



Catalytic and photocatalytic ozonation of tert-butyl alcohol in water by means of falling film reactor: Kinetic and cost-effectiveness study



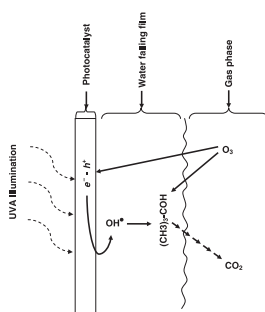
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HIGHLIGHTS

- A complete decomposition of TBA was observed after 15 min using $\text{TiO}_2/\text{O}_3/\text{UVA}$ systems.
- Presence of metallic ions increased the oxidation rate of TBA in wastewater by TiO_2/O_3 .
- Photocatalytic ozonation systems presented first order kinetics for the TBA oxidation.
- Catalytic ozonation in wastewater showed the lowest electrical energy use per order.

GRAPHICAL ABSTRACT



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ABSTRACT

This study investigates the oxidation conditions and kinetics of TBA as a pollutant in aqueous solutions using two different advanced oxidation processes; catalytic ozonation in the dark (TiO_2/O_3) and in the presence of UVA illumination ($\text{TiO}_2/\text{O}_3/\text{UVA}$). The oxidation experiments were conducted in deionised water and in a real wastewater sample using a falling film reactor. The utilised electrical energy per order (E_{EO}) under different oxidation conditions was calculated to investigate the cost effectiveness of the studied oxidation processes with regard to the electrical energy consumed by the treatments. It was observed that the oxidation of TBA by photocatalytic ozonation systems follows pseudo-first-order kinetic with respect to the concentration of TBA. The presence of metallic ions and increase of ozone and TBA concentration in the oxidation medium led to an increase of the oxidation rate of TBA in water. Catalytic ozonation in the dark showed the best cost effectiveness among the other oxidation conditions in the degradation of TBA in wastewater, with a consumed electrical energy of 12 kW/m^3 order.

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1. Introduction

tert-Butyl alcohol (TBA) has been used as a component in paint removers, industrial cleaning compounds, perfumes and a variety of cosmetics as well as an ingredient in the synthesis of some chemical substances. It is also added to gasoline as an octane booster to increase its octane rating and reduce atmospheric emissions

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[1,2]. Therefore, it is very often observed in the effluent of production units of these products.

In addition, TBA has been reported as a stable intermediate in the degradation of other fuel oxygenates such as methyl tert-butyl ether (MTBE) and ethyl tert-butyl ether (ETBE) [3,4]. Due to the tertiary structure of the molecule of TBA, it is more stable against oxidation and less reactive than other butanol isomers. Therefore, it can frequently accumulate during the biodegradation of MTBE and ETBE [5,6]. The major source of ground water contamination with TBA and other fuel oxygenates is fuel leaking from underground storage tanks [6,7].

Some evidence of carcinogenic activity in TBA-contaminated water samples has been reported in rats and mice [8,9], which implies the toxicity of this compound and health risks arising from its presence in water. Therefore, many remediation technologies have reported on the treatment of aqueous TBA solutions to characterise and evaluate its removal conditions from water [10–13]. Despite the low treatment costs of biological remediation methods, they require relatively long treatment times and have low degradation efficiencies for the treatment of this compound. Alternatively, the high reaction rate of this compound with hydroxyl radicals has attracted much attention to the elimination of TBA from water using advanced oxidation processes (AOPs) in recent years [14–16]. In advanced oxidation processes, different combinations of reagents, catalysts and illuminations are utilised to generate hydroxyl radicals as highly oxidative reagents in the oxidation medium to attack the contaminant molecules in water and oxidise them non-selectively [17,18]. One of these processes is the heterogeneous combination of ozone with a catalyst in the dark (catalytic ozonation) or in the presence of UV irradiation (photocatalytic ozonation) and utilising the increased synergy of this combination in generating hydroxyl radicals for the mineralisation of different pollutants in water [19,20].

According to our literature review, there has been no investigation of the oxidation conditions and kinetics of TBA as a contaminant in water using catalytic and photocatalytic ozonation processes. Therefore, the purpose of the present study is to investigate the oxidation conditions and kinetics of TBA in water and wastewater using catalytic ozonation in the dark (TiO_2/O_3) and in the presence of UVA illumination ($\text{TiO}_2/\text{O}_3/\text{UVA}$) by means of a falling film reactor. Pilkington Activ™ glass was used in the structure of the reactor as an immobilised catalyst. The effect of some experimental variables such as: concentration of TBA in the aqueous solution, ozone concentration in the liquid phase, presence of metallic ions in the oxidation medium and, pH on the oxidation rate of TBA are evaluated and discussed. At the end, the electrical energy per order was calculated for the investigated advanced oxidation processes under different experimental conditions to evaluate the cost effectiveness of the used oxidation methods in the treatment of water and wastewater in terms of electrical energy.

