Selective Catalytic Reduction of NO with C₂H₄ -Effect of Addition of H₂ or CO to Each Elementary Reaction-

Tatsuya Matsuoka, Noriyasu Okazaki*

Kitami Institute of Technology, Kitami, 090-8507 Japan Fax : +81-157-24-7719, E-mail : zaki@chem.kitami-it.ac.jp

Abstract: The activity of C_2H_4 -SCR-NO over alumina based catalyst added H_2 or CO more improved than the activity of additive-free. As a result of this study, it was found that the addition of H_2 or CO affects oxidation reaction of NO and combustion reaction of C_2H_4 . Especially, the positive effect of adding H_2 or CO to oxidation reaction of NO was strong. From the above, we focused on the reaction on the catalyst surface to elucidate effect of addition of H_2 or CO to oxidation reaction of NO, so oxidation reaction of NO on catalyst surface was observed in situ.

Keywords: alumina based catalyst, C₂H₄-SCR-NO, addition of H₂ or CO.

1. Introduction

NO contained in the exhaust gas discharged from factories or cars causes environmental pollution, so purification is needed. The selective catalytic reduction of NO with hydrocarbon (HC-SCR-NO) is efficacious method to remove NO included in automobile exhaust gas. By past result of research, alumina is found to have high selectivity for NO in HC-SCR-NO among metal oxide¹. It is also reported that the activity of C₂H₄-SCR-NO on Ag/alumina catalyst is rapidly improved to add H₂ to HC-SCR-NO². The various impurities are included in commercially available alumina, the kind of impurity included in it depend on the difference in the production process. But, it has not extensively researched how the various impurity will affects the activity. Also, how a lot of CO and H₂ included in automobile exhaust gas affect HC-SCR-NO. From the above, I studied about the effects the impurities mainly included in alumina have on each elementary reaction.

2. Experimental (or Theoretical)

Alumina (AKP-G015) was purchased from Sumitomo Chemical Co.,Ltd. Alumina-supported sulfur catalyst (1.0wt% S) and sodium catalyst (1.0wt% Na) was prepared by impregnation of it with ammonium sulfate and sodium hydrogen carbonate. These catalysts were pressed into pellet followed by crushing and sieving to 30-42 mesh and disc-shaped. Thereafter they calcined at 800°C for 4h in air. All the reactions were carried out with a fixed bed flow reactor at a W/F of 0.18 g·s·cm⁻³ (catalyst, 0.40 g; total flow rate, 130 ml·min⁻¹). A reactant gas (NO, 1000 ppm; C₂H₄, 500 ppm; O₂, 2.0%; H₂ and CO, 0 or 2000ppm; He, balance) was passed through the catalyst bed. The outflow gases were analyzed by GC (TCD).

3. Results and discussion

At first, it was considered the activity of C_2H_4 -SCR-NO (additive-free, added H₂, added CO) over each alumina based catalyst. As a result, the activity of C_2H_4 -SCR-NO added H₂ over Al₂O₃ catalyst was more improved than the activity of additive-free in the range of 400-500°C (Fig.1). The activity of C_2H_4 -SCR-NO added H₂ over S(1wt%)/Al₂O₃ catalyst was more improved than the activity of additive-free at 550°C. Also, the activity of C_2H_4 -SCR-NO added CO over Al₂O₃ catalyst was more improved than the activity of additivefree in the range of 400-500°C (Fig.1). the activity of C_2H_4 -SCR-NO added CO over S(1wt%)/Al₂O₃ catalyst was more improved than the activity of additive-free at 550°C. But, the activity of C_2H_4 -SCR-NO added H₂ or CO over Na(1wt%)/Al₂O₃ catalyst more decreased than the activity of additive-free. Next, it was studied that H₂ and CO have the effect on elementary reaction which constitutes C_2H_4 -SCR-NO (oxidation reaction of NO, combustion reaction of C_2H_4 and reduction of NO₂). As a result, it was indicated that addition of H_2 or CO to oxidation reaction of NO and combustion reaction of C_2H_4 had positive effect for the activity. Combustion reaction of C_2H_4 over Al₂O₃ catalyst was more restrained than it of additive-free by adding H_2 to it, Na(1wt%) catalyst was more restrained than it of additive-free by adding H_2 or CO to it. But, positive effect of adding H_2 to combustion reaction of C_2H_4 over S(1wt%)/Al₂O₃ catalyst was not observed. And the activity of oxidation reaction of NO was improved by adding H_2 or CO to it even using any alumina based catalyst, especially the maximum activity of the reaction added CO was remarkably improved (Fig.2). From the above, I focused on the reaction on the catalyst surface to elucidate effect of addition of H_2 or CO to oxidation reaction of NO.

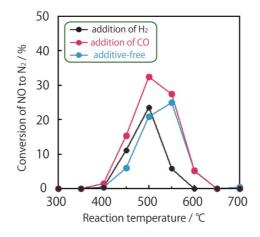


Fig. 1. The activity of C_2H_4 -SCR-NO additive-free and C_2H_4 -SCR-NO added H_2 or CO on alumina catalyst.

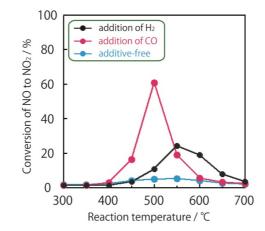


Fig. 2. The activity of oxidation of NO additive-free and oxidation of NO added H_2 or CO on alumina catalyst.

4. Conclusions

The addition of H₂ or CO had effect on oxidation reaction of NO and combustion reaction of C₂H₄. Especially, the activity of oxidation reaction of NO over Na(1wt%)/Al₂O₃ or S(1wt%)/Al₂O₃ added CO more remarkably improved than the activity of additive-free. From the above, it is think that the improvement of the activity of oxidation reaction of NO and combustion reaction of C₂H₄ relate to the improvement of the activity of C₂H₄-SCR-NO.

References

- 1. H. Hamada, Catal. Today, 22, 21 (1994).G. Rothenberg, Catalysis, Wiley-VCH, Weinheim, 2008, p. 127.
- 2. S. Satokawa, Chemistry Letters, 294 (2000).