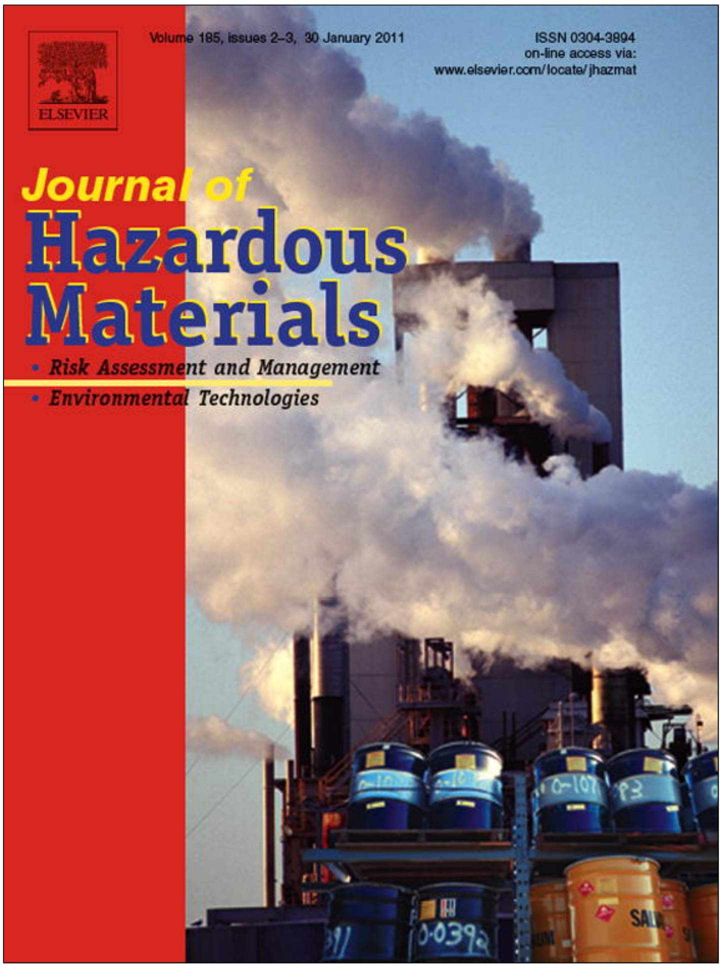
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# Micellar-enhanced ultrafiltration of cadmium and methylene blue in synthetic wastewater using SDS

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ABSTRACT

Single and simultaneous removal of $Cd^{2+}$ and methylene blue (MB) with sodium dodecyl sulfate (SDS) by micellar-enhanced ultrafiltration under different experimental conditions was investigated. In single removal process, with initial SDS concentration increasing, the removal efficiency of $Cd^{2+}$ and MB kept increasing and then decreased. When the initial concentrations of SDS and $Cd^{2+}$ were 1.0 cmc and 50 mg $L^{-1}$, respectively, the maximum removal efficiency of $Cd^{2+}$ was obtained as 99.2%. Removal efficiency of MB could achieve more than 99.9% with initial SDS concentration below 2.0 cmc. As compared with single $Cd^{2+}$ removal, the removal efficiency of $Cd^{2+}$ in the presence of MB was slightly higher with initial SDS concentration below 1.0 cmc, while decreased with the SDS concentration above 1.0 cmc. The maximum removal efficiency of $Cd^{2+}$ was 98.8% when initial concentrations of SDS and MB were 1.0 cmc and 4 mg $L^{-1}$, respectively. The removal efficiency of MB in the presence of $Cd^{2+}$ could achieve higher than 96.5%, which was only 3.4% less than the optimum result of the single removal. Meanwhile, effect of pH on removal efficiency of $Cd^{2+}$ was more significant than that of MB.

## 1. Introduction

The increasing contaminated industrial wastewater containing toxic metal ions or/and organic contaminants has caused a series of environmental problems [1,2]. Cadmium-containing wastewater has always attracted widespread attention as its significant threat to the environment and human health [3,4]. The traditional techniques for treating cadmium-containing wastewater have substantial deficiencies, such as secondary pollution of deposition, inconvenient operation, high cost, and difficulty of recycling cadmium [5–8].

