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Removal of NO_x and SO₂ by CuO/ γ -Al₂O₃ Sorbent/Catalyst in a Fluidized-Bed Reactor

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Selective reduction of NO and the simultaneous removal SO₂/NO by CuO/ γ -Al₂O₃ sorbent/catalyst were carried out in a fluidized-bed reactor. The optimum temperature ranges for NO reduction over the fresh and sulfated CuO/ γ -Al₂O₃ catalyst are found to be 250–300 and 300–450 °C, respectively. The NO removal efficiency with the sulfated catalyst is somewhat higher than that with the fresh catalyst. The amount of optimum Cu/S is found to be 1.5 in the simultaneous removal of SO₂/NO. The optimum temperature range for the simultaneous removal of SO₂/NO by CuO/ γ -Al₂O₃ sorbent/catalyst in the reactor is 350–400 °C based on the SO₂ and NO removal efficiencies (>90%) and N₂O formation (<10% of NO in feed).

Introduction

Sulfur oxides (SO_x) and nitrogen oxides (NO_x) in flue gas are the major air pollutants, which have to be removed before emitting into the atmosphere. Various processes are under operation to remove SO_x and NO_x from flue gas.¹ Numerous research works on the simultaneous removal of SO_x and NO_x have been carried out to find the cost-effective catalysts/sorbent. In simultaneous removal of SO_x and NO_x, the selective catalytic reduction (SCR) process with CuO/ γ -Al₂O₃ has distinct advantages to remove SO_x and NO_x in flue gas because of the predominant reactivity with SO₂ in the presence of oxygen and easy regeneration under the reductant gas atmosphere.^{2–5}

Recently, studies on sulfation,^{4,8} regeneration,^{5,6} and enhancement⁷ of the sorption capacity of CuO/ γ -Al₂O₃ as a sorbent to remove SO₂ and the SCR⁹ of NO over CuO/ γ -Al₂O₃ as a catalyst to remove NO_x have been reported by the present authors. It has been reported that the SO₂ removal capacity of CuO/ γ -Al₂O₃ can be largely enhanced by the sulfation conditions and alkali-salt promoters.^{4,7} NO conversion in SCR over the sulfated catalyst reaches 90% at the optimum temperature range.⁹ In the simultaneous SO_x and NO_x removal process, the amount of sulfates in CuO/ γ -Al₂O₃ sorbent/catalyst will gradually increase with reaction time. Although the honeycomb-type reactor has been used to remove NO_x, this reactor suffers from external mass-transfer limitation, nonideal gas–solid contact, laborious catalyst replacement, and difficulty of simultaneous removal of SO_x/NO_x.¹⁰

The reductions of SO_x and NO_x in fluidized-bed reactors have been studied with the following advantages: (1) Fly ash passes through the reactor without plugging that can occur in fixed beds. (2) Contacting efficiency may be insensitive compared to the honeycomb reactor with variation of gas flow rate. (3) Shut-down for replacement of catalyst is not needed. (4) SO_x and NO_x can be removed in one stage simul-

taneously.^{10–13} On the other hand, the main potential disadvantages of fluidized beds would be a higher pressure drop compared to the parallel passage reactor, possible attrition of the bed material, and the undesired NH₃ slip through bubbles.¹³

Although the CuO/ γ -Al₂O₃ process is able to remove SO_x and NO_x simultaneously, research works for the CuO/ γ -Al₂O₃ process using fluidized beds has been mainly focused on to remove SO_x from the flue gas.^{14–17} Simultaneous removal of SO₂/NO_x using CuO/ γ -Al₂O₃ in a fluidized-bed reactor was reported by Yeh et al.,¹³ but their result was limited to a narrow temperature range and N₂O produced during NO_x removal by the SCR was neglected.

