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ABSORPTION OF NO_x IN PACKED COLUMN (II)
—COMPARISON OF EXPERIMENTAL WITH THEORETICAL—

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Abstract—An absorption efficiency of packed column removing nitrogen oxides with water and NaOH solution under atmospheric pressure was studied. The efficiency and the acidity produced by absorption of NO_x were measured in a packed column. The model developed that was based on the mass-transfer information for packed column and absorption mechanism accompanying the chemical reaction was compared with experimental results. Predictions using the model presented by the previous paper (part I) was shown well to agree with from the experimental results (part II). The efficiency of NO_x absorption is largely dependent on the height of packing material and the partial pressure of NO_x in the feed gas. The efficiency of NO_x absorption decreases with the increase of the acidity produced by recycling of water as a scrubber liquid. For the recycle mode with an aqueous NaOH solution as a scrubber liquid, NO_x absorption efficiency is shown to be constant until all of the C_{OH^-} in the scrubber liquid are converted into C_{H^+}.

INTRODUCTION

An experimental study of the absorption of gaseous NO_x into water and dilute nitric acid and aqueous solution of NaOH in the packed column was carried out. The typical NO_x absorber was designed to use a water flowing in a single pass through the column for the HNO_3 industry production. There are many kinds of absorber such as packed column, bubble column and sieve plate column in the commercial scale NO_x scrubber. For this study, the packed column was selected as a NO_x absorber because it is simple and suitably resistant to corrosion. Experiments were conducted to evaluate the aqueous absorption of NO_x at atmospheric pressure and 293 K in the column packed with 9 mm ceramic Raschig rings. For varying the feed NO_x concentration in the feed gas, an air-NO_x mixture gas flow rates, the liquid flow rates and the heights of packing material, a NO_x absorption efficiency in packed column was measured and analyzed. These experiments were carried out by batch and recycle modes of the scrubber liquid. NO_x absorption efficiencies with the concentration of the scrubber liquid by increasing the recycle time were measured in recycle mode.

EXPERIMENTAL APPARATUS AND PROCEDURE

A flowsheet of the experimental system is illustrated in Fig. 1. The system consists of the scrubber, liquid supply, gas supply and gas-liquid sampling parts. The NO_x scrubber was a packed column constructed with Pyrex glass of inside diameter of 0.075 m. The column was packed with ceramic Raschig rings of 0.0095 m length, and the heights of packing material of 0.3, 0.5, 0.7 and 0.9 m, respectively. The scrubber liquid was pumped up to the top of the column from the liquid supply tank on a batch or recycle mode. This stream was metered by rotameter to the column in use. The liquid was distributed approximately 0.1 m above the top of the packing material. The feed gas was entered into the bottom of the packing material through the packing material support after mixing with air in the holding tank. The input NO_x concentration was varied from 2,000 to 12,000 ppm. Both feed and effluent liquid streams with respect to the packed column were manually sampled. The process air was metered along with the NO_x gas by a rotameter. The gas analysis for NO/NO_x could be done by passing a gas stream through NO_x

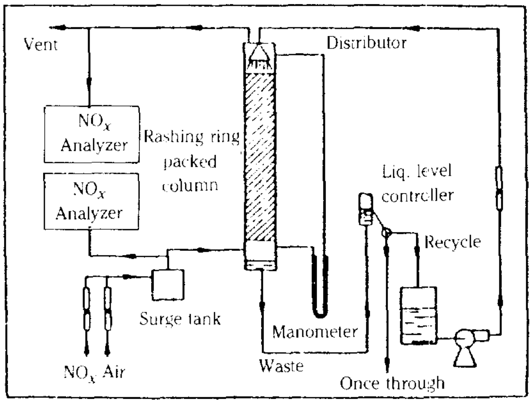

Fig. 1. Flowsheet of NO_x absorption experimental system.

analyser (model: DY-206 and Beckman NO/NO_x analyzer). The packed column was operated slightly above atmospheric pressure.

The system was allowed to reach the steady state conditions that NO_x concentration and acid concentration in the feed/effluent streams were constantly maintained before samples were taken. All liquid entering into and leaving the experimental column were sampled and analyzed for their aqueous reaction products of HNO_3 and HNO_2. Each sample from the effluent liquid stream was analyzed immediately for later analysis of total nitrous acid concentration. The nitric acid was analyzed by NaOH titration. In order to analyze C_{HNO_2} with Saltzman method [1], 1-5 ml of the sample diluted with a distillated water was put into the 25 ml volumetric flask and then 20 ml of Saltzman solution was added. The distillated water was used as a make-up solution. After 40 minutes the sample was analyzed by spectrometer (Beckman-spectra 20, wave length 550 μm). The absorption efficiencies of packed column were calculated from NO_x gas concentration differences between influent and effluent. The effluent gas stream was discharged through the fume hood in which a constant air flow is maintained.

