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Photoelectrocatalytic Electrodes with High Activity for Methylene Blue Degradation†

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In this study, a series of electrodes were prepared. The decrease of surface area compared with that of the pristine AC indicated the blocking of micropores on the surface of the activated carbons; was further supported by SEM, XRD, EDX. It was found that catalytic decomposition of methylene blue solution could be attributed to synthetic effects between the TiO_2 photocatalysis and electro-assisted activated carbons network and that photoelectrocatalytic oxidation increased with added of Co and graphene composition.

Key Words: Activated carbon/TiO_2 electrode, Photoelectrocatalysis, Visible-light, Ultrasonic, Methylene blue.

INTRODUCTION

The activated carbon/TiO_2 photocatalyst seems to have the application form of particles, which time consuming for the particle-fluid separation after radiation treatment for the reuse of the catalyst restrained its industrial applications. There were many reports on photoelectrocatalytic degradation of organic pollutants using TiO_2 composite electrodes, which were prepared by coating the surfaces of electrically conducting substrates with TiO_2 films[1,2]. In this method, a positive potential was applied on the working electrode, which could inhibit the recombination of electron/hole pairs and enhance the rate of photoelectrocatalytic degradation activity for organic compounds. Many studies have been devoted to fabrication of noble metal (such as Pt, Pd, Co, and Au) nanoparticle-decorated carbons as well as measuring their unique electrical, magnetic and optical properties[3]. Two-dimensional (2-D) graphene has emerged as high potential material and increasingly attracted attention owing to its fascinating physical properties including quantum electronic transport, extremely high mobility, high elasticity, and electromechanical modulation[4].

EXPERIMENTAL

Preparation of different composites and electrodes: Active carbon was mulled for 5 h and treated with phosphoric acid (0.1 M 1L), drying at 373 K for 5 h. Carbon granules were prepared. Since TOS was easily dissolved by oxydol solution, 6 g, 8 g, 10 g TOS were added to 300 mL of oxydol, respectively. After stirring for 1 h, TOS-H_2O_2 solution was obtained. Then the activated carbon granules were added to each TOS-H_2O_2 solution, respectively. Churn up the mixture for 1 h. Then the supported TiO_2 particles were first dried at 273 K for 6 h and calcined at 773 K for 2 h each. Activated carbon/TiO_2 photocatalyst composites were obtained. 55 g carbon granulers were mixed with 50 mL cobalt chloride solution (0.1 M). After strring 1 h and drying at 373 K, the mixture was subjected to heat treatment at 773 K. And then Co-AC compounds were formed. TOS treated method as above. The phenol resin was added into these different composites and the composites were pressed at a pressure of 250 kg/cm^2 in a mould into a dimension of 150 mm × 300 mm × 20 mm. The electrodes were obtained agree heat treatment at 673 K for 1 h.

Photoelectrocatalytic degradation: The photoelectrocatalytic degradation was performed using various electrodes in a container and then irradiating the system with UV light, visible light and ultrasonic. The distance from the light to the solution was 50 mm in darkness room. The counter electrode was artificial graphite with a dimension of 150 mm × 300 mm × 20 mm. The electrode was placed in 2 L of 1.0×10^{-5} mol/L methylene blue solution. The photoelectrocatalytic degradation of methylene blue was performed with a voltage of 6 V and irradiating. The blue colour of the solution faded gradually with time, owing to an adsorption and degradation of methylene blue solution. The concentration of methylene blue in the solution was determined as a function of irradiation time from the absorbance change at a wavelength of 660 nm.

RESULTS AND DISCUSSION

The micro-surface structures and morphologies of the four different composites were characterized by SEM (Fig. 1). The

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SEM technique is used for inspecting topographies of specimens at very high magnifications using a piece of equipment called the scanning electron microscope. Fig. 1 shows the macroscopic changes in the morphology of the composites. In Fig. 1 particles have a small particle size and a good dispersion and the fullerene particles are shown as spherical particles with small facets and a good dispersion. Zhang and Oh[4] reported that a good dispersion of small particles could provide more reactive sites for the reactants than aggregated particles.

