



Removal of nickel ions from aqueous solution by micellar-enhanced ultrafiltration, using mixed anionic–non-ionic surfactants



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ABSTRACT

The aim of this study was to investigate the performance of single and mixed surfactants, both non-ionic and anionic, for the removal of nickel from aqueous solution using micellar-enhanced ultrafiltration (MEUF). To achieve this objective, SDS and four non-ionic surfactants with different ethylene oxide (EO) group length were utilized in this work. Furthermore, the synergistic interaction of non-ionic surfactants with SDS micelles was evaluated using β interaction parameter for TX100. The synergistic behavior of non-ionic surfactants in the SDS micelles was demonstrated, in view of the fact that the interaction parameter had a negative value. In this work, the effects of parameters such as SDS and nickel concentration, pressure and pH on the nickel rejection rate were examined in VCF = 1.7 at 25 °C. The optimum SDS and nickel concentration were determined 8 mM and 10 ppm (rejection was about 100%), respectively. It was also concluded that in the non-ionic surfactant and SDS system, the nickel rejection rate was considerably enhanced from 56% to 98%, 99%, 96% and 95%, using TX114, TX100, Brij35 and TX405, respectively for a particular concentration of SDS (i.e., 3.2 mM). Furthermore, the molar mass concentration and EO group length of non-ionic surfactants in the MEUF had a considerable effect on the performance; i.e., the surfactants with lower molar mass would lead to a higher flux rate and retention.

The findings of this study also revealed that as the SDS concentration was increased, the mean micelle size in the SDS and nickel solutions would also decrease accordingly.

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1. Introduction

Micellar-enhanced ultrafiltration (MEUF) is a relatively less energy intensive and cheaper process than traditional separation techniques such as distillation, evaporation, or distillation followed by extraction [1]. MEUF is an effective separation technique for removing metal ions from aqueous environments. In this process, surfactants can be used as an additive substance for separation of metal ions and other toxic substances from water and wastewater.

Industrial wastewater very often contains nicks because it is employed in a large number of industries such as electroplating, batteries manufacturing, mining, metal finishing and forging. Nickel and its compounds are carcinogenic and may pose danger to human health and other life forms [2]. Therefore, the concentra-

tion of heavy metals in wastewater should meet the applicable water quality criteria [2].

Several methods for nickel recovery are available, namely adsorption [3], precipitation [4], membrane filtration processes such as microfiltration [5], reverse osmosis, ultrafiltration [6,7] and recently nanofiltration [8]. As the process cost-effectiveness is of the main concern; therefore, a lot of attention has been recently paid to the membrane processes and in particular MEUF.

Micellar enhanced ultrafiltration has been widely used for the separation of low molecular weight substances using surfactant at or above of the critical micelle concentration (CMC) and in particular for the removal of copper, chromate, zinc, nickel, cadmium, selenium and arsenate [9]. Yurlova and co-workers [10] studied the effect of three different polymeric membranes (OPMN-K, UPM-10, and UPM-20) on the removal of nickel. They reported that at concentration below CMC value of sodium dodecyl sulfate (SDS), the addition of OP-10 (as a non-ionic surfactant) would enhance the removal efficiency of nickel from 88% to 96% by the UPM-20.

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However, with exceeding the CMC limit for SDS, no significant changes for the removal of nickel was observed [10]. Samper et al. [11] also reported that a reduction of heavy metals (Cd^{2+} , Cu^{2+} , Ni^{2+} , Pb^{2+} , Zn^{2+}) was observed in case where NaCl was added to the SDS and linear alkyl benzene sulfonate (LAS) solutions [11]. Samper et al. [2] have also investigated the removal of nickel ions using SDS and LAS. They concluded that as the surfactant concentration increases beyond the CMC, nickel rejection with SDS would slightly be higher than with LAS. Danis and Aydinler [12] investigated the MEUF performance and its fouling mechanisms when nickel was removed from contaminated waters. They reported that sodium lauryl ether sulfate (SLES) could be utilized for removing the nickel ion. The rejection of 98.6% for nickel was obtained at 250 kPa. They also observed that increasing the pressure would enhance the nickel and SLES rejection, but causing a reduction in the permeate flux. It has also been found that the fouling strongly depends on the formation of gel layer [12]. Some researchers removed nickel in the presence of other metal ions. Karate and co-workers removed Ni^{2+} and Co^{2+} from synthetic wastewater simultaneously using polysulfone ultrafiltration membrane and SDS as a surfactant by the MEUF process [1]. They concluded that a rejection of 99% and higher would be obtained for the Ni^{2+} and Co^{2+} when the inlet flow rate was 150 mL/min in the case where the concentration was 1 mM for Ni^{2+} and Co^{2+} . They reported that even though the presence of electrolyte decreases the efficiency of MEUF; however, considerable rejection would be achieved even in the presence of salt. Channarong et al. [13] concluded that no significant changes was observed for the removal of Zn and Ni in the MEUF process and in the presence of both elements. Monem El Zef-tawy et al. [14] used the rhamnolipid biosurfactant in MEUF for the removal of copper, zinc, nickel, lead and cadmium from contaminated waters using two membranes with molecular weight cut off (MWCO) of 10 and 30 kDa, respectively. In this work, the effect of different key operating parameters on the MEUF performance was also investigated. Furthermore, the optimal conditions were successfully applied to treat six contaminated wastewaters from the metal refining industries using two membranes [14]. Landaburu-Aguirre et al. [15] also utilized MEUF to remove heavy metals from phosphorous rich drainage waters of a fertilizer company. It was also concluded that the rejection rate of zinc and nickel would cause the copper to have the lowest tendency in removing the heavy metals. For this work, the order of ion removal by the MEUF could be inscribed as $\text{Zn} = \text{Ni} > \text{Cd} > \text{Cu}$.

