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## Concentration modulation excitation QuickXAS study of $\text{LaFe}_{0.95}\text{Pd}_{0.05}\text{O}_3$ under cyclic red-ox conditions for methane oxidation

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$\text{LaFe}_{0.95}\text{Pd}_{0.05}\text{O}_3$  is currently used as a component of automotive three-way catalysis for the emission control of stoichiometric gasoline engines. It is known that  $\text{LaFe}_{0.95}\text{Pd}_{0.05}\text{O}_3$  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  $\text{LaFe}_{0.95}\text{Pd}_{0.05}\text{O}_3$  (where Pd is incorporated into the  $\text{LaFeO}_3$  lattice) and 2 wt% Pd/ $\text{LaFeO}_3$  show that highest  $\text{CH}_4$ -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  $\text{CH}_4\text{-O}_2$  and  $\text{CH}_4$  at  $500^\circ\text{C}$  was simulated using concentration modulation excitation QuickXAS. This operation revealed a practical strategy to improve the activity of  $\text{LaFe}_{0.95}\text{Pd}_{0.05}\text{O}_3$ . At each switch,  $\text{CH}_4\text{-O}_2 \rightarrow \text{CH}_4$  and  $\text{CH}_4 \rightarrow \text{CH}_4\text{-O}_2$ , activity was enhanced for a short time. Identical measurements on Pd/ $\text{Al}_2\text{O}_3$  demonstrated that an increase of  $\text{CO}_2$  production was observed only at the  $\text{CH}_4 \rightarrow \text{CH}_4\text{-O}_2$  switch. The time-resolved QuickXAS spectra of Pd/ $\text{Al}_2\text{O}_3$  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  $\text{PdxC}$  species, interpreted as the source of the enhanced  $\text{CO}_2$  production. The phase-resolved spectra of  $\text{LaFe}_{0.95}\text{Pd}_{0.05}\text{O}_3$  vary between oxidized and reduced Pd and intensity variations are more pronounced compared to Pd/ $\text{Al}_2\text{O}_3$ . The shape of the phase-resolved spectra is interpreted as the fingerprint of the continuous segregation and re-incorporation of Pd into  $\text{LaFeO}_3$ . 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.

[2] A. Eyssler, P. Mandaliev, A. Winkler, P. Hug, O. Safonova, R. Figi, A. Weidenkaff, D. Ferri, *J. Phys. Chem. C* 2010, 114, 4584.

[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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