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Catalytic reduction of SO_2 over Sn–Zr based catalysts for DSRP under high pressure

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ABSTRACT

The catalytic activity of Sn–Zr based catalysts and the production behavior of COS during SO_2 reduction were investigated for the catalytic reduction of SO_2 under high pressure condition in this study. The SnO_2, ZrO_2 and SnO_2–ZrO_2 used as the catalysts were prepared by the precipitation and co-precipitation methods. The activity tests for the Sn–Zr based catalysts were carried out in the range of 200–450 °C at atmospheric pressure and 20 atm. CO was also used as the reducing agent for the reduction of SO_2. The catalytic activity and a surface area of the SnO_2–ZrO_2 prepared by the co-precipitation methods were higher than that of the SnO_2 and SnO_2–ZrO_2 catalysts prepared by the physical mixing. It was concluded that the surface area of SnO_2–ZrO_2 catalyst prepared by the co-precipitation increased due to the formation of the lattice defects. A high yield of elemental sulfur was also maintained for 50 h in the durability test. Carbonyl sulfide is produced by the reaction between CO and the elemental sulfur produced during the reduction of SO_2, and was also produced by the gas–solid reaction between the metal sulfide and CO. The selectivity of COS was high at the intermediate level of conversion of SO_2. Since the vapor pressure of elemental sulfur decreased with increasing pressure, the production of COS was reduced to a greater extent at the high SO_2 conversion observed at high pressure. From these results, it was confirmed that SnO_2–ZrO_2 is a suitable catalyst for the catalytic reduction of SO_2 at high pressure.

1. Introduction

IGCC (integrated gasification combined cycle) systems can produce various chemicals and energy resources such as DME (dimethyl ether), synthetic gas, and electric power by gasifying coal. The coal gasification process has the high energy efficiency than the coal fire power plant, and also emits the low carbon dioxide. Therefore, these systems will contribute to the development and production of highly efficient and environmental friendly energy resources. In IGCC systems, sulfur compounds such as hydrogen sulfide and carbonyl sulfide can be produced, due to the sulfur component in coal, and the resulting hydrogen sulfide can be treated using the hot gas desulfurization process with a sorbent. An SO_2 removal process is necessary for IGCC systems, because SO_2 is produced in the regeneration process of the sulfide sorbent. Sulfur dioxide is also a toxic and corrosive sulfur compound, which damages the health, corrodes equipment, generates acid rain, and pollutes and acidifies the soil. As environmental regulations are becoming increasingly strict, there is a pressing need to develop cleanup technologies for the control of SO_x emissions. SO_2 can be removed by recovering the elemental sulfur produced in the SO_2 reduction process using a reducing agent. Various reducing agents such as hydrogen, carbon monoxide, methane, and carbonaceous materials have been used for the reduction of SO_2 [1–5]. Carbon monoxide is used in the DSRP (direct sulfur recovery process) through the reaction (1).

$$SO_2 + 2CO \rightarrow 2CO_2 + S \quad (1)$$

$$S + CO \rightarrow COS \quad (2)$$

However, using CO used as the reducing agent can lead to the production of COS as through reaction (2), which is endothermic. Therefore, the catalyst used for SO_2 reduction should have high catalytic activity at the low temperature for obtaining a high yield of elemental sulfur.

In order to find a suitable catalyst for DSRP, Sn–Zr based catalysts were prepared and their catalytic activity was examined in this study. DSRP is the unit process of the desulfurization process, and is necessary for the conversion of the SO_2 produced in the regeneration process to elemental sulfur [6]. The characteristics of the catalytic reaction for the Sn–Zr based catalysts were researched in our previous study [7,8]. SnO_2 and ZrO_2 catalysts occurs at the different catalysis for the reduction of SO_2. SO_2 is conversed to elemental sulfur through redox mechanism over SnO_2 and COS

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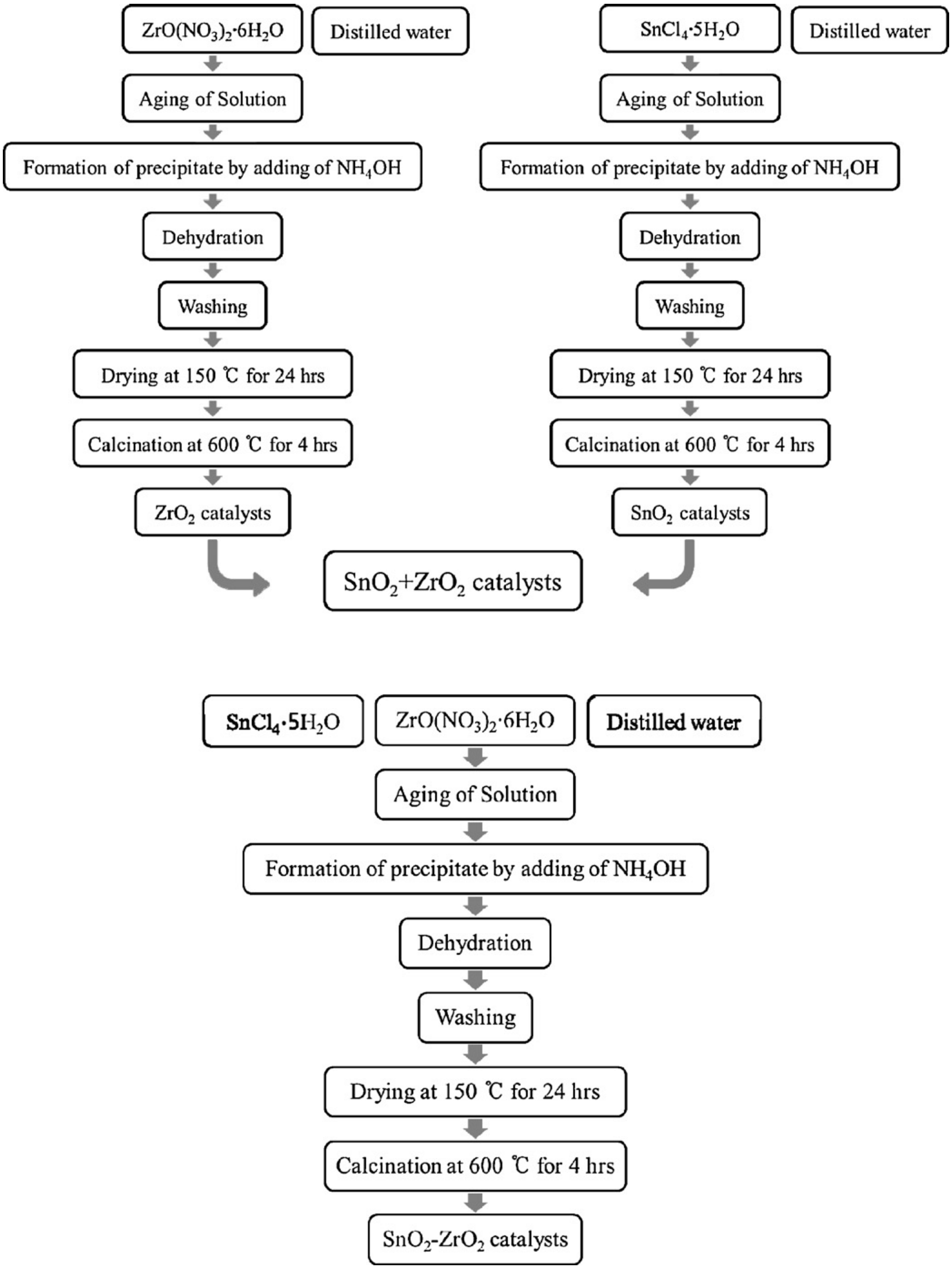

