

Chelating surfactant for the removal of heavy metals from wastewater and surfactant recovery

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ABSTRACT

The treatment of soil leaching solutions contaminated by heavy metals is challenging because it is necessary to remove the heavy metal ions while attempting to reduce washing costs through strategies like using recycled detergent. In the present study, micellar-enhanced ultrafiltration (MEUF) was used to purify simulated wastewater containing the chelating surfactant, C₁₄-ED3A3Na, and three heavy metal ions: Cu(II), Pb(II), and Zn(II). Additionally, this research also investigated the effectiveness of using MEUF to recover the used surfactant. The results showed that increasing the membrane's molecular weight cutoff (MWCO) caused an increase in the permeate flux and a decrease in the solute removal rate. The results also showed that the surfactant concentration significantly influenced the solute removal rate. The metal ion removal rate was highest when the surfactant concentration was between 5 and 10 critical micelle concentration (CMC), with peak removal rates of 93%, 95%, and 85% for Cu(II), Pb(II), and Zn(II), respectively. A peak surfactant removal rate of 88% was achieved at a concentration of 50 CMC. The results also revealed that feed solution pH had a minimal effect on surfactant and metal ion removal. Similarly, the recovery test showed that the relationship between the surfactant recovery rate and the pH value and type of acidifier was insignificant. In contrast, the removal rate of the three metal ions increased as the pH value decreased. A pH value of 1 yielded removal rates of 68%, 76%, and 90% for Cu(II), Pb(II), and Zn(II), respectively. It is suggested that, with regards to forming a chelate with C₁₄-ED3A3Na, Cu(II) was the most effective, followed by Pb(II) and Zn(II), respectively. Ultimately, the results of this research show that C₁₄-ED3A3Na MEUF technology is an effective method for treating heavy metal ion wastewater, and that surfactant can be effectively recovered by using a concentrated acidification solution.

Keywords: Chelating surfactant; MEUF; Heavy metal; Wastewater; Surfactant recovery

1. Introduction

In many parts of the world, heavy metal levels in soil and groundwater around mine sites significantly exceed

regulated standards due to the excessive and disordered development of mineral resources. Many methods have been developed and employed to remediate heavy metal contaminated soil, for example, solidification and

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stabilization [1], chemical leaching [2,3], and bioremediation [4,5]. Of these approaches, chemical leaching has proven to be the most effective for treating the soil with high concentrations of heavy metals [2]. At present, strong acid, organic acid [6], synthetic chelating agent [7,8], synthetic surfactant [9], and biosurfactant [10,11] are the most commonly used eluents for treating contaminated soil. Unfortunately, each of these eluents has certain drawbacks. For instance, strong acid can damage the physical and chemical properties of the soil, causing it to lose nutrients. Organic acid and synthetic chelating agents can effectively remove heavy metals from the soil, but it is difficult to recover them afterward [12]. In contrast, synthetic surfactants are easily recovered, but they are inefficient with respect to soil cleaning [9]. Biosurfactants are an ideal option because they are both environmentally friendly and effective in removing heavy metals from the soil, but they are not available in large quantities [10]. In recent years, Peng and Chen [13] synthesized ethylenediamine triacetate (ED3A) chelating surfactant, which proved highly effective in cleaning heavy metal contaminated soil. Moreover, it has been demonstrated that, in addition to being effective, ED3A chelating surfactants are also environmentally friendly [14]. In selecting a surfactant for use in large-scale chemical leaching to restore heavy metal contaminated soil, it is critical to identify a cleaning surfactant that is able to minimize costs with respect to soil eluent purification and surfactant recovery.

It is well-known that ultrafiltration can remove dissolved macromolecules with a molecular weight cutoff (MWCO) of between 1,000 and 100,000 Da, while nanofiltration can separate monovalent salts and organics within a molecular weight range of 200–1,000 Da [15]. As such, ultrafiltration and nanofiltration technology have often been combined for use in advanced wastewater treatment [16,17].

