Application of nanofiltration membrane in the recovery of aluminum from alkaline sludge solutions

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Abstract. Large amounts of aluminum hydroxide ($Al(OH)_3$) exist in water purification sludge (WPS) because of the added aluminum coagulant in water treatment process. Notably, $Al(OH)_3$ is an amphoteric compound, can be dissolved in its basic condition using sodium hydroxide to form aluminate ions ($Al(OH)_4^-$). However, in a process in which pH is increasing, the humid acid can be dissolved easily from WPS and will inhibit the recovery and reuse of the dissolved aluminate ions. This study attempts to fix this problem by a novel approach to separate $Al(OH)_4^-$ ions using nanofiltration (NF) technology. Sludge impurity in a alkaline solution is retained by the NF membrane, such that the process recovers $Al(OH)_4^-$ ions, and significantly decreases the organic matter or heavy metal impurities in the permeate solution. The $Al(OH)_4^-$ ion is an alkaline substance. Experimental results confirm that a recovered coagulant of $Al(OH)_4^-$ ion can effectively remove kaolin particles from slightly acidic synthetic raw water.

Keywords: aluminate; nanofiltration membrane; recovery; coagulant

1. Introduction

Industrialized and urbanized cities worldwide require large amounts of potable water. Coagulation plays an important role in most water treatment processes. Either aluminum sulfate or polyaluminum chloride (PACl) is normally used as a coagulant to remove soil particles and organic matters from raw water. The coagulation process causes tremendous amounts of Al(OH)3 and solid containing sludge, and roughly 30% of the remaining sludge is aluminum salt (Cornwell and Lemunyon 1980). It is still an issue to choose a disposal method for the water purification sludge (WPS) that would be reasonable in terms of technology and economy. There are laboratory and full scale attempts at using WPS as a component in the manufacture of concrete; as geotechnical works materials; as a potential for use in agriculture; as a primary source of aluminum and iron based coagulants through several recovery process; and for phosphorus

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reduction during wastewater treatment (Evuti and Lawal 2011, Hegazy *et al*. 2012). Among these disposal methods, the recovery of coagulant has high economic advantage and recommended as a suitable treatment option for the disposal of WPS (Evuti and Lawal 2011). Therefore, recycling aluminum from WPS will reduce the usage of new coagulant or for other purposes (Babatunde and Zhao 2007, Yang *et al*. 2014, Cheng *et al*. 2016). The main approaches for recovering aluminum ions from sludge are acidification, basification, membrane separation, and ion exchange (Sengupta and Prakash 2004, Jiménez *et al*. 2007, Huang *et al*. 2011, Cheng *et al*. 2012, Yang *et al*. 2013, Keeley *et al*. 2014, Dassanayake *et al*. 2015). Since $Al(OH)_3$ is an amphoteric compound, it can react with acid to form soluble aluminum ion (Al^{3+}) or can react with a base to form soluble aluminate ion $Al(OH)_4^-$. According to Panswad and Chamnan (1992), if the aluminum sludge pH is reduced to 1-3 by adding H_2SO_4, the aluminum recovery rate is approximately 70-90%. If NaOH is used in a recovery process, aluminum recovery is optimal when pH is 11.2-11.8 (Masschelein *et al*. 1985). These recovery processes related to pH adjustment are relatively simple. Therefore, they are commonly used in aluminum recovery studies. For instance, the pH value recently identified that maximizes aluminum dissolution efficiency (in terms of moles of aluminum dissolved per mole of H^+ or OH^- ion) is 2 and 12 for acidification and basification, respectively (Li *et al*. 2005, Huang *et al*. 2010). The major reaction for acidified sludge is expressed as

$$Al(OH)_3 + 3H^+ \rightarrow Al^{3+} + 3H_2O \qquad (1)$$

Decreasing solution pH can dissolve aluminum and thereby increase the aluminum recovery rate. However, excessive acidification may increase operational costs. Additionally, research results reported by Cornwell and Susan (1979) showed that when sulfuric acid is used in the aluminum recovery process, the amount of dissolved aluminum did not increase as the amount of acid increased at the mole ratio of the aluminum in sludge to sulfuric acid 3:2. Also, a shortcoming of acidification processes is large amounts of Fe^{3+}, Mn^{2+} and other heavy metal ions are dissolved during the acidification process. These metal ions may adversely affect the aluminum recovery efficiency (Huang *et al*. 2010). In Eq. (2), sodium hydroxide or calcium hydroxide is typically utilized to form aluminate ion $Al(OH)_4^-$