Fig. 1. Procedure for preparation of Sn–Zr based catalysts by co-precipitation method.

intermediate mechanism over ZrO_2, respectively. It was confirmed in this study and our previous study that the hybrid catalyst, which is mixed with SnO_2 and ZrO_2, has the high catalytic activity for the reduction of SO_2. It was expected that redox property of SnO_2–ZrO_2 catalyst prepared by the co-precipitation method is enhanced due to the improvement of oxygen mobility by the defect formed in the lattice of catalyst. The defect in lattice of metal oxide structure can be formed by the replacement of zirconium ions into MeO_2 (metal oxide) structure [9,10].

Meanwhile, most of the experiments in our previous study were carried out at atmospheric pressure. It could be expected with Le Chatelier's principle that the equilibrium conversion on the reduction of SO_2 is higher at high pressure condition than at an atmospheric pressure due to the low molar volume of product gases than that of the reactant gases. The content of reactant gases adsorbed over the active sites of the catalyst also increased with increasing the pressure in the catalytic reactor. If the product gases well desorbs from the active sites over the surface of catalyst, the high reaction rate can be obtained at high pressure. Therefore, the activity tests for the Sn–Zr based catalysts were performed at a high pressure of 20 atm as the operating conditions desired for the DSRP in this study. The selectivity of the products, such as elemental sulfur and carbonyl sulfide, was also researched at high pressure.

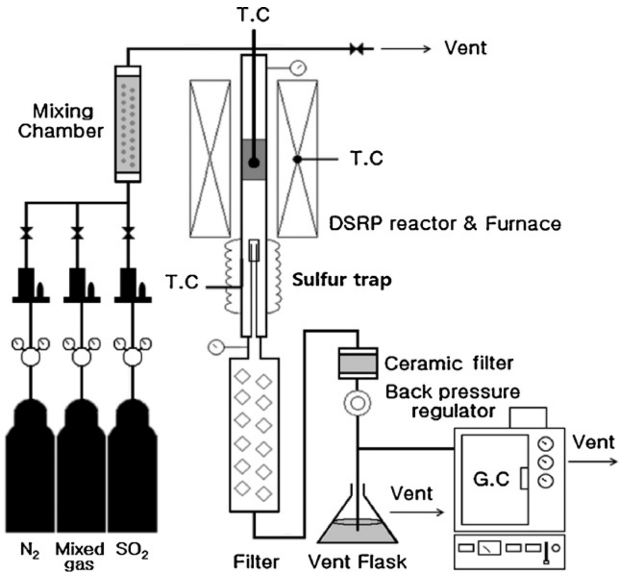

Fig. 2. Schematic diagram of experimental apparatus used for SO_2 catalytic reduction under conditions of high pressure.

2. Experiment

2.1. Preparation of Sn–Zr based catalysts

In this study, Sn–Zr based catalysts for the catalytic reduction of SO_2 were prepared by the precipitation and co-precipitation methods, as shown in Fig. 1. Tin chloride pentahydrate ($SnCl_4{\cdot}5H_2O$, Aldrich) and zirconyl nitrate hydrate ($ZrO(NO_3)_2{\cdot}6H_2O$, Aldrich) were used as the precursors of the main active materials for the Sn and Zr components, respectively. These two precursors were prepared in the solution state with distilled water, and were mixed at a Sn/Zr molar ratio of 2/1. The ammonia water (NH_4OH) at a concentration of 15.0 M used as the precipitator was slowly added to the mixture solution until a pH of 9 was obtained. The precipitate of the solid phase obtained by the precipitation was washed with the distilled water, and was calcined for 4 h at 600 °C after dried for 24 h at 150 °C. The prepared Sn–Zr based catalysts were used for the catalytic activity tests. As shown in Fig. 1, the SnO_2 and ZrO_2 catalysts were prepared by precipitation. The other SnO_2–ZrO_2 catalyst was prepared by the physical mixing of the SnO_2 and ZrO_2 prepared in this study.