Micellar-enhanced ultrafiltration (MEUF) technology is an effective, low-cost option for treating wastewater with low concentrations of heavy metals [18]. MEUF is predicated on the principle that heavy metal cations can be combined on the surface of micelles formed by anionic surfactants via electrostatic attraction that are unable to pass through the ultrafiltration membrane. As a result, most of the heavy metal ions and micelles are intercepted by the ultrafiltration membrane, thus purifying the water body. At present, MEUF technology most commonly uses synthetic surfactants, such as sodium lauryl sulfate (SDS) [19], and biosurfactants [20]. However, there are few reports on the use of chelating surfactants to treat heavy metal wastewater via MEUF. In addition to addressing this gap in the literature, the present research also investigates the effectiveness of using chelating surfactant and MEUF technology to treat wastewater with higher concentrations of heavy metals.

In this paper, the chelating surfactant, sodium N-(3-tetradecanoxo-2-hydroxypropyl) ethylenediamine triacetate (C_{14} -ED3A3Na), was added to solutions containing 50 mg L⁻¹ Cu(II), Zn(II), and Pb(II) in order to simulate wastewater containing chelating surfactant and heavy metal ions. To provide theoretical and experimental guidance for eluent purification and surfactant recovery, the use of MEUF technology for the purification of heavy metal wastewater and the use of acidification concentrate and ultrafiltration technology for the recovery of surfactant were studied.

2. Materials and methods

2.1. Materials

Concentrated sulfuric acid, concentrated hydrochloric acid, and concentrated nitric acid were purchased from Xilong Scientific Co., Ltd., China as guaranteed reagents. Other chemicals were purchased from Xilong Scientific Co., Ltd., China as analytical reagents.

The chelating surfactant, C_{14} -ED3A3Na was synthesized in our laboratory according to a previously reported method [13] (see Fig.1 for the chemical structure of C_{14} -ED3A3Na). The critical micelle concentration (CMC) for C_{14} -ED3A3Na is 5.89×10^{-4} mol L⁻¹. The ultrapure water used in the experiments was prepared using an FBZ2001-UP-P ultrapure water machine from Qingdao Fulham Technology Co., Ltd., (China). The raw solution (or feed solution) consisted of C_{14} -ED3A3Na and Cu(II), Pb(II), and Zn(II) at concentrations of 50 mg L⁻¹. The pH value of the raw solution was adjusted using a 5% sodium hydroxide solution and a 5% nitric acid solution, and was measured using a PHS-2F precision acidity meter (Shanghai Hongyi Instrument and Instrument Co., Ltd., China).

2.2. Ultrafiltration device and ultrafiltration methods

The ultrafiltration device used in this research was an MSC300 ultrafiltration cup (Shanghai Mosu Science Equipment Co., Ltd., China) with a polymethyl methacrylate body and base. The effective membrane area was 3.85×10^{-3} m². The membrane properties are presented in Table 1, and the ultrafiltration setup is illustrated in Fig. 2 [21]. Transmembrane pressure was adjusted via the total pressure valve and the partial pressure valve, and was displayed in the low voltage meter. During the ultrafiltration process, the magnetic rotor was operated at a speed of 300 r min⁻¹. The permeate solution collection and ultrafiltration time recording were conducted at the same time.

Prior to beginning the ultrafiltration experiments, the membranes were immersed in 0.5% formaldehyde solution,

Table 1
Ultrafiltration membrane properties

Material	Polyethersulfone
MWCO (kDa)	3, 5, 10, 30
Working pressure (MPa)	0.1–0.22
T_{max} (°C)	40
Company	Sepro, USA

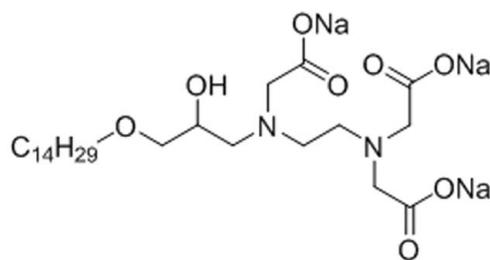


Fig. 1. Structure of chelating surfactant C_{14} -ED3A3Na.

