



Treatment of industrial wastewater produced by desulfurization process in a coal-fired power plant via FO-MD hybrid process

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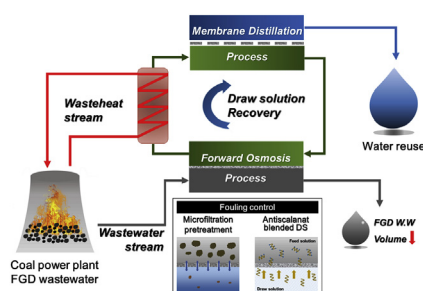
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HIGHLIGHTS

- Developed FO-MD hybrid to treat FGD wastewater with high fouling potential.
- Severe fouling, including CaSO_4 scaling, was observed in FO.
- MF pretreatment and antiscalant-blended DS controlled FO fouling effectively.
- FO with fouling control achieved the highest recovery compared to RO and MD.
- Antiscalant should be developed for the scaling control in both FO and MD.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, the feasibility of forward osmosis (FO) hybridized with membrane distillation (MD) was systematically investigated for treating flue gas desulfurization (FGD) wastewater. FO experiments were conducted using raw FGD wastewater obtained from a coal-fired power plant in Korea. Severe membrane fouling in FO was observed since FGD wastewater contained various components (i.e., particles, colloids, organics, and ions). The combined fouling layer by particulates and scales was identified via scanning electron microscope (SEM), energy dispersive X-ray (EDX) and X-ray diffraction (XRD). Therefore, fouling control strategies were suggested and evaluated. Microfiltration (MF) pre-treatment was effective in removing particulates and mitigating the initial fouling. Antiscalant-blended draw solution (DS) could inhibit the formation of membrane scaling. With such fouling control schemes, FO achieved the highest recovery rate compared to other desalting processes (i.e., RO and MD), suggesting that FO is suitable for treating wastewater with high fouling potential and high TDS. Finally, the diluted DS was recovered by MD. MD could re-concentrate the diluted DS up to 50% recovery rate with no significant flux decline. Rapid flux decline was then observed due to membrane scaling. Thus, appropriate antiscalants in DS should be considered to inhibit scaling formation in FO and MD simultaneously.

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1. Introduction

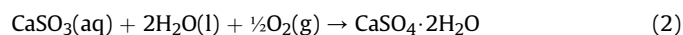
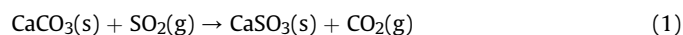
During the last several decades, energy consumption has significantly increased in the world. Electricity usage is known to be

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approximately 24,659 TWh and it is expected to double by 2040 (Stern, 2011; Kopplitz et al., 2017). To meet the demand for increasing electricity consumption, conventional fossil fuel power plants and nuclear power plants have been intensively utilized. However, recently, there have been many attempts to develop renewable energy such as biomass, wind, solar, geothermal and fuel cells, which are considered as eco-friendly energy resources for electricity production (Liserre et al., 2010). This paradigm shift was caused by the movement toward a reduction of greenhouse gas emitted from fossil fuel power plants and an escape from the concerns associated with nuclear power plants (Davison, 2007). Furthermore, coal-fired power plants used for 200 years have generated considerable air pollution from combustion ash and sulfur dioxide gas (Wu et al., 2004; Davison, 2007; Kopplitz et al., 2017).

Recently, a flue gas desulfurization (FGD) process has been applied in coal-fired power plants to remove sulfur dioxide gas and combustion ash in exhaust gas (Srivastava and Jozewicz, 2001; Wu et al., 2004). The wet limestone-gypsum FGD process is most widely utilized owing to its high desulfurization efficiency and reliability (Srivastava and Jozewicz, 2001; Srivastava et al., 2001). In the combustion process with coal, SO₂ as acid gas is produced. It can be removed by either CaCO₃ (limestone) shaped slurries or other alkaline materials used from the flue gases (Srivastava et al., 2001). During the reaction from wet scrubbing with a CaCO₃ slurry, CaSO₃ (calcium sulfite) can be formed in the dry form as in Eqn. (1) (Zhao et al., 2007). To further oxidize the CaSO₃ to produce valuable CaSO₄·2H₂O (gypsum), which can be used in wallboard and other products, dry sorbent injection systems can be employed and should be operated under forced oxidation as in Eqn. (2).