2.2. Catalytic activity tests for DSRP

The high pressure experimental setup for SO_2 catalytic reduction over the Sn–Zr based catalysts consisted of a gas flow controller, catalytic tubular reactor, vertical type furnace, sulfur trap and gas chromatograph, as shown in Fig. 2. The composition of the reactants was controlled by a mass flow controller (Brooks MFC 5850E) and the molar ratio of $[CO]/[SO_2]$ (40,000 ppmv CO and 20,000 ppmv SO_2 in N_2 balance) contained in the reactant gas was fixed at 2.0. In this process, CO was used as the reducing agent for the reduction of SO_2. The reaction temperature was controlled in the range of 200–450 °C by a temperature controller and thermocouple inserted in the catalyst bed. The pressure in the catalytic reactor was kept at atmospheric pressure or 20 atm by a back pressure regulator. A stainless steel tube (Inconel 600) with an outside diameter of 1/2 inch was used as the catalytic reactor and 6 g of the catalyst was packed in the center of the reactor. A sulfur trap was vertically connected in the reactor outlet and was used for the collection of the elemental sulfur produced by the catalytic reduction of SO_2. The gas composition discharged in the reactor outlet was analyzed by a gas chromatograph (Shimadzu GC-8A) equipped with a thermal conductivity detector and a column consisting of Hyasep Q(8 ft) and Porapak T(2 ft). Hydrogen was used as a carrier gas.

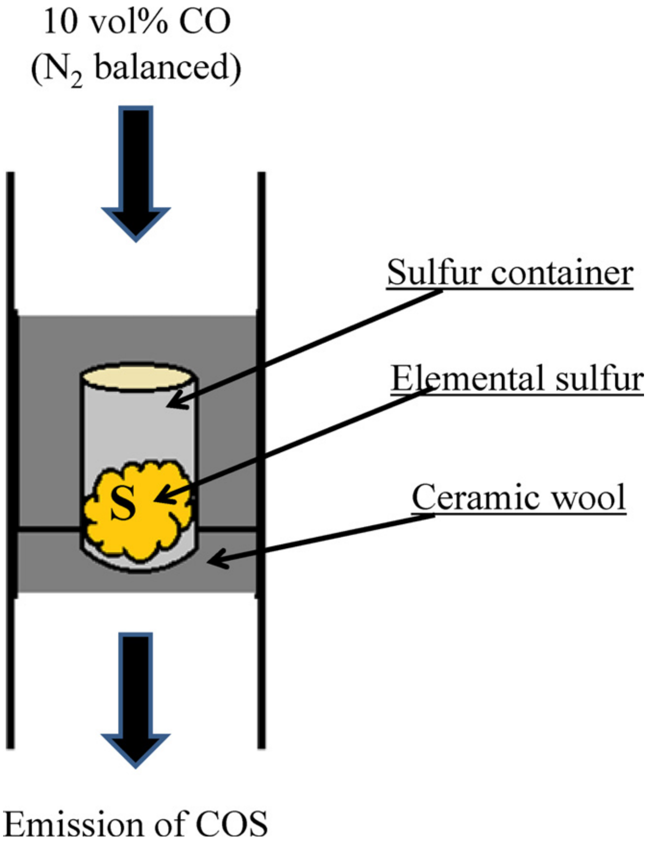

Fig. 3. Schematic diagram of experimental apparatus used for COS production by reaction of elemental sulfur with CO.

2.3. Variation of COS production with pressure

In order to investigate the characteristics of COS production from the reaction between CO and elemental sulfur, the experiments for COS production were carried out at various temperatures and pressures in the catalytic reactor. A small sulfur container, which is a glass vessel packed with elemental sulfur, was fitted in the center of the tubular reactor used for the supply of elemental sulfur in the vapor phase, as shown in Fig. 3. The amount of elemental sulfur packed in the sulfur container was 1 g. The reactor was placed in a vertical type furnace and the temperature in the reactor was kept in the range of 200–400 °C by a temperature controller and thermo-couple. The pressure in the reactor was also controlled to 1, 10, or 20 atm. 10 vol% of CO diluted with nitrogen gas was flowed at a flow rate of 100 ml/min into the tubular reactor. Finally, the concentration of COS exhausted from the reactor was measured by a gas chromatograph.

3. Results and discussion

3.1. Activity tests for the SnO_2 and ZrO_2 catalysts

The catalytic activity of SnO_2 prepared by the precipitation method for the catalytic reduction of SO_2 was investigated. The catalytic activity tests for SnO_2 were carried out in the range of 200–450 °C at atmospheric pressure and 20 atm. The molar ratio

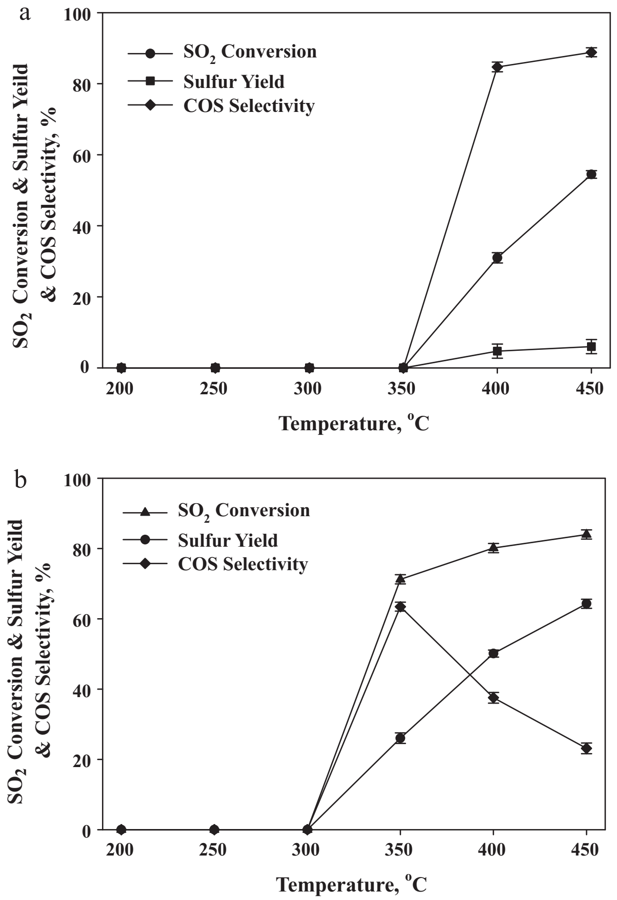

Fig. 4. Effect of reaction temperature for SO_2 reduction with CO over SnO_2 catalyst under conditions of (a) atmospheric pressure and (b) 20 atm.