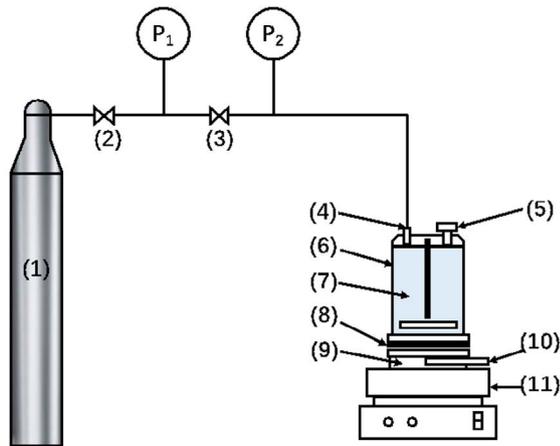


Fig. 2. Schematic diagram of ultrafiltration experimental device (1) nitrogen cylinder, (2) total pressure valve, (3) partial pressure valve, (4) intake, (5) feed inlet, (6) ultrafiltration cup, (7) rotor, (8) ultrafiltration membrane, (9) ultrafiltration cup base, (10) permeate outlet, and (11) magnetic stirrer, P_1 and P_2 are high voltage meter and low voltage meter, respectively.

and then rinsed 1–2 times with ultrapure water. After ultrafiltration, the membranes were soaked in pure water for 30 min before being successively soaked in 0.5 mol L⁻¹ citric acid and NaOH solution for 10–15 min, and then finally washed with pure water until the water was neutral.

2.3. Surfactant recovery

After adjusting the pH value of the concentrated solution (raw solution) with acidifier, the total volume was measured using a measuring cylinder. The feed solution was then poured into the ultrafiltration cup with a MWCO of 5 KDa and a transmembrane pressure of 0.2 MPa. Once the permeate solution had stopped flowing through the other end, it was collected and the retained sediment in the ultrafiltration cup was recovered.

2.4. Determination of permeate flux

Permeate flux was calculated using Eq. (1):

$$J = \frac{V_p}{\Delta t \times A} \quad (1)$$

where J represents permeate flux in L h⁻¹ m², V_p is the volume of permeate solution in L, Δt is the time of ultrafiltration in h, and A is the effective membrane area in m².

2.5. Determination of metal ion removal rate

After the raw solution (or feed solution), permeate solution, and ultra-pure water (reagent blank) had been acidified using concentrated nitric acid, an ICP-MS7800 inductively coupled plasma mass spectrometer (Agilent Science and Technology Co., Ltd., China) was used to measure their concentrations of metal ions. The removal rate of metal ions was calculated according to Eq. (2):

$$R_{M(\text{II})} = \frac{c_f - c_p}{c_f - c_0} \times 100\% \quad (2)$$

where $R_{M(\text{II})}$ denotes the removal rate of metal ions, and c_f , c_p , and c_0 denote the concentration of metal ions in the feed solution, permeate solution, and reagent blank in mol L⁻¹, respectively.

The removal rate of metal ions in the recovery test was calculated according to Eq. (3):

$$R_{r,M(\text{II})} = \frac{(c_p - c_0) \times V_p}{(c_r - c_0) \times V_r} \times 100\% \quad (3)$$

In Eq. (3), $R_{r,M(\text{II})}$ indicates the removal rate of metal ions; c_r , c_p , and c_0 represent the concentration of metal ions in the raw solution, permeate solution, and reagent blank in mol L⁻¹, respectively; and V_r and V_p are the volumes of raw solution and permeate solution in mL, respectively.

