body.

The analysis was conducted, among others, based on the changes in the volume of demineralized water (DEMW) flux, permeate, and rinsing water. Furthermore, the values of removing impurities, based on selected physicochemical parameters studied the effect of the matrix (POOLW, DEMW, and tap water (TAPW)) on compounds from the BFA solution.

2 Experimental procedures

2.1 Analyzes of physicochemical parameters

Total carbon (TC), TOC, and DOC in POOLW samples, model solutions, feeds, and permeates (after 0.45 μm filtration, PVDF syringe filter) were measured using a TOC-L series analyzer by catalytic oxidation combustion at 680 $^{\circ}\text{C}$ (Shimadzu). The UV_{254} values were measured using the UV VIS Cecil 1000 from Analytik Jena AG, with an optical path length of the cuvette equal to 1 cm. The UV_{254} value was determined based on the measurement method presented by the United States Environmental Protection Agency [59], and the final result of the analysis is presented as per meter. Turbidity was measured using EUTECH Instruments model Turbidimeter TN-100. Measurement of TN, total and free chlorine concentration in POOLW samples was made by the photometric method in cuvette tests using a VIS Spectroquant[®] Pharo 300 UV spectrophotometer (Merck). The pH and redox potential in POOLW samples, model solutions, and feeds were measured with a multi-parameter inoLab[®] 740 m (WTW, Measuring and Analytical Technical Equipment).

2.2 Data regarding the pool facility

Water samples were obtained from a swimming pool located in a primary school. The pool is constantly loaded during sports activities (from 10 to 25 people per hour). Modernization of the water treatment cycle, covering the basin, and fitting the elements in the facility took place in 2016. The water is continuously treated in a closed circuit, and its losses are supplemented up with TAPW. The facility operates during the school year. During the holidays, there is a technological break, during which the elements in the circulation are cleaned, and the water is wholly exchanged with fresh TAPW. Purified water flows through the overflow gutters and is transported to the overflow tank via pipelines. The water is treated by the process of contact coagulation (aluminum coagulant) and pressure filtration (multilayer filter: hydro-anthracite + quartz sand with different granulation). Heat exchangers heat the water. Then, the pH is automatically adjusted, and the water is disinfected with the addition of stabilized sodium hypochlorite.

Table 1 General technical and computational data for the analyzed swimming pool facility

Technical data	Value
Dimensions of the pool (m)	12.42 × 7.22
The surface of the water surface (m ²)	89.67
Pool capacity (m ³)	121.06
Personal load on the day of collection (bathers/h)	15
Bather dose (bathers/m ³ /h)	0.12
Required usable area according to DIN 19643 [4] (m ² /bather)	4.50
The length of the overflow gutter (m)	39.28
Circulation performance (m ³ /h)	30
Water displaced V _V (m ³)	1.49
Flow by waving V _W (m ³)	3.62
Total water exchange in the basin (water recirculation) (h)	4.00
The capacity of the overflow tank (m ³)	12.47
Number of filters*	2
The diameter of the filters (mm)	1250
Filter surface of a single bed (m ²)	1.23
The height of the filter bed (H, m)	1.2
The height of the gravel layer with a grain size of 3–5 mm (mm)	100
The height of the gravel layer with a granulation of 1–2 mm (mm)	100
The height of the sand layer with a grain size of 0.4–0.8 mm (mm)	400
The height of the anthracite layer with a grain size of 0.8–1.6 mm (mm)	600
Filter cycle time (day)	2
Water for rinsing filters V _R (m ³)	7.36

*They work alternately

Purified water is introduced into the basin through holes located at the bottom of the basin. The personal load on the pool at the moment of water intake was 15 people per hour. Table 1 shows the detailed technical data on the basin and circulation system.

Based on the information obtained from the technical documentation of the facility (Table 1), the total time of water overflows, which is valuable information about the exchange of water in the basin, and thus the time spent in the DBP precursors.

The selected physicochemical parameters of pool water are presented in Table 2. Significant exceedances of standards (according to the Regulation of the Polish Ministry of Health) have been demonstrated in the scope of redox potential (at least 750 mV), free chlorine (range: 0.3–0.6 mgCl₂/L), and bound chlorine (max. 0.3 mgCl₂/L) [1].

