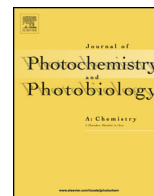




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Degradation by irradiation of an Acid Orange 7 on colloidal TiO₂/(LDHs)

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ABSTRACT

The photocatalytic efficiency of colloidal TiO₂ was considerably improved after its immobilization using synthetic layered double hydroxides (LDHs) as support of the photocatalyst. Compared to conventional colloidal TiO₂ nanoparticles (Q-TiO₂), LDHs present the advantage to enhance the contact between an organic pollutant and TiO₂. This work is focused at the same time on the synthesis of new materials (colloidal TiO₂/LDHs) and to enhance their efficiency for photodegradation in aqueous solutions of selected dye.

Q-TiO₂/Mg₂AlCO₃ and Q-TiO₂/Mg₂FeCO₃ composite photocatalysts were prepared by a direct coprecipitation method at constant pH and characterized by several analytical techniques including XRD, FTIR and BET. Subsequent to the dark adsorption studies, photocatalytic efficiency of the supported catalysts was evaluated using an azo dye, Acid Orange 7, as model compound in an aqueous medium. The supported catalysts exhibit good photodegradation efficiency of this dye and its removal was improved compared to bare colloidal TiO₂.

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1. Introduction

In recent years, colloidal solutions of titanium dioxide (Q-TiO₂) particles have been extensively investigated for the photocatalytic oxidation of organic compounds in organic media [1,2]. The assembly of nanosized TiO₂ onto porous solids represents a topic largely studied to develop improved heterogeneous supported catalysts [3,4]. The formed nanocomposites often exhibit large specific area and porosity and high adsorption capacity. These nanoparticles immobilized on a solid substrate are of great interest for the study of photocatalytic reactions in terms of the recovery of the catalyst and for limiting the aggregation of TiO₂ nanoparticles.

Advanced Oxidation Processes (AOPs) are alternative techniques of destruction of dyes as well as many other organics in wastewater and effluents. These processes generally involve UV/H₂O₂, UV/O₃ or UV/Fenton's reagent for the oxidative degradation of contaminants [5–7]. Semiconductor photocatalysis represents a newly developed AOP, which can be conveniently applied to dye pollutants for their degradation. Typically, this process is initiated by band-to-band excitation of semiconductor particles by irradiation, to generate •OH radicals derived from

valence band hole oxidation of terminal OH⁻ groups and hydration water on the particle surface [8]. Among the various photocatalysts employed, TiO₂ is the most preferable material due to its non-toxicity, insolubility, stability, high photoactivity and inexpensive nature. However, TiO₂ can only use a relatively small part (less than 5%) of the solar spectrum for photocatalytic oxidation, and artificial UV light sources are often unstable and relatively expensive, thus presenting main barriers to industrialize the photocatalytic oxidation processes of TiO₂. Moreover, commercial grades of TiO₂ usually possess small particle sizes so that TiO₂ particles are difficult to recover from the suspension, such as by filtration [9]. Recently, semiconductor/lamellar nanocomposites have been receiving much interest [10–12]. Miyoshi et al. [13] and Sterte [14] reported on the incorporation of extremely small particles of Fe₂O₃, and TiO₂, with diameters <1 nm, into the interlayer of a montmorillonite clay. These studies did not focus on the evaluation of photocatalytic degradation activities of contaminants of the retained semiconductor particles.

Layered double hydroxides (LDHs), also known as anionic or hydroxalcalite-like clays, represent a class of lamellar compounds that consist of positively charged brucite-like host layers and hydrated exchangeable anions located in the interlayer gallery for charge balance. LDH matrices can be easily prepared in the laboratory compared to cationic clays. Different synthetic methods have been reported in the literature [15,16] such as, for instance,

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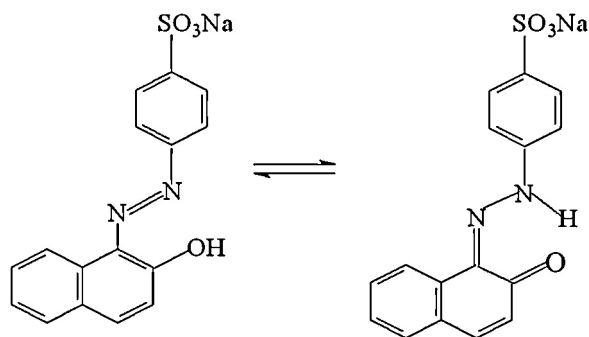


Fig. 1. Tautomers of the AO7 molecule; the azo form is shown on the left hand side and the hydrazone form is represented at the right hand side.

coprecipitation by constant pH-value, coprecipitation using a retardant base such as urea [17], or process involving preparation of polyol [18].

LDHs have been the subject of intense research because of their wide applications as catalysts [19], catalyst precursors [20], anion exchangers [21], acid absorbents [22], bioactive nanocomposites [23], and as electroactive and photoactive materials. The general formula of LDHs is given by $M^{(II)}_{1-x}M^{(III)}_x(OH)_2Y_{x/n}$ (abbreviated as $(M^{II}M^{III}-X)$ where $M^{(II)}$ and $M^{(III)}$ represent the divalent and trivalent cations, respectively; Y is the interlayer anion; and n is the charge of the interlayer anion.

In this work, we present a new method for preparation of an active colloidal TiO_2 /LDH nanocomposite photocatalyst, and results will be given for the photocatalytic degradation of Acid Orange 7. This azo dye was chosen as a model compound because it is representative of a large class of dyes used commercially.

