



Degradation of terephthalic acid in a photocatalytic system able to work also at high pressure

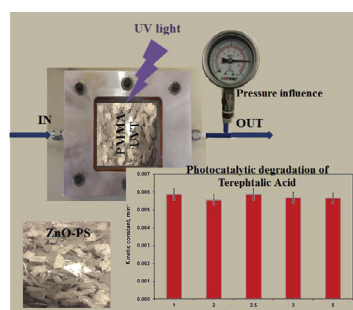
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HIGHLIGHTS

- Photocatalytic device able to work also at pressures higher than 1 atm.
- ZnO immobilized on polystyrene beads (ZnO-PS) used in a stainless steel reactor.
- ZnO-PS was active in the photocatalytic removal of terephthalic acid.
- ZnO-PS was stable after several reuse cycles.
- No worsening of photocatalytic activity by increasing the pressure up to 5 atm.

GRAPHICAL ABSTRACT



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ABSTRACT

Photocatalytic degradation of terephthalic acid (TPA) has been studied on commercial ZnO immobilized on polystyrene (PS) beads (mean size: $1.5 \times 1.5 \times 0.1$ cm) and located in a stainless steel flat plate reactor with an irradiation window realized in a UV transparent polymethylmethacrylate (PMMA UVT) in order to make the system able to work also at high pressure. In this way, it is possible to overcome, both the problems related to the photocatalysts separation after the wastewater treatment and those ones caused by the inlet pressure (which may be higher than atmospheric one) of incoming wastewater. The solvent casting-method used for photocatalyst preparation is able to induce the formation of ZnO particles in form of isles dispersed on PS macro surface. Moreover XRD analysis evidenced that the PS support didn't influence the crystalline arrangement of ZnO in wurtzite phase.

The structured composite photocatalyst ZnO-PS showed high ability in the degradation of TPA giving its total mineralization under UV irradiation at atmospheric pressure. Increasing the pressure, similar levels of TPA removal were observed. This finding demonstrated the possibility, for the developed photocatalytic system, to operate under moderate pressure without worsening the photocatalytic performances.

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1. Introduction

Terephthalic acid (TPA) is used in the production of polyester fibers, PET-bottle, PET-film, dyes, medicines, synthetic perfumes, pesticides and other chemical compounds. The produced wastewater account for 3–4 m³ each ton of TPA produced, being TPA one of

the main organic component [1]. Moreover, by recycling process of PET after the municipal collection, wastewater obtained from the washing of the collected PET could contain hydrolysis products of the PET resin as TPA [2].

TPA has been classified toxic and endocrine disruptor [3]. Wastewater containing TPA can be fed to conventional aerobic biological units. However the removal of TPA is slow and inefficient, and the biodegradation efficiency is negatively influenced by TPA presence [4].

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In recent years innovative technologies named advanced oxidation processes (AOPs), such as heterogeneous photocatalysis, have become the focus of several researches for treatment of pollutants in both aqueous and gaseous streams [5,6].

Despite TiO_2 is the most and widely studied photocatalyst for the degradation of organic compounds in aqueous and gaseous media [7,8], ZnO could be a significant alternative photocatalyst [9] because of its high photosensitivity, low toxicity, chemical stability, and optical properties [10].

ZnO has wide band gap energy (about 3.37 eV) and therefore it can be activated only by UV light [11]. The reaction mechanisms of photocatalytic degradation on ZnO are similar to that on TiO_2 [12,13]. However in some cases, ZnO was more effective than TiO_2 [14,15].

Photocatalysts studied for TPA degradation are usually used as suspended particles in the liquid reaction medium [1,16]. Because of the corrosion phenomena given by the slurry suspension, the recirculation pumps have to be substituted or repaired after a certain limited utilization time. Furthermore, in any case, after wastewater treatment, the separation of photocatalyst from treated water is achieved with equipments that require high energy demand. Consequently, the key problem of use of the photocatalytic technology in a large scale seems to be the expensive process of separation and recycling of the photocatalyst. A different type of photocatalyst may avoid the further step of separation, so the immobilization of ZnO on suitable supports is advisable. Several studies report the immobilization of ZnO nanoparticles onto different supports through different methods [9] developed to obtain ZnO based devices with optical, electrical, photocatalytic, and bactericidal properties [17,18].

However, in addition to the energy drawbacks related to the separation processes of the photocatalyst after the treatment, an efficient photocatalytic device must be composed by a photoreactor specifically designed to withstand the operating pressure anticipated for fluid circulation through a large field. In some cases, the wastewater inlet pressure to the reactor should reach values up to 5 atm [19].

