Separation and Purification Technology 122 (2014) 170–182

Contents lists available at ScienceDirect

Separation and Purification Technology

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

Treatment of oil sands process-affected water with ceramic ultrafiltration membrane: Effects of operating conditions on membrane performance

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ARTICLE INFO

Article history:
Received 4 June 2013
Received in revised form 4 November 2013
Accepted 5 November 2013
Available online 12 November 2013

Keywords:
Ultrafiltration
Ceramic membrane
Oil sands
Coagulation
Fouling mechanism

ABSTRACT

This study investigated the performance of 1 kDa ceramic ultrafiltration membrane for the removal of inorganic and organic compounds from oil sands process-affected water (OSPW) generated after thermal operations of heavy oil recovery in Alberta, Canada. The OSPW was pretreated with alum coagulant, and the effect of operating conditions on subsequent membrane filtration was studied. While permeate flux increased with increasing trans-membrane pressure (TMP) from 1.4 bar to 3.5 bar, greater permeate flux decline was observed at TMP of 3.5 bar due to increased accumulation of foulants at the membrane surface. The membrane filtration performed at cross-flow velocity (CFV) of 0.2 L/min was characterized by the lowest initial and steady-state permeate fluxes and by the highest normalized flux decline compared to higher CFVs. This effect was attributed to lower turbulence at the membrane surface which might have promoted the buildup of contaminants. According to the resistance-in-series model, no irreversible membrane fouling was observed when OSPW was pretreated with coagulation–flocculation–sedimentation. The pore blocking and cake layer formation dominated at the beginning of filtration, whereas cake layer formation was the primary fouling mechanism at later stages. The final membrane permeates met the requirements for the high pressure-driven membrane processes (i.e., nanofiltration and reverse osmosis) with respect to turbidity and silt density index (SDI_{15}) values. Up to 38.6 ± 2.7% (depending on TMP and CFV values) of chemical oxygen demand (COD) was removed, and the removal percentages of the acid extractable fraction (AEF) and naphthenic acids (NAs) were less because of the small sizes of NAs and other organic compounds contributing to the AEF as compared to the membrane pore sizes.

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1. Introduction

During thermal operations of heavy oil recovery in Northern Alberta, Canada, the caustic soda is used to extract bitumen from the oil sands, and four cubic meters of the oil sands process-affected water (OSPW) are produced per one cubic meter of oil sands [1]. The water demand for this process is met by OSPW recycling and by the fresh water uptake from the Athabasca River [2]. As a result, the quality of OSPW which is a complex mixture of solids, dissolved organic and inorganic species as well as metals [3], deteriorates due to concentrating of organic and inorganic constituents during OSPW recycle. Under no-release policy, operating companies store OSPW in tailing ponds in order to avoid its release into the receiving environment [2,4]. To achieve the sustainability of the oil sands operations, OSPW needs to be treated to decrease its toxicity and to facilitate its reclamation for safe release into the receiving environment and/or for treated-OSPW recycle for reuse to minimize the volume of the fresh water uptake. Various chemical, physical, physicochemical and biological treatments have been evaluated in order to develop a performance-efficient and cost-effective strategy for OSPW remediation [5–10].

The use of membrane filtration for water treatment has increased worldwide because of the advances in treatment efficiency and reduction in operational costs [11,12]. Low pressure-driven membrane processes such as microfiltration and ultrafiltration (UF) are being considered as attractive alternatives for municipal and industrial water treatment [13–15]. The use of ceramic membranes due to their excellent chemical resistance to inorganic acids, bases, oxidants, the tolerance to high temperatures and long-

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http://dx.doi.org/10.1016/j.seppur.2013.11.005

er life-span [16–19] has increased over the last decade [13,20–22]. Recently, they have been applied for the treatment of petrochemical wastewaters [15,23,24]. For example, Deriszadeh et al. [15] reported the application of 50 kDa TiO_2 ceramic membrane for the removal of organic contaminants from the oil-field processed waters in Alberta and California.

Due to the high concentration of solids, organic compounds and ionic species, OSPW can cause severe fouling of polymeric membranes and rapid decrease in permeate flux [7,25,26]. Thus, it was shown that chemical cleaning of polymeric nanofiltration (NF) and reverse osmosis (RO) was not capable of restoring the initial permeate flux [26]. As such, in a new approach to the treatment of OSPW we employed ceramic 1 kDa UF membrane for the removal of organic and inorganic compounds from OSPW. We hypothesized that the application of ceramic membrane filtration to the treatment of OSPW will allow to achieve high removal ratios of organic and inorganic contaminants and to maintain high permeate flux due to the better ability of ceramic membranes to sustain cleaning procedures as compared to their polymeric counterparts. The objectives of this study were as follows: (1) to investigate the effect of coagulant addition on the treatment efficiency in terms of permeate flux and membrane selectivity; (2) to evaluate the effect of hydrodynamic conditions such as transmembrane pressure (TMP) and cross-flow velocity (CFV) on the membrane filtration performance; and (3) to evaluate the membrane fouling caused by OSPW and to elucidate the fouling mechanisms. Our previous studies have shown that OSPW pretreatments such as coagulation/flocculation/sedimentation (CFS) or filtration with the direct coagulant addition reduced membrane fouling of NF and RO polymeric membranes [25,26]. The coagulation of OSPW destabilized suspended solids and colloidal matter by reducing their surface charge and promoting their coalescence leading to flocs formation during the flocculation process [6,26]. The flocs were further removed by a sedimentation (i.e., settling) process or on membranes' surfaces during direct filtration [25,26]. Although CFS followed by ceramic membrane filtration has been recently applied for the treatment of surface waters (membranes with the nominal pore sizes of 0.01 μm [13,14] and 0.1 μm [13,14]) and for secondary effluent from wastewater treatment plant (membranes with the nominal pore sizes of 0.1 μm, 0.5 μm and 1.0 μm) [21] in order to enhance the removal of particulate material and dissolved organics, this process has never been investigated with respect to the treatment of OSPW.