$$Al(OH)_3 + OH^- \rightarrow Al(OH)_4^- \qquad (2)$$

The process utilizing an alkaline solution results in majority heavy metal removal from a sludge solution by precipitating metal hydroxide. Masschelein *et al*. (1985) indicated that a relatively high aluminum recovery rate (80%) was obtained when solution pH was adjusted to the range of 11.2-11.8 by adding sodium hydroxide. When the sodium hydroxide dosage was increased, the aluminum recovery rate reached 98%; however, increasing the chemical dosage was not cost-effective. Conversely, when pH is increased, organic matter in alum sludge can be dissolved easily in a strongly alkaline solution. These dissolved organic compound inhibit the recovery and reuse of aluminum (Dhage *et al*. 1985). Either acidification or basification may enhance the dissolution of impurities in sludge. These impurities may adversely affect the quality of recovered aluminum, such that the aluminum is not suitable for reuse. Therefore, studies in recent years focused on finding a way to acquire pure aluminum ions from sludge. These recovery methods are based on ion exchange or membrane filtration. The ion exchange methods include liquid-phase and solid-phase ion-exchange methods the liquid ion-exchange process can obtain high purity aluminum salt, but the recovery process requires an extraction solvent (David *et al*. 1980), and many extraction solvents contain toxic compounds. Furthermore, optimal extraction conditions remain unknown (Yu *et al*.

2009). The exchange capacity of the solid-phase ion-exchange resin is limited to a constant and the ion-exchange process is relatively long (Petruzzell *et al.* 2000). Li and Sengupta (1995) used composite ion-exchange membranes to recover Al^{3+} ions. First, a composite membrane is placed in the acidified sludge solution to adsorb aluminum ions. Then the chelating exchanger of the composite membrane is regenerated in a stirring regeneration tank by desorbing the aluminum ions under acidic conditions. However, this approach cannot yield extremely high concentrations of Al^{3+} ions. Additionally, the composite ion-exchange membrane has not been commercialized and is not widely used. Prakash *et al.* (2003, 2004) used applied an ion-exchange membrane to recover aluminum from wastewater treatment sludge. Based on the Donnan effect, the recovery rate for this process can be derived. Although the recovery rate reported by Prakash *et al.* (2004) was extremely high, acidifying the feed solution with a strong acid is necessary to increase aluminum ion recovery rate.

Therefore, the main limitations for recovering aluminum from sludge and its reuse are the residual effects of heavy metals, organic matter, and operational cost (Xu *et al.* 2009). Typically the recovered aluminum cannot be used directly as coagulant in drinking water treatment processes. This study used a commercialized, low-priced household filtration machine with a nanofiltration (NF) membrane to recover aluminum ions from slightly acidic or basic sludge solution. An NF membrane is subject to the Donnan exclusion effect, such that it can effectively exclude high valence metal ions and let mono-valence metal ions permeate past the membrane (Yeom *et al.* 2000, Akbari *et al.* 2002). For acidification or basification methods, the high valence Al^{3+} or mono-valence $Al(OH)_4^-$ ions were dissolved in sludge. Therefore, if the sludge is acidic, the NF membrane is utilized as a barrier to obtain a high concentration of Al^{3+} ions in the retentate solution. Conversely, if the sludge is alkaline, the NF membrane filters out impurities and a purified $Al(OH)_4^-$ solution is obtained in the permeate solution. Therefore, if an NF membrane is used to concentrate the aluminum ions from an acidified sludge solution, it will increase the concentrations of aluminum salt, and heavy metals and organic matter. Hence the circulating solution was not appropriate for direct use as a coagulant. In contrast, because most heavy metals form hydroxide precipitants in neutral and alkaline solutions, very low levels of heavy metals remain in a permeate alkaline solution. In an NF membrane filtration system, the macromolecular organic matter that dissolves in an alkaline solution can be in the retentate solution and the pure aluminate ions ($Al(OH)_4^-$) can be obtained effectively in the permeate solution.

