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Effect of Ag loading on activated carbon doped ZnO for bisphenol A degradation under visible light

Khanitta Intarasuwan [a], Pongsaton Amornpitoksuk [a,b,*], Sumetha Suwanboon [c], Potchanapond Graidist [d,e], Saowanee Maungchanburi [d], Chamnan Randorn [f,g]

[a] Department of Chemistry, Faculty of Science, Prince of Songkla University, Hat-Yai, Songkhla 90112, Thailand
[b] Center of Excellence for Innovation in Chemistry, Faculty of Science, Prince of Songkla University, Hat-Yai, Songkhla 90112, Thailand
[c] Department of Materials Science and Technology, Faculty of Science, Prince of Songkla University, Hat-Yai, Songkhla 90112, Thailand
[d] Department of Biomedical Science, Faculty of Medicine, Prince of Songkla University, Hat-Yai, Songkhla 90112, Thailand
[e] The Excellent Research Laboratory of Cancer Molecular Biology, Prince of Songkla University, Hat-Yai, Songkhla 90112, Thailand
[f] Department of Chemistry, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand
[g] Materials Science Research Center, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand

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ABSTRACT

Silver modified activated carbon doped zinc oxide (Ag/AC-ZnO) was synthesized via a calcination-electroless deposition route. The samples were characterized by X-ray powder diffractometry, scanning electron microscopy, transmission electron microscopy, X-ray photoelectron spectrometry, Fourier transform infrared spectroscopy and UV–vis diffuse reflectance spectroscopy. The photocatalytic activity of the Ag/AC-ZnO was evaluated for bisphenol A degradation in the presence of H_2O_2 under visible light irradiation. The archived results showed that the photocatalytic activity of the Ag/AC-ZnO was higher than that of AC-ZnO and pure ZnO. The cytotoxicity of the bisphenol A after photocatalysis under visible light irradiation was tested using L929 mouse fibroblast cells and the obtained results indicated that the treated bisphenol A solution exhibited no cytotoxicity against normal cells.

1. Introduction

Bisphenol A (BPA) is commonly used as the starting material in the production of polycarbonate plastics and epoxy resins. It is emitted into the environment during manufacturing process and leaches from products. BPA exhibits moderate acute toxicity to vertebrates [1] and when BPA discharged into the river as a result of industrial activity, it may affect aquatic animals and whoever drinks the contaminated water. BPA is a stable compound and does not readily degrade in the ecosystem so it must be treated before being released into the environment. Nowadays, there are many treatment processes to remove BPA from wastewater such as microfiltration, ozonation and biological methods [2]. One of the most effective treatments is an advanced oxidation process that the pollutant is removed by oxidation through the reaction with reactive species such as the hydroxyl radical or the superoxide anion radical. This treatment has many advantages over conventional processes, for example, it produces harmless degradation products, does not use expensive or hazardous oxidizing chemicals and takes advantage of natural sunlight. In the field of photocatalysis, zinc oxide (ZnO) is a good candidate because of its good photocatalytic property, high stability and eco-friendliness.

In the literature, ZnO based photocatalysts such as pure ZnO [3], carbon doped ZnO [4], Ag/ZnO [5] and Ce-ZnO [6] showed a high degradation potential for BPA in aqueous media under UV irradiation but there are only a few reported investigations of these photocatalysts under visible light irradiation. Qui et al. [7] reported that 93% of BPA was degraded after 4 h under visible light irradiation using N-doped ZnO and the visible-light response of this photocatalyst was attributed to the narrow band gap of the N 2p state isolated above the valence band of ZnO. However, modification of the band gap energy of the ZnO, in order to increase the visible light absorption capacity, is too complicated compared to a coupling method. Among the many coupling materials, it is well known that Ag is a good candidate sensitizer to make a wide band gap photocatalyst that functions in visible light. After exposure to

* Corresponding author at: Department of Chemistry, Faculty of Science, Prince of Songkla University, Hat-Yai, Songkhla 90112, Thailand.
E-mail addresses: 5710230028@email.psu.ac.th (K. Intarasuwan), pongsaton.a@psu.ac.th (P. Amornpitoksuk), sumetha.s@psu.ac.th (S. Suwanboon), gpotchan@medicine.psu.ac.th (P. Graidist), msaowane@medicine.psu.ac.th (S. Maungchanburi).

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visible light, according to the energy allowed transition, electrons from Ag can transfer to the conduction band of the ZnO and these injected electrons further react with O_2 to produce the superoxide anion radical ($\cdot O_2^-$). Furthermore, the photocatalytic activity of the Ag modified ZnO can be enhanced by inhibited charge recombination in Ag due to the electron injection. Another important factor in the photocatalytic process is the substance or pollutant adsorption on the surface of the photocatalyst. It has been demonstrated that the adsorption of pollutant molecules on the photocatlyst surface can significantly enhance the photocatalytic efficiency [8]. This property would be increased by the addition of porous materials to the photocatalyst. Activated carbon (AC) is a very cheap material that can increase the adsorption ability of many semiconductor-based photocatalysts [4].

Based on the above considerations, silver modified carbon doped zinc oxide (Ag/AC-ZnO) was prepared by a calcination-electroless deposition route. Furthermore, this work also examined the influence of Ag loading on AC-ZnO on the photocatalytic degradation of BPA under visible light irradiation. The overall cytotoxicity of the treated BPA solution after photocatalysis was assessed on L929 cells.