Therefore, the SCR of NO and the simultaneous removal of SO₂/NO_x using CuO/ γ -Al₂O₃ have to be systematically studied in a fluidized-bed reactor for possible commercial application. In the present study, the effects of reaction temperature, NH₃ concentration, superficial gas velocity, and sorbent/catalyst feeding rate on the SCR of NO and the simultaneous removal of SO₂/NO_x using CuO/ γ -Al₂O₃ have been determined in a fluidized-bed reactor.

Experimental Section

Catalyst Preparation. The catalyst was prepared by impregnating a copper precursor solution [Cu(NO₃)₂·3H₂O] into γ -Al₂O₃ of 0.65 mm mean diameter (Alcoa Co., Pittsburgh, PA). The alumina support was dried at 110 °C for 24 h and allowed to cool in a desiccator. After complete evaporation of moisture from the impregnated alumina in an oven, it was taken out and then calcined at 600 °C under air flow. The concentration of copper oxide (8 wt %) in the calcined sorbent based on the dry alumina was measured by atomic absorption spectroscopy. After calcination, uniformity of CuO distribution in CuO/ γ -Al₂O₃ was verified by scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX). The surface area, average pore diameter, and pore volume of the fresh catalyst after calcination are found to be 213 m²/g, 74.6 Å, and 0.4 cm³/g, respectively. To prepare the sulfated catalyst, sulfation of CuO/ γ -Al₂O₃ was carried out in a fluidized-

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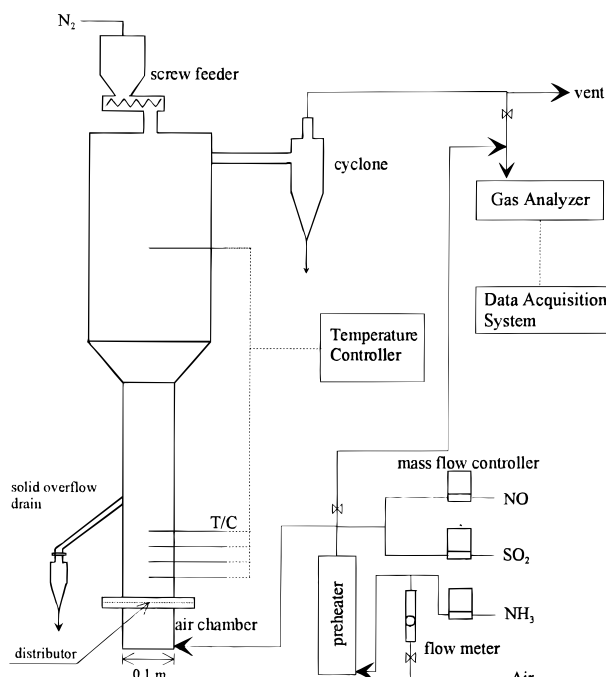


Figure 1. Schematic diagram of a fluidized-bed reactor for simultaneous removal of SO_2/NO_x .

bed reactor under 3000 ppm SO_2 /air flow rate of $2U_{mf}$ (0.74 m/s) at 400 °C.

Experimental Procedure

SCR of NO by ammonia over $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ catalyst was carried out in a fluidized-bed reactor (0.1 m i.d. \times 2.0 m high) with a sintered metal plate distributor. The experimental equipment (Figure 1) consists of three sections: reactor, gas feeding system, and gas analyzer. The reactor with $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ particles was heated to the desired reaction temperature by an electric heater with an on/off controller. The flow rates of simulated flue gas (400 ± 10 ppm NO, 1500–2000 ppm SO_2 /air) and ammonia were regulated by a flowmeter and mass flow controllers (MFC) and introduced into an air chamber via a preheater, which was heated to 300 °C to prevent the formation of ammonium sulfates and nitrates. For simultaneous removal of SO_x and NO_x , a screw feeder was installed to feed the $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ sorbent/catalyst particles at the top of the fluidized-bed reactor.