of [CO]/[SO_2] and the space velocity for the activity test were 2.0 and 10,000 ml/$g_{\text{-cat}}$ h, respectively. In the activity tests performed at atmospheric pressure, the reduction of SO_2 started at 350 °C and the SO_2 conversion increased with increasing temperature, as shown in Fig. 4(a). The conversion of SO_2 decreased to 3% at 350 °C and increased to 35% and 53% at 400 °C and 450 °C, respectively. The selectivity of COS increased by 85% at 400 °C and the elemental sulfur yield decreased due to the high selectivity of COS. However, in the activity tests performed at a pressure of 20 atm, the conversion of SO_2 increased to 70% at 350 °C, as shown in Fig. 4(b). The elemental sulfur yield was also improved, due to the low production of COS.

The activity tests for ZrO_2 prepared by the precipitation method were carried out under the same experimental conditions as those used for the activity tests of the SnO_2 catalyst. The conversion of SO_2 increased with increasing temperature and was approximately 90% at 450 °C at atmospheric pressure, as shown in Fig. 5(a). A high yield of elemental sulfur produced by the reduction of SO_2 was obtained, due to the low production of COS. The selectivity of COS was 15% and 8% at 400 and 450 °C, respectively. Meanwhile, the conversion of SO_2 and the selectivity of COS at 300 and 400 °C at a pressure of 20 atm were higher than those at atmospheric pressure, as shown in Fig. 5(b). However, the yield of elemental sulfur increased at 20 atm because the increase of the SO_2 conversion was greater than that of the COS selectivity.

The formation of COS can occur by two kinds of reactions. One is the gas and solid reaction between CO and metal sulfide [11,12]. The other is the reaction of CO and elemental sulfur in the vapor phase [10]. If we assume that COS is formed by the reaction of CO and elemental sulfur in the vapor phase, the highest production of COS can be obtained at the maximum partial pressure of both CO and elemental sulfur produced by the reduction of SO_2 over the catalyst bed. Therefore, the maximum selectivity of COS can be obtained at an intermediate level of SO_2 conversion. The curves relating (X_{SO_2} vs. S_{COS}) between the conversion of SO_2 and the selectivity of COS obtained from the activity tests for SnO_2 and ZrO_2 are shown in Fig. 6. The selectivity of COS was lowered at high and low conversions of SO_2, but was high at intermediate levels of

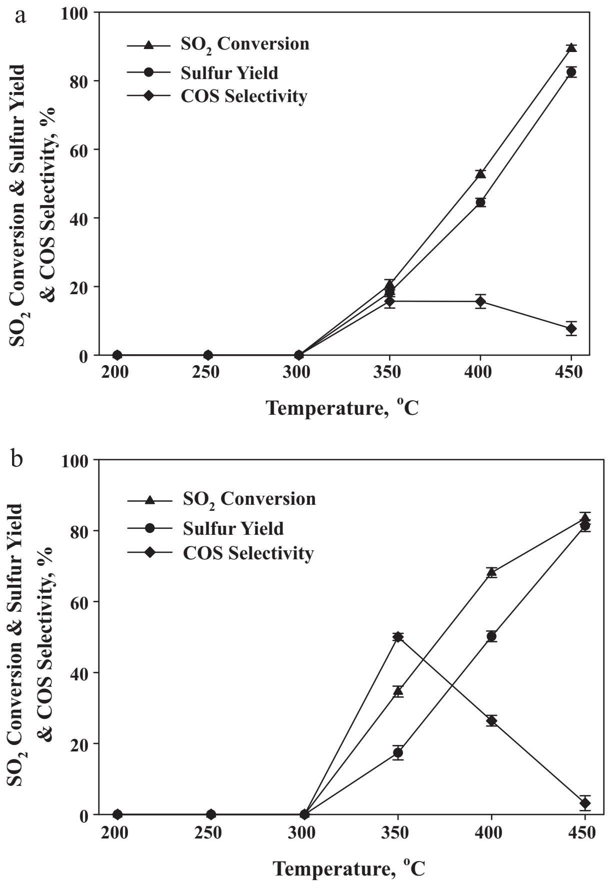

Fig. 5. Effect of reaction temperature for SO_2 reduction with CO over ZrO_2 catalyst under conditions of (a) atmospheric pressure and (b) 20 atm.

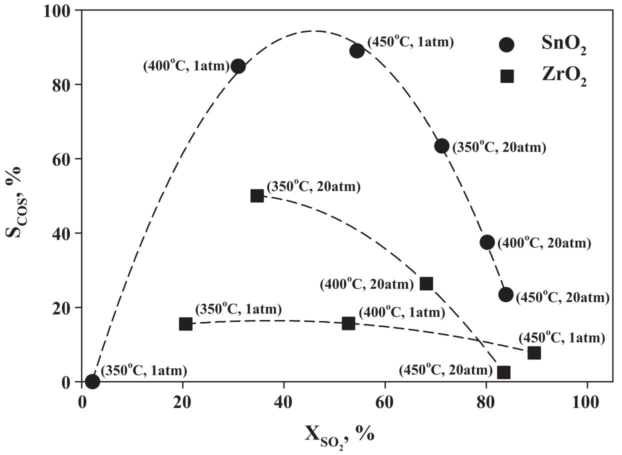

Fig. 6. Relation curves for conversion of SO_2 vs. selectivity of COS obtained from SO_2 reduction with CO over (a) SnO_2 and (b) ZrO_2 catalyst under conditions of atmospheric pressure and 20 atm.

