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Effects of Absorbents on Ammonia Removal from Wastewater Through Hollow Fiber Membrane Contactor

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ABSTRACT

The aim of this study is mainly to evaluate the effects of absorbents on ammonia removal from wastewater through hollow fiber membrane contactor. There were two absorbents used in the experiments namely sulfuric acid solution and Natural Hot Spring Water (NHSW)-based solution and the pH of absorbents are 2.3 and 0.8. In the ammonia removal experiments, wastewater solution was flowed through the shell side of the contactor, whilst absorbent solution flowed through the lumen side of the fibers. Operating variables such as pH and ammonia initial concentration of wastewater are used as tested variables for evaluating effectiveness of the absorbents. Experimental results show that the ability of NHSW-based solution can compensate the sulfuric acid solution to absorb ammonia through hollow fiber membrane contactor at the same pH of wastewater. The NHSW-based solution give higher ammonia removal efficiency and overall mass transfer coefficient than sulfuric acid solution at ammonia initial concentration of 800 ppm in the ranged pH of wastewater applied in this study. The types of absorbent, at the pH of wastewater of 11, had a negligible effect on ammonia removal efficiency and overall mass transfer coefficient in the range of ammonia initial concentration of 100-800 and 100-400 ppm for the pH of absorbents of 2.3 and 0.8, respectively.

Key words: Ammonia, mass transfer, NHSW-based solution, ammonia removal efficiency

INTRODUCTION

Ammonia is a major pollutant that comes from household and industrial waste. Wastewater containing ammonia can be derived from the fertilizer industry, coal gasification, petroleum refining, pharmaceutical and catalyst plant (Ashrafizadeh and Khorasani, 2010). Ammonia from industrial wastewater should be removed so that its content does not exceed 5 ppm due to the nature of ammonia, which is very toxic to all species of fish and is oxidized by microorganisms through a process of nitrification producing nitrate compounds, which are highly undesirable by humans. The concentration of ammonia in industrial wastewater varies from 5-1000 mg L^{-1} (Rezakazemi *et al*., 2012; Kartohardjono *et al*., 2009) and therefore, removal of dissolved ammonia from wastewater becomes a necessity to protect the environment and human health.

Ammonia removal is very important in reusing wastewaters in industry. The applicability of ammonia removal processes generally depends upon several factors such as contamination level, plant safety and regulatory consideration and availability of a heating source and chemicals (Xie *et al*., 2009). Conventionally ammonia was removed from wastewater through air stripping, selective ion exchange, chlorination and biological nitrification (Hasanoglu *et al*., 2010), but these methods are dependent on relatively large amount of energy for the operation

(Bonmatı and Flotats, 2003). There are several unconventional methods to remove ammonia from wastewater such as contact glow discharge electrolysis (Saksono *et al*., 2013) and membrane-based separation methods. Membrane-based separation methods include vacuum degassing and sweep gas through membrane contactor (Mandowara and Bhattacharya, 2009), membrane distillation (Ding *et al*., 2006; Xie *et al*., 2009), vacuum membrane distillation (Ding *et al*., 2006; El-Bourawi *et al*., 2007), combination of membrane and ozonation (Kartohardjono *et al*., 2012a) and combination of membrane, ozonation and nonthermal plasma (Kartohardjono *et al*., 2012b) methods. In this study, we will remove ammonia from wastewater through hollow fiber membrane contactor using sulfuric acid solution and Natural Hot Spring Water (NHSW) based solution as absorbents. The aim of the study is to investigate the effects of these absorbents in absorbing ammonia through hollow fiber membrane contactor. In this study we will examine the effects of pH of absorbent solutions and initial concentration of ammonia in wastewater on ammonia removal efficiencies and overall mass transfer coefficients.

