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Coagulation and nano-filtration: A hybrid system for the removal of lower molecular weight organic compounds (LMWOC)

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ABSTRACT

The removal of lower molecular weight organic compounds (LMWOC) from water is of increasing concern. While, nano-filtration (NF) is a good option, it removes only a fraction of the LMWOC. In this paper, NF experiments were conducted to remove oxalic acid and diuron in combination with coagulation using poly-aluminum chloride (PAC) as the coagulant. The results showed that this hybrid treatment system was effective in removing oxalic acid where almost a 100% removal efficiency of oxalic acid was achieved. However, using PAC as coagulant to remove diuron from water was not effective. In order to improve the removal efficiency of diuron, 0.02 M NaCl was added to diuron and a 40% increase in the removal of diuron was achieved. Higher removal of diuron was achieved when the solution was treated with reverse osmosis (RO) when compared to the nano-filtration.

Keywords: Coagulation; Diuron; Low molecular weight organic substances (LMWOC); Nano-filtration; Poly-aluminum chloride; Reverse osmosis (RO)

1. Introduction

One of the major problems in treating water is the removal of organic compounds present in those waters. The presence of organic compounds in the treated water can have great adverse influence on the quality of those effluents. Organic compounds present in the water can be divided into lower molecular weight organic compounds (LMWOC) and higher molecular weight organic compounds (HMWOC). Although HMWOC account for a major fraction of organic compounds [1], they could be removed easily by the existing treatment processes such as enhanced coagulation [2,3] and adsorption [4–6] that are applied in water treatment systems. However, LMWOC are less easily removed compared to HMWOC from water [7,8], and therefore removing LMWOC is of greater concern.

LMWOC are defined as the organic compounds with molecular weight smaller than 1000 Da [9], including bioorganic acids, vitamins, chlorophyll [1,10] and other toxic substances such as persistent organic pollutants (POP), endocrine disrupting chemicals (EDC), and pharmaceuticals and personal care products (PPCP) [11–13]. LMWOC occur both in groundwater and in the surface waters, e.g., rivers, lakes, seas, rainwater and snow ice. Each source of raw water has different concentrations of LMWOC. Nevertheless, most of the LMWOC are present at only trace levels [14,15] in these water sources, for instance, there were about several μg/L of pesticides (e.g., atrazine and chlorpyrifos) by gas chromatography/mass spectrometry analysis, which were most frequently found and in greater concentrations in the Ebro River Basin water samples [16]. However, even in trace levels, they might deteriorate the quality of water, such as color, taste and odor. In the worse case, pathogenic organisms may be produced and the formation of disinfectant by-products (DBPs) such as chlorinated organic substances

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is induced when chlorine is used as the disinfectant. In 2005, the maximum allowable concentration was 30 μg/L for diuron in drinking water [17], and are 80 and 60 μg/L for trihalomethanes (THMs) and haloacetic acids (HAAs) (the two major groups of DBPs), respectively [18]. Since LMWOC and its derivatives could create risks to human health, it is necessary to study the removal of LMWOC from water.

In drinking water production, the removal of LMWOC can be performed through (i) granular/powdered activated carbon adsorption, (ii) oxidation processes by various oxidants such as ozone, hydrogen peroxide, and chlorine, and (iii) membrane processes. The organic substances are adsorbed onto the surface of the activated carbon, thus the activated carbon columns are easily saturated and must be regenerated frequently. Adsorption is an effective method and in many cases high removal of organic compounds could be achieved. For example, 87% removal of atrazine and 94% removal of alachlor from the Maumee River water source were achieved using powdered activated carbon adsorption [19]. However the operating costs of activated carbon columns are very high. With the second technique, the major problem associated with the use of chlorine is the formation of carcinogenic by-products such as trihalomethanes (THMs). The application of ozone does not produce organohalogenated by-products and could provide efficient removal of oxalic acid [20], however, the bacterial re-growth can be induced on the inner surface of the pipe walls if the ozonated water is delivered through a distribution system due to the absence of disinfectant residual [21] and the economic cost is high.