2. Experimental

2.1. Synthesis of Ag/AC-ZnO

AC-ZnO was prepared by the precipitation-calcination method. In a typical procedure, 20 mL of 0.25 M of $Zn(NO_3)_2{\cdot}6H_2O$ containing 0.01 g of activated carbon (QRëC) were added dropwise into 60 mL of 0.166 M $H_2C_2O_4$. In the preparation of the ZnO, the activated carbon was not used in this step. Next, this mixture was heated under vigorous stirring at 70 °C for 1 h in a water bath. After cooling to room temperature, the precipitant was filtered, washed with distilled water and dried at 100 °C for 1 h in a hot air oven. After that, the powders were calcined at 500 °C for 1 h in a muffle furnace.

Ag/AC-ZnO was prepared by a reduction from $[Ag(NH_3)_2]^+$ ions. This reagent was prepared by the addition of 3 mL of conc NH_3 to 40 mL of $AgNO_3$ in various concentrations. One gram of AC-ZnO powders (1 g of ZnO was used for the preparation of the Ag/ZnO) was stirred in a solution of $[Ag(NH_3)_2]^+$ for 15 min followed by the addition of a 1 M glucose solution and this mixture was continuously stirred for 1 h at room temperature. After that, the solid powder was filtered, rinsed with distilled water and dried at 100 °C for 1 h in an oven.

2.2. Characterization

The structural identifications of all samples were carried out using an X-ray diffractometer (XRD, X'Pert MPD, Phillips) with Cu K_α radiation at a wavelength of 0.15406 nm. The morphological studies were observed using a scanning electron microscope (SEM, Quanta 400, FEI) and transmission electron microscope (TEM, JEM-2010, JEOL). Elemental compositions of samples were analyzed by an energy dispersive X-ray fluorescent spectrometer (EDX, Oxford). The Fourier transform infrared (FT-IR) spectra of the samples in transmission mode at 400–4500 cm^{-1} were recorded by the FT-IR spectrophotometer (Spectrum BX, Perkin Elmer). The diffuse reflectance UV–Vis absorption spectra were obtained on a UV–Vis spectrophotometer (UV-2450, Shimazu) using $BaSO_4$ as a reflectance standard. The surface areas of all samples were evaluated by the BET (Brunauer-Emmett-Teller) method using a surface area analyzer (Autosorb 1 MP, Quantachrome). X-ray photoelectron spectroscopy (XPS) was performed on an AXIS Ultra DLD (Kratos Analytical Ltd.) electron spectrometer and all binding energies were calibrated to the C 1s line at 284.8 eV. Degraded products after photocatalytic reaction was characterized by liquid chromatograph-mass spectrometry (LC-MS, 2690-LCT, Waters, Micromass) using electrospray ionization operating in the negative (ESI^-) mode.

2.3. Photocatalytic study

The photocatalytic activities for all samples were evaluated from the degradation of the BPA under visible light irradiation in the presence of H_2O_2 (Fig. 1). Into a 250 mL beaker containing of 150 mL of 5 ppm BPA, 0.3 g of photocatalyst was dispersed. After that, 3 mL of H_2O_2 (30%, Merck) was added into the BPA solution. This mixture was stirred in the dark in order to establish adsorption/desorption equilibrium between the BPA molecules and the surfaces of the photocatalyst. The suspension was then exposed to visible light (51 W Xe lamp) for the required time. After the required time had elapsed, 2 mL of BPA solution was pipetted and centrifuged to completely remove the particles of photocatalyst. The concentration of the BPA was determined by UV–vis spectrophotometer (UV2550, Shimadzu) and the degradation of BPA

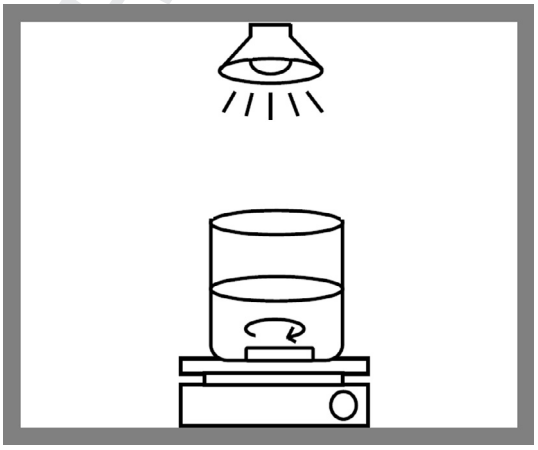

Fig. 1. Photocatalytic experimental setup.

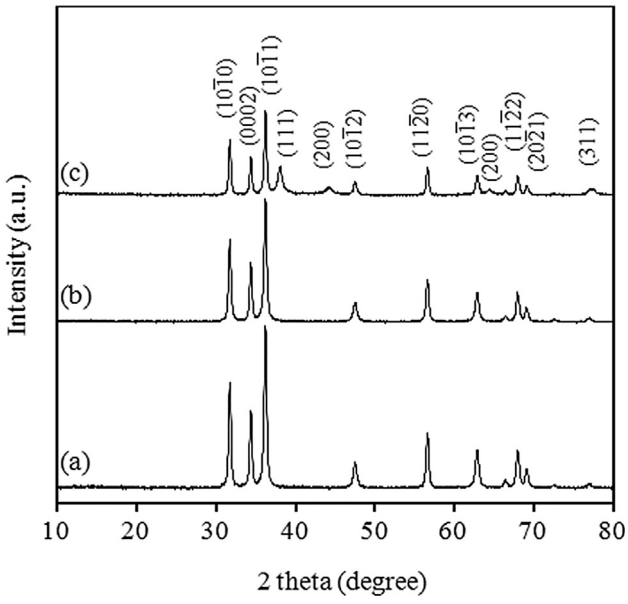

Fig. 2. XRD of (a) ZnO, (b) AC-ZnO and (c) 0.03Ag/AC-ZnO powders. The peaks labeled for three and four indices represent Ag (JCPDS 04-0783) and ZnO (JCPDS 36-1451), respectively.

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