MATERIALS AND METHODS

Polypropylene hollow fiber membrane contactor was used in this study having surface area of 1.4 m^2 (based on onside fiber diameter) and provided by Liqui-Cell Company (1.7×8.75 Mini Module®). Schematic diagram of experimental setup is shown in Fig. 1. The wastewater was prepared by dissolving a given amount of ammonium sulfate into distilled water and will be used as wastewater solution in the experiment. Sodium hydroxide solution was added to the wastewater to adjust the pH of wastewater solution. Meanwhile, the absorbent solutions used in the experiment were sulfuric acid and NHSW-based solutions. Wastewater solution flowed through the shell side of the membrane contactor, whilst the absorbent solution flowed through the lumen fiber using a peristaltic pump Longer WT600-2J. Wastewater was analyzed every 15 min to measure the concentration of ammonia in the wastewater using ammonia meter Martini Instruments Mi 4500.

The ammonia removal efficiency is calculated through Eq. 1:

$$R\ (\%) = \frac{C_o - C_t}{C_o} \times 100 \tag{1}$$

where, C_0 is the ammonia concentration in the feed solution (mg L^{-1}) at initial condition and C_t is the ammonia concentration at time t. Furthermore, the overall mass transfer coefficient, K_{OV}, for the ammonia removal process, can be determined using the Eq. 2 (Ahmed and Semmens, 1992):

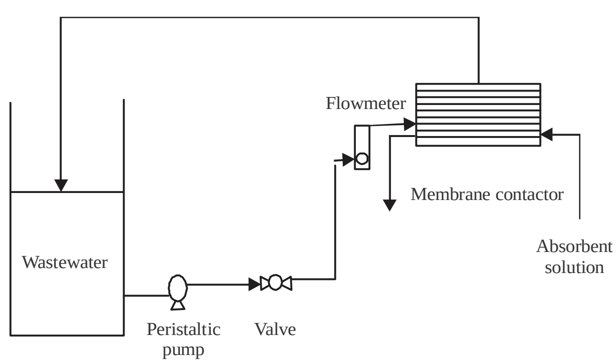

Fig. 1: Schematic diagram of experimental setup

$$\ln\left(\frac{C_t}{C_o}\right) = \frac{Q}{V}\left[\exp\left(-\left(\frac{K_{0V}aL}{v_L}\right)-1\right)\right]t \tag{2}$$

where, Q and V are wastewater circulation rate and volume of wastewater in the reservoir, respectively. Meanwhile, a, L and v_L are membrane specific area, fiber length and wastewater velocity in the membrane contactor, respectively.

RESULTS AND DISCUSSION

Figure 2a shows the ammonia removal efficiency during wastewater circulation at initial ammonia concentration in wastewater of 800 ppm, pH of wastewater of 10, 11 and 12 and pH of both NHSW and sulfuric acid solutions according to original pH of NHSW from its source i.e., 2.3. The maximum removal efficiency achieved in this experiment cannot reach 100% (max. ≈ 81%) as the acidity level of absorbent solutions are considered low. Figure 2a illustrated that ammonia removal efficiency through membrane contactor using NHSW solution is relatively higher than sulfuric acid solution at the same pH of wastewater solutions. The higher ammonia removal efficiencies could be due to various monoprotics acid present in the NHSW such as HCl, HNO_3 and HI (Kartohardjono *et al*., 2012a). These monoprotic acids are dissociated more easily than diprotic acid (H_2SO_4) in aqueous solution as there is only experience one step of ionization. Therefore, more anions are produced in monoprotic acid that can combine with ammonia to form ammonium salts, which lead to high ammonia removal efficiencies.