2.6. Determination of surfactant removal rate (or recovery rate)

The concentrations of chelating surfactant in the feed solution and permeated solution before and after ultrafiltration were determined via phase separation titration. The anionic chelating surfactant was titrated with cationic surfactant cetyltrimethylammonium bromide (CTMAB), which caused phase separation indicated by the bromocresol green alkaline [22]. A blank reagent (ultrapure water) was tested in order to provide a control. The ultrafiltration membrane's surfactant removal rate was calculated using Eq. (4).

$$R_s = \frac{V_{c,f} - V_{c,p}}{V_{c,f} - V_{c,0}} \times 100\% \quad (4)$$

where R_s indicates the surfactant removal rate, and $V_{c,f}$, $V_{c,p}$, and $V_{c,0}$ are the volumes (in mL) of CTMAB consumed by the titration feeding solution, permeate solution, and ultra-pure water, respectively.

In the recovery test, the surfactant recovery rate was calculated according to Eq. (5).

$$R_{r,s} = \frac{V_r \times (V_{c,r} - V_{c,0}) - V_p \times (V_{c,p} - V_{c,0})}{V_r \times (V_{c,r} - V_{c,0})} \times 100\% \quad (5)$$

where $R_{r,s}$ denotes the recovery of surfactants; $V_{c,r}$, $V_{c,p}$, and $V_{c,0}$ are the volumes (in mL) of CTMAB consumed by the titration of the raw solution, permeated solution, and ultrapure water, respectively; and V_r and V_p are the volumes (in mL) of the raw solution and permeated solution, respectively.

3. Results and discussion

3.1. Effect of MWCO on ultrafiltration

Fig. 3 shows the ultrafiltration effect of feed solution containing 10 CMC surfactants using different membrane

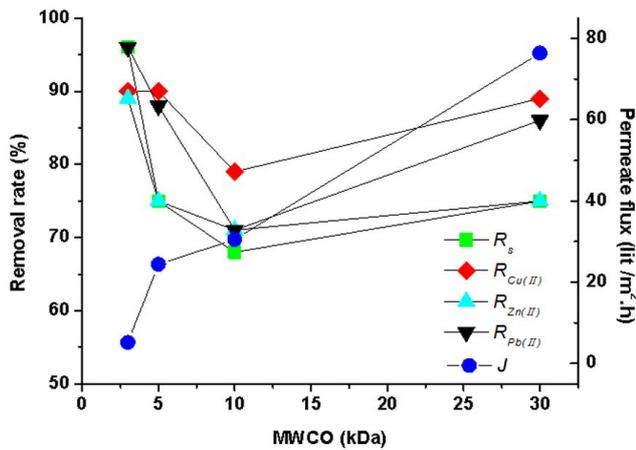


Fig. 3. Effect of MWCO on ultrafiltration.

specifications and a transmembrane pressure of 0.2 MPa. As can be seen, the permeate flux of the membrane increases alongside the MWCO. As a whole, the surfactant removal rate (R_s) and heavy metal ion removal rate ($R_{M(II)}$) all decrease as the MWCO is increased. The R_s of the 3 kDa membrane was 96%, with removal rates of 90%, 89%, and 96% for Cu(II), Zn(II), and Pb(II), respectively. The R_s decreased to 75% when the 5 kDa membrane was used, with the removal rates for Cu(II), Zn(II), and Pb(II) also decreasing to 90%, 75%, and 88%, respectively.

3.2. Effect of transmembrane pressure on ultrafiltration

Fig. 4 shows the ultrafiltration effects that were observed when a feed solution containing 10 CMC surfactant was fed through a 5 kDa membrane under different transmembrane pressures. Once again, the permeate flux increased when the transmembrane pressure was increased. On the whole, increasing the transmembrane pressure caused a slight decrease in R_s and $R_{M(II)}$ which was mainly due to the increase in the permeate flux.

