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A BRIEF REVIEW OF MICELLAR ENHANCED ULTRAFILTRATION (MEUF) TECHNIQUES FOR TREATMENT OF WASTEWATER IN INDIA

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ABSTRACT

In this paper, we have reviewed the application and efficiency of Miceller Enhanced Ultrafiltration (MEUF) method which is used to remove Heavy Metals, Dyes, Polyaromatic Hydrocarbons (PAH), Novel Compounds, Chromium, Phosphorous and Phenolic Compounds. It uses various surfactants like Hexadecylpyridinium chloride (CPC), Sodium Dodecyl Sulphate (SDS), Hexadecyltrimethyl Ammonium Bromide (CTAB), Oxyethylated Methyl Dodecanoate (OMD) and Alkylpolyglucoside (APG). These surfactants are retained after the process to be reused. Bisorption, Coagulation, polyelectrolyte enhanced ultrafiltration, Reserve Osmosis, Nanofitration are some of the methods that are used for the treatment of wastewater. Available research focused on the removal of metals, phenol, organic & inorganic materials, aromatic hydrocarbons by MEUF mainly on the type of surfactant used, surfactant concentration, applied pressure, operating time and pH. Formation of surfactant micelle and mechanisms for the attraction between micelle and metal ions are ignored during the research study on MEUF. It has been reported that MEUF combines the efficacy and simplicity of the conventional options with the operational flexibility of membrane-based separation. MEUF could be used for removal of wastewater provided the pollutants have low molecular weight. Higher reagent and electrical costs are reported in conventional MEUF method as compared to the hybrid process with MEUF. The hybrid process requires fewer surfactants. MEUF is considered a better alternative to the typically available membrane separation processes. The advantages of this method over other methods are high removal efficiency, low energy consumption, and easy operation. The selection of surfactant for the removal purpose is based on the target pollutants.

Keywords: Miceller Enhanced Ultrafiltration; Surfactant; Micelles; Polyaromatic Hydrocarbon; Novel Compounds.

INTRODUCTION

Water is becoming a scarce resource across the world. Consequent to rapid growth in population and thereby increasing water demand, stress on water resources in India is increasing and per capita water availability is reducing day by day. Providing high effluent quality of discharge in rivers and reuse of it is a viable option. However, cost-effectiveness and the low land requirement should be sufficient enough to warrant such project.

When properly managed, wastewater can be a valuable resource which horticultural and agricultural crop growers, as well as home gardeners, can benefit from. It is, after all, the same phosphorous, potassium and nitrogen making wastewater a source of pollution for lakes, rivers, and groundwater which are excellent nutrient sources for vegetation when this wastewater is made available for irrigation. Sewage wastewaters comprise of human waste, animal waste, and fecal material. Treatment of sewage wastewater is difficult because of the high concentration of organic matter, nutrients, and suspended solids. Most of these materials decompose readily in warm weather, releasing odorous gases to the atmosphere. Moreover, such constituents exert high oxygen demand in water bodies and render them incapable of supporting aquatic life. Therefore, it is required to treat such effluents by using methods that are affordable and can handle strong organic wastewater that is released intermittently. Various treatment technologies are present for the treatment of wastewater such as

a. Activated Sludge Process (ASP)
b. Trickling Filter (TF)
c. Biological Filtration and Oxygenated Reactor (BIOFOR)
d. Fluidized Aerobic Bioreactor (FAB)
e. Submerged Aeration Fixed Film (SAFF)
f. Upflow Anaerobic Sludge Blanket (UASB)
g. Miceller Enhanced Ultrafiltration (MEUF)

Industrial activities have increased heavy metal and inorganic micro-pollutants in the environment, mainly in an aquatic system (Samper et al., 2009). This is a matter of concern as they are non-biodegradable, highly toxic and probably have a carcinogenic effect. The degrading environment and ecological requirements have compelled us to look for the treatment of wastewater from industries with low energy, labor and capital costs (Schwarze, 2017). Many techniques are currently available as mentioned above; one of them is Miceller Enhanced Ultrafiltration (MEUF). Miceller Enhanced Ultrafiltration is a surfactant based separation technique as surfactants are the main ingredients in MEUF. The surfactant is added into the aqueous stream containing contaminants or solute above its critical micelle concentration (CMC). Selection of materials and operating conditions are required for this process.

There is an increase in the size of pollutant molecule by forming a complex with a surfactant. Surfactants are compound that lowers the surface tension between two liquids, liquid or gas or between a liquid and solid. They are organic compounds that are amphiphilic (both hydrophobic and hydrophilic groups) in nature. A surfactant contains both water soluble and insoluble components. At the periphery of air and water or oil and water (if oil is present), the surfactant diffuses and get absorbed. While using, a surfactant is added to the water spontaneously that forms aggregates called micelles which contains approximately 100 surfactants. A micelle is a particle of colloid dimension that exists in the equilibrium with the molecule or ions in the solution from which it is formed. The micelles reduce the hydrophobic tails of the surfactant and water, reducing the overall free energy of the system. Surfactants present above the CMC act as emulsifiers that will allow a compound (insoluble) present in the solvent being used to dissolve. This occurs because the insoluble species can assimilate into the micelle core, which is itself solubilized in the bulk solvent due to the interaction of hydrophilic groups with solvent species. The main advantages of this method are the low –energy requirements involved in the UF process (Samper et al., 2009). The size of a micelle is approximately the same in magnitude than the pore size of the membrane so that they are dispersed together with the solute through filtration. This method is in fact based on the use of surfactant which helps in the removal of substances with low molecular weight.

MEUF is an alternative way of ultrafiltration process and uses ultrafiltration membrane to remove substances from water. Ultrafiltration is a key separation process in which hydrostatic pressure forces a liquid against a semipermeable membrane. It can remove bigger substances from water with membrane pore sizes ranging from 0.01 to 0.1µm e.g. colloids and microorganisms (Asak, 2011). The high efficiency of Reverse Osmosis (RO) and high permeate flux of ultrafiltration membrane are utilized in the process. MEUF has combined the efficiency of nanofiltration (filter pore size 0.01-0.001µm) and reverse osmosis (filter pore size ranges from 0.001 to 0.0001µm) (Asak et al., 2011).