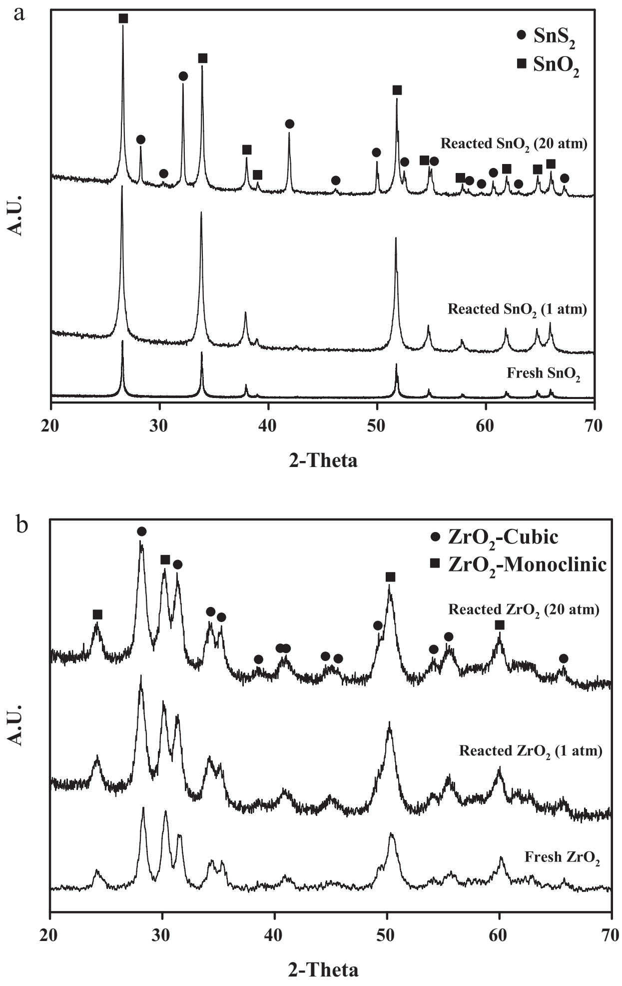

Fig. 7. XRD patterns of (a) ZrO_2 and (b) SnO_2 after reaction at atmospheric pressure and 20 atm.

SO_2 conversion. These results mean that COS is synthesized by the reaction between the un-reacted CO and elemental sulfur produced during the reduction of SO_2.

It had been reported that SO_2 adsorbed over SnO_2 catalyst is reduced elemental sulfur by redox mechanism [7]. Redox mechanism is consisted of two reactions over the metal oxide catalyst. That is the reduction and oxidation of the lattice oxygen in the metal oxide catalyst. The lattice oxygen is reduced with CO and the lattice oxygen vacancy created by the reduction is oxidized with SO_2. Therefore, COS cannot be produced by redox mechanism. However, COS was observed the most activity tests in this study. The selectivity of COS was also different over SnO_2 and ZrO_2 at intermediate levels of SO_2 conversion. It was concluded that the different selectivity of COS at the same SO_2 conversion is considered with to the different catalytic activities of the two catalysts. It is known that the synthesis of COS can occur by both non-catalytic and catalytic reactions. The catalytic reaction occurs by reactions (3) and (4) [12,14–16].

$$\text{M-s} + \text{S} \rightarrow \text{M–S} \tag{3}$$

$$\text{M–S} + \text{CO} \rightarrow \text{COS} \tag{4}$$

The peaks at 2θ values of 28.3°, 32.2°, 41.9°, and 49.9° in the XRD pattern of SnO_2 after the activity tests correspond to tin sulfide (SnS_2), as shown in Fig. 7. However, no XRD peaks related to metal sulfides were detected in the case of ZrO_2. Therefore, it was concluded that some of the SnO_2 used as the catalyst was converted into tin sulfide during the catalytic reduction of SO_2 and that the sulfur contained in the tin sulfide was converted to COS by the CO used as a reducing agent. In our previous study, it had been suggested that SO_2 over SnO_2 catalyst is catalytic reduced to elemental sulfur by redox mechanism [7]. However, it was confirmed by XRD that some SnO_2 catalyst prepared in this study is conversed to SnS_2. The sulfur contained in SnS_2 can be formed COS with CO. Thus, it was concluded that the reduction of SO_2 over SnO_2 is not only high in the conversion of SO_2 but also in the selectivity of COS because two reactions, that is the sulfidation of SnO_2 and the reduction of SO_2 by redox mechanism, occurs.

