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Removal of petroleum compounds from aqueous solutions in the aeration and reverse osmosis system

Janina Piekutin*, Iwona Skoczko

Department of Technology in Engineering and Environmental Protection, Technical University of Bialystok, 45A Wiejska Str., 15-351 Bialystok, Poland, Tel. +48 85 746 96 44; emails: j.piekutin@pb.edu.pl (J. Piekutin), i.skoczko@pb.edu.pl (I. Skoczko)

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ABSTRACT

The prevalent occurrence and use of oil lead to an increasing environmental pollution by petroleum products. Petrochemical-based products penetrate the soil and then indirectly get to water along with the sewage and stormwaters that wash away the surface of streets and roads. The object of this work was to study the removal of oil from water. The study examined the use of test system: aeration–reverse osmosis (RO) in order to remove C_7–C_{35} petroleum compounds from aqueous solutions. Tests were conducted using the model water—distilled water with the addition of diesel oil and gasoline. The effectiveness of the integrated process was evaluated based on the degree of petroleum compounds removal from the water after various stages of the process, and after completion of water purification. Hydraulic performance of the membrane was also investigated. Six test series, starting from 2,997.87 to 96.5 mg/dm^3 concentration, were performed. The test water was aerated and then subjected to the RO process. Determination of petroleum hydrocarbons was made using a gas chromatograph according to Polish Norm PN-C-04643 and expressed as mineral oil index.

Keywords: Aeration; Water; Petroleum hydrocarbons; Reverse osmosis

1. Introduction

The issue of removal of the petroleum substances from water is still associated with the search for high-efficient methods. Currently, mechanical separation of hydrocarbons, chemical methods, and filtration are widely used [1–5]. To remove the petroleum from an aqueous medium, a variety of equipment for collecting impurities is applied [1–4,6–9].

Up-to-date studies have shown that hydrocarbons dissolved in ground water are subject to a small extent to the adsorption [3,10,11]. This type of pollution belongs to the category of extremely harmful to the environment, thus to develop methods of water purifying from oil pollution is one of the leading trends [7–10]. The purification process must be rationally carried out, and the degree of environment purification has to be determined individually for each object depending on the area management type [1,4,10,11].

Petroleum compounds are a mixture of many hydrocarbons—both aliphatic and aromatic—and their removal from water is one of the biggest problems in industrial practice. The solubility of gasoline in water is in the range from 131 to 185 mg/L [2,3,8], while the constituting hydrocarbons have a very diverse

*Corresponding author.

solubility. The highest solubility characterizes aromatic hydrocarbons such as benzene, toluene, xylene, and ethylbenzene [3,10,12,13]. Removing them from water is difficult and expensive. Therefore, the authors undertook the study upon removal of petroleum from water using the aeration–reverse osmosis (RO) system

The aim of the present study was to evaluate the system applied to remove the C_7–C_{35} petroleum compounds out of the water. The test system has to ensure greater efficiency as compared to the unit processes in physical methods [14–16] during removal of petroleum substances from water.

2. Materials and methods

In the present study, removal of petroleum hydrocarbons was carried out in the aeration–RO. Contaminated water (distilled water with the addition of petroleum compounds) was subject to fine-bubble aeration using diffusers. The pore size of diffusers was 20–150 μm. After aeration, floated petroleum substances were decanted, and the remainder was applied to a RO. The RO process used water, in which the best effect of tested substances removal was achieved. The process was performed in the installation equipped with spiral aromatic-polyamide membrane. The RO system worked in a continuous system with partial recirculation of the concentrate. Membrane filtration tests were carried out at 19˚C under the pressure of 1.1 MPa and the linear velocity over the membrane of 1.0 m/s. Before the final measurements, deionized water was passed through the membrane to determine the maximum permeate flux, then the appropriate test was conducted. After the completion of test , the membrane was rewashed using distilled water, thereby the change in its after-work transport properties was evaluated. The efficiency of the infiltration water filtration process was assessed based on the measurement of volumetric permeate flux (J_v) and its chemical analysis. The analysis included determination of the mineral oil index as a fraction of hydrocarbons in the range of C_7–C_{35}. Presented results for petroleum hydrocarbons were expressed as mineral oil index.