Nanometers are measured by their Molecular Weight Cutoff (MWCO) expressed in Dalton (Da) which refers to 90% of the molecular weight retained in the membrane from a solution. In nanofiltration, MWCO is higher

than 200 g/mol. On the contrary, RO is supposed to remove almost all substances from the liquid including single valence ions. The MWCO of that membrane is below 200g/mol. It requires a pressure of 15 -100 bars. The classification of Micellar-Enhanced Ultrafiltration are as follows (Liu et al., 2010);

1. With respect to surfactants
   i. Cationic surfactant
   ii. Anionic surfactant
   iii. Nonionic surfactant
   iv. Amphoteric surfactant
2. Hybrid processes
   i. Activated Carbon Fibres (ACF)
   ii. Biological treatment
   iii. Synthetic processes

Different pollutants can be removed from wastewater like cyanide (Bade and Lee, 2011), toxic heavy metals such as Mn^{2+}, Co^{2+}, Cu^{2+}, Zn^{2+}, and Cr^{3+} (Abbassi and Mulligan, 2014), Chromium (Li and Liu, 2005), organic compounds like phenol (Gorna and Nowak, 2013) and inorganic contaminant like phosphorous (Canizares et al., 2006), and toxic acids like tannic acid (Krivorot et al., 2009). In the present study, an attempt has been made to review the published work in peer-reviewed journals and article which deals with the treatment of pollutants from wastewaters through Micellar-Enhanced Ultrafiltration. The eviction of pollutants by MEUF depends on the characteristics and concentration of the metals and surfactants, solution, pH, ionic strength and parameters related to membrane operation. Recently many membrane-based techniques have been developed for the treatment of polluted water, MEUF is one of them. But the knowledge about this technique is less among the people as this method is new and has been used by few people. Also, this technique needs to be used for the real treatment of wastewater not just in laboratories on small scale.

Chromium

Chromium is one of the pollutants that are harmful in some of its forms. It is naturally found in rocks, animal, plants, soil, and volcanic dust and gases. It comes in different forms, including Trivalent (III) and Hexavalent (VI). Trivalent Chromium is often indicated as Chromium (III) and is an essential nutrient for the body. Hexavalent Chromium, referred to as Chromium (VI), is generally produced by industrial effluents. It is widely used in electroplating, stainless steel production, leather tanning, textile manufacturing, and wood preservation. Hexavalent Chromium, if inhaled, is harmful to human life. Workers get exposed to Hexavalent Chromium in various occupations and are at increased risk of developing lung cancer, asthma or damage to the nasal epithelia and skin (Zhitkovich et al., 1996). Consumption through drinking water has been found to cause cancer in the oral cavity and small intestine (Banchhor et al., 2017). In India, leather industry, steel manufacturing industries, textiles, and some chrome plating industries are high producers of Hexavalent Chromium. According to the National Primary Drinking Water Regulation standards, maximum contaminant level for Chromium should not be more than 0.1mg/l (Sumithraa et al., 2015). Some of the existing methods for the treatment of Chromium (Cr-VI) are Ion Exchange, Granular Activated Carbon, Adsorbents and Membrane filtration. Reverse Osmosis or Nanofiltration is also a process for ion removal but is not economical as they require high transmembrane pressure which makes this process expensive and uneconomical. Many researchers have explored and experimented with MEUF techniques to remove heavy metal contaminants from wastewater.

Nura et al. (2017) removed Cr -VI by mixed micelles of CTAB (Cetyl trimethyl ammonium bromide). A cationic surfactant was mixed with Triton X-100 and Tween-80 (nonionic). Batch stirred cell was used at 160 rpm and transmembrane pressure was kept at 100 KPa. They studied the effect of initial feed concentration Cr (VI) on percentage rejection of Cr (VI). When CTAB mixed with Triton X-100, the rejection was at 97% and when it was mixed with Tween -80, the rejection was observed to be as 93%. Bade et al. (2008) removed Chromium and Cetyl Pyridinium Chloride (CPC) from artificial wastewater. They used MEUF along with ACF (Activated carbon fiber) for this removal. CPC as a surfactant and UF membrane with 100 kDa, MWCO were used. The optimum molar ratio as 1.5, retentate pressure of 0.14 MPa and pH value as 7 was considered. They used Sodium

Chromate and Cetyl (hexadecyl) Pyridinium Chloride (CPC) of 99% and 98% purity with flux as 43.7 L/m and 32.9 L/m. The highest rejection of chromium and CPC were found to be as 97 and 70%, respectively.

Phenol and its derivatives

Phenol, also known as carbolic acid, is an aromatic organic compound that is volatile in nature. It consists of phenyl groups with a hydroxyl group. It is a weak acid and requires careful handling due to its property to cause chemical burns. Phenol and its vapors are corrosive to the eyes, skin, and respiratory tract (Kariduraganavar et al., 2014). Its corrosive effect on skin and mucous membranes is due to a protein degenerating effect (Lee et al., 2006). Repeated or prolonged skin contact with phenol may cause dermatitis or even second and third-degree burns (Warner and Harper, 1985). The substance can cause harmful effects on the central nervous system and heart resulting in dysrhythmia, seizures, and coma. In regions where water resources are becoming extinct, wastewater from industries causes serious problems. For example, in the Mediterranean region, Agro-Food industries produce more than 30 million tons of phenol every year. These industries involve olive mills wastewater, table olive wastewater, winery and distillery wastewater (Abbassi and Mulligan, 2014).

There are several techniques for the removal of phenols and its derivatives from wastewater, e.g. photooxidation (Rubalcaba et al., 2007), different Advanced oxidation processes such as Wet Air Oxidation (WAO), Catalytic Wet Air Oxidation (CWAO), H_2O promoted CWAO (Qie et al., 2012), Ozonation (Hidalgo et al., 2013), Nanofiltration (Mohammed *et al.* 2011) and adsorption with activated carbon adsorbents (Reis et al., 2007). Membrane processes also contribute to the removal of phenol. Different membranes may be used like liquid membranes (Kojima et al., 1995), anion exchange (Arsuaga et al., 2006), nanofiltration/ reverse osmosis membrane (Gupta et al., 2003), pervaporation process (Sarker, 2016). MEUF combines the efficacy and simplicity of the conventional options with the operational flexibility of membrane-based separation. MEUF could be used for removal of wastewater provided the pollutants have a low molecular weight (Chaudhari and Marathe, 2010).

