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| Title:                  | Metal-free heterogeneous photocatalysts for solar energy harvesting   |
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| Abstract:               | <p>The copious amounts of waste-plastic produced per year pose a serious threat to the environment due to their non-biodegradable nature and a tendency to absorb other pollutants, menacing the marine life, soil, and freshwater quality, and the entire food cycle. Therefore, the reduction of plastic-waste has emerged as a research priority, leading to many innovative approaches to convert them into useful materials such as liquid-fuels and related carbon-based materials. Carbon-based semiconductor photocatalysts are rare and their application is limited due to low visible light absorption and high charge recombination. Photocatalytic reactions are crucial because of their potential to carry out chemical transformations that can assist renewable fuel generation and production of fine chemicals by using inexhaustible and green solar energy. Thus, there remains a lack of efficient metal-free photocatalyst to achieve high catalytic efficiency. This thesis is mainly focused on the development of carbon-based semiconductor materials such as carbon quantum dots (CQDs), graphitic carbon nitride. This thesis describes a novel rational strategy for converting waste plastic into photocatalytic CQDs in a residue-free, inexpensive and impurity-free manner. I have further demonstrated a unique property that the CQDs exhibit an ability to harvest molecular oxygen from the air, thereby resulting in ultrahigh photocatalytic oxidation efficiency in chemical transformations under ambient conditions and natural sunlight. In this direction, I will be discussing the origin of such oxygen harvesting by CQDs. I also discovered that the amount of oxygen adsorbed on the CQD surfaces are different in the dark and under light illumination. The excitons generated under light induce desorption of the O<sub>2</sub> molecules, a phenomenon that we have named as 'light-induced hypoxia'. I have further demonstrated that when not performing any reaction, the CQDs exhibit self-sensitized photo-oxidation and produce CO<sub>2</sub>, thus removing themselves from the reaction media. This is a new material property that we term as 'CQD-autophagy' and beneficial as removal of photocatalysts is an expensive process in industrial-scale applications. I will also be discussing the greener strategy to exfoliate bulk g-C<sub>3</sub>N<sub>4</sub> into g-C<sub>3</sub>N<sub>4</sub> nanosheets and the visible light enhancement upon Au NPs loading on it. The origin of high catalytic activity in Au/g-C<sub>3</sub>N<sub>4</sub> is due to the dual role of Au NPs i.e Surface Plasmon Resonance induced 'hot- electron injection' and a reverse-electron-transfer mode leading to a pronounced 'co-catalytic effect'. I will also describe our efforts for the one-step synthesis of highly visible light active g-C<sub>3</sub>N<sub>4</sub> NSs and its photocatalytic H<sub>2</sub> evolution from the splitting of water.</p> |
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