2.3 The composition of body fluids analog solution

The mixed composition of a model BFA solution has been prepared based on the literature data [7]. All chemical reagents (BFA components) were purchased as analytical grade (98–99% purity) and used without additional purification (dissolved in DEMW (pH = 9.2)). Table 3 presents the concentration of the analyzed compounds in the model solution. The analyzed doses of the model

Table 2 The values of physicochemical parameters of samples taken from the pool intended for swimming

Parameter	Value
pH	6.82
Redox potential (mV)	630
Turbidity (NTU)	0.43
UV254 (m ⁻¹)	4.70
Free chlorine (mgCl ₂ /L)	0.21
Total chlorine (mgCl ₂ /L)	1.26
Total organic carbon (mgC/L)	8.09
Dissolved organic carbon (mgC/L)	5.80
Total carbon (mgC/L)	12.73
Total nitrogen (mgN/L)	2.80

NTU nephelometric turbidity unit

solution were determined based on the German standard DIN 19643 [4] (according to which 1 bather should have an area of 4.5 m² of water surface at his disposal) and information about the total water exchange in the basin. The time of water recirculation was 1 (theoretical time of the presence of impurities in the basin) and 4 h (real time for the circulation from which the POOLW came from) (Table 3).

Table 3 Concentration of body fluid analog (BFA) components in the tested solutions

Compounds of BFA solution	CAS number	Concentration in the stock solution* (mg/L)	BFA doses ($\mu\text{L/L}$)	
			3.90	15.60
			Concentration of the component in the BFA solution (mg/L)	
Urea	57-13-6	14,800	0.0577	0.2309
Ammonium chloride	12125-02-9	2000	0.0078	0.0312
Creatinine	60-27-5	1800	0.0070	0.0281
Hippuric acid	495-69-2	1710	0.0067	0.0267
L-histidine	71-00-1	1210	0.0047	0.0189
Sodium phosphate dibasic	7558-79-4	4300	0.0168	0.0671
Citric acid	77-92-9	640	0.0025	0.0100
Uric acid	69-93-2	490	0.0019	0.0076

*25 mL of solution corresponds to the volume introduced by a single bather during an hour of physical activity

Table 4 Physicochemical parameters of BFA-enriched solutions for the nanofiltration process

Parameter	Matrix: demineralized water (DEMw)		Matrix: tap water (TAPw)		Matrix: swimming pool water (POOLw)	
	3.90 $\mu\text{L/L}$	15.60 $\mu\text{L/L}$	3.90 $\mu\text{L/L}$	15.60 $\mu\text{L/L}$	3.90 $\mu\text{L/L}$	15.60 $\mu\text{L/L}$
pH	7.25	6.86	7.45	7.10	6.80	6.72
Redox potential (mV)	480	450	360	320	260	230
Turbidity (NTU)	0.28	0.32	0.35	0.42	0.47	0.58
UV254 (m^{-1})	2.10	4.50	2.80	4.90	5.20	6.30
Free chlorine (mgCl_2/L)	0.00	0.00	0.10	0.10	0.18	0.18
Total chlorine (mgCl_2/L)	0.00	0.00	0.62	0.62	1.20	1.20
Total organic carbon (mgC/L)	0.0198	0.0842	3.26	3.32	8.29	8.62
Dissolved organic carbon (mgC/L)	0.0174	0.0799	3.09	3.16	5.96	6.02
Total carbon (mgC/L)	0.0399	0.0944	9.64	9.70	12.86	12.92
Total nitrogen (mgN/L)	>0.20	>0.20	>0.20	>0.20	2.80	2.84

NTU, nephelometric turbidity unit

2.4 Characteristics of matrices used for BFA

In addition to the analysis of POOLw, we analyzed the transport and separation properties of the NF membrane by using DEMw and TAPw, to take into account the importance of the matrices for BFA pollutants. From the prepared BFA stock solution, the designated solutions were withdrawn (3.90 $\mu\text{L/L}$ and 15.60 $\mu\text{L/L}$) and dosed to the feeds (the reaction time of the matrices with the compounds was 30 min). Then, the selected physicochemical parameters were measured. Table 4 summarizes the results of the physicochemical analysis.

