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Treatment of oil sands process-affected water by submerged ceramic membrane microfiltration system

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ABSTRACT

With the rapid expansion of the oil sands in Northern Alberta over past decade, oil sands process-affected water (OSPW) management has become a significant issue. Currently, there is an urgent need for the OSPW reuse and/or safe discharge. In this study, coagulation followed by submerged microfiltration (MF) with a range of ceramic membranes was investigated as a potential process for pretreating OSPW. The ceramic MF membranes with the average pore size of 100 nm were made of Al_2O_3 and their selective layer was further modified with SiO_2 or TiO_2 nanoparticles. Our results showed that membrane surface charge played an important role in controlling membrane fouling. The maximum fouling reduction was achieved when OSPW was treated by SiO_2-modified membrane. Surface roughness studies also demonstrated its significant effect on membranes' filtration performance and fouling. More foulants were deposited on the surface of rough TiO_2-modified membrane as compared to smoother SiO_2-modified membrane. It was found that removal of organic and inorganic fractions in OSPW was not affected neither by the type/concentration of coagulant nor by membrane's surface modification. Thus, more than 93% removal of the total suspended solids and less than 17% and 10% removals of the total organic carbon (TOC) and chemical oxygen demand (COD), respectively, were achieved regardless of the membrane type and applied treatment. When MF permeate was used as an influent to the reverse osmosis (RO), this resulted in 98% and 100% reductions in TOC and COD concentrations, respectively. Moreover, RO filtration achieved more than 95% removal of magnesium, calcium, iron, fluoride, chloride, sulfate, nitrate, sodium and potassium and silica. The results of this study showed that combination of RO and MF could be considered as an effective and feasible option for OSPW treatment.

1. Introduction

The oil sands in Northern Alberta are the third-largest oil reserves in the world [1]. For example, in 2011 marketable oil production from the Canadian oil sands was 1,617,600 barrels per day, representing 73% of the total crude bitumen production [1]. Bitumen recovery through the surface mining is achieved by the Clark hot water extraction, in which hot water is mixed with the oil sands [2], leading to a production of a large volume of the oil sands process-affected water (OSPW) [2]. OSPW is a complex mixture of suspended and dissolved solids, salts, and persistent organic compounds that are extremely toxic to aquatic ecosystems if directly discharged to the environment [2].

Because of OSPW complexity, the remediation methods usually consist of several steps including physical, chemical, and biological treatments [3]. Generally, suspended solids are removed firstly because they cause membrane fouling or aversively affect the advanced oxidation process (e.g., by scattering UV light) [4,5]. Physical and physicochemical treatments to remove suspended solids include gravity separation, centrifugal settling, granular media filtration, membrane filtration and coagulation–flocculation–sedimentation (CFS) [4]. Among these treatments, low pressure membrane filtration, such as microfiltration (MF), is advantageous because of its relatively low cost and energy consumption as well as high quality effluent [5]. Although efficiency of MF in removing suspended solids in the oil field-produced water has been demonstrated [6], MF has not been yet examined for the OSPW treatment.

The limitation of MF for the treatment of the oil field-produced water is flux decline, resulting from the adsorption or accumulation

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of suspended and dissolved solids on membrane's surface or inside the membrane pores [5]. OSPW contains high concentration of suspended solids that can be deposited on the membrane surface, increasing its hydraulic resistance and trans-membrane pressure (TMP). As a result, operational expenses such as chemical cleaning and maintenance costs are increased, and the lifespan of membrane is shortened [7].

The recent advances in nanotechnology have allowed fabrication of nanocomposite ceramic membranes in which nanoparticles are embedded into ceramic matrix [8]. Metal oxides (e.g., titanium dioxide (TiO_2) and silicon dioxide (SiO_2)) are frequently used for manufacturing nanostructured ceramic membranes for wastewater treatment [8]. Ceramic membranes have high resistance to mechanical, chemical and thermal stresses as well as cleaning agents [5]. They can also operate under high pressure differentials [5]. It has been widely reported that surface modification by nanoparticles could change membrane's surface charge, resulting in decreasing membrane fouling and increasing permeate flux of different wastewaters [9,10]. Therefore, surface-modified ceramic membranes are expected to reduce fouling load on membrane surface during OSPW filtration.

This study investigated removal of suspended solids and organic matter in OSPW which was treated by coagulation–flocculation and submerged ceramic MF (CF–MF). CF process was selected for the pretreatment of OSPW to enlarge the particle size so that they could be better retained by membrane. The pretreatment was also aimed to mitigate the membrane fouling caused by OSPW solids, such as sand, silt and clay. The sedimentation step was omitted in CF–MF process in order to reduce the operational and maintenance cost. In a separate series of experiments we also evaluated the feasibility of the sequential CF–MF and reverse osmosis (RO) for complete OSPW treatment. The membranes were cast of Al_2O_3 nanoparticles and used to evaluate the effect of different coagulants and solution pH on membranes' fouling and permeate quality. In order to investigate the effect of surface charge on filtration performance, membranes were further coated with TiO_2 or SiO_2 nanoparticles and these surface-modified membranes were tested in optimal filtration conditions. MF filtration efficiency was assessed through the trans-membrane pressure (TMP) decrease. Scanning electron microscopy (SEM), atomic force microscopy (AFM) and energy-dispersive X-ray spectroscopy (EDX) were used to investigate the membrane fouling and surface morphology. In addition, CFS tests were also conducted in order to compare the optimal coagulant concentrations required for CF–MF and CFS processes.

