Stoichiometry for NO Reduction by NH_3

In a recent publication, Vogt, Boot, van Dillen, Geus, Janssen, and van den Kerkhof (*1*, *2*) [*J. Catal.* **114**, 313 (1988); *Catal. Today* **2**, 569 (1988)] have written Reaction (1) of their text including oxygen as one of the reactants for SCR of NO with NH_3 over a V_2O_5/TiO_2 on silica catalyst. It suggests that the stoichiometry of NO, NH_3, and O_2 by Reaction (1) would then be equal to 4 : 4 : 1, indicating an ammonia consumption ratio of unity defined as the moles of NH_3 consumed per net moles of NO reduction. Many previous investigations (*3* and *4*, to name a few) observed 0.67 for this ratio for similar vanadia catalysts, unlike the value of 1.0 by Vogt *et al.* (*1*, *2*). Reaction (1′) can thus be written

$$6NO + 4NH_3 \rightarrow 5N_2 + 6H_2O. \quad . \quad . \quad . \qquad (1')$$

There is no doubt that the stoichiometry can be easily found by the quantitative measurements of ppm levels of reactants and products using the radioactive isotopes of reactants, but the continuous measurement of the ppm level of NH_3 concentration is not simple routine work. Although the authors used radioactive isotopes to quantify the reaction stoichiometry, their experimental results from Fig. 2 and Table 3 do not agree with the reaction system, particularly as suggested for NO reduction reaction (Reaction (1)). For instance, the stoichiometry of Reaction (1) can be simply confirmed for B and C catalysts (6/33/61) at 400°C as follows:

$$NO_{consumed} = 480 \text{ ppm from Fig. 2}$$
$$NH_{3,consumed} = 495 \text{ ppm from Fig. 2.}$$

From Table 3, the contribution of Reaction (5) to the overall reaction is 26% for B and C catalysts at 400°C. Then,

$$NH_3 \text{ consumed by Reaction (5)} = (495) \times 0.26 = 129 \text{ ppm.}$$

Note that NH_3 oxidation by Reactions (3) and (4) is negligible, since the 26% NH_3 oxidation produces mainly N_2 by Reaction (5) and that NO reduction by Reaction (2) is also negligible from Table 3. Thus,

$$NH_3 \text{ consumption ratio for Reaction (1)} = \frac{NH_3 \text{ consumed by Reaction (1)}}{NO \text{ consumed by Reaction (1)}} = \frac{490 - 129}{480} = 0.76.$$

The value of 0.76 for the NH_3 consumption ratio does not reflect NO reduction reaction which occurs in the manner of Reaction (1) as proposed. This result is even closer to 0.67 for Reaction (1′) from this note than 1.0 for Reaction (1) from Vogt *et al.* (*1*, *2*). Therefore, care should be taken to write the stoichiometric reaction equation so that it agrees with the experimental verification of the mass balance. Bosch *et al.* (*5*) have recently pointed out that Reactions (1) and (1′) are competitive and both reactions should be considered for the overall reaction system because of the dependence of NH_3 consumption ratio on reaction temperature.

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