A Review of Micellar Enhanced Ultrafiltration Technique in the Removal of Heavy Metals from Aqueous Solutions

Deniz ŞAHİN*

Gazi University, Ankara, Turkey

Abstract: The pollution of the aquatic ecosystems with heavy metal ions has become a global problem in recent years. Heavy metals normally occur in nature and are essential to life at trace levels. However, they can be toxic when their concentrations exceed the upper permissible limits. Heavy metal contaminated habitats have the ability to bioaccumulate in aquatic ecosystems, which, in turn, may enter into the food chain and lead to health problems. Therefore, it is necessary to remove these heavy metals from aquatic ecosystems. Several technologies are already in operation, but these conventional technologies involve high operational costs and may produce harmful impacts on aquatic ecosystems. Micellar enhanced ultrafiltration (MEUF) is an alternative technique to remove the trace concentrations of heavy metals from aquatic ecosystems. The uniqueness of MEUF is that it requires less energy due to low membrane cost and working pressure. Although various researchers have been carried out the MEUF study on the removal of heavy metal ions, few review papers indicate the factors on MEUF technique. That is the reason why this article focuses on reviewing of different parameters such as membranes, surfactants, operating conditions in the MEUF technique. In this technique, heavy metal ions' removal even at lower concentrations has reached over 99%, which is evidently demonstrated in the presented review. The use of water-soluble ligands in combination with MEUF is a hybrid process to remove selectively and enhance the recovery of heavy metals. As understood in this study, an investigation is needed to treat highly concentrated solutions and real wastewater.

Keywords: Critical micelle concentration, Heavy metal ions, Membrane types, Micellar-enhanced ultrafiltration, Surfactant.

1. INTRODUCTION

Heavy metals not only have seriously threatened human health but also severe impacts on aquatic ecosystems due to their non-biodegradable nature, toxicities, etc [1]. In natural aquatic ecosystems, heavy metals are present in low concentrations, generally at the nanogram to microgram per liter level. However, the occurrence of heavy metal contaminant has become a problem of increasing concern in recent times. This situation has arisen as a result of the rapid growth of population, increased urbanization, expansion of industrial activities, exploration, and exploitation of natural resources, an extension of irrigation, and other modern agricultural practices.

Various techniques have been employed for removing heavy metals from wastewater, such as precipitation, ion exchange, evaporation, reverse osmosis, adsorption. When used singularly, conventional chemical techniques generally generate toxic sludge or pollutants that are unable to settle within industries, while the biological techniques are prolonged slow and time-consuming. At the same time, these techniques need large areas and proper maintenance and operation. Therefore, economical and effective water treatment is still a serious problem [2-9]. Hence to overcome them, newly applicable treatment techniques are recently developed.

In the last two decades, micellar-enhanced ultrafiltration (MEUF) as a surfactant-based pressure-driven membrane separation technique has gained great notice for heavy metals removal from aqueous ecosystems. MEUF technique was initially introduced by Leung [10] in 1979 to treat industrial effluents laden with toxic heavy metal ions and organic compounds [11-15]. The main idea of this technique is to increase the size of metal ions by forming a complex with surfactant. When the surfactant is added to aqueous streams at a concentration higher than its critical micelle concentration (CMC) level, they form large amphiphilic micellar aggregates. These aggregates have a hydrodynamic diameter significantly larger than the pore diameter of ultrafiltration membrane. The metal ions and inorganic pollutants form a bond with the head group of the ionic micelles, which is oppositely charged through electrostatic interaction, as organic contaminants will entrap in micelles via Van der Waals force and will solubilize in the micelle interior [16]. Then, the micellar solution is passed through an appropriate molecular weight cut-off (MWCO) ultrafiltration membrane enough to reject the micelles. The micelles containing pollutants shall be rejected by the membrane, and in this way, the permeate side contains unbound ions, organic molecules in micelles

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*Address correspondence to this author at the Gazi University, Ankara, Turkey; E-mail: dennoka1k@hotmail.com

E-ISSN: 2409-983X/20

and surfactant monomers. This results in a clean permeate that can be recycled or discarded [17].

The major advantages of MEUF compared to other techniques are relatively low energy consumption, high removal efficiency, high flux, small space requirement and convenience to recover surfactants [17,19-29]. The efficiency of the MEUF technique on heavy metals removal depends on various parameters, such as operating conditions, surfactant properties, membrane characteristics, additives, and competing metals to be removed, and so on [30]. Many research groups have studied the aforementioned parameters and their effect on MEUF performance. However, universal experimental conditions that can be applied to MEUF experiments do not exist. Experimental parameters have to be selected based on the individual system. Also, this technique must be used for the real treatment of wastewater not just in laboratories on a small scale.

This paper aims to provide an overview of the state-of-the-art MEUF technique. In the following sections, the effects of various parameters such as membrane, surfactant, and operating properties on MEUF performance are summarized. Published studies of 91 cited references (2000–2020) are reviewed.

2. PARAMETERS AFFECTİNG EFFİCİENCY OF MEUF TECHNIQUE

Micellar-Enhanced Ultrafiltration technique has been shown to be a promising technique for the removal of multivalent heavy metal ions in a solution. The MEUF technique involved the combined use of surfactant and ultrafiltration membrane. In this technique, the surfactant is added to an aqueous solution at a concentration higher than its CMC. The CMC is the minimum concentration at which micelles of the surfactant start to form. Micelle has a high electrical potential on its surface, where pollutants can be trapped depending on the charge characteristic of the pollutants. Therefore, the heavy metal cations electrostatically adsorb on the micellar surface formed by anionic surfactants. Similarly, cationic surfactants are effective in removing hazardous anions. When the solution containing micelle is passed to the ultrafilter membrane, micelle retains on the membrane surface. Unbound ions and surfactant monomers pass through the ultrafilter membrane to the permeate side. The separation principle of the MEUF is illustrated in Figure **1**.