These levels of pressure are prohibited for traditional photoreactors built in quartz or Pyrex glass, due to the rupture of the photoreactor transparent walls or windows. Moreover also the support on which the photocatalyst was immobilized could not have the proper strength. The latter problem could be avoided by using polymeric materials. Among different polymeric materials, polystyrene (PS) is a cheap, inert, non-toxic and low density thermoplastic polymer widely used in food service and retail industry [20]. Nowadays, the literature about the use of ZnO supported on PS in the photocatalytic treatment of wastewater is still very scarce. As an example, Altin and Sokmen immobilized ZnO particles onto waste PS by thermal method and tested the obtained photocatalysts in the degradation of methylene blue [9]. However, the size of PS pieces was in the range between 500 and 800 μm and therefore they cannot be utilized in a large-scale photoreactor because in the case, they will generate very high pressure drops due to the high wastewater inlet pressure. In addition, also the papers reporting the use of a photocatalytic reactor able to work at pressures higher than 1 atm are limited in number.

Considering the previous observations, in this work it is presented a photocatalytic device (that may be considered as a proof of concept) able to overcome both the problems related to the photocatalysts separation and the high inlet pressure of incoming wastewater.

For this purpose commercial photocatalysts (ZnO or TiO_2) in powder form was immobilized on PS pieces and located in a stainless steel photocatalytic reactor specifically designed with flat plate geometry and equipped with an irradiation window made

by a UV transparent polymethylmethacrylate (PMMA) in order to make it able to work also at high pressure.

2. Materials and methods

2.1. Materials

The experiments were performed with ZnO (Sigma-Aldrich) and TiO_2 (Degussa P25 – Evonik Corporation, Germany) commercial powders. The polymeric support used for the deposition of ZnO particles was commercial polystyrene pieces (PS) (almost regular flat PS pieces with a mean size of: $1.5 \times 1.5 \times 0.1$ cm) obtained by cutting Petri dishes [21].

As model pollutant in photocatalytic tests, terephthalic acid (TPA), supplied by Sigma Aldrich, was used.

2.2. Preparation of ZnO-PS and TiO_2 -PS

Commercial PS was used as polymeric support for the immobilization of commercial ZnO powder.

For the immobilization of the photocatalyst particles, a solvent-casting method has been used. In detail, 0.5 g of ZnO photocatalyst were dispersed in 50 ml of solvent (acetone) and vigorously mixed to obtain a uniform dispersion of the particles. The suspension was then dropped on the surface of PS pieces.

ZnO particles adhere to the soft surface of the PS when solvent reached the surface. The final composite ZnO-PS was obtained after total evaporation of acetone at room temperature. In order to remove the excess of ZnO particles not immobilized on PS surface after the treatment with acetone, several cycles in an ultrasonic bath (CEIA-CP104) were effected until to reach a ZnO stable loading (Fig. 1).

The ZnO amount coated on PS has been measured using precision balance (Mettler Toledo). The final loading of ZnO in ZnO-PS sample was 3.3 wt%.

For comparison, commercial TiO_2 was immobilized on PS (TiO_2 -PS) with the same method to have the same loading of ZnO on PS (3.3 wt%).

2.3. Catalyst characterization

The ZnO, PS and ZnO-PS samples were characterized by several techniques. UV-vis reflectance spectra of the samples were recorded by a Perkin-Elmer spectrophotometer Lambda 35 using a RSA-PE-20 reflectance spectroscopy accessory (Labsphere Inc.,

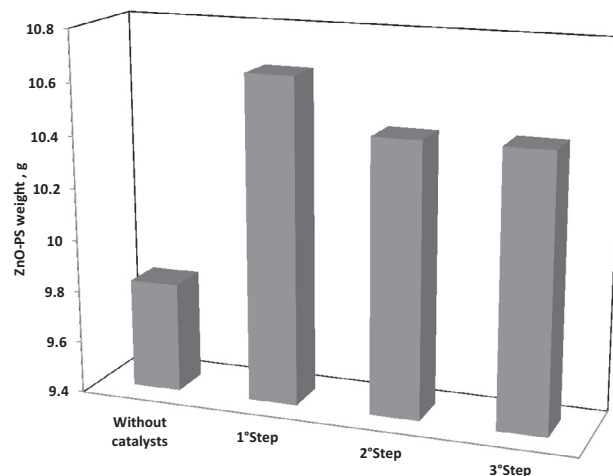


Fig. 1. Results of ultrasonic cycles on ZnO-PS photocatalyst.

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