



Effective liquid-phase photocatalytic reaction using a newly developed titanium oxide-assisted reactor with ozone microbubble treatment



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ABSTRACT

We have developed a TiO₂-assisted reactor that improved the UV light penetration into the solution during the photocatalytic decomposition of isopropyl alcohol (IPA). Increasing the UV penetration enhanced the decrease in the concentration of acetone, which is the main IPA decomposition product, and the total organic carbon. However, it did not affect the decrease in IPA concentration. Charging the solution with O₃ microbubbles significantly increased mineralization for the first 5 h of the reaction. Our results can be used to improve the efficiency of UV sources and develop an ecologically friendly, practical wastewater treatment for IPA.

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1. Introduction

2-Propanol (isopropyl alcohol; IPA) is extensively used in the semiconductor industry for cleaning the surface of silicon wafers. Large quantities of wastewater containing IPA, which can cause toxicity by ingestion, inhalation, or absorption, are generated. Therefore, effective industrial technology is needed for removing IPA from the wastewater before it is discharged into the environment [1].

Advanced oxidation processes (AOPs) are based on the production of hydroxyl radicals ($\cdot\text{OH}$) [2], and have been investigated for treating wastewater [3–5]. AOPs involving various combinations of ozone, hydrogen peroxide, sonolysis, ultraviolet (UV) radiation, and photocatalytic treatments have been used to degrade gas-phase volatile organic compounds (VOCs). The UV/TiO₂ photocatalytic reaction combined with ozone, hydrogen peroxide, and sonolysis has been used to degrade VOCs in both the gas and liquid

phases [2,6–10]. However, conventional reactors have a UV source at only the center of the reactor, which restricts the VOC reaction and the degradation of the intermediates. Thus, it is desirable to maximize the exposure of the reaction mixture to the UV source throughout the reactor. In particular, a UV source reaching the entire reactor would enhance the degradation of intermediates.

We have developed a TiO₂-assisted photocatalytic reactor, which enhances the UV light penetration into the solution, for effectively degrading IPA and its decomposition intermediates.

IPA has been frequently and widely used as a good model substance for photocatalytic oxidation experiments [1,11–15]. IPA is a major contaminant in indoor air and air streams [16–21]. It is also an important surface probe for semiconductor metal oxide photocatalytic reactions [19,22,23]. Furthermore, understanding the alcohol photocatalytic oxidation processes can enhance our knowledge of VOCs photocatalytic oxidation since VOCs are believed to oxidize photocatalytically through alcohol intermediates [24]. For a few decades, IPA adsorption and decomposition on TiO₂ has been studied at liquid phase, solid phase, and gas–solid interfaces [19,20,22–29]. The decomposition of IPA occurs by either dehydrogenation or dehydration, and its selectivity strongly depends on the reaction conditions. Under thermal oxidation, acetone, a dehydrogenation product, and propene, a dehydration product, were both detected [22]. However, only acetone and other intermediates were produced in the photooxidation process [20,23,28,29].

Abbreviations: AOP, advanced oxidation process; IPA, isopropyl alcohol; MB, microbubble; TC, total carbon; TOC, total organic carbon; UV, ultraviolet; VOC, volatile organic compound.

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Therefore, IPA was chosen as a model substance for our photocatalytic reaction.

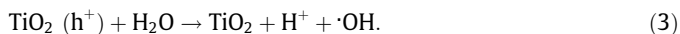
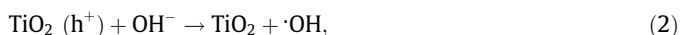
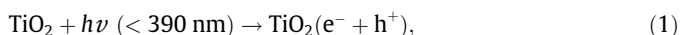
We investigated the degradation of IPA by observing its degradation intermediates, and assessed the effectiveness of the reactor for the UV₂₅₄/TiO₂ reaction and O₃ microbubble treatment. In order to develop an eco-friendly, practical treatment method we focused on the efficient use of the UV source to completely degrade IPA and its decomposition products. Our reactor treats 4 L of wastewater with an 8 W UV source, and the method is intended to be scaled up to a 40 L reactor with a 100 W UV source, or by using several low-energy UV lamps. The method is eco-friendly because the UV sources are used efficiently by increasing the UV light penetration of the solution with nine hollow quartz rods attached to the UV source casing.

2. Experimental

2.1. Reactor design

Unlike conventional UV reactors, our reactor consists of nine hollow quartz rods (50 × 10 mm) so that the UV light can penetrate the solution efficiently (Fig. 1). Initially, a UV₂₅₄/TiO₂ photocatalytic reaction was carried out to elucidate the degradation of IPA. The efficiency of the reactor was then tested for the UV₂₅₄/TiO₂ photocatalytic reaction with and without the quartz rods, by wrapping the rods with aluminum foil and Teflon tape to assess the effect of light penetration. The effectiveness of the UV/TiO₂/O₃ MB method was evaluated by charging the solution with O₃ MBs for 1 h before carrying out photocatalytic IPA decomposition with UV₂₅₄/TiO₂. The decomposition of IPA and acetone, which is the first degradation intermediate [1], were compared with the total organic carbon (TOC).