The performance of MEUF depends upon the material selection of ultrafiltration membranes and other characteristics, surfactant properties, operating conditions, and dissolved ions in the solution, as shown in Figure **2**.

2.1. Surfactant Properties

2.1.1. Surfactant Type

The literature studies presented 54.5% of Pb^{2+} removal [31] by using regenerated cellulose membrane, 15% chromate (CrO_4^{2-}) rejection of when PES mem-

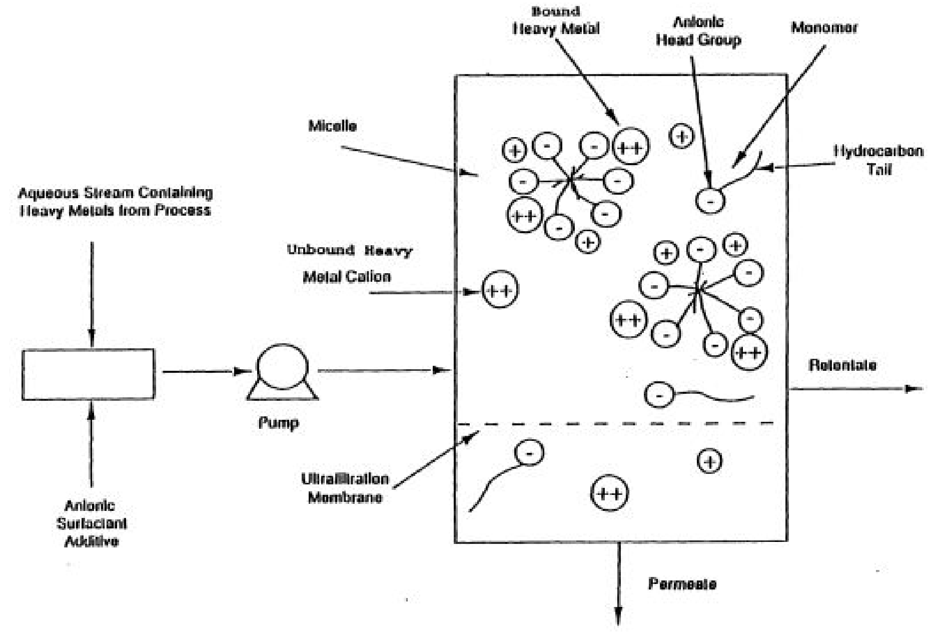


Figure 1: Schematic representation of the MEUF technique (adapted from [17]).

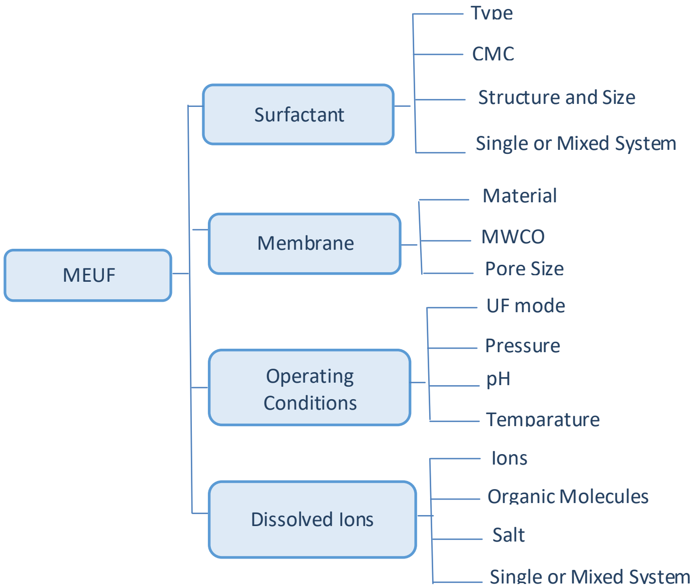


Figure 2: MEUF operating parameters block diagram.