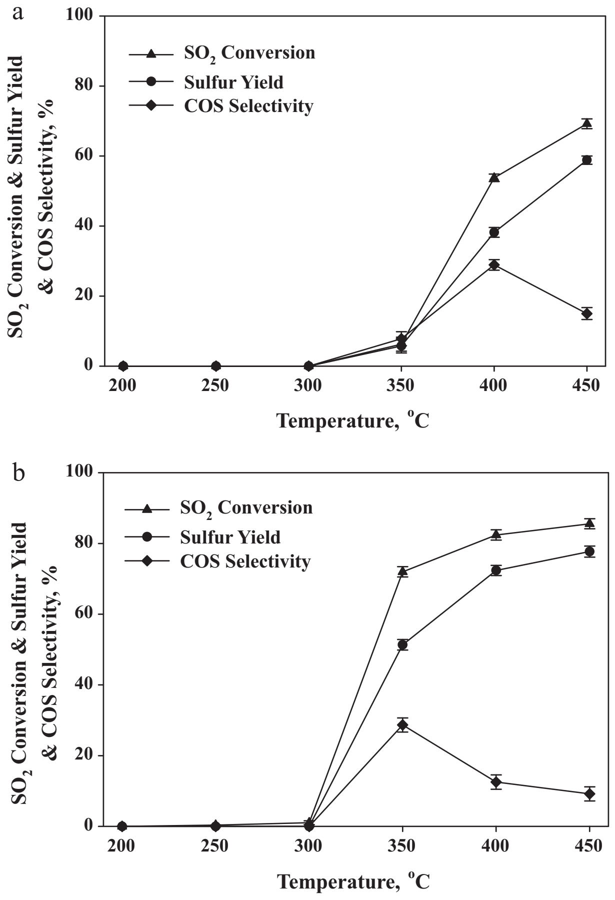

Fig. 8. Effect of reaction temperature for SO_2 reduction with CO over SnO_2–ZrO_2 catalyst prepared by physical mixing method under conditions of (a) atmospheric pressure and (b) 20 atm.

3.2. The activity tests for the SnO_2–ZrO_2 catalysts

In the activity tests, the activity of SnO_2 was higher than that of ZrO_2 at 350 °C and 20 atm. However, the yield of elemental sulfur was low due to the high production of COS. In order to improve the yield of elemental sulfur, the Sn–Zr based catalysts were prepared by the physical mixing and co-precipitation methods in this study. The molar ratio of Sn/Zr contained in the catalysts was fixed at 2.0, as in our previous study [7,17]. The activity tests for the Sn–Zr based catalysts were carried out in the range of 200–450 °C at atmospheric pressure and 20 atm. The molar ratio of [CO]/[SO_2] and space velocity were fixed at 2.0 and 10,000 ml/$g_{\text{-cat}}$ h, respectively.

The conversion of SO_2 in the reduction of SO_2 over the SnO_2–ZrO_2 catalyst prepared by the physical mixing method was higher than that over the SnO_2 catalyst and a high yield of elemental sulfur was obtained due to the low selectivity of COS, as shown in Fig. 8(a). As shown in Fig. 8(b), the conversion of SO_2 and the

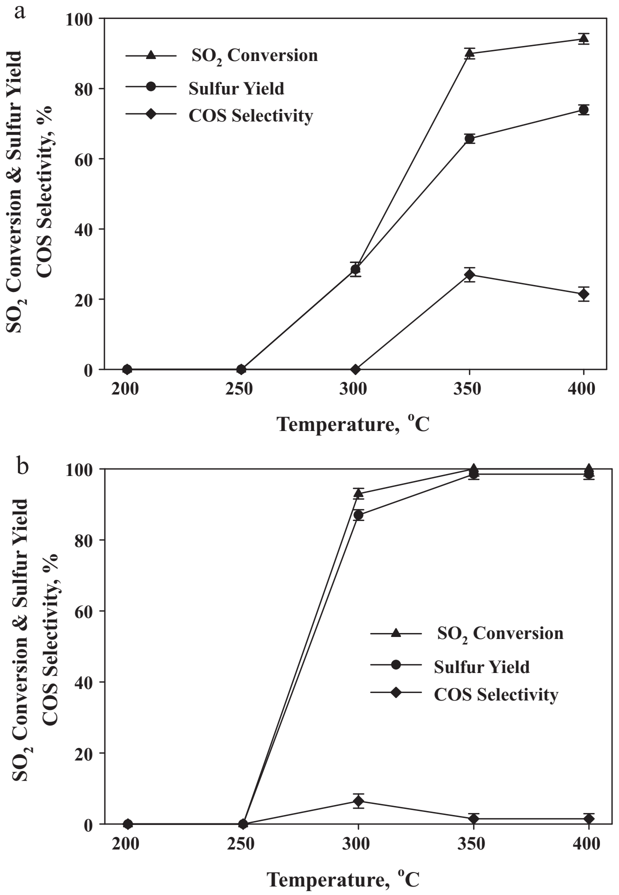

Fig. 9. Effect of reaction temperature for SO_2 reduction with CO over SnO_2–ZrO_2 catalyst prepared by co-precipitation method under conditions of (a) atmospheric pressure and (b) 20 atm.

yield of elemental sulfur also increased to 73% and 52% at 350 °C at a pressure of 20 atm, respectively. It was concluded that the selectivity of COS over the SnO_2–ZrO_2 catalyst is low than that over SnO_2, because COS formulated during the reduction of SO_2 over SnO_2 site is conversed to elemental sulfur with SO_2 fed as the reactant by COS intermediate mechanism in Lewis and Brönsted acid sites over ZrO_2 [18,19].

Table 1
Surface area of Sn–Zr based catalysts measured by BET method.

Catalysts	Surface area (m^2/g)
SnO_2	22.2
ZrO_2	43.1
SnO_2 + ZrO_2[a]	29.3
SnO_2–ZrO_2[b]	48.0

[a] The catalyst prepared by physical mixing method.
[b] The catalyst prepared by co-precipitation method.

