SCR of NO with C_3H_6 over iron modified Ag/Al_2O_3 catalysts supported on honeycomb ceramic

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Abstract: Ag/Al_2O_3 catalysts supported on honeycomb ceramic were prepared by sol-gel and impregnation methods and modified with Fe to promote the catalytic performance for SCR of NO with C_3H_6 and to improve the resistance to SO_2 and H_2O. The results showed that the NO reduction efficiency by $7.2Fe/1.9Ag/20Al_2O_3/CM$ with C_3H_6 was more than 90% and reached about 100% at 500 °C and 550°C respectively. Iron can effectively improve the ability of $Ag/20Al_2O_3/CM$ catalysts to resist SO_2 and H_2O in flue gas.

Keywords: SCR of NO, Ag/Al_2O_3 catalysts, C_3H_6, Fe

1. Introduction

Ag supported on Al_2O_3 catalyst showed good activity in the SCR of NO by HC agents [1, 2]. The resistance of Ag/Al_2O_3 catalysts to H_2O and SO_2 is a problem. More et al. [3] used Mg to modify the Ag/Al_2O_3 catalyst and found that Mg can improve the resistance of Ag/Al_2O to SO_2, but H_2O had a great influence on the reduction of NO by $Ag/Mg/Al_2O_3$, e.g., at 350°C, the addition of 9% H_2O reduced the conversion of NO by 40% and the conversion of reducing agent C_3H_6 by 10% respectively. The resistance of Ag based catalysts to SO_2 and H_2O remains to be further investigated for HC-SCR of NO.

Recent studies demonstrated that iron or iron oxides could effectively reduce NO above 850°C with HC fuels and has a good ability to resist SO_2 and H_2O [4]. Further study found that the iron-based supported catalysts can effectively reduce NO with HC at lower temperature, e.g., Fe/Al_2O_3/Cordierite with a 5.5% Fe loading (mass fraction) can achieve a NO reduction efficiency of up to 97% at 550°C and a better resistance to SO_2 and H_2O in the flue gas [5]. In this study, the Ag/Al_2O_3 catalyst was modified by Fe in order to improve its resistance to SO_2 and H_2O and cordierite honeycomb ceramic was used as the carrier. The C_3H_6-SCR of NO was tested in a flow reactor with simulated flue gas.

2. Experimental

Sol-gel and impregnation methods were used to prepare the catalysts. Raw cordierite honeycomb ceramics were first immersed into the Al_2O_3 sol for 3h followed by drying at 110 °C for 12 h and calcining at 500°C for 5 h to prepare the $20Al_2O_3/CM$ samples. Then the $20Al_2O_3/CM$ samples were immersed into the 1 mol/L $AgNO_3$ solution first for 10 hours, then dried at 110 °C for 12 h and calcined at 500°C for 5 h to obtain the $Ag/20Al_2O_3/CM$ samples, which were then immersed into the 1mol/L $Fe(NO_3)_3$ solution to finally obtain the xFe-yAg/$20Al_2O_3$/CM catalysts, where x and y note the mass fraction of loaded Fe and Ag respectively based on the raw cordierite honeycomb ceramics mass. The physical-chemical properties were characterized by SEM, XRD, Nitrogen adsorption/absorption, H_2 temperature programmed reduction (H_2-TPR) and pyridine adsorption FTIR (Py-FTIR), etc.

The C_3H_6-SCR of NO evaluation were conducted in a one-dimensional electrically heated temperature programmed ceramic tubular reactor in simulated flue gas atmosphere (total flow rate 1.5L/min, 500ppm NO, 0.5% C_3H_6 , 3% O_2, 8% H_2O, 200ppm SO_2 , N_2 balanced) at 200-700 °C.

3. Results and discussion

Figure 1 presents the NO and C_3H_6 conversion. The NO conversion to N_2 over $7.2Fe/1.9Ag/20Al_2O_3/CM$ was about 100% at 550 °C and did not decrease as the temperature increased. However, the NO conversion over $2.0Ag/20Al_2O_3/CM$ decreased from about 90% at 550°C to about 60% as the temperature increased to

700 °C. Figure 2 presents the effect of SO_2 and H_2O in the flue gas on the NO conversion. The results showed that 7.2Fe/1.9Ag/20Al_2O_3/CM had a good resistance to SO_2 and H_2O, while 2.0Ag/20Al_2O_3/CM would lost its reactivity in the presence of SO_2 and H_2O.

