Tuning Catalytic Properties via Electronic Reconstruction at the Interface of Immobilized Gold Nanoparticles on Layered Titanate Nanosheets

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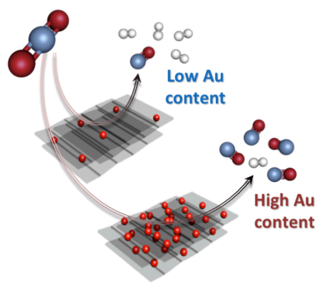
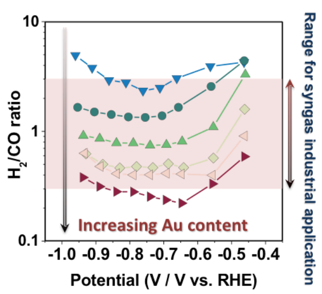
In the rational design of novel electrocatalysts, assessing the interaction between the loaded catalyst and the selected support / substrate e.g. conductive carbon black powder, metals, or metal oxides has been of increasing interest.[1,2] The synthesis of porous or nanostructured architectures providing high surface area, selection of supports / substrates with improved conductivity, and most significantly the assessment of synergetic effects arising at the formed interface affording complementary catalytic properties, reflect well-known strategies toward improved performances in a wide number of applications. Here we investigate a new electrocatalyst incorporating well-dispersed gold nanoparticles (NP) in a 0 to 93 wt.% Au range on ultra-thin titanate nanosheets (TiNS). Detailed characterization of these hybrid materials hinted to a charge transfer between the catalytic metal and the substrate. The effect of an electronic reconstruction was assessed in the reduction of CO_2 to CO, which has been highlighted in the industrial conversion of syngas (CO and H_2) to fuels. A control of the resulting electronic properties based on the interaction between Au NP and the TiNS substrate was suggested to dictate the stabilization of formed reaction intermediates and resulting product selectivity. In particular, the CO selectivity could be effectively controlled (in a CO Faradaic efficiency range from 3 to over 80%) balanced by an exclusive H_2 formation (HER), which is of pronounced industrial interest. In addition, our Au/TiNS achieved optimally high CO and H_2 production current densities, with 73 wt.% Au at the low cathodic potential region (-0.6 to -0.9 V_{RHE}). The synergism between both components sheds light in the promising potential of a tuned electronic reconstruction in the design hybrid catalysts, which is of great concern in electrocatalytic systems such as Li-air batteries.





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