Desalination 352 (2014) 166–173

Contents lists available at ScienceDirect

Desalination

journal homepage: www.elsevier.com/locate/desal

Heavy metal elimination from drinking water using nanofiltration membrane technology and process optimization using response surface methodology

Amin Maher, Morteza Sadeghi *, Ahmad Moheb

Department of Chemical Engineering, Isfahan University of Technology, Isfahan, Iran

HIGHLIGHTS

- Heavy metal removal from drinking water with a nanofiltration membrane was studied.
- Response surface statistical method was used for process optimization.
- All of the operating variables have important effects on the membrane performance.
- 93% of nickel and 86% of lead ions were removed in the optimum conditions.

ARTICLE INFO

Article history:
Received 23 June 2014
Received in revised form 27 August 2014
Accepted 29 August 2014

Keywords:
Nanofiltration
Heavy metal
Water treatment
Drinking waters
Response surface method

ABSTRACT

In the current study, the effect of operating conditions including pH value, feed flow and applied pressure on heavy metal removal of a nanofiltration membrane for drinking water production was investigated. A polyamide nanofiltration membrane with a net negative surface charge was used for the experiments. In the first series of experiments, single salt solutions containing 1 ppm lead in the form of lead (II) nitrate and 1 ppm nickel in the form of nickel (II) sulfate were used as feed. According to the results, recovery rate, rejection rate and saturation factor were all increased with increasing applied pressure. After that, optimization of operating conditions for maximizing the membrane's heavy metal rejection performance was performed. According to the results, 93% of nickel and 86% of lead ions were eliminated in the optimum condition. In the next series of experiments, the effect of heavy metal ion concentration on the performance of nanofiltration membrane was studied and finally, the effect of mixed salt solutions on the performance of the membrane was discussed. Results showed that, nickel rejection in the mixed salt solution was lower than its value in the single salt solution; but for the lead ion, rejection performance was improved due to the mixing.

1. Introduction

Nowadays, industrial growth especially in developing countries has led to increasing industrial waste discharge into the environment. These waste waters, contain dangerous toxins such as heavy metals and their discharge into the environment can cause air, soil and water pollution.

Waste waters of different industries such as metal plating facilities, paper and pesticide industries contain heavy metals. Unlike organic substances, heavy metals are not biodegradable and tend to accumulate in living organisms [1]. Heavy metals in the waste waters can also pollute the underground water resources. Two examples of heavy metals that may be present in the water are nickel and lead.

Nickel is a rigid and shiny white metal and in the water, it is usually in the form of divalent cation. Although nickel may be present in some ground waters as a consequence of dissolution from nickel ore- bearing, its major source is leaching from metals in contact with drinking water such as pipes and fittings [2]. Although use of waters with high levels of nickel for drinking, can cause serious lung and kidney problems aside from skin dermatitis and pulmonary fibrosis; the major concern about nickel is its carcinogenic properties [3]. Maximum allowable concentration of nickel in drinking water is 0.1 ppm [4].

Lead is another toxic heavy metal that may be present in drinking waters. Just like nickel, lead is usually in the form of divalent cation in water. Long time exposure to high levels of lead can cause kidney, liver, central nervous system and reproductive system damages. Also it is proved that lead is a carcinogenic material [5–7]. Maximum allowable concentration of lead in drinking water is 0.05 ppm [4].

Different methods have been discussed for removing heavy metals from waters such as: chemical precipitation, coagulation, using ion exchange resins and membrane methods [8–11]. In some of these methods for the elimination of heavy metal ions, it is necessary to add

* Corresponding author. Tel.: +98 31 33915645; fax: +98 31 33912677.
E-mail address: m-sadeghi@cc.iut.ac.ir (M. Sadeghi).

http://dx.doi.org/10.1016/j.desal.2014.08.023
0011-9164/

a secondary chemical to the water. Consequently, this chemical should be eliminated from water if the goal of treatment is drinking water production; that it involves extra cost. Membranous methods are relatively new ways that can be used in water treatment. Among different types of membrane filtration methods, reverse osmosis and nanofiltration membrane technologies can be used to eliminate metallic ions directly without the need of secondary chemicals. Reverse osmosis has the highest heavy metal ion removal efficiency among membranous methods [12,13]; but its high capital and operational costs have limited its usage in this field. In addition, because of very low content of minerals, reverse osmosis product water is not suitable for drinking.

With a nominal pore size of 2 nm, nanofiltration falls between reverse osmosis and ultrafiltration in its separation characterization. Nanofiltration needs lower operational pressures than reverse osmosis, so it has a significantly lower electrical energy consumption [14]. Nanofiltration has two separation mechanisms; separation of uncharged solutes due to size effects (sieving) and separation of charged species such as ions because of electrical repulsion (Donnan and Dielectric effects) [15]. Generally, some advantages of the nanofiltration membrane technology in the water and waste water treatment are: elimination of divalent ions such as heavy metals without the need of secondary chemicals, capability of treating waters containing more than one kind of heavy metals, continuous water treatment and having automated process [15–17]. There are some examples of using the nanofiltration membrane technology for the elimination of heavy metal ions such as lead and nickel from wastewaters in the other researches [14,15,18–24]. For example, Jakobs and Baumgarten, discussed the use of nanofiltration for lead elimination in the picture tube industry waste treatment process [24]. Murthy [19,20] and Chaudhari [18], studied the effect of operating conditions on heavy metal removal of a nanofiltration membrane from single and mixed salt solutions. Wahab Mohammad and his co-workers, have used nanofiltration for treatment of electroplating rinse water [23].