2.5 Methodology of the nanofiltration process

Asymmetric DowFilmtec™ flat membranes NF90 (YMNF903001) with the cutoff molar mass distribution of

~200–400 Da were used in this study. The NF90 membrane is a full aromatic polyamide-thin film composite membrane with support layers of polysulfone and polyester, with an average pore radius of 0.34 nm, a contact angle of 63.2°, and a root mean square roughness of 142.8 ± 9.6 nm [60].

The transmembrane pressure during the filtration was maintained at 0.80 MPa. The membranes were placed in a steel filtration cell with a volume of 380 cm^3 where the active surface of the membrane was equal to 38.5 cm^2 . Before starting the process of filtration, the new membranes were conditioned by filtering DEMw to stabilize the permeate flux (J_0). Each cycle was conducted to recover 50% of the feed. The concentrated retentate was then removed from the cell. After each cycle, the membrane was rinsed with DEMw (J_R) to document the occurrence of fouling and scaling caused by the organic and inorganic

Table 5 Characteristics of membranes and operating parameters of the filtration process

Feed	The flux of demineralized water J_0 10^{-6} , $m^3/m^2 s$	The flux of permeate J_v 10^{-6} , $m^3/m^2 s$	The flux of rinsing water J_R 10^{-6} , $m^3/m^2 s$	The flux of demineralized water after cleaning J_C 10^{-6} , $m^3/m^2 s$
DEMw, 3.90 μ L/L	1.32	1.27	1.32	1.36
DEMw, 15.60 μ L/L	1.43	1.32	1.42	1.45
TAPw, 3.90 μ L/L	1.32	1.14	1.29	1.34
TAPw, 15.60 μ L/L	1.32	1.05	1.31	1.34
POOLw, 3.90 μ L/L	1.32	1.11	1.29	1.34
POOLw, 15.60 μ L/L	1.32	0.96	1.29	1.33

*The average value of the nine recorded during the process

**The average value of three rinsing processes

DEMw, demineralized water; TAPw, tap water; POOLw, swimming pool water

pollutants. Then the cell was refilled with a new portion of the feed. The treatment process was performed in three consecutive cycles without changing the membrane.

To evaluate the transport properties of the membranes, the volumetric flow rate of DEMw (J_0), permeate (J_v) (in the course of the proper filtration process), rinsing water (J_R), carried out each time after the end of the cycle, and DEMw after cleaning (J_C) were determined using the formula: $J_0(J_v, J_R, J_C) = \frac{v}{F \cdot t} \left(\frac{m^3}{m^2 \cdot s} \right)$, where v is the volume of water or permeate (m^3), F is an active surface area of the membrane (m^2), and t is the filtration time (s).

The intensity of the reduction of the hydraulic performance of the membrane was determined by determining the value of an intermediate parameter—the relative permeate/DEMw flux (relative permeability) $\alpha = \frac{J_v}{J_0} \cdot 100(\%)$. It is the quotient of the fluxes determined during the course of filtration of the treated solutions and of DEMw (J_0). This parameter is a simple measure of the disadvantageous phenomena accompanying the process of membrane filtration. Table 5 provides a detailed information on the volume of the demineralized rinsed water flux and after cleaning the membrane with a cellulose sponge. The flux of rinsing water (J_R) and after cleaning (J_C) were determined in an analogous way.

Furthermore, based on the selected parameters, we analyzed the characteristics of pore blockage occurring during the filtration process. There was an irreversible fouling ratio (IRF (%)), the flux recovery (FR (%)), and the flux recovery after cleaning the membrane (R_C (%)) with cellulose materials, which were calculated based on the following formulas: $IRF = \frac{J_R - J_C}{J_R} \cdot 100$; $FR = \frac{J_C}{J_0} \cdot 100$; $R_C = \frac{J_C}{J_R} \cdot 100$.

To determine the separation properties of the membranes, the percentage of rejection (R (%)) was determined based on the reduction of the values of pollution indicators: $R = \left(1 - \frac{c_p}{c_f} \right) \cdot 100$, where c_p and c_f are concentrations

(indicator value) of pollutants in the permeate and feed, respectively, (mgC/L or nephelometric turbidity unit (NTU) or m^{-1}).

3 Results and discussion

Efficient removal of general and DOC from POOLw with pressure membrane processes requires membranes of low threshold limit value. Therefore, it is beneficial to use NF or reverse osmosis to remove precursors and DBPs. In addition, NF has higher values of permeate flux than that of reverse osmosis [61].