When the system reached steady state at the desired temperature and superficial gas velocity (U_g), NH_3 was injected into the bed having a static bed height of 0.2 m. The reaction temperature, U_g , NH_3 concentration, and Cu/S mole ratio (moles of Cu in the solid feed per moles of S in the inlet gas stream) were varied in the range of 200–500 °C, $1\text{--}4U_{mf}$, $0.5\text{--}2$ $[\text{NH}_3]/[\text{NO}]$, and $0.5\text{--}2.0$ Cu/S, respectively. The concentrations of NO, SO_2 , N_2O , and NH_3 were measured by gas analyzers (Chung Eng. Co., Korea; Hartmann & Braun Co., Germany; Siemens Co., Germany) using a nondispersed infrared (ND-IR) method with a vacuum pump.

Results and Discussion

Effect of Reaction Temperature on NO Removal by Fresh $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ Catalyst. The effect of reaction temperature on the catalytic activity over the fresh $\text{CuO}/$

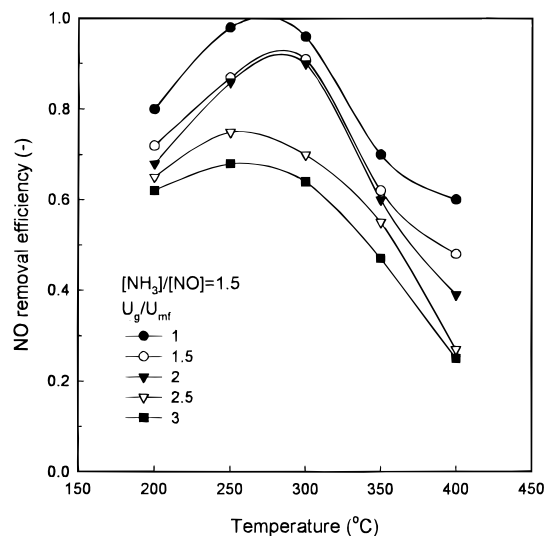


Figure 2. Effect of the reaction temperature on NO conversion over the fresh $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ in a fluidized-bed reactor.

Table 1. Basic Reactions in the Selective Catalytic Reduction of NO by NH_3

$4\text{NO} + 4\text{NH}_3 + \text{O}_2 \rightarrow 4\text{N}_2 + 6\text{H}_2\text{O}$	(1)
$4\text{NO} + 4\text{NH}_3 + 3\text{O}_2 \rightarrow 4\text{N}_2\text{O} + 6\text{H}_2\text{O}$	(2)
$6\text{NO} + 4\text{NH}_3 \rightarrow 5\text{N}_2 + 6\text{H}_2\text{O}$	(3)
$8\text{NO} + 2\text{NH}_3 \rightarrow 5\text{N}_2\text{O} + 3\text{H}_2\text{O}$	(4)
$4\text{NH}_3 + 3\text{O}_2 \rightarrow 2\text{N}_2 + 6\text{H}_2\text{O}$	(5)
$4\text{NH}_3 + 5\text{O}_2 \rightarrow 4\text{NO} + 6\text{H}_2\text{O}$	(6)

$\gamma\text{-Al}_2\text{O}_3$ as a bed material in the fluidized-bed reactor is shown in Figure 2 in which the removal efficiency for species i is defined as

$$\text{removal efficiency} = \left(\frac{C_0 - C_f}{C_0} \right) \quad (1)$$

where C_0 and C_f are the initial concentration and the final concentration of species i , respectively. NO conversion exhibits the maximum values with variation of reaction temperature regardless of U_g . The decrease of NO conversion above the maximum value may be caused by the increase of the parallel reaction rate of NH_3 oxidation to N_2 by reaction 5 in Table 1.^{9,18} As can be seen, NO conversion is higher than 0.9 at $U_g < 2U_{mf}$ and 250–300 °C. The NO removal efficiency decreases sharply with increasing U_g because of the bypassing of reactant gas through bubbles and the reduction of the gas residence time in the bed.¹⁹

Therefore, in the present study, $2U_{mf}$ was employed as the optimum U_g in all of the experimental conditions based on the NO removal efficiency and the amount of flue gas treatment.

