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Simultaneous removal of cadmium ions and phenol with MEUF using SDS and mixed surfactants

Xue Li, Guang-Ming Zeng *, Jin-Hui Huang, Dong-Mei Zhang, Liang-Jing Shi, Song-Bao He, Min Ruan

College of Environmental Science and Engineering, Hunan University, Changsha, 410082, China
Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha, 410082, China

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ABSTRACT

Micellar-enhanced ultrafiltration (MEUF) is a promising technology to remove metal ions and organic contaminants simultaneously from wastewater. A laboratory experiment was conducted to evaluate the efficiency of a MEUF operation for the removal of Cd^{2+} and phenol using pure SDS and mixed surfactants (Triton X-100/SDS). In pure SDS system, with the increase of the feed SDS concentration, a significant rise in Cd^{2+} rejection was obtained, which peaked at 97.0% with initial SDS concentration being 8.0 mM. Nevertheless, the phenol rejections only kept moderately increasing from 14.5% to 40.0%. In mixed Triton X-100/SDS system, the rejections of Cd^{2+} and phenol were both enhanced by the moderate addition of nonionic surfactant for the same total feed surfactant concentration as the pure SDS system. With the increase of the molar ratios of Triton X-100 to SDS (α), Cd^{2+} rejection increased slightly with α ranging from 0 to 0.8, and peaked at 91.3%. And the phenol rejection kept increasing from 27.7% to 42.4% when α was less than 1.0 followed by slight decrease to 40.2% at $\alpha = 1.5$. Moreover, the SDS dosage and the surfactant(s) concentration in permeate were reduced efficiently. The permeate flux of MEUF with mixed surfactants was lower than that with pure SDS.

1. Introduction

Micellar-enhanced ultrafiltration (MEUF) is a surfactant-based separation technique which has been employed to remove dissolved organics or multivalent ions from water in the past decade [1–5]. Upon a surfactant being added into the polluted aqueous phase, it aggregates and forms micelles at a concentration higher than its critical micellar concentration (CMC). Micelle can facilitate the solubilization of organic matters and integrates them into its hydrophobic core or/and adsorbs 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 contaminants [6]. In recent studies, almost all metal ions can be separated via MEUF method, including Cd^{2+} [7], Co^{2+} [8], Ni^{2+} [9], Mn^{2+}, Sr^{2+}, Cs^{+} [10], Cr^{3+} [11], Zn^{2+} [12], Pd^{2+} [4], Cu^{2+} [13], $AuCl^{4-}$ [14] and $Fe(CN)_6^{3-}$ [15]. In those studies, consistently, high removal efficiencies of metal ions with mostly more than 90% have been achieved. Besides, many scientists have studied the MEUF of organic matters in aqueous streams [2,16–19].

MEUF is a viable alternative technique which is economical and effective for the cleanup of dissolved contaminants from wastewater compared with conventional techniques since membranes can be added as a retrofit of existing plants. However, previous studies on the removal of dissolved contaminants by MEUF were mainly based on the application in single systems containing either metal ions or organic solutes. In fact, the wastewater from a number of industrial operations (including coal refining, textiles, dyes, and synfuel processing) contains unacceptable concentrations of both dissolved organics and multivalent ions (e.g., heavy metals). Theoretically, as mentioned above, micelles act on metal ions and organics in different locations. It is possible to obtain good removal efficiency of these two types of contaminants by MEUF when they co-exist in wastewater. Simultaneous removal of organic and metal ions with MEUF such as chromate and chlorinated aromatic hydrocarbons, nitrate [20], Cu^{2+} and phenol [21], Cr^{3+} and phenols [22], Cd^{2+} and methylene blue [23], uranyl ions as well as dissolved DBP and TBP [24], were investigated by several authors. Most of them focused on how to obtain high rejection for the removal of dissolved contaminants by MEUF, not including residual surfactant in permeate, which may make the process effluent stream environmentally unacceptable yet [11,25].

* Corresponding author at: College of Environmental Science and Engineering, Hunan University, Changsha 410082, China. Tel.: +86 731 88822754; fax: +86 731 88823701.
E-mail address: zgming@hnu.cn (G.-M. Zeng).

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Accordingly, it is highly desirable to use a surfactant system having a low CMC in order to reduce the surfactant concentration in permeate. Previous literatures [7,11,26] have shown addition of small amount of nonionic surfactant to an anionic surfactant, which usually results in a decrease in the CMC of the anionic–nonionic system compared with the CMC of the pure anionic.