2. Experiments

2.1. OSPW and chemicals

Raw OSPW was collected from the oil sands tailings pond in Northern Alberta. The OSPW was preserved in polyvinyl chloride barrels and stored at 4 °C in a temperature controlled room at the University of Alberta. Before experiments, OSPW was vigorously mixed with a mechanical stirrer to dissipate suspended solids and left overnight to warm to a room temperature (22 ± 1 °C). The OSPW characteristics are shown in Table 1.

Alum ($Al_2(SO4)_3{\cdot}18H_2O$), polyaluminum chloride (PAC) ($Al_2(OH)_nCl_{6-n}$ and sodium hydrochloride (NaOH) were supplied by Fisher Scientific, Fair Lawn, NJ. Sodium hypochlorite (NaOCl) was purchased from Ricca Chemicals (Arlington, TX). Chemicals were of analytical grade and were used without further purification. Milli-Q water (Millipore Corp., Bedford, MA) with a resistivity of 18.2 MΩ cm and conductivity of less than 1 μS cm^{-1} was used in all experiments.

Table 1
Physico-chemical composition of raw OSPW.

Parameter	Value
pH	6.9 ± 0.6
Conductivity (μS/cm)	3671.5 ± 227.7
Turbidity (NTU)	25.6 ± 2.1
TOC (mg/L)	41.2 ± 0.8
COD (mg/L)	134.6 ± 9.5
TSS (mg/L)	21.2 ± 3.5
TDS (mg/L)	1906.2 ± 25.9
SDI_5	6.2 ± 0.3
Magnesium (mg/L)	12.6 ± 0.7
Calcium (mg/L)	25.9 ± 1.04
Iron (mg/L)	0.2 ± 0.1
Silicon (mg/L)	5.7 ± 0.8
Fluoride (mg/L)	2.0 ± 0.3
Chloride (mg/L)	1291.3 ± 497.4
Sulfate (mg/L)	60.6 ± 14.5
Nitrate (mg/L)	0.9 ± 0.1
Sodium (mg/L)	705.1 ± 18.3
Potassium (mg/L)	13.1 ± 1.2

2.2. Membranes

The Al_2O_3 ceramic MF membranes were fabricated by Meidensha Corp. (Tokyo, Japan). Schematic of ceramic membranes used in this study is shown in Fig. 1. The membranes used in this study were rectangular-shaped with a number of internal cylindrical channels. The filtration was performed in the "outside-out" mode and permeate was collected in membrane's inner channels. The membrane consisted of two layers: a selective layer and a supporting layer. Membrane's surface was then modified with SiO_2 or TiO_2 nanoparticles The membrane modification techniques are a proprietary process of Meidensha Corporation. Flat-sheet polymeric RO membrane was supplied by the GE Osmonics (Fairfield, CT). The RO membrane was first soaked in Milli-Q water for at least 24 h and then compressed to achieve a stable permeate flux. The membranes' properties are listed in Table 2.

2.3. Coagulation–flocculation and coagulation–flocculation–sedimentation tests

CF tests were conducted with PAC and alum at a concentration range of 0.5–60 mg/L. The optimum coagulant concentrations for

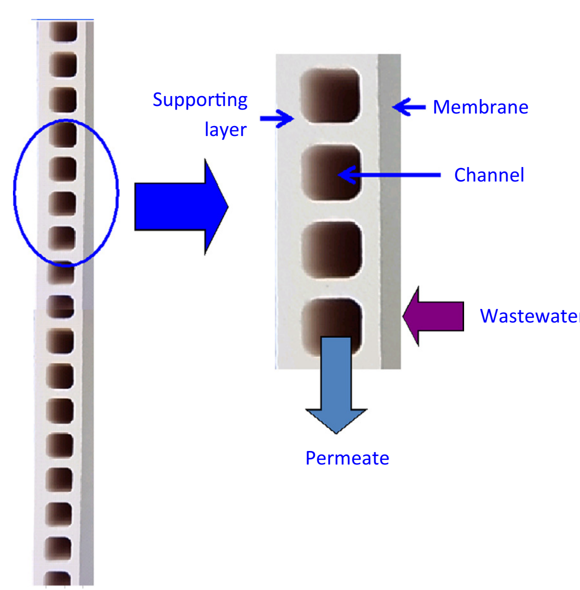

Fig. 1. Schematic of Al_2O_3 ceramic membrane.