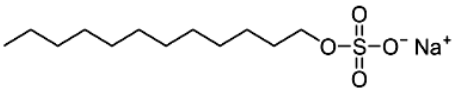
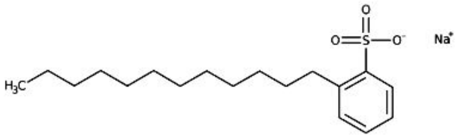
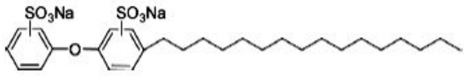
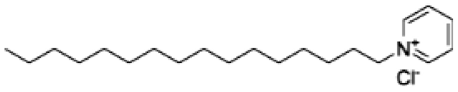
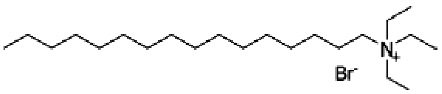
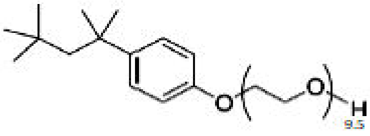
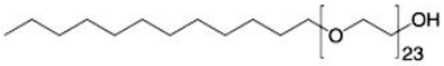
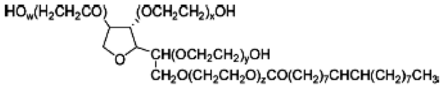
brane was used [32], rejection of polyphenols was between 5 and 28% by hydrophobic poly(vinylidene fluoride) (PVDF) membrane [33] in the absence of surfactants. These findings revealed the importance of surfactant application in the MEUF process. For this reason, the appropriate surfactant selection is one of the foremost parameters considered in the MEUF technique. Basically, the surfactant can be classified into four main types; anionic, cationic, non-ionic, and amphoteric surfactant [34]. Nevertheless, most of the researchers prefer ionic surfactants in the MEUF technique due to their ion-pair complex formation ability with oppositely charged ions for the removal of metal ions from aqueous solution via ultrafiltration. Usually, anionic surfactants have been utilized for the removal of metal ions, while cationic surfactants have been used for anions and organics. For example, sodium dodecyl sulfate (SDS) as an anionic surfactant in removing heavy metal ions from wastewater has been widely used by researchers. Samper *et al*. proved that 100% removal of ion Pb^{2+} was achieved using SDS. They had also investigated the removal of other metal ions, Cd^{2+}, Cu^{2+}, Ni^{2+}, Zn^{2+} using SDS and linear alkylbenzene sulfonate (LAS) as surfactants by MEUF. They found that metal retention is higher than 90%, except for Ni^{2+}, for both SDS and LAS surfactant [35]. Taşcıoglu *et al*. found that the Cu^{2+} ion was completely removed by MEUF performed at pH 5 in the presence of SDS [36]. Baek *et al*. demonstrated that 99 % of CrO_4^{2-} removal was achieved using Cetylpyridinium chloride (CPC) as surfactant [37]. The potential of CPC also has been tested for the removal of organic materials from wastewater by MEUF. Luo *et al*. obtained that the removal of phenol using CPC is higher (93.8%) than with the use of other cationic surfactants; hexadecyltrimethylammonium bromide (CTAB) (85.9%) and octadecyl trimethyl ammonium bromide (OTAB) (92.4%), which is attributed to the structural similarity caused by CPC and phenol that have an aromatic ring [38].

2.1.2. Surfactant Concentration

The CMC defines micellar aggregates formation over which surfactant solutions show an abrupt change in physical properties such as electrical conductivity and surface tension. Aoudia *et al*. reported that feed SDS concentration below CMC has Cr^{3+} removal efficiency of 33%. On the other hand, a marginal increase in rejection (99%) was observed at CMC [21].

A study for the removal of zinc from aqueous solutions by using SDS indicated that an effective removal (97%) was noted when the initial SDS concentration goes beyond 6 mM, although the CMC of SDS is 8.27 mM. The rejectionis attributed to the concentration polarization (CP) and adsorption of surfactant monomers at the membrane/solution interface [39]. Liu *et al*. reported Cu^{2+} removal efficiency of 93% at the molar ratio of SDS to Cu^{2+} of 5 and SDS concentration equal to CMC [40]. Juang *et al*. investigated the removal of single metal ions, including Cs^{+}, Sr^{2+}, Mn^{2+}, Co^{2+}, Cu^{2+}, Zn^{2+}, and Cr^{3+} from aqueous solutions by using SDS. They showed that complete removal of metal ions except for Cs^{+} could be achieved as long as the SDS micelles were formed [23]. Beolchini *et al*. mentioned an effective removal of arsenic (As-V) (98%) [41]. Baek

Table 1: Basic Properties of Surfactants Mentioned in the Article

Surfactant	Acronym	General Structural Formula	Molecular Weight	CMC (mM)	Type
Sodium dodecyl sulfate	SDS		288.37	8.27	Anionic
Sodium dodecylbenzene-sulfonate	SDBS		348.48	1.5	Anionic
Sodium hexadecyl diphenyl oxidedisulfonate	DPDS		598.72	0.601	Anionic
Cetylpyridinium chloride	CPC		358.01	0.90	Cationic
Hexavalent trimethyl ammonium bromide	CTAB		364.46	0.92	Cationic
Octadecyl trimethyl ammonium bromide	OTAB		392.50	0.28	Cationic
P-tertiaryoctylphenoxy polyethyl alcohol	Triton X-100/TX100		628	0.28	Non-ionic
Polyethylene glycol lauryl ether	Brij-35		298	0.28	Non-ionic
Polyoxyethylene sorbitan monooleate	Tween 80		1310	0.03	Non-ionic
Nonylphenyl ether	NP-12		230	0.078	Non-ionic

et al. demonstrated that at the molar ratio of 1:5, 98% of CrO_4^{2-} removal was achieved. In addition, increasing molar ratio to 1:5 and 1:10 CrO_4^{2-} removal efficiency through MEUF increased from 98% to over 99% [37]. Gzara *et al*. studied permeate flux and CrO_4^{2-} rejection as a function of CTAB surfactant concentration in the feed solution at a fixed CrO_4^{2-} concentration 2.10^{-4} M of feed solution. They illustrated that CrO_4^{2-} rejection was above 80%, even below the CMC of surfactant. The higher surfactant concentration causes a micelle aggregation layer (MAL) on the membrane surface which shows micelles presence and results in concentration polarization as mentioned in previous studies [42].