3.3. Effect of surfactant concentration on ultrafiltration

These tests were conducted using a membrane with an MWCO of 5 kDa and a transmembrane pressure of 0.2 MPa. The results indicated that the removal rates for surfactant and metal ions both increased rapidly as the surfactant concentration in the feed solution increased, and then decreased slowly (Fig. 5). As shown in our previous study [23], these surfactant monomers first form insoluble chelates with free soluble heavy metal ions, which can then be solubilized in chelating surfactant micelles. If the total concentration of heavy metal ions in wastewater is 1.798 mM and the surfactant concentration is 1 CMC (0.589 mM), there will be an excess of heavy metal ions regardless of whether the complexation of ions to surfactant is 1:1 [23] or 2:1 [13]. As such, the feed solution will contain no surfactant micelles, only insoluble chelate particles, and free heavy metal ions. These conditions result in a relatively low heavy metal removal rate due to the insoluble chelate particles becoming trapped on the ultrafiltration membrane and

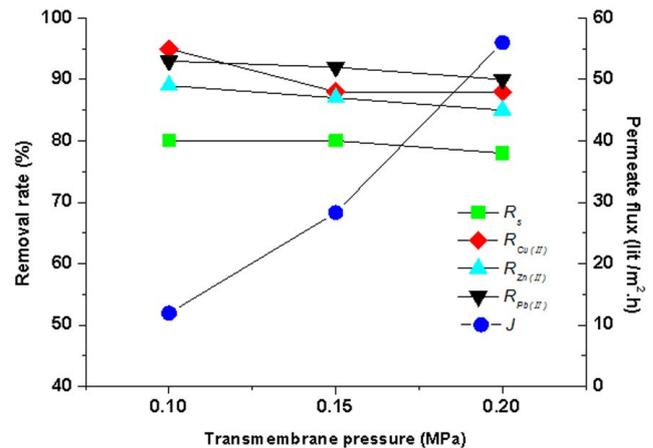


Fig. 4. Effect of transmembrane pressure on ultrafiltration.

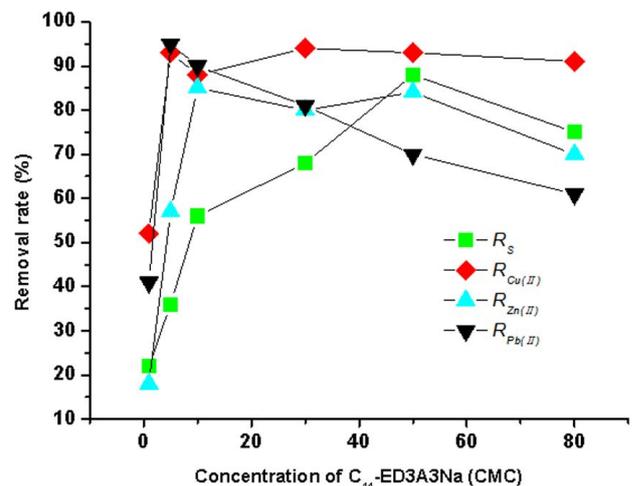


Fig. 5. Effect of surfactant concentration on ultrafiltration.

the free heavy metal ions passing through the ultrafiltration membrane. However, the removal rate of heavy metal ions increases significantly when the surfactant concentration is increased to 5 CMC (2.945 mM), as this volume of surfactant will be considerably greater compared to the concentration of heavy metal ions. The excessive surfactant in the feed liquid allows micelles to form and the insoluble chelate particles to be solubilized. Notably, only the removal rate of Zn(II) continues to increase when the surfactant concentration is increased to 10 CMC (5.89 mM), while the removal rates for Cu(II) and Pb(II) begin to decrease. Further increases in the surfactant concentration result in marginal changes in the removal rate of Cu(II) and a downward trend in the removal rates of the other two metals. This result is likely attributable to excessively high surfactant concentrations causing the micelles to deform and become smaller, thus making it easier for them to pass through the ultrafiltration membrane, which in turn results in a lower metal removal rate [24].