2. Experimental

2.1. Materials

All solutions were prepared using deionized (18 m Ω) decarbonated distilled water. The organic pollutant used as model substance was an azo dye, Acid Orange 7 (AO7), also called Orange II, which was obtained from Sigma–Aldrich and used without further purification. AO7 has two tautomeric forms, the azo-form, A, and the hydrazone form, H, which are in equilibrium in solution (Fig. 1). This dye is very soluble in water; it contains a sulphonate bond which gives him the anionic character.

In the field of visible light, the dye presents a wavelength maximum at 483 nm. This dye does not react with acids and bases, neither with electro-magnetic irradiation in the wavelength of UV and visible light which makes its degradation a difficult task.

$MgCl_2 \cdot 5H_2O$ (Acros, 99%), $Al(NO_3)_3 \cdot 9H_2O$ (Acros, 99%), $Fe(NO_3)_3 \cdot 9H_2O$ (Acros, 99%), $TiCl_4$ (Aldrich, 99%) as well as NaOH (Acros, >97%) were used as received.

The hydroxide double lamellar compounds of composition $[Mg_{2.0}Al(OH)_{6.0}](CO_3)_{0.5} \cdot 2.2H_2O$; $[Mg_{2.0}Fe(OH)_{6.0}](CO_3)_{0.5} \cdot 2.2H_2O$ were prepared by a direct coprecipitation procedure [24]. The LDHs were designated as Mg_2AlCO_3 and Mg_2FeCO_3 , respectively.

2.2. Preparation of colloidal TiO_2 /LDHs photocatalysts

TiO_2 colloids were prepared from hydrolysis of $TiCl_4$. For rinsing purposes, triple distilled water was used. Concentrated $TiCl_4$ of 3.5 mL was added to 900 mL distilled water; drop-by-drop with vigorous stirring at 1 °C, and a transparent TiO_2 solution was obtained. For the realization of dialysis, three Spectra/Por membranes with a maximum volume of 400 mL were used. They were first washed

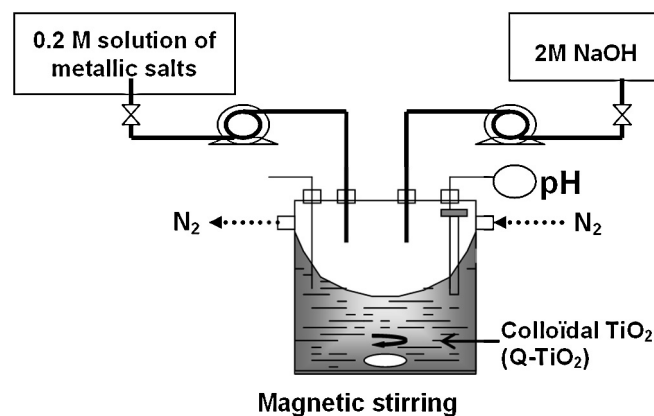


Fig. 2. Experimental setup for the synthesis of colloidal TiO_2 /LDHs.

with ultrapure water (Millipore), possessing a resistivity of 18 m Ω then 300 mL of colloids were introduced into each of the solutions. These three membranes were then placed in beakers containing 4 L of ultrapure water (18 m Ω). During dialysis, the ion exchange OH/Cl^- through the membrane leads to an increase of the pH of the colloidal solution of TiO_2 which was initially very acidic. Several replacements of the aqueous medium were carried out so that the pH of the solution was equal to 2.5. A total volume of 960 mL of the solution was maintained, corresponding to a colloidal TiO_2 concentration of $33.3 \times 10^{-3} \text{ mol L}^{-1}$. The pH adjustment to 9 using 2 M KOH was necessary to prevent the destruction of the structure of the adsorbent due to the potential reaction with acid.

The TiO_2 nanoparticles were characterized by a Zetasizer Nano instrument, yielding an average diameter of 12 nm. The composites $Q-TiO_2$ /LDHs were prepared by dropwise addition of a mixed salt solution ($M^{II} + M^{III}$) into a container with $Q-TiO_2$, paying particular attention not to exceed the stoichiometric ratio $Q-TiO_2$ /LDHs = 2/1 in order to maintain a constant pH value at precipitation. The experimental setup for the synthesis of colloidal TiO_2 /LDHs is shown in Fig. 2.

2.3. Characterization of catalysts

Powder X-ray diffraction (PXRD) patterns of the solid samples were recorded using a Siemens D501 X-ray diffractometer using $Cu K\alpha$ radiation ($\lambda = 0.154 \text{ nm}$) and fitted with a graphite back-end monochromator. The samples were scanned from 2° to 70° (2θ) using steps of 0.04° and a counting time per step of 8 s. Attenuated Total Reflectance Fourier Transform infrared (ATR-FTIR) spectra were measured in the range $400\text{--}4000 \text{ cm}^{-1}$ on a FTIR Nicolet 5700 (Thermo Electron Corporation) and a resolution of 4.0 cm^{-1} . The nitrogen adsorption/desorption isotherms were recorded on a Fison SP 1920 instrument, after outgassing of the samples at 80°C for 12 h.

2.4. Photoreactor and light source

The experiments were carried out in a photocatalytic oxidation reactor. In the center of the cylindrical reactor, three 6 W ($\lambda_{\text{max}} = 365 \text{ nm}$) black light fluorescent UV lamps were used as artificial light sources. A Pyrex cylindrical jacket located around the plugging tube was used as reactor containing a water cooling circuit to absorb IR radiation and thus to avoid heating of the solution. Constant stirring of the solution was maintained by a magnetic stirrer (Fig. 3). The disappearance of AO7 was monitored by measuring the absorbance by means of a UV–vis spectrophotometer (SAFAS UV mc 2) at a wavelength of 485 nm. The determined absorption was

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