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Research Article

A Ceramic Ultrafiltration Membrane System for Producing High Quality Drinking Water

Seramik Ultrafiltrasyon Membran Sistemi İle Yüksek Kalitede İçme Suyu Üretimi

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Abstract

In this study, ultrafiltration (UF) with ceramic membranes was used to produce safe and quality drinking water. The small scale UF membrane system had a capacity of 1.44 m3/d. The UF membrane filtration process includes two parts: a tubular ceramic membrane formed by a porous support (α-alumina) and a tube reactor chamber 10 m long and 5 cm in diameter to generate electrocoagulation.

In this study raw water treated with small-scale UF membrane systems was taken from the Alibey Lake in Istanbul City, Turkey. The system removed 75% to 85% of ferrous and turbidity contaminants. The decrease in pH, chloride and total hardness was similar, but ammonia and manganese removal was much lower than expected. Nevertheless, removal of total organic carbon (TOC) was the best—only 15% remained. The UF ceramic membrane filtration system produced water that met Turkish Standards (TS-266, regulated standards for drinking water in Turkey). Chemical cleaning with a clean-in-place (CIP) operation was successful in removing fouling and scaling materials in ultrafiltration UF ceramic membrane. The UF ceramic membrane filtration system produced water with no added chemicals as a coagulant and disinfectant. Indeed, producing water with no chemicals and disinfection byproducts (DBPs) like trihalomethenes (THMs) is better for human health than the approaches used at conventional drinking water treatment facilities.

Keywords: Ceramic membrane, Electrocoagulation, Ultrafiltration, Water treatment, Water quality

Öz

Bu çalışmada tübüler seramik membrane kullanılarak oluşturulmuş küçük ölçekli Ultafiltrasyon (UF) membran sistemi ile kaliteli bir içme suyu üretilmesi hedeflenmiştir. Bu amaçla kullanılan UF membrane filtrasyon sistemi 1.44 m3/ gün'lük bir su üretim kapasitesine sahiptir. Bu UF membrane filtrasyon sistemi, α-alumina içeren gözenekli bir destek tabakası ve elektrokoagülasyon prosesini gerçekleştirmek için 5 cm çapında ve 10 m uzunluğunda tubüler bir reactor odasından meydana gelmektedir.

Bu çalışmada UF membran sistemi ile arıtım amaçlı olarak kullanılacak ham su İstanbul şehrinin önemli içme suyu kaynağı olan Alibeyköy baraj gölünden sağlanmıştır. Bu sistem ile yapılan deneysel çalışmalarda demir ve bulanıklık giderim oranlarının sırası ile; % 75 ve %85 olduğu gözlenirken, pH, klorür ve toplam sertlik parametre değerlerinde herhangi bir değişim olmadığı ortaya konulmuştur. Bununla beraber Toplam Organik Karbon (TOK) değerlerinde o yaklaşık %15'lik bir düşüş oldğu gözlenirken, Amonyak ve Mangan değerlerinde ise tahmin edilenden daha düşük bir giderim verimi sağlandığı tespit edilmiş olup arıtma sonunda alınanan su numunelerinin bakteriyolojik olarak temiz olduğu rapor edilmiştir. Bu çalışmanın en önemli sonuçlarından biri, herhangi bir kimyasal ve dezenfektan kullanmadan UF membrane filtarsyon sistemi ile üretilen suyun TS-266 içme suyu standartlarında yer alan temel su kalite parametre değerlerini sağlamış olmasıdır. Bununla beraber konvansiyonel içme suyu arıtma tesisleri ile karşılaştırıldığında dezenfektan olarak klor kullanımı sonucu meydana gelen Trihalometanlar gibi özellikle insan sağlığı üzerinde kanserojenik etkiye sahip dezenfeksiyonyan ürünlerinin olmaması UF membrane sistemi ile üretilen içme suyunun sağlıklı, güvenli ve kaliteli olduğunu ortaya koymaktadır.

Anahtar Kelimeler: Seramik membran, Elektrokoagülasyon, Ultrafiltrasyon, Su arıtımı, Su kalitesi

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1. Introduction

The lack of safe, potable water and increased demand as well as higher standards have increased the need for membrane technologies to produce high quality drinking water (Bagga et al. 2008). Moreover, conventional water treatment processes including coagulation/flocculation, sedimentation, filtration and disinfection processes are not very effective at meeting these stringent regulations. Thus, use of pressure-driven membrane processes such as microfiltration (MF) and ultrafiltration (UF) are increasingly popular in drinking water treatment (Jacangelo et al.1995, Yuan and Zydney 1999, Zularisam et al.2007). Furthermore, the chlorine used as a disinfectant in conventional water treatment plants reacts with Natural Organic Matter (NOM) and produces disinfection byproducts (DBPs) that are carcinogenic and mutagenic (Rook 1974).