A new method for phenol removal, known as GMEUF, was given by Zhang et al. (2013). GMEUF basically uses Gemini surfactants which have a unique nature. It has two hydrophobic and two hydrophilic head groups connected with a spacer. Cationic Gemini surfactant (CG) and flat sheet UF membrane with 10 kDa MWCO were used. The rejected phenol was about 90%. Bielska and Szymanowski (2004) mentioned about the removal of Nitrobenzene and Nitrophenol (Table 1) and investigated the effect of pollutant structural parameters, type of surfactant and membranes upon the efficiency of ultrafiltration in the removal of 4- nitrobenzene and nitrophenol and also to predict its operating parameters. The membranes that were used in this experiment are Cellulose (CQ), Polyethersulphone (PES) and Polyvinylidene Fluoride (PVDF) with CTAB as a surfactant. The highest rejection for 4-nitrobenzene and nitrophenol was 92.52 and 93.53%, respectively. The mechanism of separation and solubilization of solute in the micelles optimizes the operating conditions. Abbassi and Mulligan (2014) investigated the possibility to remove some Phenolic compounds (Tysorol and Vanillic acid) from model solution using an anionic surfactant and PES membranes.

The effect of surfactant and phenolics concentration on rejection and permeate flux were studied. Removal and recovery of surfactant by precipitation from the retentate stream in the presence of p-coumaric acid was also studied. MEUF was more effective in removing phenolic compounds than nanofiltration. The rejection was 80% which was observed to be satisfactory. Shyamal and Bhattacharya (1997) performed an experimental study to characterize the filtration of surfactant solution in terms of permeate flux and permeate concentration in both stirred and unstirred and to quantify phenol solubilization in micelle under different operating conditions. The stirrer speed was kept between 110-540 rpm for stirred cell experiment and maximum pressure was taken as 483KPa and the duration of the unstirred experiment was 60- 90 minutes. In the stirred part, there was a negligible increase in permeate flux for fixed whereas, in unstirred permeate, concentration increased with the bulk concentration. Phenol solubilizes with CPC micelles. Materna et al. (2004) developed a new method called cloud point separation to remove phenol. They tried to predict the operating parameters which affect MEUF and cloud point separation of organic toxic pollutants. A hydrophilic Millipore membrane with 10 kDa MWCO and CTAB, SDS and APG (Alkylpolyglucoside) were considered. Cloud point separation is related to

the phase phenomenon of nonionic and zwitterionic surfactant micelle systems. The rejection percentage was 90 at the end of the experiment. Polyphenols from aqueous solution were removed by Ortega et al. (2017) in one of their research work. Three surfactants, namely Esterquat, dodecylbenzenesulfonic acid sodium (DBSS) and Lutensol AO 7 (cationic, anionic and non-ionic surfactants, respectively) were used for the experiment. They investigated the efficiency of removal of Polyphenol through this process. Highest rejection of Polyphenol that occurred was 95% at pH 3 which was impressive. Removal of natural Polyphenolic compounds can also be done by this filtration process.

Table 1 Properties of phenol and its removal (Bielska and Szymanowski (2004)

Phenolic compound	Rejection%	Flux(L/m^2h)	Surfactant
Tysorol	13.56	13	CMC
Vanlic compound	33.43	7	CMC
Aromatic alcohol	97	2.22×10^{-3}	CPC
Nitrobenzene and 4-nitrophenol	92.52 93.51	2.53 1.59	CTAB+OMC-10 CTAB+ONP-9
o-cresol	75	35	CTAB & Tween -20
MNP(Meta-nitrophenol) with CC (catechol) and PNP (para-nitrophenol) with BN (Beta-phenol)	93(MNP)	$7.2 \times 10{-}6$	CPC

Phenol is more effectively removed from wastewater by MEUF when compared with other filtration methods. The MEUF process can be used very effectively and suitable for the separation of trace concentrations of organic pollutants. Salts present in wastewater do not lower the performance of MEUF, despite the fact that more rejections of solutes are observed because of increase in solubilization of solutes in micelle due to the salting out effect. On the other hand when salt concentration increases while other methods are used the rejection of solute decreases rather than increasing. While the efficiency of phenol rejection by MEUF decreases when the temperature of the surrounding increased.

Heavy Metals

Heavy metals also are known as ‘trace metals' are metals with relatively high densities, atomic weight or atomic numbers. Some of the common heavy metals include copper, iron, tin and precious heavy metals are silver, gold, and platinum. The harmful heavy metals that are present in water are lead (Pb), Cobalt (Co), Copper (Cu), Cadmium (Cd), Zinc (Zn), Nickel (Ni),Chromium (Cr),Manganese (Mn), Molybdenum (Mo), Selenium (Se), Arsenic (Ar), Antimony (Sb), Tellurium (Te), Thallium (Tl), Tin (Pb) and Mercury (Hg). Individual separation of these metals may be sometimes tough from an environmental and industrial point of view. The standard concentration in drinking water of some of the heavy metals is given in Table 2. Very low concentration of these metals can cause harmful effects on the environment, organism and even in humans. It is very important to detect the presence of these metals in drinking water as it may lead to renal failure, hair loss, and chronic anemia like diseases. It may also cause a nervous breakdown. Treatment of industrial wastewater can reduce the chances of waterborne disease in the areas where industrial wastewater is discharged. Some of the conventional methods for the treatment of these metals are precipitation, coagulation, a reduction process, ionic exchange, membrane technologies and adsorption on different adsorbents (Xie et al., 2011).

When heavy metal is available in wastewater at low concentration, adsorption is an effective process and produces high quality treated effluent (Ates et al., 2007). Biosorption process was used for the extraction of Cu^{2+} and Ni^{2+} (Fu and Wang, 2011). Biosorption is known for its high effectiveness and use of inexpensive bisorbents.

Table 2 Drinking and Discharge Standards (www.nec.gov.bt)

S.No	Compound	Drinking water standard(mg/l)	Discharge standard(mg/l)
1	Lead	0.1	0.1
2	Copper	0.05	0.5
3	Nickel	0.03	3.0
4	Chlorine	0.2-1	0.5
5	Zinc	2	5.0
6	Arsenic	0.05	0.1
7	Mercury	0.002	0.001

Precipitation is the most commonly used method due to its low-cost requirement and ease at pH control. Ion exchange method is widely used for high treatment capacity, high removal efficiency, and kinetics (Huang et al., 2010). In 1985, MEUF was studied and used for the removal of 4-tert-butyl-phenol from wastewater which showed 98% effectiveness. Therefore it is an efficient alternative for wastewater treatment (Jung et al., 2008). MEUF has an efficiency of about 92- 95% when the concentration ratio of metal (cadmium, copper, and lead) and surfactant (S/M) was 10. The COD (chemical oxygen demand) value was reduced by 80% from 2500 to 500 ppm. This was observed in soil washing effluent for heavy metal removal (Hankins et al., 2005). Huang et al. (2010) used polysulfone hollow fiber ultrafiltration membrane, with SDS as the surfactant, to treat Cd^{2+} and Zn^{2+} from wastewater. The rejection level reached 99.53 % for Cd^{2+} and 98.71% for Zn^{2+}. Li and Liu (2005) experimented for removal of metal and organic contaminants using mixed surfactants. Cu was removed with an efficiency of 92%. The surfactants that were used are SDS and Triton –X (Sodium Dodecyl Sulphate).

