



Photocatalytic removal of phenol using titanium dioxide deposited on different substrates: Effect of inorganic oxidants



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ABSTRACT

Photocatalytic degradation of phenol has been investigated using a laboratory reactor and UV-A lamp (BLB) and different commercial catalysts: TiO₂PC500 fixed on cellulose paper (Ahlstrom paper) and TiO₂ (Degussa P25) on stainless steel or conducting glass. Thin layers of TiO₂ on glass and stainless steel were prepared by electrophoretic deposition. Phenol and by-products were monitored by HPLC system equipped with a phenyl column and PDA detector. The effects of various operating parameters such as initial concentration of phenol (100–500 μmol L⁻¹), initial pH (2.0–10.0) and addition of inorganic oxidants (H₂O₂, S₂O₈²⁻) were evaluated on Ahlstrom paper. The results revealed that the best conditions of phenol removal were 100 μmol L⁻¹ of initial concentration, natural pH (5.0–6.0) and absence of inorganic oxidants when using Ahlstrom paper. Also, analysis of the by-products shows their dependence on initial pH. The comparison of phenol removal efficiency shows that thin layers of TiO₂ P25, deposited on glass, present the best photocatalytic activity. Several experiments performed with the same photocatalyst show that washing layers with pure water several times and drying at low temperature allow reactivation of the catalysts.

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1. Introduction

Phenolic compounds are an important family of chemicals used in chemical, petrochemical and plastics industries. They are highly water-soluble and toxic to living organisms [1]. So they are easily found in groundwater which is one of the main sources for drinking water production [2]. Among the phenolic compounds, phenol with the simplest structure, has the highest solubility [3]. To remove from contaminated water, these toxic and refractory compounds that constitute a public health problem, various technologies are used including adsorption on activated carbon, biological treatment etc. However, these methods are not able to totally remove the pollutant. They are not efficient enough due to phenol toxicity [4]. Advanced methods of oxidation can be an alternative for the effective removal of these pollutants from liquid effluents. Photocatalysis based on titanium dioxide is an especially attractive method that has been at the center of much work due to

its effectiveness in degrading several organic compounds up to mineralization. Photocatalysis is one of the advanced oxidation processes (AOPs) based on the generation of hydroxyl radicals at ambient temperature which oxidize the organic compounds. Indeed, when the photocatalyst is illuminated with light whose energy is higher than its band gap energy, it may result in electron-hole pair production in the material [5]. The species formed will lead to the formation of primary oxidants such as hydroxyl radicals. Titanium dioxide can be used in slurry form (suspension) or as a thin layer deposited on an appropriate support. Several deposition dimensional structures can be obtained which provide different properties of these materials for novel applications [6]. Among the most used supports, we have: steel [7,8], conducting glass [9–10], cellulose paper [11], silica fiber [12] and activated carbon [13]. Despite the effectiveness of the heterogeneous photocatalytic process with the TiO₂ slurry system, the operating cost of total mineralization of hazardous organic effluents remained high. This is due to serious drawbacks found in the conventional TiO₂ slurry system that limit its practical application. Mainly, the settling velocity of aggregated TiO₂ (average diameter of 0.2 μm) is very low, requiring a long

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retention time in the clarifier or functioning of filtering system. Other drawbacks, such as the dosage of TiO_2 increase in order to raise the photocatalytic rate and the high turbidity created by the high TiO_2 concentration can decrease the depth of UV penetration [14]. This “shadowing effect” can drastically lower the rate of photocatalytic reaction. Therefore, attention has turned to the immobilization of TiO_2 on solid carriers to eliminate the need for a follow-up clarification technique and to avoid an unnecessary excess of catalyst [15]. Electrophoretic deposition (EPD) is a simple method that can be used with conducting supports [9,16,17].

The recombination of electron–hole pairs is a factor limiting the effectiveness of the catalytic phenomena [14]. In order to avoid electron–hole pair recombination which limits quantum efficiency and to increase photocatalytic efficiency, inorganic oxidants (electron acceptors) [3,18] such as hydrogen peroxide H_2O_2 and persulfate $\text{S}_2\text{O}_8^{2-}$ can be added to the reaction mixture. They are characterized by high redox potential, are traps for photogenerated electrons and can also be activated by photochemical processes resulting in the formation of radicals such as hydroxyl radical $\bullet\text{OH}$ and sulphate radical anion $\text{SO}_4^{\bullet-}$. These radicals participate in oxidation reactions according to the processes of hydrogen abstraction, hydrogen addition, or electron transfer. However, the $\text{SO}_4^{\bullet-}$ radicals are more selective in oxidation reactions (electron transfer) than $\bullet\text{OH}$ radical and can therefore increase the rate of organic compounds removal [19,20]. Several research papers reported the use of these inorganic oxidants with TiO_2 in suspension but few publications focus on their use in the case of thin layer of titanium dioxide, especially in the case of $\text{TiO}_2\text{PC500}$ coated on non woven paper (Ahlstrom paper) [11,21]. The purpose of this study is to evaluate both the effectiveness of Ahlstrom paper (commercial catalyst) in the removal of phenol from aqueous media according to different parameters (initial pHs, concentration of pollutant, inorganic oxidants) and on the other hand to compare its photocatalytic activity to that of TiO_2 P25 deposited on stainless steel and conducting glass.