Membrane-based filtration such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) have been investigated as a potential alternative to conventional water treatment options for small communities. Membrane installations are easily automated. The UF, NF and RO remove significant levels of trihalomethene (THM) precursors from drinking water supplies and deliver excellent microorganism control. Hence, membrane filtration removes turbidity, reduces THM precursors, and disinfects in a single step (Richard and Paul 2003). Small-scale membrane treatment systems such as MF and UF systems are highly effective for turbidity as well as bacteria and virus removal from surface waters such as rivers and lakes (Jacangelo et al.1991, Madaeni 1999, Neranga et al.2014, Zhu et al.2005). They also indirectly assist in DBPs control by lowering chemical disinfection requirements for the filtered water. Furthermore, the goals of small-scale treatment systems are simplicity, no chemicals, dynamic remote control, long service interval times and low energy use.

Ceramic membranes have several advantages over polymeric membranes such as high chemical, mechanical and thermal resistance as well as higher permeability rates than polymeric membranes. Nevertheless, ceramic membranes are substantially more expensive though this may be compensated by their higher fluxes and extended lifetimes (Van Der Bruggen et al.2008, Kim et al.2007, Barredo-Damas et al.2012).Porous ceramic membranes are an important membrane category that is of particular interest in applications requiring high chemical or thermal stability (Pagana et al.2006, Shams Ashaghi et al.2007). Tubular ceramic membranes are formed by a porous support (generally, a-Al_2O_3) with one or more layers of decreasing pore diameter and an active or separating layer (α-alumina, zirconia, etc.) covering the internal surface of the tube. The use of ceramic membranes for microfiltration and ultrafiltration is of great interest because they can remediate fouling problems associated with those processes and solutions, i.e., adsorption or deposition of macromolecules on the membrane pores/surface. This strongly reduces volume flow and requires harsh chemicals and high temperatures for cleaning. In turn, this damages the polymeric membranes (Richard et al. 2013, Verberk et al. 2002).

Thus, the use of these systems is still limited by fouling. It has also been suggested that viruses are etiologic agents responsible for the majority of unidentified outbreaks because they are typically more difficult to analyze than bacterial pathogens. It is difficult to remove viruses by filtration because of their small size (Tanneru and Chellam 2012, EPA 2006, Urase et al. 1996, Mi et al. 2005, Pontius et al. 2009).

Electrocoagulation (EC) has been widely studied in water and wastewater treatment to remove heavy metals, organics, bacteria, hardness, turbidity, and other contaminants (Tsouris et al. 2001, Can et al. 2003, Al malack et al. 2004, Mills 2000, Zhu et al.2005).

EC has been widely studied in water and wastewater treatment. Here, the electrodes are consumed as the coagulant is generated and precipitated. No liquid chemicals are added. No basic chemical are used, and the pH does not have to be adjusted (Mills 2000, Zhu et al.2005).

Additionally, EC pretreatment is an alternative to conventional chemical coagulation using Fe or Al salts prior to MF or UF membrane systems. In electrocoagulation, the coagulant (Fe or Al) is generated by electrolytic oxidation of an anode. The advantages of EC over conventional chemical coagulation include: (1) no addition of lime, ferric and coagulant chemicals, (2) no change in bulk pH, (3) simple operation and maintenance and (4) low sludge generation (Bagga et al. 2008, Ca^zinares et al. 2006, Hu et al.2013). The most important advantage of EC pretreatment is the reduction in fouling problems that occurs in small-scale MF and UF membrane systems (Bagga et al. 2008, Al Malack et al. 2004).

The aim of this study is to provide high quality potable water without added chemicals via a small-scale membrane treatment system consisting of UF ceramic membranes.

We compared the treatment performance of conventional treatment process in Kagithane Water Treatment Plant (KWTP) and UF membrane filtration process and characterized the pH, turbidity, total organic carbon (TOC) and total hardness.

2. Materials and Method

2.1. Source Water Quality

Water quality is an important factor in determining the treatment performance of small-scale UF membrane systems. For this study, raw water taken from the Alibey Lake in Istanbul City, Turkey, was used as feed water for small-scale UF membrane systems during the winter period (January, February and March) in 2011.

This surface water supply is one of the major drinking water sources of Istanbul City. Also, Alibey Lake is one of the most important water reservoirs in Istanbul and provides up to 700,000 m^3/day of raw water to produce drinking water.

Raw water samples were collected by plant personnel as a grab sample and shipped to a water quality laboratory (Istanbul Water Utilities Administration (ISKI)) on the same day. Samples were stored in the dark at +4^{o}C to prevent biological activity prior to analysis.