The test pattern included six series by changing the concentration of petroleum hydrocarbons and aeration time. The model water consisted of a mixture of distilled water and petroleum with diesel fuel mixture in 1:3 ratio. The proportions resulted from the amount and type of fuels sold in the domestic market in 2008–2010 [17]. The study began with adding the following amounts of fuel mixture to distilled water: 1, 0.5, 0.25, 0.125, 0.075 through 0.025 cm^3/L, achieving corresponded petroleum hydrocarbons concentrations of 2,997.87, 1,200.33, 617.67, 503.96, 335.23, and 96.5 mg/L. The aeration time for each batch was 15, 30, and 45 min, respectively. Before aeration, to obtain a homogeneous mixture, the model water was homogenized each time using a rotor homogenizer. During the RO process, the sample was collected every 10 min. Determination of monitored aliphatic petroleum C_7–C_{35} hydrocarbons was performed in accordance with the Polish Norm PN-C-04643 applying gas chromatograph coupled to the VARIAN 4000 spectrometer. The device was equipped with a VF-5 MB column with parameters 30 m × 0.25 mm × 0.25 μm. The stationary phase was composed of a polydimethylsiloxane with 5% share of phenol groups. Helium was the carrier gas. Aliphatic hydrocarbons were extracted from water using hexane liquid–liquid method. After separation of the aqueous and organic layers, the eluate was transferred to volumetric flasks and subsequently subjected to drying by means of anhydrous magnesium sulfate ($MgSO_4$, for analysis grade). The temperature program was as follows: 40˚C (5 min) to 130˚C (0 min), with increment 10˚C/min to 300˚C, the temperature of transfer line 230˚C, and ion source temperature 180˚C.

3. Results and discussion

The study revealed that the 45-min aeration (Fig. 1) gave the best results among all tested aeration times. It was evidenced by the high reduction ratio of the test oil, which amounted to about 86%. In the series I, 75.05% of petroleum hydrocarbons removal effect was achieved to yield 718.05 mg/L. In the series II, quantity of studied compounds after the process was 105.24 mg/L to give 91% reduction rate. The effect of petroleum hydrocarbons removal in series III was

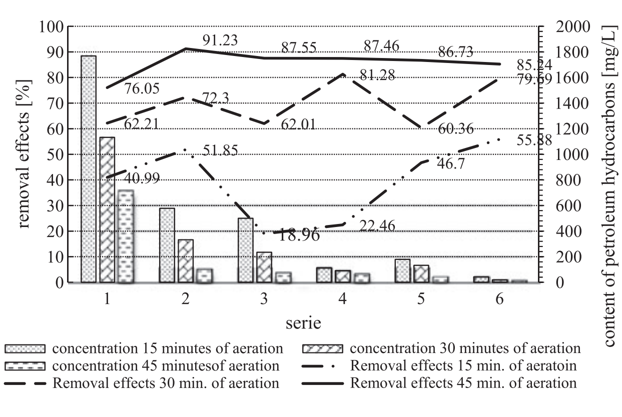

Fig. 1. The content of petroleum hydrocarbons and the degree of their removal after 15, 30, and 45 min of aeration.

86%, reaching 76.89 mg/L of petroleum hydrocarbons. In series IV, 85.46% effect of tested compounds reduction yielding 65.66 mg/L was recorded. In series V, 44.48 mg/L of petroleum hydrocarbons remained in the sample, which proved their high degree (87%) of removal. In the last series VI, 85% removal was obtained to give 14.25 mg/L of petroleum hydrocarbons in purified water. The greatest efficiency of purification after 30-min aeration was achieved in the series II, and the weakest in the series I, which was different from the best value by 14%, on average.

The achieved results (Table 1) can indicate some pattern, i.e. degree of petroleum compounds reduction depends on the time of sample aeration: the longer the duration, the better degree of the removal. It caused that dispersed petroleum substances floated in the form of large droplets.