In a membrane filtration process, liquid is pressurized to pass through a membrane (Cath *et al.* 2006). The relationships among membrane flux, rejection rate, and osmotic pressure are expressed as following Eq. (3) (Yuan and Zydney 2000)

$$J_v \equiv \frac{1}{A}\frac{dV}{dt} \tag{3}$$

where J_v is membrane flux (L h^{-1} m^{-2}); A is the area of the membrane used for filtration (m^2); V is the permeate liquid volume (L); and t is filtration time (h)

$$R_i = 100 \bullet \left[1 - \frac{C_{pi}}{C_{Bi}} \right] \tag{4}$$

where R_i is the rejection rate; C_{Pi} is the permeate concentration; and C_{Bi} is the bulk feed concentration

$$\pi = C_M RT \tag{5}$$

Where π is osmotic pressure (atm); CM is the sum of the aluminum concentrations (mole L^{-1})whose value is calculated as aluminum concentration (mgL^{-1})/27000 (mg mole^{-1})) and total organic carbon concentration (mole L^{-1}), whose value is calculated as total organic carbon (TOC) concentration (mg L^{-1})/12000(mg/mole)); R is the ideal gas constant (0.082 atm L mole^{-1} K^{-1}); and T is absolute temperature (K).

In this study, sulfuric acid or sodium hydroxide solution was added to aluminum sludge to dissolve the aluminum ions into the solutions. After the solid-phase floc (i.e., sludge) settled and separated, the liquid-phase supernatant was extracted for NF membrane filtration. Parameters, including membrane flux, retentate concentration (aluminum, aluminate, organic matter, and heavy metals) and osmotic pressure are discussed. Additionally, synthetic raw water containing kaolin was adjusted to acidic and was coagulated using three coagulants: polyaluminum chloride (PACl); aluminum sulfate (Alum); and the recovered aluminate ions. These coagulants were used to evaluate the effect of the coagulants on turbidity removal efficiency.

2. Materials and methods

2.1 Main composition of water treatment sludge

The sludge sample was harvested from a sludge-drying bed at the water purification plant of the Ming-Der Dam, Miao-Li County, Taiwan. This water treatment plant uses traditional coagulation and flocculation processes to treat roughly 35,000 tons of water/day. Polyaluminum chloride (basicity, 47.6%; Al_2O_3, 10.57%; Chung Hwa Chemical Industrial Works, Ltd., Taiwan) is added to the rapid-mixing tank. After mixing, the treated water flows into three flocculation tanks that are connected in series. Flocculation mixing was *via* a mechanical paddle. Following flocculation, the water is treated consecutively by sedimentation, filtration, and chlorination to meet drinking water standards. The sludge samples were dried at 105°C. An Energy Dispersive Spectrometer (EDS) (Fig. 1) was used to analyze the elemental composition of the sludge. Analysis showed that Si, Al, and Fe were the dominant elements. The sludge sample was also analyzed by X-ray diffraction (XRD) to identify its crystal structure. The sludge is primarily composed of SiO_2 and Al_2O_3 (Fig. 2). Additionally, the sludge may have also contained some Fe_2O_3, $KAl_3Si_3O_{10}(OH)_2$ (Muscovite), and $(Ca,Na)(Si,Al)_4O_8$ (Labradorite). The EDS and XRD results indicate that aluminum is a major metal in the sludge
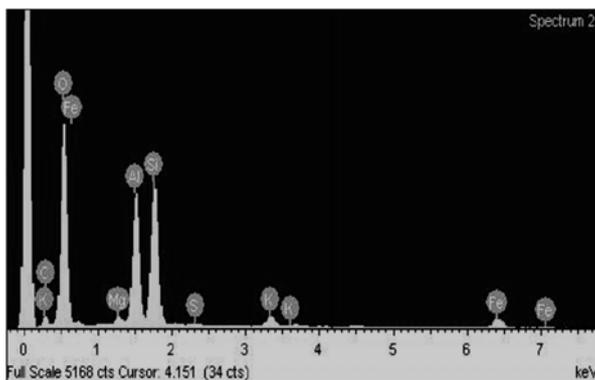