2.2. Membrane

In this study, the small-scale UF membrane filtration system used to purify Alibey Lake water was composed of tubular ceramic membranes formed by a porous support (α-Alumina) (Fig. 1).

These membranes consist of 580 mm long channels with an external diameter of 4 mm and 2 mm. Their effective pore sizes are 0.04 µm, and the effective filter area is 1.8 m^2 as surface area per volume (m^2/m^3). Table 1 lists other relevant properties of these membranes

2.3. Electrocoagulation (EC) unit

Coagulation process in the EC used a dedicated tube reactor. This reactor chamber consists of a 10 m long tube 5 cm in diameter. The rod-shaped iron anodes are 50 cm long. The cylindrical stainless steel cathodes are placed in a electrode chamber and are 1 m long. The total anode surface area was 100 cm^2, and the current density was typically 0.15 mA/cm^2. During iron EC, the following electrochemical reactions occur:

Anode: Fe^0 (s) = Fe^{3+}+3e

Fe^{3+} + $3H_2O$ = $Fe(OH)_3$ (s) + $3H^+$

Table 1. Typical characteristics of the membrane used in this study.

Parameter	Value	Unit
Material	Ceramic (a-Al_2O_3)	-
Pore size (nominal)	0.04	µm
Effective area	1.8	m^2
Feed water flux	60	$L{\cdot}h^{-1}m^{-2}$
Max. operating pressure	0.65	bar
Max. operating temperature	30	^{0}C
pH range	4-11	-

Figure 1. Tubular ceramic membrane.

Cathode: $3H_2O$ +3e^- = 3OH + 3/2 H_2

Overall: Fe^0+$3H_2O$ = $Fe(OH)_3$(s) + 3/2 H_2

2.4. Experimental UF Membrane Filtration Setup

UF filtration experiments were conducted in a multi tubular ceramic membrane. The process was designed for a flux of 60 L/m^2/hr. As seen in Fig. 2, surface lake water was taken from the 1,000 L tank with a peristaltic pump and transferred into the reactor chamber for EC processing. In the meantime, the iron electrodes are sacrificed at this step at a concentration of ~ 4 ppm. In this way, it is possible to have dynamic, in-line process control as well as a short residence time in the tube reactor for floc growth.

After coagulation, the raw water was passed into the second part including the UF ceramic membrane. The flow level and temperature sensor were located at the first part of the reactor; water levels in the reactor were held constant. Permeating and backwashing operations were performed automatically with an automatic control system. To save