Dye containing waste stream is one of the major toxic industrial waste. Methylene blue (MB) was used as a dye in paint production, wool dyeing, and microbiology, and as a sensitizer in photo-oxidation of organic pollutants [1]. The effluents containing MB are highly colored, and cause serious water pollution. The majority of technologies currently used are largely dependent on adsorption, which is inherently a slow process, and hence its performance is limited by equilibrium [9]. The flocculation treatment produces a large amount of sludge, which causes disposal problem, and thus increases the operation cost [10]. As the low biodegradability of dyes due to the complex aromatic molecular structure, conventional biological wastewater treatment processes seems not efficient enough for the treatment of dyeing waste [11]. The techniques for treating heavy metals or dye-containing wastewater continued to mature during the years. However, there are a few studies concerning the simultaneous treatment of wastewater with simultaneous occurrence of heavy metals and dyes. Fu et al. [12] found that a heavy metal precipitant, disodium N,N-bis-(dithiocarboxy) piperazine (BDP), both precipitated $Cu^{2+}$ ions and removed the dye Acid Red-73 from wastewater. For a 1:1 molar ratio of BDP/$Cu^{2+}$, 99.9% $Cu^{2+}$ ions and 73% Acid Red-73 were removed when the feed solution (50 mL) containing 50 mg $L^{-1}$ $Cu^{2+}$-50 mg $L^{-1}$ Acid Red-73, which was not shown high removal efficiency for Acid Red-73. Blanquez et al. [13] studied biodegradation of Grey Lanaset G, which consists of a mixture of metal complexed dye. Experiments were carried out in a bioreactor with retained pellets of the fungus *Trametes versicolor* that was operated under conditions of laccase production. Although decolorization was highly efficient, no direct relationship to extracellular enzyme was apparent. Moreover, the extracellular enzyme was found to be unable to degrade the dye in vitro.

Micellar-enhanced ultrafiltration (MEUF), as a viable alternative membrane technique, involves the addition of surfactant to

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contaminated aqueous solution, in order to remove metal ions and organic matters in a cost-effective manner [14]. In recent studies, almost all metal ions can be separated via MEUF method, including $Cd^{2+}$ [15,16], $Ni^{2+}$ [17], $Co^{2+}$ [18], $Cs^+$, $Sr^{2+}$, $Cr^{3+}$, $Mn^{2+}$ [19] $Pb^{2+}$, $Zn^{2+}$ [20], $Cu^{2+}$ [21], $AuCl^{4-}$ [22] and $Fe(CN)_6{}^{3-}$ [23]. In those studies, consistently, high removal efficiencies of metal ions with mostly more than 90% have been achieved. MEUF is also widely investigated for the treatment of wastewater containing MB, methyl orange, indigo carmine, amido black, titan yellow, and direct black [24–26]. When the surfactant concentration in the aqueous stream exceeds its critical micellar concentration (cmc), the surfactant monomers will assemble and aggregate to form micelles. Micelle can solubilize organic matters into its hydrophobic core or/and adsorb counter metal ions on its surface. The micellar solution is then passed through an ultrafiltration membrane with pore sizes small enough to reject micelles containing the attracted metal ions or/and solubilized organic solutes. As a result, permeate will contain little unattracted metal ions or organic molecules and surfactant monomers, which can be recycled or discarded with foam fractionation [27]. Theoretically, as mentioned above, micelles act on metal ions and organics in different locations. It is possible to obtain good removal efficiency of those two contaminants by MEUF when they co-exist in wastewater. Simultaneous removal of inorganic and organic with MEUF such as chromate and chlorinated aromatic hydrocarbons, nitrate [28], $Cu^{2+}$ and phenol [29], $Cr^{3+}$ and phenols [30], uranyl ions as well as dissolved DBP and TBP [31], were investigated by several authors.

Our primary goal is to evaluate the process performance of MEUF in the treatment of wastewater containing both metal ions ($Cd^{2+}$) and organic dye (MB). Single and simultaneous removals of $Cd^{2+}$ and MB with sodium dodecyl sulfate (SDS) by micellar-enhanced ultrafiltration under varying experimental conditions were investigated in this study. For single removal process, the effects of initial concentration of SDS and $Cd^{2+}$/MB on separation characteristics, retentate concentration of $Cd^{2+}$/MB, and permeate flux were detected and discussed. In simultaneous removal process, the influence of initial SDS level, interaction between contaminants, and pH value on separation characteristics and retentate concentrations were investigated. The differential removal efficiency in these two processes was also comparatively studied.

## 2. Materials and methods

### 2.1. Chemicals

All chemicals were of analytical agent grade. $Cd(NO_3)_2{\cdot}4H_2O$ was selected as heavy metal ions which was purchased from Shanghai Tingxin chemical factory in China. Anionic surfactant, sodium dodecyl sulfate (SDS) with a purity of 99%, was obtained from Tianjin Kermel chemical factory. Methylene blue was supplied by Tianjin DaMao Chemical Agent Company. Deionized water was used as a solvent in all runs.

### 2.2. Membrane

The hollow fiber ultrafiltration membrane used in this study was offered from Yidong Membrane Engineering Equipment Ltd., Dalian, China. The membrane material is polysulfone which is hydrophobic in nature. Its characteristic was shown in Table 1.

### 2.3. Procedure

MEUF experiments were conducted at room temperature between 20 and 25 °C. According to the experimental design, the synthetic wastewater was made by adding pre-determined amount of $Cd^{2+}$, MB and SDS into deionized water. After adequately mixed, the aqueous solution was subjected to ultrafiltration. A laboratory-scale ultrafiltration system employed was shown in Fig. 1. The transmembrane pressure (TMP) was invariably maintained at 30 kPa, while the volume of feed solution for ultrafiltration was 3.0 L through all experiments. The experiments were performed with the retentate being recycling back into the feed tank and the permeate solution reserved in the permeate tank. When the volume of the permeate stream was 2.6 L, namely the volume of the retentate stream was 0.4 L, the experiment was ended. The cmc of SDS (2.16 g/L) was obtained by conductivity measurement (not shown).

