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Effect of Precious Metal Species and Ceria-Zirconia Composition on Oxygen Storage Distribution in Three-Way Catalyst

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In three-way catalysts (TWC) for gasoline engines, ceria-zirconia solid solutions (CZ) function as oxygen storage materials, buffering fluctuations in exhaust air-fuel ratio under transient operating conditions. Elucidating the spatial oxygen storage and release mechanism inside the TWC is essential for improving catalyst performance and constructing predictive numerical models. In this study, oxygen isotope labeling experiments were conducted using model catalysts consisting of dense CZ substrates coated with a precious metal (Pd or Pt), and the effects of precious metal species and CZ composition on the internal oxygen storage distribution were visualized and analyzed.

Two CZ compositions were examined: Sample A (CeO_2:ZrO_2 = 80:20 wt%) and Sample B (CeO_2:ZrO_2 = 44.4:55.6 wt%). After reducing the sample in H_2/Ar at the target temperature (400 °C) for 40 minutes, $^{18}O_2$ was introduced for 2 minutes, followed by rapid quenching by He gas impinging jet to freeze the ^{18}O distribution in its reacting state. Cross-sections polished by cross-section polisher and surfaces of the quenched samples were analyzed by NanoSIMS 50L (CAMECA, maximum spatial resolution ≈ 50 nm) to map the spatial distribution of ^{18}O concentration along with the Ce/Zr signal ratio and precious metal signal.

NanoSIMS imaging of the cross-sections and surfaces revealed that the oxygen storage distributions differ between precious metal species (Fig. 1). In Pt/CZ, elevated ^{18}O concentration spread broadly across the surrounding CZ surface away from the Pt particle. In Pd/CZ, by contrast, high ^{18}O concentration was localized directly beneath and on the Pd particle surface. A gap observed between the Pd particle and the CZ substrate, identified from the $^{12}C_2^-$ signal, suggests that $^{18}O_2$ diffused through this path to the Pd/CZ interface. Surface imaging confirmed that the high-^{18}O region extended farther from the metal particle in Pt/CZ than in Pd/CZ. Regarding CZ composition, Sample A with higher Ce content exhibited a larger high-^{18}O region than Sample B across both precious metal types.

To further investigate the relationship among ^{18}O concentration, distance from the precious metal interface, and local Ce/Zr signal ratio, a grid-based heatmap analysis was applied to the NanoSIMS data (Fig. 2). The results showed that Pt maintains high ^{18}O concentration far from the metal interface with low distance-dependence, while Pd shows a steep concentration gradient with rapid decay over distance. In addition, a peak in ^{18}O concentration was observed near a Ce/Zr signal ratio of approximately 0.5 in the Pd/CZ heatmap at 500 °C.

The findings obtained in this study provide valuable insights into the effects of precious metal species and CZ composition on the ^{18}O storage distribution inside the model three-way catalyst.

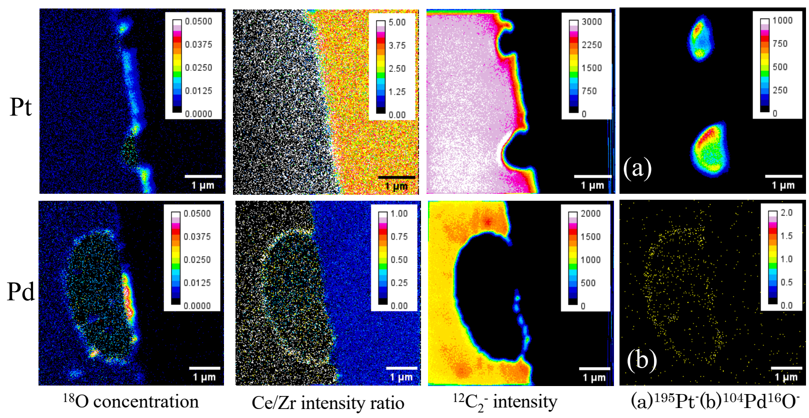

Fig.1 Cross-sectional distribution of ^{18}O concentration and phase mapping for Pd/CZ (top) and Pt/CZ (bottom) at 400 °C. The CZ composition is CeO_2:ZrO_2 = 80:20 wt% (Sample A).

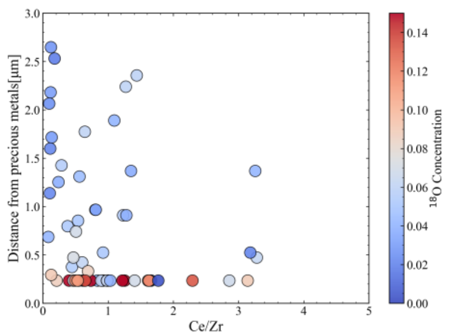

Fig.2 Heatmap of ^{18}O concentration with respect to distance from precious metals and Ce/Zr signal ratio (Pd/CZ, 500 °C).

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