2.1.3. Surfactant Size, Shape, and Mixed Surfactants

The micelles size has an important role in the removal of a targeted ion in the MEUF technique. Xu *et al*. expressed that normal micelle size is 5.07 nm at CMC of SDS, while beyond CMC value, micelle size decreased, and its shape was also changed [43]. Linear molecule passes through a membrane that will retain globular molecules of the same molecular mass. Several factors affect the CMC of a surfactant, including temperature, pressure, presence of non-ionic surfactant, and inorganic salt. Ionic surfactants have much higher CMC than non-ionic ones even though they contain equivalent hydrophobic groups in an aqueous medium [44]. Theoretically, less removal of ionic contaminants by the MEUF technique can be expected using nonionic surfactants. Yenphan *et al*. studied on removing Pb^{2+} ion using TX-100 and nonyl phenyl ether (NP12) as nonionic surfactants and found that the removal of the Pb^{2+} ions was low, about 9% to 18% for TX-100 and 30% to 37% for NP12. They explained that the low rejection of ions was due to the complex formation between the Pb^{2+} ions and ethylene oxide (EO) groups of TX-100 and NP12 [45].

These findings supported the importance of surfactant selection with large size micelles formation, low CMC, the high solubility of the solute, lower adsorption ability to the membrane surface, and biodegradable are considering properties for better MEUF performance. However, all at the same time is not possible, therefore, a strong interaction between solute and surfactant is considered to be the basic criteria. Furthermore, surfactants with lower CMC and readily biodegradable are preferred in the MEUF technique in order to reduce the surfactant concentration in permeate. For example, Lee *et al*. reported that CrO_4^{2-} removal reduced from 93.7% to 84.8% when the concentration of Tween 80 increased from 10 to 25 mM in CPC and CrO_4^{2-} solution [46]. This aspect of the technique could be managed by reducing the CMC of ionic surfactant by adding anon-ionic surfactant, but the removal efficiency decreases slightly [47,48]. Zhang *et al*. demonstrated that removal efficiencies of Cu^{2+} were up to the maximum values 98.3 and 95.8% when the molar ratios of Brij 35 and TW80 to SDS were 0.3, and it was 93.5% given 0.7 molar ratio of TX100 to SDS. They also revealed the concentration of SDS in the permeate decreased dramatically with the addition of these nonionic surfactants [49]. Xu *et al*. showed that at ahigh molar ratio of non-ionic surfactant Brij in the solution not only the CMC of anionic surfactant (SDS), decreased but Cd^{2+} removal efficiency was also decreased [43].

2.2. Membrane Properties

Choosing the appropriate membrane is one of the main parameters to efficiently link reaction and metal rejection by the MEUF technique. Important factors in membrane selection include membrane material, surface charge, and MWCO/pore size. An ideal membrane should have high hydraulic permeability to water, which enables high permeate flux under a moderate transmembrane pressure (TMP), molecular sieving features so that the membrane can completely retain solutes with molecular weight higher than a particular MWCO value and completely release those with lower ones, high stability against chemical/thermal changes, and solute types or concentrations changes, high fouling resistance to ensure longer membrane life, and high manufacturing reproducibility for better performance of MEUF technique. Organic MEUF membranes are made of polyethersulfone (PES), regenerated cellulose (RC), polysulfone (PS), cellulose acetate (CA), polyvinylidene difluoride (PVDF), polyacrylonitrile (PAN), polytetrafluoroethylene (PTFE), and polyamide (PA). In addition, ceramic membranes are also a desirable system because they are capable of withstanding under high temperatures and highly acidic or basic environment [23,26].

2.2.1. Membrane Material

Metal rejection and permeate flux can vary depending on the nature of membrane material that is hydrophilic or hydrophobic. These membranes studies have demonstrated that micelles adsorb preferably on the hydrophilic surface of PA membranes rather than on the hydrophobic surfaces of ceramic or PS membranes [50]. Hydrophilic membranes consist of capillaries large enough to display for more than one layer; thus, either a second layer is adsorbed on the first one, or adsorbed clusters are formed on the surface. In both these structural models, hydrophilic head groups are in the outer layer and make the surface more hydrophilic. On the other hand, the surface aggregates consist either of monolayer or semi-spherical clusters in a hydrophobic membrane. The tail groups are adsorbed to the surface. In contrast to a hydrophilic membrane, head groups arrange in the outer coat so that they are in direct contact to the aqueous medium as shown in Figure **3** [51].

Chung *et al*. investigated the effect of membrane hydrophilicity on the performance of the MEUF process. They were prepared from polysulfone blends containing various amounts of a hydrophilic copolymer, poly(1-vinylpyrrolidone-co-acrylonitrile) (P(VP-AN)). An increase in the permeate flux was observed with an increase in the membrane hydrophilicity [52]. Taşcıoglu *et al*. found that the Cu^{2+} ion was entirely removed by (with) hydrophilic (RC) membrane and SDS as surfactant [36].

