Electrochemical Plasma for Treating 2,4,5-Trichlorophenoxyacetic Acid in a Water Environment Using Iron Electrodes

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ABSTRACT: Herbicide compounds containing aromatic rings and chlorine atoms, such as 2,4,5-trichlorophenoxyacetic (2,4,5-T), cause serious environmental pollution. Furthermore, these compounds are very difficult to decompose by chemical, physical, and biological techniques. Fortunately, the high-voltage direct current electrochemical technique can be controlled to form a plasma on metallic electrodes. It creates active species, such as H_2, O_2, and H_2O_2, and free radicals, such as $H^\bullet$, $O^\bullet$, and $OH^\bullet$. Free radicals that have a high oxidation potential (e.g., $OH^\bullet$) are highly effective in oxidizing benzene-oring compounds. Iron electrodes are used in the study to combine the dissolving process of the iron anode electrode to create Fe^{2+} ions and the electrochemical Fenton reaction. In addition, the flocculation process by $Fe(OH)_2$ also occurs and the plasma appears with a voltage of 5 kV on the iron electrode in a solution of 30 mg L^{-1} of 2,4,5-T. After a period of time of the reaction, the aromatic-oring compounds containing chlorine were effectively treated, and the electric conductivity of the solution increased due to the amount of Cl^- ions released in the solution and the decrease in the pH value. The degradable products of 2,4,5-T were qualitatively characterized by gas chromatography—mass spectrometry (GC−MS), and it was determined that straight-chain carboxylic acids are formed in the solution. These compounds are easy to oxidize thoroughly under appropriate conditions in a solution via $OH^\bullet$ free radicals. Moreover, 2,4,5-T was also quantitatively analyzed using a calibration curve from GC−MS and high-performance liquid chromatography (HPLC). Furthermore, this work also suggests that the performance of the treatment process can be optimized by controlling the technological factors, such as the input voltage, the distance between anodic and cathodic electrodes, the initial concentration of 2,4,5-T, and flowing air through the solution that represents an approximately 99.83% degradable efficiency. Finally, the work demonstrates a potential technology for treating the 2,4,5-T compound, particularly for environmental pollution treatments.
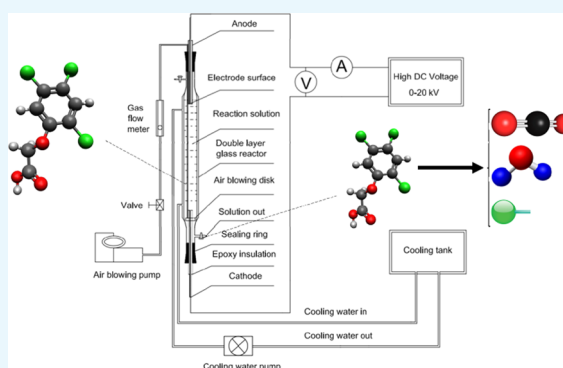
INTRODUCTION

Plasma is the fourth state of matter. It is classified based on temperature and is divided into high thermal plasma and cold plasma. Thermal plasma occurs at high temperatures ranging from 10^6 to 10^8 K. Thermal plasma is the fusion reaction, solar wind, and stars. Cold plasma occurs at a low-temperature range from $300 \div 2 \times 10^4$ K.[1−3] It usually is the process of electric discharge at a high voltage from $0 \div 40$ kV[3] and can be generated by many different techniques, such as AC voltage, DC voltage, and pulse voltage.[4]

