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## Enhanced performance in phenol removal from aqueous solutions by a buoyant composite photocatalyst prepared with a two-layered configuration on polypropylene substrate



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#### ABSTRACT

A buoyant composite photocatalyst was prepared by immobilizing powdered activated carbon (PAC), followed by  $TiO_2$  nanoparticles (P25), in a two-layered configuration on the polypropylene granule (PPG) substrate. The PAC layer was anchored onto PPG surface through a thermal bonding process, and then the P25 layer was loaded on the PAC layer by a suspension deposition method. The PAC layer not only served as a protection layer for PPG but also provided the composite photocatalyst with adsorptive property to enhance its capability of collecting organic pollutants from the solution and thus enhance the photocatalytic degradation efficiency. The performance of the prepared composite photocatalyst was evaluated through a series of experiments, including its stability against mechanical attrition and photocatalytic degradation, its adsorptive performance and photocatalytic degradation efficiency in phenol removal. The results showed that the developed composite photocatalyst (denoted as PPG-PAC-P25) was much more mechanically and photocatalytically stable, as compared to those composite photocatalysts prepared from immobilizing a mixture of PAC and P25 together on the PPG, and also showed better photocatalytic degradation performance in phenol removal. A synergistic effect was clearly observed with the composite photocatalyst that had the two layered PAC and P25 configuration. PPG-PAC-P25 was also tested for 20 recycles in a batch process for phenol removal and the overall photocatalytic degradation performance was only slightly decreased by less than 7%, indicated that the developed buoyant composite photocatalyst in this study has a great prospect for possible actual applications in the removal of organic pollutants from water or wastewater.

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### 1. Introduction

Titanium dioxide (TiO<sub>2</sub>) has been extensively studied as one of the best photocatalysts, attributing to its high activity and stability, nontoxicity and low cost [1,2]. Various progresses in applying TiO<sub>2</sub>-based photocatalytic degradation process have been made in the last several decades [3–6], but there remain a number of major limiting factors that hindered the practical application of the technology in water and wastewater treatment so far. Those factors include the high cost and difficulty for post separation of the used TiO<sub>2</sub> nanoparticles from the treated effluent, the low UV light utilization efficiency commonly encountered in the slurry reactor system, and the relatively high rate of recombination of the electron/hole pairs associated with the TiO<sub>2</sub> photocatalyst [7–9]. To solve some of these

http://dx.doi.org/10.1016/j.jece.2015.10.034 2213-3437/© 2015 Elsevier Ltd. All rights reserved. problems, buoyant photocatalysts have been developed and studied, which can provide higher light utilization efficiency and easier posttreatment separation [10]. The floating photocatalysts were often prepared by immobilizing TiO<sub>2</sub> micro- or nano-particles onto a larger substrate of lower apparent density than water, such as hollow glass microsphere [11,12], polyethylene sheets [13], polystyrene beads [14,15], and polypropylene granules or fabrics [10]. Buoyant photocatalysts can allow photocatalytic degradation to take place on water surface and thus achieve greater utilization efficiency of the light provided because light attenuation is much lower in air than in water medium [10,14]. Enhanced oxygenation of buoyant photocatalysts at the water/air interface is also obtained due to the higher oxygen content at the water/air interface than that in water. Beside, buoyant photocatalysts can be easily separated from the treated water and thus eliminate their post separation concern. However, the use of those macro substrates may result in lower performance of the prepared photocatalysts, due to the limited amount of TiO<sub>2</sub> immobilized on the substrate [16,17], and the buoyant photocatalysts

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can face slower mass transfer rate for the organic pollutants in the bulk solution to the photocatalysts on the water surface, attributed to the large transport distance in the system and the non-adsorptive feature of the TiO<sub>2</sub> photocatalyst used [13,18]. The detachment of immobilized TiO<sub>2</sub> particles from the substrate sometimes may also become a concern during long periods of usage [19–21]. Although hollow glass microspheres as the substrate may provide excellent stability to photocatalytic degradation for the prepared product, the use of such substrate was generally expensive in the material as well as in the immobilization of TiO<sub>2</sub>, besides being very fragile in nature for handling. In contrast, low density thermoplastics, such as polypropylene (PP), have been more preferred for TiO<sub>2</sub> immobilization to prepared buoyant photocatalysts. The plastic substrates can offer the advantages of low price, reasonably good mechanical strength, good UV and/or chemical stability, and excellent processing flexibility in the final shape and dimension of the products. To minimize the effect of low mass transfer rate and thus increase the photocatalytic efficiency, co-adsorbents have been used together with the  $TiO_2$  photocatalyst, particularly such as combining  $TiO_2$ with powdered activated carbon (PAC) [8,22]. In our previous work, we have successfully prepared buoyant composite photocatalysts through a thermal bonding process, by immobilizing TiO<sub>2</sub> nanoparticles as the photocatalytic component and PAC as the coadsorbent in a mixture simultaneously onto the polypropylene granule (PPG) substrate [23]. The study showed that the PAC component in the composite photocatalysts helped concentrating the organic pollutants in the bulk solution to the vicinity of the photocatalyst particles and thus made the photocatalytic degradation process of phenol more efficient and less dependent on its concentration in the bulk solution. However, the buoyant composite photocatalysts prepared by simultaneously immobilizing PAC and P25 together showed some detachment of the P25 nanoparticles and thus unstable performance after extended long periods of usage, attributed to the slow photocatalytic degradation of the PPG substrate immobilized with the P25 particles in the application process of removing other organic pollutants. Therefore, it is of great interest to improve the photocatalytic degradation stability of the developed buoyant composite photocatalyst for its potential use in long term practical applications for organic pollutant removal in water or wastewater treatment.