2.2.2. Membrane MWCO

Larger pore-sized membranes are known to cause earlier development of the concentration polarization and reduce the surfactant's release at the permeate

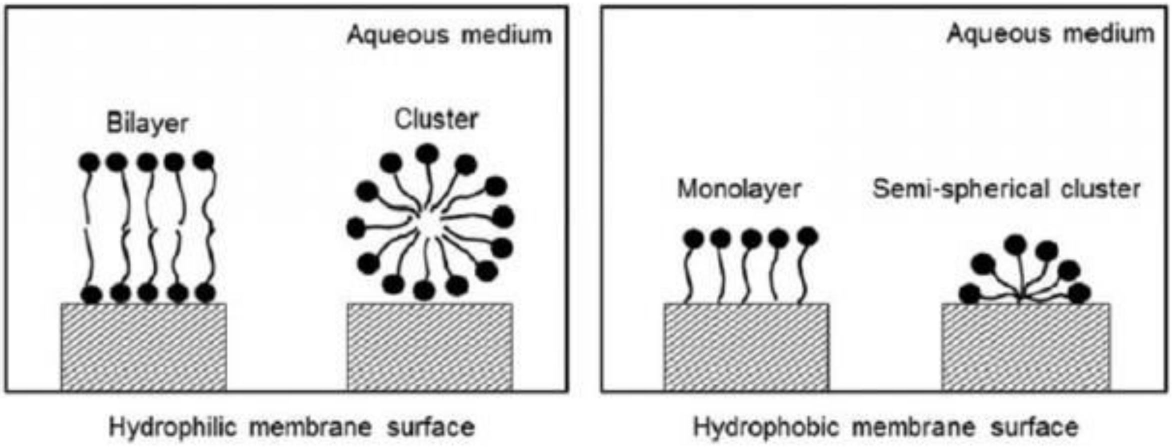


Figure 3: Surfactant adsorption on hydrophilic and hydrophobic membrane surfaces (adapted from [51]).

[21]. Şahin *et al*. demonstrated that Pb^{2+} ion was almost wholly removed from aqueous solution at pH 6 in the presence of SDS at relatively low concentrations using RC membrane. In their experiment, they further observed two times increase in permeate flux with an increase in the membrane porosity by 1000 Da to 5000 Da [31]. Karate and Marathe explained that Ni^{2+} and Co^{2+} were simultaneously removed from aqueous solution using cross-flow micellar enhanced ultrafiltration. They used 20 kDa PS membrane and obtained the rejection of more than 99% [53]. Rafique and Lee reported average Cd^{2+} removal (74.6%) using PAN membrane [54]. In another study, Rafique *et al*. obtained higher Ni^{2+} rejection (83.0%) using membrane MWCO of 300 kDa than that of 100 kDa (72.3%) [55]. Similar results were reported by Landaburu-Aguirre *et al*., reporting that the ultrafiltration membrane with 3-kDa pore size exhibited more Zn^{2+} removal than the 10-kDa pore size membrane [56].

2.3. Operating Conditions

The MEUF can be applied in two operating modes: a dead-end (unstirred batch system, stirred batch, stirred batch with feed reservoir) or crossflow mode (with and without retentate recycling), which can be a pressure (or vacuum)-driven system (Figure **4**). A feed solution is applied perpendicular to a membrane in dead-end mode without any flow along the membrane. As the process proceeded, an accumulation of rejected solutes occured near the membrane surface, which decreased permeate flux. Therefore, the feed solution was stirred to reduce polarization effects. In crossflow mode, shear forces are applied on the membrane

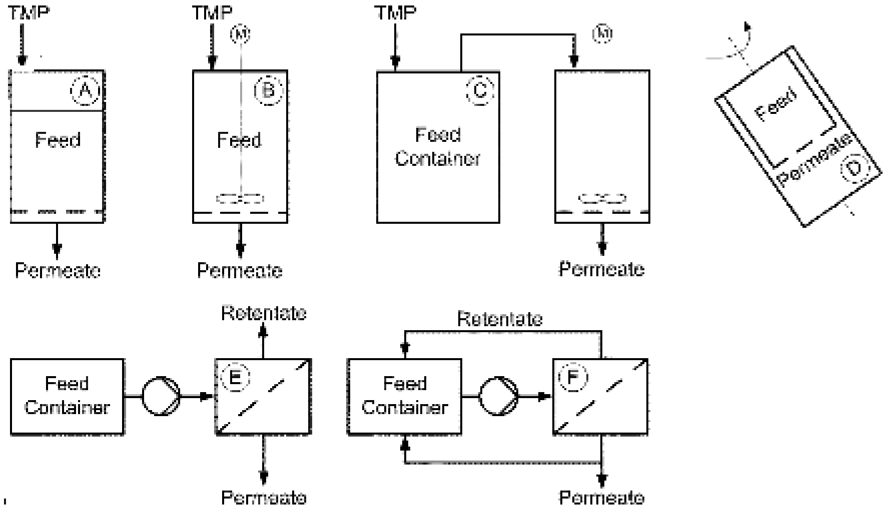


Figure 4: Schematic representation of operating modes; dead-end (A-unstirred batch system, B-stirred batch, C-stirred batch with feed reservoir) or cross-flow mode (E-with and F-without retentate recycling) (adapted from [17]).

surface through a flow of the feed along the membrane [17].

For example, Xiarchos *et al*. investigated the removal of Cu^{2+} from aqueous solutions via batch stirred cell mode MEUF. They obtained the maximum rejection coefficient of 98.4% for the following optimal conditions: stirrer speed: 100 rpm, applied pressure: 3 bar which was adjusted by pressurized air, and operating temperature: 25±2 °C [57]. Kim *et al*. reported that the order of removal efficiency was Cd>Cu>Co≈Zn by using a solvent-resistant stirred cell filtration [58]. Das *et al*. treated an aqueous solution containing copper and potassium permanganate (MnO_4^{2-}) by cross-flow mode ultrafiltration using a mixed micellar system comprising of SDS and CPC. They reported that the retention of Cu^{2+} was in the range of 90-100% and that of potassium permanganate was 96-99% [59]. While cross-flow filtration has been used widely in industrial applications, the stirred batch system has been preferred in laboratory studies because it is easy to set up and requires less equipment of feed volume.