NF90 membranes are highly efficient in removing DBPs [62–65]. Moreover, NF membranes have been shown to remove contamination from waters with low concentrations of DOC and specific ultraviolet absorbance (SUVA) was confirmed [60, 62, 63]. In this study, the retention coefficients were found to be diversified—high for halo ketone creatinine and low for haloacetic acids and glycine [61, 64, 65]. Moreover, NF90 membrane is classified as hydrophobic, which is good for removing hydrophilic and neutral compounds [64].

Our results show that NF is highly efficient in removing selected pollutants related to human secretions (Fig. 1 a–f). The average value of TOC retention ranged from 57.06% (DEMw, 3.90 μ L/L) to 87.56% (DEMw, 15.60 μ L/L). NF showed low efficiency in removing turbidity (Fig. 1 a, b). The value of average retention coefficients (turbidity) ranged from 52.58% (POOLw, 3.90 μ L/L) and 69.54% (POOLw, 15.60 μ L/L) (Fig. 1 e, f). NF showed the highest efficiency based on the values of UV_{254} , with an average value ranging from 67.72% (DEMw, 3.90 μ L/L) to 96.65% (POOLw, 15.60 μ L/L).

Moreover, separating properties were found to be different with regard to the applied doses of BFA. For example, in feeds with DEMw as the matrix, the average value of retention coefficient for organic carbon increased from