Membrane filtration is becoming increasingly popular and more viable as an alternative treatment for the removal of LMWOC from water sources as there is no formation of by-products in the membrane filtration processes. Moreover, reuse of organic substances such as pesticides is possible. Nano-filtration (NF) and reverse osmosis (RO) are two of the membrane processes and NF is becoming widespread because it could produce higher permeate flux at lower operating pressures relative to RO. The membranes used for NF have a molecular weight cutoff (MWCO) in the range of 200–1000 Da. Therefore, research on NF membranes has gained considerable interest over the last few years. In order to obtain a higher removal of organic compounds while improving the fouling of the NF membrane an hybrid system composed of coagulation and NF has been introduced in the field of water treatment [22–24].

The objective of this study is to investigate the performance of NF with and without coagulation in the removal of two model compounds (oxalic acid and diuron). The effectiveness of poly-aluminum chloride as coagulant and the corresponding optimum dosage are determined for the model compounds.

2. Materials & methods

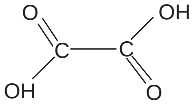
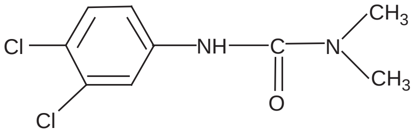
2.1. Model compounds

Oxalic acid and diuron were used as model compounds for the experiments in this study. The characteristics of those compounds are shown in Table 1.

Oxalic acid is a relatively strong organic acid with a molecular formula of $C_2H_2O_4.2H_2O$. It can increase the solubility of heavy metals such as Pb^{2+} in soil [25]. If it occurs in water, the di-anion of oxalic acid (known as oxalate) combines with metal ions such as Ca^{2+}, Fe^{2+}, and Mg^{2+} to form corresponding precipitates, which irritate the gut and kidneys. A prominent example is that calcium oxalate is known as a primary constituent of kidney stones. These factors lead in the selection of oxalic acid as one of the model compounds.

Pesticides are generally toxic and non-degradable, as well as may pollute surface water and groundwater. Diuron is a non-selective herbicide with the molecular formula of $C_9H_{10}Cl_2N_2O$ and is mainly used to control weeds in agriculture. It was chosen as the model

Table 1
Characteristics of model compounds.

Model compounds	Molecular structure	Molecular weight (g/mol)	Size of molecule (nm)
Oxalic acid		126.07	0.25
Diuron		233.09	0.31

compound because it has become a popular herbicide after the ban of atrazine and is less easily removed than the other pesticides [15].

In this study, oxalic acid and diuron were purchased from Ajax Finechem Pty Ltd and Sigma Aldrich (Australia), respectively. The synthetic solutions of model compounds were prepared with distilled water at different concentrations. The oxalic acid concentrations were chosen to be 100, 200 and 500 mg/L for various experiments. As diuron has low solubility in water compared to oxalic aicd, about 10 mg/L of diuron solutions were prepared for the experiments.

2.2. Experiments

2.2.1. Coagulation

Prior to the NF experiments, jar tests were carried out to evaluate the efficiency of coagulation and to determine the optimum dosage of the coagulant. Poly-aluminum chloride (PAC) with a concentration of 13 wt% Al_{total} was used as coagulant, which was supplied by Water Treatment Services (Australia) Pty Ltd.

A jar test apparatus (Flocculator SW1) equipped with six mixing blades and beakers was used to conduct coagulation experiments. Each beaker contained 500 ml of model solution. Coagulant with appropriate doses was added into the beakers and mixed with the model solutions for 3 minutes at 120 rpm. Rapid mixing was followed by the slow agitation for 15 minutes at 40 rpm. Then the solutions were allowed to settle for 20 minutes before the supernatant solutions were collected from the beakers for analysis of parameters such as TOC and UV-absorption.

