

Ir(111) Adsorbate Structure Induced Selectivity During PROX Reaction

Niclas Johansson^{1,2}, Joachim Schnadt^{1,2}, and Jan Knudsen^{1,2}

¹Division of Synchrotron Radiation Research, Lund University, BOX 118, 221 00 Lund, Sweden

²MAX IV Laboratory, Lund University, BOX 118, 221 00 Lund, Sweden

Niclas_V.Johansson@maxiv.lu.se

With an increasing interest in fuel cell technologies, much scientific effort is directed towards widening the use by overcoming its inherent efficiency thresholds. As the fuel cell anodes often contain Pt, they are very susceptible to CO poisoning and hence, the supply of clean, CO free, Hydrogen gas to the anodes is one of the biggest challenges [1]. Most of the H₂ gas today is produced from natural gas [2] and the resulting fuel gas typically contain a few thousand ppm CO where a concentration below 10 ppm is desirable for an effective and sustained fuel cell operation using air as O₂ feed. There are many approaches to reduce the CO concentration after H₂ synthesis, for instance, steam reforming, water-gas-shift, and preferential oxidation (PROX). During the PROX reaction, CO is selectively oxidized into CO₂ while inactive in water formation. PROX is a very effective method for cleaning the H₂ feed gas but it requires a very selective catalyst due to high H₂ to CO ratio, which is typically of the order of 100:1.

Here we report on an ambient pressure x-ray photoelectron (APXPS) study of PROX over Ir(111) in gas mixtures of O₂, H₂ and CO using the case of Pt(111) as reference reaction. We show that in the Pt(111) case, there is a step-like transition from inactive to active in both the CO oxidation and water formation reactions, cf. Figure 1(a) and (c). In contrast, the Ir(111) surface exhibits a phase where it is active in CO oxidation, but no water formation is observed as shown in Figure 1(b) and (d). This distinction can be explained in terms of the adsorbate surface structures present during the employed conditions. As recently demonstrated, the Ir(111) surface exhibits a phase where both chemisorbed O and CO is present on the surface [3], which leads to an increased CO oxidation activity compared to the inactive regime. Here, O₂ dissociation is preferred over H₂ dissociation and a surface phase related to the one over CO oxidation is formed, enabling CO oxidation prior to water formation.

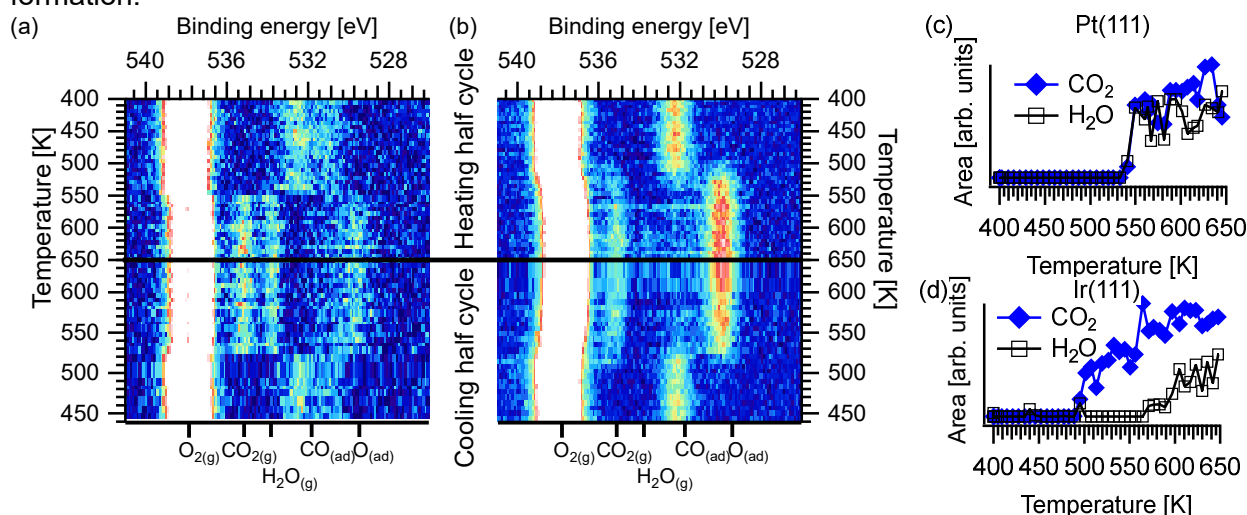


Figure 1: (a) and (b) show image plots of O 1s APXPS spectra acquired over Pt(111) and Ir(111), respectively, during the temperature ramp where the measured species are labelled in the bottom. (c) and (d) show the integrated areas of the products over the Pt(111) and Ir(111) surface demonstrating the PROX capabilities of the Ir(111) surface.

References:

- [1] R. Farrauto, S. Hwang, *Annu. Rev. Mater. Res.* **33**, 1 (2003)
- [2] J. N. Armor, *Appl. Catal. A-Gen.* **176**, 159 (1999)
- [3] N. Johansson, *J. Phys.: Condens. Matter* **29**, 44 (2017)