



Photochemical degradation of trichloroethylene by sulfite-mediated UV irradiation



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ABSTRACT

This study investigates the degradation of trichloroethylene using advanced reduction processes (ARP), which was developed by combining activation methods and reducing agents to produce reactive reducing radicals that can destroy persistent oxidized contaminants. Screening experiments were performed using a combination of three reducing reagents (sulfite, sulfide, and dithionite) and three UV light sources (monochromatic light at 254 nm and two UV lamps with primary energy peaks at 365 and 312 nm, respectively). The screening test results showed that the highest TCE removal efficiency was obtained by the combination of sulfite with low-pressure UV light, monochromatic at 254 nm. The effectiveness of sulfite/UV-L ARP for dechlorinating TCE with regard to the initial TCE concentration, sulfite dose, solution pH, and light intensity was evaluated. Additionally, the reaction mechanisms and kinetics of TCE degradation were proposed. The photochemical degradation of TCE in ARP is attributed to sulfite radicals and hydrated electrons and generally followed first-order decay kinetics. At high pH and high sulfite dose, TCE was completely degraded, with chloride ions as the major reaction product. The TCE dechlorination rate constant increased as the sulfite dose, pH, and light intensity increased. This study provides the effectiveness of sulfite/UV-L ARP for TCE dechlorination and suggests optimum conditions that result in complete TCE conversion to a nonhazardous byproduct, chloride ions, and a rapid TCE dechlorination rate.

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Introduction

Trichloroethylene (TCE) is a chlorinated hydrocarbon that is commonly used as a dry cleaning and degreasing solvent. TCE is often found in soil and groundwater, and surface water as a result of leaks from underground storage tanks and inappropriate usage and disposal [1,2]. The United States Environmental Protection Agency (U.S. EPA) has listed TCE as a priority pollutant and established a maximum contaminant level (MCL) for TCE of $5 \mu\text{g L}^{-1}$ in drinking water supplies [3]. Because TCE is not easily biodegraded by natural attenuation and can be converted to more potent carcinogenic compounds, such as vinyl chloride (VC), several remediation technologies for TCE degradation in water have been proposed. These treatment technologies include bioremediation, air stripping, adsorption on activated carbon,

thermal treatment, dechlorination, and chemical oxidation [4–6]. Recently, various functionalized materials including nanoscale zero valent iron, mesoporous silica, and magnetic maghemite/chitosan nanocomposite films have been developed and applied to environmental remediation for organic pollutants [7–9]. Photo-induced advanced oxidation processes (AOPs) to treat TCE were introduced as alternative environmental degradation methods [6,10]. Also, catalytic photolysis of various contaminants that contain TCE using UV light has been reported by many researchers [11–23]. TiO_2 has commonly been used as a photocatalyst with UV light for TCE removal [11–20]. Efficient TCE photodegradation methods have been developed with the addition of hydrogen peroxide (H_2O_2) [5,24–26], ozone (O_3) [18], chlorine [25], titania supported on silica [27], Fe/TiO_2 [28,29], and a nano-ZnO/Laponite composite [30], along with UV irradiance.

Monochromatic UV light at 254 nm was widely used for TCE photolysis. Chu and Jia investigated TCE photolysis using three different monochromatic UV lamps at 254, 300, and 350 nm [10]. This study showed that UV light at 254 nm resulted in the greatest TCE removal rate compared with that at the other two UV lamp wavelengths (300 and 350 nm) [10].

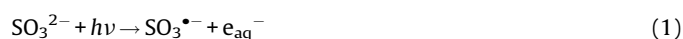
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Recently, there has been interest in chemical degradation using advanced reduction processes (ARP). In ARP, a reducing agent is combined with an activating method to produce highly reactive reducing radicals. ARPs have been successfully applied to destroy various persistent and oxidized contaminants, such as VC, 1,2-dichloroethane (1,2-DCA), perchlorate, nitrate, bromate, perfluorooctanoic acid, and 2,4-dichlorophenol [31–39]. The advantage of ARP is to destruct the target contaminants, not transfer to other phase, and convert the chemicals to environmentally benign compounds, showing high efficiency and fast removal rate [31–33]. Table S1 summarizes the published data for TCE degradation by UV photolysis with respect to degradation products and removal efficiency (see Supplementary material).

Previous studies of ARPs have shown that an ARP that combines sulfite with a low-pressure UV mercury lamp that is monochromatic at 254 nm (UV-L) could rapidly and efficiently destroy chlorinated organics [32–34]. When sulfite is irradiated by UV light, it produces sulfite radicals ($\text{SO}_3^{\bullet-}$) and hydrated electrons (e_{aq}^-) (Eq. (1)) [40]. It may also produce hydrogen radicals (H^\bullet) when HSO_3^- is predominantly at low pH, as shown in Eq. (2) [40,41]:



It was reported that sulfite radicals and hydrated electrons are reactive species to degrade chlorinated organic compounds in a sulfite/UV process [40,41], and these are expected to act as a potential reductant to degrade TCE in ARP. The objective of this study is to investigate the kinetics and efficiency of TCE dechlorination in sulfite/UV-L ARP and to understand the influence of process variables such as sulfite dose, pH, initial TCE concentration, and light intensity. Additionally, a TCE dechlorination mechanism was proposed, and quantum yields of TCE photodegradation were evaluated.

