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In situ XAS characterization of Pd/ZnO and Pd/Ga2O3 methanol steam reforming catalysts: correlating structure and reactivity

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Methanol steam reforming (MSR) is a promising reaction for H2 production for use in PEM fuel cells. In contrast to Pd on inert support oxides, which catalyzes methanol decomposition to CO and H2, Pd supported on ZnO and Ga2O3 has shown excellent activity and selectivity to the steam reforming reaction yielding H2 and CO2 [1]. Based on ex-situ characterization after reaction or reduction, this substantial difference in reactivity has been attributed to the formation of Pd-Zn and Pd-Ga alloys.
In this contribution we investigated the dynamic formation and decomposition of the selective alloy phases in different reaction atmospheres and correlated the properties with the catalytic performance. Time resolved in situ x-ray absorption spectroscopy at the Pd K edge was performed at the SuperXAS beamline with simultaneous analysis of formed products by mass spectrometry. Complementary information was obtained by utilizing a range of situ spectroscopic methods of different bulk/surface sensitivity, such as FTIR spectroscopy, in situ XRD and in situ XPS. We have monitored the dynamic formation of the active Pd-Zn and Pd-Ga alloy phases under relevant reaction conditions. PdZn alloying occurred already in methanol/water via spillover and reduction of the ZnO by H2 generated in the reaction [2], while oxidative conditions resulted in the decomposition of the alloyed surface and partial decoration of the Pd nanoparticles by patches of ZnO [2]. Upon alloying, the reactivity changed from methanol decomposition (CO/H2) to MSR (CO2/H2), confirming that PdZn alloy is in fact the selective phase for MSR. Similar spectral changes occurred on Pd/Ga2O3. A full EXAFS analysis identified the formation of a Pd2Ga intermetallic compound, which was confirmed by XRD. The extent and conditions of alloy formation under reduction/reaction atmospheres, the catalytic performance and the stability with respect to surface and bulk structure of PdZn and Pd2Ga nanoparticles will be discussed and compared.

[1] N. Iwasa, N. Takezawa, Top. Catal., 22, 215 (2003).
[2] K. Föttinger, J.A. van Bokhoven, M. Nachtegaal, G. Rupprechter, J. Phys. Chem. Lett. 2, 428 (2011).

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