Moreover, the removal rate for Cu(II) and Pb(II) are significantly higher than that of Zn(II) when the surfactant

concentration is lower than 10 CMC. This result is due to the competitive chelation relationship between the three metal ions and C_{14} -ED3A3Na, with Cu(II) and Pb(II) demonstrating superior ability to form chelates with C_{14} -ED3A3Na compared to Zn(II). Similar to EDTA and phenylenediamine tetraacetic acid (PDTA) [8], the stability coefficients of three metal ions for forming a chelate with C_{14} -ED3A3Na is as follows: Cu(II) > Pb(II) > Zn(II).

The surfactant removal rate reaches its highest value (88%) when the surfactant concentration is increased to 50 CMC, and then decreases with further increases in concentration. This may be due to two factors affecting the surfactant removal rate. (1) Once reaches the surfactant concentration CMC, a dynamic equilibrium forms between the monomolecules and micelles in the solution; that is, the solution becomes a saturated solution of surfactant. If more surfactant is added to the solution, it will mainly exist in the form of micelles, which means a reduction in the proportion of single surfactant molecules in the solution. This increases the surfactant removal rate during ultrafiltration. (2) Micelles tend to deform and become smaller at higher surfactant concentrations, which allows them to pass through the ultrafiltration membrane easily, causing a decrease in the surfactant removal rate [24]. It is possible that the first factor is dominant at surfactant concentrations of 50 CMC and below, while the second factor is dominant at surfactant concentrations of greater than 50 CMC.

3.4. Effect of pH value of feed solution on ultrafiltration

Fig. 6 illustrates the ultrafiltration results for feed solutions with different pH values. These tests used solutions containing 10 CMC surfactant, a 5 KDa membrane, and a transmembrane pressure of 0.2 MPa. As can be seen, pH value had low effect on the removal rates of the three metal ions. This result is due to the anionic ED3A chelating surfactant that was used, as this surfactant can form chelates with heavy metal ions in both neutral and alkaline environments, which can then be solubilized in micelles. The removal rates for Cu(II) and Pb(II) were slightly higher when the pH value of feed solution was 9.35, with removal

rates of 97% and 94%, respectively. However, the highest removal rate for Zn(II) (88%) was obtained when the pH of the feed solution was raised to 11.2. Within the experimental pH range, the removal rates for three metal ions were as follows: $R_{Cu(II)} > R_{Pb(II)} > R_{Zn(II)}$. The lowest surfactant removal rate was obtained at a feed solution pH of 11.2. This result may be due to the use of a ED3A surfactant, which is insoluble in water when pH value of aqueous solution is 2.0 and can be soluble in alkaline aqueous solution [13]. Therefore, we have reason to speculate that when the pH value is higher the surfactant has better water solubility and larger CMC, and there are more surfactant monomers in the feed liquid, which makes the removal rate of surfactant lower.

3.5. Effect of acidifier type on recovery

Adjusting the pH value of the concentrate to 2.0 using hydrochloric acid, nitric acid, and sulfuric acid resulted in surfactant recovery rates of 83%, 77%, and 76%, respectively. The use of nitric acid as an acidifier produced the best removal efficiencies for the three metal ions, with removal rates of 57%, 97%, and 97% for Cu(II), Pb(II), and Zn(II), respectively. In contrast, the use of sulfuric acid as an acidifier resulted in the worst removal efficiencies for the three metal ions. Obviously, the reason for this disparity is that the anions of the three acidifiers are different, which may be related to the volume and oxidation of three anions.

