## Controlling the Electronic State of Platinum Single-Atoms for Maximizing Catalytic Activity

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Single atom catalysts (SACs), in which all the metal atoms are spatially isolated on a support, have emerged as promising materials in heterogeneous catalysis. Previous studies reported controversial results about the relative level in catalytic activity for SACs and nanoparticles (NPs). These works have focused on the effect of metal atom arrangement, without considering the electronic state of SACs. Here, we immobilized platinum single atoms on defective ceria deposited on alumina (Pt<sub>1</sub>/CeO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>), and controlled the electronic state of Pt SACs, from oxidized state (Pt<sup>0</sup>: 16.6 at%) to highly metallic state (Pt<sup>0</sup>: 83.8 at%), by increasing reduction temperature up to 500 °C. The Pt SACs with controlled electronic state were employed for CO oxidation, CH<sub>4</sub> combustion, and NO oxidation, and their catalytic activity was compared with Pt NPs. The highly oxidized Pt SACs presented lower activity, whereas metallic Pt SACs showed superior activity than Pt NPs. The Pt SAC reduced at 300 °C (Pt<sup>0</sup>: 73.7 at%) showed the maximum activity for all the oxidations. This work clearly demonstrates that the electronic state of Pt SACs can play a major role in determining the catalytic activity.