To increase the removal efficiency, the acidity level of absorbent solutions were increase so the pH of solutions around 0.8, which is almost equivalent to 0.1 M H_2SO_4 solution. To achieve this pH for NHSW absorbent, we added concentrated H_2SO_4 solution to the absorbent until the desired pH. Figure 2b shows the ammonia removal efficiency during wastewater circulation at pH of wastewater of 10, 11 and 12 and pH of both NHSW-based and sulfuric acid solutions 0.8. The maximum removal efficiency was achieved using pH of wastewater 12 at 105 and 120 min of circulation for NHSW-based and sulfuric acid solution, respectively. This indicates that NHSW-based solution shows better performance than sulfuric acid solution. Figure 2b also shows

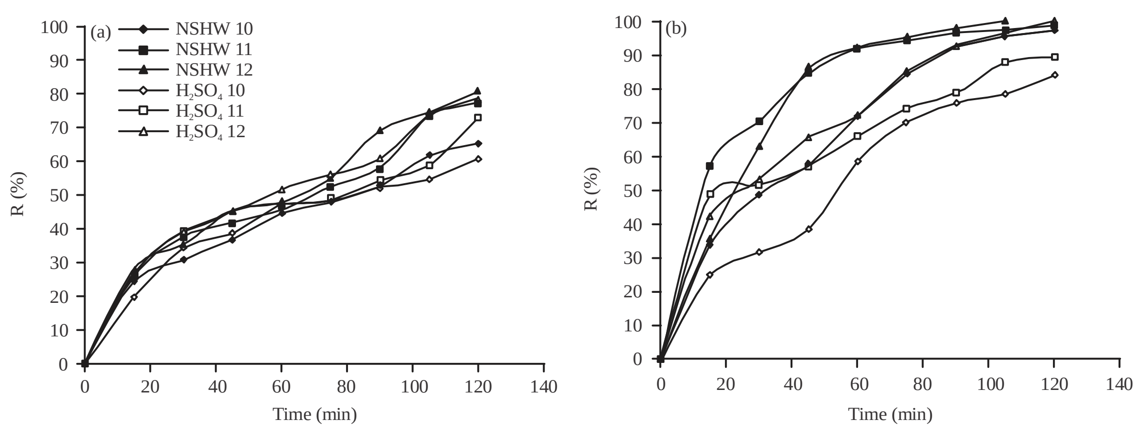

Fig. 2(a-b): Variation of ammonia removal efficiencies, R, with wastewater circulation time, at initial ammonia concentration of 800 ppm, various pH of wastewater (10, 11 and 12) and pH of absorbent, (a) 2.3 and (b) 0.8

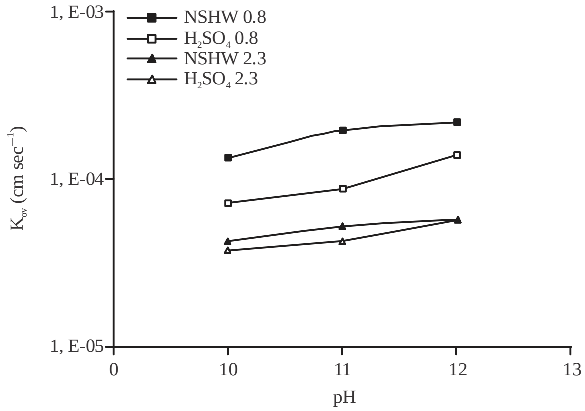

Fig. 3: Variation of overall mass transfer coefficients, K_{OV}, with pH of wastewater, at ammonia initial concentration of 800 ppm and pH of absorbent 2.3 and 0.8

Table 1: Conductivity of absorbent solutions

Solution	Conductivity (μS)
NHSW 2.3	17.28
H_2SO_4 2.3	5.73
NHSW 0.8	63.21
H_2SO_4 0.8	18.86

that the ammonia removal efficiencies of NHSW-based solution are relatively higher than sulfuric acid solution at the same pH of wastewater solution due to more monoprotics acid present in the NHSW-based solution. To prove further we measured the conductivity of the solution as shown in Table 1. Table 1 shows that both NHSW solutions at pH 2.3 and 0.8 have higher conductivity than sulfuric acid solution at the same pH, indicating that there are more anions in NHSW-based solutions.

