

## Simultaneous removal of SO<sub>2</sub>, NO and particulate by pilot-scale scrubber system

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**Abstract**—SO<sub>x</sub> and NO<sub>x</sub> have both previously been identified as primary precursors of acid rain, and thus the abatement of SO<sub>x</sub> and NO<sub>x</sub> emissions constitutes a major target in the field of air pollution control. In this study, the efficacy of a pilot-scale scrubber was evaluated with regard to the simultaneous removal of SO<sub>2</sub>, NO and particulate with wet catalysts. The removal efficiencies of particulate were measured to be 83, 92 and 97% with catalyst flux of 0.5, 0.8 and 1.5 L/min, respectively. The average removal efficiencies of particulate with different nozzles were approximately 94 and 90% with FF6.5 (5/8 in.) and 14 W (1.0 in.) nozzles, respectively. At least 96-98% of particulate and SO<sub>2</sub> were removed, regardless of the stage number of reactor. In a one-stage scrubber, 83.3% removal efficiency of NO was achieved after 48 hours; however, the two-stage scrubber achieved an NO removal efficiency of 95.7%. Regardless of the liquid-gas ratio, SO<sub>2</sub> and particulate were removed effectively, whereas NO was removed about 84% and 74% under liquid-gas ratio conditions of 39.32 L/m<sup>3</sup> and 27.52 L/m<sup>3</sup>, respectively. In experiments using STS and P.P. pall ring as packing material, particulate and SO<sub>2</sub> removal efficiency values in excess of 98% were achieved; however, NO removal was correlated with the different packing materials tested in this study. With the above optimum operation conditions, even after 20 hours, the removal efficiency for NO stayed at 95% or higher, the removal efficiency for SO<sub>2</sub> stayed at 97% or higher, and the removal efficiency for particulate stayed at 92% or higher. In accordance, then, with the above results, it appears that this process might be utilized in scrubber systems, as well as systems designed to simultaneously remove particulate, SO<sub>2</sub> and NO from flue gas.

Key words: Scrubber, SO<sub>2</sub>, NO, Particulate, Simultaneous Removal

### INTRODUCTION

Sulfur oxides (SO<sub>x</sub>) and nitrogen oxides (NO<sub>x</sub>) are both major causes of acid rain, and are both emitted from stationary combustion facilities such as power plants, incinerators and combustors. Recently, a number of research efforts have sought to develop a method for the removal of these oxides, in an attempt to protect the atmosphere against pollution [1,2].

Currently, the most commonly used techniques for the removal of SO<sub>x</sub> and NO<sub>x</sub> employ physicochemical processes. These processes, however, tend toward high operating costs and also generate chemical waste by-products which must be disposed of prior to discharge. The most effective technology for the removal of SO<sub>2</sub> is flue gas desulfurization (FGD). Commercial processes for SO<sub>2</sub> removal utilize limestone slurry as a scrubbing solution [2-4]. These wet FGD processes have been widely implemented, as a consequence of their generally low cost, ease of operation, and high SO<sub>x</sub> removal efficiency, in comparison to other processes [2-8]. Although the wet FGD system has been proven to efficiently remove a high degree of SO<sub>2</sub>, it does not remove NO<sub>x</sub> with a comparable efficiency. The reason for this is that NO is extremely insoluble in water [1].

Technologies utilized for the removal of NO<sub>x</sub> can be divided into two types: combustion control and post-combustion treatments. Post-combustion treatments include a variety of techniques, including selective non-catalytic reduction (SNCR), selective catalytic reduction (SCR), thermal De-NO<sub>x</sub>, scrubbing and several others [2]. Among these technologies, scrubbing methods are the most economically competitive, and also carry the advantage of simultaneously controlling other acid gases and particulates [2,9].

A number of wet and dry processes appropriate for NO<sub>x</sub> removal have been developed, and some of these wet scrubbing processes are capable of simultaneously removing SO<sub>x</sub> [10]. Despite the many successful commercial implementations of individual desulfurization and denitrification processes, a considerable amount of attention has recently been focused on the simultaneous removal of SO<sub>x</sub> and NO<sub>x</sub> within a single reactor, considering the capital investment, operating cost and space for equipment inherent to the processes [2]. A minor adjustment of the FGD system may prove sufficient for the successful operation of a simultaneous SO<sub>x</sub>/NO<sub>x</sub> removal system, and should also prove cost-effective. In general, additives must be added to a scrubbing system in order to effect the oxidation of the relatively water-insoluble NO to soluble NO<sub>2</sub>, which can then be removed via alkaline absorbents [1,2]. Therefore, in the development of efficient simultaneous SO<sub>x</sub>/NO<sub>x</sub> removal systems, chemical reagents, including FeSO<sub>4</sub>/H<sub>2</sub>SO<sub>4</sub>, FeSO<sub>4</sub>/Na<sub>2</sub>SO<sub>3</sub>, Fe(II)-EDTA,

