Contribution ID: **181** Type: **Talk**

Concentration modulation excitation QuickXAS study of LaFe0.95Pd0.05O3 under cyclic red-ox conditions for methane oxidation

Friday 16 September 2011 11:55 (25 minutes)

LaFe0.95Pd0.05O3 is currently used as a component of automotive three-way catalysis for the emission control of stoichiometric gasoline engines. It is known that LaFe0.95Pd0.05O3 is able to segregate metallic palladium to its surface when reduced and to re-incorporate it to form a solid solution when re-oxidized, which is described as the self-regenerative function [1]. Catalytic activity measurements during methane combustion performed on LaFe0.95Pd0.05O3 (where Pd is incorporated into the LaFeO3 lattice) and 2 wt% Pd/LaFeO3 show that highest CH4-conversion rates are obtained for the impregnated material, where most of the Pd exists in the form of well dispersed nano-particles [2]. Cyclic operation between red-ox conditions of CH4-O2 and CH4 at 500℃ was simulated using concentration modulation excitation QuickXAS. This operation revealed a practical strategy to improve the activity of LaFe0.95Pd0.05O3. At each switch, CH4-O2⟶CH4 and CH4⟶CH4-O2, activity was enhanced for a short time. Identical measurements on Pd/Al2O3 demonstrated that an increase of CO2 production was observed only at the CH4⟶CH4-O2 switch. The time-resolved QuickXAS spectra of Pd/Al2O3 show that Pd reversibly changes between partially oxidized and reduced states. The phase-resolved spectra obtained by phase sensitive detection (PSD) display a simple reduction-partial oxidation process that may be accompanied by the formation of a PdxC species, interpreted as the source of the enhanced CO2 production. The phase-resolved spectra of LaFe0.95Pd0.05O3 vary between oxidized and reduced Pd and intensity variations are more pronounced compared to Pd/Al2O3. The shape of the phase-resolved spectra is interpreted as the fingerprint of the continuous segregation and re-incorporation of Pd into LaFeO3. This reversible process is responsible for the activity improvement at every red-ox switch [3].

[1] Y. Nishihita, J. Mizuki, T. Akao, H. Tanaka, M. Uenishi, M. Kimura, T. Okamoto, N. Hamada, Nature 2002, 418, 164.
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[3] A. Eyssler, E. Kleymenov, A. Kupferschmid, M. Nachtegaal, M. Santhosh Kumar, P. Hug, A. Weidenkaff, D. Ferri, J. Phys. Chem. C 2011, 115, 1231.

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Heterogeneous catalysis

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Session Classification: Heterogeneous Catalysis

Track Classification: Heterogeneous Catalysis