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Study of synergetic effect, catalytic poisoning and regeneration using dielectric barrier discharge and photocatalysis in a continuous reactor: Abatement of pollutants in air mixture system

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ABSTRACT

In the present work the abatement of butyraldehyde (BUTY), dimethyl disulfide (DMDS) and their mixtures in gas phase was studied in continuous reactor at three different configurations: photocatalysis (TiO_2 + UV), dielectric barrier discharge (DBD) plasma and their association in the same system (DBD+ TiO_2 + UV). The effect of some operating parameters such as inlet concentration of pollutant and flowrate on planar reactor performance in term of (i) BUTY removal (ii) selectivity of CO and CO_2, selectivity of byproducts has been also investigated. Moreover, ozone formation has been studied to evaluate the performance of the combined process. A synergetic effect was observed by combining (DBD) plasma and photocatalysis on BUTY removal but has not been present when it was in air mixture with dimethyl disulfide (DMDS) due to the poisoning of the catalyst. Additionally, degradation was observed as a consequence of by-products accumulation on the surface of the catalyst. Moreover, the regeneration/recovery of the initial photocatalytic activity was explored in details. A significant regeneration has been occurred by combining photocatalysis and nonthermal plasma. This trend of nonthermal plasma on catalytic surface can explain the synergetic effect during the pollutant degradation time. Moreover, the catalyst was concomitant with the time required for the hydrophobic to hydrophilic transition on the catalyst surface as followed by contact angle measurement (CA). Redox catalysis was detected by X-ray Photoelectron Spectroscopy (XPS) showing Ti^{4+}/Ti^{3+} switching during the degradation, poisoning and regeneration times.

1. Introduction

The emission of volatile organic compounds (VOCs) is a major source of air pollutants, coming largely from industrial sectors especially by the petrochemical, and therefore, poses serious damages to environment and public health [1–3]. Sulfur containing volatile organic compounds (SVOCs) constitutes one of the important families of atmospheric pollutants such as methyl mercaptan (CH_3SH),dimethyl sulfide (CH_3SCH_3) and dimethyl disulfide (CH_3SSCH_3) [4,5]. These mentioned compounds are coming either from natural processes like anaerobic biological activities or from anthropogenic emissions sectors such as pulping processes and chemical industries. First and foremost, these sulfur compounds are known by their high toxic and corrosive effect [4,6,7]. Besides, they are causing significant environmental problems. In fact, their oxidation in the atmosphere will lead to the production of sulfur dioxide (SO_2), which be consequently converted into sulfuric acid and returns to the earth when it rains [4]. Therefore, the abatement of those odorous emissions in the atmosphere is an important issue that needs an efficient and least-cost process. Adsorption [8], thermal oxidation [9], biological treatment [10], ozonation [11], and catalysis/photocatalysis [12] have been proposed by researchers as conventional VOCs treatment methods.

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Photocatalysis is a promising technology for air purification because of its ability to break down a wide range of pollutants to non-toxic end-products such as CO_2 and H_2O under ambient conditions [13,14]. The latter processes based on photoreactions which take place at the surface of a semiconductor usually titanium dioxide (TiO_2) under ultraviolet (UV) irradiation [15–18]. Some significant results have been achieved in the photocatalytic oxidation of many organic compounds like VOCs and SVOCs which includes dimethyl sulfide [19], ethyl mercaptan [20], hydrogen sulfide [21], BTEX (Benzene, Toluene, Ethylbenzene and Xylenes) removal [22] and isovaleraldehyde [23]. Moreover, several researchers have investigated non-thermal plasma (NTP) as one of the technologies which can be used to treat polluted air [24–26]. This techniques characterized by its high efficiency in the formation of reactive chemical species like hydroxyl radicals, atomic oxygen and ozone, which may react and degrade organic molecules at room temperature under atmospheric pressure [26–28]. In fact, dielectric barrier discharge (DBD), one of the NTP techniques, has been employed successfully for the abatement of various aromatic VOCs: toluene [29] and benzene [30], trimethylamine [31], hydrogen sulfide [32], and sulfur dioxide [33]. Several studies have reported on the combined process between non-thermal plasma (NTP) and photocatalysis leading to improve the efficiency of VOCs degradation [34–39] over a synergistic effect [29–32].

The aim of the present work is to explore the removal efficiency of butyraldehyde (from now as BUTY), dimethyl disulfide (from now as DMDS) and their mixtures in planar reactor, using dielectric barrier discharge (DBD) plasma (from now as DBD-plasma) and photocatalysis processes separately and their combinations in the same reactor. The influence of inlet concentration and flowrate on the pollutants removal, nature of the by-products and ozone formation inside the process are studied in details. Moreover, an interesting challenge of this work is to study the presence of synergetic effect between DBD-plasma and photocatalysis during the oxidation of mixture of VOCs at pilot scale. A special attention is paid to TiO_2 poisoning and its regeneration with different process oxidation (plasma, photocatalysis and combined system). The evidence for redox catalysis responsible for the photocatalytic activity was also investigated.