This study aimed at investigating the dependence of rejection and flux on feed SDS concentration and/or the molar ratio of Triton X-100 to SDS, as well as optimizing this specific molar ratio. The differential rejection of contaminations and surfactants as well as flux in pure SDS system and anionic–nonionic surfactants system were also comparatively studied. Our primary goal is to experimentally test the ability of MEUF to simultaneously remove phenol and a divalent metal Cd^{2+} from water with anionic–nonionic surfactant and reduce the residual surfactant in the permeate.

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. Phenol was supplied by Tianjin Fuchen chemical reagent factory in China. Sodium dodecyl sulfate (SDS) with a purity of 99%, was obtained from Tianjin Kermel chemical factory. Triton X-100 was supplied by Wako Pure chemical industries company in Japan. Their chemical characteristics were shown in Table **1**. Distilled water was used as a solvent in all experiments.

2.2. Membranes

The ultrafiltration experiments were carried out in a cross-flow ultrafiltration unit. The hollow fiber ultrafiltration membrane was used, which 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 2.

2.3. Procedure

$Cd(NO_3)_2{\cdot}4H_2O$ and phenol were added into the deionized water to produce the synthetic wastewater with a Cd^{2+} concentration of 0.45 mM, and a phenol concentration of 1.06 mM, respectively. Then the surfactant with its concentration pre-determined was added into the synthetic wastewater. After adequately mixed, the aqueous solution was subjected to the ultrafiltration process. The ultrafiltration experiments were conducted at room temperature (around 20 °C). All of the experiments were run under neutral pH. 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 recycled back into the feed tank, and the permeate solution was 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.

After each run, the membrane had to be thoroughly washed to recover its permeability. First, tap water without pressure was used to rinse out the residual synthetic wastewater for 10 min. Then, it was washed with 0.1 mol L^{-1} NaOH, and 0.1 mol L^{-1} HNO_3 at 30 KPa for 10 min, respectively. Subsequently, the ultrapure water of 45 °C was recycled at 30 KPa for 20 min. Finally, 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^{-1} as the membrane was thoroughly washed.

2.4. Mechanisms

Fig. 1 shows the schematic diagram of MEUF technique using SDS which can be used to remove Cd^{2+} and phenol simultaneously. The dominant mechanisms for the rejection of metal ions and organic solutes were different. Due to the different forms of aggregates, organic solutes can be solubilized in different locations in the micelles. The nonionic organic solute will tend to solubilize in a hydrophobic core of micelles by ion–dipole interactions and the divalent cationic metal will bind or adsorb on the surface of the oppositely charged micelle [6,22,24].

2.5. Analysis

The concentration of SDS was measured by the methylene blue spectrophotometric method (ISO-7875-1-1996) with Shimadzu UV-2550 (P/N206-55501-93) spectrophotometer from Japan [5]. The concentration of Triton X-100 was measured by UV absorption at 274 nm with a UV-2550 spectrophotometer (Shimadzu) [27]. The concentration of Cd^{2+} was measured by flame atomic absorption spectrophotometer (PerkinElmer) [28]. The concentration of phenol was determined by HPLC and measured spectrophotometrically at 270 nm [19].

3. Results and discussion

Rejection of Cd^{2+}/phenol and surfactant R was defined as:

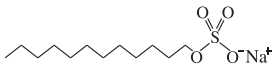
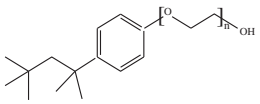
$$R(\%) = \left(1 - \frac{c_p}{c_f}\right) \times 100\% \quad (1)$$

where c_p is the solute concentration in the permeate and c_f is the solute concentration in the feed solution, respectively.

The fresh membranes were compacted at 30 kPa for 10 min using deionized water before experiments conducted. Their permeabilities were measured using deionized water at various pressures. When deionized water is filtered, the permeate flux follows the equation:

$$J_w = \frac{\Delta p}{\mu_w R_m} \quad (2)$$

Table 1
Properties of the surfactants used in this work.

Surfactant	Formula	Molecular structure	Molecular weight (g/mol)	Type	CMC (mM)
SDS	$C_{12}H_{25}OSO_3Na$		288.38	Anionic	8.0
Triton X-100	$(C_2H_4O)_nC_{14}H_{22}O$		646.86	Nonionic	0.25