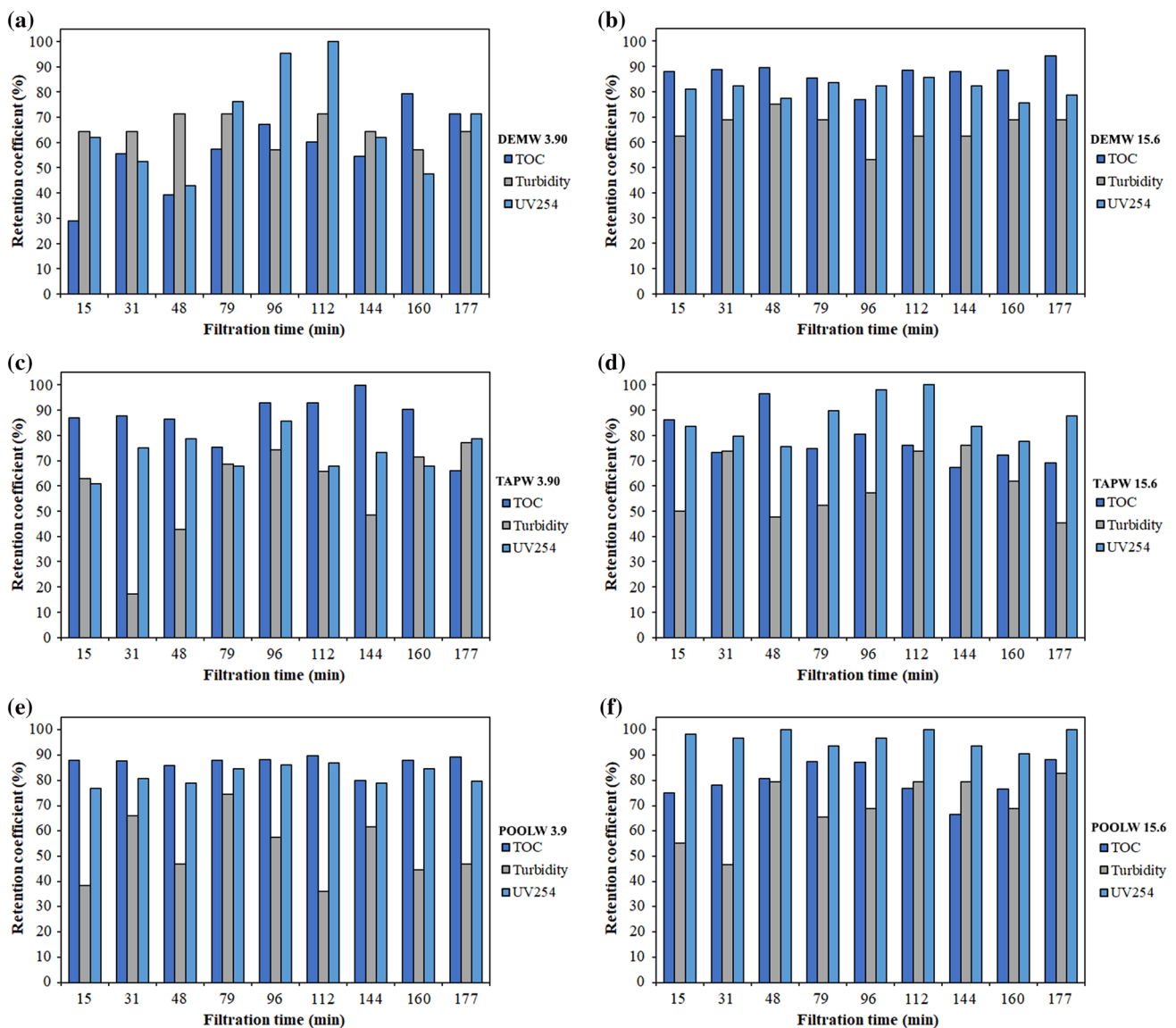


Fig. 1 Separation properties of NF90 membranes during filtration of solutions: **a** demineralized water with a BFA (3.90 $\mu\text{L/L}$); **b** demineralized water with BFA (15.60 $\mu\text{L/L}$); **c** tap water with BFA (3.90 $\mu\text{L/L}$); **d** tap water with BFA (15.60 $\mu\text{L/L}$); **e** swimming

pool water with BFA (3.90 $\mu\text{L/L}$); **f** swimming pool water with BFA (15.60 $\mu\text{L/L}$). BFA, body fluid analogs; DEMW, demineralized water; TAPW, tap water; TOC, total organic carbon; POOLW, pool water

57.06% (for 3.90 $\mu\text{L/L}$) to 87.56% (for 15.60 $\mu\text{L/L}$) (Fig. 1 a, b). Meanwhile, UV_{254} values increased from 67.72% (3.90 $\mu\text{L/L}$) to 80.94% (15.60 $\mu\text{L/L}$). There was a significant increase in the values of turbidity and UV_{254} in addition to BFA after filtration of POOLW (even up to $R=100\%$) (Fig. 1 e, f). At the same time, the value of retention coefficients for the total concentration of organic carbon decreased from 87.21% (for 3.90 $\mu\text{L/L}$) to 79.67% (for 15.60 $\mu\text{L/L}$).

The effect of the matrix on the filtration due to the presence of BFA was also significant. There was a difference in the transporting and separating properties of the membranes [54, 55]. The level of complexity of the

matrix affects the efficiency of NF membranes [62]. In the analyzed processes, the lowest values of contaminants' retention coefficients were obtained for DEMW (3.90 $\mu\text{L/L}$). This feed showed the lack of foreign compounds from the matrix and low concentration of compounds from the model solution (BFA). BFA (15.60 $\mu\text{L/L}$) caused an increase in the separation properties of the membrane. This phenomenon was observed for processed with the use of matrices, i.e., TAPW and POOLW, for turbidity retention and UV_{254} absorbance coefficients. Meanwhile, separation properties of membranes with respect to reducing the total concentration of organic carbon were not improved.

