

Experimental research and characteristics analysis of alumina-supported copper oxide sorbent for flue gas desulfurization[†]

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ABSTRACT: The dry regenerative sorption process using the γ -alumina-supported copper oxide sorbent is considered an alternative to the conventional once-through limestone scrubbing process for flue gas desulfurization. To define the characteristics of the γ -alumina-supported copper oxide sorbent in the sulfation and regeneration reactions, the wet-impregnation method was applied to prepare γ -alumina-supported copper oxide sorbent, and cycles of sulfationregeneration reactions were carried out in a quartz tube reactor. The effects of the physical properties of the used supports and the concentration of the impregnation solution on the SO₂ sorption capacity of the sorbent, as well as the dispersed form of the copper oxide on the support and the stability of the sorbent were determined by means of Brunauer, Emmett and Teller (BET), X-Ray Diffraction (XRD), electron probe microanalyser (EPMA) and scanning electron microscope (SEM) techniques. The results show that the γ -alumina used for the sorbent should have both a large surface area and an ideal pore size. As the supports were impregnated with 2 mol/l Cu(NO₃)₂ solution, the loading amount of the active copper oxide coated on the sorbent was optimum and the copper oxide was dispersed in the desired form. The prepared γ -alumina (DS)-supported copper oxide sorbent exhibited high SO₂ sorption capacity and the desired sulfation-regeneration properties. © 2007 Curtin University of Technology and John Wiley & Sons, Ltd.

KEYWORDS: flue gas desulfurization; copper oxide; sorbent; SO₂

INTRODUCTION

Emission of SO_2 from the combustion of fossil fuels causes air pollution. The technologies developed for the control of emission of SO_2 from the flue gas can be categorized into either dry and wet, or recovery and throwaway processes. Most of the desulfurization processes are based on the throwaway wet-scrubbing technique, using limestone as absorption reagent, because of the low costs of these processes. But they suffer from a common deficiency: they convert the air pollution problem into a solid or liquid pollution problem (Henzel *et al.*, 1982). Dry regenerative sorption processes based on chemical reaction of SO_2 with a metal oxide such as CuO (Yeh *et al.*, 1985; Centi *et al.*, 1990; Kiel *et al.*, 1992; Deng and Lin, 1995a) are considered as alternatives for flue gas desulfurization

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(FGD) because of their several advantages over the conventional wet-scrubbing, throwaway processes (Harriott and Markussen, 1992; Frey, 1993; Centi *et al.*, 1995).

In a dry regenerative copper oxide flue gas desulfurization process, the flue gas is brought into contact with CuO, whereupon the SO₂ reacts with CuO and O₂ to form copper sulfate (CuSO₄) at 350-500 °C. The sulfated sorbent can be regenerated with a reducing gas such as methane or carbon monoxide in the same temperature range as used in the sulfation stage, the copper sulfate being reduced to elemental copper. The concentration of the sulfur dioxide in the regeneration of gas is high enough for further processing to generate sulfuric acid or elemental sulfur (Satriana, 1981). After regeneration, the metallic copper in the sorbent can be easily oxidized in the air at the same operating temperature and used in the next sulfation stage.

Selecting the right sorbent is the key to the success of the dry regenerative sorption FGD processes. In the past years, a large number of metal oxide active species have been studied for their reaction rates with sulfur dioxide (DeBerry and Sladek, 1971). It was found that the oxides of Cu, Cr, Fe, Ni, Co and Ce can react with



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sulfur dioxide present in the flue gas at an economically feasible reaction rate (Uysal et al., 1988). Among these favorable active species for flue gas desulfurization purposes, copper oxide is the most promising and extensively studied (Cho and Lee, 1983). Besides, different porous materials including γ -alumina, α - alumina, silica and titania have been used as the sorbent supports (Centi et al., 1990; Wolff et al., 1993) y-Alumina is the most commonly accepted support because of its large area and excellent characteristics (Buelna and Lin, 2001). Therefore, the CuO/ γ -Al₂O₃ sorbent for FGD is worth studying. In addition, several different methods, such as wet impregnation, vacuum impregnation and sol-gel, have been employed to disperse the active copper oxide on the surface of the supports. Among these sorbent preparation methods, the sol-gel method may be the best (Deng and Lin, 1995b), but it is very complicated and can be applied only in the laboratory at present, whereas the impregnation method is economically more feasible (Dautzenberg and Nader, 1971). Several investigators have studied the characteristics of the CuO/ γ -Al₂O₃ sorbent prepared by the wet impregnation method in sulfation reaction (Cull, 1978; Kyung et al., 1994). However, the influence of the factors of the wet impregnation method on the SO₂ sorption capacity of the sorbent, such as the influence of the support and concentration of the impregnation solution, needs to be determined. Besides, the criteria for use and the form of the copper oxide coated on the support, as well as the stability of the sorbent in the cycles of sulfation-regeneration reaction, also need to be studied further.