Fig. 1 Element analysis by Energy Dispersive Spectroscopy (EDS)
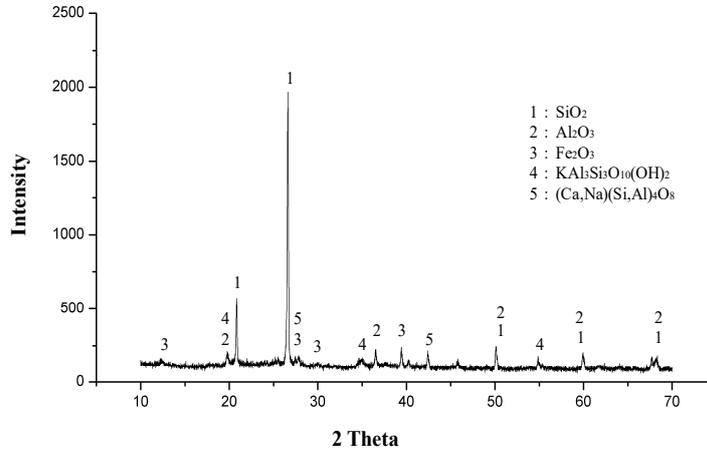
Fig. 2 Crystal structure analysis of the sludge by X-ray diffraction (XRD)

sample. The total amount of aluminum that can leach out in the acidification process was determined using the standard method by Taiwan's Environmental Protection Administration (NIEA R353.00C; dissolved in nitric acid (1:1)). The quantity of aluminum was 40.03 mg Al/g sludge.

2.2 Experimental layout and membrane pre-treatment

Fig. 3 shows the layout of an NF filtration experiment. The part number on the NF membrane was HL1812T (GE, USA). The membrane was composed of three layers of aromatic polyamide fibers. The cross-sectional area was 0.5 m^2; operational pressure was 4-16 atm; and cut off molecular weight was 150-300 MW. The operational pH is suggested as 2-12. The pH should not be <1 or >13. Before the new membrane was used, it was immersed in de-ionized water for 24 hrs and then flushed with pressurized water for 2 hrs.

(1) Valve   (2) 20 L Feed tank   (3) Pressure gauge
(4) UF or NF membrane unit   (5) Pre-filtration unit   (6) Pump
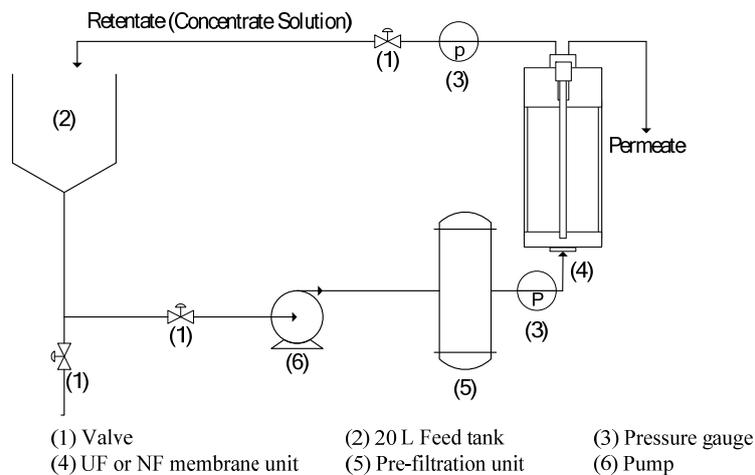
Fig. 3 Experimental setup

2.3 Experimental procedures

2.3.1 Acidity and alkalinity effects on nanofiltration

In total, 300 g sludge obtained from the water treatment plant at the Min-der Reservoir was added to 5L of 0.3N sulfuric acid solution or 5L of 0.5N sodium hydroxide solution and stirred for 3 hrs. Solution pH of the 0.3N sulfuric acid solution and 0.5N sodium hydroxide solution were 2.2 and 11.7. After settling, 4L supernatant was extracted and analyzed for the aluminum ion concentration. The supernatants were separately filtrated by the NF membrane at a pressure of 7.0 atm. In Fig. 3, the retentate liquid (bulk) in filtration process was continuously circulated into the feed tank and the permeate liquid was collected in a 5L (PE) bottle. Filtration continued until the membrane flux of the sulfuric acid solution was reduced to 1 (L h^{-1} m^{-2}) and the flux value for sodium hydroxide test was <3 (L h^{-1} m^{-2}). During the filtration, samples of solutions in both the feed tank and PE bottle were taken every 5 minutes. They were filtrated and analyzed by atomic absorption spectrometry (Z-5000; Hitachi Co. Japan) to determine their aluminum concentrations (mg L^{-1}). A total organic carbon (TOC) analyzer (Phoenix 8000; Tekmar Dohrmann Co., USA) was used to determine the concentration of organic matter (mg L^{-1}).

