Removal of heavy metals from wastewater by membrane processes: A review

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Abstract

In this paper a general review on removal of heavy metal from wastewater by different membrane processes wasdone. As the main aim of this paper is to review the application of membrane processes in removal of heavy metalfrom wastewater, processes such as composite membranes, Polymer-supported ultrafiltration, multiple membrane,complexation –ultrafiltration, surfactants-enhanced ultrafiltration, combining process, MEUF technique, flotationwith membrane were reviewed.

Keywords : heavy metals, membrane proesses , wastewater, removal, ultrafiltration, nanofiltration, reverseOsmosis

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1. Introduction

Pollution of water sources is usually due to human activities that lead to the presence of toxic species like pesticides, micro organisms, dyes and heavy metals in surface water, and eventually in ground water. Heavy metals are well known for their high toxicity for human beings. They are soluble in water as ions or chemical complexes and they could be ingested if water is not correctly treated [1].

Due to the discharge of large amounts of metal-contaminated wastewater, industries bearing heavy metals, such as Cd, Cr, Cu, Ni, As, Pb, and Zn, are the most hazardous among the chemical-intensive industries. Because of their high solubility in the aquatic environments, heavy metals can be absorbed by living organisms. Once they enter the food chain, large concentrations of heavy metals may accumulatein the human body. If the metals are ingested beyond the permitted concentration, they can cause serious health disorders [2].

It is not possible to discharge industrial wastewater contaminated with heavy metals directly into rivers, water reservoirs or into the sea because the heavy metals pose a serious risk to humans, animals and the environment. Indirect discharge of this wastewater into municipal sewage treatment plants may have a major effect on the activated sludge and hinder the efficiency of the plants [3].

However, increased concentrations of heavy metals in wastewater result in the reduction of the biological activity of microorganisms and finally in the total containment of microbial growth that this item means reduce the efficiency of treatment [4].it is necessary to treat metal-contaminated wastewater prior to its discharge to the environment. Heavy metal removal from inorganic effluent can be achieved by conventional treatment processes such as chemical precipitation, ion exchange, and electrochemical removal. These processes have significant disadvantages, which are, for instance, incomplete removal, high-energy requirements, and production of toxic sludge [5].

Membrane is defined essentially as a barrier, which separates two phases and restricts transport of various chemicals in a selective manner. A membrane can be homogenous or heterogeneous, symmetric or asymmetric in structure, solid or liquid; can carry a positive or negative charge or be neutral or bipolar. Transport through a membrane can be affected by convection or by diffusion of individual molecules, induced by an electric field or concentration, pressure or temperature gradient. The membrane thickness may vary from as small as 10 microns to few hundred micro meters[6].

Membrane separation has been increasingly used recently for the treatment of inorganic effluent due to its convenient operation. There are different types of 2 membrane filtration such as ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) [7].This article presents an overview of various membrane processes for removal of heavy metals from wastewater.

2. Heavy metal and quantity of toxicity

Heavy metals are generally considered to be those whose density exceeds 5 g per cubic centimeter. A large number of elements fall into this category, but the ones listed in Table 1 are those of relevance in the environmental context. Arsenic is usually regarded as a hazardous heavy metal even though it is actually a semi-metal. Heavy metals cause serious health effects, including reduced growth and development, cancer organ damage, nervous system damage, and in extreme cases, death. Exposure to some metals, such as mercury and lead, may also cause development of autoimmunity, in which a person's immune system attacks its own cells. This can lead to joint diseases such as rheumatoid arthritis, and diseases of the kidneys, circulatory system, nervous system, and damaging of the fetal brain. At higher doses, heavy metals can cause irreversible brain damage. Children may receive higher doses of metals from food than adults, since they consume more food for their body weight than adults. Wastewater regulations were established to minimize human and environmental exposure to hazardous chemicals. This includes limits on the types and concentration of heavy metals that may be present in the discharged wastewater. The MCL standards, for those heavy metals, established by USEPA are summarized in Table 1 [8].