In this study, γ -alumina-supported copper oxide sorbent was prepared by the wet impregnation method. The effects of the physical properties of the used supports and the concentration of the impregnation solution on the SO₂ sorption capacity of the sorbent, as well as the dispersed form of the copper oxide coated on the support were determined. In addition, the thermal and chemical stability of the prepared CuO/ γ -Al₂O₃ sorbent were also examined after several sulfation–regeneration reaction cycles.

EXPERIMENTAL

Sorbent preparation

The γ -alumina-supported copper oxide sorbents were prepared by the wet impregnation method. The support used for sorbent was γ -Al₂O₃ pellets with a diameter of 4 mm. It was dried at 160 °C for 24 h and allowed to cool to room temperature in a desiccator. A definite mass of dried γ -Al₂O₃ was soaked in a known concentration of Cu(NO₃)₂ solution for a few hours. Then the impregnated γ -alumina particles were dehydrated in a drying oven where temperature rose gradually from

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50 °C to 200 °C. After the impregnated γ -alumina had been cooled to room temperature, it was calcined at 500 °C in a muffle furnace for 5 h to form the γ -alumina-supported copper oxide sorbent.

Experimental procedure

The sulfation reaction was carried out in an electrically heated quartz tube reactor, as shown in Fig. 1 (Ma et al., 2002). The experimental system consists of two sections: a preheating section and a reaction section. The preheating section heated up the simulated flue gas, which consisted of air and SO_2 , and the SO_2 concentration of the simulated flue gas was 1000 ppm. The reaction section provided the desired sulfation reaction temperature, which was 350-450 °C. The γ alumina-supported copper oxide sorbent was placed on a quartz plate in the reaction section, and a number of 1 mm diameter eyelets were evenly distributed on the quartz plate. The distance between the top of the quartz tube and the quartz plate was 600 mm. The flow rate of the simulated flue gas was 3.8 l/min. The stack height of the sorbents was about 50 mm, and the residence time of the simulated flue gas in the sorbent bed was about 1 s.

The experimental system for regeneration was similar to the sulfation reaction system, as shown in Fig. 2 (Deng *et al.*, 2003). The sulfated sorbent was taken out from the sulfation reaction system and regenerated with the reducing gas in the regeneration reaction system. The reducing gas consisted of methane and a balancing gas (N₂), and the flow rate of the reducing gas was 2.4 l/min. The temperature of the regeneration reaction was the same as that of the sulfation reaction.



SO₂; 2. Air; 3. Rotameter; 4. Mixer; 5. Flue gas analysis meter;
 Gas switch valve; 7. Quartz tube reactor; 8. Exhaust gas absorption cell;
 9. Reactor section temperature controller;
 10. Preheating section temperature controller

Figure 1. Schematic of the desulfurization experimental system.

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1.CH₄; 2. N₂; 3. Rotameter; 4. Preheater; 5. Thermocouple; 6. Temperature controller; 7. Flue gas analyser; 8. Quartz tube reactor



A KM900 flue gas analyzer was used to measure the gas from the exit of the reactors. The SO₂ measurement range of the KM900 analyzer was 0-5000 ppm. After the sulfation or the regeneration reaction, physical properties of the sorbent samples (surface area, pore volume) and their crystal structures, as well as the dispersed form of the active species coated on the sorbent were determined by BET (ASAP-2000), XRD (Rigaka), EPMA (JXA-8800R) and SEM techniques, respectively.

RESULTS AND DISCUSSION

Effects of the physical properties of the support on the sorbent

It is obvious that the supports used in the sorbent should ideally possess a large surface area, uniform pore size distribution with sufficiently large pores and large pore volume. But these factors are sometimes contradictory. In order to clarify the effects of the physical properties of the support on the sorbent, two types of γ -alumina, named XP and DS, were used to prepare the sorbent, and the two types of sorbents prepared were correspondingly named XP-sorbent and DS-sorbent. The physical properties of the different supports are summarized in Table 1.

