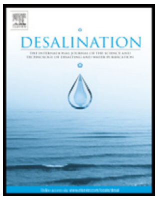
Desalination 267 (2011) 133–138

Contents lists available at ScienceDirect

Desalination

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



Electro-microfiltration treatment of water containing natural organic matter and inorganic particles

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ARTICLE INFO

Article history:
Received 6 May 2010
Received in revised form 9 September 2010
Accepted 10 September 2010
Available online 13 October 2010

Keywords:
Natural organic matter (NOM)
Inorganic particles
Membrane fouling
Electro-microfiltration (EMF)
Critical electrical field strength (E_{critical})

ABSTRACT

Natural organic matter (NOM) and inorganic particles are ubiquitous in many natural surface waters that serve as sources of drinking water. In addition to reacting with chlorine to form chlorinated disinfection by-products (DBPs), NOM and NOM sorbed on inorganic particles cause serious membrane fouling problems. We evaluated the feasibility of using a novel laboratory-scale electro-microfiltration (EMF) system to remove NOM and inorganic particles from water. Synthetic water samples containing 5 mg/L humic acid (HA) only (Kao0), 5 mg/L HA and 5 mg/L kaolin (Kao1), and 5 mg/L HA and 50 mg/L kaolin (Kao2) were used in the EMF study. Kao2 demonstrated the largest flux decline in the absence of an electrical field. When an electrical field was applied, the filtration flux, NOM removal, and decrease in specific ultraviolet absorbance (SUVA) were enhanced because of electrophoretic and electroosmotic effects. When the applied electrical field strength was greater than a critical electrical field strength (E_{critical}), system operations and efficiency did not increase further. These results suggest that the applied electrical field should be close to E_{critical} for maximum efficiency and cost-effectiveness. This study demonstrates that an EMF system is a highly effective way to treat water containing NOM and inorganic particles.

1. Introduction

Natural organic matter (NOM) and inorganic particles are ubiquitous in the natural surface waters that typically serve as sources of drinking water. However, NOM can react with chlorine to form chlorinated disinfection by-products (DBPs) such as trihalomethanes (THMs) and haloacetic acids (HAAs) that present serious health concerns. Therefore, removing the NOM from source water before the chlorination process is essential for minimizing the formation of DBPs. Additionally, inorganic particles such as clays in surface waters cause turbidity that either reduces the water treatment efficiency or increases the dosage of chemical coagulant needed for effective coagulation.

Membrane technology has been widely used in water purification in order to meet more stringent water quality standards. Compared with traditional water treatment processes, membrane technology has the advantages of requiring less space for treatment and producing a higher-quality effluent in a shorter treatment time [1]. The membrane used in microfiltration (MF) has relatively large pore sizes and does not remove all NOMs [2], whereas alternative techniques such as nanofiltration (NF) and reverse osmosis (RO) can remove NOM efficiently. Moreover, the most important problem with membrane filtration processes is fouling, which reduces the filtration flux and results in a poorer-quality effluent, thus increasing costs of operation. Previous studies reported that the presence of NOM alone or NOM mixed with inorganic particles in water was one of the most significant factors causing membrane fouling [3–5]. Most components of NOM, such as humic acid (HA, which comprises a major fraction of total NOM), are negatively charged due to dissociation of the carboxylic acid (COOH) and hydroxyl (OH) functional groups [6]. Additionally, inorganic particles in the natural environments commonly have negative surface charges due to dissociation of the functional groups on their surfaces, isomorphic substitutions (e.g., in the tetrahedral sites, with replacement of Si^{4+} by Al^{3+}), or adsorption of organic compounds (such as NOM) and surfactants by van der Waals' forces or hydrogen bond [7].

As mentioned above, the performance of MF processes could be enhanced by applying an electrical force to attract charged water-borne NOM and inorganic species. When the electrical field is applied through the membrane, two important electrokinetic phenomena, electrophoresis and electroosmosis, occur during electro-microfiltration (EMF) [8–10]. Electrophoresis refers to the convective movement of the charged species relative to the liquid phase in an electrical field, whereas electroosmosis is the convective movement of liquid dragged from the anode towards the cathode under the influence of the electrical field. Some studies have reported that electrofiltration can efficiently decrease fouling and increase rejection of NOM [9,11,12].

To the best of our knowledge, the mechanisms of fouling by inorganic particles mixed with NOM are poorly understood, and in

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doi:10.1016/j.desal.2010.09.015

fact, no data are available that describe EMF treatment of water containing NOM and inorganic particles. In this study, the effect of these water matrices on filtration performance was examined using synthetic water containing HA and kaolin, standing in for NOM and inorganic particles, respectively. Three sets of samples containing 5 mg/L HA only (Kao0), 5 mg/L HA and 5 mg/L kaolin (Kao1), and 5 mg/L HA and 50 mg/L kaolin (Kao2) were used to simulate natural surface waters. The effect of different ratios of HA to kaolin on the performance of EMF under various trans-membrane pressures (49 or 98 kPa) and electrical field strengths (0, 21.7, 60.8, or 73.3 V/cm) was also investigated.

2. Materials and methods

2.1. Feed solution

The HA stock solution was prepared by dissolving 1 g of HA (Sigma-Aldrich, St. Louis, MO, USA) in 1 L of Milli-Q water. The solution was filtered through a 0.45-μm filter to remove particulates and stored in the refrigerator (4 °C) before use. Sodium chloride was used as the background electrolyte. The concentration of dissolved organic carbon (DOC) in the feed solution was adjusted to 5 mg/L; kaolin was then added to yield working solutions containing 5 mg/L HA only (Kao0), 5 mg/L HA and 5 mg/L kaolin (Kao1), or 5 mg/L HA and 50 mg/L kaolin (Kao2), each with a conductivity of 100 μS/cm and pH 7.