3. Membrane processes

Membrane separation processes are increasingly used as an alternative to conventional industrial wastewater treatment. There are several advantages of using membranes in water and wastewater treatment [9]. Reuse of water from industrial processes is an important business goal, especially when the industry is using large amounts of water. This goal can be achieved through industrial wastewater treatment by membrane processes. Using these processes will also enable the industry to comply with the effluent standard limits imposed by environmental regulations [10].

Membrane filtration Depending on the size of the particle that can be retained, various types of membrane filtration such as ultrafiltration, nanofiltration and reverse osmosis can be employed for heavy metal removal from wastewater.

Ultrafiltration (UF) utilizes permeable membrane to separate heavy metals, macromolecules and suspended solids from inorganic solution on the basis of the pore size (5–20 nm) and molecular weight of the separating compounds (1000– 100,000 Da). These unique specialties enable UF to allow the passage of water and low-molecular weight solutes, while retaining the macromolecules, which have a size larger than the pore size of the membrane [11].

Reverse osmosis (RO) and nanofiltration (NF) are relatively new processes, which were initially developed for the production of potable water from saline and brackish water. However, the development of these processes has been accelerated in recent years and has begun to find applications in the treatment of industrial wastewater. Recent research indicates that wastewater reclamations by RO offers great promise for a sustainable reduction in cost, conserving natural resources, as well as marked improvements of pollutant removal efficiency.[12]

Abu Qdaisa and Moussab [12] investigated the application of both reverse osmosis (RO) and nanofiltration (NF) technologies for the treatment of wastewater containing copper and cadmium ions. The results showed that high removal efficiency of the heavy metals could be achieved by RO process (98% and 99% for copper and cadmium, respectively). NF, however, was capable of removing more than 90% of the copper ions existing in the feed water.

Table 1: The MCL standards for the most hazardous heavy metals (Babel and kurinawan, 2003)

Heavy metal	**Toxicities**	**MCL(mg/l)**
Arsenic	Skin manifeststions, visceral cancers, vascular disease	0.050
Cadmium	Kidney damage, renal disorder, human carcinogen	0.01
Chromium	Headache, diarrhea, nausea, vomiting, carcinogenic	0.05
Copper	Liver damage, Wilson disease, insomnia	0.25
Nickel	Dermatitis, nausea, chronic asthma, coughing, human carcinogen	0.20
Zinc	Depression, lethargy, neurological signs and increased thirst	0.80
Lead	Damage the fetal brain, diseases of the kidneys, circulatory system, nervous system	0.006
Mercury	Rheumatoid arthritis, and diseases of the kidneys, circulatory system, nervous system	0.00003

3.1. Composite membrane

Juang and Shiau [13] studied the removal of Cu(II) and Zn(II) ions from synthetic wastewater using chitosan-enhanced membrane filtration. The amicongenerated cellulose YM10 was used as the ultrafilteration. About 100% and 95% rejection were achieved at pH ranging from 8.5 to 9.5 for Cu(II) and Zn(II) ions, respectively. The results indicated that chitosan significantly enhanced metals removal by 6– 10 times compared to using membrane alone. This could be attributed to the major role of the amino groups of chitosan chain, which served as coordinationsite for metal-binding. Steenkamp et al [14] to manufacture a composite alumina/chitosan membrane, which could remove Cu2+ ions in large quantities to values below the 1.3 mg/L level.

Saffaj et al [15] employed low-cost ZnAl2O4–TiO2 UF membranes to remove Cd(II) and Cr(III) ions from synthetic solution. They reported that 93%Cd(II) rejection and 86% Cr(III) rejection were achieved. Such high rejection rates might be attributed to the strong interactions between the divalent cations and the positive charge of the membranes. These results indicate that the charge capacity of the UF membrane, the charge valencies of the ions and the ion concentration in the effluent,played major roles in determining the ion rejection rates by the UF membranes. Depending on the membrane characteristics, UF can achieve more than 90% of removal efficiency with a metal concentration ranging from 10 to 112 mg/L at pH ranging from 5 to 9.5 and at 2– 5 bar of pressure [16].