The TMP and CFV are factors that significantly influence the separation properties and permeate quality [27–29]. Therefore, it is important to evaluate the operating parameters of ceramic membrane filtration in order to achieve maximum removals of organic and inorganic compounds from OSPW while keeping high permeate flux. A resistance-in-series model [27,30] was used in this study to analyze different types of fouling and foulant/membrane interactions caused by the complex nature of OSPW matrix. The fouling mechanisms were elucidated based on four classical models (cake filtration, complete, standard or intermediate pore blockings) which are commonly applied to describe the fouling behavior of both natural waters and solutions of model compounds [31–33]. The three-mechanism model recently developed by Duclos-Orcello et al. [34] was also used to evaluate the internal and external membrane fouling. Thus, unlike previously published studies on membrane filtration of OSPW, this is the first study which comprehensively examines all aspects of membrane filtration including removal of contaminants, effect of hydrodynamic conditions on process efficiency as well as evaluation of OSPW fouling behavior. Considering membrane's performance, this may decrease operational expences as well as membranes' cleaning and maintenance costs in a long-term prospective.

2. Materials and methods

2.1. OSPW and chemicals

OSPW was collected from an active OSPW recycling pond in Fort McMurray, Alberta, Canada. OSPW was stored in polyvinyl chloride containers at 4 °C and was warmed up to the room temperature (21 ± 1 °C) before conducting the experiments. Due to its high turbidity, raw OSPW was stirred with a mechanical mixer (Model L, Mixing Equipment Co., Inc., Rochester, NY, USA) for 1 min to ensure sample homogeneity. The OSPW characteristics are shown in Table 1. All solutions used in this experimental study were prepared with Milli-Q water (Millipore Corp., Bedford, MA, USA). Solutions used in NAs analysis by ultra performance liquid chromatography were prepared with Optima-grade water (Fisher Scientific, Fair Lawn, NJ, USA). The details of the chemicals can be found in Supplementary Information (SI).

2.2. Jar tests

The optimum coagulant dose was determined in a six-paddle jar-test apparatus (PB-700 Jar Tester, Phipps & Bird, Richmond, VA, USA) by using the procedure developed in previous studies [6,25]. Aluminum sulfate octadecahydrate ($Al_2(SO_4)_3 \cdot 18H_2O$) in the range of 20–145 mg/L (as alum) was added to raw OSPW and the water was rapidly mixed at 120 rpm for 30 s, followed by slow mixing at 30 rpm for 10 min. As the flocs were formed, the samples were allowed to settle for 1.5 h [6,25]. Each alum dose was tested in triplicate.

2.3. Membrane filtration system

Fig. 1 shows a schematic of the membrane filtration system. The system consisted of a membrane module, recirculation pumps, a feed tank and a data acquisition system. The membrane housing and fittings were purchased from TAMI Industries (Nyons, France). The volume of the feed OSPW tank was 3 L. The system was operated in a total recycling mode in which permeate was recycled, using a Master Flex L/S peristaltic pump (ColeParmer, Chicago, IL, USA), into a feed tank once the permeate volume reached 200 mL. The TMP and CFV were set up using a mechanical pump (Model 75211-10, ColeParmer, Montreal, QC, Canada) and a back-pressure valve (Swagelok, Solon, OH, USA). The mass of permeate was measured by an electronic balance (Scout Pro, Ohaus Corp., Parsippany, NJ, USA). CFS-pretreated OSPW (using the optimum coagulant dose) served as a feed water to the membrane filtration system (i.e., CFS treatment followed by membrane filtration (denoted as "CFS-filtration")). To evaluate the effect of sedimentation alone on membrane filtration, OSPW treated by coagulation/flocculation without sedimentation step was tested in a separate experiment, denoted as "direct filtration".

2.4. Ceramic membranes

Two tubular 1 kDa membranes with seven channels (inner diameter: 0.1 cm; hydraulic diameter of each channel: 0.02 cm; membrane length: 25 cm) (Inside CéRAM™, TAMI Industries, Nyons, France) were used for CFS-filtration and direct filtration, respectively. The total effective filtering area of each membrane was 132 cm^2. The membranes' selective and support layers were TiO_2/ZrO_2 and TiO_2, respectively [16]. Upon receiving the membranes were cleaned with 0.4 M sodium hydroxide (NaOH) and phosphoric acid (H_3PO_4) according to the manufacturer's technical directions [16]. The membranes' flux and removal were measured according to Eqs. S1 and S2, respectively (see SI). After each exper-

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