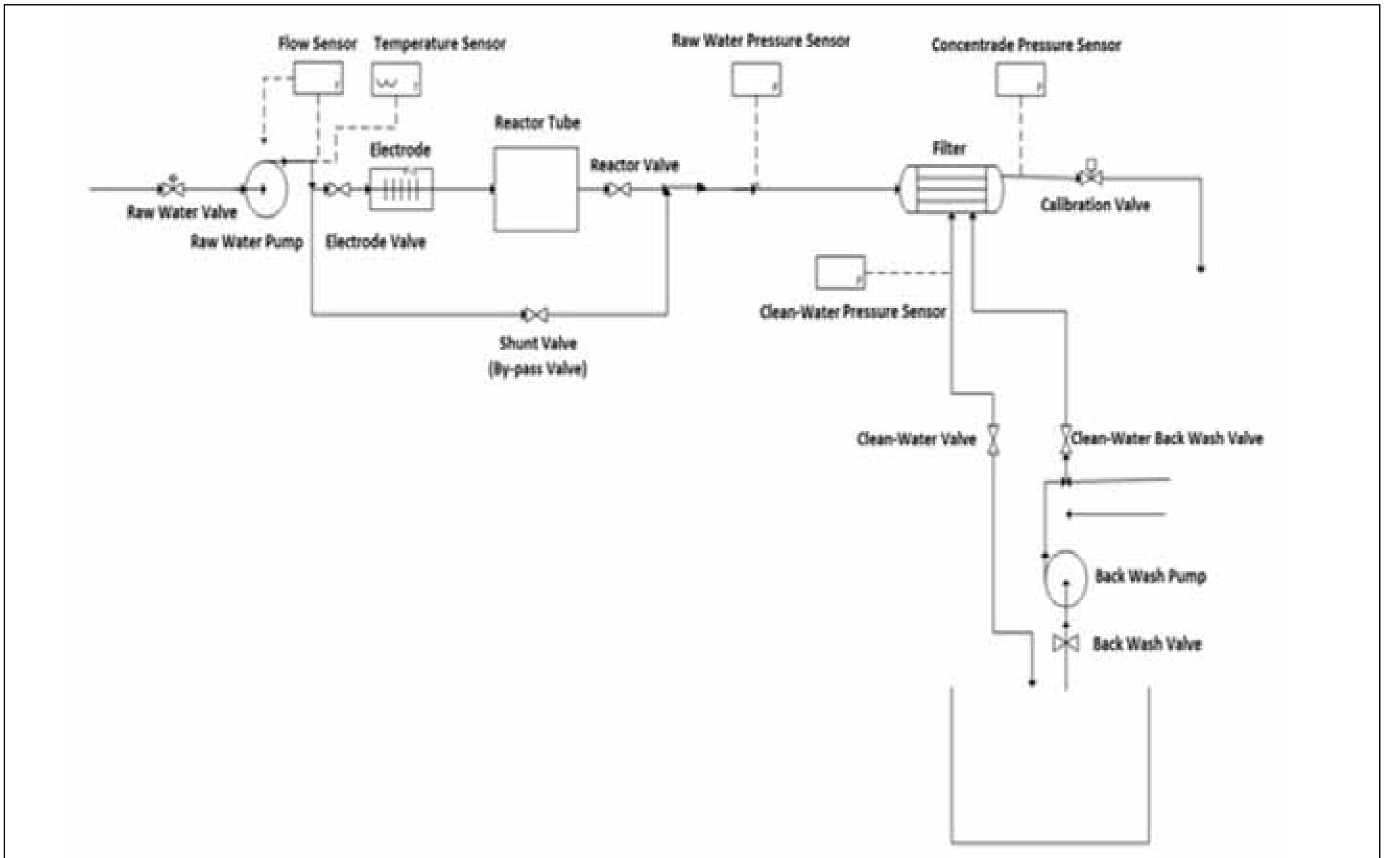

Figure 2. A schematic diagram of the UF ceramic membrane filtration system.

energy, the filtering process was planned at a low trans-membrane pressure (TMP). Up to 0.25 bar of TMP was used for the expected floc-sizes. The membrane cleaning process used filter backwashing and chemical cleaning with an automatic control system. Filter backwashing was automatically performed every 20 minutes with water treated by the UF membrane system. Chemical cleaning of the membrane was automatically carried out using 200 ppm NaOCl and 500 ppm H_2O_2 using chemical dosage pumps every 1.5 hours. This avoids membrane fouling from microbial contamination.

2.5. Analytical Methods

The TOC analysis used high temperature combustion according to Standard Methods (SM) 5310 B using a Shimadzu TOC-VCPH analyzer equipped with an auto-sampler (APHA 1998).The total iron was measured using atomic absorption spectroscopy (AAnalyst 300, Perkin Elmer Corp., CT) after acidifying the samples to pH<2 using HNO_3 according to Method 3111 in the Standard Methods (APHA 1998). Turbidity was determined by with a Thermo turbidimeter according to Standard Methods. The pH, total hardness, chloride, manganese and ammonia were measured according to the literature (APHA 1998).

3. Results

3.1. Treatment performance of the UF ceramic membrane filtration system

In this study, we measured the quality of the water produced by the UF ceramic membrane filtration system. Alibey Lake water was treated with the UF ceramic membrane filtration system, and Table 2 details physicochemical characteristics of raw Alibey Lake water versus the treated water.

As seen in Table 2, the turbidity values drop from 6.5 NTU to below 1 NTU. The concentrations of ferrous and manganese were 0.03 and 0.05 mg/L, respectively, in clean water. In other words, the removal percentage of turbidity wasapproximately 85%. The conductivity was 740 µS/cm^{-1}, and the pH was 7.71 on average in treated water. The total hardness and chloride in treated water remained relatively constant (Table 2).

The drinking water was produced from Alibey Lake water with the UF ceramic membrane filtration. Fig. 3 illustrates that turbidity of Alibey Lake water did not exceed 0.3 NTU at the effluent of the UF ceramic membrane filtration system. To better compare the turbidity values, we plotted the data (Fig. 3) ten-fold.

As shown Fig. 4, the pH of the water was not significantly influenced by treatment—the results were within the known target limit for Turkish Standards -266 (TS-266, regulated standards for drinking water in Turkey).

Table 2. The relevant parameters used to evaluate the UF ceramic membrane filtration system including pre- and post-treatment water.