The experiment was carried out with a batch and recycle mode of the scrubber liquid. In the batch mode, water is used as a scrubber liquid to remove NO_x gas in packed column. NO_x absorption in the recycle mode was completed with water and NaOH solution, respectively. NO_x absorption efficiency and the concentration of the scrubber liquid were measured with the increase of recycle time. In the recycle mode, the total volume of the scrubber liquid in the NO_x gas absorption system was 1.5l.

Table 1. The experimental conditions for NO_x scrubbing experiment

feed gas temperature	293 K
inlet liquid temperature	283 K
scrubber liquid flow raten	0.04-0.1 m^3/hr
feed gas flow rate	2-4 m^3/hr
NO_x gas concentration	2,000-12,000 ppm
height of packing material	0.3-0.9 m

RESULTS

The final evaluation of the model which was developed in the previous paper (part 1) [2] lies in its comparison with the experimental data. Experimental results using the apparatus previously described were obtained under the experimental conditions in Table 1. These conditions were subject to variation for the purpose of the study.

NO_x absorption efficiency in packed column is defined as [3]

$$X_{NO_x}=\frac{1-(P_{NO_x})_{out}/(P_{NO_x})_{in}}{1+\in(P_{NO_x})_{in}/(P_{NO_x})_{out}} \quad (1)$$

If NO_x concentration is low, $\in(P_{NO_x})_{in}/(P_{NO_x})_{out}$ is lower than unity. Therefore, X_{NO_x} is represented as follows;

$$X_{NO_x}=1-\frac{(P_{NO_x})_{out}}{(P_{NO_x})_{in}} \quad (2)$$

The experimental data obtained with the 0.075 m ID column packed with 9.5 mm ceramic Raschig rings. The experimental NO_x absorption efficiency is compared with the predicted gaseous NO_x absorption efficiency from the previous paper [2]. The absorption efficiencies predicted by the model are very close to the experimentally determined efficiencies. The NO_x absorption efficiencies predicted by the model are then compared with those experimentally determined for the variation of scrubber liquid flow rates, feed gas flow rates, NO_x concentration in feed gas, and the heights of packing material at references conditions in Fig. 2-5. In most cases, the model used to predict NO_x removal efficiencies in the packed column is conservative with respect to absorption efficiencies. Increasing the flow rate of the scrubber liquid results in a higher overall NO_x absorption efficiency, X_{NO_x} as shown in Fig. 2 due to the increase of the gas-liquid interfacial area and a liquid-phase mass-transfer coefficient with the liquid flow rate. The higher gas flow rate results in

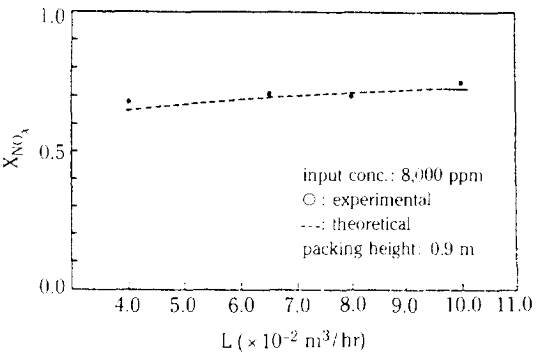

Fig. 2. NO_x absorption efficiency for various liquid flow rates at 2.0 m^3/hr gas flow rate.

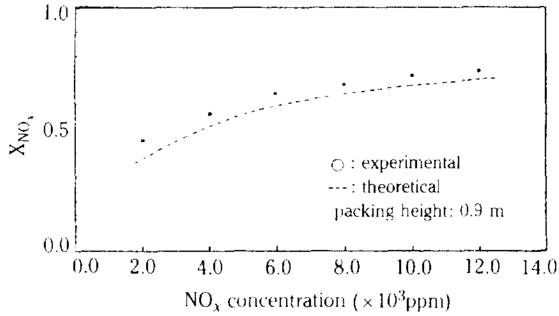

Fig. 4. NO_x absorption efficiency for concentration of NO_x entering the packed column at 2.6 m^3/hr gas flow and 0.065 m^3/hr liquid flow rates.