Figure 3 shows the desulfurization efficiency of the XP-sorbent and the DS-sorbent. As shown in Fig. 3, the SO₂ sorption capacity of the DS-supported copper oxide sorbent is higher than that of the XP-supported one.

Sorbents with a large surface area support generally have a correspondingly high loading of copper oxide and consequently a high SO_2 sorption capacity. The surface area of the XP support is larger than that of the DS support, as shown in Table 1, but the SO_2 sorption

Table 1. Physical Properties of XP Support and DS Support.

Support name	BET surface area (m ² /g)	Mean pore size (Å)	Pore volume (cm ³ /g)
XP	269.3	38.6	0.26
DS	146.8	89.7	0.33



Figure 3. Desulfurization efficiency of the different sorbents.

capacity of XP-sorbent is lower than that of DS-sorbent. This may be because the median pore size of DS support is larger than that of XP support. As a result, the dispersion of the active copper oxide coated on the surface of the DS support is much more uniform and the SO_2 gas diffuses into the pores of the DS-sorbent more easily. In order to confirm this hypothesis, the two types of sulfated sorbent particles were analyzed by the EPMA technique. Figure 4 illustrates the superficial element dispersion of the sulfated XP-sorbent and DS-sorbent along the radial direction on the sorbent particle.

As shown in Fig. 4(a), it is obvious that the amount of elemental sulfur dispersed on the XP-sorbent gradually decreases from the surface to the side of the pellet. But in Fig. 4(b), the dispersion of elemental sulfur on the DS-sorbent is more uniform. Figure 4 indicates that SO_2 gas could easily diffuse into the inner core of the DS-sorbent and react with the copper oxide, but the diffusion of SO₂ gas in the XP-sorbent is more difficult. As a result, the SO₂ sorption capacity of the XP-sorbent, in spite of having a larger surface area, is lower than that of the DS-sorbent. These complex results show that not only a large surface area of the support is needed for the sorbent but also the appropriate pore size of the support is necessary for the high SO₂ sorption capacity of the sorbent. The large pore size of the support is favorable for the dispersion of the active copper oxide on the sorbent and for the SO_2 gas diffusion in the sorbent.



Figure 4. EPMA analysis of the XP-sorbent and DS-sorbent.. This figure is available in colour online at www.apjChemEng.com.

So the γ -alumina used as support of the sorbent should have both a large surface area and an ideal pore size.

Effects of concentration of the impregnation solution on the sorbent

In the preparation of the sorbent for flue gas desulfurization, the active copper oxide coated on the surface of the γ -alumina support in the monolayer or submonolayer form is highly desired in order to maximize the amount of active species that can react with SO₂ and enhance the absorption capacity of SO₂ of the sorbent, according to Xie and Tang (1990) and Strohmeier et al. (1985). The loading amount of active copper oxide for the monolayer or submonolayer coverage is mainly dependent on the pore characteristics of the support and the factors of the wet impregnation process, especially, the concentration of impregnation solution. In order to examine the effects of the concentration of the impregnation solution on the SO2 sorption capacity of the sorbent, several Cu(NO₃)₂ solutions with various concentration were used as the impregnation solution to prepare the DS γ -alumina-supported copper oxide sorbents. Figure 5 illustrates the absorptivity of sulfur dioxide of the sorbents impregnated with different concentrations of $Cu(NO_3)_2$ solution.

The absorptivity of sulfur dioxide (η) is defined as:

$$\eta = \frac{C_{\rm i} - C_{\rm o}}{C_{\rm i}} * 100\%$$

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Figure 5. Desulfurization efficiency of different concentrations of the sorbent.

where C_i and C_o are the initial and final concentrations of SO₂, respectively.

As shown in Fig. 5, the SO₂ sorption capacity of the sorbent impregnated with 2 mol/l Cu(NO₃)₂ solution is the highest among these sorbents. The results suggest that the active copper oxide coated on the sorbent impregnated with 2 mol/l Cu(NO₃)₂ solution may be in the form of a monolayer or submonolayer. As the concentration of Cu(NO₃)₂ solution exceeds 2 mol/l, the coating amount of the active copper oxide is more than that required for a monolayer coating.