2.3.1. Operating Pressure

MEUF, a pressure-driven membrane separation technique, uses less pressure than other filtration techniques for the removing of small-sized molecules, which makes MEUF a good removing process. Various studies were conducted to investigate the effect of operating pressure on MEUF performance for removing inorganic and organic pollutants. Muthumareeswaran *et al*. found no such significant effect of pressure on CrO_4^{2-} removal, which suggests that there may be negligible CP [60]. Another study done by Juang *et al*. the removal of a single metal ion by using SDS presented similar results as aforementioned [23]. Huang *et al*. reported that the TMP and feed concentration of SDS had significanty influenced on the permeate flux. They discovered that increase in TMP resulted in higher flux while SDS retention was decreased [61]. Ghazi *et al*. found that the permeate flux increased almost linearly with operating pressure, within the range from 0.35 L $m-^{2}$ min^{-1} at 1 bar to 1.79 L $m-^{2}$ min^{-1} at 4 bar. They also showed that the Mn^{2+} rejection increased slightly with an increase of the operating pressure, ranging from 89.658 at 1 bar to 97.971 at 4 bar [62]. Rafique and Lee investigated the effect of input pressure on Cd^{2+} removal under different initial pressures. Cd^{2+} removal increased with an increase in initial retentate pressure, similar to previous studies [54]. With an increase in retentate pressure, TMP also increased. At higher pressure, micelles might be compacted and would cause more retention of a broken micelle [63]. Hence, as a result of pressure increase, secondary resistance to the solutes transporting through the membrane to the permeate solutions was increased [64]. Luo *et al*. obtained that the retention of phenol remains almost independent of pressure nearly within the range of 0-0.15 MPa [38]. In conclusion, a very high operating pressure is not necessary for a high permeate flux. The operation of the MEUF technique at low TMP is an important issue in terms of minimizing operating costs.

2.3.2. pH

The pH affects the interaction of solute and micelle; thereby, pH of the solution has a significant influence on solute removal through the MEUF technique. The influence of pH variations in the MEUF technique is dependent on the nature of solute and surfactant. Juang *et al*. reported that the removal efficiency of cationic metals (Mn^{2+}, Co^{2+}, Cu^{2+}, Zn^{2+}, and Cr^{3+}) was increased at higher pH values with the help of SDS [41]. The removal efficiency was decreased at lower pH due to the competition between H^{+} ions and cationic metal ions to get adsorbed on the micelle surface. Xu *et al*. observed that there was no effect of pH on the removal efficiency of Sr^{2+} and Cr^{3+}, but rejection of Cd^{2+} increased from 83 to 99% when the pH increased from 3 to 11 in the feed solution at a fixed Cd^{2+} and SDS concentration of 100 mg/L and 8 mM, respectively [43]. Şahin *et al*. reported that the removal of Pb^{2+} sharply increased with the increase of pH (3-7). The increase in the removal was mainly due to the formation of metal hydroxides at the higher pH. The $Pb(OH)^{+}$ begins to form at pH 5; the percentage of its formation gradually increases with the increase of pH and reaches its maximum value at pH 8.5. At higher pH, $Pb(OH)_2$ and $Pb(OH)_3^{-}$ and $Pb(OH)_4^{2-}$ complexes are formed, respectively. They have also achieved that Pb^{2+} ion was almost completely removed from a battery plant's wastewater by adding 0.1 mM SDS at pH 6 [31]. Chen *et al*. observed the effect of pH on arsenate As (V) removal by adding 20 mM CPC prior to UF. Rejection of As (V) increased from 0.39 to 0.89 as the pH increased from 5.0 to 8.2. Higher pH leads to a higher As removal because As (V) is increasingly present as a divalent anion with a higher binding capacity to the micelles of surfactant [65].

2.3.3. Operating Temperature

From previous literature, it is known that temperature influences the CMC of the surfactant, the in viscosity of solution, solubility, and micelle properties likethe size of the micelles. The CMC of surfactants is a function of temperature. Kowalska *et al*. reported that the temperature increase of SDS solutions resulted in an increase CMC of the SDS due to the demicellization occuring at a higher temperature because of the breakage of the palisade layer of the micelle, and thus, detachment of surfactant ions from micellar bulks occurred [66]. As the temperature is raised, the solubility increases until the CMC is reached at the Krafft temperature. Below the Krafft temperature, precipitation of the ionic surfactant occurred. For nonionic surfactant, when the temperature is increased to a spesific value, the nonionic surfactant solution will separate into surfactant-rich and water-rich phases and will become turbid. The temperature at which the solution becomes turbid is termed cloud point (CP). The cloud-point temperature for nonionic surfactants depends upon the hydrophilic character of the surfactant. Purkait *et al*. observed that nonionic surfactants have CP temperature below 100°C, e.g., TX-100 (65°C) or TX-114 (24°C) [67], and this temperature undergoes changes due to the addition of solutes. In the case of ionic surfactants, CP temperature is higher than 100°C because of the solubility of ionic surfactants increases with an increase in temperature. In addition, increasing temperature results in a decrease in viscosity which ultimately increases flux through the membrane and affects the filtration process [17]. Urbański *et al*. reported that CTAB and alkyl polyglucoside (APG) surfactants caused a small additional resistance, but SDS added a strong resistance to the membrane at high temperatures. This is because CTAB solutions have a high krafft point 250°C, while the krafft point of SDS was 210°C [64].

