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High photocatalytic efficiency of spouting reactor compared with fluidized bed with top irradiation source

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ABSTRACT

The removal of volatile organic compounds by photocatalytic degradation is one of the safest and most effective ways of removing pollutants from the air. This process is highly affected by the type of reactor, light exposure, and hydrodynamics. For scale up purposes, continuous reactors with high capacity are required for treating large amounts of feedstock. In this work, two types of reactors based on different hydrodynamics, fluidized and spouted reactors, were designed to work under light irradiation inside the reactor. The efficiency of the reactors for volatile organic compound removal from high flow rates of air under Hg lamp irradiation using N–F-TiO_2 photocatalyst was investigated. The performance of the fluidized bed and spouted bed were evaluated and compared at the same weight hourly space velocity of feed stream through the reactor. The results revealed that 80% of the initial acetaldehyde was removed in the fluidized bed after about 200 min, while in the spouted bed the acetaldehyde was totally removed after about 120 min.

Introduction

The emission of volatile organic compounds (VOCs) is one of the major sources of indoor and outdoor air pollution (Oppenländer, 2003; Seinfeld & Pandis, 1998). The photocatalytic degradation of VOCs is an oxidation process that uses a catalyst (mostly TiO_2), light irradiation, and oxygen to convert these organic pollutants to carbon dioxide and water.

During the photocatalytic oxidation (PCO) of VOCs, the catalyst generates electron-hole pairs on its surface upon adsorption of light. The holes can create very reactive hydroxyl radical ($OH^\bullet$):

$$h^+ + OH^- \rightarrow OH^\bullet.$$

This radical assists with the decomposition of VOCs on the catalyst surface. The oxidant (O_2) prevents the recombination of the electron-hole pairs by reacting with the electrons:

$$e^- + O_2 \rightarrow O_2^-.$$

More details of the mechanism of the photocatalytic decomposition of VOCs can be found elsewhere (Agustina, Ang, & Vareek, 2005; Mo, Zhang, Xu, Lamson, & Zhao, 2009). In summary, the PCO reaction can be described by the following:

$$VOC + O_2 \xrightarrow{\text{Catalyst+Light}} CO_2 + H_2O.$$

Beside the catalyst type, the efficiency and rate of PCO reaction can be significantly affected by the type of reactor and operating conditions. Various types of photocatalytic reactors with different contact regimes between gas, solid, and light source have been reported in the literature. These include plate, honeycomb, annular, packed bed, and fluidized bed reactors. However, most of the synthesized nano-photocatalysts have been evaluated using batch reactors (Habibi, Fatemi, Izadyar, & Mousavand, 2012; Izadyar, Fatemi, & Mousavand, 2013; Jokl, 2000; Obee & Hay, 1997; Raillard et al., 2005; Shiraishi, Yamaguchi, & Ohbuchi, 2003; Yamazaki, Matsunaga, & Hori, 2001). Because the concentration of VOC contaminants in industry is usually high (several hundreds of ppm) and they are continuously generated and emitted in flue gas, a process for the continuous photocatalytic oxidation of VOCs with high capacity is necessary. The continuous PCO reactors can be classified into two major classes; packed bed and fluidized bed reactors (Dashliborun, Sotudeh-Gharebagh, Hajaghazadeh, Kakooei, & Afshar, 2013). Effective photocatalytic reactors must provide appropriate irradiation to the surface of catalyst and optimized contact area between the reactant and photocatalyst. The packed bed reactor is insufficient owing to the low accessibility of the pho-

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tocatalyst to the light source. Fluidized bed reactors can provide better contact between gas, solid, and light source than packed bed reactors. Dibble and Raupp (1992) designed a bench scale flat plate fluidized bed photoreactor for continuous trichloroethylene (TCE) oxidation. The conversion of trace TCE (<10 ppm) in a humid gas stream with a flow rate of 360 cm^3/min was as high as 99% for 46 h. Dashliborun et al. (2013) designed a fluidized bed photo reactor using nano-TiO_2 supported on gamma-Al_2O_3 for the oxidation of gaseous methyl ethyl ketone (MEK). They studied the effects of the relative humidity, initial concentration of MEK, and superficial gas velocity on the MEK degradation. Tasbihi et al. (2011) studied the oxidation of gaseous toluene over titania/mesoporous silica powder in a fluidized-bed reactor. The conversion of 15 ppm toluene in a 2 dm^3/min gas stream was as high as 90% for 7 h. Kuo, Wu, and Hsu (2009) studied the photodegradation of gaseous toluene over AC/TiO_2 in a fluidized bed reactor. They studied the effects of the relative humidity, gas flow rate, and initial concentration of toluene on the PCO process. Yao and Kuo (2015) studied the removal of gaseous toluene over SiO_2/TiO_2 in a fluidized bed photoreactor. For 20 g catalyst, a flow rate of 5 L/min, and 1000 ppm toluene, the conversion in their system was 30%–40%.