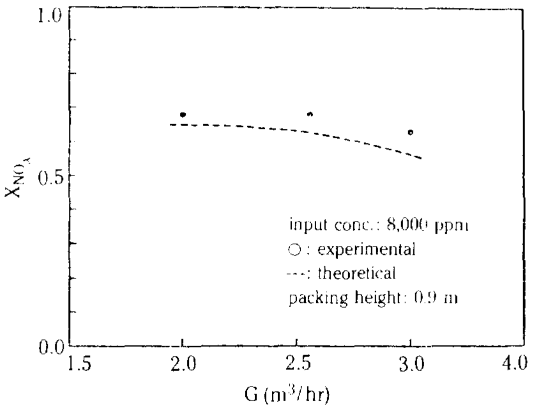

Fig. 3. NO_x absorption efficiency for various gas flow rates at 4.0 × $10^{-2}m^3$/hr liquid flow rate.

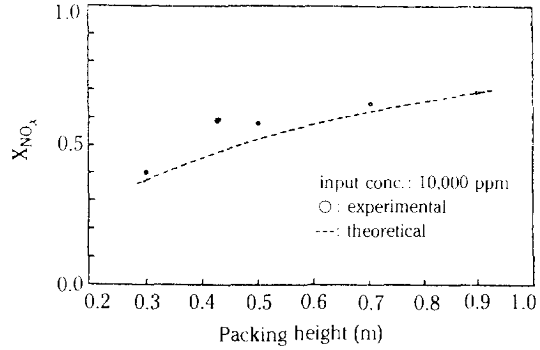

Fig. 5. NO_x absorption efficiency for the various packing height at 2.0 m^3/hr gas flow and 0.04 m^3/hr liquid flow rates.

a slight decrease of NO_x absorption efficiency as shown in Fig. 3, due to a reduced gas residence time for absorption process and NO oxidation. The results for the variation of NO_x partial pressures in the gas feed and height of packing material are shown in Fig. 4 and 5, respectively. The increased NO_x gas concentration is consistent with increased absorption rates of NO_2^* and oxidation of NO due to higher driving force concentrations of NO_2^* and NO, respectively. The above results have a same tendency with that of the previous studies of NO_x absorption by a sieve plate column [4,5]. From Fig. 3,4 and 5 the experimental results are higher than the NO_x absorption efficiencies predicted by model because this model was not considered the absorption of N_2O_3 into water. If N_2O_3 absorption is considered in NO-NO_2 mixture system, the theoretical values by model will be a little higher. Also, the increase of the height of packing material results in a higher NO_x absorption efficiency.

The effects of NO_x absorption efficiency for recycle mode of scrubber liquid with 0.9 m height of packing material at 8,000ppm of NO_x in the feed gas are shown in Fig. 6 and 7. As shown in Fig. 6, the NO_x absorption efficiency for the increase of the recycle time of scrubber solution through the packed column decreases exponentially. After about 2 hours in the recycle mode

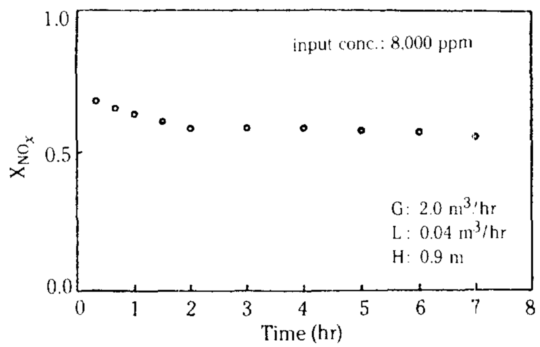

Fig. 6. NO_x absorption efficiency for recycling time of scrubbing solution.

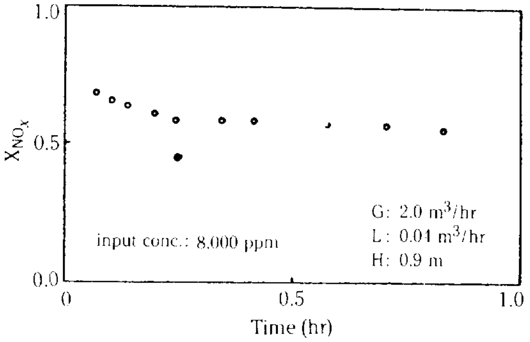

Fig. 7. NO_x absorption efficiency for C_{H^+} in the recycled scrubbing solution.

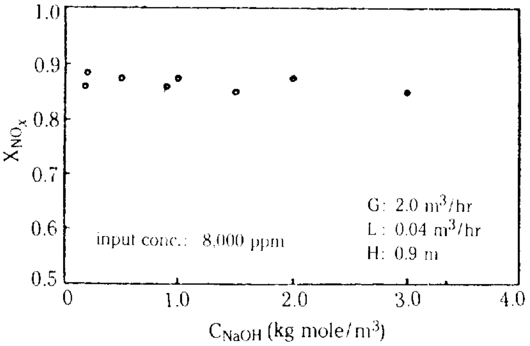

Fig. 8. NO_x absorption efficiency for the concentration of NaOH scrubbing solution.