2.4. Dissolved Ions

The addition of salt decreases the surfactant leakage to permeate due to drops in the CMC of the surfactant, which leads to efficient removal of surfactant through membranes. Miyagishi *et al*. determined that the drop is much lower for nonionic surfactants than for ionic surfactants [68]. In the presence of salt, the repulsive forces between the head groups of ionic surfactant, which are usually fighting against the aggregation, decrease due to the electrostatic shielding effect. Thus, in the presence of an electrolyte, micelles formation is comparatively easy [44,69]. In this study, researchers presented that Cd^{2+} removal efficiency decreased from 95% to 75% with the increase of the NaCl concentration from 10 to 100 mM. This may be attributed to the cation Na^+ which occupies the available binding sites. Additionaly, Cl^- anions can form complexes with Cd^{2+} metal ions [44]. Aoudia *et al*. reported that Cr^{3+} removal was reduced from 99.65 to 92.21% with the addition of 0.05 M NaCl and even up to 53.50% by adding 0.9 mM NaCl at the rate of 0.9 mmol/dm^3 [21]. When the salt is added to a solution, some of the water molecules are attracted by the salt ions which enhancing the attaching of organics with the micelles due to the salting-out effect as demonstrated for phenols [70]. On the contrary, Gzara and Dhahbi determined that as long as the NaCl feed concentration is less than or equal to 100 mM, more than 88% of Cr^{3+} are retained and surfactant leakage was reduced [42]. Generally, permeate flux decreases by adding salt but metal removal efficiency depends upon the nature of solute.

Various researchers reported that the retention of metal in the MEUF technique was inhibited in the presence of other inorganic pollutants. Baek and Yang reported that the rejection of nitrate (NO_3^-) from the nitrate/chromate/CPC system was below 40% with the molar ratios of 1:1:1, 1:1:2, and 1:1:3 (nitrate:chromate: CPC). They explained that when the ratio was increased to 1:1:5 and 1:1:10, rejection of NO_3^- reached 65% and 80%, respectively. The rejection of NO_3^- was inhibited in the presence of CrO_4^{2-} due to differences in binding efficiency towards ionic micelles between counter ions, which depends on the valence of counter ions; the valence of CrO_4^{2-} was higher than that of NO_3^-. The rejection of NO_3^- was suppressed significantly by co-existence of CrO_4^{2-} compared to the single component NO_3^-/CPC system. However, the rejection of CrO_4^{2-} was increased as compare to single component CrO_4^{2-} /CPC system. In the mixed system, the rejection of CrO_4^{2-} increases from 50%, 71%, 90%, and 98% at the molar ratios of 1:1:1 to 1:1:2, to 1:1:3, and to 1:1:5 (nitrate:chromate:CPC), respectively, as compared to CrO_4^{2-} /CPC system [71]. The same author did another study for NO_3^-, CrO_4^{2-}, and ferric cyanide [$Fe(CN_6)^{3-}$] removal using CPC and clearly observed the inhibition of [$Fe(CN_6)^{3-}$] on the removal of NO_3^- and CrO_4^{2-}, but the inhibition of CrO_4^{2-} on the removal of NO_3^- was lower than that of ferricyanide [72]. Lee *et al*. Investigated simultaneous removal of CrO_4^{2-} and Trichloroethylene (TCE) by MEUF in the mixed surfactants. They stated that the removal of TCE and CrO_4^{2-} was not hindered by each other's presence, since the removal mechanism is different [46]. Similarly, Li *et al*. reported that the removal of Cd^{2+} and phenol using pure SDS and mixed surfactants (Triton X-100/SDS) [73]. In another experiment, Tung *et al*. found that the co-existence of phenol slightly enhanced Cu^{2+} removal while phenol removal was approximately 27% due to its relatively hydrophilic characteristics [48].

In literature, simultaneous removal of multiple ions through the MEUF technique has also been investigated. For example, Karate and Marathe explained that Ni^{2+} and Co^{2+} were simultaneously removed from aqueous feed using cross-flow micellar enhanced ultrafiltration [53]. Channarong *et al*. reported that the simultaneous removal of Ni^{2+} and Zn^{2+} from aqueous solution by the MEUF and activated carbon fiber (MEUF-ACF) hybrid process. They observed that the concentration polarization of micelles played a major role in the removal of Ni^{2+}, Zn^{2+}, and SDS. In the case of single ion removal, ions are attracted by the charged surface of the micelles until the binding capacity is attained. If multiple ions are present in the solution, they compete for the binding sites at the surface of the micelle [74].

3. SELECTIVELY REMOVAL: LİGAND MODİFİED MİCELLAR ENHANCED ULTRAFILTRATION (LM-MEUF)

Traditional MEUF has the disadvantage of not providing a high selectivity in removing metal ions from aqueous solutions. In recent years, this problem has been overcome by adding a ligand to an aqueous solution. The vast majority of the ligand forms a complex with the target ion of interest and solubilizes or dissolves inside the hydrophobic core of the micelles. This solution containing surfactant/ligand/ion is then forced through an ultrafilter membrane. Thus, the macro-ligand and its associated ions will be rejected by the membrane, unlike the uncomplexed ions that can pass through the membrane. This process is called the "ligand-modified MEUF" (LM-MEUF) technique. The main advantages of the LM-MEUF technique over the ultrafiltration system are to have more efficient separation with better selectivity, recovery of desire metals, and complexing agent can be chosen in order to be easily regenerated and reuse. The efficiency of the LM-MEUF technique depends on the ligand to metal ion mole ratio, the nature of the ligand, and the pH of the solution [36,85,86]. Based on the literature review, the removal efficiency of LM-MEUF for different heavy metals is summarized in Table **2**.