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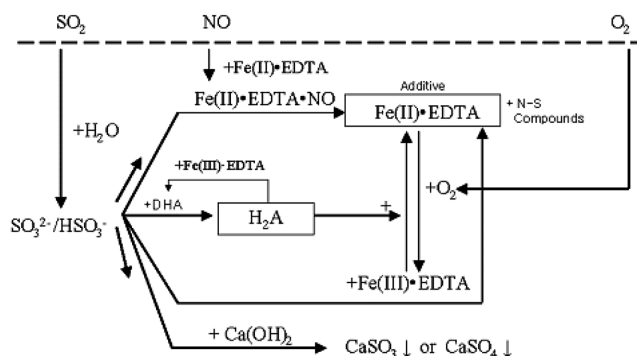


Fig. 1. Reaction mechanism of simultaneous removal of SO<sub>2</sub>/NO by ferrous chelate.

KMnO<sub>4</sub>/NaOH, NaClO<sub>2</sub>/NaOH, Na<sub>2</sub>S/NaOH, Na<sub>2</sub>S<sub>2</sub>O<sub>4</sub>/NaOH, H<sub>2</sub>O<sub>2</sub>, Na<sub>2</sub>SO<sub>3</sub>, CO(NH<sub>2</sub>)<sub>2</sub>, NaOH, Na<sub>2</sub>CO<sub>3</sub>, peracides and ferrous-chelating agents, have been added to the liquid phase in a number of experiments [1,2,9,11-14]. However, the wet scrubbing method carries the distinct disadvantage of inevitable waste generation. Considering this disadvantage, it appears that an aqueous solution of Fe(II)-chelate might prove to be a promising NO absorbent, due to the ease of regeneration associated with these solutions. Recently, it has been reported that an aqueous solution of Fe(II)-EDTA reacts rapidly with dissolved NO, and has a very substantial NO absorption capacity [10].

This study preceded following experiments that examined the reaction mechanism with an aqueous solution of Fe(II)-EDTA for simultaneous SO<sub>2</sub>/NO removal, as shown in Fig. 1 [15-17]. There has been, thus far, a relative paucity of attempts to determine the efficiency of a wet scrubbing system using Fe(II)-EDTA solution with some oxidizing chemicals, with regard to the ability of the system to simultaneously remove particulate, SO<sub>2</sub> and NO.

In this study, we have investigated the effects of a variety of parameters on particulate, SO<sub>2</sub> and NO removal efficiency using pilot-scale scrubber system employed a Fe(II)-EDTA solution with additives utilized to augment the oxidizing ability of the wet catalyst.

## MATERIALS AND METHODS

### 1. Chemicals

Reagent grade disodium ethylenediaminetetraacetic acid (EDTA-2Na), FeSO<sub>4</sub>·7H<sub>2</sub>O, ascorbic acid, adipic acid and sodium sulfite were used in this study. All other reagents were of analytical grade.

### 2. Preparation of Wet Catalyst

The wet catalyst was prepared as the protocols established by Hishinuma [18], via dissolution in distilled water at the following modified composition [17]: Fe(II)-EDTA 0.03 M, ascorbic acid 0.024 M, adipic acid 0.024 M, sodium sulfite 0.09 M. The initial pH 5 value of the wet catalyst was adjusted to the desired value via the

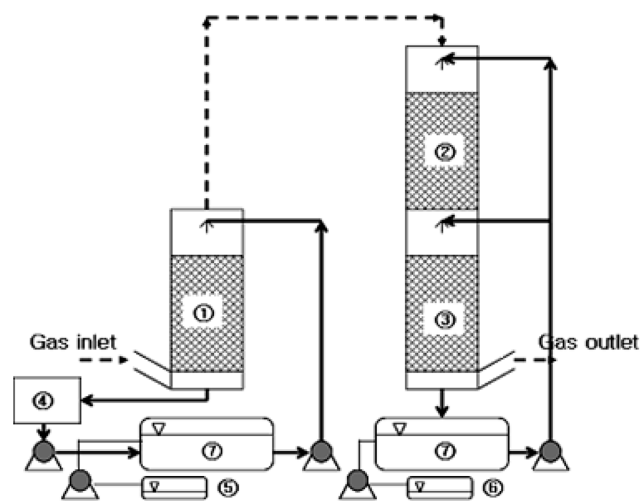


Fig. 2. Flow diagram of experimental apparatus system.

- ①, ②, ③ Reactor
- ④ Filter Press
- ⑤ Ca(OH)<sub>2</sub> solution bath
- ⑥ Additives bath
- ⑦ Tower sump

addition of either HCl or NaOH.

### 3. Experimental Set-Up

In order to remove the particulate, SO<sub>2</sub> and NO generated by a small-scale incinerator, we applied a pilot-scale scrubber system. The experimental system employed in this work included a scrubber, a filter, and a control box, as shown in Fig. 2.