According to our knowledge, in the relevant researches, heavy metal elimination from wastewaters with nanofiltration membranes has been discussed and there is no report of using this technology for drinking water production in the literature. In addition to these researches, the effects of change in one of the operational variables on the membrane performance have been discussed, keeping the other factors constant. So because of its importance, this study was aimed to discuss the nanofiltration performance in heavy metal removal from water to produce drinking water and for the goal of process optimization, nanofiltration performance variation due to simultaneous changes in the operating variables was discussed with the help of response surface statistical method. As the heavy metal concentration in the water that could be used for drinking after purification is significantly lower than in wastewaters, in the current research, solutions containing trace amounts of heavy metals were used as feed, so in the absence of concentration polarization phenomenon, membrane performance variations due to operating parameter changes could be seen obviously. Again based on our knowledge, in the other researches, only flat sheet membranes have been used for the experiments and there is not any reports of using conventional spiral wound nanofiltration membrane modules in the literature. Thus because of its direct applicability in household uses and also because of its flow pattern similarity to industrial nanofiltration modules, it could be useful to use a spiral wound membrane module to perform the experiments.

2. Theory

2.1. Membrane performance determination parameters

Membranes split influent feed stream into two streams, a part of feed that passes through the membrane called permeate or product and residual called concentrate or reject [25]. A schematic of a membrane and its influent and effluent streams are shown in Fig. 1.

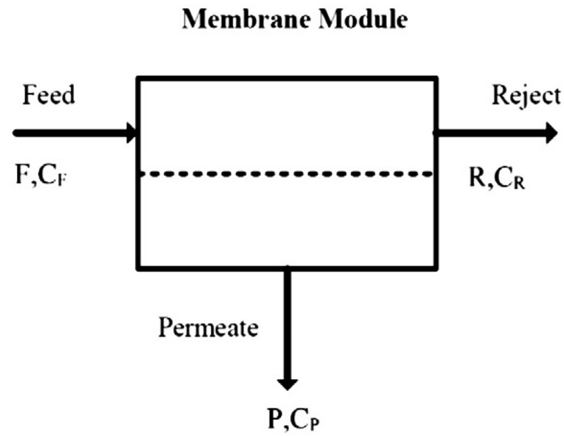

Fig. 1. Schematic of a membrane and its influent and effluent streams.

Where *F*, *P* and *R* are feed, product and reject flow rates in L/h and C_f, C_P and C_R are their concentrations in mol/L respectively.

For the above membrane, recovery of the membrane is given by:

$$\%\,Recovery = \frac{P}{F}\times 100 = \frac{P}{P+R}\times 100. \quad (1)$$

And heavy metal ion rejection is defined as follows:

$$\%\,Rejection = \left(1-\frac{C_p}{C_F}\right)\times 100. \quad (2)$$

The saturation factor of concentrate stream is a parameter for determining the probability of metal hydroxide scale formation in the concentrate stream and it is defined as follows:

$$Saturation\ Factor = \frac{C_{M(OH)_2,conc.}}{C^{sat}_{M(OH)_2,conc.}} \quad (3)$$

where $C_{M(OH)_2,conc.}$ and $C^{sat}_{M(OH)_2,conc.}$ are molar concentrations of metal hydroxide in the concentrate stream and in the saturated conditions respectively. $C^{sat}_{M(OH)_2,conc.}$ is given by:

$$C^{sat}_{M(OH)_2,conc.} = \frac{K_{sp}}{\left(10^{-(14-pH_{Conc.})}\right)^2} \quad (4)$$

where pH_{conc} is the pH value of concentrate stream and K_{sp} is the solubility product constant of the metal hydroxide.

2.2. Design of experiments

In the current study, the face centered response surface method has been used for the determination of the relation between the input variables containing pH value, feed flow and applied pressure and output responses containing system recovery, heavy metal ion rejection and the concentrate stream saturation factor. RSM uses the least square method for this purpose. A quadratic model which also includes the linear terms is given by [26]:

$$\eta = \beta_0 + \sum_{j=1}^{n}\beta_j x_j + \sum_{j=1}^{n}\beta_{jj}x_j^2 + \sum\sum_{i<j=2}^{n}\beta_{ij}x_i x_j + e_i \quad (5)$$

where η is the response, x_i and x_j are variables, *n* is the number of independent variables, β_0 is the constant coefficient, β_j, β_{jj} and β_{ij} are coefficients of linear, quadratic and the second order terms, respectively and e_i is the error term.