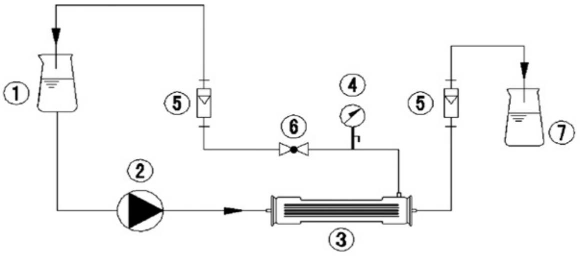

**Fig. 1.** Schematic diagram of ultrafiltration system: (1) feed tank, (2) peristaltic pump, (3) membrane, (4) manometer, (5) rotameter, (6) pressure control valve, (7) permeate tank.

After each run, the membrane was washed successively with deionized water, 0.1 mol $L^{-1}$ NaOH, deionized water, 0.1 mol $L^{-1}$ $HNO_3$ and finally with deionized water. After then, deionized water was filtered to determine the permeate flux in order to check the permeability of membrane. The permeate flux of deionized water was 16 L/h as the membrane was thoroughly washed.

### 2.4. Analysis

The concentration of MB was measured by Shimadzu UV-2550 (P/N206-55501-93) spectrophotometer at 663 nm. The surfactants (SDS) used in the work does not contain an aromatic ring and, consequently does not interfere the determination of methylene blue [1]. The concentration of $Cd^{2+}$ was analyzed by flame atomic absorption spectrophotometer (PerkinElmer).

### 2.5. Calculations

Removal efficiency of $Cd^{2+}$/MB $R$ was defined as:

$$R\ (\%) = \left(1 - \frac{c_p}{c_f}\right) \times 100\%$$

where $c_p$ is the concentration of $Cd^{2+}$ or MB in the permeate solution (mg $L^{-1}$); $c_f$ is the concentration of $Cd^{2+}$ or MB in the feed solution (mg $L^{-1}$).

## 3. Results and discussion

### 3.1. Single removal of $Cd^{2+}$

The variation of $Cd^{2+}$ removal efficiencies showed a strong concentration-dependent manner with SDS ranging between 0 and 10 cmc, as shown in Fig. 2a. In Fig. 2a, as the initial SDS concentration increased, the removal efficiencies all sharply increased near to 100%, followed by decreasing with initial $Cd^{2+}$ concentrations being 20 mg $L^{-1}$, 50 mg $L^{-1}$, 100 mg $L^{-1}$. The best removal efficiency of $Cd^{2+}$, i.e. 99.2%, was achieved with initial SDS and $Cd^{2+}$ concentrations of 1.0 cmc and 50 mg $L^{-1}$, respectively. Theoretically, there were no micelles formed at the SDS concentration below 1.0 cmc, and no $Cd^{2+}$ removal was expected. However, as shown in Fig. 2a, removal efficiency was observed when SDS concentration

**Table 1**
Characteristics of the used hollow fiber ultrafiltration membrane module.

| Type | Material | MWCO (Da) | Effective area of membrane ($m^2$) | Max operating pressure (MPa) | pH operating range | Operating temperature (°C) | Max power of pump (W) |
|---|---|---|---|---|---|---|---|
| ZU503-22 | Polysulfone | 10 K | 0.3 | 0.25 | 1–14 | 5–45 | 40 |

was below 1.0 cmc. The unanticipated rejection could be primarily attributed to the concentration polarization and direct adsorption by membrane [32]. The concentration polarization is an important characteristic of all ultrafiltration systems. It is caused by the accumulation of retained solutes or particles on the membrane surface. Some level of concentration polarization may benefit the rejection of solute. The increased concentration of the solute in the vicinity of the membrane surface has been shown to act as a “secondary membrane” and aids in rejecting solutes. The rejected SDS molecules during the process of concentration polarization gradually deposited onto the membrane surface and formed the gel layer near the cmc and the micelles formed to bind $Cd^{2+}$. These micelles provided more adsorption sites for the $Cd^{2+}$ in the initial feed solution, and then reduced the fraction of $Cd^{2+}$ passing through the membrane to the permeate side. Meanwhile, it was noted that $Cd^{2+}$ concentrations of the permeate, the retentate and the feed did not meet the mass balance. This means partial $Cd^{2+}$ were adsorbed directly on the membrane surface and in the membrane pores all over the set of experiments, which is the main reason for $Cd^{2+}$ removal in low concentration of SDS. This could also explain why above 30% of $Cd^{2+}$ was removed in absence of SDS. The removal efficiencies of $Cd^{2+}$ decreased gradually as the concentration of SDS > 2.0 cmc. Besides, SDS concentration in permeate increased significantly under these conditions. The similar conclusion has been reported in other literatures [24,33]. This could be explained by the fact that micelles near the membrane surface deformed and changed their shape due to the high SDS concentration. Consequently, some micelles passed through the membrane increasing the permeate $Cd^{2+}$ concentration. It is supposed that beyond the second cmc (about 7.0 cmc), spherical micelles associate to build so-called “rodlike” aggregates that pass through the membrane with metal ions [34–36].

