Abstract

Solar energy is by far the most abundant renewable resource available to mankind. However, it is diffused and intermittent, and often geographically separated from that of the production results in underwhelming utilization of this resource. Inspired by photosynthesis, various efforts were made to store solar energy in form of chemical bonds than can be used when the sun is not shining. A promising approach is to produce hydrogen, a carbon-neutral energy carrier is via water splitting which requires an electrocatalysts to accelerate the two half-cell reactions, hydrogen evolution reaction (HER) and oxygen evolution reaction (OER). The state-of-art catalysts used for HER is Pt and for OER is IrO_2/RuO_2 that are prohibitively expensive which seriously undermines the potential of water electrolysis.

We have developed new synthesis methodologies for various earth-abundant electrocatalysts supported heteroatom-doped carbon nanostructures and exploited for water splitting. An in-situ solid state route was developed to integrate ruthenium nanoparticles with N-doped graphene sheets which exhibited an HER activity rivalling state-of-art Pt/C over a wide pH range. In order to find further cost-effective materials, we sought inspiration from NiFe-hydrogenase (the most efficient catalyst for HER) to develop a general solid state method for bimetallic MFe@ N-doped carbon core-shell nanostructures (M = Ni, Cu, Co, Zn, Mn) as efficient total water splitting catalyst. Thereafter, a new, phosphine-free, solid state method to hybridize Co_2P with N, P co-doped CNTs was developed as an efficient bifunctional electrocatalyst for both HER as well as OER, thereby effecting total water splitting. Moreover, glucose oxidation was attempted as a possible replacement for the kinetically sluggish OER half-cell reaction, wherein Co_2P/N, P-CNTs were demonstrated to be an efficient non-enzymatic glucose sensor for the first time. Thereafter, as an attempt to maximize the exposed edge sites in MoS_2 (which are considered to be active for HER),

we synthesized vertically aligned MoS_2 nanosheets directly on a Mo foil. The foil not only acted as a conducting support but also as a source of Mo during the reaction. When directly used as a cathode for HER, excellent HER activity was observed. Finally, an in-situ strategy was developed to hybridize N-doped graphitic carbon sheets with Ni and Mo_xC (Mo_2C and MoC) nanoparticles which exhibited resilient HER activity besides effectively accelerating OER, thereby resulting in overall water splitting that can be attributed to favorable electronic modulation between various strongly coupled components.