N_2O Formation and NH_3 Slip. The amounts of N_2O formation and unreacted NH_3 slip in the SCR of NO in a fluidized-bed reactor are shown in Figure 3. The formation of N_2O by reaction 2 in Table 1 increases up to about 20% of NO in the feed stream with increasing temperature as found previously in a fixed-bed reactor.⁹ However, the amount of produced N_2O in the SCR exhibits a constant value regardless of U_g at a given temperature. It may indicate that U_g does not affect N_2O formation in the SCR of NO at a given NH_3 concentration. The amount of NH_3 slip is significant at 200 °C at the given U_g because NH_3 oxidation does not actively occur at the given temperature.⁹ Also, the amount of effluent NH_3 from the reactor increases with increasing

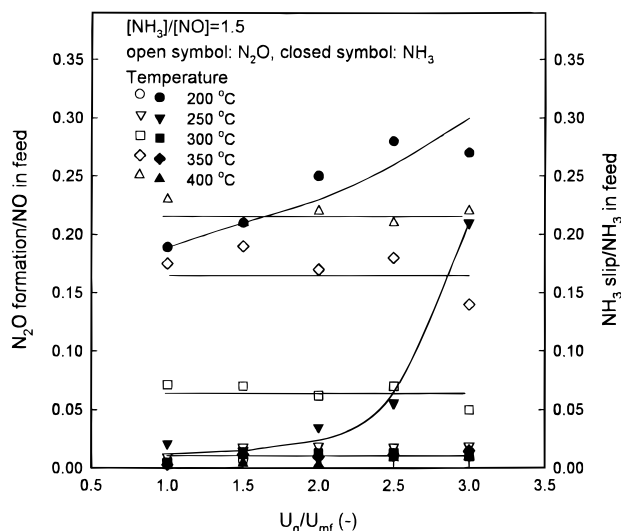


Figure 3. Effect of U_g on N_2O formation and NH_3 slip with a variation of the reaction temperature over the fresh $CuO/\gamma-Al_2O_3$ in a fluidized-bed reactor.

U_g because of an increase of NH_3 bypassing through bubbles. At 250 °C, NH_3 slip exhibits the lower values ($<0.03 NH_3$ slip/ NH_3 in the feed) up to $2.0U_{mf}$ and increases sharply up to about 25% of NH_3 in the feed stream with a further increase in U_g . However, NH_3 slip is not affected by U_g above 300 °C. The amount of NH_3 slip is very low at the temperature above 300 °C because of the active NH_3 oxidation above 300 °C.⁹ Nevertheless, more detailed study is needed to measure the trace level of NH_3 in the flue gas to meet the regulatory emission level of NH_3 . In the practical operation, NH_3 slip may decrease with increasing catalyst bed height because $CuO/\gamma-Al_2O_3$ is an effective catalyst for NH_3 oxidation.⁹ The higher catalyst bed height may provide higher NO_x and SO_x removal efficiencies with higher operating cost because higher pressure drop in the bed. Therefore, further study is needed to determine the optimum operating condition.

SCR over Sulfated Catalyst. In the simultaneous removal of SO_x and NO_x , the amount of sulfates in $CuO/\gamma-Al_2O_3$ sorbent/catalyst will gradually increase with the reaction time. Therefore, it is important to determine the SCR characteristics of NO over the sulfated catalyst in a fluidized-bed reactor.

