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Visualization of Oxygen Storage in Pd/CeO_2-ZrO_2 Three-Way Catalyst through Isotope Quenching Technique

-Effect of Temperature-

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In three-way catalyst (TWC) for automotive SI engine, high purification performance can be maintained at lean or rich condition as well as stoichiometric condition through oxygen storage/release process in oxygen storage material. For material development toward higher catalytic activity and thermal durability, and construction of precise numerical model for model-based development, precise understanding of the detailed TWC reaction mechanism including oxygen storage-release process is an important issue. In order to clarify the local oxygen release/storage kinetics, isotope-quenching technique was applied to a model TWC, in which the catalyst is quenched within 1.5 sec by He gas jet after oxygen isotope is supplied at elevated temperature. Distribution of oxygen isotope at surface and inside bulk of the quenched sample is measured by secondary-ion mass spectroscopy (SIMS) with a spatial resolution of 50 nm. In this study, the effect of temperature on oxygen isotope distribution of model TWC consisting of dense oxygen storage material (CZ: CeO_2-ZrO_2) substrate with Pd on the surface was investigated. Fig. 1 shows cross-sectional ^{18}O concentration (c_{18O}) distributions in the model TWC at (a) 600 ℃, (b) 500 ℃, and (c) 400 ℃. Before $^{18}O_2$ supply, the sample was exposed to 50%H_2/Ar for reduction (oxygen release). The lower figures show $^{12}C_2^-$ (cyan) and $^{91}Zr^{16}O^-$ (yellow) signals, which corresponds to gas phase and CZ, respectively. The Pd positions on the CZ surface were identified from these figures. From Fig. 1, ^{18}O incorporation from the CZ surface to bulk is enhanced around Pd at any temperature. With decreasing temperature, ^{18}O diffusion scale becomes shorter and high c_{18O} area is more localized around Pd, resulting in higher maximum c_{18O} in the cross section at 500 ℃ and 400 ℃ compared to 600 ℃. At 400 ℃, ^{18}O incorporation from the Pd-CZ-gas triple phase boundary was clealy observed. In addidon, ^{18}O penetration depth from the bare CZ surface decreases with decreasing temperature, while c_{18O} near the CZ surface is highest at 500 ℃, followed in order by at 600 ℃ and 400 ℃.

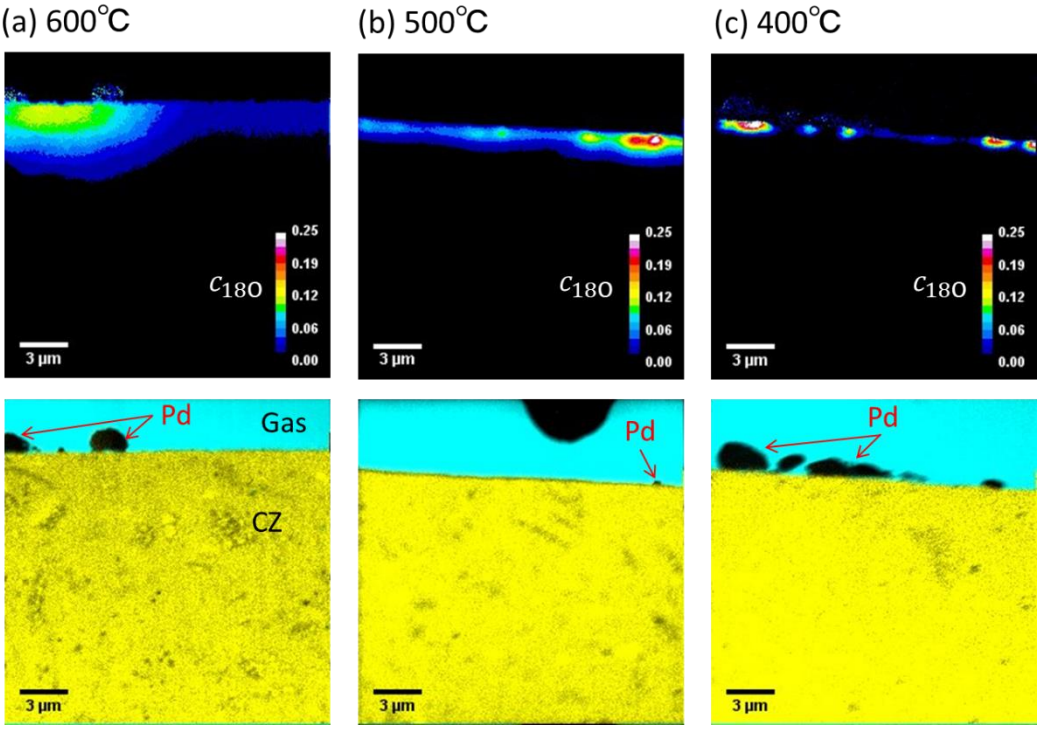

Fig. 1 Cross-sectional ^{18}O concentration (c_{18O}) distributions in the model TWC at (a) 600 ℃, (b) 500 ℃, and (c) 400 ℃. Lower figures show $^{12}C_2^-$ (cyan) and $^{91}Zr^{16}O^-$ (yellow) signals, which corresponds to gas phase and CZ, respectively.