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Direct Measurement System for N_2O Emissions from NH_3-SCR Reaction over V_2O_5/TiO_2-Based Catalysts

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1. Introduction

Nitrous oxide (N_2O) having global warming potential value much greater, by 310 times, than that of CO_2 [1], would be significantly produced from NH_3-SCR $DeNO_x$ (selective catalytic reduction of NO_x by NH_3) processes [2] that are widely used for abating NO_x from fossil fuel-fired power plants. The use of grab sampling for N_2O analysis gives secondary formation in the sample during transport and storage, because of the simultaneous presence of NO, SO_2 and H_2O in the sample [3]. Thus, an on-line analysis technique is required to quantitatively determine the extent of the production of N_2O at on-site SCR units.

2. On-line N_2O measurement system development

To adequately simulate N_2O emissions from many commercial NH_3-SCR processes, a continuous flow-type fixed-bed reactor system has been made [4]. The homogenizer equipped with this reactor system was specially designed to facilitate complete mixing between feed gases, such as NO, NH_3, O_2, and N_2O, in flowing He. A 180-mesh glass frit was inserted into the Pyrex reactor to hold fine powder samples. Pressures less than 5.5 psi were indicated for this test reactor into which a fine powder sample of a commercial V_2O_5-WO_3/TiO_2 catalyst was charged to be ca. 0.52 g under a pure He flow of 1,500 cm^3/min, corresponding to a gas hourly space velocity of 110,000 h^{-1}.

A Nicolet Model 6700 FTIR spectrophotometer with a specially-designed gas cell and data processing software setups was installed for direct measurements of N_2O formation during NH_3-NO-O_2 reaction with commercial SCR catalysts (Fig. 1). The gas cell has a volume of 0.75 L, and a pathlength of max. 10 m that can be adjusted using the micrometer feedthrough adjustment assembly. The whole surface of the wall inside the multipath gas cell was coated with Ni and Au to allow N_2O measurements

even under harsh conditions. The inside of the gas cell was designed to easily clean up via frequent temperature cycling in high vacuum, when contaminated by NH_4^{+1} salts such as ammonium nitrates and (bi)sulfates. The outer surface of the cylindrical stainless steel body of the gas cell was uniformly winded by heating wires that are able to heat it up to 200°C at which no condensation and deposition of reactants and their derivative salts for SCR reaction may occur.

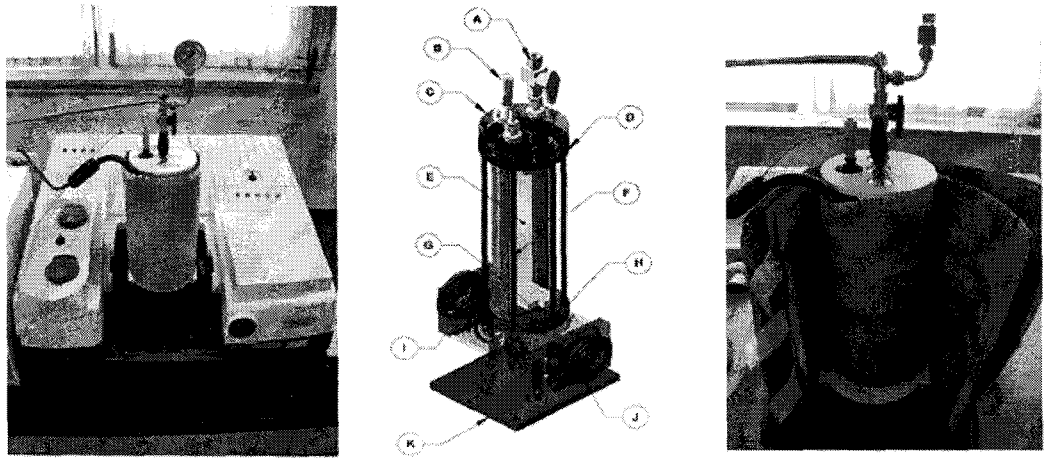

Fig. 1. On-line N_2O measurement system with a multipath gas cell.

3. Gases impurities determination and system validity tests

Ultrahigh purity N_2O, NH_3, O_2, He (99.999%) and NO (99.99%) were employed here, but a level of impurities, such as H_2O, CO_2, CO, CH_4, and other trace species, present in each cylinder gas was determined at the Specialty Gases Analysis Center, Scott Specialty Gases USA as a unit of ppm to acquire possible interference to direct N_2O measurements. All the impurities were less than 1.5 ppm, depending on the pure gases, and 10.3 ppm of N_2O as an impurity was detected for the pure NO gas, which is due to auto-oxidation reaction between NO gases even in the aluminum cylinder.

The instrument with the gas cell was calibrated using a flowing mixture consisting of different ppms of N_2O in ultrahigh purity He. This system had a detection limit of 0.3 ppm for N_2O based on the most intense peak at 2237 cm^{-1}. Substantial presence of 5% O_2 in a flowing mixture of N_2O/He gave no change in N_2O concentrations, as shown in Fig. 2. Consequently, this IR-based on-line N_2O measurement system will allow us continuous monitoring and measurements for N_2O levels produced during the course of SCR reaction over commercial TiO_2-supported V_2O_5 catalysts.

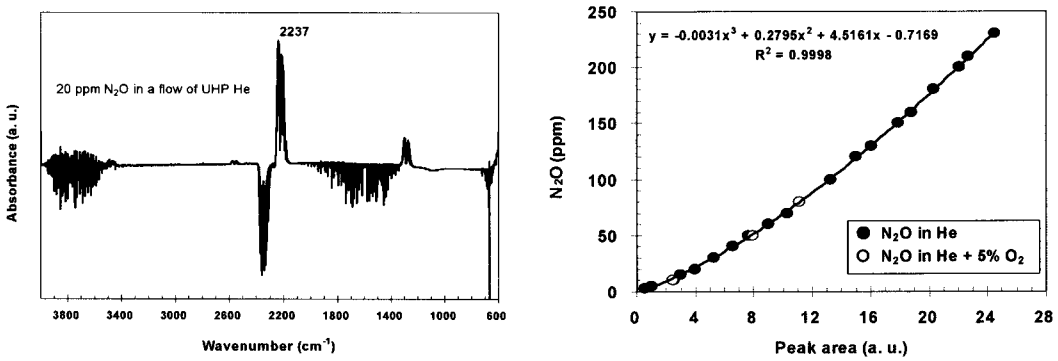

Fig. 2. Typical gas-phase spectrum for N_2O and its calibration using on-line IR analysis system..

4. Conclusions

An on-line IR-based system with a multipath gas cell was developed and had a detection limit of tens of ppbs. The gas cell with this system is available for directly measuring N_2O emissions from NH_3-SCR reaction on V_2O_5/TiO_2-based catalysts, because it has a strong tolerance to harsh conditions.

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