Ghadge et al. (2015) studied mathematical modeling for the removal of heavy metals by MEUF. They separated mixture of heavy metals ions like Cu (II), Cd (II), Ni (II), Zn (II) using SDS and Tween-20 for surfactants. It aimed to estimate the factors affecting the series model of this experiment. It used a flat sheet cross flow UF as a membrane. The nonionic to ionic surfactant mole fraction was indicated by α. The rejection of was 81% and 98.5% when α was kept 1.5 and 0.3, respectively. Nickel ions (Ni-II) were separated from wastewater by Chhatre and Marathe (2006) in one of their experimental studies. They recorded the changes, occurred by the effect of pH and by adding monovalent salts (NaCl, NaBr, and NaI). This separation utilized CTAB and SDS surfactants and polysulfone membrane. The rejection of Ni (II) ions reached 99.5 % efficiently. Das et al. (2008) study the separation of both cation and anion by mixed surfactants.

They investigated to remove the copper ions (cation) and permanganate ions (anion) and SDS and CPC were used as surfactants. Also, the effect of operating conditions like TMP, cross flow rate and composition of the feed on the rejection and permeate flux characteristics were observed. A thin film composite (TFC) membrane with 5kDa MWCO was used. The treatment was performed in 4 steps. In the first step, SDS was treated with NA_2CO_3; in the second step, Calcium Dodecyl Sulphate dissolution percent was calculated. In the 3rd step, KI to CPC ratio was subjected to the filtrate of the SDS treatment (1st step) and in the last step, the precipitated CPI was then treated with $CuCl_2$ and finally CPC was calculated. The cationic (Cu) ion rejection was 90-100% and anionic (PP) ion rejection reached 96-99%. Beolchini et al. (2006) removed arsenic (Ar) from wastewater. This study aimed at the effects of operating conditions like TMP, pH, surfactant concentration and membrane pore size on permeate flux and rejection of arsenic. CPC as a surfactant and cross-flow ceramic membrane was used. Permeability tests were done to investigate the operating conditions. The novelty was in evaluating the possible advantages of using large-pore membranes (20 and 50 nm) and reduced surfactant concentrations (1-3mM) for treating high fluxes of concentrated arsenic-bearing solutions (10-40 mg/l). Dynamic simulations were also performed.

Ahmadi et al. (1994) removed heavy metals by using Lecithin. Lecithin is a natural, nontoxic and biodegradable surfactant which was recovered by soyabean oil refining. They carried out the prediction of flux rate, rejection of metal ions, rejection of Lecithin (surfactant) and pH of the solution. The metals that were used in this experiment are Copper, Cadmium, Zinc, and Nickel. Lecithin exhibits some binding behavior in relation to Cadmium, Copper, Nickel, and Zinc. The copper recovery was noted as more than 99%. Pramauro et al. (1992) removed Uranyl from aqueous solution. They used CTAB as surfactant and cellulose membrane served the purpose of the requirement of the membrane. It was concluded that new amphiphilic ligands having selective chelating groups and modulated hydrophobic substituent could improve the efficiency of MEUF as a separation technique for metal ions.

Palladium was removed and recovered from the water by Ghezzi *et al* (2008). The surfactant and membrane treat was used in the removal were dodecyl-trimethylammonium chloride (DTAC) and UF cellulose membrane, respectively. The rejection of palladium ion was 66%. The recovery of surfactant has not been focused on this study which would make this study expensive. The surfactant that was used would go full waste after the experiment. Copper was recovered using MEUF from the waste stream by Liu and Li (2009). They used Deoxycholic acid (DCA) as a surfactant which was also recovered in this process. In the research work, the effect of ionic strength, voltage, and pH over the process was also observed. The efficiency of recovery was more than 90.5%. Gzara et al. (2000) attempted to remove lead ions from aqueous stream by micellar-enhanced ultrafiltration. He used SDS as a surfactant. Experiments were done at 318^0K and 1-3 bar of pressure. The inlet flux was up to $0.5ms^{-1}$ which was kept constant. Rejection of Lead was 99% under optimal conditions of pressure, feed concentration in SDS, tangential velocity of the feed, and percent filtered volume. Here ionic exchange model was used to study the interaction between lead ions and SDS. Also to know the performance of MEUF studies were done at constant cation concentration and various pH values. Still, the rejections of lead were more than 95%.

Rafique et al. (2016) in one of their researches investigated the optimal operating conditions for removing cadmium from wastewater. He used SDS as a surfactant and 10 kDa MWCO membrane. This was a hybrid experiment in which MEUF combined with ACF. Also, the adsorption isotherm of ACF was studied at a different concentration of SDS. When 10 kDa MWCO membrane was used, the highest rejection efficiency of cadmium was 99.6% whereas when 30 kDa MWCO membrane was used, the rejection was 99.5%.Here SDS rejection also took place with an efficiency of 90%. Danis and Aydiner (2009) experimented for the removal of nickel ions from wastewater in one of the studies. This experiment was carried out at low TMP of 250 kPa. Sodium lauryl ether sulfate (SLES) was used as a surfactant. The final flux $0.304m^3/m^2$ h was obtained. The % rejection of nickel was 98.6%. This study was performed for determining the performance of MEUF which was proved to be suitable for the removal of nickel ions.

Rahmanian et al. (2012) gave an experimental paper which was aimed at the removal of lead ions. The main objective of this study is to investigate the performance of MEUF process for lead removal and application of experimental design approach to find the optimum operating conditions of the proposed process. A statistical experimental design was applied for the investigation. The rejection obtained was 99.47% at pH=7.84. Xiarchos et al. (2008) worked for the separation of copper from aqueous solution. Here SDS as a surfactant and cellulose membranes with 10,000 MWCO were used. The pressure and temperature of the experiment were maintained at 3 bar and 25 ± 2^0C respectively. The rejection of 98.4% was obtained.

Dyes

It is a colored substance that has an affinity to the substrate to which it is being applied. It is generally applied to an aqueous solution. The dyestuff is organic compounds present in a solution which gets mixed chemically with objects. Industries like textile, paper, and plastic use synthetic dyes for coloring their products and consume water for this purpose. This wastewater is then discharged in water bodies along with these toxic dyes. The pollution caused due to these industries has become a serious problem for the environment and living beings. The presence of color is an indication of the presence of dye in the water bodies. The presence of this color

prevents the penetration of sunlight inside the water bodies where it is discharged thereby interfering with the photosynthesis process.