#### 3-1. Flue Gas Simulation System

The flue gas simulation system was comprised of a pure SO<sub>2</sub> cylinder, a pure NO cylinder, a pure N<sub>2</sub> cylinder, a pure O<sub>2</sub> cylinder, a mass flow controller (F-201C, Bronkhorst) and a gas mixing system. The simulated flue gas was prepared to a constant concentration and flow rate via the mixing of pure SO<sub>2</sub>, NO, N<sub>2</sub> and O<sub>2</sub> gases, by using a gas mixing system. The flow rates of SO<sub>2</sub>, NO, N<sub>2</sub> and O<sub>2</sub> were controlled by the mass flow controller. The simulated flue gas was then introduced into the scrubber.

#### 3-2. Scrubber

The scrubber used in this study was comprised of a tower sump for the containment and distribution of the wet catalyst, a particulate generator for particulate removal, a reactor for SO<sub>2</sub> and NO removal, a nozzle for the spraying of the wet catalyst and sampling ports. The pilot-scale scrubber was composed of acrylic material. The internal and external diameters of the reactor were 0.18 m and 0.20 m, respectively. The height of the reactor was 2.25 m. The reactor for the stacking of the packing material was comprised of a two-stage structure, each stage reaching a packing height of 0.65 m. The packing materials utilized in this study were applied to two types of pall ring, 5/8 in. (STS) and 1.0 in. (P.P.), both manufactured by Solmaro Engineering (Korea). The height of the particulate generator was 0.6 m. The characteristics of the pall ring and nozzle of

Table 1. Characteristics of packing materials (used in this work)

Material	Nominal size	Wall thickness (mm)	Q'ty (EA/m <sup>3</sup> )	Weight (kg/m <sup>3</sup> )	Surface area (m <sup>2</sup> /m <sup>3</sup> )	Void fraction (%)	Packing factor (m <sup>-1</sup> )
Metal (STS)	5/8 in.	0.4	234,000	465	362	92	230
P.P.	1.0 in.	1.5	50,100	88	205	90	170

**Table 2. Characteristics of nozzle of scrubber**

Nozzle kind	Orifice diameter (mm)	Flow rate (L/min)				Spraying angle (°)		
		0.5 kg/cm <sup>2</sup>	1 kg/cm <sup>2</sup>	1.5 kg/cm <sup>2</sup>	2 kg/cm <sup>2</sup>	0.3 kg/cm <sup>2</sup>	0.7 kg/cm <sup>2</sup>	1.5 kg/cm <sup>2</sup>
FF 6.5	2.31	2.1	2.9	3.5	4.0	45	45	5
FF 10	2.9	3.2	4.5	5.4	6.1	58	58	67
14 W	3.6	-	6.2	7.4	8.4	114	120	120

**Table 3. Experimental operating conditions of pilot-scale scrubber system**

Input gas concentration	Particulate	200±20 mg/Sm <sup>3</sup>
	SO <sub>2</sub>	375±25 ppm
	NO	155±20 ppm
Scrubber operation parameter	Gas flow rate	0.10 m/s
	Wet catalyst flow rate	4.2-9 L/min
	Liquid-gas ratio	27.52-58.98 L/m <sup>3</sup>
	Operation stage number	1 or 2 stage (0.5 or 1 m)

the scrubber are given in Tables 1 and 2.

#### 4. Experimental Procedure

For the continuous wet scrubbing in the De-particulate, De-SO<sub>2</sub> and De-NO studies, the range of operating conditions is shown in Table 3. Both the gas flow rate and wet catalyst flow rate were continuous. The operating temperature in the studies was maintained consistently at 35 °C. For the corresponding liquid-gas (L/G) ratio conditions, the gas flow rate was kept constant and the wet catalyst flow rate was adjusted. The O<sub>2</sub> concentration of the simulated gas was 4%.

#### 5. Analysis Method

The input and output concentrations of simulated flue gas (SO<sub>2</sub> and NO) were measured with a PGM-35 Toxic gas monitor (Rae System, USA), a detection tube (Gastec Co., Japan) and a Green Line MK2 (Eurotron, Italy). In order to analyze the particulate concentrations, a DustMate (Turnkey Instruments Ltd., England) was employed.

The removal efficiency (RE) was determined in accordance with the variables used to determine the treatment capacity of the scrubber. Removal efficiency was expressed as the content (%) of gas eliminated by the scrubber.

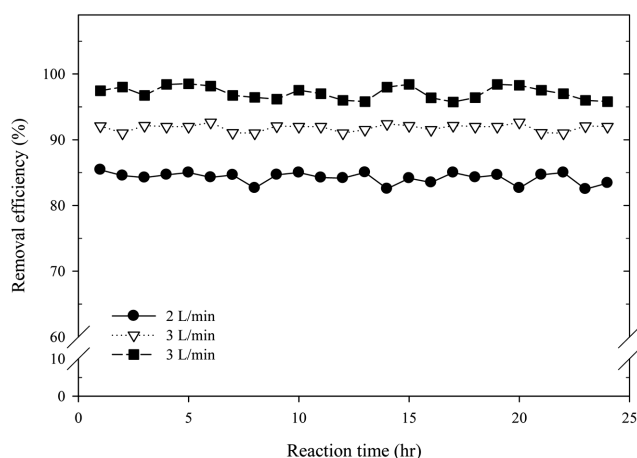
$$RE [\%] = (C_{Gi} - C_{Go}) / C_{Gi} \times 100$$

Where  $C_{Gi}$  and  $C_{Go}$  were the inlet and gas concentrations (ppm; g/m<sup>3</sup>).

