Highly Dispersed Noble Metal-Based Carbon Nitride as Efficient Electrocatalytic/Photocatalytic Hydrogen Evolution Reaction Catalysts

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Abstract

Hydrogen (H_2) , with a high energy density and environmental friendliness, is considered one of the most promising clean energy carriers. Water splitting to generate H₂ by electrocatalytic/photocatalytic processes is a promising technology for renewable energy storage. However, the widespread deployment of water electrolyzers requires the development of efficient and stable catalysts containing minimal platinum group metals (PGMs) for the hydrogen evolution reaction (HER). Electrocatalytically active catalytic species can be used as co-catalysts in photocatalysis processes [1]. Hence, advanced catalysts combining high electrocatalytic performance with high photocatalytic performance are highly desired. Ruthenium and iridium (Ru, Ir)-based materials have drawn increasing attention for catalyzing the HER, which have been demonstrated both theoretically and experimentally to possess good HER performance [2,3]. Nevertheless, the utilization efficiency of noble metals seems still unsatisfactory, which is restricted by the relatively large particle size, wide size distribution, and high loading mass to achieve decent HER performance. Despite some recent progress in electrocatalytic HER using Ru and Ir-based catalyst, the use of Ru and Ir in visiblelight-driven photocatalytic HER processes has been hindered by the inefficient transfer of electrons from the photosensitive material to the metal. Graphitic carbon nitride (gCN) has attracted increasing interest owing to its suitable bandgap (2.7 eV), excellent stability, and the abundance of free N-containing groups on its surface, which can anchor the metal species [4]. However, there are by far only a few reports on highly dispersed Ru and Ir stabilizing on the gCN support for photocatalytic HER. In this presentation, we will show our recent progress in making atomically dispersed Ru and Ir catalysts supported on gCN (Ir-gCN, Ru-gCN) by a simple and cost-effective hydrothermal approach. Thanks to the strong metal-support interaction (SMSI) between the noble metal and gCN, Ru and Ir are dispersed on gCN in the form of single-atoms and ultrafine sub-nanometric clusters. Impressively, the Ir-gCN and RugCN catalysts with ultra-low metal loading exhibit superior catalytic activities for both electrocatalytic and photocatalytic HER, which can be attributed to the ultra-small catalyst size and high degree of dispersion of the active sites. Significantly, excellent durability has been also achieved benefiting from the SMIS. This study provides a potential strategy for developing high-efficiency, stable and multifunctional catalysts for electrochemical energy conversion.

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