The amount of monolayer dispersion with CuO is $0.19 \text{ g}/100 \text{ m}^2$. The theoretical amount of CuO is lower

than 31.48%, when the surface area of the Al_2O_3 support is 165.68 m². In the experiment, the amount of CuO is 6.2% in the sorbent that was impregnated with the 2 M solution. The amount of CuO is 12% in the solution of 4 M. We feel that the CuO is coated on the support in the form of a monolayer or a submonolayer. It can be confirmed by the XRD technique.

According to the monolayer dispersion theory (Friedman et al., 1978; Sirvaraj and Kantarao, 1988; Xie and Tang, 1990), if the active species is coated on the surface of the support in the form of a monolayer or a submonolayer coverage, the active species phase will not be detected by XRD. Otherwise, isolated active species in the crystalline form will deposit on the surface of the active monolayer species, which can be detected by XRD. For exploring the form of the active copper oxide coated on different sorbents and examining the effects of concentration of the impregnation solution on the coating amount of the active copper oxide, the pure DS alumina (base), the sorbent impregnated with 2 mol/l $Cu(NO_3)_2$ solution (2 M) and sorbent impregnated with 4 mol/l Cu(NO₃)₂ solution (4 M) were used as examples and analyzed by the XRD technique. The analysis results of XRD are shown in Fig. 6.

In Fig.6, the X-ray diffraction characteristic peaks of the pure DS alumina (base) appear at $2\theta =$ 44.0°, 53.7° and 79.5°. The characteristic peaks of copper oxide, which appear at $2\theta = 32.5^{\circ}$, 41.5°, 45.6°, 57.3° and 72.9°, occur in the XRD powder patterns of the sorbent impregnated with 4 mol/l Cu(NO₃)₂ solution (4 M), and there are no detectable XRD peaks of copper oxide in sorbent impregnated with 2 mol/l Cu(NO₃)₂ solution (2 M). It can be inferred from these results that the active copper oxide coated on the 2 M sorbent is almost in the form of a monolayer or a submonolayer coverage. The coating amount of active copper oxide on the 4 M sorbent exceeds that required for monolayer coverage, and the copper oxide may be in the crystalline form. So the optimum concentration of impregnation solution is very important for the dispersion form of the active species coated on the support. The impregnation solution used in the wet impregnation process should have the optimum concentration so that the active copper oxide is coated on the support in the form of a monolayer or a submonolayer, and consequently the sorbent has a high SO₂ sorption capacity.

Characteristics of the sorbent in the sulfation-regeneration cycles

In order to further examine the SO_2 sorption capacity and the stability of the optimum alumina-supported copper oxide sorbent, the DS γ -alumina was impregnated with the 2 mol/l Cu(NO₃)₂ solution to acquire the optimum sorbent, and then four cycles of sulfation-regeneration reaction experiments were carried out with the sorbent. The active copper oxide loading in the prepared sorbent was about 6 wt% on the basis of dry alumina.

Figure 7 illustrates the desulfurization efficiency of the sorbent of the first, second and fourth sulfation cycles. The maximum desulfurization efficiency of the sorbent can reach 90%, and the SO₂ sorption capacity of 100 g sobent is about 4 g SO₂ for single sulfation cycle by calculation. As shown in Fig. 7, the SO₂ sorption capacity of the sorbent for the first sulfation cycle is higher than that of the subsequent cycles. However, the SO₂ sorption capacity of the fourth sulfation is similar to that of the second sulfation. These results suggest to some extent that the characteristics of the sorbent tend to stabilize.

Figure 8 shows the SO₂ concentration curves measured at the exit of the regeneration reactor for four cycles of the regeneration reaction. By applying integral calculus to these curves, the released volumes of SO₂ in the four cycles of regeneration reaction can be found, that is, 0.63 1 (1st), 0.57 1 (2nd), 0.59 1 (3rd) and 0.56 1 (4th), respectively. The sorbent in the first



Figure 6. XRD comparison of different sorbents.

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Figure 7. Desulfurization efficiency of the sorbents during sulfation–regeneration cycles.



Figure 8. release of SO₂ from sorbents during the sulfation-regeneration cycles.

sulfation-regeneration circulation is fresh, and both the SO₂ sorption capacity and the regenerative capability of the sorbent are the best. Because the aluminum sulfate cannot be regenerated by CH₄ after the first cycle, the released SO₂ volume of the sorbent in the later three cycles (2nd, 3rd and 4th) of the regeneration reaction are approximate, which indicates the regenerative process with methane is stable.

