Nanomaterials for Heterogeneous Catalysis in Environmental Remediation: From Metal to Metal-free Carbon

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Abstract

The increasing discharge of wastewater containing high concentration of organic contaminants and heavy metal ions has post great threat to human beings because of their intrinsic toxicity and reluctance to natural biodegradation. One of the emerging protocols is the advanced oxidative processes (AOPs). However, most of the studies were carried out using metal catalysts, secondary contamination might occur because of the inevitable metal leaching during the catalytic oxidation. Recently, metal-free carbon materials, such as graphene, carbon nanotubes, nanodiamonds, or mesoporous carbons, have been demonstrated to be excellent alternatives to metal-based catalysts, like transition metal ions or oxides, for activation of peroxymonosulfate (PMS) to produce reactive radicals. The produced sulfate radicals can show appealing characteristics over hydroxyl radicals, e.g., a flexible pH range, a higher oxidation ability and the free of sludge, for degradation of organic pollutants in water. We firstly discovered that reduced graphene oxide (rGO) can provide an effective activation performance, which is comparable to cobalt oxide (Co_3O_4), in PMS activation. The active sites were proposed to be (i) sp^2 carbon framework, (ii) defective sites, and (iii) oxygen functional groups. Delicate material design and comprehensive experiments were carried out to prove the three proposed active sites. For example, nanodimonds with tailored graphitic shells were used to investigate the effects of sp^2 and sp^3 carbon. rGO with controlled defective degrees were employed to show the influence of edge defects of graphene on PMS activation. Amorphous carbon nanospheres were also prepared and used to study the oxygen groups as one of the origins of PMS activation, with combination of theoretical calculations. The hypothesis was well proved by the experiments and computational studies. The findings are critical for understanding the insights of carbon materials as alternatives to metal-based materials for environmental catalysis, leading to the development of novel, metal-free remediation technologies.