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Title: Investigating the Sustainable Reutilization of Sulphuric Acid in Conversion of Waste Plastic into Luminescent Carbon Dots

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Abstract:

Over the decades, synthetic plastics have become everyday-use materials because of their low production cost, durability, moldability, etc. However, the rapid growth of plastic production combined with its short self-life leads to a significant accumulation of waste plastics, which causes severe environmental pollution. Usually, waste plastic comprises a substantial amount of carbon, especially olefines, which contain more than 86% of carbon atoms. Hence, transforming waste plastics into high-value-added carbon functional materials is a promising and yet not much-explored approach that Favors decreasing environmental pollution and resource recycling. Among different functionalized carbon materials, luminescent carbon dots are a new class of materials and have gained considerable attention from the scientific community due to their excellent and adjustable photoluminescent, in addition to extreme potential in diverse applications, including photocatalysis, optoelectronic devices, solar energy harvesting, biosensing, and imaging, etc. Our group recently explored and demonstrated a facile synthesis strategy for transforming waste plastics into highly photocatalytic active luminescent CDs. The methods involve the initial acid-mediated polyethylene (PE) plastic mass charring and a chemical oxidative fragmentation process. In reflux conditions, sulfuric acid converts PE plastic into a graphitic carbon with ample sp 3 defects. The graphitic regions are separated as ultra-small CDs by adding large quantities of KMnO 4 and H 2 O 2 to the charr. These PE-CDs exhibit three novel properties, i.e., (i) when a small quantity of CDs dispersed in an aqueous medium, it absorbs a large quantity of oxygen from the ambient air in a reversible manner, which can be utilized further to carry out efficient oxidative reactions under light conditions (ii) when there were no other reactants in the CDs solutions, it can attack the neighbouring CDs and converts them into simple molecules, i.e., CO 2 (self-elimination or autophagy), and (iii) under light conditions, the quantity of the O 2 on the CDs surfaces can be controlled (light-induced hypoxia). In the current approach, a large amount of conc. H 2 SO 4 and KMnO 4, along with water, are used to transform PE plastic mass into CDs and extract CDs from the reaction medium. The recovery and reutilization of acid in the subsequent batches of PE charring may not be possible due to dilution. Also, KMnO 4 converts to harshly toxic Mn +4 species during the process, and easy disposal is unfeasible. Therefore, we developed an ex-situ two-step approach to make this method sustainable and more industry-favourable. Initially, the PE plastic mass was transformed into graphitic charr by acid treatment, followed by separation from the acid and oxidization of the same using a green chemical oxidizer, i.e., H 2 O 2, to make XIICDs. We successfully reused the acid for seven cycles. However, the charring quality decreases as we increase the acid reutilization cycles. We studied the detailed synthesis and properties of the generated charr and CDs produced at different acid cycles. Compared them photocatalytic activities by taking rhodamine dye as a model pollutant. Finally, we showed the possibility of a large-scale synthesis of graphitic char and luminescent CDs employing this approach The potential of polyethylene-derived carbon dots (CDs) has two-fold solution: mitigating plastic pollution and enabling novel applications. We report a scalable method for synthesizing carbon dots using sulfuric acid. The H 2 SO 4 was successfully reused for seven cycles, demonstrating a sustainable approach. While the carbonization efficiency of H2SO4 decreased with reuse, the core properties of the CDs, as evidenced by UV-visible absorption and photoluminescence, remained consistent. Furthermore, we explored the scale-up feasibility by increasing precursor and H 2 SO 4 amounts by 50 and 5 times, respectively. This approach yielded highly efficient CDs (80%) that exhibited promising photocatalytic dye degradation and autophagy activity.

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