### RESULTS AND DISCUSSION

According to Harriott [19], in order to facilitate the removal of NO via a wet process, the height of the NO wetting tower was most efficient at a height 30 times that of the general SO<sub>2</sub> absorption tower, and the ratio of the diameter and packing materials was correlated at approximately 1 : 4. In this study, in an attempt to remove particulate, SO<sub>2</sub> and NO efficiently, the effects of the wet catalyst flow rate, the number of stages of the scrubber, the liquid-gas ratio and the type of packing materials used were evaluated.

#### 1. Removal of Particulate in Pilot-Scale Scrubber



**Fig. 3. Effect of the flow rate of wet catalyst on particulate removal. Fe(II)-EDTA 0.03 M, ascorbic acid 0.024 M, adipic acid 0.024 M, sodium sulfite 0.09 M, particulate 150±10 mg/Sm<sup>3</sup>, initial pH 5, operating temperature 35 °C, 4% O<sub>2</sub>.**

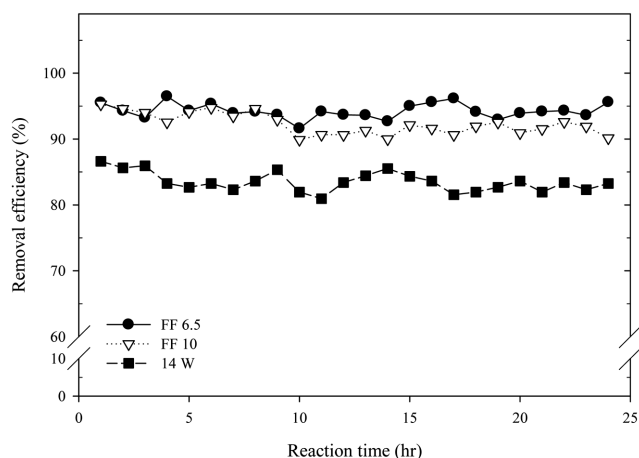
The factors that affect particulate removal include the shape and organization of the scrubber, the distribution and spray condition of the wet catalyst, the particulate input condition and several other factors. In this study, the shape and organization of the scrubber was fixed, and the particulate removal properties were analyzed relative to the distribution and spray condition of the wet catalyst, as well as the particulate input condition.

#### 1-1. Effect of Wet Catalyst Flow Rate on Particulate Removal

As shown in Fig. 3, the effect on particulate removal of the spray amount of the wet catalyst was determined via the operation of the particulate eliminator, at a 0.1 m/s gas flow rate. In this experiment, the wet catalyst was sprayed at flow rates of 2, 3 and 4 L/min at the top of the reactor by using a 5/8 in. FF6.5 (STS) nozzle. The input particulate concentration was 150±10 mg/Sm<sup>3</sup>. The particulate removal efficiency did not alter the reaction time; however, a change of the removal efficiency was noted with increases in the flow rate of the catalyst. The removal efficiencies were 83%, 92% and 97% at catalyst flow rates of 0.5 L/min, 0.8 L/min and 1.5 L/min, respectively. In the particulate removal experiments, as particulates can be removed without a chemical reaction as a result of physical contact with a wet catalyst, the removal efficiencies were found to be affected by the catalyst flow rates. This system is designed for the proper performance of particulate removal, via the control of the supplied wet catalyst flow rates and spray conditions.

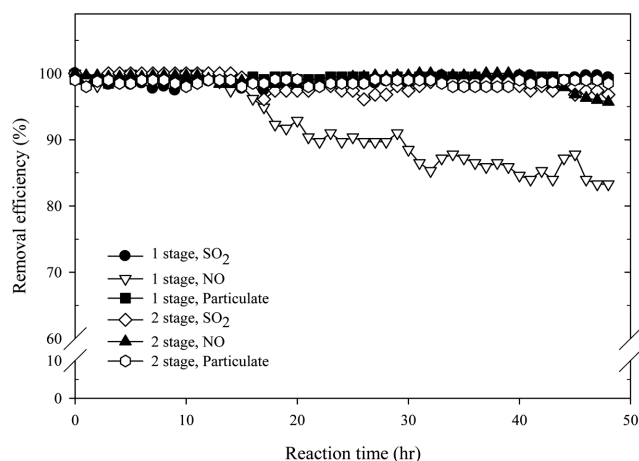
#### 1-2. Effect of Nozzle on Particulate Removal

As shown in Fig. 4, the effects of different nozzles on particulate removal were assessed under different particulate eliminator operation conditions, with a gas flow rate of 0.1 m/s. In order to deter-



