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Carbon nanotube/titania composites prepared by a micro-emulsion method exhibiting improved photocatalytic activity

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ABSTRACT

A micro-emulsion method was used to prepare TiO_2-coated carbon nanotubes (ME-TCNTs). The physico-chemical properties of these composites were characterized by modern analytical tools. Crystalline TiO_2 within the composites was composed solely of anatase. Carbon nanotubes (CNTs) within the composites were virtually uniformly covered with TiO_2. CNT addition increased the surface area and the amount of hydroxyl groups on the composite surface, which suppressed the recombination of photo-generated electron/hole pairs (excitons). The composite photoactivity was greater than that of pure TiO_2 and the commercial photocatalyst P25 for methylene blue degradation, predominantly due to the effect of CNT addition. An optimal TiO_2 loading of 12% was found to result in the highest photoactivity in comparison with other two loadings (6% and 15%). Too little TiO_2 or excessive CNT addition shielded the TiO_2 and reduced the UV intensity, due to photon scattering by the bare CNTs. However, a high TiO_2 content was found to be ineffective in suppressing exciton recombination because of the large distance between the TiO_2 and CNTs. The photoactivity of ME-TCNTs was greater than that of TiO_2 coated carbon nanotubes (TiO_2-CNTs) prepared by the sol–gel method. The uniform nanometer-scale TiO_2 layer on the CNT surface provided a high concentration of surface hydroxyl groups, a low exciton recombination rate and a high surface area.

1. Introduction

TiO_2 photocatalysis has been extensively investigated for the removal of organic contaminants in water and air, because of its strong oxidizing power, low toxicity and long-term photostability [1–5]. The photocatalytic activity of pure TiO_2 is insufficient for many industrial purposes [3]. Mixed-phase TiO_2-composites tend to show higher photoactivity than pure-phase materials, an example of which is commercial Degussa P25. This is due to the formation of solid–solid interfaces that facilitate charge transfer and separation, and reduce electron–hole (exciton) recombination and interfacial defect sites that act as catalytic “hot spots” [2–4].

Carbon nanotubes (CNTs) are considered to be a good support material for TiO_2, because they provide a large surface area support and also stabilize charge separation by trapping electrons transferred from TiO_2, thereby hindering charge recombination [5]. TiO_2 coated carbon nanotubes (TiO_2-CNTs) are widely investigated functional materials [6–10]. They have been fabricated by various methods, including the mechanical mixing of TiO_2 and CNTs [4], sol–gel synthesis of TiO_2 in the presence of CNTs [3], electro-spinning methods [6,7], electrophoretic deposition [7] and chemical vapor deposition [5]. The selected preparation method strongly affects the product’s physico-chemical properties [1]. The high surface area of TiO_2 is beneficial for photocatalytic activity, as it provides for a high concentration of target organic substances around sites activated by ultraviolet (UV) radiation [5]. CNTs coated with a uniform layer of anatase TiO_2 by a modified sol–gel process have been reported to exhibit an increased rate of aqueous phenol degradation compared with the individual components or simple component mixtures [7]. However, it was unclear why the TiO_2-CNT composite exhibited higher photoactivity. The uniformity of the oxide coating varies with preparation method, and uniform TiO_2 coatings on CNTs can be achieved by chemical vapor deposition and electro-spinning methods [5–7]. However, these techniques are not straightforward and require specialized equipment, and quantifying the composite ratio can be difficult. Sol–gel methods are generally preferable, although they usually lead to a heterogeneous, non-uniform TiO_2 coating with regions of bare CNT surfaces and random TiO_2 aggregation [3,9,10]. The micro-emulsification technique provides a controlled reaction rate, and is promising for synthesizing composites with high surface areas [11,12]. In the current study, we have demonstrated the feasibility of the micro-emulsion method for coating a uniform and well-defined nanometer-scale TiO_2 layer on CNTs. The photocatalytic activity of the composites was evaluated by the degradation of methylene blue (MB) in aqueous suspension. The

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