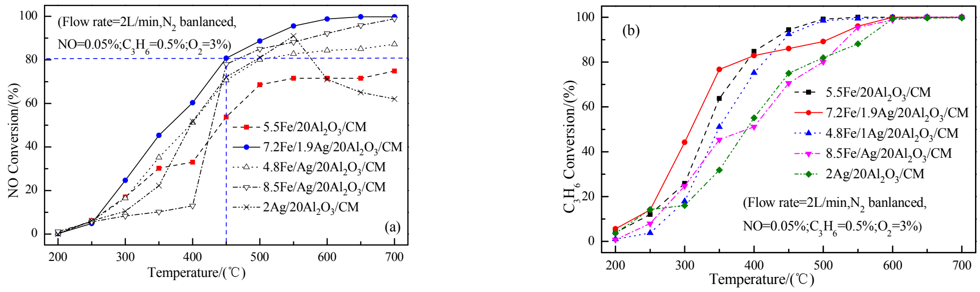


Figure 1. NO and C_3H_6 conversion over different catalysts

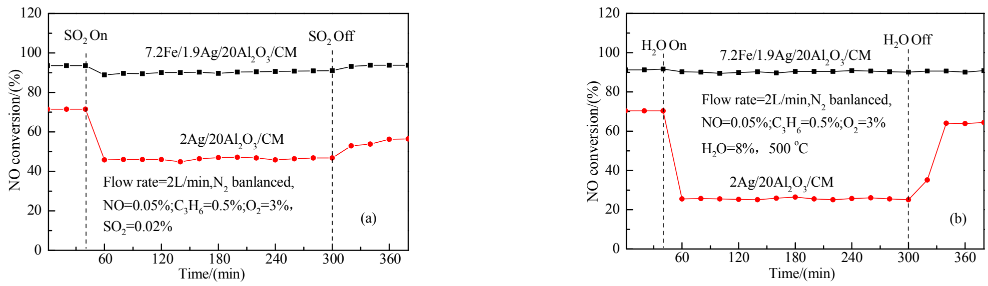
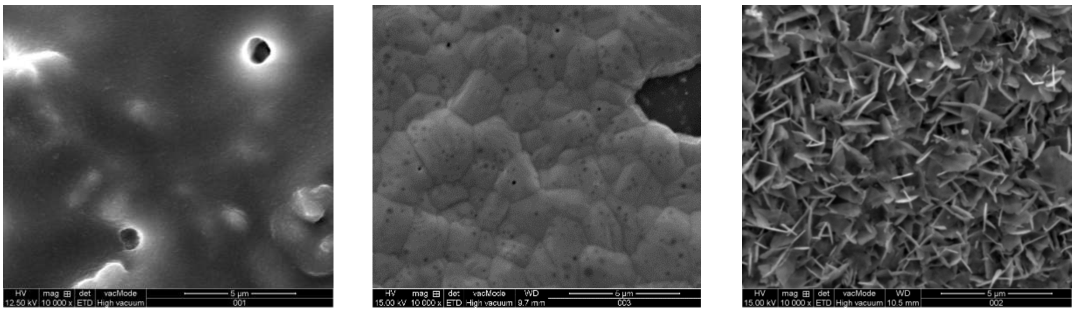


Figure 2. Effect of SO_2 (a) and H_2O (b) on NO conversion at 500 °C



(a) (b) (c)

Figure 3. SEM images of (a) 20Al_2O_3/CM; (b) 2Ag/20Al_2O_3/CM; (c) 7.2Fe/1.9Ag/20Al_2O_3/CM

Figure 3 presents the SEM images of the catalysts. The surface of 2Ag/20Al_2O_3/CM was relatively smooth and a layer of Ag_2O was formed on the surface of the carrier, 20Al_2O_3/CM. When iron was used to modify the 2Ag/20Al_2O_3/CM catalyst, e.g., 7.2Fe/1.9Ag/20Al_2O_3/CM, the catalyst surface became porous and needle-like and sheet-like crystals whose main phases were Fe_3O_4 and $AgFeO_2$ based on XRD patterns were formed. The other characterization was also conducted to investigate the influence of Fe on the physical and chemical properties of the catalysts. H_2-TPR results showed that 7.2Fe/1.9Ag/20Al_2O_3/CM has better reduction properties than Ag/20Al_2O_3/CM in a wider temperature range. Pyridine adsorption spectroscopy (Py-FTIR) results showed that Fe increased the Lewis acid sites on the catalyst surface.

4. Conclusions

Fe loading can effectively improve the resistance of Ag/Al_2O_3 catalysts to SO_2 and H_2O. NO conversion to N_2 was higher than 90% at 500 °C when there was 200 ppm SO_2 and/or 8% H_2O and reached about 100% at 550 °C when 7.2Fe/1.9Ag/20Al_2O_3/CM was used as the catalyst for C_3H_6-SCR of NO.

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