2.2.2. Nano-filtration (NF)

A laboratory scale filtration unit used in this study consisted of a membrane unit (RNF-0460), and a 15 L stainless steel feed tank with a built in heat exchange jacket. A set of OHAUS electronic scale connected to a computer was used to measure the mass of permeate.

The feed solution was pumped to the membrane at a constant flow rate of 8 L/min by a 1.5 kW plunger pump with a 380 W, 50 Hz power supply. The operating pressure of the membrane can be varied from 0 to 70 bar with a manual valve. The feed, permeate and concentrate solutions were collected for analysis.

A spiral wound DK-1812 NF membrane module purchased from General Electric Company was used in this study, which was a thin film composite membrane with the molecular weight cut-off (MWCO) of 200 Da. To improve the removal of diuron, reverse osmosis filtration was performed using a SG-1812 RO membrane purchased from General Electric Company.

2.3. Analysis of samples

A total organic carbon (TOC) analyzer (TOC-V, Shimadzu Corporation) was used to determine TOC of samples before and after treatment. In this paper, TOC is expressed as the concentration of the model compounds. Moreover, ultraviolet absorbance (UV) was measured using a Hewlett-Packard 8453 ultraviolet-visible spectrometer to determine the relative concentration of model compounds. The pH of treated water was measured by a TPS WP-80 pH meter.

2.4. Calculation of flux

Besides TOC and UV removals, the flux of permeate from membrane filtration is also an important aspect that needs consideration. The membrane filtration experiments yielded the mass of total permeate collected every 3 minutes. This data was used to calculate the flux, J_i, at the i^{th} time interval using the following equation

$$J_i = \frac{m_i - m_{i-1}}{\rho A (t_i - t_{i-1})} \tag{2.1}$$

where m_i is the mass (kg) recorded at the i^{th} time interval, ρ is the density of the feed solution (kg.m^{-3}), A is the active surface area of the membrane (m^3), and t_i is the time recorded at the i^{th} time interval.

The density, ρ, was assumed to be 1000 kg.m^{-3} for all solutions. Although this value is larger than the actual density of 997 kg.m^{-3} at 22°C to 23°C, a difference of 3 kg.m^{-3} would produce errors in the flux values which will be less than 1%. The active membrane surface area was 0.46 m^2.

3. Results and discussion

3.1. Evaluation of the performance of coagulation

3.1.1. Removal of oxalic acid in terms of TOC and UV

Fig. 1 shows the removal of oxalic acid in terms of TOC by coagulation for 100, 200 and 500 mg/L of oxalic acid solutions, respectively. In all those three cases, TOC removal increased with the increase in PAC dose, although the maximum removal of TOC was less than 7%. The highest TOC removal obtained by PAC coagulation were only about 4.0, 3.5, and 6.8% for 100, 200 and 500 mg/L of oxalic acid solutions, respectively and the corresponding optimum PAC doses were 300, 300 and 600 mg/L. The optimum PAC doses that gave the highest TOC removals were selected for the membrane filtration experiments.

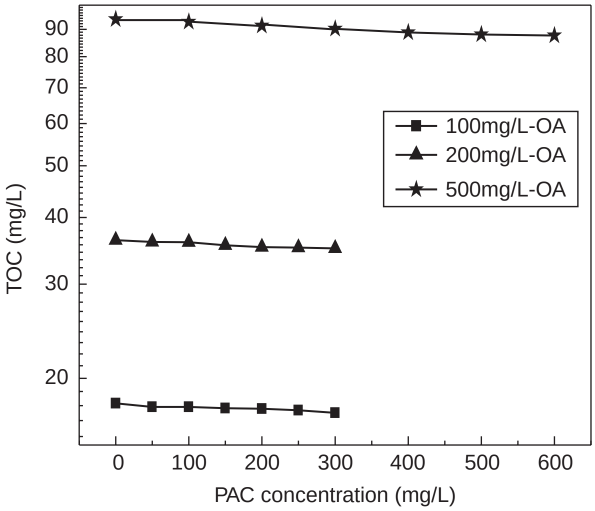


Fig. 1(a). The TOC values of tested oxalic acid solutions after coagulation (initial concentrations of oxalic acid are 100, 200 and 500 mg/L).