The optimum amount of NH_3 supply in the SCR process is important for NO conversion, unreacted NH_3 slip, and the operating cost. The effect of NH_3 concentration on NO conversion and N_2O formation is shown in Figure 4. As can be seen, NO conversions over the fresh and sulfated catalysts increase gradually up to 90% with increasing NH_3/NO mole ratio up to 1.5 and remain constant with a further increase in the mole ratio. Yeh et al.¹³ reported that the NO removal efficiency is higher in a deep bed than in a shallow bed over the NH_3/NO range of 0.3–0.9. Although NO conversion in the present study at a given NH_3 concentration is somewhat lower than that of Yeh et al.,¹³ who carried out a DeNO_x test at lower U_g ($\approx 1.1U_{mf}$) in a deep bed (0.457 m), the general trend is similar. Also, it can be noticed that NO conversion over the sulfated catalyst is higher than that over the fresh catalyst because of the increase of acidity that is caused by the surface ion of SO_4^{2-} on the catalyst.^{9,20}

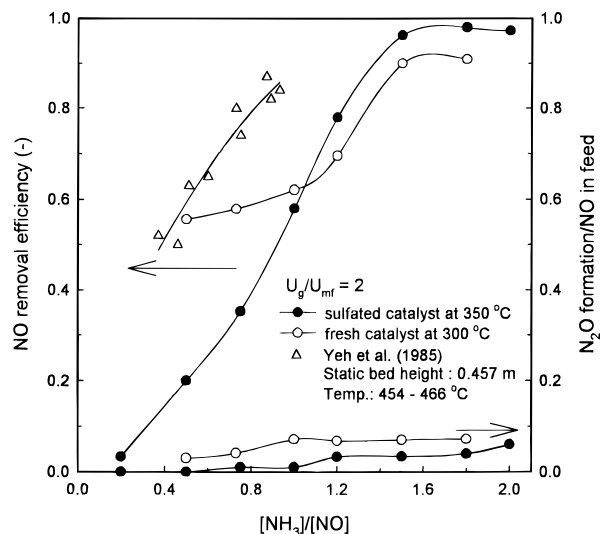


Figure 4. Effect of NH_3 concentration on NO conversion and N_2O formation over the fresh and sulfated $CuO/\gamma-Al_2O_3$ with a variation of the reaction temperature.

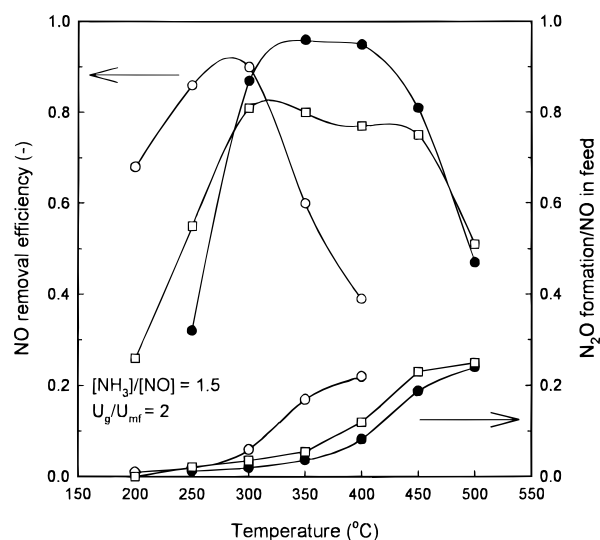


Figure 5. Effect of the reaction temperature on NO conversion and N_2O formation over the fresh and sulfated $CuO/\gamma-Al_2O_3$: ○, fresh catalyst; ●, sulfated catalyst; □, fresh + sulfated catalyst (1:1).

The amount of N_2O produced in the SCR of NO by NH_3 over the fresh and sulfated $CuO/\gamma-Al_2O_3$ in the reactor exhibits less than 0.1 of NO in the feed stream at the given temperatures, which may indicate no relation to NH_3 concentration.

The effect of reaction temperature on NO conversion and N_2O formation over the fresh and sulfated $CuO/\gamma-Al_2O_3$ catalysts is shown in Figure 5. As can be seen, the temperature at which NO conversion exhibits the optimum value (about 95%) with the sulfated $CuO/\gamma-Al_2O_3$ catalyst is about 300–450 °C, which is 100 °C higher than that with the fresh catalyst. Also, the optimum temperature window for SCR over the sulfated catalyst is much wider than that over the fresh catalyst. These results in a fluidized bed exhibit a trend similar to those in a fixed-bed reactor.⁹ This wider optimum temperature window may be able to ensure the flexible operation of the DeNO_x process.