3.6. Effect of degree of acidification on recovery

When acidifier (hydrochloric acid) is added to the concentrated solution, its pH value decreases gradually, thus changing its state. The appearance of the concentrated solution does not change significantly when the pH value is adjusted to 5 and 4; however, when the pH value is lowered to 3, the solution becomes gelatinous in texture and lighter in color. Finally, adjusting the pH of the concentrated solution to 2 and 1 causes it to precipitate and become clear.

On the whole, the surfactant recovery rate (75%–84%) does not change much as the feed solution pH decreases, as the micelle and precipitate do not pass through the ultrafiltration membrane. However, the removal rates for the three metal ions do increase as the feed solution pH decreases. Indeed, the best removal rates for the three ions were obtained at a feed solution pH of 1, with rates of 90%, 76%, and 68% for $R_{r,Zn(II)}$, $R_{r,Pb(II)}$ and $R_{r,Cu(II)}$ respectively. This shows that hydrogen protons can reduce the stability of the chelate formed by heavy metal ions and C_{14} -ED3A3Na, just as increasing acidity can reduce the chelating ability of EDTA, and it also proves that the stability coefficient (K) of the chelates formed by C_{14} -ED3A3Na and the three metal ions should be: $K_{Cu(II)-C_{14}-ED3A3Na} > K_{Pb(II)-C_{14}-ED3A3Na} > K_{Zn(II)-C_{14}-ED3A3Na}$.

4. Conclusion

The experiments presented in this paper explored the effectiveness of using MEUF technology to remove heavy metal from wastewater containing chelating surfactant C_{14} -ED3A3Na. The results showed that the permeate flux increased alongside the MWCO and transmembrane

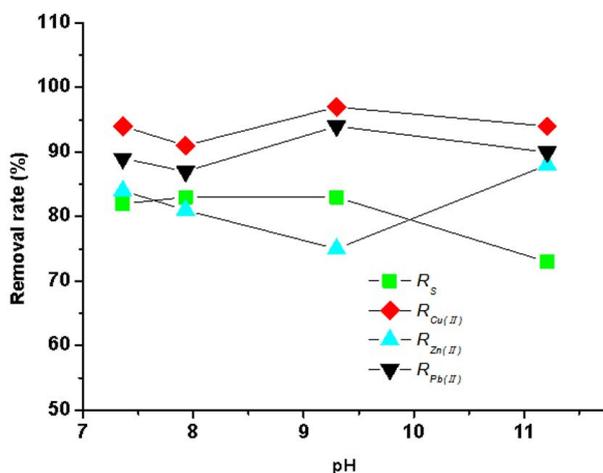


Fig. 6. Effect of pH value of feed solution on ultrafiltration.

pressure. Additionally, the results also showed that the solute removal rate decreased in response to an increase in the MWCO but had little relation to changes in the transmembrane pressure. Furthermore, we found that the concentration of surfactant significantly influenced the removal of solute. The removal rates for the metal ions were highest when the surfactant concentration was in range of 5–10 CMC, with removal rates of 93%, 95%, and 85% for Cu(II), Pb(II), and Zn(II), respectively. As for the surfactant itself, a peak removal rate of 88% was achieved when its concentration in the feed solution was 50 CMC. The pH of the feed solution was also found to have a minimal effect on the removal of surfactant and metal ions. Moreover, the recovery test showed that, in addition to having little relation to pH, the surfactant recovery rate was also not strongly affected by acidifier type. The removal rates of the three metal ions increased as the pH value decreased: when the pH value was 1, the removal rates of Cu(II), Pb(II), and Zn(II) were 68%, 76%, and 90%, respectively. Therefore, we suggest that, at the laboratory scale, C₁₄-ED3A3Na MEUF technology can be used to effectively treat wastewater polluted with heavy metal ions, and that acidification treatment can be an effective approach for recovering the surfactant from the concentrated solution. Finally, our results demonstrate that, of the three investigated metal ions, Cu(II) possesses the greatest ability to form chelate with C₁₄-ED3A3Na, followed by Pb(II) and Zn(II), respectively.

Acknowledgments

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