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Experimental study of ammonia removal from water by modified direct contact membrane distillation

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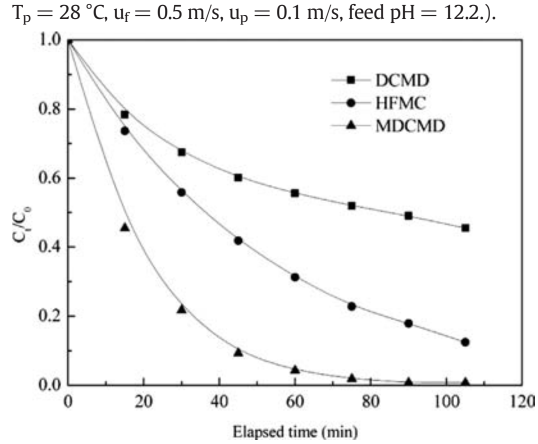
HIGHLIGHTS

- MDCMD with receiving solution in permeate was developed for ammonia removal.
- Ammonia removal efficiency of DCMD, HFMC and MDCMD was investigated.
- The feed concentration, pH, temperature and flow rate was studied.
- The highest ammonia removal efficiency was observed in MDCMD.
- Feed pH was the most dominant factor in MDCMD.

GRAPHICAL ABSTRACT

Variation of feed ammonia concentration in HFMC, DCMD and MDCMD (HFMC: $u_f = 0.5$ m/s, $u_p = 0.1$ m/s, feed pH = 12.2; DCMD: $T_f = 50$ °C, $T_p = 28$ °C, $u_f = 0.5$ m/s, $u_p = 0.1$ m/s, feed pH = 12.2; MDCMD: $T_f = 50$ °C, $T_p = 28$ °C, $u_f = 0.5$ m/s, $u_p = 0.1$ m/s, feed pH = 12.2.).



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ABSTRACT

In this work, a modified direct contact membrane distillation (MDCMD) with receiving solution in permeate was developed for accelerating ammonia removal from aqueous solution. The ammonia removal efficiency by means of DCMD, hollow fiber membrane contactor (HFMC) and MDCMD was comparatively investigated. And the effect of feed pH, temperature, flow rate and concentration on ammonia removal efficiency and the permeate flux in MDCMD process was studied. Experimental results showed that the ammonia removal efficiency of DCMD, HMC and MDCMD was 52%, 88% and 99.5% within 105 min, respectively. In MDCMD, feed pH value was proved to be the most dominant factor and the optimal feed pH was 12.20. Increasing feed temperature and flow rate led to higher ammonia mass transfer coefficient, ammonia removal efficiency and permeate flux within the range studied. The initial ammonia concentration had a negligible effect on ammonia removal efficiency.

1. Introduction

Ammonia is a common pollutant in industrial and municipal wastewaters and its accumulation in water leads to eutrophication and depletion of oxygen due to nitrification and thus harms the waterborn organisms [1]. Some conventional techniques have been applied to remove ammonia from water and wastewater such as air stripping [2], break-point chlorination [3], ion-exchange [4] and biological nitrification–denitrification [5]. However, these techniques cannot satisfy the increasing disposal need, or solve the problem with high efficiency and acceptable cost. Thus, there is a continuing need for an alternative separation technique for more efficient ammonia removal from water.

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Recently, hollow fiber membrane contactors (HFMCs) have been developed for volatile component removal from water such as ammonia [6–10], CO_2 [11], chloroform [12], and toluene [13]. In such operations, hydrophobic membrane is used as a barrier between gas and liquid, and the volatile component diffuses through membrane pores and reacts with the receiving solution for removal. HFMCs are often operated at room temperature and no temperature difference exists between the feed side and the permeate side. Compared with conventional air stripping processes, the membrane-base stripping process provides many advantages including larger interfacial area, independent control of gas and liquid flow rate without any flooding or foaming, etc. [14–16]. Some researchers experimentally and theoretically investigated the effect of operating parameters and configurations on the ammonia removal efficiency in HFMCs [6–10]. Results showed that feed pH had a significant effect on ammonia removal efficiency while the feed ammonia concentrations had negligible effects on ammonia removal [6–8]. Tan et al. developed a mass transfer model based on plug flow behavior for the ammonia removal in PVDF hollow fibers. At any cross-section of the lumen, overall mass transfer coefficient was empirically estimated [9]. Rezakazemi et al. developed an unsteady state two-dimensional mathematical model implemented in linked MATLAB–COMSOL Multiphysics, which could be used to evaluate the effective parameters involved in the ammonia removal by HFMCs [10].

