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Enhancement of the tartrazine photodegradation by modification of silicon nanowires with metal nanoparticles

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ABSTRACT

In this work we investigated the tartrazine photodegradation by modified and unmodified silicon nanowires (SiNWs). SiNWs were elaborated by one-step metal-assisted electroless chemical etching of silicon substrate in $HF/AgNO_3$ aqueous solution. The modification of SiNWs was carried out by nanoparticles of platinum, palladium, silver, gold and copper in chemical solutions of PtO_2, $PdCl_2$, $AgNO_3$, $AuCl_3$ and $CuSO_4$, respectively. The results show that Cu-modified silicon nanowires give the highest photocatalytic activity compared to the unmodified SiNWs and SiNWs modified with nanoparticles of Au, Pt, Pd and Ag.

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1. Introduction

Dye pollutants in industrial waste water are the principal source of environmental aqueous contamination. There are several established ways to remove such contaminants namely: biological treatment, adsorption on activated carbon, hydrogen peroxide/UV light, Ozone/UV/hydrogen peroxide and γ_radiolysis [1,2]. Recently, heterogeneous photocatalytic oxidation, one of the Advanced Oxidation Processes (AOPs), has attracted much attention and has proved to be a promising technology for remediation of organic pollutants at ambient conditions [3–11]. Its advantage is the complete destruction compared with the other more established water purification methods. In this process, photogenerated charge carriers in a semiconductor and the formation of highly reactive chemical species such as hydroxyl radicals could mineralize a broad range of organic pollutants quickly and non-selectively.

In the present work, we employed this process for the photodegradation of tartrazine using modified and unmodified silicon nanowires as photocatalysts. The tartrazine is a synthetic lemon yellow azo dye known as E102, C.I. 19140, or FD&C Yellow 5 and trisodium-5-hydroxy-1-(4-sulfonatophenyl)-4-(4sulfonatophenylazo)-H-pyrazole-3 carboxylate. It is used as food colorant, in cosmetics, drugs, electroplating, pharmaceuticals and the textile industry [12,13]. Its structure is shown in Fig. 1. It is very soluble in water; this is the reason why it is commonly found in waste water. It seems to cause the most allergic and intolerance reactions, particularly among asthmatic patients, migraines, eczema, thyroid cancer, and lupus [12].

In the literature, there are very few reports on the tartrazine photodegradation. Indeed, Fragoso et al. investigated the degradation of tartrazine by oxidation with H_2O_2 in an alkaline solution [14]. Patel and Suresh investigated the decolonization of azo dyes (sunset yellow and tartrazine) using a magnesium/palladium system [15]. Teimouri et al. synthesized nikel-doped TiO_2 by a sol–gel method and studied the influence of ultrasonic irradiation on the photocatalytic degradation of tartrazine. Pertruta et al. studied the kinetics of tartrazine photodegradation by UV/H_2O_2 in an aqueous solution [16,17]. Wang et al. demonstrated that Pt/SiC NWs exhibit an enhanced photocatalytic activity (more than 88%) for water splitting compared with the pure SiCNWs, they found that Pt/SiC NWs will be a promising photocatalyst for water splitting [18]. Zhang et al. synthesized 3C-SiC nanowires with hierarchical structure which give an enhanced photocatalytic activity for the degradation of methylene blue solution under visible light irradiation [19]. However, the tartrazine photodegradation by SiNWs has not yet been reported in the literature.

In this study, silicon nanowires elaborated by metal-electroless etching were used as catalysts for the tartrazine photodegradation.

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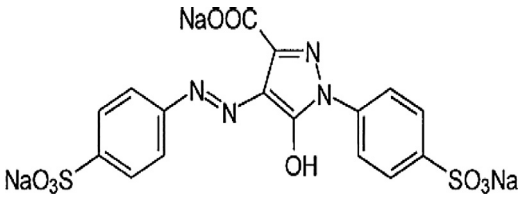

Fig. 1. Molecular structure of tartrazine.

The silicon nanowires were either unmodified or modified with metal nanoparticles using an electroless metal deposition technique (EMD).

2. Experimental

2.1. Materials

Tartrazine powder with a purity ≥95% was purchased from S.A color and was used without further purification. Silicon obtained from Siltronix was used to elaborate the photocatalysts. In this study, all chemicals such as copper (99.99%), palladium (II) chloride (99.99%), platinum oxide (99.99%), hydrogen fluoride (50%), oxygen peroxide (30%), nitrates of silver (99.8%), acetone (98%), ethanol (99.8%), sulfuric acid (95–97%) and nitric acid (69%) were obtained from Sigma–Aldrich.

