Selective catalytic reduction nitrogen oxides with methane over nanosized CuO supported on Al_2O_3. Part 3. Reaction kinetic behavior study

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The primary sources of NO_x, including several stationary industrial fossil fuel high temperature combustion processes and automobile exhaust sources, are of concern because these pollutants are significant precursors to acidic precipitation, which may acidification of the aquatic and terrestrial systems, and because the pollutants react in the atmosphere to generate ozone and photochemical smog. Moreover, the kinetic behavior of NO reduction with catalysis can be accounted by using the rate expression of the kinetic model. Kinetic parameters are also developed on the basis of the differential reactor data in this study. The adsorptive and catalytic behavior of a catalyst is strongly influenced by the capacity and texture of its exterior surface. Figure 1 presents the XRD patterns on the nanosized Cu/γ-Al_2O_3 catalysts, verifying the presence of CuO phases of the nanosized Cu/γ-Al_2O_3 catalyst. Dominant CuO diffraction peaks appeared near 2θ=35.5, 38.8, 48.7, 61.4, and 68.0° for a catalyst. As reported in the previous work established that CuO is the most active phase. However, under reduction conditions over the nanosized Cu/γ-Al_2O_3 catalyst, the oxidation state of copper may vary between Cu^{2+} and Cu^{+}, which might also govern the efficiency of the deNO reaction. Figure 2 compares the ATR-FTIR spectra on the catalysts, and also confirms the presence of the CuO-like phase on the surface of the nanosized Cu/γ-Al_2O_3 catalyst. It reveals that the peaks associated with the CuO-like phase on the framework are associated with a peak at around 2348 cm^{-1}. Therefore, the highly catalytic activity of the catalyst system in reduction NO with methane may be explained by the reversible redox behavior of CuO-Al_2O_3 couples in promoting the functional mechanism. Moreover, a kinetic rate expression was developed to describe the data over the range of conditions investigated. Furthermore, correlation equations in the form of Arrhenius's law are derived from experimental data to predict the destruction efficiencies of NO from the temperatures and residence period in a catalytic process. A Langmuir-Hinshelwood model incorporating a first-order reaction appears to adequately represent the NO reduction, which shows fairly good agreements with experimental data. This kinetic model is appropriate for oxygen presence and high concentrations of NO.

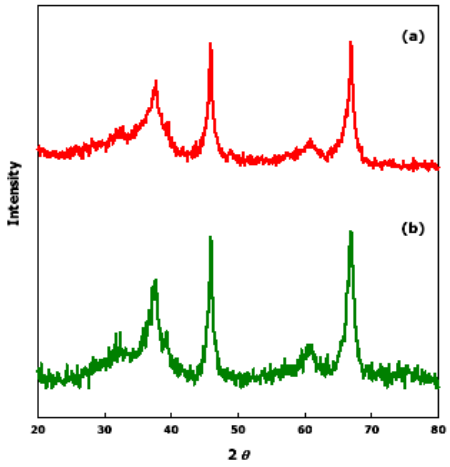

Figure 1. XRD pattern of the nanosized Cu/γ-Al_2O_3 catalyst. (a) fresh, (b) after activity test.

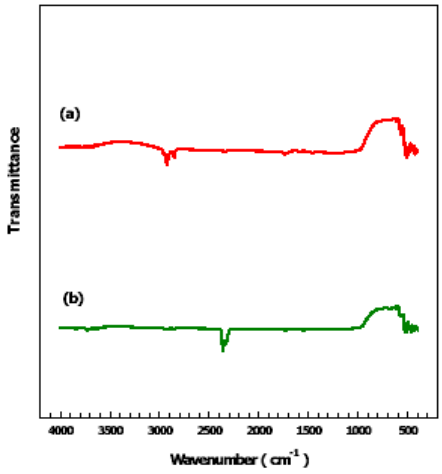

Figure 2. ATR-FTIR pattern of the nanosized Cu/γ-Al_2O_3 catalyst. (a) fresh, (b) after activity test.