



# Computational fluid dynamics modeling and parameterization of the visible light photocatalytic oxidation process of toluene for indoor building material



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## ABSTRACT

Poor indoor environmental conditions caused by the presence of gas-phase/aerosol-phase contaminants are thought to be potential triggers of illnesses, leading to productivity losses. In recent decades, the photocatalytic oxidation (PCO) process has attracted attention because of its potential to purify indoor air polluted with volatile organic compounds (VOCs), especially at low concentration levels. Titanium-dioxide (TiO<sub>2</sub>)-bound building materials have been extensively studied for the oxidation of indoor VOCs. In this study, kinetic studies were carried out to evaluate the PCO of toluene in the gas phase, over TiO<sub>2</sub>-bound building materials, using a 20 l small test chamber. A Langmuir–Hinshelwood (L–H)-type model, for reproducing the PCO process over TiO<sub>2</sub>-bound building materials, was developed as a function of the toluene concentration, illumination intensity, and humidity levels. The parameters of the L–H model were determined using the hybrid identification procedure of computational fluid dynamics (CFD) simulations and a 20 l small test chamber experiment. By using this hybrid identification method, PCO kinetic parameters were estimated with reference to the concentration in viscous-sub layer. Finally, CFD simulations were also carried out under the same boundary conditions as those used in the chamber experiments to evaluate the prediction accuracy.

## 1. Introduction

The importance of indoor air quality (IAQ) for residents/workers is well-recognized because IAQ directly or indirectly affects the health, comfort, and performance of occupants/workers (Wargoeki, Wyon, Sundell, Clausen, & Fanger, 2000). Generally, control procedures for IAQ levels are roughly classified into two parts: ventilation control and source control. Ventilation control is primarily intended to directly control the contaminant concentration level by allowing clean/fresh outdoor air into the indoor environment for dilution. Hence, the increase in ventilation rate can contribute to lower contaminant concentration levels. On the other hand, the increase in ventilation rate corresponds to an increase in the HVAC (heating, ventilation, and air-conditioning) loads (Fan & Ito, 2012; Fan, Kameishi, Onishi, & Ito, 2014). From the viewpoint of source control, for example, emissions of volatile organic compounds (VOCs) from building materials in indoor environments are considered to lower IAQ, as the minimization of emission sources directly contributes to lower contaminant concentration in the indoor environment (Wolkoff, 1995).

In recent decades, a passive IAQ control method with low energy requirements has been actively discussed to design a healthy indoor environment (Day & Gunderson, 2015). The introduction of building materials with the capacity to adsorb indoor gas-phase contaminants might be a good example of a passive IAQ control method (Ataka et al., 2004; Murakami et al., 2001; Murakami, Kato, Ito, & Zhu, 2003). Among the technologies related to the passive control method, the photocatalytic oxidation (PCO) reaction has been recently recognized as a potential method for contributing towards resolving IAQ issues (Fujishima & Honda, 1972; Mo, Zhang, Xu, Lamson, & Zhao, 2009; Zhao & Yang, 2003). In particular, PCO processes initiated by TiO<sub>2</sub>-based catalysts have been confirmed to be highly effective in the purification of air polluted with VOCs (Chen & Poon, 2009; Kim & Hong, 2002; Sopyan, Watanabe, Murasawa, Hashimoto, & Fujishima, 1996). Newly developed TiO<sub>2</sub>-coated/bound building materials have been used indoors, and qualitative and quantitative reports of their performance with respect to reduction in indoor contaminant concentration through photocatalytic decomposition effect are available. While PCO based technologies and their kinetic studies have been reported, studies

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that intended for application to indoor air quality control and integration into computational fluid dynamics (CFD) based indoor environmental analysis are limited (Zhong & Haghighat, 2015; Zhong, Haghighat, & Lee, 2013; Zhong, Branco, Batterman, Bartlett, & Godwin, 2017). In typical kinetic studies of PCO reactions, tubular-type reactors are used, and the reaction rates are evaluated under conditions where the reaction rates are not affected by the diffusion and convection of gas phase molecules, and ideal laminar type plug flow in the reactor is assumed (Davis & Davis, 2003). In some cases, the reactors cannot be assumed as ideal plug flow; the convection and diffusion of the substrates and formation of concentration boundary layer in the photochemical reactor should be investigated to precisely determine the kinetic parameters of surface reactions.

Against this background, kinetic studies were carried out to determine the PCO of gas-phase toluene over photocatalyst-bound building materials, using a 20 l small test chamber in this study. The kinetic equation for toluene photo-oxidation over TiO<sub>2</sub>-coated building material could be expressed by Langmuir–Hinshelwood (L–H)-type kinetics (Einaga, Tokura, Teraoka, & Ito, 2015; Mills, Wang, & Ollis, 2006; Noguchi, Fujishima, Sawunyama, & Hashimoto, 1998; Ollis, 2005; Turchi & Ollis, 1990). The objective of this study is to develop a mathematical model, which can reproduce the PCO process on photocatalyst-bound building materials as a function of indoor environmental parameters, like contaminant concentration, illumination, and absolute humidity levels, in addition to being integrated into computational fluid dynamics (CFD) and illuminance distribution analyses as a wall surface boundary condition. In this study, we propose an improved L–H-type mathematical model for integrating the CFD simulation intended for indoor air quality analysis. Moreover, the hybrid identification procedure for kinetic parameters, present in the improved L–H-type mathematical model, is also proposed. In the proposed method, kinetic parameters for surface photocatalytic reaction are initially determined by using experimental results, being subsequently optimized via the corresponding precise CFD simulations considering the development of the concentration boundary layer and non-uniform distribution of gas-phase toluene on photocatalyst-bound building material surfaces. The illuminance distribution of photocatalytic building material surfaces was also calculated beforehand based on the radiosity method, and applied as wall surface boundary condition.