2.3.2 Acidic raw water treated with different coagulants

Synthetic raw water was prepared in the laboratory by adding 1 g kaolin to 1 L de-ionized water and mixed rapidly. The mixture was then allowed to settle for a 10-minute period. After settling, the top 600 mL of the supernatant was decanted and some de-ionized water was added to make the turbidity of this kaolin solution about 100 NTU. Then, 1.224 g L^{-1} NaClO$_4$ was added to the kaolin solution to increase ionic strength to 10^{-2} M. The pH of the kaolin solution was then adjusted to 5. This solution was then used as synthetic raw water in the jar test experiments. Coagulant doses in the jar tests were 1.6 mg L^{-1} (as Al) for all three coagulants, PACl, Alum and recovered aluminate. Each coagulation experiment involved used a speed-adjustable 75 mm×25 mm flat paddle as an impeller and a 1 L beaker. After adding raw water and coagulant to the beaker (i.e., jar), the content was mixed rapidly at 100 rpm for 1 min, mixed slowly at 20 rpm for 10 minutes, and allowed to settle for 60 minutes. Samples were taken at 3 cm below the water surface every 10 minutes. Sample turbidities were analyzed by an HACH 2100 Turbidimeter (Hach co., USA).
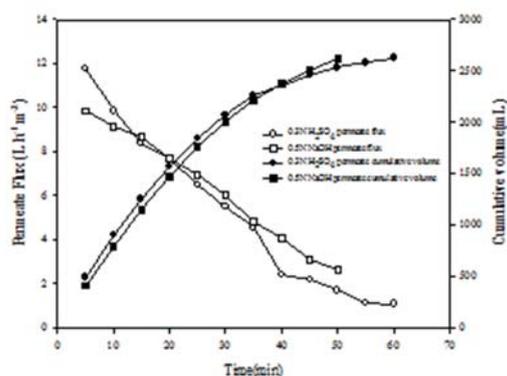

Fig. 4 A plot of membrane filtration flux and accumulated volume as functions of time
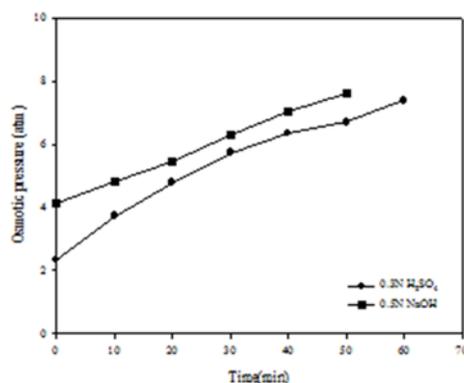
Fig. 5 Changes in osmotic pressure as a function of time during filtration of two solutions

3. Results and discussions

3.1 Acidity and alkalinity effects on NF membrane filtration

In total, 300 gm of sludge was added and mixed in either 5L of 0.3N sulfuric acid or 5L of 0.5 N NaOH solutions for 3 h. After mixing, each supernatant was collected for the NF membrane filtration test. When operational pressure remained at 7.0 atm, the cumulative volume of the permeate liquid increased and the volume of the retentate liquid decreased (Figs. 4 and 5). Therefore, osmotic pressure calculated by Eq. (5) increases with filtration time due to the increase of amount of dissolved aluminum ions and organic compounds in the retentate liquid. When osmotic pressure exceeded 7.0 atm in the test to filtrate 0.3N sulfuric acid solution, membrane flux was <1 ($L\ h^{-1}m^{-2}$) and the flux of the test to filtrate 0.5 N NaOH solution was <3 ($L\ h^{-1}m^{-2}$). As the permeate volume could not be increased further, the NF membrane test did not continue. The organic matter solubility was higher in the alkaline solution than that in acidic solution (Fig. 6). Therefore, initial membrane flux of the alkaline solution was less than the flux of the acidic solution (Fig. 4). Additionally, when both solutions have the same initial aluminum concentration (Fig. 7), analytical results show that the aluminum concentration in the acidic retentate liquid was greater than that in basic solution. For the 0.3N sulfuric acid solution test, the aluminum concentration was increased from 2610 mg L^{-1} to 6123 mg L^{-1} in the retentate solution. For the 0.5N NaOH solution test, the aluminum concentration was only increased from 2520 to 3022 mg L^{-1} in the retentate solution. After NF membrane filtration, the aluminum concentrations in both retentate liquid volumes were similar.