Lv et al [17] investigated amphoteric polybenzimidazole nanofiltration hollow fiber membrane for both cations and anions removal NF membranes perform separation in between those of UF and RO ones. The molecular weight of the solute that is 90% rejected by NF membrane range from 200 to 1000 Da with pore diameters varying from 0.5 to 2 nm.

Boricha and murthy [18] prepare new blend membranes with different compositions of acrylonitrile butadiene styrene (ABS) and chitosan (CHS) on polyethersulfone (PES) substrate membrane. These membranes are used to separate mercury and sodium ions from aqueous solutions at different operating conditions. The results showed that the maximum rejections of mercury and sodium ions are 96.25% and 89.74% for ABS membrane.

3.2. PSU technique

Polymer-supported ultrafiltration (PSU) technique has been shown recently to be a promising alternative for the removal of heavy metal ions from industrial effluent (Rether and Schuster, 2003).This method employs proprietary water-soluble polymeric ligandsto bind metal ions of interest, and the ultrafiltration technique to concentrate the formed macromolecular complexes and produce an effluent, essentially free of the targeted metal ions. Polyamidoamine dendrimers (PAMAM) have been surface modified, using a twostep process with benzoylthiourea groups to provide a new excellent water-soluble chelating ion exchange material with a distinct selectivity for toxic heavy metal ions. Studies on the complexation of Co(II), Cu(II), Ni(II), Pb(II) and Zn(II) by the dendrimer ligand were performed using the PSU method. The results show that all metal ions can be retained almost quantitatively at pH 9. Cu(II) form the most stable 4 complexes with the benzoylthiourea modified PAMAM derivatives (can be completely retained at pH >4), and can be separated selectively from the other heavy metal ions investigated [19].

3.4. Multiple membrane

A multiple membrane process was developed for selective separation to reduce cost and mitigated the increasing heavy metal pollution. The process was divided into three stages: firstly, microfiltration (MF) and UF were used to separate the possible organic and suspended matters, secondly, electrodialysis (ED) was carried out for effective desalination, and thirdly, the concentrate from ED was treated by NF and RO separately to increase the recovery rate of water. Results showed that filtration characteristics of UF membrane here was not so good as is usually, even if compared with MF membrane. And RO performed better than NF in wastewater separation, especially in anti compacting ability of membrane [20].

3.5. Complexation–ultrafiltration

Recently, the complexation–ultrafiltration technique has been shown to be a promising technique for removal of heavy metals in solution. this technique, proves to be a promising alternative to technologies based on precipitation and ion exchange. The use of water-soluble metal-binding polymers in combination with ultrafiltration (UF) is a hybrid approach to concentrate selectively and to recover valuable elements as heavy metals. In the complexation–UFprocess cationic forms of heavy metals are first complexed by a macroligand in order to increase their molecular weight with a size larger than the pores of the selected membrane that can be retained whereas permeate water is then purified from the heavy metals [21].

Water-soluble polymeric ligands have shown to be powerful substances to remove trace metals from aqueous solutions and industrial wastewater through membrane processes. Carboxyl methyl cellulose (CMC) [21]; diethylaminoethyl cellulose [22], polyethyleneimine (PEI) [23] polyvinylethylenimine [24,25] and polyvinyl alcohol [26], poly(acrylic acid) [27,28] were used as efficient water-soluble metalbinding polymers in combination with ultrafiltration (UF) for selective removal of heavy metals from water. Barakat [29] investigated the removal of Cu(II), Ni(II), and Cr(III) ions from synthetic wastewater solutions by using Carboxy methyl cellulose and polyethersulfon ultrafiltration membrane. Llanos et al [30] employed a polymerenhanced UF system to remove copper fromsynthetic wastewater, achieving removal efficiencies higher than 97%. Borbely and Nagy [31] applied a polymer-enhanced UF process combined with complexation to remove nickel and zinc from industrial wastewater achieving removal efficienciesof93%andhigherthan99% respectively at pH=9.