The effect of initial SDS concentration on $Cd^{2+}$ concentration in the retentate during MEUF processes was shown in Fig. 2b. With the increment of initial SDS concentration, the retentate $Cd^{2+}$ concentration kept increasing linearly (approximately), and then decreased slightly above 2.0 cmc. Obviously, with the increment of the initial SDS concentration, the quantity of micelle increased, which resulted in more $Cd^{2+}$ absorbed by micelles into retentate. However, as the initial SDS concentration increased beyond 7.0 cmc, spherical micelles with metal ions associate to build so-called “rodlike” aggregates that pass through the membrane. As

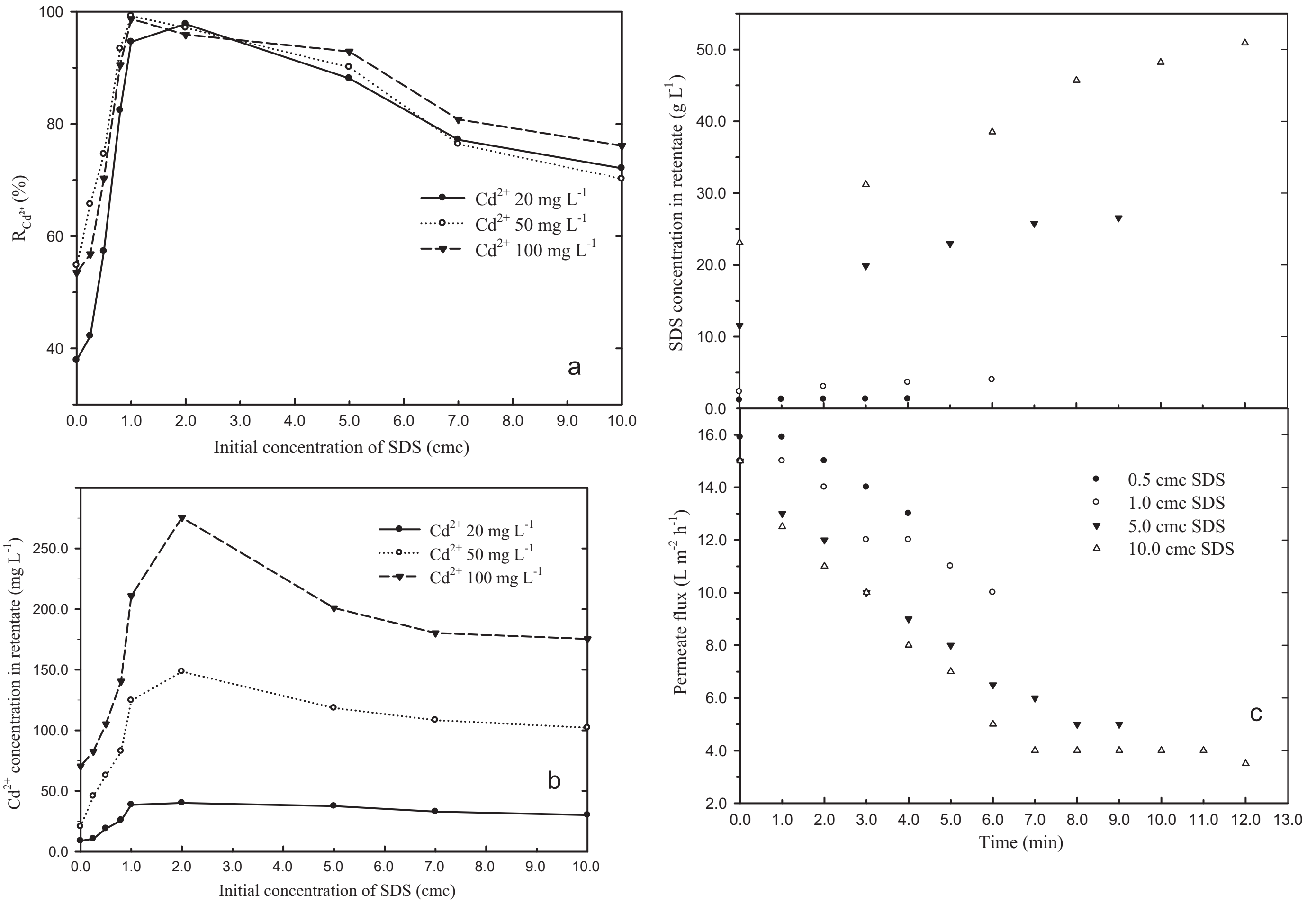

**Fig. 2.** Effects of initial SDS concentration on removal efficiency (a), retentate concentration of $Cd^{2+}$ (b), permeate flux and SDS concentration in feed tank (c) in single removal process.