Parameters	Units	Raw Water (Average)	Product water (Average)	Standarts for drinking water in Turkey (TSI-266)
pH	-	7.82	7.71	6.5-8.5
Turbidity	NTU	6,54	0.97	5
Conductivity	μS/cm	651	664	650-2000
Total Hardness	mg $CaCO_3$/ L	153.35	155.5	300
Chloride	mg/L	80.14	79.82	250
Ammonia	mg/L	0.32	0.24	0.5
Dissolved Oxygen	mg/L O_2	10.52	11.1	Not defined
TOC	mg/L	6.12	5.18	Not defined
Iron	mg/L	0.11	0,03	0.2
Manganese	mg/L	0.067	0.05	0.05
T.Coli.Bacteria	cfu/100 mL	>20000	None	None

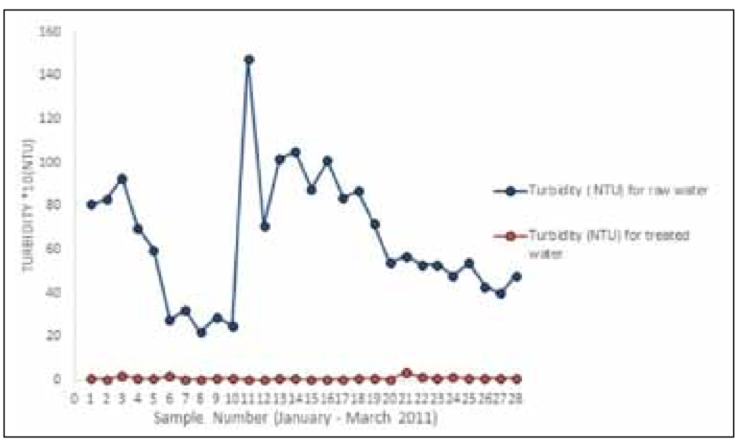

Figure 3. Turbidity values (NTU) in raw water and treated water with UF membrane filtration system.

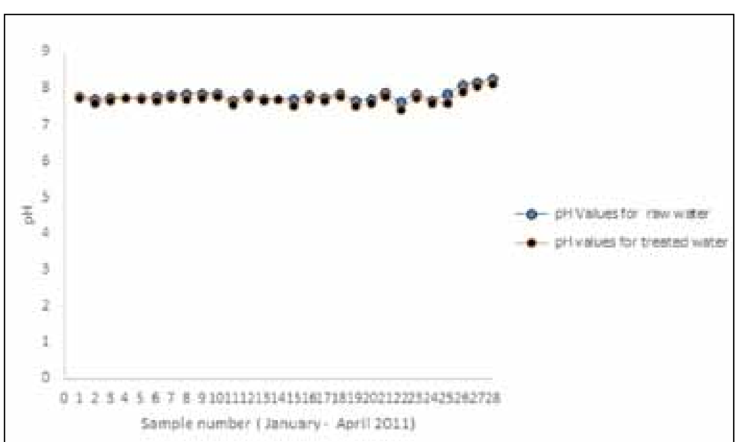

Figure 4. pH values in raw water and treated water with a UF membrane filtration system.

There was no change in total hardness during the membrane filtration as occurs with chloride treatment (Fig. 5a, b). In other words, both parameters had similar removal percentages.

Ammonia and manganese removal through the ceramic UF membrane filtration was much lower than expected—only 25% was removed (Fig. 6a,b). Of all of the water quality metrics, turbidity had the highest removal ratio at 85% (Fig. 7); the TOC removal ratio was only 15%. Moreover, the performance of the UF membrane system was much lower for the TOC parameter (Fig. 8). This result is expected because the electrocoagulation time is very short. This also means that it is not enough time for the needed floc-growth in the EC due to low retention time. No bacteria were found in the UF-filtered water despite the lack of chlorine. Thus, no disinfection byproducts were present in the UF water that might result in adverse health effects.

3.2. Operation of the EC unit

After designing the EC unit, it was tested for its ability to generate iron in proportion to the current by operating it continuously at different current values.

The process has been designed for a flux of 60 L/m^2/hr. The Fe electrodes of the system are sacrificed during the process at a concentration of ~4 ppm. This gives dynamic in-line process control and a short detention time as needed for floc growth. To determine the current efficiency, the amount of iron generated was calculated using Faraday's Law (Eq.1):

$$m = I \times t \times MW / Z \times F \qquad \textbf{(Eq.1)}$$

Where m is the mass in grams of Fe generated at a specific current (I, amps) over a time interval (t, seconds). Term Z is the number of electrons transferred per Fe atom, MW is the molecular weight (55.85 g mol^{-1}), and F is Faraday's constant (96,486 C eq^{-1}).