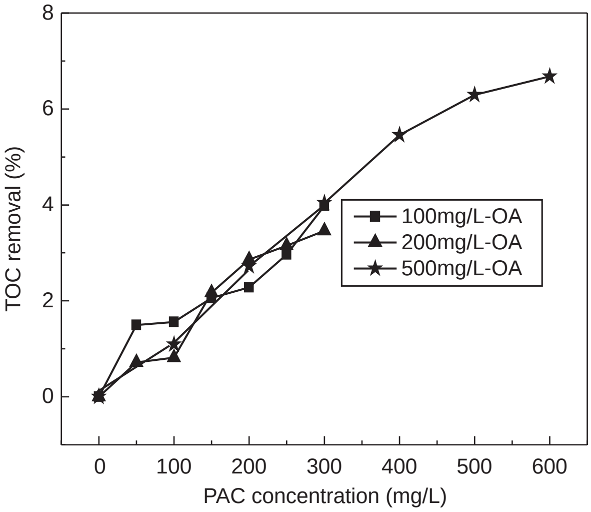


Fig. 1(b). The TOC removal percentage of tested oxalic acid solutions after coagulation (initial concentrations of oxalic acid are 100, 200 and 500 mg/L).

The performance of coagulation was also reflected by UV-199 absorbance measurements. The effect of PAC doses on UV-199 absorbance is presented in Fig. 2. After coagulation, the UV-199 absorption of those three concentrations of oxalic acid solutions showed similar trends where the UV-199 absorption decreased slightly as PAC dose increased. Moreover, it was found that the oxalic acid solutions with concentrations of 200 and 500 mg/L exhibited the lowest UV-199 absorptions at the corresponding optimum doses.

From the results of TOC and UV removals, it can be seen that the removal of oxalic acid is quite limited by the PAC coagulation alone.

3.1.2. Diuron solution-TOC and UV removal

Fig. 3 presents the concentrations of diuron in terms of TOC after PAC coagulation. There was no significant TOC removal observed in the tested range of PAC doses. Only about 3% of TOC was removed at a PAC dose of 800 mg/L.

The effect of PAC doses on UV absorbance is summarized in Fig. 4. It was found that there were two peaks for absorption, one at 211 nm and another one at 248 nm.

The results showed that coagulation using PAC as coagulant was not effective for removing diuron from water.

Oxalic acid and diuron are LMWOC with molecular weights less than 500 Da. These results indicated that coagulation with PAC alone was not good at removing LMWOC, as previous studies have shown that $FeCl_3$ coagulation preferentially removed organic compounds that are higher than 1000 Da [26].

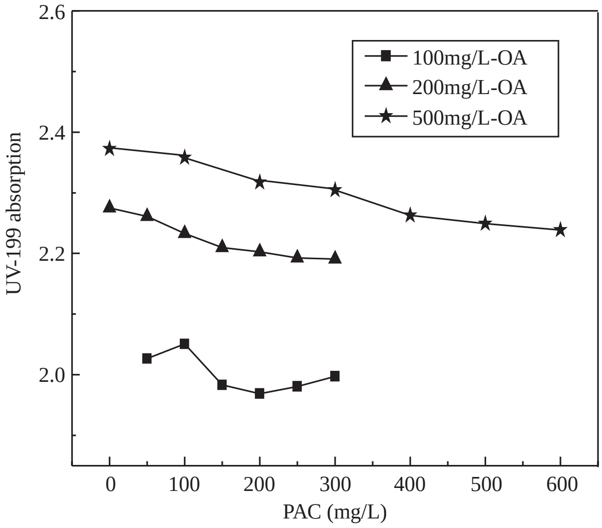


Fig. 2. The effect of PAC doses on UV-199 absorption of the oxalic acid solutions with initial concentrations of 100, 200 and 500 mg/L.

