

Fundamental Studies of Fuel Cell Catalysis Using X-rays

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Electrocatalytic energy conversion processes in fuel cells are expected to play a major role in the development of sustainable technologies to mitigate global warming and to lower our dependence on fossil fuels. I will demonstrate how electron and x-ray spectroscopy can be used to address fundamental questions regarding the reaction mechanism of the oxygen reduction reaction (ORR) on Pt and probe the electronic structure of Pt and adsorbed species of model system Pt catalysts. From time resolved x-ray photoelectron spectroscopy (XPS) we have determined the activation barrier for the O₂ dissociation process on Pt to be in the range around 0.25 eV [1]. Using in-situ studies under real electrochemical conditions of single crystal surfaces using high resolution Pt L-edge spectroscopy we have identified oxide growth at certain potentials that could have some major influence on the rate of the ORR reaction [2]. Recent dealloyed Pt-Cu catalysts have shown an enhanced activity in comparison to pure Pt by factor of five tested in a real fuel cell [3]. We demonstrate that this activity enhancement comes from compression strain of the Pt lattice that leads to modifications of the Pt electronic structure and subsequently weakens the surface bond of adsorbed oxygen species [4].

[1] D. J. Miller et al., *J. Chem. Phys.*, 22, 224701 (2010).

[2] D. Friebe et al., *Phys. Chem. Chem. Phys.*, 13, 262 (2011).

[3] R. Srivats et al., *Angew. Chem. Int. Ed.*, 46, 8988 (2007).

[4] P. Strasser et al., *Nature Chemistry*, 2, 454 (2010).