Theoretically, the aluminum concentrations in both retentate liquids should be the same. However, due to the charge of the aluminum ions, the aluminum ions presented as Al^{3+} in the acidic solution and as $Al(OH)_4^-$ in the alkaline solution. The NF membrane typically captured high-valence ions and released low-valence ions, such that the NF membrane retained aluminum ions (Al^{3+}) in the retentate liquid and permitted aluminate ions ($Al(OH)_4^-$) to pass through to the permeate liquid. Hence, after NF membrane filtration, the aluminum concentrations in the permeate solutions of the 0.3N sulfuric acid solution and 0.5N NaOH solution were 24.4 and 836 mg L^{-1}, respectively.
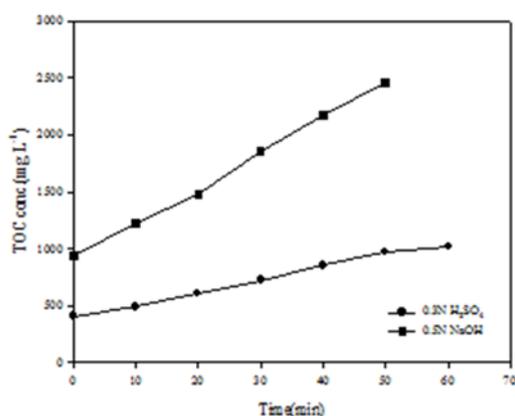

Fig. 6 Concentrations of total organic carbon in retentate solutions as a function of time
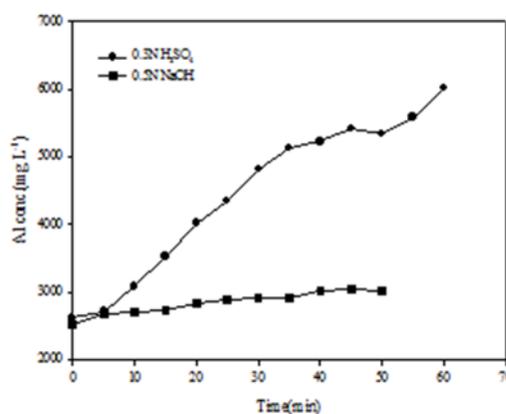
Fig. 7 Aluminum concentrations in retentate solutions as a function of time

Although aluminum was concentrated in the acidified solution, the NF membrane captured and concentrated not only high-valence aluminum ions, but also other metal ions and organic matter (Table 1). If the concentrated solution was recovered and reused as the coagulant, the amount of dissolved chemicals in the coagulant should be limited according to regulations set by the Taiwan Water Corporation. Table 2 lists the limitation of heavy metals in aluminum sulfate. The Mn concentration in the 0.3N sulfuric acid solution exceeded the allowable Mn concentration by 30 times (Tables 1 and 2). Moreover, TOC was 1016.44 mg L^{-1}. Therefore, the recovered solution was not suitable for direct use as the coagulant in drinking water treatment process. Conversely, under the alkaline conditions, almost all metal ions precipitate as hydroxide compounds. Hence, the metal concentration in the alkaline permeate solution was lower than that in the acidic permeate solution and the large molecules in the organic matter were effectively removed by the NF membrane filtration process (Table 1). Furthermore, mono-valance aluminum ion $Al(OH)_4^-$ and sodium ions penetrated the NF membrane. Hence, a sodium aluminate $NaAl(OH)_4$ solution containing a low concentration of metal ions and TOC were obtained. This relatively pure aluminate solution cannot be obtained by simply adjusting the pH of the water treatment sludge to recover aluminate.

Table 1 The metals and TOC concentrations under acidity and alkalinity conditions

	$0.3N(H_2SO_4)$ retentate solution	$0.5N(NaOH)$ permeate solution
Al	6123	836
Cd	ND	ND
Cr	0.21	ND
Pb	0.11	ND
Fe	91	ND
Mn	830	ND
Cu	0.43	ND
TOC	1016.44	2.72

Unit: mg L^{-1}

Table 2 The limitation of heavy metals in aluminum sulfate used in drinking water treatment of Taiwan, R.O.C.