Classification of dye

1. Anionic dye
    - Direct dye
    - Acidic dye
    - Directive dye
2. Cationic dye
    - Basic Dyes
3. Nonionic dye
    - Disperse Dyes

Some of the methods used for the removal of dyes are electrocoagulation (Popuri et al., 2016), adsorption (Coro and Laha, 2001), enhanced softening, biosorption (Lachheb et al. 2002), UV-irradiated titania (Mondal et al., 2012), Polyelectrolyte Enhanced Ultrafiltration (Purkait et al. 2006), Reverse Osmosis and Nanofiltration are already registered as a good technique for the separation of several pollutants from wastewater. MEUF is a new technique used for the separation which a step further than NF. MEUF can successfully remove toxic dye from wastewater. Purkait et al. (2006) studied the separation of eosin dye using hexadecyl pyridinium chloride. The aim of this study was the separation of toxic eosin dye from the aqueous stream based on MEUF using continuous cross-flow cell is studied.

This experiment was carried out for almost 60 minutes and CPC was used as the surfactant. The membrane that was used is Organic Polyamide membrane with 1000 MWCO This membrane was used for both batch and cross-flow cell. Up to 74% of eosin was retained by using MEUF. Khosa et al. (2011) gave an experimental study on the removal of Alizarin Red S (ARS) Dye by the combination of UV-Visible Spectrometric Study and Micellar-Enhanced Ultrafiltration. In this experiment, 3 distinct surfactants were used: CTAB, SDS, and TX-100. UV-visible absorption spectra were recorded in which matched pair of quartz cells with 1 cm optical length was employed at 25^0C. Regenerated cellulose membrane of 76 mm diameter with 30 kDa MWCO was used. The rejection of ARS for CTAB surfactant was maximum with 98.8% at various concentrations. The presence of $CuCl_2$ is helpful dye rejection.

Sharma and Purkait (2016) investigated the blending of $_D$-TA and $_{DL}$- TA in PSn membrane and their performance in the removal of crystal violet dye (CVD) from aqueous solution. Here 3 membranes with different compositions were used and they are named as M1, M2, and M3.M1 and M2are almost identical but the pore size distribution (PSD) of M3 is different. The highest rejection was obtained while using M2 membrane i.e. 99.2% with SDS at pH 11. Zaghbani et al. (2008) discussed the removal of Safranin T (dye) through MEUF. Safranin T is a water-soluble phenazine dye which is reddish-brown powder. It is mainly used for flavoring of food, coloring candies, and cookies. It may also be used for dying tannin, cotton, bast fibers, wool, silk, leather, and paper. The objective of this paper was to evaluate the efficiency of MEUF for dye removal. SDS (cationic in nature) was used as a surfactant. Regenerated cellulose membrane with 10 kDa in cross-flow filtration was used. The retention % for the Safranin T was 99%. This percentage shows that MEUF is efficient enough in the extraction of Safranin T from wastewater.

Zghbani et al (2009) have studied the effect of chemical parameters such as ionic strength, the chain length of surfactant and pH on the extraction of direct blue 71(DB71). DB71 is an anionic industrial dye. CTAB, SDS, and Triton-100 as surfactants and cellulose membrane of 10 kDa MWCO were used. The rejection of RB71 that was recorded is 98%. Zaghbani et al. (2009) Eriochrome removed Blue Black R (EBBR) from dye-polluted wastewater. The effects of dye and surfactant concentrations, ionic strength, pH and transmembrane pressure were investigated in this paper. A hydrophilic membrane made of cellulose (molecular weight cut-off 10 000 Da) was used here and N-Alkyltrimethylammonium bromide i.e. dodecyltrimethylammonium bromide (C12TAB), tetradecyltrimethylammonium bromide (C14TAB), cetyl-trimethylammonium bromide (C16TAB) and octadecyltrimethylammonium bromide (C18TAB) were taken as cationic surfactants. From the experimental results, the retention rate of dye was about 99% in presence of C18TAB and C16TAB. Khosa et

al. (2011) aimed to study the working principle of MEUF from the perspective of osmotic or Donnan equilibrium between two pseudo phases and the thermodynamic aspect. SDS, CTAB & CPC were used as surfactants. The rejection of organic dye reached 99% when removed by MEUF.

Phosphorous and Phosphate Removal

Baek et al. (2004) removed phosphorous by using MEUF. They studied the removal efficiency and flux decline of phosphorous removal and also the effect of pH on the efficiency. CPC was used as a surfactant in this experiment. The area of the membrane was 45.6 cm^2 of regenerated cellulose acetate with molecular weight cut-off (MWCO) of 3,000 and 10,000, and transmembrane pressure was adjusted to 2 bar. The pH of Na (H_4PO_4) solution was adjusted to 5 and 9 for $Na_3(HPO_4)$ solution. With 97% efficiency, phosphorous was retained at the end. They opined that MEUF is a good alternative for phosphorous removal.

Camarillo et al. (2009) used CTAB as a surfactant to remove phosphate ions from wastewater. They aimed at the removal of phosphorous along with the operating conditions like pH, a temperature which governs the experiment. The rejection of phosphorous was 95%. CTAB was concluded as best among surfactants like CPC and ODA. Misra et al. (2009) performed an experiment to remove di-butyl phosphate (DBP) and tributyl phosphate (TBP) as well as uranyl ions from aqueous solutions by using Micellar-Enhanced Ultrafiltration Technique (MEUF). The 47 mm diameter Millipore ultrafiltration membranes of 3000, 5000 and 10,000 MWCO pore sizes were used and the pressure of the ultrafiltration cell was kept at 1.7 bar for all the experiments. DBP and TBP with an efficiency of 90% and > 80% of uranyl ions.

Technology Development

The effect on the fluorescence characteristics of 2, 3-diaminophenazine (DAPN), which is the product of the lactase-catalyzed reaction with o-phenylenediamine (1, Zdiaminobenzene) as the substrate is studied in an experimental paper by Huang et al. (1995). This paper showed that the fluorescent intensity of DAPN can be improved by the different surfactant. Brij-85 is considered as the best one so far. Also, MEUF can be used to assay for laccase activity with high sensitivity activity with high sensitivity. SDS and CTAN were used as surfactants. An automated flow-injection stopped-flow kinetic spectrofluorimetric method for determination of lactase activity was used here. It is based on the oxidation of o-phenylenediamine (1, Zdiaminobenzene) catalyzed by lactase during the stopped-flow period, yielding 2, 3-diaminophenazine, which is determined by a micelle enhanced spectrofluorimetric method in a non-ionic surfactant medium.

