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A Study on Oxygen Storage Capacity Characteristics during Thermal Degradation of Three Way Catalyst

Sota Aoyama [1)] Yunosuke Kubo [1)] Jin Kusaka [2)]

1) The University of Waseda, Graduate School of Creative Science and Engineering
3-4-1 Okubo, Shinjuku, Tokyo, 169-8555, Japan (E-mail: sota-bluemt18@fuji.waseda.jp)
2) The University of Waseda, School of Creative Science and Engineering
Address is the same as above

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Three-way catalysts (TWC) simultaneously purify carbon monoxide (CO), nitrogen oxides (NOx), and hydrocarbons (HC), which are regulated emission components in vehicles equipped with spark-ignition engines. To extend the purification performance range, ceria-zirconia (CeO2-ZrO2:CZ) with oxygen storage capacity(OSC) is used as an auxiliary catalyst. However, it was found that thermal degradation occurs when exposed to high temperatures due to high-load engine operation, and that thermal degradation progresses from the quantitative factor of particle size and the qualitative factor of reaction rate constant, resulting in a decrease in purification performance. Particle size increased with degradation, and a decrease in purification performance was confirmed by experiments. When only the change in particle size, a quantitative factor, is considered in numerical calculations, discrepancies arise between the experimental results and the results, and previous studies have suggested the need to consider qualitative factors as well. Therefore, the objective of this study is to focus on OSC, to obtain experimentally the reaction rate constants, and to develop a model that takes into account the qualitative degradation factors of OSC.

The reactions within the TWC were assumed to occur at the four active sites shown below. As shown in Figure 1, Precious group metal (PGM), Three Phase Boundary (TPB), Fast OSC, a highly active site where rapid absorption and release of oxygen occur, and Slow OSC, a low active site that reflects the long distance from the TPB vicinity and the oxygen absorption property of ceria itself. In this study, we will conduct experiments and analysis on the Fast OSC, which has the highest contribution to the purification performance among the auxiliary catalysts. Since the OSC function may be lost due to endurance treatment, catalysts that have not lost their OSC function were used in the experiments and simulations. To elucidate the phenomena in Fast OSC, CO-TPR(Temperature Programmed Reduction) and CO_2 production tests related to the oxygen release reaction in Fast OSC were conducted.

First, CO-TPR tests with CO and O_2 were conducted using two catalysts in different states of degradation. As a result of confirming the formation behavior of carbon dioxide (CO_2) by transiently increasing the temperature, it was found that the temperature range in which the OSC functions shifted to the high-temperature side as thermal degradation progressed. From the results of this test, peaks associated with OSC can be discriminated. Therefore, an Ozawa plot was created as shown in Figure 2. Then, the activation energy was calculated using the slope of the approximate line obtained from the created Ozawa plot. From these experimental analyses, it was found that the OSC-related activation energy increases with the progress of thermal degradation, and the OSC function also decreases.

Furthermore, the CO_2 production test confirmed the CO_2 production at steady-state temperatures, and the results showed a decrease in purification performance due to degradation and differences in the temperature range of OSC onset in O_2 release and absorption. In the future, I also plan to verify the activation energy calculated by the CO-TPR test analysis based on the test results.

The findings from the above are as follows. First, CO-TPR tests confirmed that OSC characteristics change with catalyst degradation. Second, Ozawa plot analysis confirmed that the activation energy of the OSC reaction increases with degradation.

The activation energies calculated from the test results were only applied to the catalysts used in the experiments. In the future, we plan to develop a more accurate OSC model by quantifying the changes in active point density and activation energy with degradation. To this end, we plan to conduct experiments and validate the model using new types of degraded catalysts.

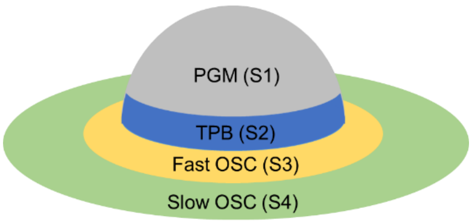

Fig.1 Schematic diagram of TWC

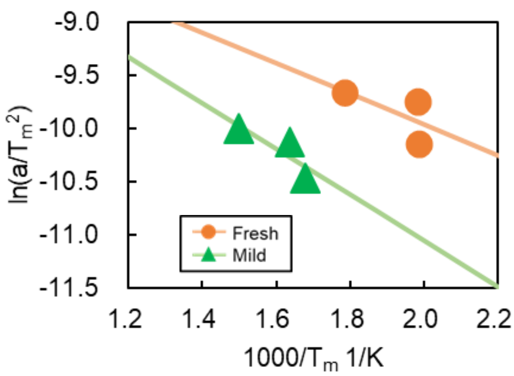

Fig.2 Results of analysis by Ozawa plot