2.2. Elaboration of SiNWs

Silicon nanowires were formed by one-step Ag-assisted electroless chemical etching of p-type (100)Si wafer with a resistivity of 7.7–8.66 ohm cm. The silicon wafers were cut into samples of $10 \times 10\,mm^2$ size. Before etching, the samples were cleaned with acetone and deionized water and then immersed into a solution of H_2SO_4 and H_2O_2 (volume ratio 3:1) to remove the organic contaminants from the surface. The thin oxide layer formed on the surface was then etched in a 10% HF solution for 3 min. The etching of samples was performed in 9.65 M HF-0.033 M $AgNO_3$ aqueous solution at 50 °C for 10 min. After the etching, the samples were covered with Ag dendrites and nanoparticles, which were removed by immersion into aqueous HNO_3 solution (69%) during 5 min. Finally, the samples were rinsed with deionized water and dried under a gentle stream of nitrogen.

2.3. Modification of SiNWs

Before the deposition of metal nanoparticles of Cu, Ag, Pd, Pt and Au, the SiNWs were immersed into a 10% HF solution for 3 min to remove the native oxide and provide hydrogen-terminated SiNWs (H-SiNWs). The H-SiNWs were then immersed into the solution containing the salt or the oxide of the element to be deposited. The depositions were carried out using the following chemical solutions:

- 0.14 M $CuSO_4$/1.35 M HF for 2 min at room temperature.
- 0.01 M $AgNO_3$/0.5 M HF for 1 min at room temperature.
- 1 mM $PdCl_2$/0.15 M HF for 40 min at room temperature.
- 3 mM PtO_2/0.15 M HF for 1 h at 50 °C.
- 1 mM $AuCl_3$/0.15 M HF for 10 min at room temperature.

2.4. Photocatalytic experiment

The tartrazine solution with an initial concentration of 1.035×10^{-5} M was prepared by dissolving tartrazine powder in deionized water under magnetic stirring at room temperature for 10 min. The photocatalytic degradation reaction was carried out at room temperature by immersing the photocatalysts into a 4 mL aqueous solution of tartrazine with an initial concentration of 1.035×10^{-5} M. The solution was irradiated with UV light ($\lambda = 365$ nm) for 200 min. The photocatalytic performance was measured by the decay of the absorption peak of tartrazine at 428 nm every 20 min.

2.5. Characterization

Scanning electron microscopy (SEM) images were obtained using a PHILIPS XL30 electron microscope. Diffraction spectra were obtained by means of a diffractometer (BRUKER axs D8 ADVANCE) with Cu Kα radiation ($\lambda = 1.54060$ Å). All diffraction peaks were identified using JCPDS databases. The measurements were carried out at a grazing angle. The absorption spectra of the tartrazine solutions contained in quartz cuvettes with an optical path of 10 mm were recorded using a Varian Cary 500 UV–vis spectrophotometer in the wavelength range 200–600 nm. The X-ray fluorescence technique was used for elemental analysis and chemical analysis (WDXRF Philips 2440 with anode Rh).

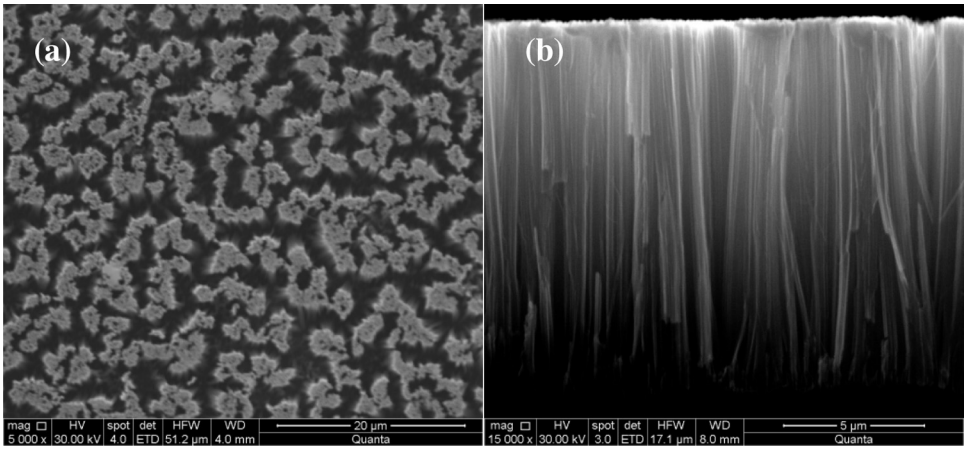

Fig. 2. Plan (a) and cross-sectional (b) view SEM images of SiNWs formed by chemical etching.

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