Ammonia transfer through the membrane fiber in the contactor can be explained by means of a resistances-in-series: Resistance in the shell side, in the membrane and in the lumen side of the fiber (Hasanoglu *et al*., 2010). The overall mass transfer coefficient, K_{OV}, in the hollow fiber membrane contactor can be expressed as follow:

$$K_{OV} = \frac{1}{k_s} + \frac{1}{k_m} + \frac{1}{k_L} \tag{3}$$

where, in the experiments conducted in this study K_{OV}, k_s, k_m and k_L are the overall, wastewater side, membrane and absorbent side mass transfer coefficients, respectively. Therefore, based on the experiments the transfer of ammonia followed sequence of steps: transfer from the bulk of the wastewater solution through the boundary layer to the wastewater-membrane interface, diffusion through the gas-filled membrane pores to the absorption solution interface, reaction on the interface and diffusion of the ammonium salt into the absorption solution. Figure 3 shows the overall mass transfer coefficients as a function of pH of wastewater for both NHSW-based and sulfuric acid solutions at pH of 2.3 and 0.8 and ammonia initial concentration of 800 ppm. The overall mass transfer coefficients of NHSW-based solutions are relatively higher than sulfuric acid solution, as shown in Fig. 3, due to more anions formed in the NHSW that can react with

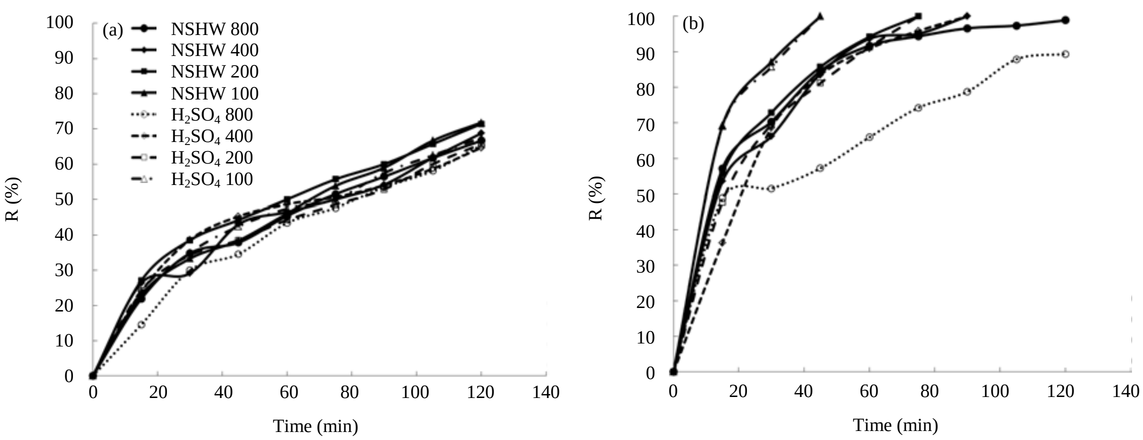

Fig. 4(a-b): Variation of ammonia removal efficiencies, R (%), with wastewater circulation time, t, at various ammonia initial concentrations (100, 200, 400 and 800 ppm), wastewater's pH of 11 and pH of absorbent, (a) 2.3 and (b) 0.8

ammonia on the interface of absorbent solution. Figure 3 also shows that increasing acidity level of absorbent will increase mass transfer coefficient at the same pH of wastewater.