Manchalwar et al. (2010) did a simulation of Micellar-Enhanced Ultrafiltration by multiple solute models. This model is characterized by the parameters, membrane resistance R_m, membrane permeability Pm, back transport coefficient K_b, K_{bi} and mass transfer coefficient k_i. In this model, cross currents are formed and some solutes get removes by membrane surface and go into the bulk known as back transport. This back transport is significant in micellization. They aimed to choose a suitable model for predicting the capacity of MEUF. And the parameters are estimated like pH, chelating agent /metal ratio (C/M). SDS was used as a surfactant and polyethersulfone having 30kDa MWCO were used here. The effective membrane area used 0.02m^2. The separation of copper and cobalt is done here with the help of MEUF. Naphthenic acid (NA) was removed was removed by Venkataganesh et al. (2011) in one of their experiments.

They aimed at the removal of NA using hollow fiber membranes and ceramic membranes with the help of MEUF. The effect of the electric field for the reduction in concentration polarization is also investigated. Dc power was passed across the membrane externally. Six different powers were used for experiment 0, 200, 400, 600, 800, 1000 V/m.This was applied in two phases. One in which the experiment is started at 0 V/m and is then increased till 1000V/ m and in the other phase the electric field is kept constant. SDS was used as a surfactant. The rejection of NA obtained was 98% after the experiment. Lipe et al. (1996) used air stripping which was merged with MEUF for the removal of contaminants. They aimed to reuse the surfactant that was used during the experiment and to predict the operating conditions. Air stripping is shifting of toxic contaminants of a liquid into an air stream. It is a technique used for the treatment of wastewater and

groundwater containing toxic materials. At the top of a packed air stripping tower, an aqueous stream is cast and the air is introduced at the bottom that creates concurrent flow. Air stripping follows two assumptions:

- Only the extra micellar portion of the contaminant determines the mass transfer driving force in air stripping.
- The surfactant has no significant effect on the interfacial rate of mass transfer of volatile materials across the air/water interface.

Overdevest et al. (2002) separated enantiomer in a cascaded MEUF system. They tried for model development which would explain the separation in multiple stages in that system using the enantioselective equilibrium complexation model. In their model, at first, a cascaded system that contained five stages was used then one single-stage bench-scale system was used to simulate the separation in a cascaded system of 60 stages. Model calculations show that the separation of enantiomers in a cascaded system is only successful within a certain_-window Z senantiomer feed concentration multiplied by its affinity constant. The study showed the suitability of microheterogeneous media Z in case of micelles in cascaded ultrafiltration systems for molecular separations.

Sabate et al. (2002) conducted an experiment for the removal of phenol by MEUF and different membrane properties were compared for its effect over the separation process. CPC was used as a surfactant and 2 polysulfone membranes with 5 (P5) and 50 (P50) kDa MWCO and 2 ceramic membranes with 15 (C15) and 50 (C50) kDa MWCO was used in the removal process. The comparison between these two membranes materials shows how they can affect the filtration process.C-50, the combination of a pores size and the electrical charge of membrane and micelle. Total filtration area of 120.9 cm^2**.** Baek and Yang (2004) used CPC as a surfactant for removal of nitrate and chromate. Competitive binding of CPC micelle was established between chromate and nitrate. The retention of nitrate and chromate was 91% and >99%, respectively. Cross-flow Micellar-Enhanced Ultrafiltration was used for this removal. It is better than the ion exchange or reverses osmosis processes through competitive binding.

Jadhav and Marathe (2013) gave a comparative experimental paper which provided information about the filtration capacities of ceramic and polymeric membranes. It also shows which membrane material affects the MEUF process more. Polysulfone membrane was more suitable for MEUF. However, the hollow fiber membrane gave higher rejection, low operating cost and better separation quality than ceramic membranes. Cetyltrimethyl ammonium bromide was used as a surfactant. A pilot scale plant was used for all experimental purposes. The membranes used were, tubular hollow fibre ultrafiltration Microza module having MWCO 6 kDa with polysulfone membrane and ceramic membrane Membralox module having pore diameter 50nm with an active layer of microporous zirconia (ZrO_2) supported on α-alumina, respectively The rejection of phenol and o-cresol reached 95% using tubular polysulfone hollow fiber and zirconia (based on alumina) ceramic membranes .

Li and Liu (2005) combined electrolysis with MEUF process which is useful for metal removal. The proposed hybrid system is operated as continuous adsorption (by MEUF) and regeneration (by electrolysis) process. Adsorption is present in many natural, physical, biological and chemical systems and is applicable in industries such as heterogeneous catalysts, activated charcoal This study compares the removal efficiency of metal by given hybrid system with other methods of metal removal like electrolysis membrane, electrolysis only and MEUF. The operating condition such as pH, the concentration of surfactant, current density, and the HRT (hydraulic residence time) that affects the system is also investigated. SDS is used as a surfactant here. MEUF was used to retain metals inside the reactor, the deployment of the electrolytic process the removal in the reactor. The samples were measured using a flame atomic absorption spectroscopy. MEUF shows that this hybrid process has better efficiency in the removal of copper ions as compared to other processes. The hybrid process showed 70 to 96% of rejection while MEUF alone showed 64.34- 95.8% of rejection. Krivorot et al. (2009) mentioned Centrifugal MEUF in one of his papers. They aimed at the partition coefficient of TBNPA between an aqueous phase and micelles by two methods: centrifugal MEUF and SPME in the presence and absence of 1% NaCl and toluene. For Centrifugal MEUF Amicon Centriprep YM membrane tubes made of regenerated cellulose were used. The centrifugal method is used to separate particles from a solution according to their size, shape, density, the viscosity of the medium and rotor speed. This process is used to separate two miscible

substances, but also to analyze the hydrodynamic properties of macromolecules. They have an effective filtration area of 2.84 cm^2, MWCO of 10 kD, and a maximum volume of 15 ml. Two methods were used here. SPME and Centrifugal MEUF. The rejection was 85%.

Materna et al. (2004) mentioned cloud point technique with MEUF in one of his papers. Cloud point separation technique separates the micellar solution into two phases. Any solution heated above the cloud point temperature during a particular time two phases are formed, one which contains most of the surfactant which is a concentrated phase and another low concentrated aqueous phase. This paper aimed at the removal of phenol and phenylamines by using the above two methods. Cloud point separation (CP) technique is better at filtration when compared to the UF technique. CTAB and SDS are the surfactants that were used. The retention was 60% by MEUF. CP technique gives better results in the separation of pollutants.