Roach and Zapien examined the specific separation of U(VI) from Sr^{2+}. For both metal ions rejection exceeded 99.9% [86]. Şahin and Taşcıoglu attained the removal of Pb^{2+} at pH 3, ~82%, in the presence of Dithizone and SDS as a surfactant. Another study done by the same author for removing of Cu^{2+} ions from both single-component and Cd^{2+} containing solutions in the presence of SDS was determined. Complete removal of Cu^{2+} ions from Cd^{2+} containing solutions could be achieved 2,4,6-tri(2-pyridyl)-triazine (TPTZ) out of 20 ligands [36]. Complexation behaviors of 20 ligands with Cu^{2+} and Cd^{2+} by co-existence of CTAB and TX100 micelles at different pH values. The most effective

Table 2: Heavy Metal Removal Efficiency in LM-MEUF Technique

Metal Ions	Ligand	Surfactant	Removal %	Ref.
Cd^{2+}	pyridine-2-azo-*p*-dimethylaniline (PADA)	SDS	~99%	75
Cr^{3+}	etylenediaminetetraacetic acid (EDTA)	SDS	99%	76
Mg^{2+},Ni^{2+}, Cd^{2+}, Cu^{2+}, Fe^{2+}, Zn^{2+}	nonaoxyethylene oleylether carboxylic acid (RO90)	SDS	> 95%	77
Ni^{2+}/Co^{2+}	iminodiacetic acid (IDA)	SDS and SDS/TX100	Co^{2+}:84% Ni^{2+} :93%	78
Rh	triphenylphosphine (TPP)	nonylphenol ethoxylate(Marlopen NP9)	~100%	79
Rh	SulfoXantPhos (SX)	poly(oxyethylene)-5-nonylphenol ether (NP5), poly(oxyethylene)-9-nonylphenol (NP9)	93%	80
Cu^{2+}	citric acid>NTA>EDTA	SDS	citric acid>NTA>EDTA	81
Pd^{2+},Pt^{2+}	pyridine-2-azo-*p*-dimethylaniline (PADA)	SDS	Pd^{2+}:98%	82
Pu^{4+}	trioctylphosphine oxide (TOPO)	polyethylene glycol ether, Tergitol 15-S-9 (Tergitol)	90%	83
U(VI)	etylenediaminetetraacetic acid (EDTA)	SDS	89%	84
Am^{3+}	2-ethylhexyl phosphonic acid mono-2-ethylheyxl ester (H_2A_2)	SDS and Tergitol	~100%	85

ligand was found to be in the presence of CTAB and 2-hydroxy-1-(2-hydroxy-4-sülfo-1-naphthylazo)-naphthalene-3-carboxylic acid. Complete removal of Cu^{2+} ions from Cd^{2+} could be achieved [87]. Roach *et al*. used nitrilotriacetic acid (NTA) derivatives to remove of Pb^{2+} from an aqueous solution. Ligands provide excellent separation of Pb^{2+}, with R_{Pb} = 99.0-99.9% in the pH range 6 to 8 [88]. Vieira *et al*. showed that water soluble polymeric ligands to be powerful substances to remove trace metals from industrial wastewater through ultrafiltration [89]. Leclercq *et al*. investigated the complexation of Cu(II) with original alkylamidotartaric acids (C_xT) in the presence of Brij 58 and demonstrated that the extraction by LM-MEUF is very efficient technique [90]. Rahmanian *et al*. studied the effect of nonionic surfactant, pH and ligand and electrolyte concentration on Zn^{2+} rejection and permeate for first time [91].

4. CONCLUSION

Micellar enhanced ultrafiltration is a versatile technique with manifold applications. As shown by the multiple examples in this review, MEUF allows for an efficient treatment of aqueous solutions containing solutes using surfactant at the CMC. The selection of appropriate parameters, e.g., surfactant properties, membrane characteristics, operational conditions, and dissolved ions, is the key to a successful application of MEUF. Researchers are more attracted to the different types of membranes and their applications. Usually, metal ions removal increased with the increased pressure, pH, and increased concentration of ionic surfactant. The rejection of solutes does not dependent on the initial amount of surfactant used but depends on its concentration near the membrane surface. However, the flux is determined by the interaction of the surfactant with the membrane under proposed conditions, and relative flux dramatically decreases when surfactant concentration increased. Many examples showed that a variety of metal ions could be applied for which a desired interaction with the surfactant can be established, finally resulting in rejection coefficient > 99%. These high values are not limited to aqueous solutions of single ion only and can be extended to mixtures of different solutes, e.g., multiple ions. As understood in this review, for applying the MEUF technique in real wastewater still needs some work, such as improving the performance in terms of permeate flux, and recovering metals, organics and surfactant from the retentate solution. Metal removal has also enhanced in the MEUF + ligand technique.

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Received on 28-09-2020 Accepted on 25-10-2020 Published on 10-11-2020

DOI: https://doi.org/10.15377/2409-983X.2020.07.5