In this work, a new method of preparing buoyant composite photocatalyst with better stability and performance was developed through a novel two-layered configuration, i.e., a PAC layer followed by a P25 layer, on the PPG substrate. PAC was used in this study not only as a good co-adsorbent, but also as an inert material that can resist radical attack to wrap and therefore protect the PPG substrate. Instead of directly immobilizing a mixture of PAC and P25 onto the PPG surface, an entire PAC layer was first anchored onto the PPG surface through a thermal bonding process that is similar to the one used before [23]. Then, another layer of P25 nanoparticles was loaded onto the PAC-immobilized PPG (denoted as PPG-PAC) through a suspension hydrothermal deposition method. The prepared buoyant composite photocatalyst (denoted as PPG-PAC-P25) was used for phenol degradation to evaluate its performance. A series of characterization analyses and photocatalytic degradation tests were conducted and the advantages of the buoyant composite photocatalyst with the two-layered immobilization configuration were examined.

#### 2. Materials and methods

#### 2.1. Preparation of buoyant composite photocatalysts

PPGs as the substrate were purchased from the Polyolefin Company (Singapore), with a cylinder shape of d = 3.5 mm and h = 2 mm. The TiO<sub>2</sub> nanoparticles (denoted as P25, Degussa) from

Germany was used as the photocatalyst component. P25 was specified with both the anatase (70–85%) and rutile (15–30%) composition, having a specific surface area of  $50 \text{ m}^2 \text{ g}^{-1}$  and an average particle size of 21 nm. Powdered activated carbon (denoted as PAC), obtained from Jacobi Carbon under the trade name of Aquasorb<sup>®</sup> CP1-F, was used as the adsorbent component. PAC was steam-activated from coconut shells and had a specific surface area of  $1050 \text{ m}^2 \text{ g}^{-1}$  and a nominal particle size of 325 meshes (around 44  $\mu$ m).

For easy comparison, three types of buoyant composite photocatalysts were prepared in this study. Type 1, denoted as "25%P25-PPG", was obtained by thermally immobilizing P25 and PAC simultaneously from a mixture (with P25:PAC = 15:45, i.e., 25% as P25 and the other 75% as PAC) onto the PPGs. Type 2, denoted as "100%P25-PPG", was obtained by thermally immobilizing P25 only onto the PPGs. The details on the procedures of the thermal immobilization method can be found elsewhere [23]. Type 3, denoted as PPG-PAC-P25, was the new buoyant composite photocatalyst with the two-layered configuration specifically developed in this study. In brief, PPGs were first immobilized with a layer of PAC only through the thermal immobilization method as for Type 1 and Type 2. The intermediate, denoted as PPG-PAC, after thoroughly washed and dried, was further loaded with P25 nanoparticles by a suspension hydrothermal deposition method. To do this, a well-dispersed P25 nanoparticle suspension was prepared in a hydrothermal reactor (900 mL, Berghof Br900, Germany) with isopropyl alcohol (IPA, AC grade, from Tedia) as the solvent [10]. 15 g of P25 and 300 mL of IPA were added into the reactor vessel. The reactor was tightly closed and the contents in the reactor were stirred with a PTFE lined stirrer bar at 500 rpm and heated to 180 °C on a hotplate stirrer (Heidolph, Germany). The process was continued for 4 h and then the contents in the reactor were slowly cooled down to the room temperature (22–23 °C). The hydrothermal treatment of P25 with IPA was to increase the stability of the suspension [24], and also to possibly enhance the interaction of P25 with PAC for immobilization [25-27]. Then, 20 g of the PPG-PAC were soaked into 50 mL of the above prepared P25 suspension for 30 min in a 250 mL beaker that was covered with a piece of aluminum foil and stirred at 250 rpm with a PTFE lined magnetic stirrer for P25 particles to deposit on the PAC. Finally, the P25-loaded PPG-PAC granules were separated from the solution, slowly dried in the fume hood with medium ventilation, and subsequently cured in an oven, with a programmed heating process from 80 to 145 °C, and then stayed at 145 °C for 90 min. The soak-dry-cure cycle was repeated for another round for more P25 nanoparticles to be loaded onto PPG-PAC. All the three types of prepared buoyant composite photocatalysts were thoroughly washed with tap water to remove any possible loosely loaded particles on the PPGs before they were dried in an oven at 80 °C to constant weights and stored in a desiccator for further uses in analyses or phenol removal experiments.

#### 2.2. Characterization of prepared composite photocatalysts

The actual compositions of the prepared PPG-PAC-P25, 25%P25-PPG and 100%P25-PPG buoyant composite photocatalysts, as well as those of the PPG substrate and PPG-PAC intermediate, were analyzed using a thermal gravimetric analyzer (TGA, TGA2950, DuPont Instruments, USA). A sample of around 7 mg of each type of the materials was examined and their weight variations as a function of the furnace temperature were obtained from the builtin balance. The TGA furnace was first heated to 400 °C at a rate of 20 °C min<sup>-1</sup> with nitrogen as the carrier gas for the decomposition of easily degradable organic constituents (i.e., PPG substrate and other organic impurities in PPG). The carrier gas was then quickly changed to compressed air and the furnace was further heated to Download English Version:

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