**Fig. 4. Effects of nozzles on particulate removal.** Fe(II)-EDTA 0.03 M, ascorbic acid 0.024 M, adipic acid 0.024 M, sodium sulfite 0.09 M, particulate 170±10 mg/Sm<sup>3</sup>, initial pH 5, operating temperature 35 °C and 4% O<sub>2</sub>.

mine the most appropriate nozzle, a wet catalyst was supplied to the upper part of the reactor, and we tested the following nozzles: FF6.5 (5/8 in., STS), FF10 (5/8 in., STS) and 14W (1.0 in., P.P.). The characteristics of each of these nozzles are shown in Table 2. The input particulate concentration was 170±10 mg/Sm<sup>3</sup>. The particulate removal efficiency when different nozzles were used did not alter the reaction time; however, the kind of nozzle used did have an effect on the removal efficiency. The most appropriate nozzle was found to be the FF6.5 nozzle, with an approximately 94% removal efficiency, whereas a 90% removal efficiency was achieved with the 14 W nozzle. This difference in removal efficiency was attributed to the spraying shape of the nozzle. The FF6.5 nozzle had a lower spray angle, 45°, than did the FF10 (58°) and 14W (120°) nozzles, which induced a low wall stream, which could create sufficient physical contact between the particulate and the wet catalyst. Based on the above experimental results, the FF6.5 (5/8 in., STS)



**Fig. 5. Effect of the number of scrubber stages on SO<sub>2</sub>, NO and particulate removal.** Fe(II)-EDTA 0.03 M, ascorbic acid 0.024 M, adipic acid 0.024 M, sodium sulfite 0.09 M, SO<sub>2</sub> 375 ppm, NO 155 ppm, particulate 210 mg/Sm<sup>3</sup>, initial pH 5, operating temperature 35 °C and 4% O<sub>2</sub>.

nozzle was selected for the particulate eliminator, and the 14 W nozzle was selected for the scrubber for the removal of SO<sub>2</sub> and NO, as this application required a high spray angle, due to the substantial height of the scrubber.

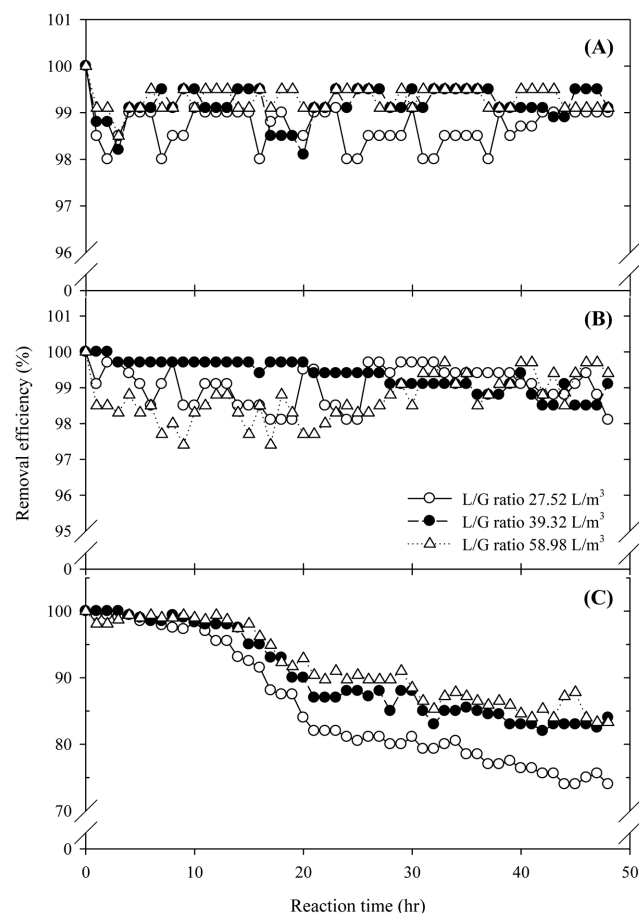
## 2. Effect of Stage Number of Scrubber on Simultaneous Removal of SO<sub>2</sub>, NO and Particulate

The effects of the number of stages of the scrubber on simultaneous removal of SO<sub>2</sub>, NO and particulate were observed in the experimental pilot-scale scrubber (Fig. 5). The first stage of the scrubber was 0.65 m high, and the packing height was 0.5 m. In order to remove the particulate, the particulate eliminator was operated under all conditions, and the gas flow rate and liquid-gas ratio were set to 0.10 m/s and 58.98 L/m<sup>3</sup>, respectively. In this study, the liquid-gas ratio was higher than the generally accepted value, due to the supply of a sufficient reaction time for the formation of the NO complex, as well as the considerable increase in the packing height. The input concentrations of SO<sub>2</sub>, NO, particulate and O<sub>2</sub> were 375 ppm, 155 ppm, 210 mg/Sm<sup>3</sup> and 4%, respectively.