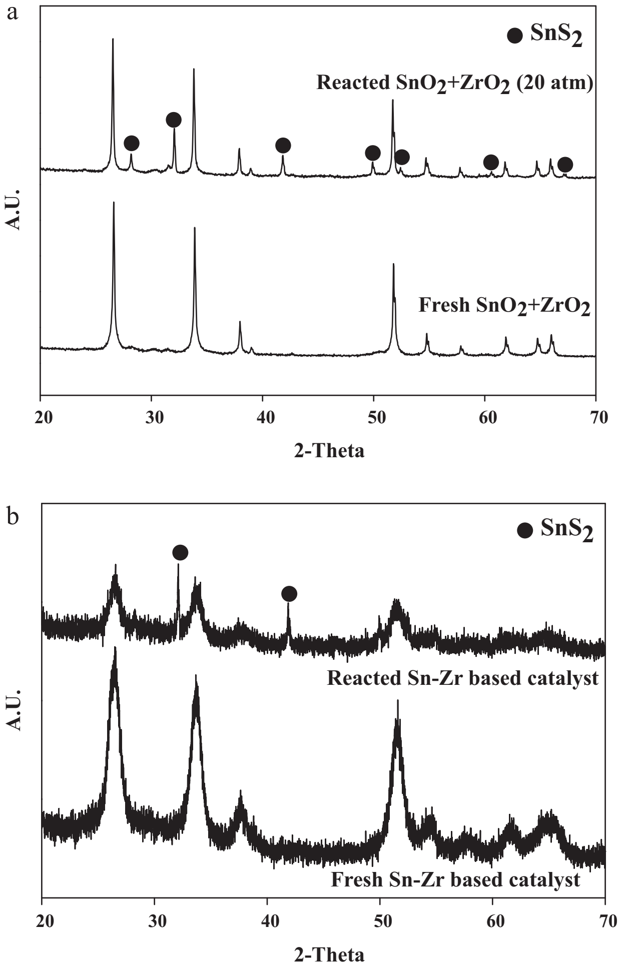

Fig. 10. XRD patterns of SnO_2–ZrO_2 catalysts prepared by (a) physical mixing and (b) co-precipitation methods, fresh and after reaction at 20 atm.

The conversion of SO_2 increased to 90% at 350 °C and atmospheric pressure with the use of the SnO_2–ZrO_2 catalyst prepared by the co-precipitation method, as shown in Fig. 9(a). A very small amount of COS was observed in the reactor outlet. Both the conversion of SO_2 and the yield of elemental sulfur reached approximately 100% above 350 °C at a pressure of 20 atm, as shown in Fig. 9(b). The surface area of the catalysts measured by the BET method was shown in Table 1. The surface area of SnO_2 and ZrO_2 prepared by the precipitation method was 22.2 m^2/g and 43.1 m^2/g, respectively. The surface area of SnO_2–ZrO_2 catalyst prepared by the physical mixing of SnO_2 and ZrO_2 was 29.3 m^2/g. The surface area of the SnO_2–ZrO_2 catalyst prepared by the co-precipitation method was also higher than that of the catalyst prepared by the physical mixing method. Therefore, it was concluded that the activity of SnO_2–ZrO_2 prepared by the co-precipitation method was enhanced by the synergetic effect afforded by the hybridization of SnO_2 and ZrO_2 and the improvement of its surface area. The catalyst prepared by the co-precipitation is the solid solution of SnO_2 and ZrO_2. The defect in the lattice of this catalyst can be also formed due to Zr ions replaced in the lattice of SnO_2. It had been known that the defects formed in SnO_2–ZrO_2 catalyst enhance the oxygen mobility in the lattice of metal oxide. Therefore, the catalytic activity of SnO_2–ZrO_2 catalyst can be improved due to the enhancement of redox property resulted from the increased oxygen mobility. It had been confirmed in H_2-TPR tests of our previous study that the redox property of SnO_2–ZrO_2 prepared by the co-precipitation method was improved than other catalysts [7]. Thus, it was concluded that the high conversion of SO_2 and the low selectivity of COS was obtained due to both the increase of surface area and redox property of SnO_2–ZrO_2. Specially, its catalytic activity for the reduction of SO_2 was increased at high pressure. XRD patterns of SnO_2–ZrO_2 catalysts prepared by the physical mixing and the co-precipitation was shown in Fig. 10. While XRD pattern (Fig. 10(a)) of the catalyst prepared by the physical mixing shown the high intensity peaks of SnO_2 and the low intensity peaks of ZrO_2, that of SnO_2–ZrO_2 catalyst prepared by co-precipitation shown the amorphous phase as shown Fig. 10(b). It was concluded that the amorphous phase shown in XRD pattern is formed due to the creation of the defects by Zr ions replaced into the lattice of SnO_2. The peaks of SnS_2 over the SnO_2–ZrO_2 catalyst

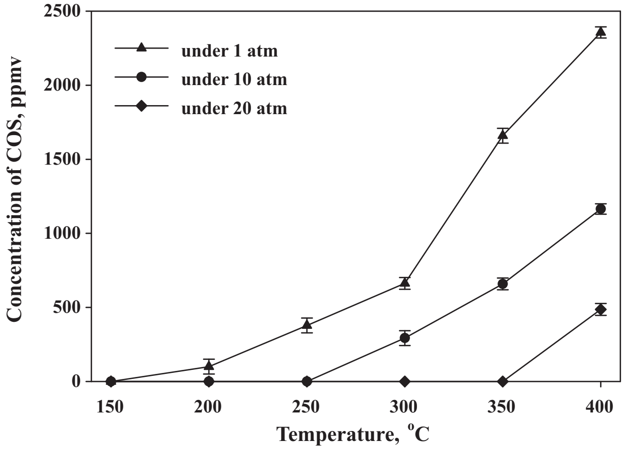

Fig. 11. COS production from reaction of elemental sulfur with CO in temperature range of 200–400 °C and pressures of 1, 10, and 20 atm.

prepared by the co-precipitation also shows the low intensity compared with that over the catalyst prepared by the physical mixing method. These results were well matched with the selectivity of COS obtained on the activity tests of two Sn–Zr-based catalysts.

