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Abatement of volatile organic compounds using an annular photocatalytic reactor: Study of gaseous acetone

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Abstract

Photocatalytic oxidation of organic compounds in gas phase appears to be a promising process for remediation of polluted air. In the present work, the photocatalytic degradation of acetone, which is a typical pollutant of indoor air, was investigated by using an annular photoreactor. After a modelling by a cascade of elementary continuously stirred tank reactor, the annular photoreactor was assimilated to a plug flow reactor (PFR). No transfer limitation (external and internal) has been demonstrated for this reactor with the fibreglass photocatalytic support. The influence of several kinetic parameters has been studied such as pollutant concentration, incident light irradiance, contact time and humidity content. The Langmuir–Hinshelwood model has been verified for acetone. It can be noticed that no by-products have been detected by FID suggesting almost total mineralization. The possible minor gaseous by-products have been accumulated into a mixture of ethanol–liquid nitrogen at -50 °C then a sample of it has been injected into a GC/MS for analysis. A mechanistic pathway is then proposed for the photocatalytic degradation of acetone. © 2008 Elsevier B.V. All rights reserved.

Keywords: Annular reactor; Radiation field; Kinetic modelling; By-products; Photocatalytic degradation mechanism

1. Introduction

Photocatalytic oxidation of airborne contaminants appears to be a promising process for remediation of air polluted by volatile organic compounds (VOCs) or by bacteria [1]. VOCs are well known to be malodorous, toxic and some of them can be considered as carcinogenic, mutagenic and teratogenic. There are a number of available photocatalytic reactor configurations for the abatement of airborne pollutants: (1) flat plate fluidized bed reactor, (2) fluidized bed reactor, (3) fixed layer photocatalytic reactor, (4) photocatalytic reactor with fibre optic bundles, and (5) annular photocatalytic reactor [2]. This annular geometry is widely used for controlling VOC emissions in indoor air. A large diversity of reactor configurations has been considered to study the efficiency of photocatalytic degradation of air pollutants. Photocatalytic reactors require several of important reactor parameters such as [2,3]:

- UV source;
- reactor configuration;
- catalyst type, distribution and impregnation;
- interaction between the light, catalyst and reacting flowing.

This work focuses on the photocatalytic degradation of acetone. Natural pollution sources: acetone is mainly produced via oxidation of humic substances and it occurs naturally as a metabolic by-product of plants and animals. Artificial pollution sources: acetone is widely used as solvent for fats, oils, waxes, resins, rubbers, plastics and pharmaceuticals. Indoor air, the presence of acetone is due to the use of domestic materials and products as paints and cleaning products. Acetone was detected at concentrations between $14.9 \text{ ng } \text{L}^{-1}$ (35.8 ppb) and 66.0 ng L^{-1} (158.4 ppb) at dwelling houses. This ketone has a threshold limit value (TLV) in air of 1.2 mg L^{-1} (500 ppm) [4]. The threshold limit value is the maximum permissible concentration of a pollutant generally defined in workplace atmospheres. Our photocatalytic reactor could be used to reduce VOC emissions in workplace atmospheres or in dwelling house indoor air.

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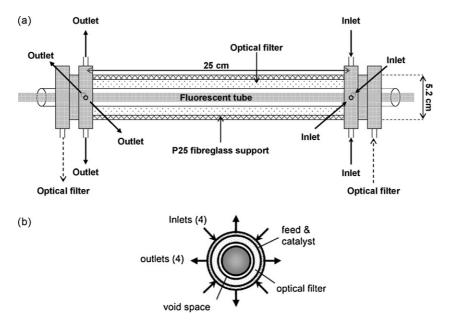


Fig. 1. Schematic representation (a) and sectional drawing (b) of the annular photoreactor.

The first part of this work deals with the results of the acetone kinetic study. Our annular photocatalytic reactor was modelled by a cascade of continuously stirred tank reactors (CSTRs) in order to classify the category of reactor. The second part of this work consists in summarizing the study of by-products, which were formed during the photocatalytic degradation of acetone. A mechanistic pathway was proposed for its photocatalytic degradation.

2. Experimental

2.1. Experimental set-up and procedure

The annular photocatalytic reactor was equipped with four inlets and four outlets in order to ensure a good flow distribution (Fig. 1). A fibre lass support (effective porosity $\varepsilon = 0.95$) impregnated of TiO₂ Degussa P25 was inserted between two Pyrex glass tubes with a thickness of 1.8 mm. This low thickness provides a best contact between polluted air and photocatalyst. A commercial Philips[®] TLD 18 W/08 fluorescent tube is placed in the centre of the unit offering the best conditions of light irradiance. It can be noticed that the UV lamp has a spectral peak centred at about 365 nm. In a previous work, the 3-W light power has been verified by actinometry [5]. The fluorescent tube and the photocatalyst were separated by a liquid filter in order to control both temperature and light irradiance during the degradation process. The light transmission was attenuated by a specific nigrosine concentration in aqueous solution. The total diameter, the volume and the photoactive length of the annular reactor were, respectively 5.2 cm, 66.4 cm³ and 25 cm. The diameter of the space for the fluorescent tube was 30.5 mm. The thickness available for the photocatalyst support was about 1.8 mm. The fibreglass support apparent area exposed to UV was 300 cm². The experimental unit permits to generate a polluted air with a specific VOC concentration and humidity content. The functioning of the experimental set-up has been widely detailed in previous works [5,6]. Several kinetic parameters can be tested on the photocatalytic degradation efficiency as initial concentration of pollutant, light irradiance, contact time and humidity content.

A gas chromatograph equipped with a flame ionization detector (FID) was used to follow acetone concentration during photocatalytic oxidation. For this pollutant, the response of the FID was linear and proportional to the amount of acetone. Finally the acetone conversion X in the reactor is expressed by the following equation:

$$X = 1 - \frac{C_{\text{out}}}{C_{\text{in}}} = 1 - \frac{A_{\text{out}}}{A_{\text{in}}} \tag{1}$$

The GC is a Hewlett Packard 5890 Series II apparatus equipped with a FID. The GC operational parameters were as follows: analytical column, Porapak Q column 1/8 in. (1 m) at 180 °C; carrier gas, nitrogen at 21 mL min⁻¹; injected volume, 1 cm³; FID detector at 250 °C supplied with air/hydrogen at 276 and 38 mL min^{-1} , respectively.

The by-products generated during the photocatalytic degradation of acetone were identified by GC/MS. The GC/MS is an Agilent 6850 Series apparatus equipped with a mass selective detector (MSD) Agilent 5973 Network. The GC/MS operational parameters were as follows: analytical column, HP Plot Q ($30 \text{ m} \times 0.32 \text{ mm}$ i.d.); carrier gas, helium at $1.5 \text{ mL} \text{ min}^{-1}$; program temperature, $40 \degree \text{C}$ for 10 min, $5 \degree \text{C} \text{min}^{-1}$ and $240 \degree \text{C} 10 \text{ min}$; temperature of injector, $250 \degree \text{C}$ (split = $4.5 \text{ mL} \text{ min}^{-1}$); injected volume, $1 \ \mu\text{L}$; detector, MSD at $250 \degree \text{C}$.

2.2. Catalyst preparation

The catalyst consisted of TiO₂ P25 Degussa deposited on a Sintomat[®] fibreglass support ($250 \text{ mm} \times 120 \text{ mm}$). A single

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