IOP Conf. Series: Earth and Environmental Science **105** (2018) 012123 doi:10.1088/1755-1315/105/1/012123

Utilization of super hydrophobic membrane contactor for NOx Absorption

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Abstract. NOx is produced from the reaction between nitrogen, oxygen and even hydrocarbons (during combustion), especially at high temperatures. Nitrogen oxide compounds are one of the harmful emissions that can result from fuel combustion. This research aims to find out the performance of super hydrophobic membrane contactors in the absorption process of N_2O from its mixture with air using acid solution and hydrogen peroxide. N_2O gas must be reduced from the exhaust gas especially to meet the regulations applicable to the environment due to the hazardous nature of the N_2O gases. The experimental results showed that the amount of NOx absorbed and the absorption efficiency increased with increasing absorbent flow rate in the membrane contactor. Meanwhile, the concentration of NOX in the outlet gas decreased with increasing absorbent flow rate.

1. Introduction

NOx is a term derived from oxide mono-nitrogen NO and NO_2 (nitric oxide and nitrogen dioxide) [1]. The amount of nitrogen oxide emitted into the atmosphere as air pollution can be significant in areas where vehicles are heavily congested, such as in big cities. NOx gas is formed whenever there is combustion in the presence of nitrogen - as in a vehicle engine, and can also be produced naturally by lightning [2]. The reduction of NOx in the exhaust gases such as those from boilers and the nitric acid industry is currently attracting much attention due to increasingly stringent environmental regulations. For this purpose, both dry and wet processing techniques are widely used and studied. In the case of the wet process, a major obstacle to absorption is the very low solubility of nitric oxide. To make the gas more soluble is by oxidizing NO to NO_2 in the gas phase with the help of ozone or chlorine dioxide. Oxidation may also be carried out in a washing liquid by adding an oxidizing agent such as potassium permanganate, sodium chlorite or sodium hypochlorite [3].

The gas absorption into the liquid can be carried out in the gas-liquid contactor on the hollow fiber membrane module. In recent years membrane contactors have attracted the attention of many researchers as an alternative technology for gas absorption process because it can overcome the weaknesses of conventional methods [4]. Hollow fiber membrane contactors have been widely used as gas-liquid contactors as they have large ratio between surface area for gas-liquid contact and volume of equipment [5]. In addition, gas absorption through membrane contactor also integrates separation and absorption processes to exploit the benefits of both processes [6]. The type of solvent and the membrane material is essential to maximize the efficiency of the process, wherein the membrane must be maintained so as not to be wetted by the solvent used, so that the membrane material used must be hydrophobic [7]. Based on research conducted by some previous researchers for gas absorption through membrane contactors, the hydrophobic property of the membrane is not enough. This membrane is still experiencing wetting at a certain period so that the mass transfer coefficient that occurs drastically due to membrane wetting [8, 9]. It is necessary that the membrane material is very resistant to not wetted by the absorbent solution for long periods, so that the advantage of gas absorption process through the membrane can be maximally exploited. The choice of membrane material is critical to the efficiency of the process [10]. To improve the hydrophobicity, The membrane can be incorporated with other material such as graphene nano sheets [11] or prepared as super-hydrophobic membrane [12]. This study aims to evaluate the performance super hydrophobic membrane contactor to absorb NOx, which will be represented by N_2O, using a mixture of nitric acid and hydrogen peroxide as absorbent.

