Enhanced Photocatalytic Activity of Graphitic Carbon Nitride Modified with Single Atom Nickel Studied by X-ray Spectro-microscopy

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Abstract

Recently, use of single atom metals to modify greatly the graphitic carbon nitride (g-C_3N_4) has been widely studied. Single-atom metals possess the ability to separate the charges in aromatic conjugated rings by breaking coordination bonds in the microstructure. However, metal ions based relative polymerization and its subsequent charge separations have been sparsely reported. In this work, single-atom Ni is used as a polymerization capping agent, and the capping agent is usually selective to synthesize the specific noble metal nanocrystals by chemical adsorption route to control interfacial free energy. Scanning Transmission X-ray Microscopy (STXM) and Near-Edge X-ray Absorption Fine Structure (NEXAFS) was performed to measure the chemically imaged carbon, nitrogen and oxygen K-edges which illustrate that the monoatomic Ni as a capping agent for the polymerization reaction. A novel coordination structure is generated in the midgap state for the reduction reaction in order to prevent the recombination of electrons and holes. By tuning sp^2 coordination allows the efficient charge separation and charge transfer for enhanced photocatalysis. This study demonstrates the monoatomic Ni/g-C_3N_4 to be a potential candidate and versatile component for future overall water splitting from atomic and electronic perspectives.