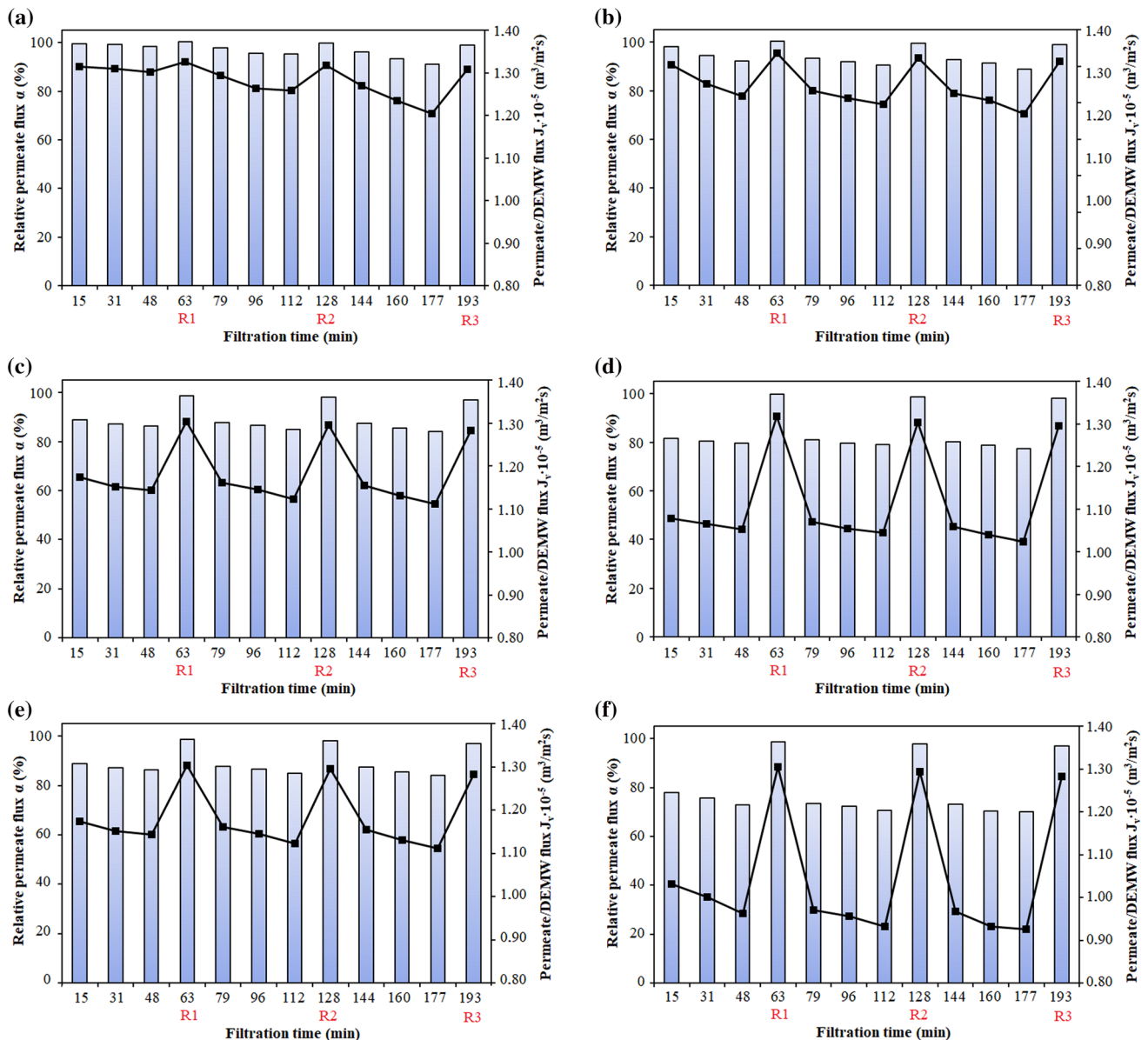


Fig. 2 Transport properties of membranes during nanofiltration processes: **a** demineralized water with a body fluid analog (BFA) (3.90 $\mu\text{L/L}$); **b** demineralized water with BFA (15.60 $\mu\text{L/L}$); **c** tap

water with BFA (3.90 $\mu\text{L/L}$); **d** tap water with BFA (15.60 $\mu\text{L/L}$); **e** swimming pool water with BFA (3.90 $\mu\text{L/L}$); **f** swimming pool water with BFA (15.60 $\mu\text{L/L}$)

The level of complexity of the matrix affects the efficiency of NF membranes [62]. The already-mentioned increase in separation properties is most probably associated with the blockage of pores of the membrane. A significant effect of the matrix on the value of the relative permeability was shown in this study. Furthermore, there was a reduction in the transporting properties of the membrane with an increase in the concentration of BFA (Fig. 2a–f).

The highest value of relative permeability was recorded for NF membranes filtering DEMW ($\alpha_{3.90} = 99.39 \div 91.01\%$; $\alpha_{15.60} = 98.16 \div 88.84\%$). The highest intensity of blockage

of membrane pores was recorded for feed with POOLW as the matrix ($\alpha_{3.90} = 88.31 \div 79.32\%$; $\alpha_{15.60} = 77.94 \div 70.04\%$) (Fig. 2e, f). Our preliminary analysis showed that POOLW with BFA as the contaminants showed the highest capacity to block the pores of NF membranes. The turbidity of POOLW was 0.43 NTU, and the concentration of TOC exceeded 8 mgC/L. Addition of BFA model solution contributed to an increase in the pollutants in the feed.