Membrane distillation (MD), as a desalination technique, may also be regarded as a membrane-based stripping process if it is used to remove volatile component from water. Being different from HFMCs, MD is a thermally driven process and temperature difference exists between the feed and permeate side, so the water vapor as well as the other volatile component can transfer across the micropores to the permeate side [17]. Different MD configurations have been investigated for ammonia removal from water [18–20]. Ding et al. compared the mass transfer coefficient and selectivity of different MD configurations for ammonia removal [18]. Vacuum membrane distillation (VMD) showed the highest mass transfer but the lowest selectivity, while direct contact membrane distillation (DCMD) enabled the highest selectivity and moderate mass transfer. The sweeping gas membrane distillation (SGMD) gave moderate selectivity and the lowest mass transfer. EL-Bourawi et al. investigated the applicability of VMD with polytetrafluoroethylene membrane for ammonia removal. Experimental results showed that high feed temperatures, initial feed concentration and pH levels enhanced ammonia removal efficiency [19]. Another SGMD applied for ammonia removal from wastewater containing 100 mg/L ammonia had been studied at pH 11.5. Up to 97% removal of ammonia was achieved on the condition of highest temperature and fastest gas flow rate, resulting in an ammonia concentration in the treated water being as low as 3.3 mg/L [20].

As reported in the literatures [6–10], the receiving solution in permeate can significantly accelerate the ammonia removal efficiency in HFMCs. And also, in a DCMD process for ammonia stripping, water vapor as well as ammonia can both transfer across the membrane to the permeate side due to the temperature difference, which may lead to wastewater volume minimization.

So in the present work, a modified DCMD (MDCMD) with receiving solution in permeate was developed for accelerating ammonia removal from water. To the best of authors' knowledge, few works had been focused on the comparison of DCMD and HFMC, as well as the developed MDCMD. Thus, the ammonia stripping efficiency by means of DCMD, HFMC and MDCMD were investigated at first. After that, the effects of feed ammonia concentration, pH, temperature, flow rate on ammonia stripping efficiency and permeate flux in MDCMD process were experimentally investigated.

Table 1
Properties of the PVDF membrane used in the experiments.

Parameter	Value
Outer diameter (mm)	1
Inner diameter (mm)	0.8
Fiber thickness (mm)	0.1
Porosity (%)	80%
Average pore radius (μm)	0.22
LEPw (kPa)	250
Contact angle θ (°)	87°

2. Experimental

2.1. Membrane and membrane module

The hydrophobic polyvinylidene fluoride (PVDF) capillary membranes used in the experiments were self-made and the characteristics are shown in Table 1 and Fig. 1. The membrane module was made by a polyester tube and two UPVC T-tubes. The outside/inside diameters of the module were 20 mm/15 mm and the effective length of the module was 150 mm. The module was equipped with 50 hydrophobic hollow fiber PVDF membranes. The total efficient area of the module was calculated based on the inner diameter and amounted to 141.3 cm^2.

2.2. Experimental setup

The experimental setup for ammonia removal is schematically illustrated in Fig. 2. The feed ammonia solution was circulated through the lumen of the membrane module, while the permeate was circulated through the shell of the membrane module. Both the feed and permeate were pumped to their reservoirs via two magnetic pumps (MP-15RN, Shanghai Seisun Pumps, China), respectively. The feed solution could be heated through a thermostat (XMTD-2202, Yongshang Instruments, China) while the permeate could be cooled by a cooler (SDC-6, Nanjing Xinzhi Biotechnology, China) if needed. There were 4 thermometers equipped on both the inlet and the outlet of the feed and the permeate sides. Two flow meters were equipped in the feed and the permeate side, respectively.

In this study, ammonia removal experiments based on the HFMC, DCMD and MDCMD were all investigated using the above installation. In the HFMC, the ammonia stripping was investigated at room temperature without heating and cooling, but the receiving solution containing 0.01 mol/L sulfuric acid was in permeate. In the DCMD process, the feed was heated via the thermostat and the permeate was cooled via the cooler and no receiving solution was in permeate. While in a MDCMD process, receiving solution containing 0.01 mol/L sulfuric acid was used in the permeate side.

2.3. Experimental procedure

Solutions containing ammonia were prepared through different additions of ammonia chloride into distilled water. The pH was adjusted by adding HCl and NaOH to the feed solution. Samples of the feed solution were taken from the feed tank every 15 min, and the ammonia concentration was calculated by Nessler's reagent colorimetric method using a UV–vis spectrophotometer (DR/4000U, HACH, USA). The average permeate flux (J_{aver}) was measured by the overflow volume of the permeate reservoir, and could be calculated as:

$$J_{aver} = \frac{V_p}{A \cdot t}. \qquad (1)$$

The ammonia removal efficiency (R) could be defined as:

$$R = \left(1 - \frac{C_t}{C_0}\right) \times 100\%. \qquad (2)$$