2. Materials and methods

2.1. Reactor design and setup details

The reactor consists of a rectangular glass chamber (length L = 1000 mm, width l = 135 mm and height H = 135 mm). Two glass plates are installed parallel to the length of the reactor and permit to maintain the catalyst, the two electrodes (1 mm thick and 800 mm in length) and high voltage grids (stainless steel, 2 mm thick). To generate the plasma, high voltage is applied to the reactor. The applied voltage is delivered by a generator (BFi OPTILAS) as a sinusoidal waveform up to 10 V and then amplified by an amplifier (TREK 30/40) to achieve 30 kV. This amplifier is connected to the electrodes. A coil of capacitors with a total capacity of 2.5 nF (C_m) was positioned between the copper electrode and the ground connection in order to collect the charges in the reactor. The distance between the two plates could be adjusted to modify the space/air gap. The voltage applied (U_{app}) and the high voltage capacity (U_m) are measured by high-voltage probes recorded with a digital oscilloscope (Lecroy wave surfer 24Xs, 200 MHz). The reactor can be used also as a photocatalytic reactor and as a plasma DBD–photocatalytic reactor. Eight lamps (Philips under reference PL-S 9W/10/4P) are placed equidistant from each other in air gap of the reactor in order to ensure a good radiation distribution (Fig. 1). The photocatalyst is maintained between the stainless steel grid electrode and the dielectric barrier in the plasma active area. The UV lamps arranged in the reactor, permit the activation of the photocatalyst. Two openings with septum downstream and upstream of the reactor allow outlet and inlet gas to be sampled with a 500 μL syringe.

2.2. Catalysts

The supported material is further named Glass Fiber Tissue (GFT) containing 13 g/m^2 of colloidal silica, a 13 g/m^{2}of titanium dioxide nanoparticles and inorganic fibers. The coating process consists in an impregnation of glass fibers by SiO_2 and TiO_2 nanoparticles suspension in pure water using an industrial size-press (PC500 Millennium). The specific area of TiO_2 nanoparticles is 300 m^2/g. The

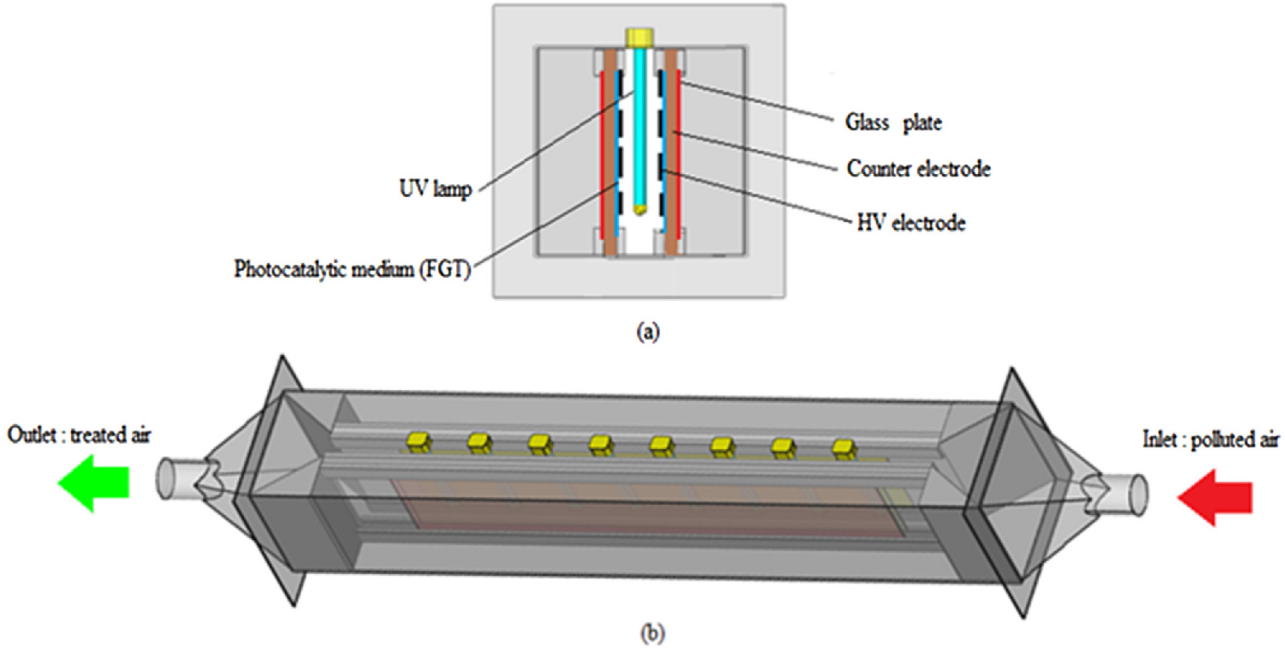

Fig. 1. Schematic illustration of the used set-up: (a) sectional drawing and (b) planar reactor.