Single-atom Catalysts on Metal Oxide Nanostructure Supports for High Performance Chemical Gas Sensors

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Single-atom catalysts (SACs) are heterogenous catalysts with atomically dispersed metal atoms that are stabilized exclusively on the surface of support materials. The key advantage of these catalysts is that every atom can take part in catalytic reactions to maximize the catalytic activity, up to levels which had only been possible with homogeneous molecular catalysts.[1] Of the SAC systems, those based on inorganic supports such as metal oxides or metals draw particular interest due to their superior thermal and chemical stabilities over that of organic supports. Recent studies on SAC systems based on inorganic materials as supports have focused largely on increasing the catalyst loading, while avoiding the clustering of catalyst atoms. Inorganic supports conventionally possess relatively low capacities for SACs loading (< 1 wt%) due to their small specific surface area and lack of defect sites for stabilization. Moreover, SACs are susceptible to surface diffusion and agglomeration during synthesis and/or catalytic reactions as a consequence of their high surface energy. Therefore, there is high demand for the development of a general synthetic approach for SAC systems based on inorganic metal oxide and metal supports that is reliable, facile, and can stabilize a large concentration of SACs.

From a design perspective, the metal loading can be maximized on metal oxide supports with nanometer-scale features and high specific surface areas. In this regard, we introduce two rational design strategies for precisely controlled synthesis of nanostructured metal oxide support-based SAC systems:

(1) Pt SACs-anchored carbon nitride is combined with electrospinning technique to stabilize the Pt SACs at heterojunctions between one-dimensional (1D) carbon nitride and SnO_2 nano-heterostructure (**Figure 1**).[2] The Pt SACs and carbon nitride simultaneously catalyze the HCHO gas sensing properties of 1D SnO_2 nanostructure (response = 33.9 at 5 ppm), and the spatial confinement effect of the heterojunction trapping endowed the Pt SACs with high stability without agglomeration (7.1% degradation in sensing performance after >170 h of operation at 275 °C).

A general approach is presented for preparing SACs on metallic, metal oxide and perovskite nanosheet (NS) supports to reach a high metal loading of up to 3.94 wt%, by utilizing N-doped graphene as a sacrificial template that spatially confines the SACs (**Figure 2**). As a proof-of-concept, Pt SACs on SnO_2 NSs exhibited high catalytic activity toward chemiresistive sensing of acetone gas (response = 95.4 at 10 ppm, 7.6-fold enhancement compared with pristine SnO_2 NSs) and unprecedented stability under highly humid conditions (27.4% response deterioration at 95% relative humidity).

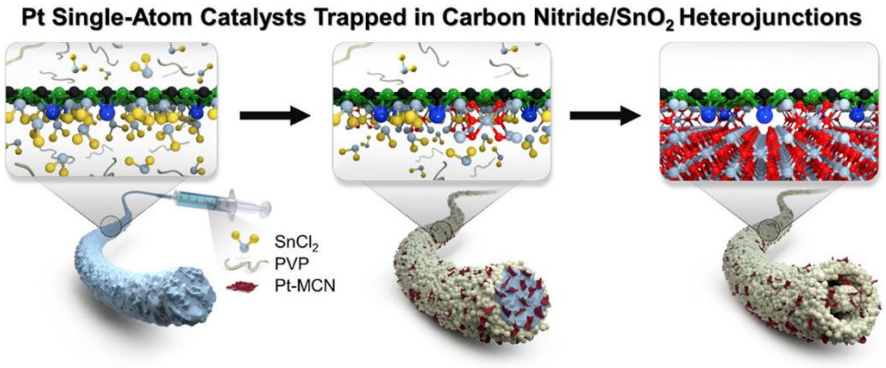

Fig. 1. Schematic illustration of the synthesis procedure for Pt SACs trapped at 1D carbon nitride/SnO_2 nano-heterostructure.

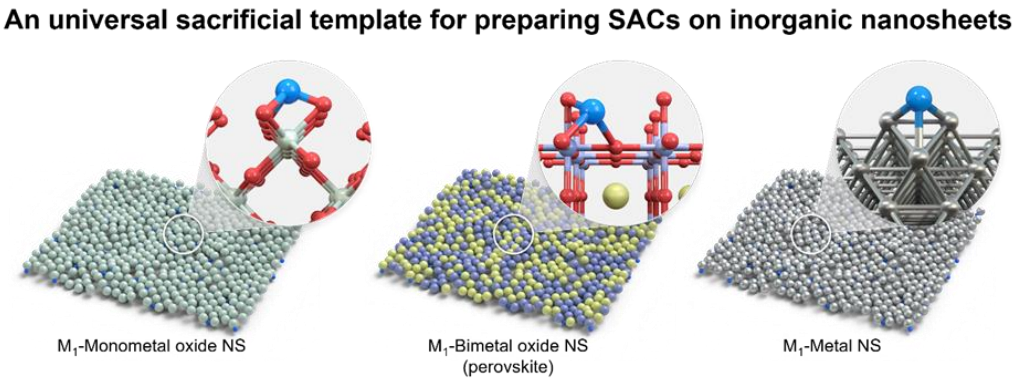

Fig. 1. Schematic illustrations of the inorganic NS supports for SACs, synthesized via sacrificial templating route.

[1] Wang A, Li J, Zhang T. Heterogeneous single-atom catalysis. Nat. Rev. Chem. 2, 65-81 (2018).
[2] Shin H, et al. Single-atom Pt stabilized on one-dimensional nanostructure support *via* carbon nitride/SnO_2 heterojunction trapping. ACS Nano 14, 11394-11405 (2020).