N_2O starts to produce over the sulfated catalyst at higher temperatures compared to that over the fresh

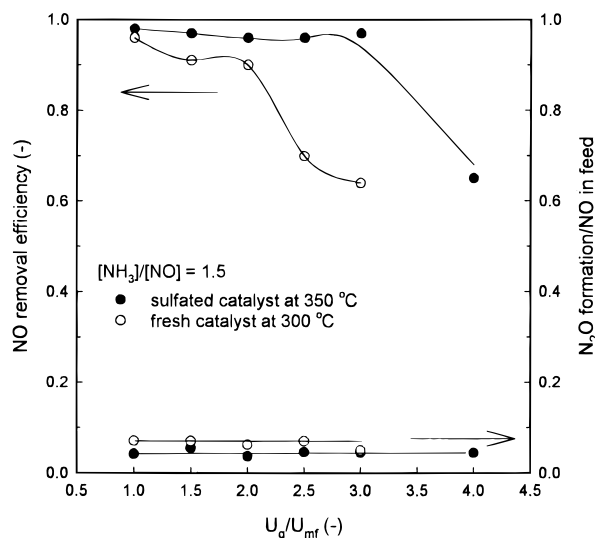


Figure 6. Effect of U_g on NO conversion and N_2O formation over the fresh and sulfated $CuO/\gamma-Al_2O_3$.

catalyst. The amount of produced N_2O exhibits lower values (<0.1 of NO in the feed stream) at the optimum temperatures with the fresh and sulfated catalysts.

With physically mixed fresh and sulfated catalysts (1:1) as the bed material, the behaviors of NO conversion and N_2O formation are similar to those of the sulfated catalyst, although the maximum NO conversion in the bed of mixed catalysts is lower than that in the bed of sulfated catalyst. From this result, it can be claimed that the presence of a small amount of sulfate in the bed of $CuO/\gamma-Al_2O_3$ may bring about the optimum temperature shift to the higher values and widening of the optimum temperature window.

The effect of superficial gas velocity (U_g) on NO conversion is shown in Figure 6 to determine the optimum U_g for SCR in a fluidized-bed reactor. As expected, the treatment capacity of the flue gas from a stationary source increases with increasing U_g . As can be seen, NO conversions over the fresh and sulfated $CuO/\gamma-Al_2O_3$ catalysts maintain above 0.9 below $2U_{mf}$ and $3U_{mf}$, respectively, and decrease with a further increase in U_g . This sharp decrease may result from bypassing of reactant gas through bubbles and the reduction of the gas residence time through the bed.¹⁹ Higher NO conversion over the sulfated catalyst compared with that over the fresh catalyst at higher U_g ($2-3U_{mf}$) may be caused by strong adsorption of the reactant gases (NH_3 and NO) onto the sulfated catalyst.^{9,20} It is reported that gas interchange between bubble and dense phases can be greatly enhanced by adsorption.²¹ Sulfate in the catalyst may increase diffusion of NH_3 to the dense phase from the bubble phase by supporting the adsorption site and, as a result, the high NO conversion may be maintained over the sulfated catalyst even at higher U_g (up to $3U_{mf}$). From these results, it can be claimed that the sulfated $CuO/\gamma-Al_2O_3$ catalyst is more profitable than the fresh catalyst for SCR of NO based on the throughput of the flue gas and NO removal efficiency. The amount of N_2O produced is independent of U_g . Therefore, N_2O formation over the sulfated catalyst as well as over the fresh catalyst may be a function of only the reaction temperature.