The results of the above experiments indicated that the particulate and SO<sub>2</sub> removal efficiencies, which were not found to be related to the number of stages in the scrubber, were 98% and 96%, respectively. Incidentally, the NO removal efficiency was 83.3% for 48 hours in the one-stage design, but in the two-stage scrubbing system, a removal efficiency of 95.7% was achieved. Via the application of the two-stage scrubbing system, an approximately 15% increase in removal efficiency was achieved. In addition, after 16 hours of operation of the one-stage scrubbing system, the NO removal efficiency was shown to have decreased. As a result, the NO removal efficiency was augmented by the increase in packing height, which increased the contact time and the area of the wet catalyst and NO gas. The particulate removal efficiency in this case was in excess of 98%, which was higher than that of the particulate eliminator, and this was attributed to the removal of particulate by the scrubber. Lee et al. [2] previously reported that the maximum SO<sub>x</sub> and NO<sub>x</sub> removal efficiencies achieved under typical operating conditions of commercialized FGD systems using sodium chlorite solution in wetting-wall column systems, were approximately 100% and 67%, respectively. Hsu et al. [20] conducted an NO absorption study using a packed column. The results of that study indicated that the NO removal efficiency could be as high as 61.5%.

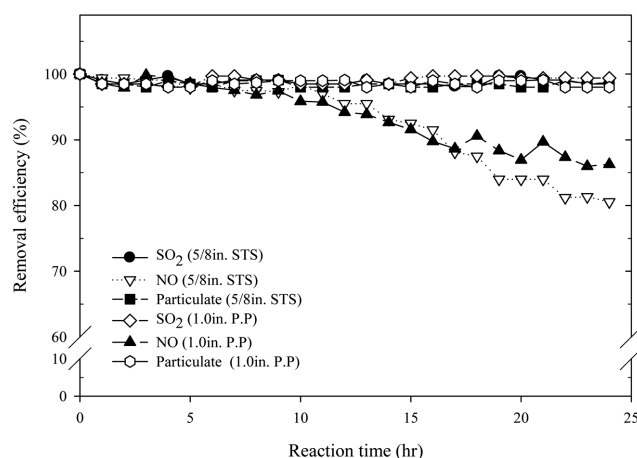
## 3. Effect of Liquid-Gas Ratio on Simultaneous Removal of SO<sub>2</sub>, NO and Particulate

The effects of the liquid-gas ratio on the simultaneous removal of SO<sub>2</sub>, NO and particulate were observed by using a pilot scale scrubber system (Fig. 6). In order to remove the particulates, the particulate eliminator was operated under all conditions, and the gas flow rate was 0.1 m/s. In this study, in order to determine the effects of the liquid-gas ratio, the scrubber was operated by using a one-stage system, and the applied liquid-gas ratio was set at 27.52 L/m<sup>3</sup>, 39.32 L/m<sup>3</sup> and 58.98 L/m<sup>3</sup>. Input concentrations of SO<sub>2</sub>, NO, particulate and O<sub>2</sub> were 393 ppm, 168 ppm, 213 mg/Sm<sup>3</sup> and 4%, respectively. As shown in Fig. 6(A) and 6(B), regardless of the liquid-gas ratio, after 48 hours of operation, the SO<sub>2</sub> and particulate removal efficiencies were in excess of 98% and 99%, respectively. With regard to the NO removal characteristics, the removal efficiencies were found to be 84% and 74% at liquid-gas ratios of 39.32 L/m<sup>3</sup> and 27.52 L/m<sup>3</sup>, respectively. These results evidenced a 14% decrease



**Fig. 6.** Effect of liquid-gas ratio on the simultaneous removal of  $\text{SO}_2$ , NO and particulate. Fe(II)-EDTA 0.03 M, ascorbic acid 0.024 M, adipic acid 0.024 M, sodium sulfite 0.09 M,  $\text{SO}_2$  393 ppm, NO 168 ppm, particulate 163 mg/ $\text{Sm}^3$ , initial pH 5, operating temperature 35 °C and 4%  $\text{O}_2$ . (A)  $\text{SO}_2$ , (B) particulate, (C) NO.

in removal efficiency consequent to the reduction in the experimental liquid-gas ratio. However, no significant differences in the NO removal characteristics of the system were noted as a result of an increase in the liquid-gas ratio from 39.32  $\text{L}/\text{m}^3$  to 58.98  $\text{L}/\text{m}^3$ . These results allowed for two conclusions. Firstly, in these NO removal experiments, the effect of the scrubber height proved to be more important than the effects of the liquid-gas ratio. As shown in Fig. 5, the NO removal efficiency at a liquid-gas ratio of 58.98  $\text{L}/\text{m}^3$  increased proportionally with the number of operating stages. Nevertheless, in the case of the one-stage scrubber, the liquid-gas ratio did not greatly affect NO removal efficiencies. This suggests that the height of the scrubber was the most important operating parameter. Secondly, under the same scrubber operation conditions, the NO removal efficiency might not differ substantially under high liquid-gas ratio conditions, because a liquid-gas ratio of 38.32  $\text{L}/\text{m}^3$  was already sufficient. The slight observed increase in the mass transfer rate occurring along with the increase of the liquid-gas ratio in a system might be responsible for the increase in the NO removal efficiency. Lee et al. [2] assessed the effects of differing liquid-gas ratios on the simultaneous removal of  $\text{SO}_2$  and NO via sodium chlorite solution in a wetted-wall column.  $\text{SO}_2$  removal efficiency was