| Items	Allowable chemical concentration in drinking water treatment process
As	<10ppm
Ag	—
Cd	<2ppm
Cr	<10ppm
Cu	—
Fe	<200ppm
Hg	<0.2ppm
Mn	<25ppm
Ni	—
Pb	<10ppm
Zn	—
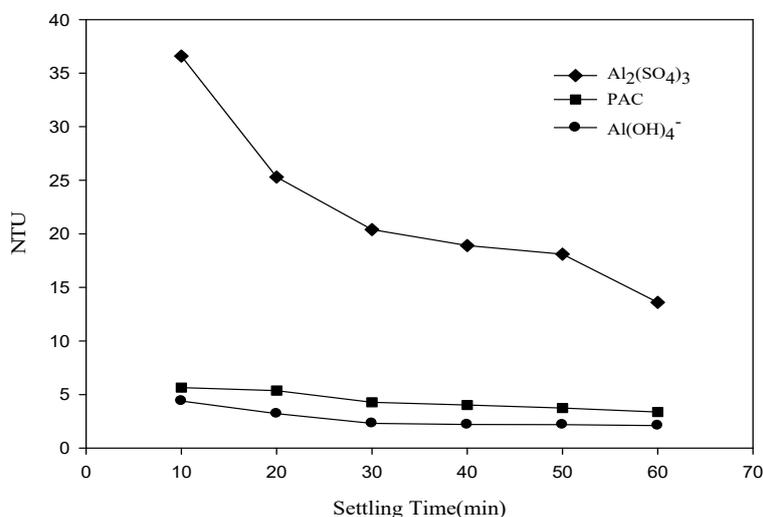
Fig. 8 Supernatant turbidity of synthesized solutions treated by $Al_2(SO_4)_3$, PACl, and $Al(OH)_4^-$

3.2 Acidic raw water treated by different coagulants

Sodium aluminate, $Na[Al(OH)_4]$, can be utilized as a coagulant in drinking water treatments as a supplement to alum and may be useful in soft, low-alkalinity water (Sahu and Chaudhari 2013). Therefore, low-alkalinity synthetic raw water containing kaolin was used to identify the influence of turbidity on removal efficiency. The synthesized raw water was prepared with a turbidity of 100 NTU and a pH of 5.0. Jar tests were conducted using the same coagulant dosage (1.6 mg L^{-1} as Al). The coagulants used for the jar tests were aluminum sulfate ($Al_2(SO_4)_3$), polyaluminum chloride (PACl) and the recovered coagulant ($Al(OH)_4^-$). After coagulation and precipitation, a water sample was taken from 3 cm below the water surface at 10-min intervals and its turbidity measured. When three sets of raw water samples were treated individually by the three coagulants, $Al_2(SO_4)_3$, PACl and $Al(OH)_4^-$, the pH values of raw water after coagulation were 4.71, 4.99, and 6.23, respectively. Experimental data indicate that the best coagulation result was when using $Al(OH)_4^-$ (Fig. 8). When aluminum sulfate ($Al_2(SO_4)_3$) was used to remove turbidity, the best pH for coagulation was 6-7 (Baghvand *et al*. 2010). In this test, after the aluminum sulfate coagulation process, solution pH reduced to 4.71, not in the suggested pH range. Hence, residual turbidity was still high even after coagulation. When using PACl as the coagulant, since PACl is a pre-polymerized coagulant, for efficient use in a wide pH range (Wu *et al*. 2009). Residual turbidity measured from PACl-treated water at pH 4.99 was lower than that measured from aluminum sulfate coagulated water. Conversely, $Al(OH)_4^-$ is an alkaline coagulant whose operational pH range is not yet documented. When $Al(OH)_4^-$ was used to treat slightly acidic raw water, it increased solution pH to a neutral range, such that the $Al(OH)_4^-$ could remove turbidity more effectively did aluminum sulfate or PACl. Hence, the lowest turbidity occurred with raw water treated with $Al(OH)_4^-$ (Fig. 8).

4. Conclusions

In this study, when sludge was pretreated with acidic or alkaline chemicals before conducting the NF membrane filtration experiment, the following findings were acquired.

1. As the NF membrane was applied to increase the concentration of aluminum (Al^{3+}) in the retentate acidic solution, the recovered aluminum solution containing excess heavy metals and organic matter is not suitable for direct using as a water treatment coagulant.

2. Due to the selective characteristics of the NF membrane, mono-valence ions easily penetrate the membrane, such that the aluminate ion ($Al(OH)_4^-$) in the alkaline solution is purified in the permeate solution.

3. The recovered $Al(OH)_4^-$ in the alkaline solution can be used as the coagulant when treating slightly acidic raw water without adding any alkalinity.

Acknowledgments

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