3.6. surfactants-enhanced ultrafiltration

Ferella et al [32] examined the performance of surfactants-enhanced ultrafiltration process for removal of lead and arsenic by using cationic (dodecylamine) and anionic (dodecylbenzenesulfonic acid) surfactants. The removal of lead ions was >99%, while with arsenate ions it was 19%, in both the systems. Modified UF blend membranes based on cellulose acetate (CA) with polyether ketone [33], sulfonated polyetherimide (SPEI) [34], and polycarbonate[35] were recently tested for heavy metals removal from water.

Yang et al [36] employed ultrafiltration effectively for removal of heavy metals and organic pollutants from contaminated-groundwater and wastewater, as part of a surfactant enhanced aquifer remediation process to concentrate and reuse the surfactant.

3.7. Combining process

A new integrated process combining adsorption, membrane separation and flotation was developed for the selective separation of heavy metals from wastewater. The process was divided into the following three stages: firstly, heavy metal bonding (adsorption) by a bonding agent, secondly, wastewater filtration to separate the loaded bonding agent by two variants: crossflow microfiltration for low-contaminated wastewater , or a hybrid process combining flotation and submerged microfiltration for highly contaminated wastewater , and thirdly, bonding agent regeneration. Synthetic zeolite R selected as a bonding agent, was characterized and used for the separation of the zeolite loaded with metal [3]. Bloocher et al [37] Nenov et al [38] developed a new hybrid process of flotation and membrane separation by integrating specially designed submerged microfiltration modules directly into a flotation reactor. This made it possible to combine the advantages of both flotation and membrane separation. The feasibility of this hybrid

process was proven using powdered synthetic zeolites as bonding agents. The toxic metals, copper, nickel and zinc, were reduced from initial concentrations of 474, 3.3 and 167 mg/L, respectively, to below 0.05 mg/L, consistently meeting the discharge limits. matis et al[39] investigated the removal of two metal ions (copper or chromates) by employing a hybrid flotation-microfiltration process. Promising results were obtained: the concentration in the outlet was 0.5 mg /L for copper and 0.1 mg /L for chromium, from initially 50 mg /L.

3.8. MEUF technique

One of the developed water treatment processes is micellar-enhanced ultrafiltration (MEUF) that has beenshown to be an efficient technology for the removal of heavy metals even in low concentrations [40]. MEUF was introduced by Scamehorn in the 1980s to remove copper ions from wastewater. In this process, an anionic surfactant at a concentration higher than its critical micellar concentration (CMC) is added to the aqueous stream containing the dissolved solutes. Micelles with negative charges cause the cations to bind to or adsorb onto the micelle interface. Because of the strong tendency of the metal ions to be attracted by the micelle surface, a large fraction of the metal ions can be trapped by micelles. The stream is then treated with an ultrafiltration system using a membrane having pore sizes, small enough, to obstruct the passage of micelles, thereby rejecting the adsorbed cations. The unbound cations and the surfactant monomers pass through the ultrafiltration membrane to the permeate side [41].

In MEUF, anionic surfactants are added into the wastewater and the surfactant monomers aggregate and form micelles above the critical micelle concentration (CMC). The heavy metal cations will mostly be trapped in the outer part of the micelles due to electrostatic interaction and will be retained by the ultrafiltration membrane. The untrapped heavy metals and the free surfactant monomers readily pass through the UF membrane. The advantages of this method are the low energy requirements involved in the UF processes and the high removal efficiency owing to the effective trapping of metals by the micelles [42].