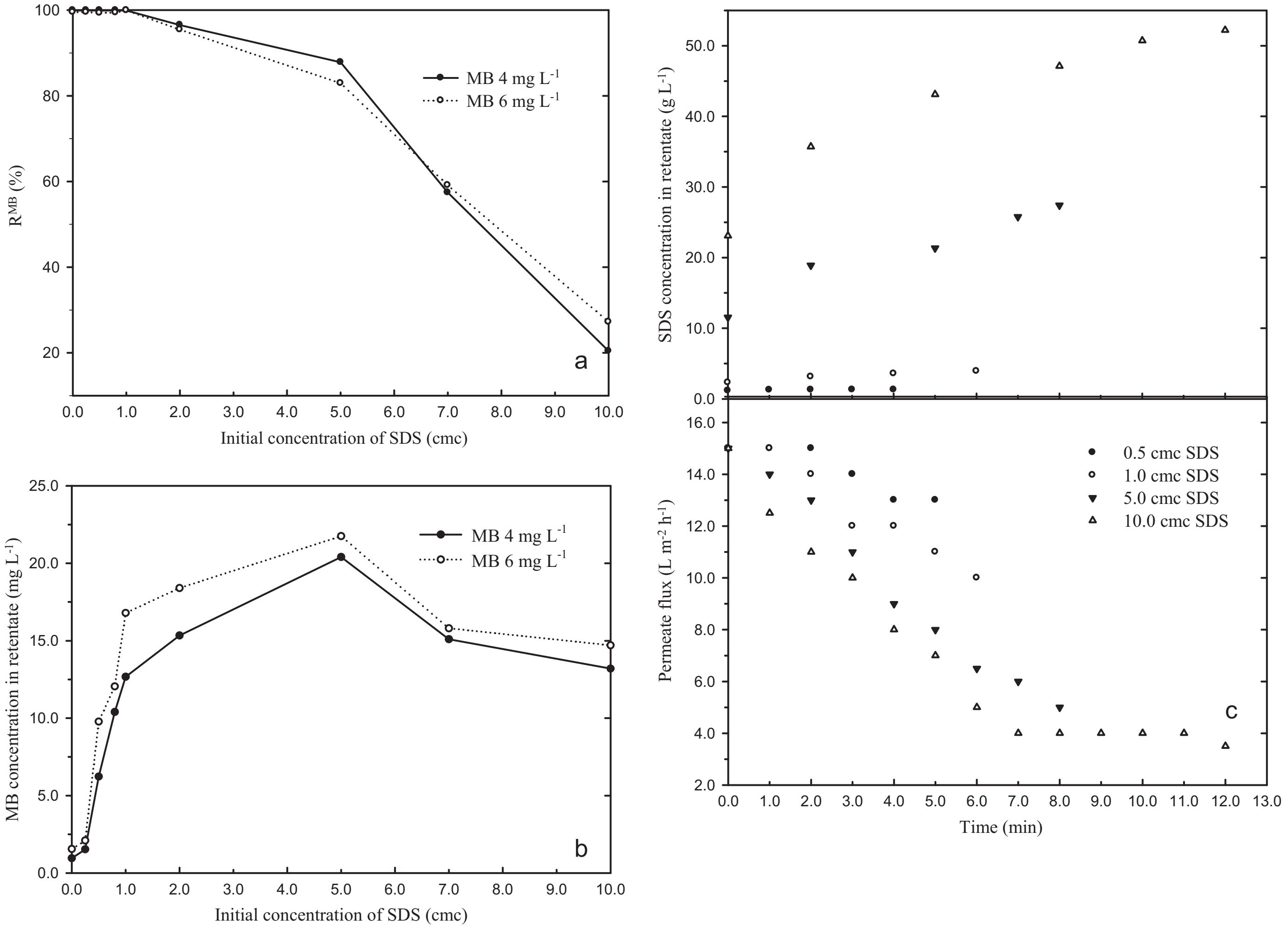

**Fig. 3.** Effects of initial SDS concentration on removal efficiency (a), retentate concentration of MB (b), permeate flux and SDS concentration in feed tank (c) in single removal process.

a result of it, amount of $Cd^{2+}$ in permeate increased, and that in retentate reduced as well as the removal efficiency.

Time evolutions of permeate flux and SDS concentrations in the feed tank were shown in Fig. 2c. The initial concentration of $Cd^{2+}$ was fixed at 50 mg $L^{-1}$ with varying SDS being supplied at 0.5, 1.0, 5.0 and 10.0 cmc. In Fig. 2c, the higher initial concentration of SDS was, the faster permeate flux decreased. As final volume of permeate fixed, more time was spent at higher initial SDS concentration. The permeate flux decreased remarkably as the process, first quickly, then slowly. This behavior is attributed to the concentration polarization, namely SDS micelles deposits quickly on the membrane surface in short time leading to mass transfer resistance increase. In addition, the retentate stream was recycled into the feed tank, the feed volume decreased and SDS concentrations increased continuously, more and more SDS micelles deposit on the membrane surface and block in the membrane pores, so that membrane fouling assembled into stable cake formation along the time. As a result, SDS concentration in feed tank increased more and more slowly and tended to be constant, as well as permeate flux.

