

Impact of operation relevant treatments on the chemical structure of Pt in GaPt SCALMS

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Abstract

Supported Catalytic Active Liquid Metal Solutions (SCALMS) with small amounts of transition metals (TMs) dissolved in a liquid metal matrix have been reported as efficient thermocatalysts for non-oxidative dehydrogenation reactions.¹⁻³ Our detailed investigation of GaPt SCALMS with lab-based and synchrotron-based photoelectron spectroscopy (PES) reveals the presence of multiple Pt species at the alloy surface. According to previous studies on GaRh alloys,⁴⁻⁶ these Pt species can be attributed to isolated Pt atoms implemented in the Ga matrix or in residual GaO_x (preserved after having performed a H₂ reduction procedure at 923 K), and to agglomerated Pt clusters. Upon GaPt alloy liquefaction at 823 K, while the Ga 3d feature remained mainly unchanged (Fig. 1a), two new Pt 4f_{7/2} features at 73.0 eV and 71.9 eV appear indicating the formation of new Pt species at the sample surface (Fig. 1b). These new features mostly disappear upon sample oxidation at room temperature, while a significant Ga 3d GaO_x feature appears (see Fig. 1a, b). The relative occurrence of the different Pt species at the sample surface and (near-surface) bulk upon performing different operation relevant treatments is shown in Fig. 1c, suggesting a dynamic evolution of the different Pt species and indicating the possibility of a chemical structure control via deliberate sample treatments. Our findings are in good agreement with catalytic performance tests, revealing a correlation between reactivity, the catalyst deactivation mechanism, and the H₂ pretreatment temperature.

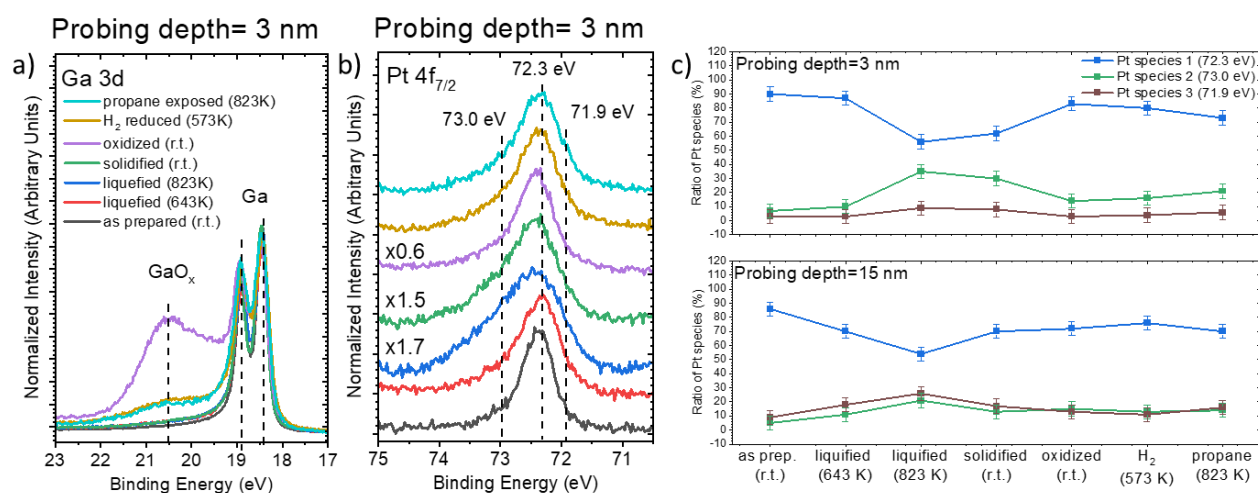


Figure 1. Synchrotron-based Ga 3d (a) and Pt 4f_{7/2} (b) XPS spectra of 1 at% Pt containing GaPt alloys that have undergone different treatments recorded at a fixed photoelectron kinetic energy of 500 eV, translating into a probing depth of 3 nm. The measurement temperatures are stated in the legend. (c) The evolution of the quantified occurrence of the different Pt species at the sample surface (top panel) and in the near-surface bulk (bottom panel) of GaPt alloys.

References

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