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A novel microwave assisted photo-catalytic membrane distillation process for treating the organic wastewater containing inorganic ions

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ABSTRACT

A novel microwave assisted photo-catalytic membrane distillation (MPMD) process was proposed in this study to treat organic wastewater containing inorganic ions. Humic acid (HA) solution containing Ca^{2+} was used as feed to investigate the effects of microwave irradiation and ultraviolet (UV) radiation on the MD process. The membrane surface and composition of the fouling layer was studied using scanning electron microscopy (SEM) coupled with energy dispersion spectrometry (EDS). Ca^{2+} affected the permeation flux by forming complex compounds with humic acids and resulted in coagulation on the membrane surface in a conventional MD process. Microwave irradiation effectively decreased the deposit on membrane surface by destabilizing complex compounds. Organic matter was photo-degraded by the microwave assisted photo-catalysis so that the membrane fouling was prevented. In the MPMD process, the flux remained nearly constant. After 45 h of operation with the continuously concentrated solution, the concentration of the solution in photocatalytic reactor was about 2.8 times than the initial and J/J_0 equaled to 94.5%. In the end of MPMD process, J/J_0 equaled to 98.5%. The conductivity of the distillate over the operation time was less than 4 μS cm^{-1}. MPMD was used to treat the coal gasification wastewater. The results clearly demonstrated that the MPMD process was sufficient to treat the coal gasification wastewater with a removal rate higher than 96% for CODcr and 98% for NH_4^+-N after an operation over 120 h.

1. Introduction

Membrane distillation (MD) has many advantages including low energy consumption, attainable operating conditions, high product purity and simple maintenance over other membrane-based water treatment processes. Several pilot scale studies using MD process such as seawater desalination, oily wastewater purification, and natural organic matter (NOM) removal for potable water production have been investigated in the past.

Membrane distillation is considered to be a promising technology for desalination of high concentration salty water. However, there are still some obstacles, such as inadequate permeate flux for amplified reactor, membrane fouling and membrane wetting phenomena, which impede the large-scale application of membrane distillation. Similar to other membrane-based processes, membrane fouling is one of the major concerns for the MD process. As a result, many research efforts have been dedicated to this topic, and several strategies to alleviate and/or prevent fouling have been proposed. Among these studies, Srisurichan et al. [1] reported that the high concentration of divalent cations (Ca^{2+}) could form complexes with humic acids which coagulated on the membrane surface, thereby leading to a thick fouling layer. Consequently, the heat transfer resistance increased, and the number of pores available for water vaporization reduced. Hsu et al. [2] carried out a comparison study of the effects of NaCl solution and seawater on membrane fouling in a MD process. The results revealed that the NaCl feed solution showed much less concentration polarization, and therefore substantially slowed down the build-up of membrane scaling compared to the case when seawater was used as the feed solution. In order to tackle the issues with membrane fouling in MD

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processes, ultrasonic cleaning technique was introduced to remove the fouling layer and subsequently restore the permeation flux. Hou et al. [3] designed a novel ultrasonic assisted direct contact membrane distillation hybrid process to utilize ultrasonic irradiation for membrane scaling mitigation. The results revealed that ultrasonic irradiation mitigated the $CaSO_4$ induced membrane scaling and prevented the decline of permeate flux. The microstreaming, shock wave and acoustic vortex streaming brought by ultrasonic wave stimulated the interaction on liquid-membrane interface, and therefore prevented the deposition of $CaSO_4$ crystals. The application of appropriate pretreatment enabled the removal of foulants from the feed, and performed well on preventing fouling [4,5].

Nowadays the development of innovative photo-catalytic membrane reactors (PMRs) has received considerable attentions, which combines the photo-catalysis with membrane processes and takes advantage of the photo-degradation of pollutants and membrane separation technology. However severe fouling was found on this particular type of membrane reactor when used in pressure-driven membrane processes such as microfiltration (MF) and ultrafiltration (UF). In this regard, it was postulated that the use of photocatalytic membrane reactor in a thermal-driven MD process can potentially mitigate the fouling behavior [6,7]. In this line of development, the use of photo-catalytic membrane distillation (PMD) process for the degradation of azo dyes and separation of by-products was examined [6,8], and clear evidence was found that the use of MD process showed much less fouling behavior in comparison with the pressure-driven membrane processes.

However the major concern around the present PMRs is that the photo-catalytic degradation and the membrane process have to take place in different reactors since traditional UV lamps cannot be placed in water. Therefore the ultraviolet radiation cannot directly work on membrane surface. On the other hand, microwave electrodeless lamp (MWL), which has been reported in many microwave assisted photo-catalytic studies [9,10], was suitable to be installed in the water. Furthermore, the reaction under microwave irradiation was found to be more efficient [11,12]. In addition, MWL, powered by microwave, has been widely used in photo-catalytic processes because of its low cost, simple equipment, and high photo-chemical efficiency [13–15].

