Field-assisted oxidation of a Fe single nanoparticle, nanoscale observations by Operando Atom Probe

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Mechanisms governing surface chemical reactions involved in heterogenous catalysis fundamentally depends on the synergistic interactions between the reactants and the different surface structures present at the surface. Recently, special attention has been raised regarding the influence of intense electric fields on these mechanisms [1]. An increasing number of analytical surface science techniques are achieving their conversion to their respective in-situ/operando version to study surface reactions at the "applied" conditions. Amongst them, Atom Probe Microscopy (APM) techniques are particularly interesting for their inherent use of intense electric fields and their capability to image matter at the nanoscale. In this work, we will present a nanoscale study of the field-assisted oxidation of a single Fe nanoparticle using Field Ion Microscopy (FIM) and Operando Atom Probe (OAP).

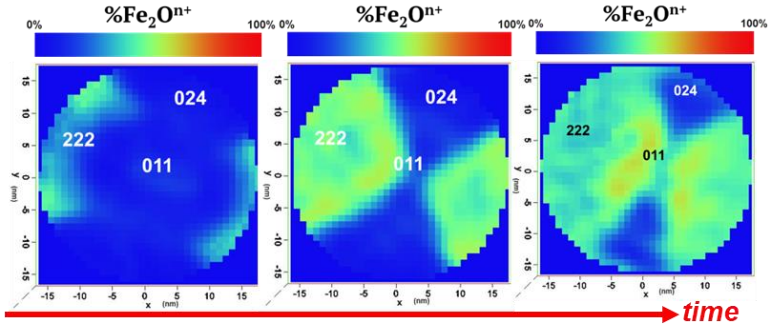

Figure 1. Fe oxidation imaged by OAP.

APM techniques are capable of imaging the apex of sharp needles, mimicking model nanoparticles, with nanometric lateral resolution. FIM is used to image apices with atomic resolution and to identify the crystal orientations with their Miller indices using stereographical projection. OAP relies on the thermally assisted field evaporation of positively charged ions from a needle shaped specimen [2]. Once the FIM characterization is complete the sample is maintained at 300K with an applied electric field of ~20V/nm, before starting OAP analysis and introducing 1.1×10^{-7}mbar of pure O_2. As soon as the O_2 is introduced, Fe_2O^{n+} ion species formation are observed starting from open facets structures, such as Fe{244} and {112}, towards the central Fe(011) and {024} (Fig.1). OAP results allow us to reconstruct the full movie of the surface oxidation in real-time and show how intense electric fields (>10V/nm) play a central role in surface chemistry.

[1] Che F. et al. ACS Catalysis, **8(6)**, 5153(2018)

[2] Lambeets S.V. et al. Topics in Catalysis, **63(15-18)**, 1606(2020)

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