The Formation of N_2O from NH_3-SCR Reaction over Commercial V_2O_5/TiO_2-Based Catalysts

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Introduction

Nitrous oxide (N_2O) is a strong greenhouse gas with global warming potential (GWP) which is much greater, by 310 times, than that of CO_2, and chronic exposure of humans to it even with low concentrations has been known to cause miscarriages in an early gestation period, liver disorders, kidney troubles, cancers and troubles of nervous systems [1-3]. N_2O having such effects and high GWP value would be produced from NH_3-SCR (selective catalytic reduction of NO_x by NH_3) processes that are widely used for abating NO_x from coal-, oil- and natural gas-fired power plants [4]. Thus, the extent of the formation of N_2O upon SCR reaction with commercial V_2O_5/TiO_2-based catalysts is of particular interest.

Materials and Methods

Direct measurements for N_2O formation during NH_3-SCR reaction over commercial V_2O_5/TiO_2-based systems were conducted using an on-line infrared (IR) analysis system as described earlier [5]. A Thermo Nicolet 6700 IR spectrophotometer equipped with a gas cell having an optical path of 10 meters is connected directly to a laboratory-designed SCR reactor, and the whole wall surface inside the gas cell was coated with Ni and finally Au to prevent undesired reactions between reactants on the wall surface. The IR system has a detection limit of 0.3 ppm for N_2O based on the most intense peak at 2237 cm^{-1}.

A "*standard*" 1.41% V_2O_5-7.98% WO_3/TiO_2 catalyst was employed for this study with reference SCR catalysts, and physicochemical properties of these samples were determined, as listed in Table 1. A model SCR reaction was conducted using a continuous flow fixed-bed type I-shaped Pyrex reactor. Typically 0.5 g catalyst was placed above a frit in the Pyrex reactor and calcined for 1 h at desired temperatures. A typical gas mixture consisting of 500 ppm NO, 500 ppm NH_3 and 5% O_2 in He at a total flow rate of 1,000 cm^3/min, corresponding to a GHSV value of 76,200 h^{-1}, was passed over the catalyst bed for measuring N_2O production as a function of reaction temperature.

Results and Discussion

When directly measuring N_2O concentrations during the course of SCR reaction, the extent of N_2O formation depended significantly on reaction temperatures, as shown in Fig. 1. The "*standard*" catalyst gave a decrease in NO_x conversions at temperatures greater than 350°C from which N_2O formation was easier. Other commercial V_2O_5/TiO_2-based catalysts with lower V_2O_5 amounts facilitated N_2O production even at the same temperatures, which was in excellent agreement with earlier studies [6]. A sample of the Com-C catalyst without WO_3 possessed higher capability of producing N_2O for NO_x reduction. Consequently, further optimization of frequently-used commercial SCR catalysts without any loss of $DeNO_x$ performances is required to reduce N_2O emissions from SCR processes.

Table 1. Physicochemical properties of commercial V_2O_5/TiO_2-based catalysts

Catalyst	Amount (wt%)		T_{calc} (°C)	S_{BET} (m^2/g)	Designated to:
	V_2O_5	WO_3			
TiO_2	-	-	-	83	
V_2O_5-WO_3/TiO_2	1.41	7.98	500	70	Com-A
	1.68	7.60	500	75	Com-B
	2.02	-	500	56	Com-C

Significance

This study clearly presents significant N_2O formation for SCR reaction with a commercial V_2O_5-WO_3/TiO_2 catalyst and the need to develop a new catalyst having lower N_2O production even at high temperatures.

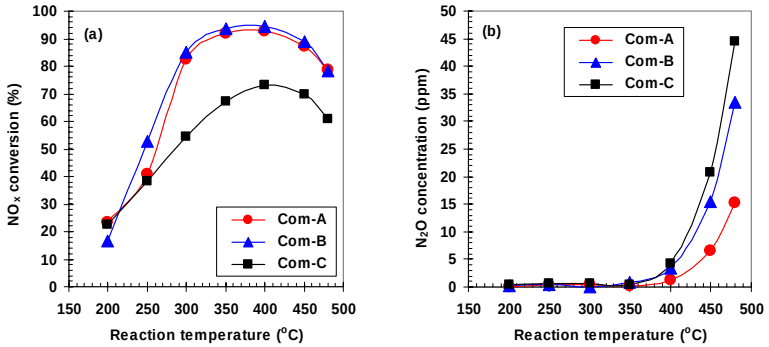

Figure 1. (a) NO_x reduction activity and (b) N_2O formation for NH_3-SCR reaction over a commercial V_2O_5-WO_3/TiO_2 catalyst calcined at 500°C.

References

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