3.2. Evaluation of the performance of membrane filtration

3.2.1. Oxalic acid

The NF membrane filtration experiments were conducted with coagulation at different doses of PAC for three concentrations (100, 200 and 500 mg/L) of oxalic acid solution.

3.2.1.1. TOC removal Table 2 summarizes the removal of TOC after NF with the addition of different concentrations of PAC. It is observed that PAC coagulation prior to NF improved TOC removal. With increase

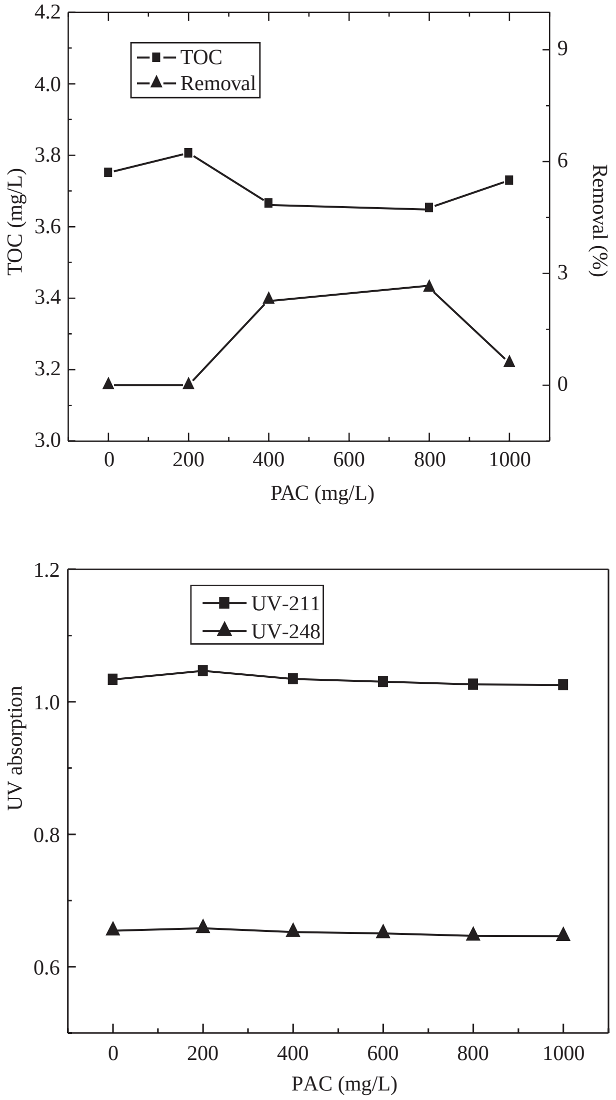


Fig. 3. TOC removal of diuron solutions after coagulation.

in PAC dose, the removal efficiency increased for these three tested solutions. When PAC was used at the corresponding optimum doses, excellent removal (~100%) of TOC obtained. It is interesting to note that, for the two lower concentrations of oxalic acid solutions (100 and 200 mg/L), TOC removal higher than 90% were obtained even at 100 mg/L of PAC dose. Therefore, good removal efficiency can be achieved when combining coagulation with NF to remove oxalic acid.

The general rejection mechanisms of NF membrane include sieving effect based on size exclusion, charge effect and other physicochemical effects such as dipole interactions [27,28]. The charge effect plays a significant role in the removal of organic compounds [29], especially for oxalic acid as the oxalate ion occurred in the synthetic solution. The charge interaction between the NF membrane surface and the oxalate ion resulted in the rejection of oxalic acid. After PAC was added to the feed water, the hydrolyzing aluminous species with positive charge flocculated oxalic acid to form flocs with larger sizes than oxalic acid, which were removed easily based on the size exclusion [30]. On the other hand, the high removal of oxalic acid resulted from the adsorption of oxalic acid on the surface of the NF membrane. This is owing to the formation of the electric double layer on the membrane surface [31]. After the addition of PAC, an electric double layer also occurred on the coagulant surface, and thus the interaction between two electric double layers leads to the adsorption of oxalic acid on the membrane surface.