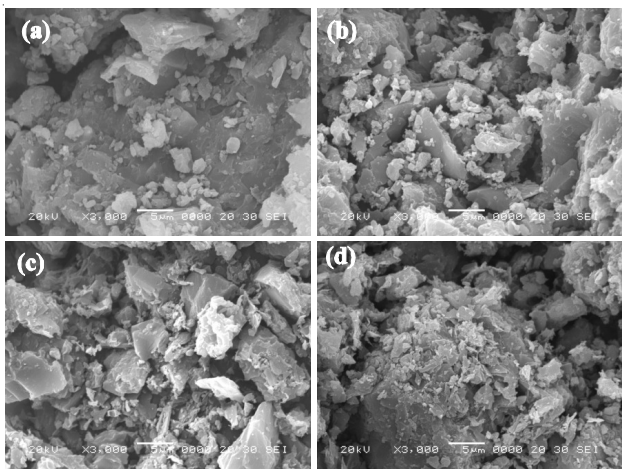

Fig. 1. SEM images of (a) AP3, (b) AOP3, (c) AGP2 and (d) ACGP

XRD was used to determine the crystallographic structure of the inorganic component of the composite. Fig. 2 shows the XRD patterns of the samples. In Fig. 2, A is anatase, C is carbon and G is graphite. The structure of AOP3, AGP2 and ACGP composites showed anatase TiO_2. The crystal structure of TiO_2 is determined mainly by the heat treated temperature. The peaks at 25.3, 37.5, 48.0, 53.8, 54.9, and 62.5º 2θ were assigned to the (101), (004), (200), (105), (211) and (204) planes of anatase. The peaks assigned of Co and Co_3O_4 can be found at ACGP composites.

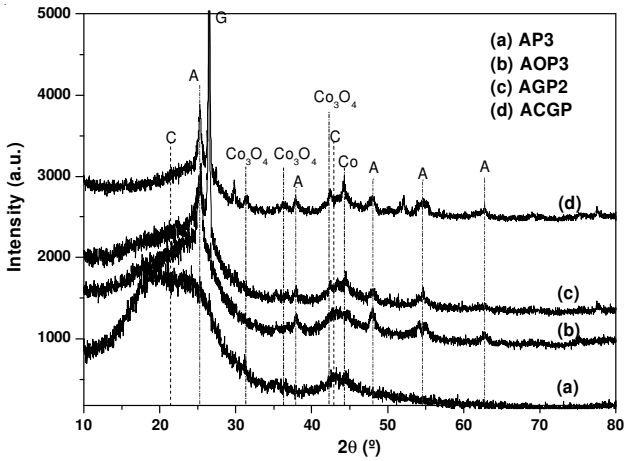

Fig. 2. XRD patterns of (a) AP3, (b) AOP3, (c) AGP2 and (d) ACGP, A is anatase, C is carbon and G is graphite

Photocatalytic studies: Fig. 3 shows the changes in relative concentration (C_t/C_0) of the different composites in methylene blue concentration of 1×10^{-5} under different conditions in the aqueous solution. Fig. 3(a) shows different electrodes degradation of methylene blue solution under electrolysis, sonolysis and photolysis condition (VL light). Fig. 3(b) shows different electrodes on the degradation of methylene blue solution under electrolysis, sonolysis and photolysis condition (UV light).

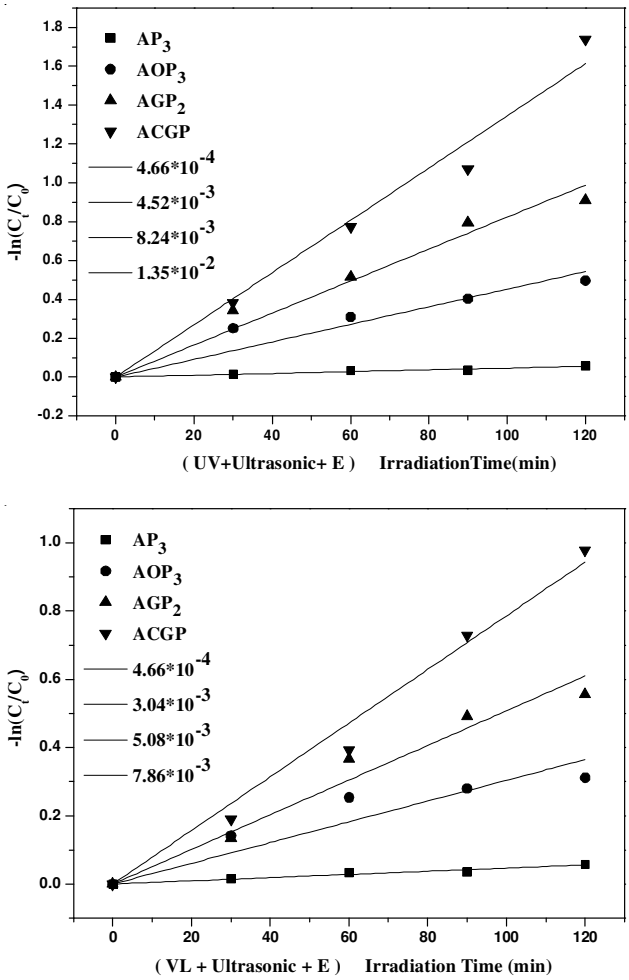

Fig. 3. Methylene blue solution on time of adsorption and visible light irradiation for different samples

Conclusion

The fabrication and characterization of the composites electrodes were presented. The BET surface areas for the AC/TiO_2 composites decreased with added of TiO_2 components. Among the four samples, the photoelectrocatalytic degradation efficiency of the sample ACGP was the best. The results demonstrated that the photoelectrocatalytic degradation of methylene blue solution could be attributed to synergetic effects of photodegradation of TiO_2, electro-assistance of AC networks, enhancement of Co, graphene and function of applied potential.

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