New Technologies

Fig. 1 shows a five-stage system was operated in the countercurrent mode. Each stage consisted of a stirred vessel with 0.5 L micellar solution and a hollow-fiber membrane module. This is the bench scale crossflow system which was used in the Enantiomer Separation (Overdevest et al., 2002). This Fig. 2 represents the systematic diagram of Ultrafiltration unit which was used in the separation of separation of phenol with the help of Gemini surfactant and the whole process was named as Gemini Micellar-Enhanced Ultrafiltration (GMEUF) (Zhang et al., 2013).

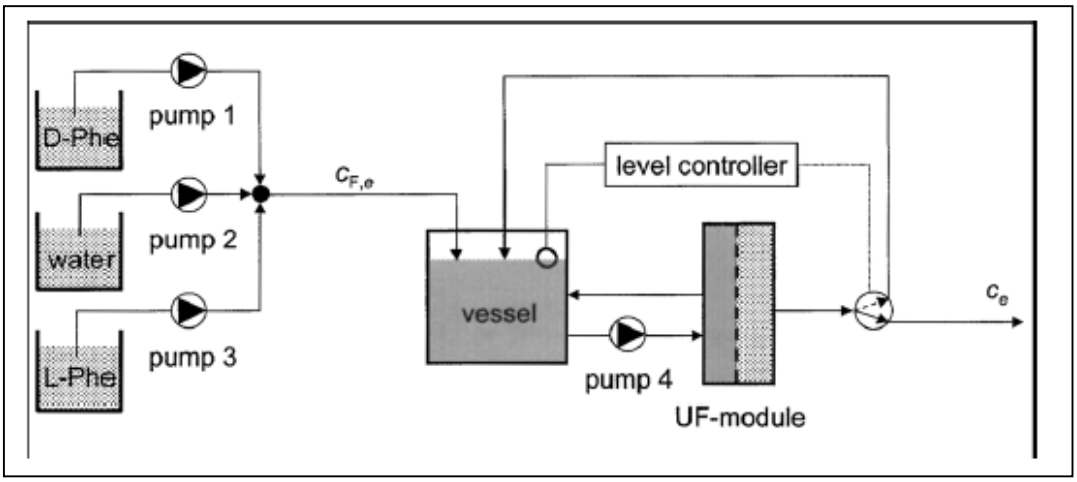

Figure 1. Cascaded System (Overdevest et al., 2002)

CRITICAL ANALYSIS

Operational difficulties

With the increase of the concentration of surfactant, the rejection of the contaminant increases with the passage of time. Researchers made an attempt to remove contaminants without surfactant but the retentions were found to be very low and therefore, the mixing of surfactant was required for maximum retention of pollutants. For this, there are several membranes available which can be used. To recover the initial permeability of the membrane, it is washed with deionized water after conducting each experiment. The difference in flux may affect the removal efficiency of the contaminant. The sufficient CMC (Critical Micelle Concentration) of surfactant must reach for better removal of pollutant. When the CMC is below or above the required amount, there is a need of addition of surfactant. If there is an increase or decrease in CMC concentration, the surface tension also increases or decreases with it. To avoid membrane clogging, the reducing agent concentration is increased from the required concentration. There are three types of problems that are faced by membrane surfaces: hydraulic resistance, gel type layer and pore blocking. Blocking filtration laws consist of four different filtration mechanisms: complete blocking, standard blocking, intermediate blocking, and cake filtration. These laws are used in membrane filtration.

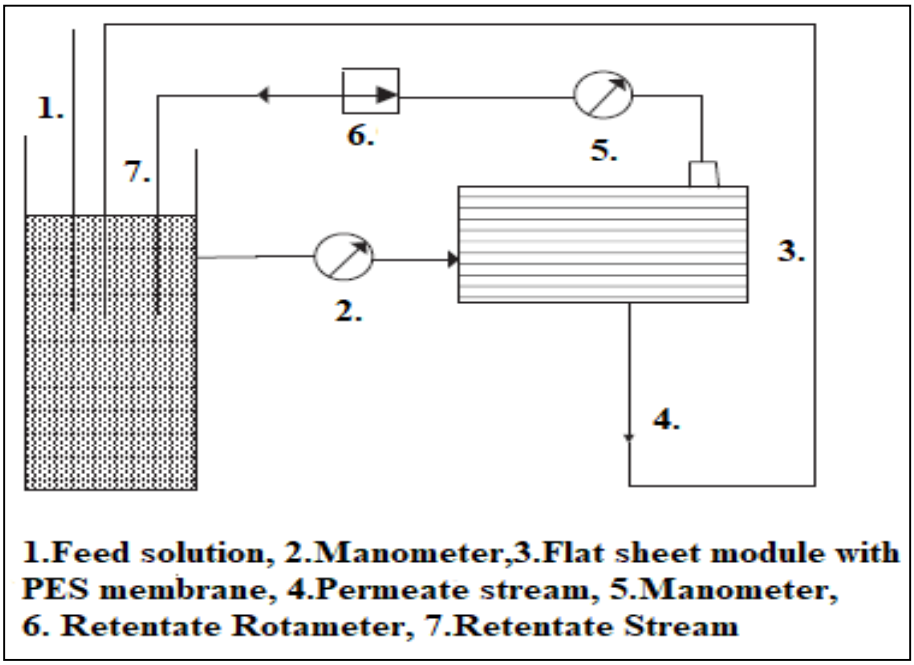

Figure 2. Gemini Miceller Enhanced Ultrafiltration (GMEUF)

The initial pore blocking frequently causes the irreversible fouling of membranes, resulting in a decrease in the efficiency of membrane cleaning. Once a sufficient number of pores on the membrane gets clogged there is a formation of an external cake-like structure over the fouled membrane (Nabetani et al., 2009). This layer that is formed after the filtration process over the membrane is known as the Gel type layer. This layer exhibits rejection ability for solutes and little resistance for permeate flow (Sahin and Taşcıoglu 2016). The resistance due to pore blocking is higher than the gel type layer which is responsible for the decrease of flux. Gel type layer is the smallest resistance but has its effects. It increases with the increase in feed concentration and the concentration of micelles also increases which provides resistance against the solvent flux. Resistance series has been mentioned in this paper.

