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Site Selective Growth of Noble Metal Atoms on Two-dimensional MoS_2 Nanosheets

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Two-dimensional (2D) MoS_2, with high surface area and unique electronic properties, has recently attracted intensive attention for applications in nanoelectronics and optoelectronics [1]. Such materials, however, have been utilized for catalytic reactions for decades [2]. The recent development of single-atom catalysis [3] has created opportunities for tuning the performance of conventional catalytic materials. For example, by doping MoS_2 nanolayers with Co [4] or Pt [5] the catalytic performance of the MoS_2 nanolayers is significantly modified. It has been shown that the active sites of MoS_2 nanosheets are the edges, making large, perfect MoS_2 layers ineffective for catalytic applications [5]. We report here the synthesis of edge-rich MoS_2 nanolayers and the interaction of noble metal atoms with these nanolayers. Our results show that Ir atoms preferentially occupy the sites on the basal planes of MoS_2 and may form sub-nanometer clusters. On the other hand, Pt atoms only decorate the edges of MoS_2 nanolayers. Such a different adsorption behavior of metal atoms warrants targeted approaches to synthesizing MoS_2 supported single-atom catalysts (SACs).

The MoS_2 nanosheets were prepared via reported procedures [6]. Typically, 1 mmol of hexaammonium heptamolybdate tetrahydrate ($(NH_4)_6Mo_7O_{24}.4H_2O$) and 30 mmol of thiourea were dissolved in 35 mL of deionized water under vigorous stirring to form a homogeneous solution. Afterwards, the solution was transferred into a 45 mL Teflon-lined stainless steel autoclave and maintained at 220 ^oC for 18 h. After the reaction system was cooled to room temperature, the precipitates were thoroughly washed with water and ethanol, followed by drying at 60 ^oC overnight. The as-prepared MoS_2 nanosheets were re-dispersed in water and sonicated for several hours to form MoS_2 nanoflakes with high number density of exposed edges. Strong adsorption method was used to disperse Ir or Pt salt precursors onto the MoS_2 nanoflakes. Aberration-corrected STEM-HAADF, indispensable for investigating the atomic structure of nanostructured materials, especially SACs [3, 7], was used to examine the atomic structures of the MoS_2 nanoflakes and the spatial distributions and locations of the dispersed Ir and Pt atoms.

Figure 1a shows the typical morphology of the synthesized 2D MoS_2 nanoflakes, revealing the thicknesses of the 2D layers (inset). Figure 1b displays the dispersion of the Pt atoms (indicated by the red arrows) which preferentially decorated the edges of each layer of the MoS_2 nanoflakes while no Pt atoms were anchored onto the MoS_2 basal planes. When Ir atoms were dispersed onto the MoS_2 nanoflakes they selectively grew on the MoS_2 basal planes and formed small clusters with a tendency of epitaxial relationship. The Ir atoms did not decorate the edges of the MoS_2 nanoflakes. Figure 2 shows schematic illustrations of the Pt and Ir adsorption sites. Such differences in adsorption of metal atoms can be utilized for tuning catalytic performances of targeted catalytic reactions. The catalytic behavior of such site-selective noble metal atoms on MoS_2 nanoflakes will be discussed [8].

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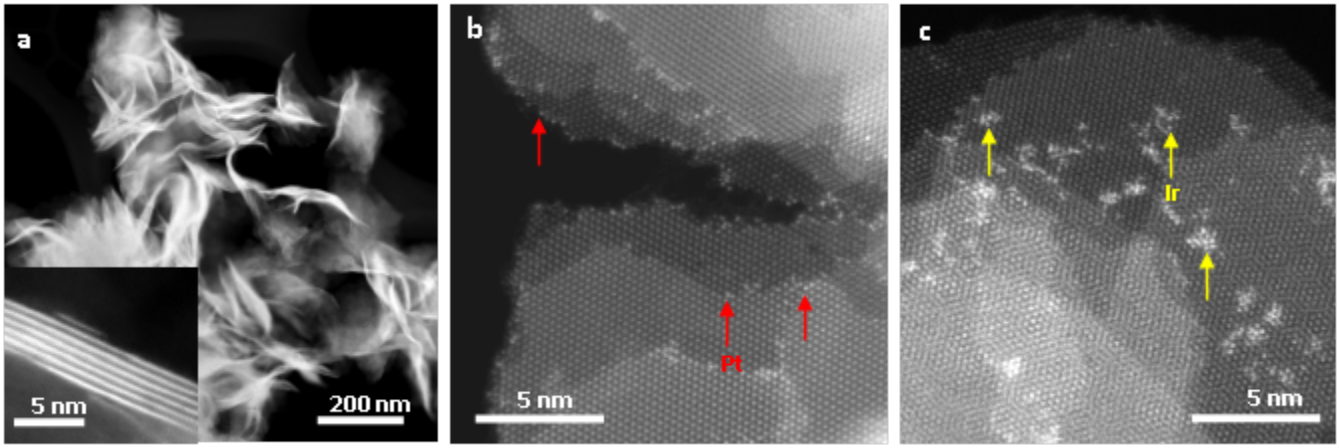

Figure 1. Low magnification HAADF images (a) clearly show the morphology and layers (inset) of typical 2D MoS_2 nanoflakes. High magnification HAADF images of Pt/MoS_2 (b) and Ir/MoS_2 (c) nanosheets clearly show that the Pt atoms decorated the edges while the Ir atoms and clusters positioned on the basal planes of the MoS_2 nanosheets.

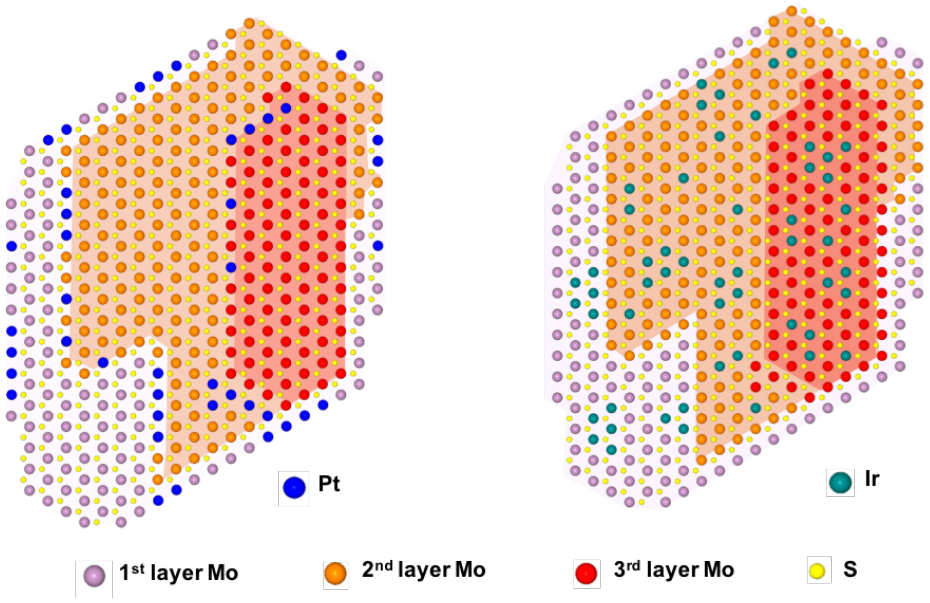

Figure 2. Schematic illustrations of Pt/MoS_2 and Ir/MoS_2 nanostructured catalysts: Pt atoms only decorate the edges of the MoS_2 monolayers while Ir atoms position themselves onto the basal planes of the MoS_2 nanoflakes.

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