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Carbon removal from the anodes of solid oxide fuel cells: insights from ab initio calculations.

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Hydrocarbon fuels, used for operation of solid oxide fuel cells (SOFCs), cause chemical deactivation of metallic (Ni) part of anodes due to carbon deposition, known as coking. Attractive approaches for carbon removal from the Ni surface via water-mediated mechanism have been proposed recently.[1,2] In both anodes Ba-containing oxides have been used for favorable adsorption of water molecules with subsequent release of hydroxyl species and oxidation of surface carbon atoms. Moreover, favorable water splitting on a Ni/BaO interface and carbon oxidation have been confirmed by ab initio calculations.[2]

Although the mechanism of carbon oxidation in these anodes and the role of water are reasonably understood, several questions still need to be addressed. These include: a) A comparison of the rates of carbon removal from Ba-containing and conventional Ni/YSZ anodes; b) The role of stoichiometric vacancies of doped oxides (i.e. YSZ) in water adsorption; c) The underlying reasons of favorable water adsorption on BaO surface (as compared to YSZ and CeO_2); d) Possible alternative oxides that can favorably adsorb and split water molecules for carbon oxidation.

In this presentation we shall attempt to provide the answers to the aforementioned questions based on our recent ab initio DFT+U calculations. We shall present the atomic model of Ni/$BaCe_{1-x}Y_xO_{3-\delta}$ as well as Ni/YSZ interfaces and show that carbon removal is indeed more favorable in case of Ba-containing oxide. Moreover we will demonstrate that the rate of carbon oxidation is significantly higher for Ba-containing anodes as compared to conventional Ni/YSZ. We will also show that water adsorption and splitting can be very favorable on a YSZ surface, if it is mediated by stoichiometric vacancies of YSZ. The explanation as to why carbon removal from Ni/YSZ anodes via interaction with steam is not observed experimentally will be provided.

The factors, which contribute to favorable adsorption of water on a BaO surface, will be analyzed. We will also show that there exists a clear correlation between the electronic properties of the oxide and the ability to favorably adsorb and split the water molecules. This finding allows formulation of a general principle for a design of coke-tolerant SOFC anode.

References.

[1] Yang L., Wang S., Blinn K., Liu M., Lui Z., Cheng Z., Liu M., Science **2009**, 326, 126.
[2] L. Yang, Y. Choi, W. Qin, H. Chen, K. Blinn, M. Liu, P. Liu, J. Bai, T. A. Tyson and M. Liu, Nature Communications **2011**, 2, 357.