The desired iron concentration was obtained by adjusting the operating current and flow rate of the source water. For example, when the feed flow rate was 250 mL/min and the operating current was 0.15 A, the iron concentration was

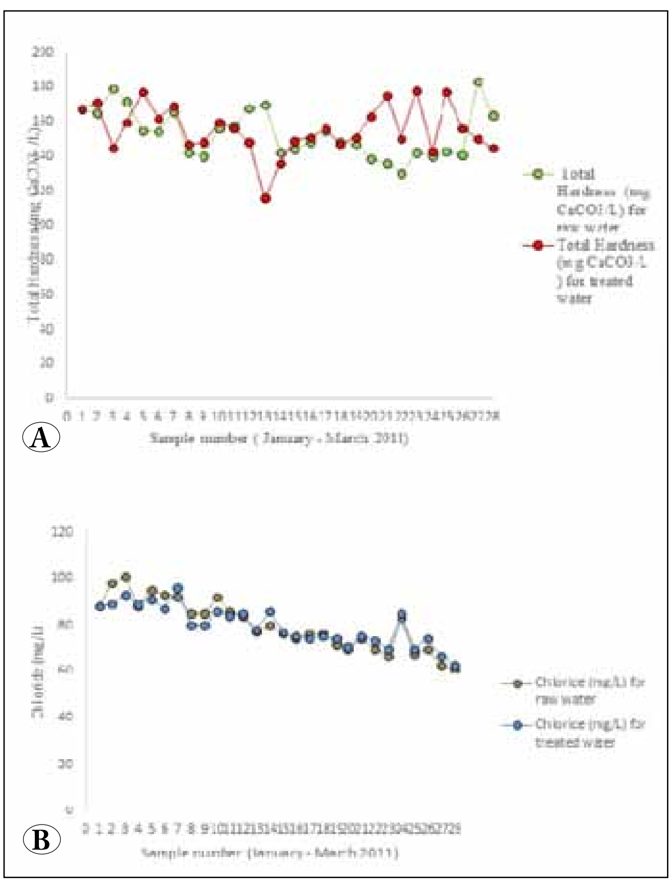

Figure 5. **A)** Total hardness (mg $CaCO_3$/L) and **B)** Chloride (mg/L) values in raw water and treated water from the UF membrane filtration system.

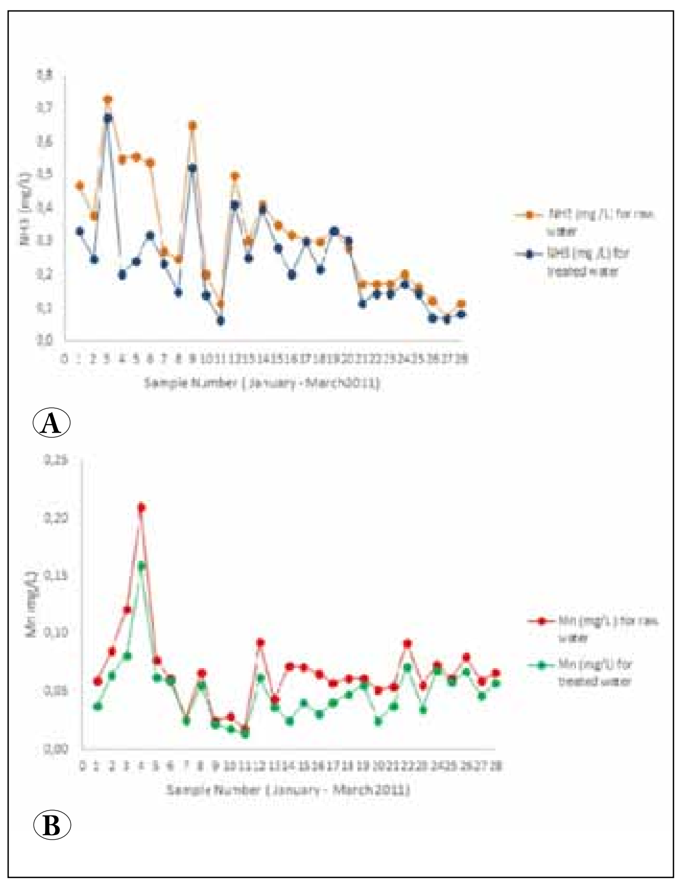

Figure 6. **A)** Ammonia (mg/L) and **B)** manganese (mg/L) in raw water and treated water.

3.6 mg/L. Fig. 9 presents Fe(III) concentration as well as the EC unit operating current. The Fe (III) formation at 250 mL/min is a function of EC operating current.