### *3.2. Single removal of methylene blue*

The variation of removal efficiency and retentate concentration of MB with initial concentration of SDS were shown in Fig. 3a and b, respectively. As shown in Fig. 3a, the removal efficiencies achieved more than 97.0% with the SDS initial below 2.0 cmc followed by decreasing in a linear pattern. The highest removal efficiency approached more than 99.9%, while the lowest was only 20.4%. After passed through the membrane, the efflux was observed to become colorless completely when initial SDS below 2.0 cmc. However, it is observed from Fig. 3b that MB concentration in the retentate was low at SDS concentration less than 1.0 cmc. This can be attributable to the partial adsorption of MB on membrane surface or in the membrane pores during MEUF as well as those rejected into retentate stream with SDS micelles. It is obvious that the less SDS used, the more MB adsorbed on polysulfone membrane as reflected by the rapid presence of blue color on the polysulfone membrane within several minutes that was difficult to clean up, indicating serious staining occurred. With the increment of SDS micelles, more MB molecules could be adsorbed on the micelles and rejected into retentate, which reduced the membrane staining and improved the performance of MEUF greatly. Nevertheless, removal efficiency decreased from 87.8% to 20.4% along with the initial SDS concentration ranging from 5.0 up to 10.0 cmc. As mentioned above, high concentration of SDS could induce deformation of micelles near the membrane surface and facilitate micelles with MB molecules passing through the membrane.

Time evolutions of permeate flux and SDS concentrations in the feed tank were shown in Fig. 3c. The initial concentration of MB was fixed at 6 mg $L^{-1}$ while initial SDS varied to be 0.5, 1.0, 5.0 and 10.0 cmc. The permeate flux and SDS concentrations in the feed tank were observed to change in similar pattern as that of $Cd^{2+}$. It

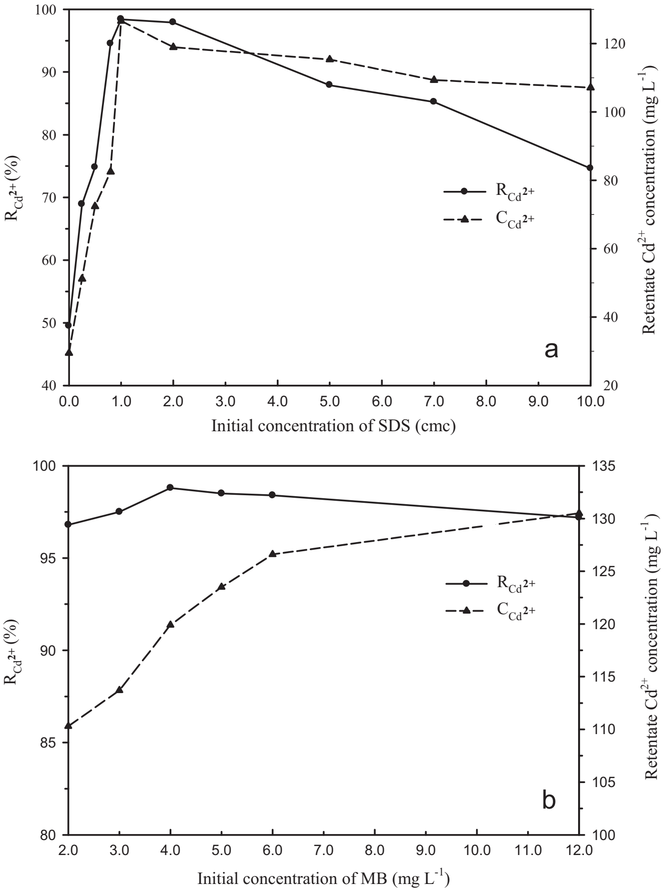

**Fig. 4.** Effects of initial SDS (a) and MB (b) concentrations on removal efficiency and retentate concentration of $Cd^{2+}$ in simultaneous removal process.