2.2. Membrane and module

A schematic diagram of the laboratory-scale EMF system is shown in Fig. 1. The filtration module consists of a housing, a cathodic mesh, a co-centric anodic rod of 1.3 mm diameter, and a tubular membrane with a 6-mm diameter covered with membrane pores of nominal size 0.1 μm. The anode and cathode were made of platinum and titanium, respectively. The membrane's filtration surface area was 20.7 cm^2.

2.3. Filtration experiments

Five liters of the feed solution containing HA or kaolin at preselected concentrations were used for each experiment. The electrical field was applied across the MF membrane using a power supply (model 6210K—600/1000 W, Chroma, CA, USA). A peristaltic pump was used to transfer the solutions into the EMF cell at a crossflow velocity of 0.18 m/s, while the trans-membrane pressure was kept at 49 or 98 kPa. The electrical balance was used to record the amount of permeate flux. Both the permeate and concentrate streams were recycled back to the feed tank to maintain a constant feed concentration. After each experiment, the membrane was cleaned with 0.1 N sodium hydroxide (NaOH), 0.01 N citric acid and Milli-Q water. NaOH solution, citric acid solution and Milli-Q water were pumped into the system sequentially. Each washing cycle lasted for 10 min. The membrane was then soaked in a 0.1 N NaOH solution for 12 h, kept in a 0.01 N citric acid solution for another 12 h, and subsequently immersed in Milli-Q water to remove the residual citric acid and NaOH. Finally, the membrane was cleaned with Milli-Q water for 1 h. The cleaning procedure recovered 98–100% of the original flux.

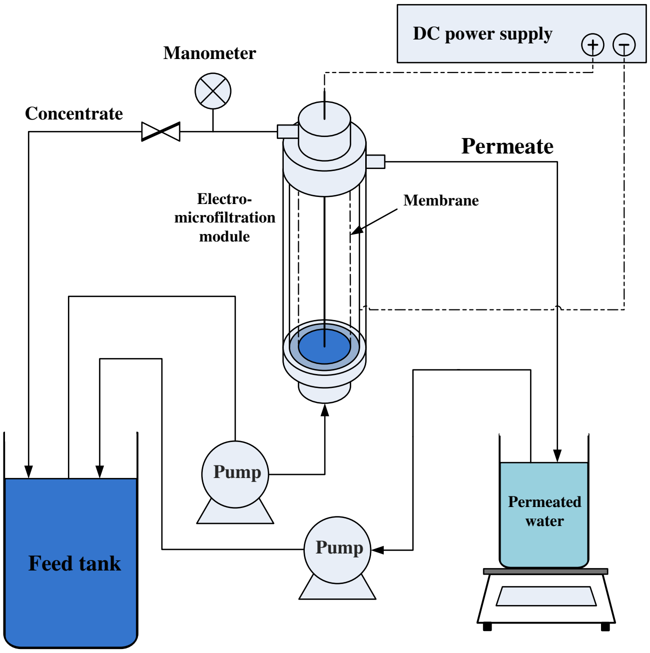

Fig. 1. Schematic diagram of the laboratory-scale electro-microfiltration system.

2.4. Chemical analyses

DOC was analyzed with an organic carbon analyzer (model 1010, OI Analytical, TX, USA). Measurement of pH and conductivity was carried out with a Consort model C831 pH meter and a Suntex model SC-120 conductivity meter, respectively. UV$_{254}$ was measured with a Cintra 20 UV–Visible spectrometer. The average electrophoretic mobility measurements for each sample were obtained with a Malvern Zeta Sizer 3000 zetameter. Particle size distribution and turbidity were determined with a Beckman model Coulter LS230 particle size analyzer and a HACH model 2100P turbidimeter, respectively. Distribution of the elements on the surface of kaolin particles was assessed with a scanning electron microscope/energy dispersive spectrometer (SEM/EDS) technique (LEO 1530 FEG-SEM+EDS).

2.5. Filtration resistance and applied electrical field strength

The filtration resistance was calculated according to Darcy's law as shown in Eq. (1).

$$R_f = \frac{\Delta P}{J\mu} \tag{1}$$

In this equation, R_f is the total filtration resistance (m^{-1}), ΔP is the applied pressure (Pa), J is the permeate flux (m/s), and μ is the viscosity (N s/m^2).

The applied electrical field strength during electrofiltration was calculated using Ohm's law as shown in Eq. (2)

$$E = \frac{I}{\kappa A} \tag{2}$$

where E is the applied electrical field strength (V/cm), I is the applied electrical current (A), κ is the solution conductivity (S/cm), and A is the filtration area (cm^2).

3. Results and discussion

3.1. Characterization of the colloids

Fig. 2 shows the average electrophoretic mobility (AEM) of colloids in the synthetic solutions as a function of solution pH. The AEM values of the colloids were negative between pH 3 and 10, indicating a strong potential for EMF treatment. In addition, the HA and kaolin mixture had a more negative absolute AEM than either HA or kaolin alone, demonstrating the synergistic effect of mixing HA and kaolin on AEM. Shen [13] previously pointed out that the pH$_{zpc}$ of kaolin was less than 3, giving kaolin a negative charge at neutral pH. Moreover, kaolin is a type of clay with a 1:1 configuration of aluminum and silica, with a