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「課程博士用」

学 位 論 文 の 要 旨

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学位論文題目	Cu_2O based Visible-Light Photocatalysts for Degradation of Organic Pollutants (Cu_2O をベースとした可視光応答型光触媒による有機汚染物質の分解に関する研究)		

学位論文要旨

Nowadays, water contamination caused by organic pollutants is one of the main environmental issues. Its degradation over photocatalysts has been regarded as one of efficient and environmentally friendly methods. Among the numerous photocatalysts, Cu_2O is a p-type semiconductor with narrow band gap energy ranging from 2.0-2.2 eV, which could effectively utilize the large amount of visible light in the sunlight. Thusly, the Cu_2O based photocatalysts have been widely applied for the organic-pollutant decontamination nowadays. However, there are still many constraints on its application, such as the instability in oxidizing conditions, the rapid recombination of photo-generated holes and electrons. Therefore, further improvement of the Cu_2O photocatalytic performance is still required.

In this study, to improve the activity and stability of Cu_2O photocatalysts under visible light, a series of Cu_2O based photocatalysts were developed. Firstly, a kind of hyper-cross-linked polymer, named as KAPs-B (Knitting Aromatic compound Polymers-Benzene) with high surface area and special benzene rings structure, was loaded on Cu_2O to form a composite of KAPs-B/Cu_2O by using the precipitation method for the first time. The activities of various KAPs-B/Cu_2O composite photocatalysts with different loading amounts of KAPs-B were tested by methyl orange (MO) degradation. Comparing with the bare Cu_2O, significantly enhanced photocatalytic performances were found for the degradation of methyl orange (MO) over KAPs-B/Cu_2O under visible light. In particular, the 7% KAPs-B/Cu_2O showed the highest photocatalytic performance, by which approximately 92% of MO was decomposed within 60 min. Also, it showed an excellent reusability. It is found that the loading of KAPs-B on Cu_2O can not only effectively enhance the adsorption of organic matter on the catalyst ascribed to its high surface area and meanwhile stabilize the Cu_2O in oxidizing environments, but also enhance the separation rate of the photoinduced electron-hole pairs, leading to the photocatalytic activity and stability enhancement. These results indicate that it is a feasible method to improve the visible light photocatalytic performance of Cu_2O by compositing with the hypercrosslinked polymer.

Secondly, in order to further improve the performance of Cu_2O based photocatalysts under visible light, a kind of Metal-Organic Framework (MOF) named UiO-66-NH_2 with high surface area, good chemical durability and the adsorption ability for visible light was combined with Cu_2O to form the UiO-66-NH_2/Cu_2O composites with Z-scheme heterojunction. UiO-66-NH_2/Cu_2O composites with different loading amounts of UiO-66-NH_2 were successfully synthesized by using a facile impregnation process. The photocatalytic performances of the composites were measured by the MO degradation. It is found that the developed UiO-66-NH_2/Cu_2O composites possessed better photocatalytic behavior than the pure Cu_2O, and

the 20 wt% UiO-66-NH_2/Cu_2O photocatalyst showed the excellent photodegradation activity, by which 98.6% of MO could be decomposed within 50 minutes. In addition, it showed an excellent reusability since the severe photocorrosion issue of Cu_2O was suppressed significantly by combining with UiO-66-NH_2. The excellent photodegradation efficiency of 20 wt% UiO-66-NH_2/Cu_2O should be due to the synergistic effect between Cu_2O and UiO-66-NH_2, in which efficient interfacial charge transfer improved the separation effect of photoinduced electron-hole pairs within the Z-scheme heterojunction. The active species trapping experiments indicated that the photo-generated h^+ and $\bullet O_2^-$ radicals were the major active species involved in the MO degradation under visible light.

Meanwhile, in order to further enhance the dispersity of Cu_2O based photocatalysts in the organic pollution, Cu_2O with cubic structure was studied. In addition, for practical applications, tetracycline (TC) antibiotics was used as the degradation target. In the following work, a heterostructured graphitic-carbon-nitride-nanosheets/copper(I) oxide (g-C_3N_4 nanosheets/Cu_2O) composite was successfully prepared using a facile precipitation method and used for the degradation of tetracycline (TC) antibiotics. It is found that this composite had higher photocatalytic activity than the pure Cu_2O as well as the pure g-C_3N_4 nanosheets. Especially, the 30 wt% g-C_3N_4 nanosheets/Cu_2O composite exhibited the highest photocatalytic activity, by which 92.1% of TC was decomposed within 100 minutes and 83.3% of total organic carbon (TOC) was removed after the reaction for 120 min. In addition, it showed an excellent reusability, and the severe photocorrosion issue of Cu_2O was suppressed significantly by combining it with g-C_3N_4 nanosheets. The high photodegradation efficiency of g-C_3N_4 nanosheets/Cu_2O composite could be ascribed to the efficient interfacial charge transfer at the p-n heterojunction and synergistic effect between Cu_2O and g-C_3N_4 nanosheets, which resulted in the enhanced separation efficiency of photogenerated electron-hole pairs. In addition, the active species trapping experiments confirmed that the photo-generated h^+ and $\bullet O_2^-$ radicals were the main active species involved in the TC degradation under visible light.

All in all, in this work, Cu_2O based photocatalysts were successfully developed for photocatalytic degradation of organic pollutants and the corresponding photocatalytic mechanisms were also proposed. It is expected to give the guidance for the design, fabrication and application of Cu_2O based photocatalyst in the practical degradation processes of environmental pollutants.

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