Water, after 45 min of aeration and decanting the floated layer of petroleum hydrocarbons, was introduced to the RO system. The retention ratio in the RO process ranged from 50 to 99% (Table 2). For series I with the highest concentration, the worst removal results were achieved in RO recording 52% effect, on average. In other series, much higher retention rates were recorded, which were larger by 30–50% than for series I, except from the single result of 57% in the series II, because a failure in RO system occurred that time. Worse removal effect at the end of purification in RO was probably associated with the surface load of the membrane [13,18,19] and the adsorption of compounds on its surface. A decrease in the retention of removed substances occurred as a result of saturation of the membrane surface charge and the mechanism turned into the dissolution and diffusion. Disruption of the separation mechanism was observed during contamination with organic sediments as well as due to concentration changes associated with the micro-pollutants removal (filtration under concentrating conditions increases in the process pressure) [15,19,20]. A decrease in the retention factor was observed along with the increase in the diesel oil concentration in a portion.

The increase in the trans-membrane pressure induced higher concentrations of the mixture on membrane and its structures, which reduced the efficiency of its removal as a result of saturation on the membrane surface.

A decrease in the retention rate and thus a higher concentration of the permeate was observed along with the increase in oil concentration in the feed (Fig. 2).

The study showed (Table 3) that the greater degree of recovery assumed the higher average volume permeate flux (Table 3) and the higher retention rate.

Table 1
The efficiency of model water aeration process

	Content of petroleum hydrocarbons (mg/L)					
	Series					
	I	II	III	IV	V	VI
Raw water	2,997.87	1,200.33	617.67	503.6	335.23	96.55
15 min	1,769.11	577.95	500.53	113.21	178.68	42.6
30 min	1,132.96	332.46	234.68	91.3	132.9	19.61
45 min	718.05	105.24	76.89	65.66	44.48	14.25

Table 2
Efficiency of RO process for samples after 45 min of aeration.

	Content of petroleum hydrocarbons (mineral oil index) (mg/L)					
	718.05	105.24	76.89	65.66	44.48	14.25
No. sample RO (min):	Retention coefficient [%]					
10	54	91	98	97	94	98
20	52	89	86	97	88	95
30	54	57	89	86	83	97
40	50	82	77	78	71	82
50	50	86	75	88	75	87

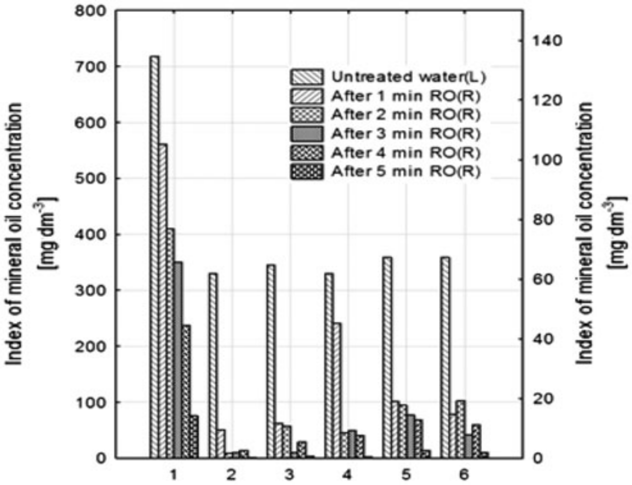

Fig. 2. Concentrations of petroleum hydrocarbons in permeate.

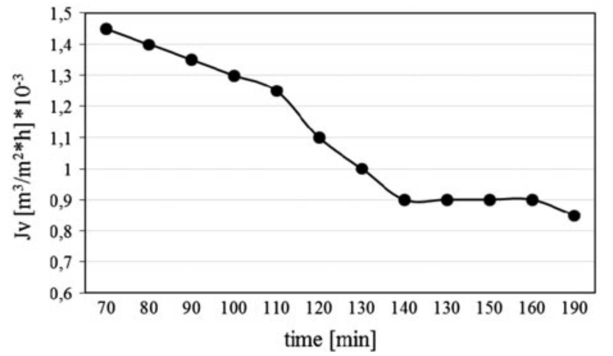

Fig. 3. Permeate stream in the time for primary concentration of mineral oil index 14.25 mg/L.

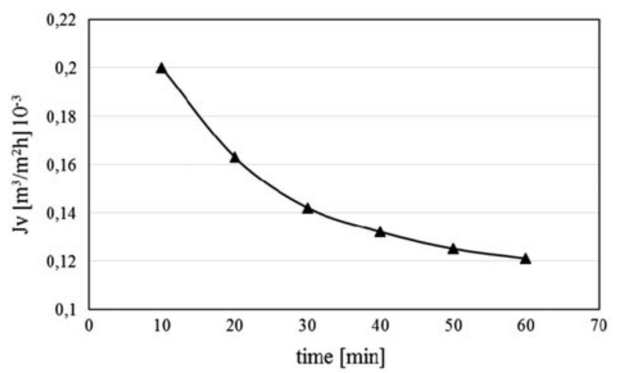

Fig. 4. Permeate stream in the time for primary concentration of mineral oil index 718.05 mg/L.