More importantly, another benefit can be brought from microwave irradiation is that it can produce micro disturbance in solution containing polar molecules [16], and therefore provides homogeneous heating which minimizes the undesired temperature polarization normally observed in MD process. In our previous work, the effect of microwave irradiation on mass transfer and crystallization in membrane distillation had been discussed [17,18]. The results demonstrated that the microwave irradiation increased the mass transfer coefficient by 27.7% during a vacuum membrane distillation (VMD) process thereby resulting in high permeate fluxes even at a low feed flow rate.

In this work, a novel microwave assisted photo-catalytic membrane distillation (MPMD) reactor was set up using MWL as light source and TiO_2 as the catalyst to couple the microwave assisted photo-catalysis and VMD process in the same reactor. From above, in addition to the benefits of a conventional PMR, such as high quality permeate and alleviated membrane fouling, a significant advantage of the developed MPMD is that microwave irradiation can enhance both the photo-catalysis efficiency and the VMD mass transfer [19].

In this extending study, two calcium salts and humic acid were selected as research objects to investigate the effect of microwave on the VMD process and the photocatalytic membrane distillation process, the membrane fouling behavior in MPMD was also be studied. Furthermore, the use of MPMD set-up for the treatment of coal gasification wastewater was also studied in this work.

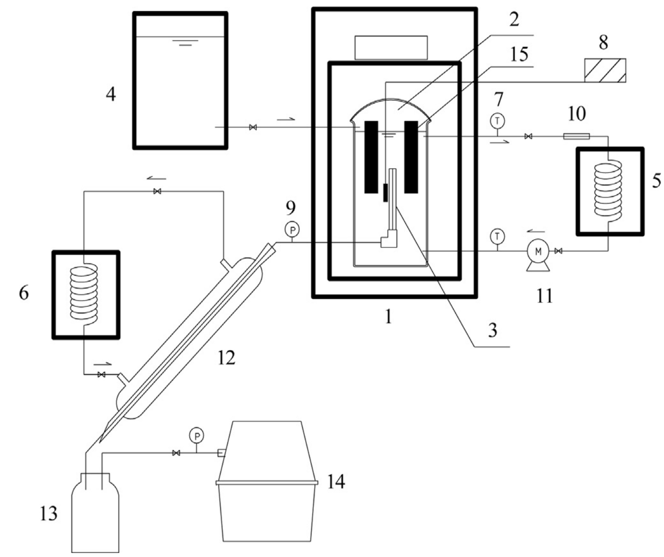

Fig. 1. MPMD experimental setup: 1 microwave oven, 2 photo-catalytic reactor, 3 capillary hollow fiber membrane module, 4 feed reservoir, 5 heat exchanger, 6 cooling system, 7 thermometer (T), 8 optical fiber thermometer, 9 vacuum meter (P), 10 flowmeter, 11 magnetic force driving pump, 12 condenser, 13 permeate reservoir, 14 vacuum pump, 15 MWL.

2. Materials and methods

2.1. Materials

Polypropylene (PP) hollow fiber membranes were supplied by Hangzhou Kaijie Membrane Separation Technology Co., Ltd, China with an outer diameter of 0.43 mm and inner diameter of 0.33 mm. These membranes had pore sizes from 0.1 to 0.2 μm, and porosity around 50%. The chemicals (NaCl, $CaCl_2$, Na_2SO_4, $CaSO_4 \cdot 2H_2O$, NaOH and hydrochloric acid) used in this work were all of analytical reagent grade, and supplied by Beijing Chemical works (China). Humic acid (HA) was purchased from Sinopharm Chemical Reagent, China. As HA dissolves well under alkaline conditions, a stock feed solution of HA at about 300 mg/L was prepared by dissolving 1.5 g of HA in 5 L of NaOH (2 mmol/L) solution, and the insoluble substance was removed by filter paper after magnetic agitation of this solution during 24 h, and then was conserved at 4 °C in the absence of light. The pH of the feed was adjusted to the desired values by adding 1 mol/L HCl or 1 mol/L NaOH. A commercially available TiO_2 (Aeroxide P25, Evonik, Germany) was used as the photo-catalyst.

2.2. Experimental set-up

The schematic representation of the experimental set-up is shown in Fig. 1. The microwave assisted photocatalytic reactor was installed in a microwave oven (Haier. Co. Ltd.; power, 750 W; frequency, 2.45 GHz, China), and the immersion hollow fiber module was fixed in the photocatalytic reactor. The hollow fiber membrane module contained 40 PP hollow fiber membranes with an effective area of 0.0119 m^2. The U-shaped MWL was made of quartz and filled with mercury and argon. MWLs (Jimin Illumination Equipment Factory, Shanghai, China), filled with 20 mg Hg and 5.0 Torr Ar, were used as the light source. The MWLs emitted UV and visible lights upon microwave irradiation. The irradiation spectrum showed peaks at 254, 312, 365, 405 and 435 nm, and the main UV wavelength was 254 nm (provided by the manufacturer). The MWL was suspended on the surface of the solution and about 60% was immersed. During the reaction, the power of the MWL measured to be less than 28.1 W [9].