Simultaneous Removal of SO_2/NO_x in a Fluidized-Bed Reactor. One of the advantages of $CuO/\gamma-Al_2O_3$ for flue gas treatment is to remove SO_x and NO_x

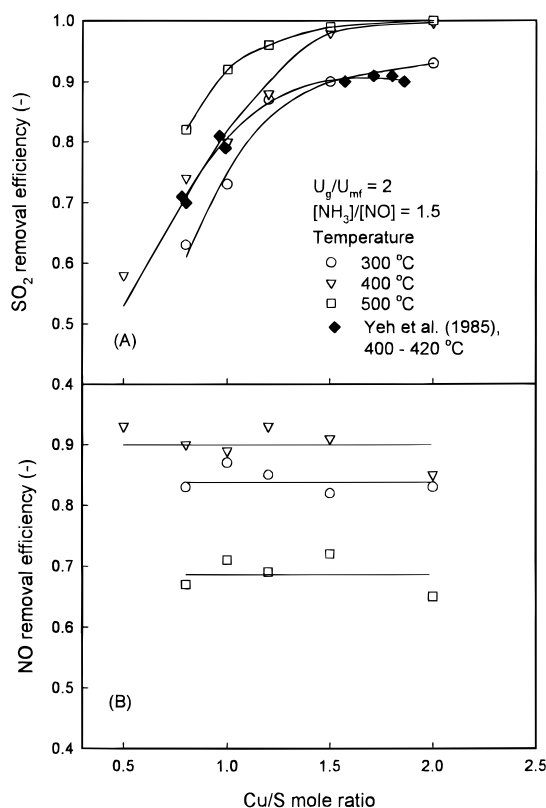
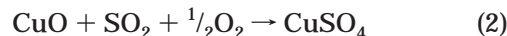


Figure 7. Effect of Cu/S mole ratio on the (A) SO_2 and (B) NO removal efficiencies in the simultaneous removal of SO_2/NO using $CuO/\gamma-Al_2O_3$ sorbent/catalyst.

simultaneously. In the simultaneous removal of SO_2/NO_x using $CuO/\gamma-Al_2O_3$ sorbent/catalyst, SO_2 reacts with CuO and Al_2O_3 to form $CuSO_4$ and $Al_2(SO_4)_3$ as follows:⁴



NO_x is reduced to N_2 by NH_3 over the sulfated surface of sorbent/catalyst following reactions in Table 1.

The effect of the amount of $CuO/\gamma-Al_2O_3$ feeding rate on the SO_2/NO removal efficiency is shown in Figure 7 to determine the optimum Cu/S mole ratio (moles of Cu in the solid feed per moles of S in the inlet gas stream) in a fluidized-bed reactor. As can be seen in Figure 7A, the SO_2 removal efficiency increases with an increase in the Cu/S mole ratio up to 1.5 Cu/S and then remains constant. This $DeSO_x$ trend with Cu/S is similar to that in a previous study.¹³ Also, SO_2 conversion increases with the reaction temperature because of good reactivity at higher temperatures. As can be seen, the SO_2 removal efficiency in the simultaneous removal of SO_2/NO exhibits very higher values ($>95\%$) in the condition of 1.5 Cu/S above 400 °C. The NO removal efficiency is independent of the feeding rate of $CuO/\gamma-Al_2O_3$ in the simultaneous removal of SO_2/NO (Figure 7B).

The effect of the reaction temperature on the simultaneous removal of SO_2 and NO is shown in Figure 8. With a S/Cu mole ratio of 1.5, the SO_2 removal efficiency reaches 90% at 300 °C and slightly increases with a further increase in the reaction temperature. On the other hand, the SO_2 removal efficiency with 1.0 S/Cu