2. Materials and Methods

The schematic diagram of N_2O absorption from its mixture with air using a membrane contactor is shown in Figure 1. The super hydrophobic hollow fiber membrane contactors used were supplied by PT GDP Filter Bandung. The feed gas (N_2O = 15000 ppm), and HNO_3 and H_2O were purchased from BOC Gases and Merck, respectively. The hollow fiber membrane contactors used sized of 6 cm and 34 cm in diameter and length, respectively, which consists of 2000 fibers. The fibers are polypropylene-based sized of 525 and 235 µm in outer and inner diameter, respectively. During the experiment, the absorbent (a mixture of HNO_3 and H_2O_2 1:1) was pumped through the lumen side of the membrane fibers, whilst the feed gas was sent to the shell side of the membrane contactor. The inlet and outlet gas compositions to and from membrane contactor were analyzed using gas chromatography Bruker Scion 436-GC. Meanwhile, the inlet and outlet gas flow rates to and from membrane contactor was measured using mass flow meter Sierra Top Trak Instruments. The amount of N_2O absorbed, N_2O_{abs}, by the absorbent and the absorption efficiency, $\%R$, from the experiment were calculated using equation (1) and equation (2), respectively.

$$N_2O_{abs} = (x_iF_i - x_oF_o)\frac{P}{RT} \quad (1)$$

$$\%R = \frac{N_2O_{Abs}}{x_iF_i} x100\% \quad (2)$$

where the x_i and F_i and x_o and F_o are N_2O concentration and flow rate in inlet and outlet membrane contactor, respectively. P, T and R, are pressure, temperature and gas constant, respectively. Meanwhile, Q_G and A_m are feed gas flow rate and membrane fibers surface area, respectively.

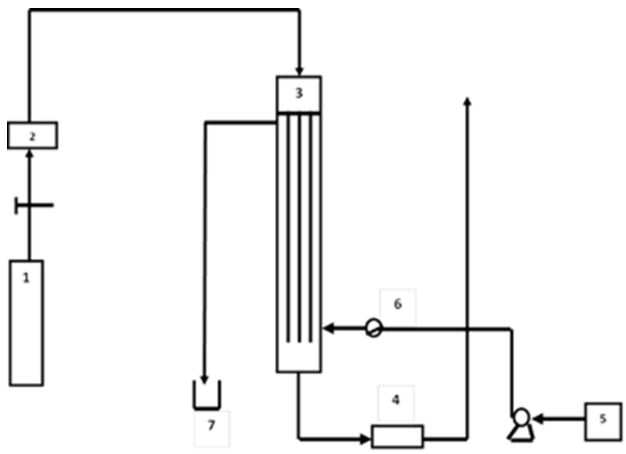

Figure 1. Schematic diagram of experimental set up:
1. Feed gas;
2. Mass flow meter;
3. Superhydrophobic hollow fiber membrane contactor;
4. Mass flow meter;
5. Absorbent reservoir;
6. Liquid flow meter

3.**Results and Discussion**

The transport mechanism of N_2O from the bulk gas in the feed side through the absorbent in the membrane contactor based on the experiment is following three steps:

- Firstly, the N_2O from the bulk gas is transferred to the fiber membrane surface in the gas phase
- The N_2O is then diffused to the other side of the fiber membrane surface in the liquid phase
- Finally, the N_2O is absorbed by the absorbent in the liquid phase through oxidation by H_2O_2 in the presence of HNO_3.

The amount of N_2O absorbed by the absorbent in the membrane contactor as a function of absorbent flow rate at various feed gas flow rates is shown in Figure 2. The N_2O absorbed increased with increasing absorbent flow rate due to more turbulence of the absorbent at higher flow rate. The turbulence in the liquid phase will reduce the resistance to the mass transfer in the absorbent side, which will lead to the increase in the amount of N_2O absorbed. Meanwhile, the increase in feed gas flow rate will increase the turbulence in the gas phase, that will reduce the mass transfer resistance in the gas phase, which also lead to the increase in the amount of N_2O absorbed.

The N_2O absorption efficiency increased with increasing the absorbent flow rate due to the increase in the amount of N_2O absorbed as shown in Figure 3. The absorption efficiency also increased with the increasing gas flow rate especially at the lower absorbent flow rate. The absorption efficiency at the higher absorbent flow rates (400 and 500 cc/min) are almost similar for feed gas flow rates of 100cc/min and 160cc/min, respectively. This is due to the N_2O absorption increment at absorbent flow rates of 400 cc/min and 500cc/min for feed gas flow rate of 160 cc/min was insignificant, so that the absorption efficiency almost constant. Meanwhile, the absorption efficiency for feed gas flow rate of 100cc/min increased in the range of absorbent flow rate observed in this study.