Mixtures of anionic and non-ionic surfactants and their synergistic interactions are valuable parameters in industrial applications since it could reduce the amount of anionic surfactant [16,17] and consequently reduce the cost and environmental concerns. To study the measure of interactions, based on regular solution approach and theoretical methods, CMCs values were used to calculate the interaction parameter (β) [18]. This parameter gives information about the interaction of surfactants in the mixed solution compared to the single solution [17]. But description of the non-ideal behavior of surfactant mixtures by β interaction parameter is a limitation in this measurement. Three cases for β interaction parameter are available: $\beta < 0$ elucidates the synergistic interaction behavior of mixed micelle formation, $\beta > 0$ is an indication of antagonism and $\beta = 0$ shows the ideal formation of mixed micelles [17] where β could be evaluated by the correlation presented by Clint [18] and Rubingh [19].

In view of the fact that anionic surfactants have a relatively high CMC, large quantities of surfactant must be used to achieve an effective separation. Therefore, a non-ionic surfactant could be used to lower the concentration of the anionic surfactant. Few studies has so far been conducted with non-ionic surfactants and anionic surfactant in order to remove the nickel from the wastewater as a high risk contaminant [10]. However, some researchers

used OP-10 as non-ionic surfactants in order to remove the nickel from the aqueous solution [10].

Therefore, in this work, we had a comprehensive study on the removal of nickel ions by MEUF process using mixed SDS and four non-ionic surfactants (i.e. TX100, 114, 405 and Brij35). The hydrophilic group (ethylene oxide or EO) lengths of selected non-ionic surfactants are different, significantly, and we investigated its effect on the MEUF performance. In addition, the synergistic interaction of non-ionic surfactants with SDS micelles was evaluated using β interaction parameter for TX100. In this regard, the nickel rejection rate and the permeate flux have examined, using non-ionic surfactants/SDS. Furthermore, the effects of several parameters including SDS concentration, nickel concentration, pressure and pH on the nickel rejection and the permeate flux in a single SDS system have also been investigated, using a UFX5 membrane. Moreover, the mean micelle size in the nickel/SDS system and for two different SDS concentrations was also assessed.

2. Materials and methods

2.1. Surfactants and membranes

In this work, SDS as an anionic surfactant was used. The polyethylene glycol lauryl ether (Brij35) and polyethylene glycol tert-octyl phenyl ether (i.e., Triton series containing X100, X114 and X405) were utilized in this work as non-ionic surfactants. The properties of all used surfactants are shown in Table 1. All of the non-ionic surfactants are different in the ethylene oxide group number. The ethylene oxide number of these non-ionic surfactants (i.e., EON) are not similar, TX114 (EON = 7–8) < TX100 (EON = 9–10) < Brij35 (EON = 23) < TX405 (EON = 40). All surfactants and $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ were supplied from Sigma Aldrich and J.T. Baker Company, respectively. In order to prepare the solution, deionized (DI) water with a conductivity of about 0.8 $\mu\text{S}/\text{cm}$ was used. The membrane utilized in this work was polysulfone (trade name UFX5-pHT) from Alfa Laval Company. Its cut-off value was approximately 5 kDa and the maximum operational pressure and temperature were 15 bar and 75 °C, respectively.

2.2. Ultrafiltration procedure

In this work, filtration was carried out using a dead-end Amicon stirred cell (model 8400) with a capacity of 300 mL. A schematic flow diagram of the ultrafiltration set-up is shown in Fig. 1.

The membrane utilized in this work had an effective surface area of 40 cm^2 . In each experiment, the membrane was first placed in DI water for 2 h. Then, the pre-treated membrane was put in the cell and compressed up to 540 kPa for approximately one hour using DI water. To work out the membrane permeability, pure water fluxes were measured at various pressures. Prior to each run, the pure water fluxes were measured at a pressure of 100 kPa. Then the membranes were washed in order to measure fouling from the changes made in the pure water permeabilities.

In this work, the feed solution was prepared by dissolving the surfactant and nickel in a 250 mL of deionized water. To ensure that the solutes were evenly dispersed in the feed, the solution was mixed for at least one hour. For the filtration, the cell was filled with the prepared feed solution and stirred for 15 min under atmospheric pressure. It was then placed under the designated trans-membrane-pressure (TMP) with nitrogen gas. For the filtration procedure, 100 mL of permeate under a constant pressure of 100 kPa was collected with a volume concentration factor of 1.7 (VCF = 1.7). All experiments were carried out three times and their average values were used in the calculations. Furthermore, it was also concluded that a simple cleaning procedure with alkaline

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