2. Materials and methods

2.1. Chemicals and reagents

The tert-butyl alcohol utilised in this study was analytical grade product purchased from Merck, Germany. It was diluted to the required concentrations and used in all experiments without any further adaption.

Ozone was produced from pure oxygen (>99.5 vol.% and $\text{H}_2\text{O} < 200$ ppmv, Air Liquide) using an ozone generator model OZ 502/10 manufactured by Fischer, Germany.

On demand, NaOH, Lachema (Czech Republic) was used to increase the pH of the aqueous solution of TBA.

Pilkington Activ™ glass (PAG) was used as immobilised photocatalyst in the present study. This kind of glass is well-known as a self-cleaning glass. It is coated with a thin, colourless, and invisible layer of TiO_2 . The photocatalytic and superhydrophilic activity of PAG, which prompted its utilisation in the photocatalytic treatments, have been reported elsewhere [20–22]. The surface of the PAG was illuminated and activated by UVA lamps to initiate the oxidation of TBA solutions by the $\text{TiO}_2/\text{UVA}/\text{O}_3$ systems therein. The UVA lamps (KG, LT 15 W/009 UV) were produced by Narva Lichtquellen GmbH & Co., Germany.

In addition to performing the oxidation experiments on TBA in deionised water under controlled synthetic reaction conditions,

these experiments were also conducted on TBA as a constituent of a real wastewater sample to investigate its oxidation under realistic conditions. For this aim, a wastewater sample was obtained from a production site of methyl tert-butyl ether (MTBE) in Schwedt, Germany to be used as a matrix for the oxidation of TBA in this series of experiments. Some characterisation results of the real wastewater are presented in Table 1. This wastewater was chosen as an oxidation matrix for TBA oxidation due to its containing MTBE in its composition. TBA is considered one of the toxic degradation products of MTBE, which can accumulate in water. Thus, the results of these experiments can provide valuable information about the removal of TBA from a potential wastewater using catalytic and photocatalytic ozonation systems.

To conduct these experiments, TBA was spiked into the above-mentioned wastewater sample to a prepared concentration of 0.1 mM with respect to TBA. Then, the oxidation of this compound was followed as a constituent of wastewater using catalytic and photocatalytic ozonation. The amount of TBA spiked into the wastewater ($C_0 = 0.1$ mM) was about 10 times greater than the amount of TBA that could be produced from the degradation of MTBE existing in wastewater. Therefore, the latter could not significantly influence the oxidation results under these conditions.

2.2. Falling film reactor and setup details

The oxidation experiments were performed in aqueous TBA solutions using a falling film reactor. The structure of the falling film reactor included a rectangular frame made of aluminium which was enclosed on both sides by two sheets of Pilkington Activ™ glass. These glass sheets were part of the structure of the falling film reactor, forming the reactor walls (Fig. 1). The internal volume of reactor was 0.01 m^3 .

For each oxidation experiment, 1 L of an aqueous solution of TBA was injected into the reactor through the top cap so that it would fall over the surface of the PAG sheets, forming liquid falling films, and would leave the reactor through an output in the reactor bottom. The TBA solution was recycled continuously through the reactor using a gear pump during the oxidation time. The recirculation rate was 1 L/min. An ozone analyser was placed between the reactor and the gear pump in order to measure the ozone concentration in the aqueous TBA solution online. At the same time, a gas stream of ozone/oxygen flowed at a rate of 10 L/h from the bottom input points into the reactor and left the reactor from the top outlet to an ozone analyser for determining the ozone concentration in the gas phase.

Seven UVA lamps were fixed inside the reactor to illuminate the surface of the Pilkington active glasses. The incident light intensity of UVA lamps was $1 \text{ mW}/\text{cm}^2$ with a maximum wavelength at 360 nm. The photoactive surface area of the reactor walls was about 0.5 m^2 .

Table 1
Characteristics of the real wastewater.

TOC	26.1	mg/L
DOC	19.2	mg/L
MTBE	850	μg/L
NH_4^+	600	μg/L
Cl^-	40,000	μg/L
Br^-	<1000	μg/L
NO_2^-	<1000	μg/L
NO_3^-	<1000	μg/L
PO_4^{3-}	<1000	μg/L
SO_4^{2-}	23,000	μg/L
Mn^{2+}	2400	μg/L
$\text{Fe}^{2+}, \text{Fe}^{3+}$	15,500	μg/L
pH	≈8	

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