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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 LaFeO_3 lattice) and 2 wt% Pd/ LaFeO_3 show that highest 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 CH_4 at 500°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/ Al_2O_3 demonstrated that an increase of 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/ Al_2O_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 PdxC species, interpreted as the source of the enhanced 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/ Al_2O_3 . The shape of the phase-resolved spectra is interpreted as the fingerprint of the continuous segregation and re-incorporation of Pd into 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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Primary author: Dr FERRI, Davide (EMPA Duebendorf, Switzerland)

Presenter: Dr FERRI, Davide (EMPA Duebendorf, Switzerland)

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