The effects of ammonia initial concentration in wastewater on ammonia removal efficiency is shown in Fig. 4a and b for pH of wastewater 11 and pH of absorbents 2.3 and 0.8, respectively. As can be seen from Fig. 4a, ammonia removal efficiency tends to slightly decrease with the increasing ammonia initial concentration in wastewater for both NHSW-based and sulfuric acid solutions. This reveals that types of absorbents and ammonia initial concentration had a negligible effect on ammonia removal for the pH of absorbents of 2.3. The maximum removal efficiency achieved for the pH of absorbents of 2.3 cannot reach 100% (max. ≈ 72%) due to the acidity level of absorbent solutions. Meanwhile, for the pH of absorbents of 0.8, the ammonia removal efficiency tends to increase with the decreasing ammonia initial concentration in the wastewater as shown in Fig. 4b. Complete ammonia removal were achieved at 45, 75, 90 and 120 min of wastewater circulation for ammonia initial concentration of 100, 200, 400 and 800 ppm, respectively for both NHSW-based and sulfuric acid solutions. Types of absorbent had a negligible effect on ammonia removal efficiency at ammonia initial concentration of 100, 200 and 400 ppm for absorbents' pH of 0.8. Meanwhile, at ammonia initial concentration of 800, the NHSW-based solution exhibits higher ammonia removal efficiency than sulfuric acid solution due to more monoprotics acid present in the NHSW-based solution.

The effects absorbents and ammonia initial concentration on overall mass transfer coefficient is shown in Fig. 5. The overall mass transfer coefficients are relatively constant with the increasing ammonia initial concentration and tend to slightly higher for NHSW-based solutions compare to sulfuric acid solution for absorbents' pH of 2.3. These facts reveal that the types of absorbent and the ammonia initial concentration had a negligible effect on the overall mass transfer coefficient at the pH of absorbents of 2.3. Meanwhile, at the pH of NHSW-based solution of 0.8, the overall mass transfer coefficient decreases with the increasing ammonia initial concentration in the ranged of 100-200 ppm and tends to constant at ammonia initial concentration in the ranged of 200-800 ppm. This reveals that initial ammonia concentration had a negligible effect on overall

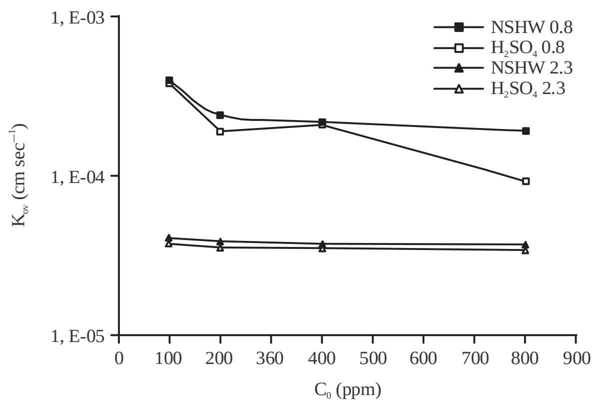

Fig. 5: Variation of overall mass transfer coefficients, K_{OV}, with ammonia initial concentration, C_0, at pH of wastewater of 11 and pH of absorbents 2.3 and 0.8

mass transfer coefficient in the ranged of 200-800 ppm for NHSW-based absorbent at pH of 0.8. Sulfuric acid solution had similar trend of the overall mass transfer coefficient to NHSW-based solution in the range of ammonia initial concentration of 100-400 ppm at the pH of absorbents of 0.8. However, the overall mass transfer coefficient of NHSW-based solution is higher than sulfuric acid solution at ammonia initial concentration of 800 ppm.

CONCLUSION

Experiments have been conducted to examine the effects of absorbents on ammonia removal efficiency and overall mass transfer in hollow fiber membrane contactor system. The pH of wastewater and absorbent solutions and ammonia initial concentration in wastewater were used as tested variables for evaluating effectiveness of the absorbents. Experimental results showed that, at the ammonia initial concentration of 800 ppm, NHSW-based solution had higher ammonia removal efficiency and overall mass transfer coefficient than sulfuric acid solution in the range of pH of wastewater applied in the system. The types of absorbent had a negligible effect on the ammonia removal efficiency and the overall mass transfer coefficient in the range of ammonia initial concentration applied in the experiments at the pH of absorbents of 2.3. Meanwhile, at the pH of absorbents of 0.8, the types of absorbent had a negligible effect on ammonia removal efficiency and overall mass transfer coefficient only in the range of ammonia initial concentration of 100-400 ppm.

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