However, the operation of the FGD process generates wastewater including dissolved fine dust in exhaust gas, inorganic sludge from surplus calcium carbonate and calcium sulfate particles formed by the reaction between calcium carbonate solution and sulfur dioxide gas (Meij, 1994). Owing to its harsh composition, the discharge and recycling of FGD wastewater are limited. Therefore, conventional physical and chemical treatments such as coagulation-precipitation and biological process to remove nitrogen have been widely used for the reuse of FGD wastewater (Dahl et al., 1997; Zhang et al., 2010). Recently, membrane processes such as microfiltration (MF) or ultrafiltration (UF) and zero-valent iron process have been evaluated for better treatment efficiency, but treated effluent still cannot be discharged or reused because of its high TDS (Enoch et al., 1990; Huang et al., 2013).

Nowadays, zero liquid discharge (ZLD) has attracted industrial and municipal organizations as well as the environment due to its lack of discharge, and thus, the concentration of wastewater is becoming more important. Therefore, desalting membrane processes such as reverse osmosis (RO), membrane distillation (MD), and forward osmosis (FO) have been studied for the concept of ZLD (Koppol et al., 2004; Ji et al., 2010; Tong and Elimelech, 2016). Although they could successfully produce a permeate with low TDS, however, they also suffered from severe membrane fouling owing to high fouling potential, which can limit efficient operation (Hong and Elimelech, 1997; Choi et al., 2016). Therefore, it can be concluded that effective fouling control is a key factor in maintaining efficient and continuous operation for the treatment of FGD wastewater.

FO is considered an alternative to RO, which is widely used for desalting wastewater with high TDS. Since FO utilizes the osmotic

pressure difference as a driving force between feed and draw solutions, it can achieve low energy consumption as well as high fouling reversibility thorough non-pressurized operation. As a result, FO can be beneficial for controlling membrane fouling. In addition, high rejection rate induced by the reverse solute flux (RSF) of draw solution is a strength in the treatment of FGD wastewater (Lee et al., 2010; Choi et al., 2017). Furthermore, FO can theoretically achieve a high recovery rate using DS at a high concentration (Kim et al., 2017c). Based on these merits, the application of FO has been expanded from desalination to value-added industries and the wastewater treatment industries (Coday et al., 2014). For instance, the FO technology was applied for juice concentration and recovery of valuable materials (Garcia-Castello and McCutcheon, 2011; Phuntsho et al., 2011). In addition, the treatment of shale gas produced water, landfill leachate and RO brine having high membrane fouling potential was carried out by FO due to high fouling reversibility (Achilli et al., 2009; Coday et al., 2014; Lee et al., 2016). Besides, FO can be integrated with MD for the efficient recovery of DS (Xie et al., 2013, 2014). Based on these previous studies, FO can be expected to efficiently concentrate FGD wastewater for ZLD. The provider of commercial FO systems (OASYS Water, Inc., USA) installed the FO system to treat FGD wastewater from a coal-fired power plant in China, but they have not reported their operation results yet. In addition, there has been no fundamental study on the treatment of FGD wastewater by FO.

The objective of the present study is to fundamentally evaluate the feasibility of FO integrated with MD for the treatment of FGD wastewater. Raw FGD wastewater was collected from the real-scale coal-fired power plant in Samcheonpo, Korea and was treated by developing an FO-MD hybrid process systematically. Membrane fouling in FO during the treatment of raw FGD wastewater was evaluated and the cleaning strategies were further investigated by employing not only typical hydraulic washing but also MF pre-treatment and antiscalant-blended DS. Then, the FO performance was compared with those of RO and MD. Lastly, the recovery of the diluted DS by MD was evaluated and the water quality of the final product was examined for reuse in power plants.

2. Materials and methods

2.1. FO membrane and draw solutions

The thin-film composite (TFC) polyamide (PA) FO membrane used in this study was provided by Porifera, Inc. (Hayward, CA, USA). The membrane was fabricated with a thinner flat-sheet form and for operating under non-pressurized osmosis. The detailed specifications of this membrane are given elsewhere (Choi et al., 2017). The membranes were stored in a dried form in a desiccator according to the manufacturer's recommendations. The membrane coupon was immersed in deionized (DI) water for 30 min prior to all FO experiments.

Sodium chloride (NaCl) was received at reagent grade (Junsei Chemical, Japan) and used as draw solutes. Poly (aspartic acid sodium salt) (PAspNa) (CAS-No. 181828-06-8) in powder form was supplied by Lanxess AG (Cologne, Germany) for preparation of the antiscalant-blended DS. The DS was prepared by dissolving in DI water.

2.2. FO experiments

All FO experiments were carried out using a lab-scale FO system similar to the one described in our previous studies (Kim et al., 2015). The FO cell had two symmetric channels consisting of 77 mm long, 26 mm wide and 3 mm deep on both sides of the membrane for each FS and DS. Variablespeed gear pumps

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