The cost and efficiency of ARP for TCE dechlorination will vary with many factors, such as reducing agent doses, solution pH, water constituents, and UV irradiance. Therefore, this study will provide a fundamental knowledge in order to support development of ARP to treat TCE as a practical water treatment method.

Materials and methods

Experimental procedures

The trichloroethylene ($\geq 99.5\%$), methanol ($\geq 99.9\%$), and hexane (95%) used in this study were purchased from Sigma-Aldrich. Analytical reagent grade anhydrous sodium sulfite and sodium chloride were purchased from Fisher Scientific. All solutions were prepared in an anaerobic chamber (Coy Laboratory Products) filled with nitrogen gas. All aqueous solutions were prepared using deionized deoxygenated water (DDW). DDW was prepared by purging high purity nitrogen gas (99.9995%) into deionized water for at least 2 h. To compare the results obtained with sulfite/UV ARP, batch experiments were conducted with a blank control (only TCE, no reagent, no UV), a reagent control (TCE and sulfite, no UV), and an irradiation control (TCE and UV, no reagent).

TCE stock and standard solutions were prepared in amber glass vials (nominally 40 mL) and closed using a screwed cap with PTFE/silicon septa. And clear quartz vials (42 mL \pm 0.1, nominally 40 mL) were used as batch reactors (J&J Science, Korea) with butyl rubber septa and aluminum caps. A stock solution of TCE in methanol was daily prepared to prevent TCE loss. The volume of 250 μL of the methanolic stock solution of TCE was spiked using a gas-tight syringe into a batch reactor containing sulfite solution to yield a target organic concentration of 0.23 mM.

The sulfite solution was prepared in 5 mM phosphate solution and transferred into a batch reactor. 0.1 M phosphate buffer (pH 7.0 buffer BDH[®]) was used to adjust the pH and was diluted to 5 mM phosphate in the reactor. The sulfite dose in experiments with TCE was ranged from 0.46 to 11.5 mM. For experiments to evaluate pH effects, 1 N HCl or 1 N NaOH were added to samples to obtain the targeted pH. The prepared samples were shaken at 250 rpm for 30 min then taken to the UV light chamber.

The ultraviolet light source included T-8C, T-8M, and T-8L lamps purchased from the Vilber Lourmat Company. The T-8C lamp was monochromatic at 254 nm (UV-L), and the T-8L and T-8M lamps had primary wavelengths at 365 nm in the range of 320–380 nm (UV-M) and 312 nm in the range of 280–320 nm (UV-B), respectively. Five 8 W UV lamps were placed in the irradiation chamber (14.5(H) \times 33(D) \times 26(W) cm, BioLink, Vilber Lourmat). The distance between the lamps and the reactor was 15 cm, and the light intensity was measured using either an ST-512 light meter (UVC, 220–275 nm, calibration point 254 nm) or an ST-513 (UVAB, 280–400 nm, calibration point 365 nm).

Analytical methods

To extract TCE, 4 mL sample of supernatant after UV irradiation was rapidly transferred using a gas-tight syringe into amber glass vials (nominally 20 mL) containing 4 mL hexane. After shaking at 250 rpm for 30 min, the extractant of 2 mL was transferred into GC autosampler vials (2 mL, amber glass). TCE was analyzed using an Agilent Technologies 7890A gas chromatograph (GC) equipped with a micro-electron capture detector (μECD) with a DB-1 column (Agilent Technologies J&W 123-1035, 30 m \times 320 μm \times 5 μm). The amount of each injected sample was 0.1 μL with a split ratio of 10:1. The oven temperature program began at 40 $^\circ\text{C}$, which was held for 3 min, was increased at 10 $^\circ\text{C min}^{-1}$ to 150 $^\circ\text{C}$ and held for 2 min, and was increased at 20 $^\circ\text{C min}^{-1}$ to 180 $^\circ\text{C}$ and held for 1 min. The temperatures of the injector and detector were 210 $^\circ\text{C}$ and 280 $^\circ\text{C}$, respectively. Helium was used as a carrier gas with a flow rate of 45 mL min^{-1} , while nitrogen was used as a makeup gas with a flow of 20 mL min^{-1} . TCE degradation byproducts were analyzed using mass spectrometry GC (Varian GC-MS) with a DB-1 column and an oven program used in GC- μECD .

To analyze the concentration of chloride ions produced from TCE dechlorination, a Dionex ICS-5000 ion chromatograph equipped with a dual gradient pump, AS autosampler, and eluent generation module was used. The eluent used was 4.5 mM Na_2CO_3 /0.8 mM NaHCO_3 with a flow rate of 0.25 mL min^{-1} . The injection volume was 1200 μL at 30 $^\circ\text{C}$, and was detected with suppressed conductivity using an anion self-regenerating suppressor (ASRS 300, 2 mm) with a 7 mA applied current. UV-vis absorbance was measured using a PerkinElmer (Lambda 25) UV-vis spectrophotometer.

Results and discussion

Screening test results

Screening experiments were conducted for each combination of three different reducing agents (sulfite, sulfide, and dithionite) and three UV light sources (UV-L, UV-M, and UV-B), resulting in nine experiments. The irradiation time in these experiments was fixed at 3 h. In these experiments, no pH buffer was used. Fig. 1 shows the results of the screening experiments and indicates that UV-L generally showed high TCE removal above 90%, regardless of the types of reducing agent used. Additionally, the use of UV-L alone without a reducing agent showed a high TCE removal that approached 94%. Chu and Jia investigated the decay of TCE by direct photolysis using three monochromatic UV lamps at different

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