There was an obvious effect of oxalic acid concentration on the removal of TOC. An increase in the initial oxalic acid concentration resulted in a decrease in the removal of TOC in all instances. Buffle et al. [32] also reported similar results for the removal of LMWOC. The result can be explained by the Donnan exclusion effect where a shield is produced with increasing ionic concentration as found by Bartels et al. [33].

3.2.1.2. Flux of oxalic acid solutions through NF membrane For the three tested oxalic acid solutions, the flux through NF membrane after the addition of PAC was calculated according to equation (2.1) and the results obtained have been presented in Figs. 5, 6 and 7.

These results indicate that a better TOC removal at the optimum dose of PAC resulted in greater fouling of membrane compared to other doses. It is possibly

Table 2
TOC removal for oxalic acid solutions after NF.

PAC (mg/L)	TOC removal (%) 100 mg/L-oxalic acid (OA)	TOC removal (%) 200 mg/L- OA	TOC removal (%) 500 mg/L- OA
0	80.94	66.97	46.27
50	89.15	74.37	50.59
100	96.47	91.04	54.93
300	99.27	98.37	77.70
600			97.74

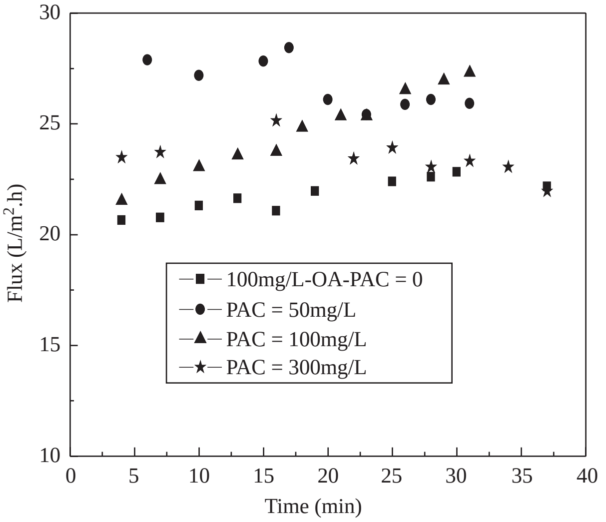


Fig. 4. Flux of 100 mg/L oxalic acid solution (operating pressure = 5 bar).

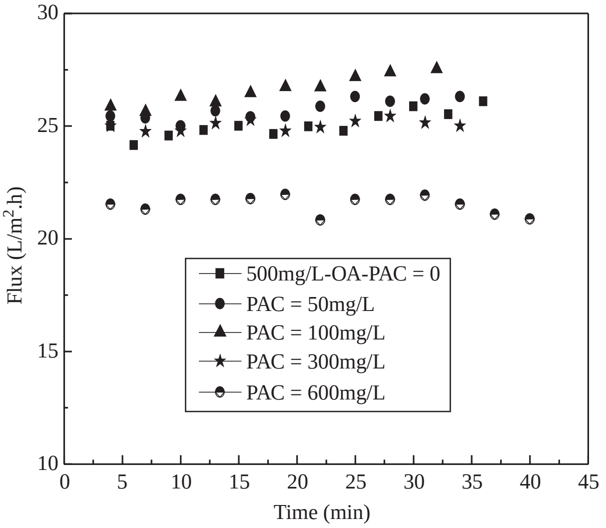


Fig. 6. Flux of 500 mg/L oxalic acid solution (operating pressure = 5 bar).