The N_2O concentration at outlet gas from the membrane contactor decreased with increasing the absorbent and the feed gas flow rates as shown in Figure 4. The increase in the absorbent flow rate will increase the amount of N_2O absorbed as shown in Figure 2, which lead to the decrease in the N_2O concentration at the outlet membrane contactor.

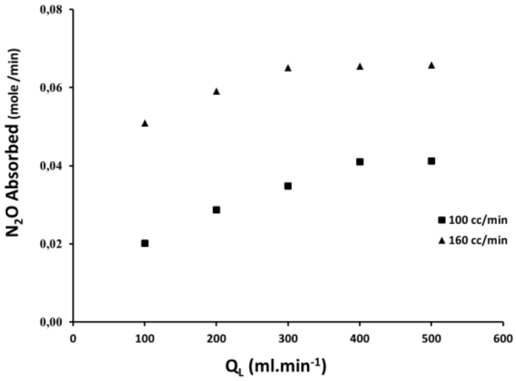

Figure 2. The amount of N_2O absorbed as a function of the absorbent flow rate, Q_L, at feed gas flow rates, Q_G, of 100 cc/min and 160 cc/min

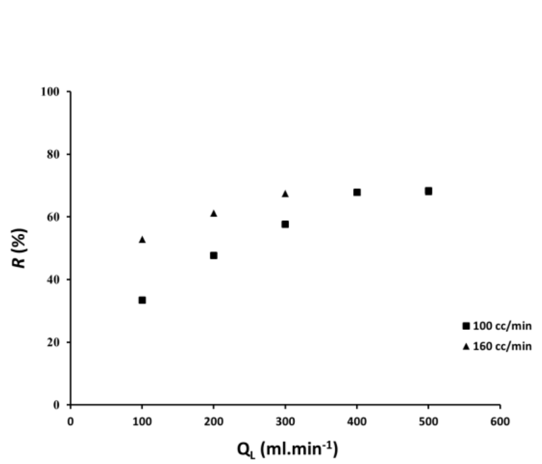

Figure 3. The N_2O absorption efficiency, %*R*, as a function of the absorbent flow rate, Q_L, at feed gas flow rates, Q_G, of 100 cc/min and 160 cc/min

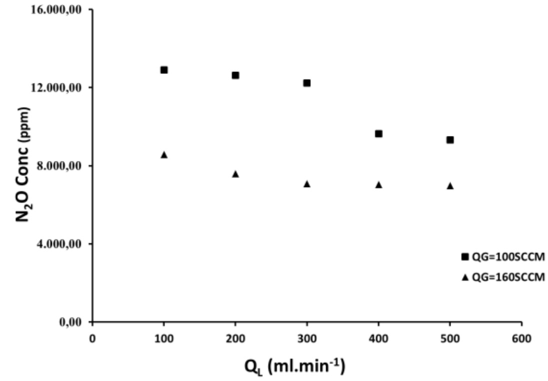

Figure 4. The N_2O concentration at the out let gas from the membrane contactor as a function of the absorbent flow rate, Q_L, at feed gas flow rates, Q_G, of 100 cc/min and 160 cc/min

4. Conclusion

Experiments have been conducted to see the effect of the absorbent and the feed gas flow rates on the N_2O absorbed, the N_2O absorption efficiency and the N_2O concentration at the outlet gas from the membrane contactor. The experimental results showed that the N_2O absorbed and the N_2O absorption efficiency increased with increasing the absorbent and the feed gas flow rates. Meanwhile, the N_2O concentration decreased with increasing the absorbent and the feed gas flow rates.

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