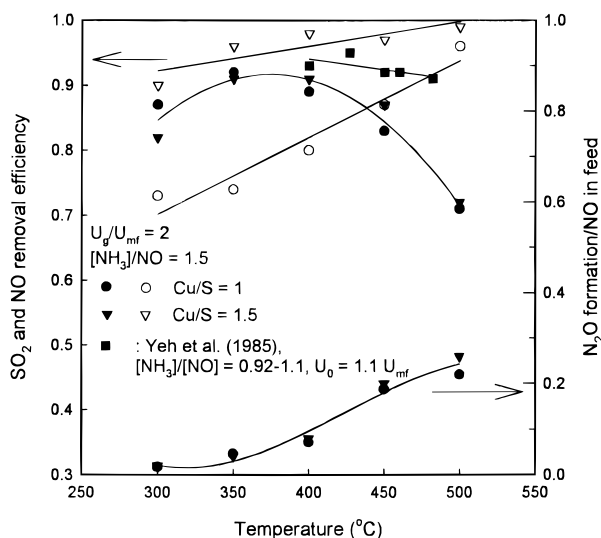


Figure 8. Effect of temperature on the SO_2 and NO removal efficiencies and N_2O formation in the simultaneous removal of SO_2/NO using $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ sorbent/catalyst. Open symbol: SO_2 . Closed symbol: NO and N_2O .

mole ratio exhibits 70% at 300 °C and sharply increases with a further increase in the reaction temperature compared to that with 1.5 S/Cu mole ratio. The increasing SO_2 uptake of $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ with increasing reaction temperature may be due to the rapid reaction rate and sulfation of $\gamma\text{-Al}_2\text{O}_3$ at higher temperatures. It is reported that the amount of $\text{Al}_2(\text{SO}_4)_3$ in the bulk of sorbent/catalyst gradually increases because of the sulfation of alumina support with an increase in the reaction temperature.⁴

Although the NO removal efficiency in the simultaneous removal is slightly lower than that over the sulfated catalyst in the SCR of NO only (Figure 5) because of the competitive adsorption of NH_3 and SO_2 onto $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$, the efficiency exhibits above 85% at the temperature range of 300–450 °C. The NO removal efficiency in the present study coincides with the result of Yeh et al.¹³

With an increase in the reaction temperature, N_2O formation increases up to 25% of NO in the feed. Over the sulfated catalyst, the behaviors of NO removal and N_2O formation in the simultaneous removal are similar to those in the SCR of NO only. It can be concluded that the optimum temperature range for the simultaneous removal of SO_x/NO_x using $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ sorbent/catalyst in a fluidized-bed reactor is 350–400 °C based on the SO_2 and NO removal efficiencies (>90%) and N_2O formation (<10% of NO in the feed). Therefore, the $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ process in a fluidized-bed reactor is an effective way to remove SO_x/NO_x simultaneously from the flue gas.

Conclusions

The removal characteristics of NO_x and SO_2 by using $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ sorbent/catalyst have been determined in a fluidized-bed reactor. The optimum temperature range for NO reduction over the sulfated $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ catalyst is 300–450 °C, which is 100 °C higher and much wider than that over the fresh catalyst. Also, the NO removal efficiency with the sulfated catalyst is somewhat higher than that with the fresh catalyst. It is found that N_2O formation in the SCR of NO is a function of only the

reaction temperature and it increases with an increase in the reaction temperature. NH_3 slip with the fresh catalyst does not nearly occur above 300 °C, which is the optimum temperature for NO reduction.

In the simultaneous removal of SO_2/NO , the amount of optimum Cu/S is found to be 1.5 and the NO removal efficiency is independent of the feeding rate of $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$. With a S/Cu mole ratio of 1.5, SO_2 and NO_x removal efficiencies reach above 90% at 300–500 °C and above 85% at the temperature range of 300–450 °C, respectively. The optimum temperature range for the simultaneous removal of SO_x/NO_x by $\text{CuO}/\gamma\text{-Al}_2\text{O}_3$ sorbent/catalyst is 350–400 °C based on the SO_2 and NO removal efficiencies (>90%) and N_2O formation (<10% of NO in feed).

Acknowledgment

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