3.3. Effects of backwashing and chemical cleaning on the UF ceramic membrane filtration

The backwashing and chemical cleaning processes were conducted automatically in the UF membrane system to control membrane fouling. Ceramic membranes have a higher permeability versus traditional polymeric membranes if the backwash interval is extended (Zhu et al. 2005, Jegatheesen et al. 2009). In this study, the UF ceramic membrane filtration was operated with Alibeyköy Lake water for 24 hours. One cycle required between 15 and 20 minutes. After 5 cycles of operation, the system was automatically backwashed for 2 minutes. As shown in Fig. 10, when the TMP was 0.65 bar, the system automatically switches to chemical cleaning mode to remove bacteria and/or viruses from the membrane.

Chemical cleaning with clean-in-place (CIP) operation is the usual method to restore the membrane permeability. There are several reagents including alkalis, acids, oxidants, chelating agents and surfactants that could be used for CIP (Zhu et al.2005, Jacb and Jaffrin 2000). Many aspects should be considered when selecting CIP reagents. The two main factors are feed composition and the composition of the fouling layer (Zhu et al.2005). In this study, H_2O_2 and

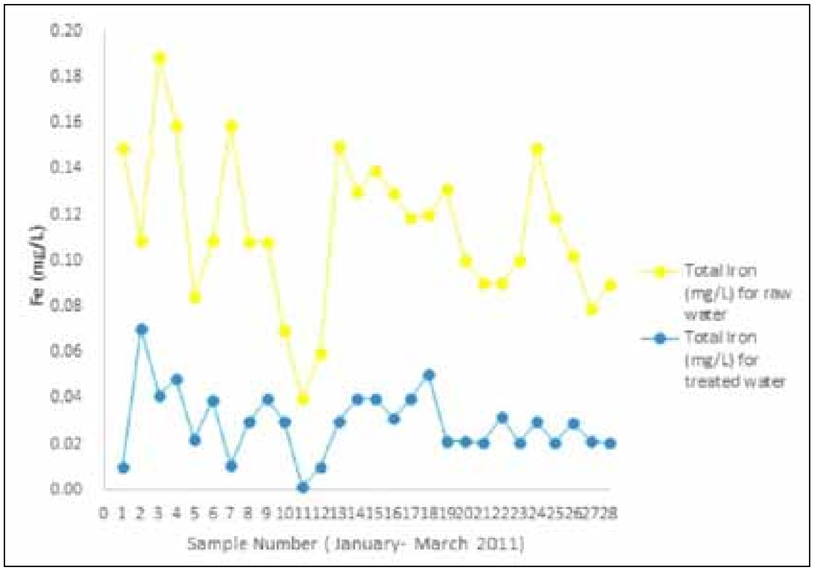

Figure 7. Total iron values (mg/L) in raw water and treated water.

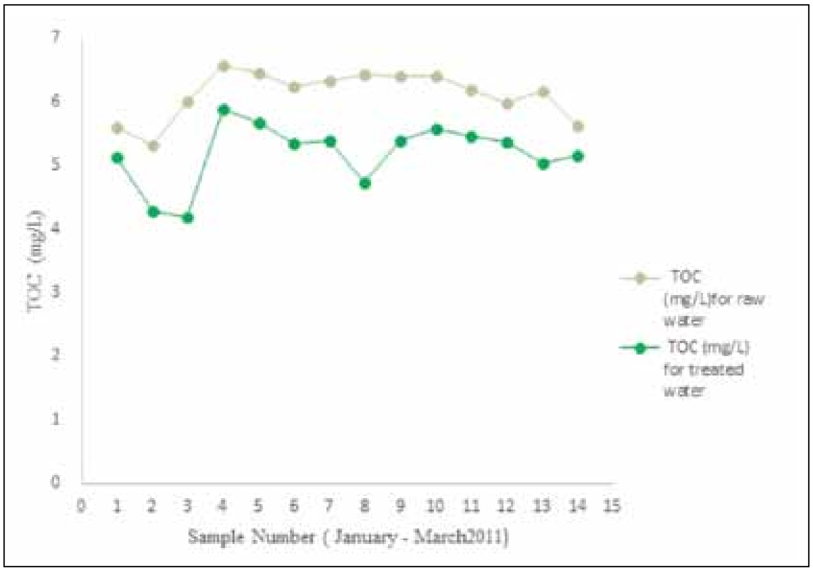

Figure 8. TOC (mg/L) values in raw water and treated water treated.

NaOCl were selected because they have strong chemical inertia and do not affect the thermal stability of the ceramic membrane.

The use of chemicals was limited to cleaning, and the total amount needed can be extrapolated from the volume of the filter elements. During this experiment, the chemical concentration was adjusted to pH 2 and 500 ppm H_2O_2 and 200 ppm NaOCl. Under normal circumstances we used 15 minutes of soaking. Figure 10 shows changes to the TMP as a function of time (0-432 hours) during operation. As shown the Figure (10), the TMP increases after chemical cleaning. The TMP decreased from 0.65 to 0.1 bar. It also demonstrated that a large number of micro-organisms and colloids resulting from membrane fouling were removed by chemical cleaning (Fig. 10).