3.3. *Effect of pressure*

It was confirmed that a high conversion of SO_2 in the reduction of SO_2 over all of the catalysts was obtained by increasing the pressure. The yield of elemental sulfur also increased at high pressure due to the decrease in the production of COS. Commonly, the conversion of the reactants is increased by the increased number of collisions of the reactants at high pressure. If the conversion of SO_2 increases, the production of COS will be decreased due to the increase in the consumption of CO, which is the amount of CO used for the reduction of SO_2. However, the selectivity of COS in the activity tests for ZrO_2 was 17% at a SO_2 conversion of 55% and 28% at a SO_2 conversion of 70%, as shown in Fig. 6. These experiments were performed at 350 °C at both atmospheric pressure and 20 atm. The selectivity of COS at high pressure was higher than that at atmospheric pressure. Meanwhile, the selectivity of COS was 3% at a SO_2 conversion of 85% and 10% at a SO_2 conversion of 90%. In these experiments, the selectivity of COS at atmospheric pressure was lower than that at high pressure. A different trend variation of the selectivity was obtained with the operating pressure was observed in the catalytic reactor at a similar SO_2 conversion. COS is generated by the reaction between CO and the elemental sulfur, which is produced by the reduction of SO_2 [13]. Thus, the production of COS is increased due to the increase in the number of molecular collisions between CO and elemental sulfur at high pressure. However, the elemental sulfur in the vapor phase can be reduced, because its vapor pressure decreases with increasing pressure in the catalytic reactor. In order to investigate the reaction between CO and elemental sulfur, experiments for the production of COS were carried out at various temperatures and pressures in the catalytic reactor. A small sulfur container, which is a vessel packed with elemental sulfur, was placed at the center of the tubular reactor to supply elemental sulfur in the vapor phase, as shown in Fig. 3. 10 vol% of CO diluted with nitrogen gas was flowed into the tubular reactor. The concentration of COS measured in the reactor outlet is shown in Fig. 11. The concentration of COS decreased with increasing pressure, while it decreased with increasing temperature. It was confirmed that the sulfur packed in the container was completely consumed after the reaction under conditions of high temperature and low pressure and that some sulfur remained after the reaction under conditions of low temperature and high pressure. These results mean that the elemental sulfur in the solid phase is well evaporated to sulfur in the vapor phase at high temperature, but is not well evaporated at high pressure. Therefore, the production of COS can be reduced by increasing the pressure in the catalytic reactor used for the reduction of SO_2, due to the effect of the vapor pressure of sulfur. It was also concluded that the effect of the sulfur vapor pressure is accentuated under conditions of a low COS selectivity and high SO_2 conversion.

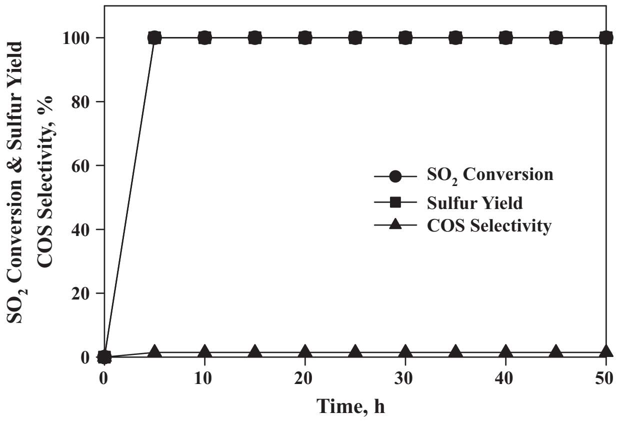

Fig. 12. Durability test for SO_2 catalytic reduction over SnO_2–ZrO_2 catalyst prepared by co-precipitation.

3.4. *The durability of SnO_2–ZrO_2*

The durability test for the SnO_2–ZrO_2 catalysts prepared by the co-precipitation method was performed for 50 h at a temperature of 350 °C and pressure of 20 atm. The molar ratio of $[CO]/[SO_2]$ and the space velocity in the durability test were 2.0 and 10,000 ml/$g_{\text{-cat}}$ h, respectively. The conversion of SO_2 and the yield of elemental sulfur were maintained at approximately 100% and 98% for 50 h, respectively, as shown in Fig. 12. The high durability of the SnO_2–ZrO_2 catalyst prepared by the co-precipitation method was confirmed by the results of this study.

4. Conclusion

The catalytic activity of Sn–Zr based catalysts for the catalytic reduction of SO_2 was investigated in this study. The activity of the hybrid catalysts prepared by the physical mixing and the co-precipitation methods was higher than that of the SnO_2 and ZrO_2 catalysts. Specially, the SnO_2–ZrO_2 catalyst prepared by the co-precipitation method has excellent activity for the catalytic reduction of SO_2 at a temperature of 350 °C and pressure of 20 atm. A high yield of elemental sulfur was also maintained for 50 h in the durability test. It was concluded that the activity of the SnO_2–ZrO_2 catalyst prepared by the co-precipitation was enhanced due to the synergetic effect and increasing a surface area of the catalyst formulated with the solid solution of SnO_2 and ZrO_2.

The production behavior of COS during the reduction of SO_2 was investigated with the change as a function of the temperature and pressure in the activity tests, and as well as the production of COS from CO and elemental sulfur. Carbonyl sulfide is generated by the unreacted CO and elemental sulfur produced during the reduction of SO_2, and is also produced by the gas–solid reaction between the metal sulfide and CO. The amount of COS produced over SnO_2 was higher than that over ZrO_2, because tin oxide can be converted to tin sulfide by elemental sulfur produced during the catalytic reduction of SO_2. A high selectivity of COS was observed at a high partial pressure of CO and vapor phase sulfur which is produced during the reduction of SO_2. Thus, the selectivity of COS was high at an intermediate level of conversion of SO_2. Since the vapor pressure of elemental sulfur decreases with increasing pressure, the amount of COS produced was reduced to a greater extent at a high SO_2 conversion under conditions of high pressure. From these results, it was confirmed that SnO_2–ZrO_2 is a suitable catalyst for the catalytic reduction of SO_2 at high pressure.

Acknowledgements

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