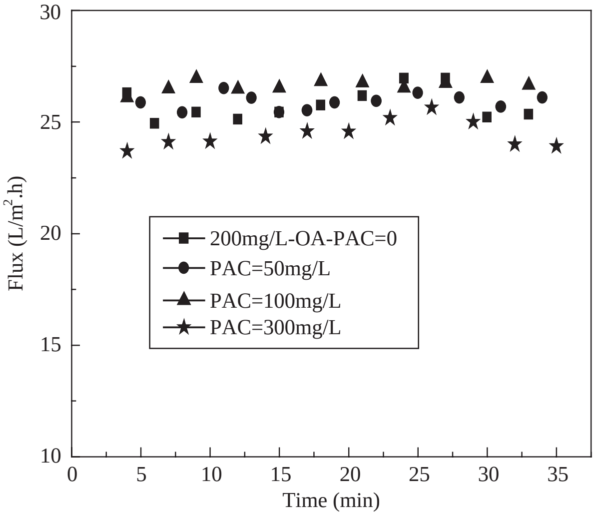


Fig. 5. Flux of 200 mg/L oxalic acid solution (operating pressure = 5 bar).

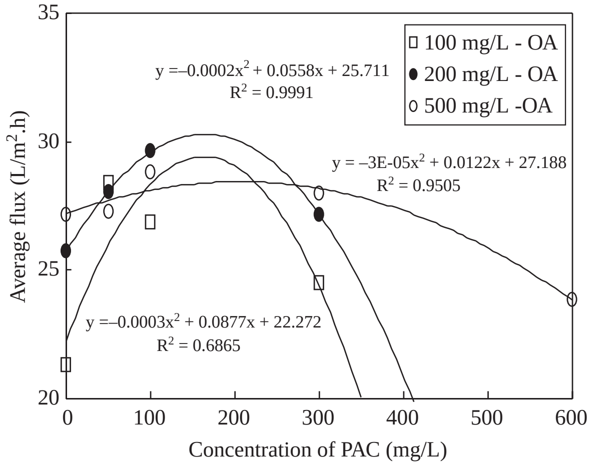


Fig. 7. Average flux of 100, 200 and 500 mg/L oxalic acid solutions (operating pressure = 5 bar).

owing to the formation of flocs due to the addition of PAC. Large numbers of smaller flocs were produced by PAC at the optimum dose [34,35] and these smaller flocs could block the membrane pores easily. Therefore, the use of PAC at the optimum dose resulted in more pore blocking and increased membrane fouling.

The average flux through NF membrane for 100, 200 and 500 mg/L oxalic acid solutions was shown in Fig. 8. For these oxalic acid solutions, the highest average flux was always achieved at the PAC doses lower than the corresponding optimum doses.

3.2.2. Diuron

To determine the performance of NF in removing diuron, NF membrane filtration experiments were carried out. Filtration experiments using RO membrane were also conducted to improve the removal of diuron.

3.2.2.1. TOC removal Van der Bruggen et al. [36,37] reported that the concentration of pesticides (at several mg/L level) did not influence the removal efficiency of pesticides through the membrane filtration treatment. In this study, five concentrations (less than 10 mg/L) of diuron solution was prepared and treated using (i) NF at different pressures, (ii) NF after the addition of PAC and NaCl, and (iii) RO. Table 3 summarizes the removal of TOC and the pH of the solution for different initial concentrations of diuron.

First, it can be seen that the RO treatment showed best results for the removal of diuron with a 95.47%

Table 3
TOC removals and pH for diuron after NF and RO.