During the course of filtration processes, interim rinsing of the membrane was planned at 63, 128, and 193 min of the cycle (marked on Fig. 2 under R1, R2, and R3). This allowed us to reset the initial value of relative permeability

to a large extent. The relative permeability for DEMW with the addition of 3.90 and 15.60 $\mu\text{L/L}$ BFA amounted to 100% and 99.30%, respectively. The α values for demineralized water matrices after rinsing the membrane was TAPW + BFA and was 97.73% and 99.24%. In the case of POOLW, the lowest value of α was obtained for both analyzed doses of BFA (97.73%). However, pore blocking is reversible to a large extent. Mechanical cleaning with the use of cellulose sponge allowed to remove the cake layer formed on the surface of the membrane.

The FR value of DEMW after mechanical cleaning exceeded 100% in both the analyzed doses of BFA. The designated irreversible fouling coefficients assumed negative values, which confirms that we are dealing with a reversible phenomenon. The filter cake is probably caused by the adhesion of colloidal particles. Moreover, limiting transporting properties of NF membranes by organic compounds is associated with their adsorption on the surface of the membrane [66]. For the purposes of further analyses of the intensity of blocking pores and the level of its reversibility, it is necessary to extend the process in time. Observing phenomena and analysis of recovery of the flux after rinsing will allow to evaluate the necessity of introducing more invasive cleansing methods, including chemical methods.

In the context of POOLW purification, further analysis is necessary to develop a complete procedure for the NF process. Limitation of fouling can be achieved by pretreatment, change in the membrane material (including surface modification), change in operating parameters, and the use of cleaning methods [66].

It should be borne in mind that in actual POOLW purification cycles, the biofouling phenomenon caused by the presence of microorganisms may have a significant role in blocking the pores [66]. Therefore, one of the possibilities to reduce biofouling is to modify the surface of the membrane with graphene, which limits the multiplication of microorganisms [67]. However, it is necessary to ensure the stability of nanoparticles, which will reduce the risk of their penetration into the environment.

Maintaining the transport properties and reducing the effect of biofouling on pressure membranes can also be achieved by using the pre-ozone process [68, 69]. It has been confirmed that ozone at a concentration of 0.2:0.4 $\text{mgO}_3/\text{mgDOC}$ contributes significantly to improve the separation and transport properties of NF membranes [69]. It is necessary to consider the economic costs between the use of pre-ozonation and cleaning with chemicals. It is also necessary to take the factors that damage the structure and decrease the life of NF membranes such as free chlorine, free oxygen, and microorganisms into account while deciding on the method of purification [70, 71].

The chemical cleaning methods for NF membranes could include the use of citric acid, hydrochloric acid, or sodium hydroxide, taking into account the diverse nature of the contaminants present in the POOLW [72]. In this study, the effect of only a part of organic compounds present in human body secretions was analyzed. The analysis of the influence of BFA compounds on inorganic compounds, which may contribute to the irreversible blocking of pores, should be extended.

4 Conclusions

- NF is a highly efficient process in terms of separation properties of compounds from the BFA solution. Depending on the matrix in which the BFA model solution was dissolved, there was different removal efficiency recorded. Demineralized water matrices with BFA (3.90 $\mu\text{L/L}$) demonstrated the lowest efficiency in terms of reducing the pollutants. An increase in the separation capacities in the presence of complex matrices is associated with the partial blockage of the pores of the membrane (with NOM and colloidal particles).
- As the number of matrix components increased, the diaphragm transport properties decreased. The highest intensity of membrane pore blockage (by producing a cake layer) was observed for the pool water matrices filtration process with BFA (15.60 $\mu\text{L/L}$). The process of rinsing the membrane with DEMW, as well as cleaning with a cellulose sponge, allowed for the recovery of the initial flux value.
- Further analyses are necessary to know the individual compounds associated with human body secretions commonly found in pool water matrices. Expanding the observation of phenomena causing the reduction in the life of membrane and elaborating full procedures of NF is highly warranted (pretreatment—treatment—cleaning).

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Compliance with ethical standard

Conflict of interest The authors declare that there is no conflict of interest.

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