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Influence of H_2O on the low-temperature NH_3-SCR of NO over V_2O_5/AC catalyst: An experimental and modeling study

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HIGHLIGHTS

- Influence of H_2O content on the SCR of NO over V_2O_5/AC catalyst was investigated.
- The microscopic reaction mechanism in the presence of H_2O was identified.
- A 3D mathematical model of monolithic reactor model was established.

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ABSTRACT

The selective catalytic reduction (SCR) of nitric oxide (NO) by ammonia (NH_3) at 393–473 K over AC (activated carbon) supported V_2O_5 (vanadium pentoxide) (V_2O_5/AC) catalyst in the presence of H_2O was investigated. In order to understand the influence of H_2O on reaction rate and reaction mechanism, the kinetic measurement was performed. Both mechanistic and empirical models were applied to correlate the experimental data. The results showed that the SCR reaction follows the Eley–Rideal mechanism rather than Langmuir–Hinshelwood or Mars–van Krevelen mechanisms whether H_2O was contained or not. And the pre-exponential factor decreased and activation energy increased with the increasing amount of H_2O. But at high H_2O content, the kinetic parameters were almost unchanged with the further increase of H_2O content. Finally, the intrinsic kinetic models obtained in this work were incorporated into a 3D mathematical model of monolithic reactor. It was found that the Eley–Rideal model was more suitable for describing the SCR of NO by NH_3 in the presence of H_2O than the empirical models, and thus should be adopted in the actual reactor design and scale-up for controlling the emission of NO_x.

1. Introduction

The emission of NO_x must be reduced because it can result in many global environmental problems. As the regulation for NO_x emission becomes more and more strict, much effort has been focused on the development of efficient NO_x control ($DeNO_x$) technology nowadays [1–4]. One of the most effective technologies is the SCR of NO by NH_3 over various catalysts, and it is generally accepted that under flue gas conditions, i.e. dilute mixtures of NO and NH_3 with O_2 in large excess, the SCR reaction occurs according to the following stoichiometry:

$$4NO + 4NH_3 + O_2 \rightarrow 4N_2 + 6H_2O \quad (R1)$$

Many metal oxide catalysts [5–7], such as CuO/Al_2O_3, V_2O_5–WO_3/TiO_2, and MoO_3/TiO_2. have ever been developed and successfully commercialized for NO removal. However, these catalysts are usually operated at temperatures higher than 623 K so as to avoid the deactivation by SO_2 and H_2O which takes place at low temperatures [8], while the flue gas temperature for many burners is in the range of 393–523 K, which will have a great energy consumption if the units for reheating flue gas are added [9]. It is known that the V_2O_5/AC catalyst has exhibited quite high activity at low temperatures for NO removal, and SO_2 in the flue gas does not deactivate the V_2O_5/AC catalyst but improves the activity in the absence of H_2O [10]. So the V_2O_5/AC catalyst may be an excellent option for SCR of NO with NH_3. Some researchers go a further step to investigate the kinetics over low-temperature catalysts. By far there have been two kinds of model proposed based on Eley–Rideal mechanism and Langmuir–Hinshelwood mechanism, respectively. Some authors believed that the reaction occurs through an Eley–Rideal type mechanism in which NH_3 is adsorbed on the vanadium-based catalyst in the first step, and then the reaction proceeds with NO from the gas phase [11–14]. Meanwhile, it has also been reported that the Langmuir–Hinshelwood mechanism which considers that NO and NH_3 are both adsorbed on the catalyst surface and then react, is more suitable than Eley–Rideal mechanism. For instance, Valdes-Solis et al. [15] measured the kinetics

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