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Study on the methods of catalytic activity evaluation, accelerated deterioration, and reactivation of diesel exhaust gas catalysts

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Copper zeolites have been used as SCR catalysts in diesel heavy-duty vehicles compliant with the 2016 regulations. In the catalyst development process, accelerated deterioration has been performed to simulate the deterioration conditions of long-distance driving at the laboratory level to evaluate the durability of the catalyst. If the deteriorated catalyst can be reactivated by certain treatments, it will not need to be replaced with a new one, which is desirable from both economic and resource perspectives. In this study, we investigated the methods for accelerated hydrothermal deterioration in the laboratory which can be performed in a relatively short time, the reactivation of the deteriorated catalysts, and for evaluating their catalytic activity.

Fig. 1 summarizes the correlation between treatment time and relative activity at various temperatures and water vapor concentrations for hydrothermal deterioration conditions. 700°C-10%H_2O-168h resulted in a relative activity of 0.73, and increasing the water vapor concentration to 30% at the same treatment temperature resulted in the same degree of deterioration after 20h, and further increasing the temperature to 750°C after 5h. The water vapor concentration generally used as a condition for accelerated deterioration by hydrothermal treatment is around 10%, the same level as the average exhaust gas from a vehicle, but by applying a much higher water vapor concentration condition than this, durability against hydrothermal deterioration can be determined in a relatively short time.

The reactivation treatment conditions were investigated for a model deteriorated sample (750°C-30%H_2O-10h, relative activity about 0.6) in the initial stage, in which the zeolitic crystal structure is mostly retained. When treated in a 75% water vapor containing H_2, a trend toward increased activity was observed in the 200-300°C range (Fig. 2). This was inferred to be due to the restructuring of Brønsted acid (H^+) and the active Cu sites. By the examination of a commercial catalyst, it was confirmed that the reactivation treatment is effective in the initial stage of deterioration.

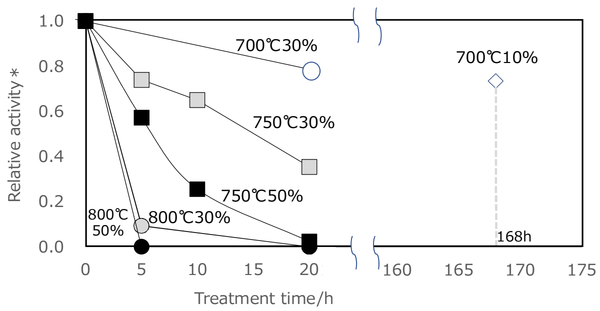

Fig. 1 Effects of water vapor concentration, temperature and time of the hydrothermal treatment on the relative catalytic activity for NOx conversion at 150℃ in standard SCR reaction. Sample weight: 40 mg; gas condition: 400 mL･min^{-1}, 200 ppm NOx + 220 ppm NH_3 + 1% H_2O + 10% O_2.

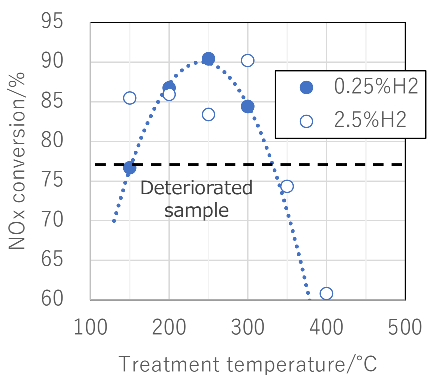

Fig. 2 Effects of addition of H_2 and temperature in reactivation treatments under 75%H_2O (N_2 balance) on the catalytic activity for NOx conversion in standard SCR reaction. Sample weight: 10 mg; gas condition: 400 mL･min^{-1}, 200 ppm NOx + 220 ppm NH_3 + 1% H_2O + 10% O_2. The y-axis shows average NOx conversion from 260 to 380°C.