Furthermore, TMP during the filtration tests varied between 0.17 Bar and 0.23 Bar. The flow was kept constant during filtration. The TMP during backwash never exceeded 0.35 Bar. The TMP after every backwash at the start of the filtration cycle varied between 0.17 and 0.19 bar.

4. Conclusion

In this study, we produced potable water in accordance with EC standards using UF ceramic membrane filtration system with no added chemicals. We studied the performance metrics of the UF membrane. Except for the TOC and ammonia, all of the relevant water parameters including pH, turbidity, Fe, and manganese met the required specifications. Moreover, the Fe and turbidity were removed at nearly 75% and 85%, respectively. Bacteria were not found in the treated water despite the lack of chlorine.

During EC, the Fe electrodes are consumed at concentration of ~4 ppm. Moreover, the Fe (III) formed at 250 mL/min is a function of the EC unit operating

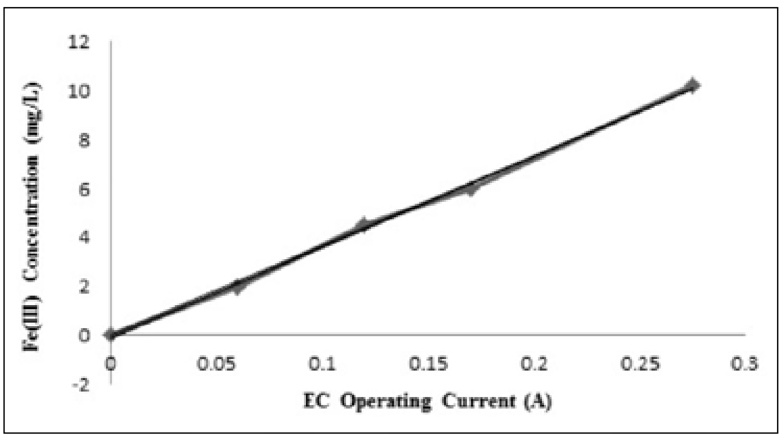

Figure 9. Fe (III) generated at 250 mL/minute as a function of EC operating current.

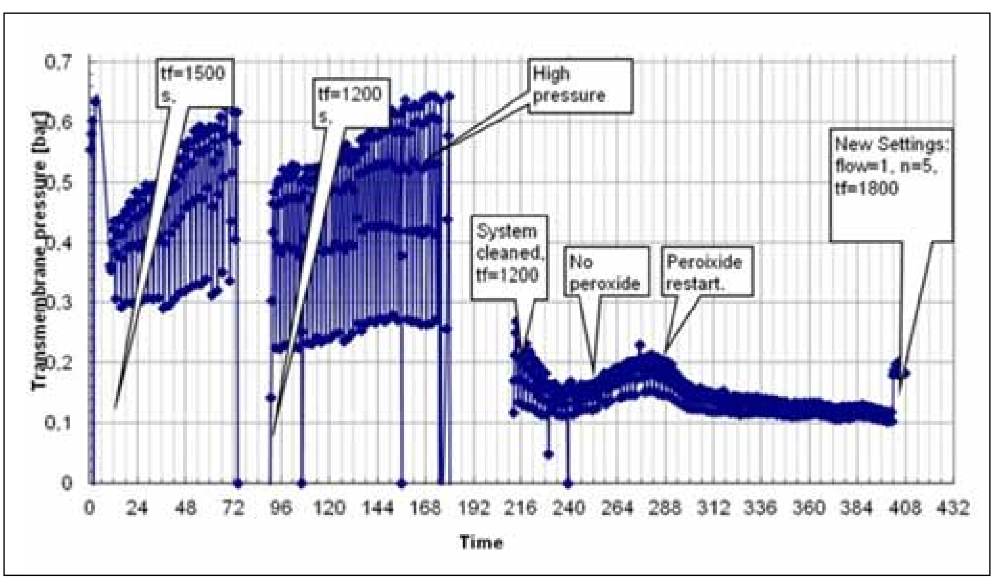

Figure 10. TMP changes as a function of time during UF ceramic membrane filtration.

current. The backwashing and chemical cleaning processes were conducted automatically by UF membrane system to control membrane fouling. After chemical cleaning, the TMP decreased from 0.65 to 0.1 bar. In summary, the UF ceramic membrane filtration system produced drinking water that met TS-266 standards with no added chemicals for coagulation and disinfection. Producing water without DBPs like THM offers better safety and quality for humans than water produced by conventional treatment systems.

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