The degradation of pollutants by TiO₂ mainly occurs via oxidative decomposition by hydroxyl radicals, which can be generated from water under UV irradiation (<390 nm) in the presence of TiO₂ [8,30–32]:



The global reaction for the UV/O₃ process [33–35] is



2.2. Experimental procedure and analytical methods

A 254 nm UV light (8 W, Sankyo Denki Co., Ltd., Kanagawa, Japan) was used with a TiO₂ suspension (P25-TiO₂, Nippon Aerosil,

Tokyo, Japan). IPA (99.9%, Wako Pure Chemicals Industries, Ltd., Osaka, Japan) and acetone (99.5%, Wako) were used to prepare the IPA solution and to perform gas chromatography. The IPA concentration was set to approximately 10, 20, or 40 ppm in distilled water (4 L). The experiment was carried out as a batch reaction, and was mixed with the TiO₂ suspension (1 g/L) with a magnetic stir bar [6].

The effect of catalyst amount on the degradation of target chemical compounds is an important factor for the generation of active species as well as for the penetration of UV light. In our previous study, the effect of TiO₂ concentration in water suspension on the removal ratios of gaseous toluene and liquid aldehydes were investigated by varying the catalyst amount and the result indicated that 1.0 g/L of TiO₂ catalyst is the optimal catalyst amount. [6,36]. Furthermore, Wu and Yu [31] also investigated the effect of TiO₂ dosage in their UV/US/TiO₂ system for the decolorization of C.I. Reactive Red 2. Their study indicated that decolorization rate increased markedly with TiO₂ loading up to 0.5 g/L; however, only a slight increase to decolorization rate existed when the TiO₂ concentration was further increased to 2 g/L indicating reduced TiO₂-specific activity from light scattering and screening. Increasing TiO₂ catalyst amount over 1.0 g/L may decrease the light penetration into the reactor. Thus, we applied 1.0 g/L of TiO₂ catalyst as the optimal catalyst amount based on our previous study.

Pure air was used as the carrier gas between the UV lamp and the quartz rods. The total temperature change in the solution in the reactor from the initial reaction (26 °C) to the final reaction (33 °C) was 7 °C (2.6 × 10⁻³ °C/min). The degradation of IPA as a percentage of TOC was calculated from Eqs. (5) and (6):

$$\text{IPA degradation (\%)} = \frac{C_{(\text{IPA})0} - C_{(\text{IPA})t}}{C_{(\text{IPA})0}} \times 100, \quad (5)$$

$$\text{Mineralization (\%)} = \frac{C_{(\text{TOC})0} - C_{(\text{TOC})t}}{C_{(\text{TOC})0}} \times 100, \quad (6)$$

where $C_{(\text{IPA})0}$ and $C_{(\text{TOC})0}$ are the initial IPA and TOC concentrations, and $C_{(\text{IPA})t}$ and $C_{(\text{TOC})t}$ are the IPA and TOC concentrations after time t . The conversion of IPA and acetone concentration to carbon concentration (μg C/mL) was calculated from Eqs. (7) and (8):

$$\text{IPA (\mu g C/mL)} = \left(\frac{\text{Carbon MW} \times \text{IPA}_{(\text{C}3)}}{\text{IPA MW}} \right) \times C_{(\text{IPA})t} (\mu\text{g/mL}), \quad (7)$$

$$\text{Acetone (\mu g C/mL)} = \left(\frac{\text{Carbon MW} \times \text{ACE}_{(\text{C}3)}}{\text{ACE MW}} \right) \times C_{(\text{ACE})t} (\mu\text{g/mL}), \quad (8)$$

where carbon MW, IPA MW, and ACE MW indicate the molecular weight of carbon, IPA, and acetone respectively. $\text{IPA}_{(\text{C}3)}$ and $\text{ACE}_{(\text{C}3)}$ indicate the carbon number of the compounds. $C_{(\text{IPA})t}$ and $C_{(\text{ACE})t}$ are the concentrations of IPA and acetone after time t .

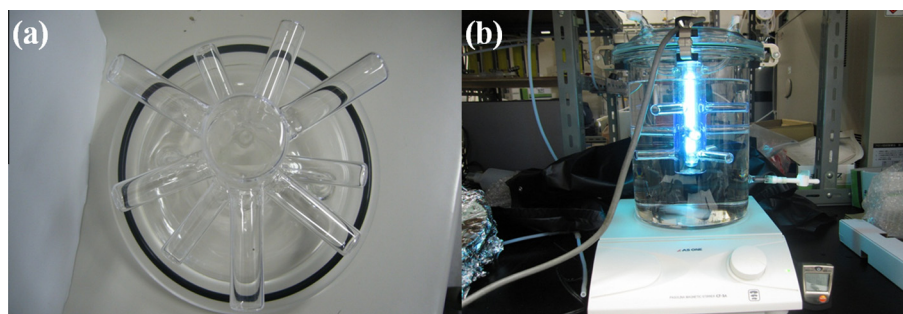


Fig. 1. Photographs of the UV reactor. (a) The UV reactor with the nine hollow rods, and (b) the UV reactor in the reaction vessel.

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