MEUF, using anionic surfactants at concentrations much higher than the CMC, has proven to be very effective in removing metals from aqueous streams. Since anionic surfactants have a relatively high CMC, large quantities of surfactant must be used to bring about an effective separation. Consequently, the economic viability of the MEUF process strongly depends on the ability to recover or recycle a significant portion of the surfactant from the retentate [43]. An additional limitation of the MEUF process is the inevitable leakage of surfactant monomers (surfactant molecules not in the micelle form) through the ultrafiltration membrane. Previous MEUF studies indicated that the permeate concentration of the surfactant is nearly equal to the surfactant concentration in the retentate or the surfactant CMC. Clearly, this may increase the cost of the separation process and/or make the process effluent unacceptable on environmental grounds [44].

Yurlova et al [45] employed MEUF to treat water contaminated by Ni(II), using SDS and a nonionic surfactant, monoalkylphenol polyetoxilate (OP-10). They studied three different membranes made of the polymers OPMN-K, UPM-10, and UPM-20. The best retention efficiency (near 100%) was obtained with OPMN-K membranes using an SDS concentration equal to the CMC. For UPM-20 membranes the maximum retention coefficient was 88% at an SDS concentration of 1 g/L. At the same SDS concentration (1 g/L), the retention coefficient for UPM-10 membranes was 99.2%. Where the SDS concentration was lower than the CMC, adding OP- 10 raised the Ni(II) retention coefficient from 88.0% to 96.0%.

3.9. Flotation with membrane

Flotation is a hetero-phase systems separation method, using gas bubble attachment. The attached particles (droplets) are floated and separated from the liquid dispersing medium [46].Water and wastewater applications of flotation are metal coating wastes purification, washing and lubricating liquids treatment, surface water clarification and thickening of activated and coagulated sludge, etc [47].

The flotation methods are traditionally classified based on the method of gas (air) bubbles production [7]. The methods can be differentiated in three groups: (1) dispersed-air flotation, air bubbles are formed by flow through a diffuser (pneumatic flotation) or by mechanical shearing action (mechanical flotation); (2) dissolved-air flotation: pressure flotation (dissolving air in water at overpressure and flotation at atmospheric pressure) and vacuum flotation (dissolving air in water at atmospheric pressure and then applying a vacuum for flotation); and (3) other flotation methods: electro flotation using controlled mild electrolysis for production of hydrogen and oxygen bubbles, biological flotation using gas of biological origin (fermentation), etc [47].

Flotation is used in water and wastewater pretreatment following membrane filtration [48] and in integrated units combining dispersed air flotation and immersed membrane filtration [49]. The examples mentioned above allow us to predict a good outcome of combining flotation with membrane-based techniques in water and wastewater treatment.

The first way of combining membranes and flotation is the use of membranes for air sparging and application of the formed bubbles for flotation. According to the traditional classification system, this type of flotation was termed membrane flotation [50]. The second way is application of membrane filtration to treat flotation effluents or use of flotation for membrane filtration concentrate treatment. The integration of flotation and membrane filtration allows us to treat industrial wastewater up to potable-grade level [47].

4. Conclusions

new adsorbents and membrane processes are the most frequently studied and widely applied for the treatment of the heavy metal-contaminated wastewater. It is clear that in near future, membranes will have an outstanding role in separation industries. Membranes work with very low energies, do not need any heating-cooling systems and do not have any environmental pollution. it is important to note that the selection of the most suitable membrane.

5. References

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[50] Kagramanov GG, Kolesnikov VA, Sudilovskiy PS.,2004 . Newflotation processes and equipment for wastewaters purification.In: 2nd Meeting of the NATO– CCMS Pilot Study-Phase II. Budapest, Hungary, Abstract Booklet, pp 19–20processes for removal of heavy metal from wastewater depends on some basic parameters such as pH, initial metal concentration and etc. technical applicability, plant simplicity and cost-effectiveness are the key factors that play major roles in the selection of the most suitable membrane processes for removal of heavy metal from wastewater treatment system for inorganic effluent.