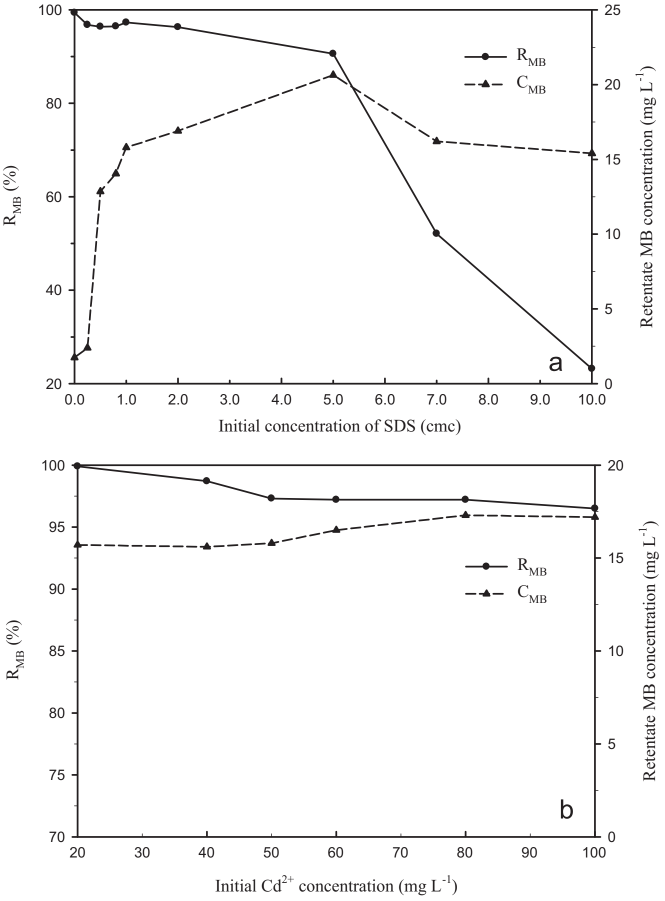

**Fig. 5.** Effects of initial SDS (a) and $Cd^{2+}$ (b) concentrations on removal efficiency and retentate concentration of MB in simultaneous removal process.

was shown that the limiting factor of permeate flux was the surfactant concentration. The influence of pollutant was negligible in our experiments.

### *3.3. Simultaneous removal of metal ions and methylene blue*

#### *3.3.1. Removal of $Cd^{2+}$ in presence of methylene blue*

Effects of the initial SDS and MB concentrations on the removal efficiency and retentate concentration of $Cd^{2+}$ in the presence of MB were investigated in Fig. 4. The initial concentrations of $Cd^{2+}$ and MB were fixed at 50 mg $L^{-1}$ and 6 mg $L^{-1}$, respectively, and the initial concentration of SDS varied to 0, 0.25, 0.5, 0.8, 1.0, 2.0, 5.0, 7.0, 10.0 cmc (Fig. 4a).

Removal efficiency and retentate concentration of $Cd^{2+}$ kept increasing and then decreased with the initial concentration of SDS increasing, as shown in Fig. 4a. The highest removal efficiency of $Cd^{2+}$ in the presence of MB was 98.4% with SDS concentration being 1.0 cmc, which was a little higher as compared with that in single removal run at SDS initial concentration below 1.0 cmc, but lower at initial SDS concentration above 1.0 cmc. This may be explained by the fact that the addition of organic solute may reduce the cmc of the surfactant since a large fraction of the surfactant will reside in micellar forms than in the absence of the organic [28]. When the cmc was reduced, more SDS molecules became available in the micellar form to attract $Cd^{2+}$ [29]. Therefore, it is not necessary to increase the initial SDS concentration excessively considering the removal efficiency and economic feasibility.

When the initial concentrations of $Cd^{2+}$ and SDS were 50 mg $L^{-1}$ and 1.0 cmc, respectively, removal efficiency of $Cd^{2+}$ was around 96.8–98.8% with the initial concentration of MB increasing from 2.0 mg $L^{-1}$ to 12.0 mg $L^{-1}$, while no significant difference was observed (Fig. 4b). However, as the initial concentration of MB increasing, retentate $Cd^{2+}$ concentration increased significantly even though the difference of removal efficiency of $Cd^{2+}$ was not remarkable. As mentioned above, the addition of organic solute may reduce the cmc, and the aggregation number and size of micelle increase. As a consequence, more SDS micelles could go to the retentate. On the other hand, adsorption of hydrophilic MB on membrane surface was much easier than that of $Cd^{2+}$. With the increased of MB concentration, more dissociative MB molecules could be adsorbed on membrane surface to inhibit the adsorption of $Cd^{2+}$. However, this influence was slight as the initial concentration of MB above 6.0 mg $L^{-1}$. Therefore, it may be concluded that moderate MB even was beneficial to $Cd^{2+}$ removal.

#### *3.3.2. Removal of methylene blue in presence of $Cd^{2+}$*

Fig. 5a shows the variation of MB removal efficiency and retentate concentration with the initial SDS concentration ranging from 0 up to 10 cmc in presence of $Cd^{2+}$. The initial concentrations of $Cd^{2+}$ and MB were fixed at 50 mg $L^{-1}$ and 6 mg $L^{-1}$, respectively. As shown in Fig. 5a, the removal efficiencies were able to achieve higher than 96.5%, at the SDS initial below 2.0 cmc and then decreased approximately linearly to 23.2%, which was similar to the results in Section 3.1.