In general, the pores of the sorbent can be sintered during the cycles of sulfation-regeneration reactions. For observing the changes of the pore structure and examining the thermal stability of the sorbent, the pore texture data of the sorbent samples, including the fresh sorbent, the first sulfation-regeneration sorbent and the fourth sulfation-regeneration sorbent, were measured by a nitrogen adsorption porosimeter (Micromeritics, Asap2000). The pore texture data, including BET surface area, pore volume and median pore size, are summarized and compared in Table 2.

As shown in Table 2, the pore structure data of the selected sorbents fluctuate owing to the effects of both sintering and chemical reaction. However, the pore texture data of the sorbent after the sulfation-regeneration cycles is still fairly similar to that of the fresh sorbent. The results show that in the four sulfation-regeneration

Table 2. Pore texture data of different sorbent samples.

Sorbent samples	Fresh	First sulfation– regeneration	Fourth sulfation– regeneration
Median pore size (Å)	64.26	67.91	61.92
Pore volume (cm ³ /g)	0.24	0.25	0.26
BET surface area (m ² /g)	170.58	163.37	176.87

reaction cycles the sorbent did not sinter and that the sorbent has good thermal stability under the experimental conditions. The same conclusion can be drawn from the SEM photographs of the selected sorbents, as shown in Fig. 9. It can be inferred from these results that the pore structure of the sorbent remains essentially unchanged for more sulfation-regeneration cycles.

The copper oxide active species coated on the support are likely to coalesce and lose their activity during the cycles of sulfation-regeneration reactions. In order to examine whether the copper oxide coated on the sorbent coalesce or not, the fresh sorbent is compared with that after the sulfation-regeneration cycles by means of the XRD technique, as shown in Figure 10.

In Figure 10, there are no detectable XRD characteristic peaks of copper oxide in both the fresh sorbent and the fourth sulfation-regeneration sorbent. The X-ray diffraction peaks of the fourth sulfation-regeneration sorbent are quite similar to those of the fresh sorbent. It can be concluded from the results that coalescence did not occur during the four cycles of sulfation-regeneration reactions under the experimental conditions, and the copper oxide active species remain in a highly dispersed state. During the first cycle of sulfation-regeneration, some sulfate species were formed, and these sulfate species were not eliminated during the first regeneration step. The presence of these sulfate



a. fresh sorbent

b. sorbent after the first cycle



c. sorbent after the fourth cycle

SEM photograph of the different sorbents during sulfa-Fiaure 9. tion-regeneration cycles (×5000).



Figure 10. XRD comparison between the fresh sorbent and sorbent after the fourth cycle.

species on the alumina probably inhibits the sintering of the copper oxide particles, thus limiting the surface reconstruction. Therefore, the sorbent shows good thermal and chemical stability during the four cycles of sulfation-regeneration. This result is consistent with the conclusions of the work reported by Waqif *et al*. (1991).

CONCLUSIONS

The γ -alumina (DS)-supported copper oxide sorbent prepared by the wet impregnation method exhibits a high SO₂ sorption capacity due in part to the good physical properties of the DS support, which has both large surface area and ideal pore size. Among the impregnation solutions with different concentration of $Cu(NO_3)_2$, the sorbent impregnated with 2 mol/l $Cu(NO_3)_2$ solution exhibited the highest SO₂ sorption capacity. It was found that the active copper oxide coated on the sorbent impregnated with 2 mol/l Cu(NO₃)₂ solution was in the form of a monolayer or a submonolayer. As the concentration of the $Cu(NO_3)_2$ solution exceeded 2 mol/l, the coated copper oxide might be in crystalline form and could be detected by XRD. The DS γ alumina-supported copper oxide sorbent showed good thermal and chemical stability in the four cycles of sulfation-regeneration reaction under the experimental conditions. The pore of the sorbent did not sinter and the copper oxide active species remained in a highly dispersed state instead of coalescing during the four cycles of sulfation-regeneration reaction.

However, the prepared copper oxide sorbent need to be evaluated under the conditions of more sulfation-regeneration cycles and the real flue gas. Further studies are needed to fully understand the characteristics of the DS γ -alumina-supported copper oxide sorbent by means of advanced techniques.

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