According to resistance-in-series models, the flux decline is due to the combined effects of (i) fouling of the membrane (reversible and/or irreversible pore blocking) and (ii) concentration polarization over the membrane surface (Hurwitz et al., 2015). Concentration polarization refers to the emergence of concentration gradients at a membrane/solution interface resulted from the selective transfer of some species through the membrane under the effect of transmembrane driving forces. Concentration polarization leads to increased salt leakage through the membrane, increased probability of scale/fouling development. This concentration polarization is caused by the accumulation of retained solutes or particles on the membrane surface. Fouling can occur at the membrane surface due to scale formation, where the surface of the membrane is coated by a thin non-removable deposit of chemicals. A common type of scale formation mechanism is through precipitation of inorganic material like calcium sulfate or silica when their solubility limits are exceeded (Zeng et al., 2008).

There are various types of difficulties which are faced by membrane while the filtration process. They are as follows: cake filtration, intermediate blocking, standard blocking, and complete blocking. The cake filtration refers to a cake like a layer formed over the membrane after the experiment. In intermediate blocking the particles settle over already blocked or occupied membrane. Settlement of particles occurs inside the pores of the membrane in standard blocking. While in complete blocking, the membrane gets fully blocked by the settling of the particles in the pores (Fillipi et al., 1998). Each filtration cycle consists of the filtration and the backwashing/backflushing steps. While filtration the surfactants along with the metal ions gets clogged on the membrane which is removed by backwashing/backflushing (Liu and Li, 2005). Backwashing/Backflushing refers to pumping water backward through the filters media, sometimes including intermittent use of compressed

air during the process. Backwashing is a form of preventive maintenance so that the filter media can be reused. Fouling can limit the permeate flux significantly and add to the frequency and cost of membrane cleaning. Hence, it is of crucial importance to adjust the hydrodynamic conditions of the system such that fouling is controlled and possibly eliminated. Biofouling is a membrane fouling which is a critical issue. This occurs due to the formation of low permeability biofilm on the surface of the membrane.

This includes (a) the adsorption of organic species and suspended particles on the wetted membrane surface to form a conditioning film; (b) the transport of the microbial cells to the conditioning film; (c) the attachment of the microbial cells to the membrane surface; (d) the growth and metabolism of the attached microorganisms and biofilm development; (e) the limitation of biofilm growth by fluid shear forces (detachment process) to achieve a steady state fouling resistance. Due to this biofouling, there is an increase in pressure and energy consumption and decline of flux occurs (Deriszadeh et al., 2008)

Economic and energy analysis

Higher reagent and electrical costs are higher in conventional MEUF when compared with the hybrid process with MEUF. The hybrid process requires less surfactant. Fillipi et al. (1998) compared the cost of the LM-MEUF copper separation method with the conventional copper solvent extraction process for 1105 gal/day (gpd) plant with feed copper concentration of 190 ppm (3mM) and with a discharge stream copper concentration of 4 ppm (0.063 mM). It was concluded that the operating cost for ligand modified MEUF (LM-MEUF) is higher than the solvent extraction mainly due to higher reagent and power costs. One of the main disadvantages of MEUF is the leakage of surfactant. The surfactant contains a major part of the operating cost and surfactant leakage increases the cost of the whole process. Anionic surfactants are major when it comes to cost. To reduce this cost and make the process cost efficient, the surfactants should be further reused. Larger pore sized membranes are preferred in UF to obtain larger fluxes.

This results in the requirement of lower membrane surface area, capital and membrane replacement costs. The preferred flow rate of permeate is thrice the retentate to reduce the cost associated with the process. A big issue is the emission of ligand and surfactant in a large amount with the stream. Hence foam fractionation can be used to avoid this situation and it is stated to recover up to 90% of a surfactant. The annual cost of ligand is much higher than the surfactant. The hollow fiber membrane gives greater separation with less power consumption than the ceramic membrane as calculated energy consumption for hollow fiber and the ceramic membrane is 6401 and 10167.5 kJ/year. Also, the hollow fiber membrane is more cost efficient than the ceramic membrane (Zhang *et al.* 2016). Markels et al. (1995) discussed an alternative based on a physically consistent permeate flux model that applies over the entire operating range. By using large pore tubular module in the first stage the prefiltration cost can be minimized. There is also a depreciation cost for the membrane and pump which needs to be considered.

CONCLUSIONS

In this paper, we have reviewed the application of Miceller Enhanced Ultrafiltration (MEUF) method in removing the various kinds of dissolved toxic and foreign pollutants in the water. The efficiency of removing the extra products from water by MEUF has its own advantages and disadvantages. The experiment becomes uneconomical due to the surfactants and the membranes. It has been reported that due to prolonged uses, the pores of the membranes get blocked and needs to be cleaned regularly. In some cases, the surfactants are not recovered after the experiments. The efficiency of MEUF is also affected by the operating conditions of the experiment e.g. pH, temperature, turbidity, Total suspended solids and concentration of surfactants. Some of the researchers have combined Activated Carbon Fibres (ACF) with MEUF to form hybrid and more efficient treatment. A hybrid process of combining electrolysis and MEUF were also successfully applied. Higher reagent and electrical costs are reported in conventional MEUF method as compared to the hybrid process with MEUF. The hybrid process requires fewer surfactants.

Available research focused on the removal of metals, phenol, organic & inorganic materials, aromatic hydrocarbons by MEUF mainly on the type of surfactant used, surfactant concentration, applied pressure,

operating time and pH. Formation of surfactant micelle and mechanisms for the attraction between micelle and metal ions are ignored during the research study on MEUF. The MEUF process showed its use not only in synthetic streams but also in combination with the other processes to carry out removal in the waste stream. It showed its usefulness for aqueous and non- aqueous streams when combined with the other processes. Researchers are more attracted to the different types of membranes and its applications. For the removal of contaminants, MEUF is considered as a better alternative to the typically available membrane separation processes. The advantages of this method over other methods are high removal efficiency, low energy consumption, and easy operation. The selection of surfactant for the removal purpose is based on the target pollutants. The characteristic of the membrane used also has an effect on the removal efficiency of the process. The membrane fouling is affected by the chemical nature of the membrane materials.

The rejection of target pollutants does not dependent on the initial amount of surfactant used but depends on its concentration near the membrane surface. MEUF is successfully applied for single contaminants with or without surfactants. For the removal of mixture contaminants, the addition of the mixed surfactant system, ligands, and chelating agents showed better rejection than using a single surfactant. The CMC of surfactant decreased when mixing with another surfactant to show a better result. The separation of a mixture of pollutants or other compounds using MEUF depends on the valance of the species. The species with greater valance attracts more towards micelles than the species having a lower valance to cause better rejection of it. Due to the requirement of less amount of surfactant, the combination of MEUF with other processes needs the attention of the researchers. More hybrid methods that combine MEUF and other separation methods could be used for better results in the rejection of solutes.

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