2. Materials and methods

2.1. Reagents

All reagents were used without pre-purification. Phenol (ACS grade), methanol (HPLC grade) and sodium persulfate (99%) were supplied by Carlo Erba. Hydroquinone ($\geq 99.5\%$), resorcinol (98%), catechol (99%) and benzoquinone ($\geq 98\%$) were purchased from Sigma–Aldrich. Hydrogen peroxide (30%w/w), the acetylacetone ($\geq 99.5\%$) and the surfactant Triton X 100 are from Fischer Scientific. TiO_2 P25 with 80% anatase and 20% rutile was obtained from Degussa. The Ahlstrom reference 1048 paper (P) is provided by Ahlstrom Company (France). It is made of $\text{TiO}_2\text{PC500}$ (anatase > 99%) coated on a cellulose support (non woven fibers) with silica as binder and zeolite as specific surface area extender. The content of TiO_2 is about 18 g m^{-2} of the paper. The paper weight is about 70 g m^{-2} corresponding to 25% TiO_2 per unit weight of the whole paper. Other characteristics of these catalysts are shown in Table 1.

Table 1
Specific surface and particle size of catalysts [3,22–24].

Catalyst	TiO_2 P25	TiO_2 PC500
Surface area (m^2/g) (BET)	54	300–375
Particle size (nm) (TEM and SEM)	15–25	5–10
Composition	Anatase (80%) Rutile (20%)	Anatase (>99%)
Isoelectric point	7	6.8
Point of zero charge	~6.25	6.20

The solutions were prepared with ultrapure water ($\text{TOC} \leq 0.2 \text{ mg L}^{-1}$, $18 \text{ M}\Omega \text{ cm}$). Sodium hydroxide and sulphuric acid (98%) were used to adjust the pH.

2.2. Experimental procedure

2.2.1. Preparation of thin films

Thin layers of TiO_2 were prepared by electrophoretic deposition (EPD) (Fig. 1) from a sol–gel suspension. An initial concentrated suspension of TiO_2 in water is obtained (500 g L^{-1} of TiO_2 P25, 16.35%(v/v) of acetylacetone, 0.83%(v/v) of Triton X-100). This suspension is then diluted with methanol to obtain 12.5 g/L of TiO_2 and used for deposition. The measured pH is about 4. Conducting glass ($\text{SnO}_2:\text{F}$ by CVD, ASAHI 100, from Solems, France) with a resistance of 70–100 Ω and stainless steel (304 L), as photocatalyst support, were cleaned with a mixture of hydrogen peroxide and sulphuric acid and washed several times with pure water. For the deposition, they were used as cathode in the electrophoresis cell. Anode and cathode plates, separated by a length of 1 cm, have the same immersion area. A source of DC voltage (10 V) ensures an electrical field causing the movement of TiO_2 particles toward the cathodic plate. This voltage is applied for 40 s. The obtained deposits are then dried in air and calcined in an oven at 450°C for 60 min.

2.3. Photocatalytic reactor and degradation protocol

All experiments were performed in a laboratory flow loop reactor (Fig. 2) provided with a reflector at the top. A UV-A lamp (15 W) type Black Light Blue with maximum irradiation at 365 nm was used for photocatalyst illumination. The light intensity transmitted to the catalyst in the absence of the solution is measured at different points of the irradiation reactor (length $44 \text{ cm} \times$ width $8 \text{ cm} \times$ height 6 cm) using a photo-radiometer (HD 2102.2, DELTA OHM, with the probe LP 471 UVA). The received irradiance at the bottom of reactor is rated on average at $3.5 \pm 0.5 \text{ W m}^{-2}$. The incident photonic flux was estimated at $0.66 \times 10^{-6} \text{ Einstein s}^{-1}$ by ferrioxalate actinometry.

Before each experiment, wet Ahlstrom paper ($241 \pm 2 \text{ cm}^2$) is thoroughly rinsed with water and dried in an oven at 50°C . It is then attached to a glass plate and placed in the irradiation reactor. In the case of TiO_2 P25 deposited on conducting glass or stainless steel, two plates were used provided with the surface area of photocatalyst (irradiated surface of $250 \pm 3 \text{ cm}^2$). 900 mL of the target pollutant solution is introduced in the tank. Before turning on the lamp, adsorption study was done for 1 h ($t = -60 \text{ min}$). A recirculation flow of about 200 L h^{-1} was done with a gear pump (Ismatec).

For the study of aging, the catalysts deposited on steel (SS) and conducting glass (G) were carefully rinsed with ultrapure water between successive experiments and dried in an oven at 105°C for

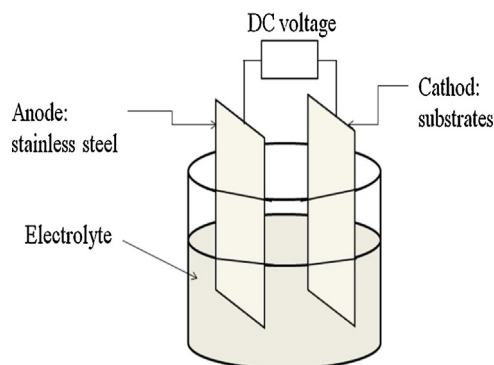


Fig. 1. Electrophoretic deposition setup.

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