Selective Catalytic Reduction of NO_x with Methane over Ag-alumina Catalysts in High-content SO_2 Gas Streams

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Abstract

In this work, we investigated the performance of Ag-alumina catalysts for the SCR of NO with methane in gas streams with a high concentration of SO_2, typical of coal-fired power plant flue gas. Ag-alumina catalysts were prepared by co-gelation and nitric-acid leaching was used to remove weakly bound silver species from the catalyst surface [1, 2]. We found that SO_2 has a dramatic inhibitory effect on these catalysts, essentially quenching the SCR reaction at T <600°C. At temperatures higher than 600°C, the CH_4-SCR of NO occurs without catalyst deactivation even in 1000 ppm SO_2, the SO_2 effect was reversible, and quick activity recovery was observed after removal of SO_2 (Fig.1). Further, co-addition of water vapor (6.2%) does not change the above findings.

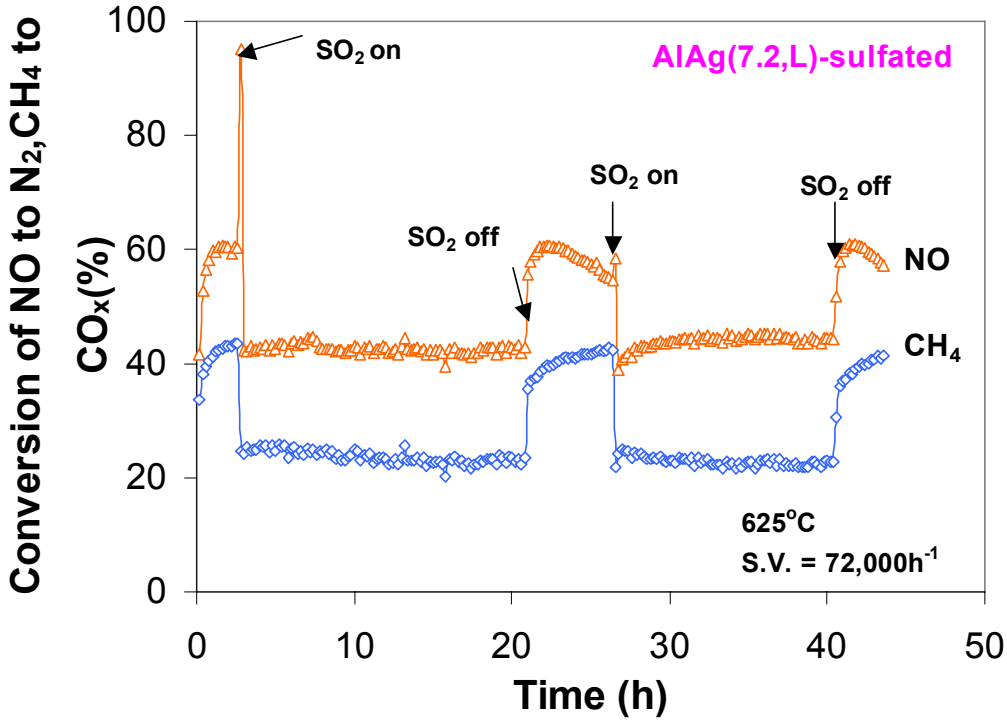

Fig. 1 SCR of NO with CH_4 over sulfated AlAg(7.2,L). Sulfation: 0.25%NO-2%CH_4-5%O_2-2000ppm SO_2-He, 625°C, 24h. Catalyst: 0.1027g. Feed gas: 0.25%NO-2%CH_4-5%O_2-0/1000ppm SO_2-He, 200ml/min. T=625°C. SV = 72,000h^{-1}.

At low temperatures, SO_2 adsorbs strongly on the catalyst surface forming sulfates on both alumina and silver. We determined that sulfates are not active for CH_4-SCR of NO_x. The reaction light-

off coincides with the onset of silver sulfate decomposition, indicating the critical role of silver, which is associated with methane activation [2]. SO_2 is reversibly adsorbed on silver above 600^oC, while alumina sites remain sulfated, and sulfated alumina decreases the extent of adsoption of NO_x. Kinetics results indicate that SO_2 increases the apparent activation energy for CH_4-SCR of NO_x on Ag-alumina catalysts, suggesting the participation of SO_2 in the reaction pathway. Our results also indicate a partial contribution of gas phase reactions to the formation of N_2 above 600^oC.

References:

1. A. Keshavaraja, X. She and M. Flytzani-Stephanopoulos, "Selective catalytic reduction of NO with methane over Ag-alumina catalysts". *Applied Catalysis B: Environmental*, 2000. 27(1): p. L1-L9.
2. X. She and M. Flytzani-Stephanopoulos, "The role of AgOAl species in silver-alumina catalysts for the selective catalytic reduction of NOx with methane". *Journal of Catalysis*, 2006. 237(1): p. 79-93.