	NF 5 bar	NF 10 bar	NF-800 mg/L PAC 5 bar	NF- 0.02 M NaCl 5 bar	RO 35 bar
Initial concentration of diuron in terms of TOC (mg/L)	7.59	8.07	7.77	9.24	4.28
pH	7.16	7.54	4.75	6.91	6.76
TOC removal (%)	22.78	23.52	23.74	60.02	95.47

removal of TOC. This can be explained by the size exclusion mechanism. On the contrary, the NF membrane, which has a MWCO of 200 Da, gave very low removal of diuron at operating pressures of 5 and 10 bars. The removal of diuron, which is an uncharged compound, was mainly decided by the molecular size and the dipole moment [38–40]. For the NF membrane used, the size of diuron molecule (0.31 nm) is similar with the pore size of membrane. Furthermore, diuron has a higher dipole moment and a linear form of molecule [37].

The second observation is that the low removal of diuron was also obtained by NF together with PAC coagulation. Compared to the results obtained by NF treatment alone, very little improvement was found in the rejection of diuron due to the addition of 800 mg/L of PAC. From the pH results, it can be concluded that there was almost no interaction between PAC and diuron because most of PAC present in the diuron solution permeated through the membrane.

As for the effect of NaCl on the diuron removal, it exhibited a dramatic improvement compared to the results obtained by other NF treatments. This is due to the adsorption between the ions and the negatively charged membrane surface [41,42]. This adsorption behavior resulted in the reduction of membrane pore size and thus increased the rejection of diuron.

3.2.2.2. Flux of diuron solution through NF and RO
Figure 9 shows the flux of diuron solution through NF and RO membrane. It can be found that the flux through the NF membrane at 10 bar was twice as the flux obtained at 5 bar. RO membrane showed higher flux than NF membrane at 5 bar, but lower flux than NF membrane at 10 bar.

Although PAC coagulation did not reduce the TOC (Table 3), the flux decreased. It means more membrane fouling occurred with the addition of PAC. In contrast, the flux did not decrease or increase with the presence of NaCl, even if the TOC removal was improved significantly. These results indicate that the addition of NaCl was a good choice for the NF membrane to improve the rejection of diuron, not PAC.

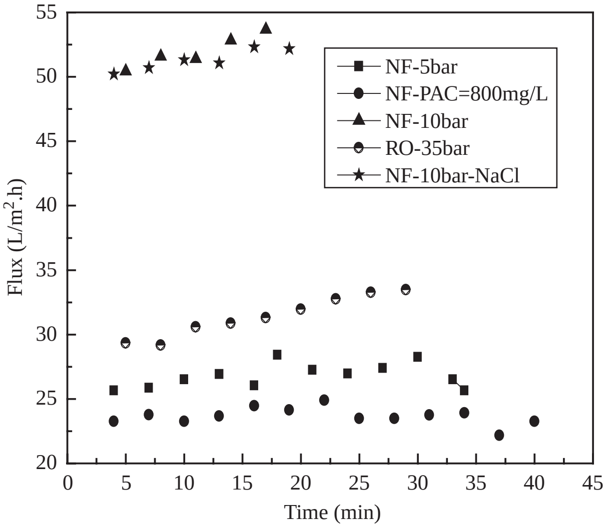


Fig. 8. Flux of diuron solution through NF and RO.

4. Conclusion

The experiments with oxalic acid concentrations of 100, 200 and 500 mg/L clearly indicate that NF in combination with coagulation was quite an effective treatment to remove the lower molecular weight organic acid from water. During the NF treatment, PAC coagulation was very helpful for removal of oxalic acid especially at higher concentrations. However, membrane fouling became more serious if the optimum doses of PAC were used. Therefore, PAC doses which were lower than the optimum but still would provide a better TOC removal, can be used to achieve a less membrane fouling. This consideration will reduce the operating cost while maintaining the higher removal efficiency.

For the tested diuron solutions, removal efficiency around 23% was obtained by using the DK-1812 NF membrane even with the addition of PAC. Nevertheless, the use of NaCl in the NF experiments enhanced the removal efficiency up to 60%, with no additional fouling of membrane. RO membrane has a good capacity to remove diuron if its relatively high operating cost compared to NF is ignored.

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