In this study, lowering of the water recovery rate with increasing concentrations of petroleum hydrocarbons in the feed was also observed. The increase in the trans-membrane pressure induced higher concentrations of the mixture on membrane and within its structure, thus reducing the efficiency of its removal by saturating the membrane surface [7–9,19–22].

Water containing large amounts of organic compounds of hydrophobic character can be effectively removed during the membrane filtration process; however, it is associated with the occurrence of adverse fouling phenomenon, i.e. clogging of the membrane pores and coating its surface with contaminants. Figs. 3 and 4 show the decrease in permeate stream according to the increase in feed water concentration. Otherwise, it was blocked by previous water aeration and additional petroleum compounds removal.

Numerous references and own studies [17,21,23,24] demonstrated that organic substances contained in water significantly contributed to the decline in the membrane performance. Therefore, to prevent such adverse situation, the membrane processes should be combined with conventional water treatment processes.

It was found (Table 3) that the greater degree of recovery assumed the higher average permeate stream volume is achieved (Fig. 1) and the higher retention rate can be recorded. The study also revealed a decline in the degree of water recovery along with increasing concentrations of petroleum hydrocarbons in the portion.

Water purification (Table 4) of petroleum hydrocarbons in the aeration–RO system allowed to obtain a high degree of removal at the level of 89–99%. Integrated system was more efficient solution than individual processes, during which a far worse removal effects were recorded.

In the case of hydrophobic compounds, e.g. petroleum compounds, the separation efficiency may

Table 3
The effect of RO process conditions on the removal of petroleum hydrocarbons

Mineral oil index (mg/L)	718.05	105.24	76.89	65.66	44.48	14.25
Pressure (MPa)	1.4	1.2	1.1	1.1	1.1	1.1
Average permeate stream volume J_v ($dm^3/m^2 \times h$)	27	27	28	28	30	34
Degree of recovery (%)	70	70	71	71	77	81

Table 4
Summary of the results obtained during the test of petroleum hydrocarbons removal from water in an integrated aeration–RO system

Water model (mg/L)	Mineral oil index (mg/L)		Removal effects (%) after whole process
	After 45 min of aeration	After RO	
2,998	718.05	312.08	89
1,200	105.24	26.25	98
617	76.89	13.86	98
503	65.66	15.8	97
335	44.48	8.9	97
96	14.25	0.85	99

depend on the adsorption process occurring on the membrane surface and its structure [19,23]. Reduction in the retention rate (R) with an increase in the trans-membrane pressure observed in RO was confirmed in the literature [14,17,20–22] and commonly reported for the separation of organic compounds characterized by a high affinity to the membrane polymer [12,16,19,20]. Presented results confirmed the effect of trans-membrane pressure on the adsorption phenomenon.

The typical unit process combined with membrane processes improved the efficiency of the membrane as well as the quality of purified water. Applying the configurations of integrated systems of activated carbon—coagulation type [6,10,11], stripping–RO, or aeration–RO—quite high efficiency can be achieved [15,17,19–25].

This study also produced satisfactory results of water treatment at high concentrations of petroleum hydrocarbons. The use of a hybrid combination of classical and membrane system allowed to reach approximately 99% removal of impurities.

4. Conclusions

The study confirmed the possibility to treat water from petroleum hydrocarbons during the aeration–RO. Studies have shown that the water purification by means of RO was possible and the effectiveness of this method was high. Performed aeration and removal of petroleum hydrocarbons from water in RO was an efficient method for the effective reduction of their content in water to a safe level [1–4].

Efficiency of petroleum hydrocarbons removal from water solutions in high-pressure membrane processes was dependent on the compounds concentration in the portion and the recovery degree.

The combination of aeration and RO process increased the efficiency of petroleum hydrocarbons removal from water. In this regard, however, it is necessary to continue the research upon the optimization of these processes.

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