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Development of TiO_2/graphene nanocomposites as visible-light active photocatalysts for water treatment

Doctor of Philosophy (Materials Science)

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Research Content

Heterogeneous photocatalysis using titanium dioxide is a well-known advanced oxidation process for water treatment. However, the large band gap, a short lifetime of photo-excited electron-hole pairs, and the ability as an absorbent limit its applications. Hybridization of TiO_2 with graphene emerges as a promising approach to diminish these drawbacks. Many efforts have been reported on the preparation of TiO_2/graphene composites, but most of them utilized graphene oxide (GO) as a starting material. Subsequent reduction of GO into so-called reduced graphene oxide (rGO) leads to the formation of defect-rich graphene with disadvantageous electronic properties. Furthermore, the aggregation of TiO_2 is usually observed because the sensitivity of titanium alkoxide to water (GO usually contains) significantly impedes the uniform and controlled growth of TiO_2 on graphene. Hence, the aim of this thesis is to explore a novel and effective approach for the preparation of the TiO_2/graphene nanocomposites to obtain excellent visible-light photocatalysts for water treatment application.

In Chapter 2, a novel GO-free route for the fabrication of TiO_2/graphene nanocomposites was explored. This route involved the ultrasonication-assisted exfoliation of graphite in a titanium tetra-*n*-butoxide and subsequent sol-gel reaction to form TiO_2 using the graphene dispersion. Featured with various advantageous characteristics including i) a morphology in which TiO_2 nanolayers uniformly and thinly covered graphene sheets, ii) a trace amount of

defects on the graphene frameworks, iii) a dramatic inhibition of charge carriers recombination, and iv) a significant extension of absorption edge into visible-light area, the obtained the TiO_2/graphene nanocomposites exhibited an excellent performance for the visible-light photocatalytic decomposition of methylene blue in an aqueous medium.

Chapter 3 concentrated on the exploration of new solvents for liquid-phase exfoliation of graphite via ultrasonication. Various new exfoliating solvents were found through screening of different solvents and their mixtures. Particularly, chlorobenzylamine, which is a combination of the molecular structure of chlorobenzene and benzylamine, known as famous exfoliating solvents, presented the highest graphene yield. More importantly, a synergistic effect among different functional groups e.g. aromatic, amine, and halogen groups was identified, and this was more effectively exploited in a form of solvent mixtures. In addition, the preparation of a graphene dispersion in the presence of different metal alkoxides was demonstrated, which could be useful as a direct precursor of various oxide@graphene nanocomposites without mediating GO.

In Chapter 4, further improvement in the visible-light photocatalytic performance of the TiO_2/graphene nanocomposites was achieved by chlorine doping. The chlorine-doped TiO_2/graphene nanocomposites were synthesized based on the synthetic method established in Chapter 2. With the aid of chlorine radicals in accelerating the photodecomposition of target organic compounds and a significant reduction of the amount of graphene defects, the chlorine-doped TiO_2/graphene nanocomposites exhibited a significant improvement in the photocatalytic performance compared to that of the undoped TiO_2/graphene nanocomposite (Figure 1).

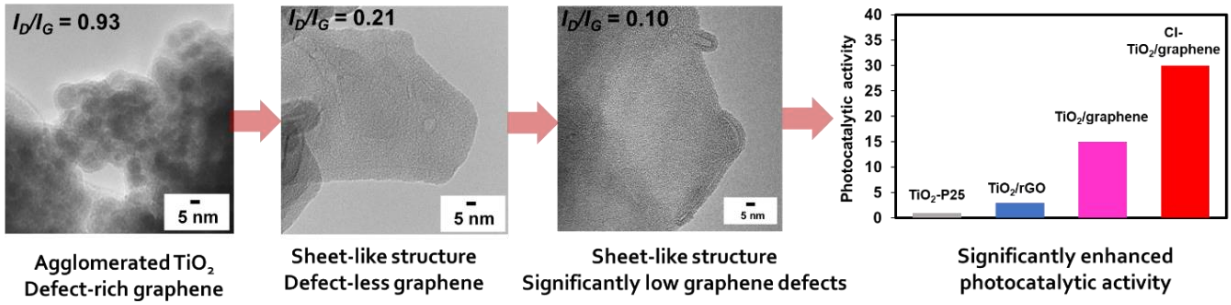

Figure 1. Development of TiO_2/graphene nanocomposites for the enhancement of visible-light photocatalytic activity.

Research Purpose

Because of the limitations of the conventional TiO_2/rGO nanocomposites which restrained their photocatalytic activity, in this research, I have found a novel GO-free route to synthesize TiO_2/graphene nanocomposites. Eliminating most of the problems of TiO_2/rGO, I have synthesized the TiO_2/graphene nanocomposites with an excellent visible-light photocatalytic activity.

Owing to my finding about the importance of solvents in the preparation of the nanocomposites, in which their ability to stabilize graphene played a key role, I have investigated deeper about the solvents for the liquid-phase exfoliation of graphite under ultrasonication. In this research, I have explored a series of new exfoliating solvents. More importantly, the relation between functional groups of solvents and their exfoliation ability has been found. A synergy between effective functional groups introduced not only in the molecular structure of single solvents but also more exploited through solvent mixtures. This finding is meaningful for chemists in investigating a proper exfoliation medium for specific purposes in term of the availability, diversity, and tunability.

According to the finding of the synthetic method for TiO_2/graphene nanocomposites as well as the available solvents for the preparation of the nanocomposite. With the ambitious to further improve the photocatalytic activity of the TiO_2/graphene nanocomposites by chlorine doping, I have investigated a novel synthetic method for Cl-doped TiO_2/graphene

nanocomposite. The Cl-doped TiO_2/graphene nanocomposites successfully prepared by this new method exhibited an excellent enhancement in the visible-light photocatalytic activity compared to the undoped ones.

In general, this thesis has established a novel and effective route for the synthesis of the TiO_2/graphene nanocomposites and demonstrated its usefulness in the field of water treatment based on excellent visible-light photocatalysis. It was also suggested that a similar method is applicable for the synthesis of different oxide@graphene nanocomposites with advantageous features.

Keywords

TiO_2/graphene, Chemical exfoliation, Sol-gel, Photocatalysis, Water treatment