Spouted bed reactors were first developed at the National Research Council (NRC) of Canada by Ernest and Mathur (1957) in 1954 as an alternative method of drying to a badly sluggish fluidized bed of moist wheat particles. The first commercial spouted bed units in Canada were installed in 1962 for the drying of peas, lentils, and flax, that is, drying of granular particles undergoing spouting. Since then, units have been built in other countries for a variety of other drying duties, including the evaporative drying of solutions, suspensions, and pastes in a spouted bed of inert particles, as well as for the blending, cooling, coating, and granulation of solids. Most commercially successful spouted bed installations have involved such physical processes, but a wide variety of chemical processes have also been subjected to laboratory- and bench-scale spouting investigations. Some of these, including electrolysis in a liquid-spouted bed (Siu & Evans, 1999), show considerable promise for further development. Conical spouted beds have been used for drying suspensions, solutions, and paste-like materials (Markowski, 1992; Markowski & Kaminski, 1983; Passos, Massarani, Freire, & Mujumdar, 1997; Passos, Oliveira, Franca, & Massarani, 1998; Pham, 1983). Chemical reaction applications of such beds include catalytic polymerization (Bilbao, Olazar, Romero, & Arandes, 1987), coal gasification (Uemaki & Tsuji, 1986), waste pyrolysis (Aguado, Olazar, San José, Aguirre, & Bilbao, 2000; Arabiourrutia et al., 2007), and selectivity-conditioned catalytic reactions (Marnasidou, Voutetakis, Tjatjopoulos, & Vasalos, 1999). Spouted bed reactors can provide appropriate contact between gas and particles as well as proper exposure to the light source, and provide a hydrodynamic regime with good particle mixing and circulation with the least amount of mass and heat transfer resistances (Bahramian & Kalbasi, 2010).

In the current study, we have applied a spouted bed to a photocatalytic process for the first time. Fluidized bed and spouted bed photoreactors were designed and compared to evaluate the efficiency of these two types of reactors for the decomposition of acetaldehyde from an air flow with high flow rate using N–F co-doped TiO_2 under an Hg lamp.

Experimental

Synthesis of photocatalyst and characterization

In this study, we prepared N–F co-doped TiO_2 by a sol gel method. NH_4F and NH_4OH (25%) were dissolved in a solution of (1:5 V/V) distilled water and ethanol under vigorous stirring. Next, tetraisopropyl orthotitanate was added dropwise to the solution. The final solution was stirred for 24 h at 40 °C and aged for 16 h at room temperature. The solution was then dried in an oven and calcined in a furnace at 500 °C for 2 h. The obtained catalyst was characterized by X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FT-IR; Model TENSOR 27, Bruker, Germany), scanning electron microscopy (SEM; SIGMA VP, Zeiss, Germany), and diffuse reflectance spectroscopy (DRS). XRD patterns were recorded in the range of $2\theta = 10^\circ$–80° with a Philips Xpert diffractometer (Philips, Netherlands) using CuKα ($\lambda = 0.15406$ nm) radiation. DRS was used to measure the absorbance of the samples using a UV–Vis spectrophotometer (model Ava-spec-2048-TEC), and $BaSO_4$ was used as a reference.

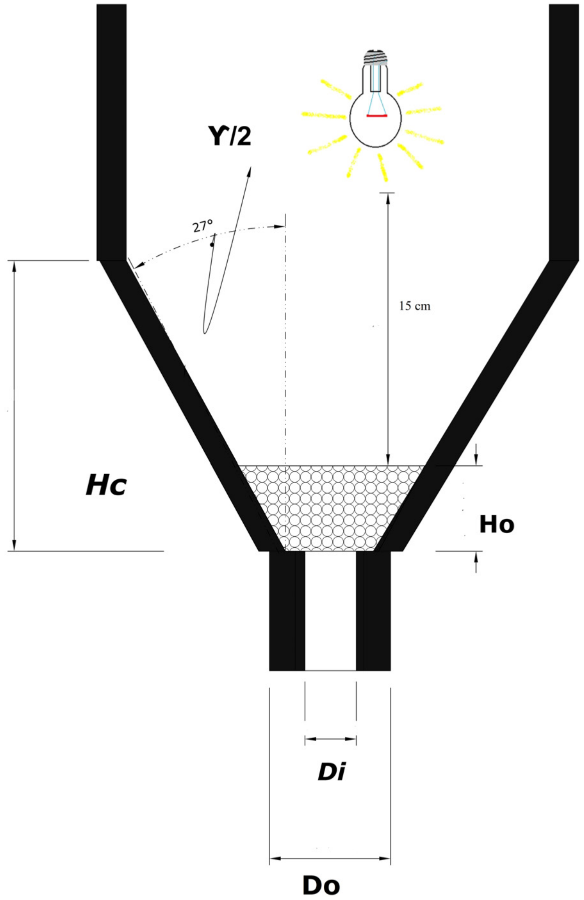

Fig. 1. Schematic diagram of spouted bed photoreactor (SPBR).

Spouted bed photoreactor

The schematic of the spouted bed photoreactor (SBPR) is shown in Fig. 1 and the specifications of the SBPR are listed in Table 1. The SBPR was made of glass with an upper cylindrical section. An 80 W Hg lamp was installed in the center of the reactor 15 cm above the catalyst. A sintered glass frit was used as the air distributor to spout

Table 1
Specification of SPBR.

Gas inlet diameter, D_i (cm)	0.7
Cone diameter of spouted reactor, D_O (cm)	1
Static bed diameter, D_b (cm)	3
Height of conical section, H_C (cm)	14
Height of static bed, H_O (cm)	2.5
Angle of conical section, γ (°)	45
Total height of reactor, H_T (cm)	37 + 14
Cylindrical diameter, D_C (cm)	4.2