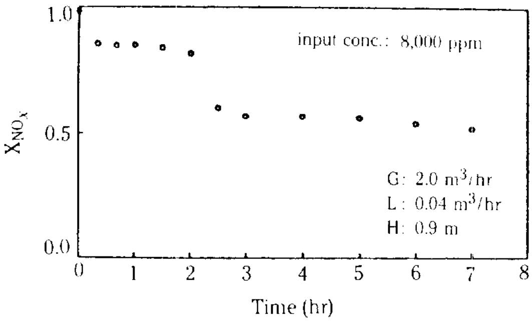

Fig. 9. NO_x absorption efficiency for recycling time of NaOH scrubbing solution.

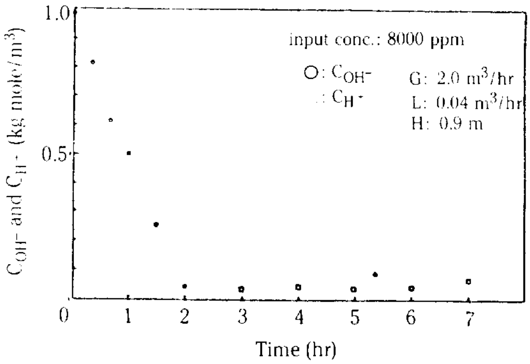

Fig. 10. The disappearance of C_{OH^-} and appearance of C_{H^+} for recycling time of NaOH scrubbing solution.

the absorption efficiency becomes nearly a constant. In Fig. 7, the absorption efficiency exponentially decreases with the increase of the acidity produced by recycle of scrubber solution. From these results NO_x absorption for recycle mode may be dependent on the acid concentration in the scrubber solution.

In the case of NO_x absorption using aqueous solution of NaOH as a scrubber solution, NO_x absorption efficiency for NaOH concentration in liquid phase is shown in Fig. 8. NO_x absorption efficiency in the 0.9 m packed column is shown to be nearly constant for the various of NaOH concentration. This result points out that NO_x absorption efficiency is scarcely affected by the concentration of NaOH. The absorption of NO_2^* into NaOH solutions may be described as following equation [6].

$$2NaOH + N_2O_4 \rightarrow NaNO_2 + NaNO_3 + H_2O \quad (3)$$

As shown in the above equation, HNO_2 is not produced. For using water as a scrubber liquid the decomposition of HNO_2 is occurred in the liquid phase and NO gas is generated. Therefore, NO_x absorption efficiency of aqueous NaOH solution shows to be higher than that of water as a scrubber liquid.

The results of NO_x absorption efficiency and the appearance of C_{H^-} and disappearance of C_{OH^-} in the scrubber solution by recycle of 1 mole NaOH scrubber solution (1.5 *l*) at 8,000 ppm of NO_x in the feed gas are shown in Fig. 9 and 10. With increasing recycle time the concentration of NaOH in the scrubber solution is sharply decrease but the absorption efficiency in packed column shows constant until 2 hours at recycle mode of the scrubber solution. After about 2 hours operation all of the C_{OH^-} in the scrubber solution was converted into C_{H^+} due to absorption of NO_x as shown in Fig. 10, and NO_x absorption efficiency decrease with the operation time and with the increase of C_{H^+} due to absorption of NO_x as shown in Fig. 10, and NO_x absorption efficiency decrease with the operation time and with the increase of C_{H^+} with recycle of the scrubber solution.

CONCLUSION

1. By moderate heights of packing material it can be achieved fairly high nitrogen oxides absorption efficiencies at similar conditions to those studied.

2. NO_x absorption efficiency in recycle of water and dilute acid decreases exponentially with the operation time.

3. The NO_x absorption efficiency in the packed column varies directly as some function of the liquid flow rate, the concentration of NO_2^* and the height of packing material. However, the higher gas flow rate results in decrease in the absorption efficiency.

4. The NO_x absorption efficiency with NaOH solution would be independent on the concentration of NaOH in the scrubber solution.

NOMENCLATURE

C_j : liquid-phase concentration of component j [kg-mol/m^3]
G : gas flow rate [m^3/s]
L : liquid flow rate [m^3/s]
NO_2^* : $NO_2 + 2\ N_2O_4$
NO_x : $NO + NO_2^*$
P_j : partial pressure of gas component j [atm]
X_{NO_x} : the efficiency of NO_x absorption in the packed column
ϵ : fractional change in volume of the gas due to absorption

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