Fig. 5b illustrated the effect of initial $Cd^{2+}$ concentration on removal efficiency and retentate concentration of MB in presence of $Cd^{2+}$. The initial concentrations of MB and SDS were fixed at 6 mg $L^{-1}$ and 1.0 cmc, respectively, and the initial concentration of $Cd^{2+}$ varied to be 20, 40, 50, 60, 80 and 100 mg $L^{-1}$. As observed from Fig. 5b, the removal efficiencies of MB were all above 96.5%,

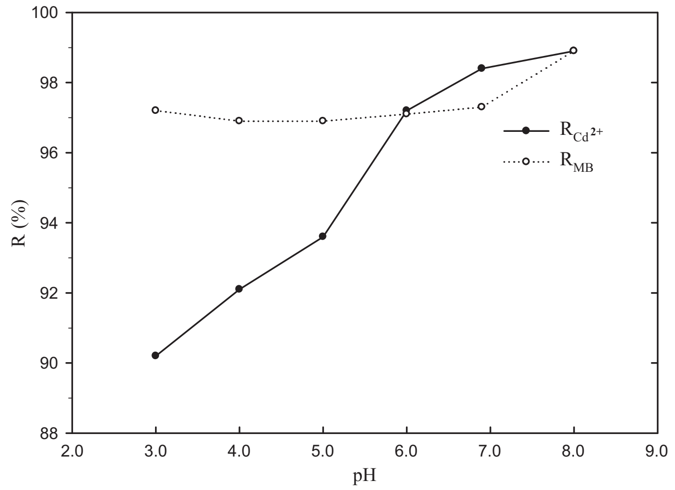

**Fig. 6.** Effect of pH values on removal efficiency of MB and $Cd^{2+}$ in simultaneous removal process.

which was only 3.4% less than the highest removal efficiency in the single process. Theoretically, due to different forms of aggregates, organic solutes can be solubilized in different locations in the micelles. Micelles of anion surfactant bind metal ions on the opposite-charged surface by electrostatic interaction [30,31,33]. Whereas it seems that hydrophobic interactions contribute to the binding between MB and SDS micelles. Large polar molecules, such as MB, are believed to be solubilized mainly between the individual molecules of surfactant in the palisade layer with the polar groups of the solubilizate oriented toward the polar groups of the surfactants and the nonpolar portions oriented toward the interior of the micelle [24]. Therefore, the competition between $Cd^{2+}$ and MB could be ignored. Therefore, simultaneous removal MB and $Cd^{2+}$ with no interference is feasible to simplify the process. According to Misra et al. [31] the removal of the DBP and TBP was not influenced by the presence of uranyl ions. Similarly, Baek and Yang [28] reported that the presence of organic solutes such as 1-chlorobenzene and 1,2-dichlorobenzene did not inhibit the removal of chromate with CPC.

### 3.3.3. *Effect of pH value on removal efficiency of $Cd^{2+}$ and MB*

Fig. 6 illustrates the effect of pH on the removal efficiency of MB and $Cd^{2+}$ at the SDS concentration of 1.0 cmc, MB concentration of 6 mg $L^{-1}$ and $Cd^{2+}$ concentration of 50 mg $L^{-1}$.

The initial pH value in the range of 3.0 and 9.0 was controlled by the addition of dilute nitric acid or sodium hydroxide solutions. The pH of feed solution before adjusting was 6.9. As shown in Fig. 6, the $Cd^{2+}$ removal efficiency increased sharply with the increment of pH. This is due to the competition of $H^+$ trapped on the surface of the micelles with $Cd^{2+}$. $H^+$ can be bound to SDS micelles with positive charge and occupies the binding sites. At lower pH, more $H^+$ in the solution corresponds to comparatively lower removal efficiency. With the pH of the solution increasing, the decreased amount of $H^+$ in the solution [37] could result in a dramatic increment of the removal efficiency of $Cd^{2+}$. By contrast, the removal efficiency of MB was around 97.0% in the whole range of pH, indicating the lower insensitivity of MB removal to pH compared with cations.

## 4. Conclusions

Single and simultaneous removal of $Cd^{2+}$ and MB were investigated using an anion surfactant, i.e. SDS. In single removal process, with the increase of the initial SDS concentration, removal efficiency of $Cd^{2+}$ and MB kept increasing and then decreased. The maximum removal efficiency of $Cd^{2+}$ was obtained as 99.2% and that of MB could reach more than 99.9% with initial SDS concentration below 2.0 cmc. In the single system of MB, it was shown that more serious membrane staining occurred by using less SDS due to competitive adsorption between polysulfone membrane and SDS. As compared with single removal, the removal efficiency of $Cd^{2+}$ in the presence of MB was slightly higher when the SDS initial concentration below 1.0 cmc, while the reverse trend was obtained at initial SDS concentration above 1.0 cmc. With the initial concentration of MB increasing, removal efficiency of $Cd^{2+}$ was between 96.8 and 98.8% with no significant difference. The removal efficiencies of MB in the presence of $Cd^{2+}$ could achieve higher than 96.5%, which indicated there were no significant interrelations between $Cd^{2+}$ and methylene blue with regard to their separation effects. With the increase of pH, removal efficiency of $Cd^{2+}$ increased sharply, while that of MB changed slightly.

These results demonstrate the potential of the MEUF for simultaneous removal of heavy metals such as cadmium ions and organic pollutants such as methylene blue and provide the scientific and technical basis for the practical application of the simultaneous removal by MEUF technique.

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