These kinetic parameters would be identified as functions of gas-phase toluene concentration, illumination intensity, and humidity levels.

Finally, a CFD simulation was carried out using the same boundary conditions as those used in the 20 l small test chamber experiment, and the prediction accuracy of CFD simulation with the improved L–H-type model was discussed in quantitative terms.

## 2. Kinetic modeling of photocatalytic degradation

### 2.1. Langmuir–Hinshelwood mechanism

Studies on photocatalytic reaction have been continuing since Fujishima and Honda (Fujishima & Honda, 1972) discovered the phenomenon of photoinduced water cleavage on TiO<sub>2</sub> electrodes. Since the 1990s, much of the fruitful research and development on the degradation and removal of harmful substances from the environment has focused on the fundamentals and application of the PCO reaction, with some of the techniques having been put to practical use in fields such as building materials and sanitary equipment (Chen & Poon, 2009; Davis & Davis, 2003; Einaga et al., 2015; Fujishima & Honda, 1972; Kim & Hong, 2002; Mills et al., 2006; Mo et al., 2009; Noguchi et al., 1998; Ollis, 2005; Sopyan et al., 1996; Turchi & Ollis, 1990; Zhao & Yang, 2003; Zhong & Haghighat, 2015; Zhong et al., 2013; Zhong et al., 2017). PCO processes require catalysts that exhibit high oxidation activities under photo-irradiated conditions. TiO<sub>2</sub>-based materials have been extensively applied to the oxidation of a variety of air

pollutants (Mo et al., 2009; Zhao & Yang, 2003). According to this photocatalytic reaction mechanism, the reaction begins with the irradiation of light over TiO<sub>2</sub>. Molecular water and oxygen adsorbed on the photocatalyst will react with the positive holes and electrons, consequently generating active oxygen in the form of OH· and superoxide (O<sub>2</sub><sup>−</sup>), respectively. The highly active oxygen will decompose organic compounds, eventually converting them into carbon dioxide (CO<sub>2</sub>) and water. This reaction generally occurs in proportion to the light intensity, proceeding under normal indoor temperature conditions. In this study, we considered toluene as the representative indoor air pollutant. The experiments reported by Obee and Brown (Obee & Brown, 1995) showed a constant PCO reaction rate, regardless of the amount of OH; under low toluene concentration (1 ppmv or below). Therefore, we focus here on the reaction between toluene and oxygen (O<sub>2</sub>).

In general, the PCO reaction mechanism is expressed as the Langmuir–Hinshelwood (L–H) mechanism, which is formulated as follows:

$$r_{PCO} = k_{PCO} \theta_{VOC} \theta_{O_2} \quad (1)$$

where  $r_{PCO}$  is the PCO reaction rate (kg m<sup>−2</sup> s<sup>−1</sup>),  $k_{PCO}$  is the reaction rate constant (kg m<sup>−2</sup> s<sup>−1</sup>), and  $\theta_{VOC}$  and  $\theta_{O_2}$  are the fractions of surface covered by toluene and O<sub>2</sub>, respectively. The simplest fraction is written as a Langmuir isotherm:

$$\theta_{VOC} = \frac{K_{VOC} C_{VOC}}{1 + K_{VOC} C_{VOC}} \quad (2)$$

$$\theta_{O_2} = \frac{K_{O_2} C_{O_2}}{1 + K_{O_2} C_{O_2}} \quad (3)$$

where  $K_{VOC}$  and  $K_{O_2}$  are the equilibrium constants of toluene and O<sub>2</sub>, respectively.

Obee and Brown (Obee & Brown, 1995) also reported toluene shared a competitive adsorption relationship with water. Assuming that the fraction of water is also expressed as same formula,  $\theta_{VOC}$  can be rewritten as follows:

$$\theta_{VOC} = \frac{K_{VOC} C_{VOC}}{1 + K_{VOC} C_{VOC} + K_W C_W} \quad (4)$$

Furthermore, assuming that there is sufficient O<sub>2</sub> (order of %), compared to toluene (order of ppb in the indoor environment),  $\theta_{O_2}$  can be approximated to be constant. Then, the PCO reaction rate is expressed as follows:

$$r_{PCO} = k \frac{K_{VOC} C_{VOC}}{1 + K_{VOC} C_{VOC} + K_W C_W} \quad (5)$$

### 2.2. Modeling of illumination intensity and modified L–H mechanism on photocatalytic building materials

Based on the pioneering work by Ollis et al. and other researchers (Mills et al., 2006; Ollis, 2005; Turchi & Ollis, 1990), the reaction rate constant  $k$  is expressed as a function of photon flux density  $I$ :

$$k = a_1 I^\beta \quad (6)$$

where  $a_1$  and  $\beta$  are the model constants. Assuming a low photon flux density of the general indoor environment,  $\beta$  is proposed to be 1.0 (Turchi & Ollis, 1990). Furthermore, Thimijan and Heins (1983) revealed that photon flux density  $I$  (photons m<sup>−2</sup> s<sup>−1</sup>) is simply proportional to the illumination,  $E$  (lx) (see Appendix A).

Generally, photocatalytic building materials are not perfectly covered with TiO<sub>2</sub> and their surfaces consist of a combination of TiO<sub>2</sub> and building materials (especially with porous structure contributing to physical adsorption) as the base substrate. This indicates that the reaction rate might exist under the condition of zero illumination level. By adopting the simple modeling to incorporate this background reaction into reaction rate constant  $k$ , Eq. (7) is derived as a combination of

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