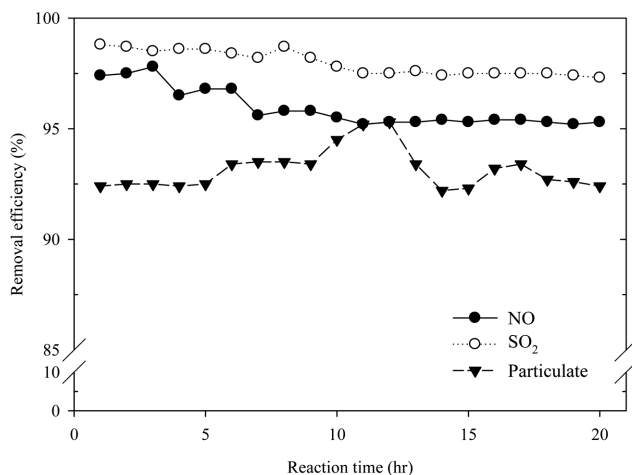
**Fig. 7.** Effect of packing materials on the simultaneous removal of  $\text{SO}_2$ , NO and particulate. Fe(II)-EDTA 0.03 M, ascorbic acid 0.024 M, adipic acid 0.024 M, sodium sulfite 0.09 M,  $\text{SO}_2$  380 ppm, NO 160 ppm, particulate 209 mg/ $\text{Sm}^3$ , initial pH 5, operating temperature 35 °C and 4%  $\text{O}_2$ .

assigned a constant value of approximately 100%, and NO removal efficiency was shown to increase slightly with increases in the liquid-gas ratio, in the range of 3–12  $\text{L}/\text{m}^3$ . Chien and Chu [1] reported that the major factor affecting NO removal was the liquid-gas ratio. A higher liquid-gas ratio (10  $\text{L}/\text{m}^3$ ) resulted in a higher NO removal efficiency, but also resulted in a shorter reaction period than was achieved at a lower liquid-gas ratio (5  $\text{L}/\text{m}^3$ ) in the lower NO concentration experiments.

#### 4. Effect of Packing Material on Simultaneous Removal of $\text{SO}_2$ , NO and Particulate

The wet catalyst sprayer and packing materials also affected the operational efficiency of the scrubber. In this study, the effects of packing materials on the operational efficiency of the scrubber were evaluated during the simultaneous removal of  $\text{SO}_2$ , NO and particulate (Fig. 7). The scrubber introduced the gas at a flow rate of 0.10 m/s, and a catalyst flow rate of 7 L/min. The removal efficiencies of  $\text{SO}_2$  and particulate were in excess of 98%, regardless of the type of packing materials utilized. The NO removal efficiency evidenced no substantial differences with the two tested packing materials, after 17 hours. After 24 hours, the pall ring (1.0 in., STS) resulted in a higher removal efficiency, 86%, than was achieved with the pall ring (5/8 in., P.P.). The pall ring (5/8 in., P.P.) had been packed at 234,000/ $\text{m}^3$  with a specific surface area of 362  $\text{m}^2/\text{m}^3$ , which was higher than that of the pall ring (1.0 in., STS). However, the high packing height resulted in low efficiencies of sprayed wet catalyst, due to streaming. Therefore, the pall ring (1.0 in., STS) proved to be an appropriate packing material for the pilot-scale scrubber system, due to its positive effects on the operational efficiency of the scrubber, as well as its relationship with removal efficiency via the catalyst flow rate, which required a HEPT setting. In this study, NO removal efficiency was affected principally by gas-liquid contact time and area. Further study should be conducted regarding the kinetics of the simultaneous absorption of  $\text{SO}_2$  and NO in aqueous mixed solutions of Fe(II)-EDTA and additives.

#### 5. Simultaneous Removal of $\text{SO}_2$ and NO at Optimum Operation Condition



**Fig. 8. Time course of the simultaneous removal of SO<sub>2</sub>, NO and particulate at optimum operation condition. Fe(II)-EDTA 0.03 M, ascorbic acid 0.024 M, adipic acid 0.024 M, sodium sulfite 0.09 M, SO<sub>2</sub> 544 ppm, NO 158 ppm, particulate 351 mg/Sm<sup>3</sup>, initial pH 5, operating temperature 35 °C, 4% O<sub>2</sub>, liquid-gas ratio 39.32 L/m<sup>3</sup>, gas flow rate 10 L/min and wet catalyst flow rate 0.5 L/min.**

The simultaneous removal of SO<sub>2</sub> and NO at optimum operation condition was observed by using a pilot-scale scrubber system (Fig. 8). In order to investigate the removal efficiency of SO<sub>2</sub> and NO, the scrubber was operated by using a two-stage system, and the applied liquid-gas ratio was set at 39.32 L/m<sup>3</sup>. The gas flow rate and wet catalyst flow rate were 10 L/min and 0.5 L/min, respectively. Input concentrations of SO<sub>2</sub>, NO, particulate and O<sub>2</sub> were 544 ppm, 158 ppm, 351 mg/Sm<sup>3</sup> and 4%, respectively.

With the above optimum operation conditions, even after 20 hours, the removal efficiency for NO stayed at 95% or higher, the removal efficiency for SO<sub>2</sub> stayed at 97% or higher, and the removal efficiency for particulate stayed at 92% or higher.

## CONCLUSION

This study developed a scrubber system that could simultaneously treat both SO<sub>2</sub> and NO in a single process by researching the optimum operation condition for the pilot-scale scrubber system. Using the combined SO<sub>2</sub>, NO and particulate removal system described in this study, the maximum SO<sub>2</sub> and particulate removal efficiencies were determined to be in the range of 96-98%. In the case of NO treatment, a removal efficiency of 83.3% was achieved after 48 hours under one-stage operational conditions. However, the removal efficiency reached 95.7% in the two-stage scrubber. Regardless of the liquid-gas ratio, the SO<sub>2</sub> and particulate removal efficiencies were in excess of 98% and 99%, respectively. However, the NO removal characteristics were not greatly altered by an increase in the liquid-gas ratio from 39.32 L/m<sup>3</sup> to 58.98 L/m<sup>3</sup>. However, the removal efficiencies were 84% and 74% at liquid-gas ratios of

39.32 L/m<sup>3</sup> and 27.52 L/m<sup>3</sup>, respectively. With regard to the application of packing material, particulate and SO<sub>2</sub> removal efficiencies were higher than 98%, without regard to the type of packing material utilized. However, NO removal differed with different tested packing materials. The above results indicate that this process might be employed in scrubber systems, and also might prove useful in the development of systems for the simultaneous removal of particulate, SO<sub>2</sub> and NO from flue gas.

## REFERENCES

1. T. W. Chien and H. Chu, *Journal of Hazardous Materials*, **80**, 43 (2000).
2. H. K. Lee, B. R. Deshwal and K. S. Yoo, *Korean J. Chem. Eng.*, **22**, 208 (2005).
3. M. Kang, J. H. Park, J. S. Choi, E. D. Park and J. E. Yie, *Korean J. Chem. Eng.*, **24**, 191 (2007).
4. J. H. Choi, J. H. Kim, Y. C. Bak, R. Amal and J. Scott, *Korean J. Chem. Eng.*, **22**, 844 (2005).
5. C. D. Cooper and F. C. Alley, *Air pollution control: A design approach*, 2nd Ed., Waveland Press Inc., Illinois, USA (1994).
6. G. T. Jeong, G. Y. Lee, D. H. Park and J. M. Cha, *Applied Biochemistry and Biotechnology*, **129-132**, 1077 (2006).
7. W. H. Yang and M. H. Kim, *Korean J. Chem. Eng.*, **23**, 908 (2006).
8. I. G. Jung, O. H. Park, H. J. Woo and C. H. Park, *Biotechnol. Bio-process Eng.*, **10**, 34 (2005).
9. C. L. Yang, H. Shaw and H. D. Perlmutter, *Chem. Eng. Comm.*, **143**, 23 (1996).
10. E. Sada, H. Kumazawa, I. Kudo and T. Kondo, *Ind. Eng. Chem. Process Des. Dev.*, **20**, 46 (1981).
11. J. L. De Pavia and G. C. Kachan, *Ind. Eng. Chem. Res.*, **37**, 609 (1998).
12. H. Chu, T. W. Chien and S. Y. Li, *Science of the Total Environment*, **275**, 127 (2001).
13. H. Perlmutter, H. Ao and H. Shaw, *Environ. Sci. Technol.*, **27**, 128 (1993).
14. Y. Shi, H. Wang and S. G. Chang, *Environ. Prog.*, **16**, 301 (1997).
15. Y. S. Ko, *Simultaneous removal characteristics of NO<sub>x</sub>, SO<sub>2</sub> from combustion gases using plasma chemical reaction*, MS thesis, Kyungnam University (2000).
16. S. G. Jin, J. M. Cha, I. W. Lee, S. J. Yoon and S. W. Kim, *Journal of The Korean Industrial and Engineering Chemistry*, **7**, 177 (1996).
17. S. H. Jung, *Simultaneous removal of SO<sub>2</sub>/NO using aqueous-catalyst in scrubber*, Ph.D. Dissertation, Chonnam National University (2007).
18. Y. Hishinuma, R. Kaji, H. Akimoto, F. Nakajima, T. Mori, T. Kamo, Y. Arikawa and S. Nozawa, *Bull. Chem. Soc. Jpn.*, **52**, 2863 (1979).
19. P. Harriott, K. Smith and L. B. Benson, *Environmental progress*, **12**, 110 (1993).
20. H. W. Hsu, C. J. Lee and K. S. Chou, *Chem. Eng. Commun.*, **170**, 67 (1998).