Cold plasma can be classified into various categories, such as electric discharge in air, electric discharge in solution, electric discharge at low pressures, or electric discharge at high pressures, and depends on the characteristics of the dielectric layer. On the other hand, under various electric discharge conditions, it will lead to the generation of $OH^\bullet$, $O^\bullet$, and $H^\bullet$ free radicals as well as O, H, O_3, and H_2O_2 active species.[5−12] These are the agents that have high redox properties.[13] For these reasons, cold plasma has many practical applications, such as in surface treatment, materials manufacturing, and food preservation. It is commonly used in environmental treatment, such as wastewater treatment, exhaust gas treatment, and drinking water treatment.[14−20] One of the ways it is used to treat pollutant compounds is a method using high-voltage DC combined with an iron electrode. The process of electric discharge directly in a solution creates free radicals and other active species, such as $OH^\bullet$ and H_2O_2.[21,22] In addition, the corrosion process on the iron anodic electrode also produces Fe^{2+} ions, which play the role of Fenton agents that increase the degradable efficiency of pollutant organic substances.[23−27]
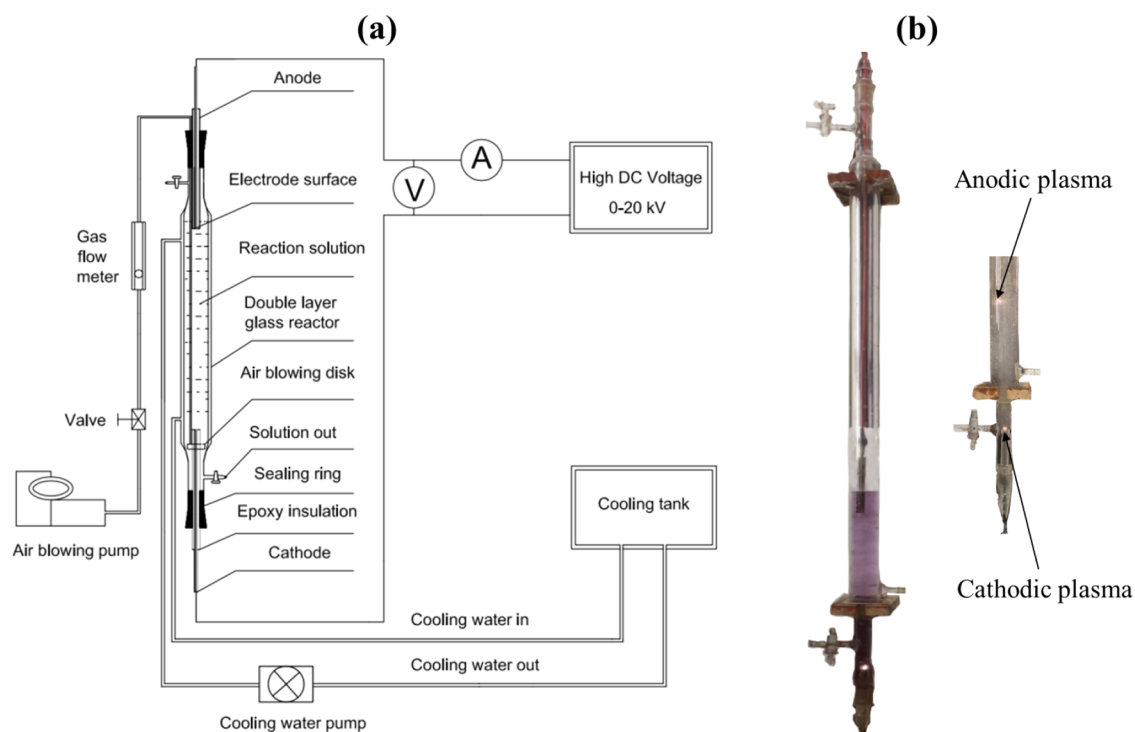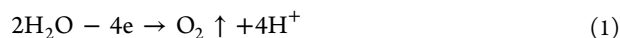
(a)   **(b)**

Figure 1. Diagram of the high-voltage DC power source including the (a) full components and (b) experimental cell with plasma appearances.

The free radicals and other active species can be formed according to the following mechanisms:

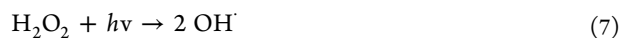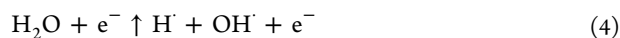The electrolysis reaction releases oxygen gas on the anodic electrode

$$2H_2O - 4e \rightarrow O_2 \uparrow + 4H^+ \tag{1}$$

The electrolysis reaction releases hydrogen gas on the cathodic electrode

$$2H_2O + 2e \rightarrow H_2 \uparrow + 2OH^- \tag{2}$$
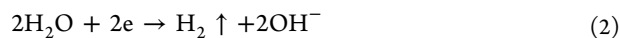
The electrolysis reaction dissolves the iron anodic electrode

$$Fe - 2e \rightarrow Fe^{2+} \tag{3}$$

The mechanisms generate the free radicals and the active species[28−30]

$$H_2O + e^- \uparrow H^. + OH^. + e^- \tag{4}$$

$$OH^. + OH^. \rightarrow H_2O_2 \tag{5}$$

$$2H_2O \rightarrow H_2O_2 + H_2 \tag{6}$$

$$H_2O_2 + hv \rightarrow 2 OH^. \tag{7}$$

Plasma decomposes water by the following reaction mechanism[31]

$$6H_2O \rightarrow 4H_2 + O_2 + 2H_2O_2 \tag{8}$$

$$2H_2O \rightarrow 2H_2 + O_2 \tag{9}$$
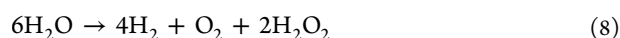
The Fenton reaction produces $OH^\bullet$ free radicals according to the reaction[25,32]

$$Fe^{2+} + H_2O_2 \rightarrow OH^. + OH^- + Fe^{3+} \tag{10}$$

The concentration of H_2O_2 is determined by reacting with titanyl ions to form a yellow complex by the following reaction

$$Ti^{4+} + H_2O_2 + 2H_2O \rightarrow TiO_2 . H_2O_2 + 4H^+ \tag{11}$$

the maximum absorption wavelength λ_{max} was 407 nm.[33]

It indicates that the Fenton reaction products are potentially used to oxidize contaminants and/or wastewater, as well as degrade the organic compounds. Based on these advantages, this work aims to use the electrochemical plasma for treating 2,4,5-trichlorophenoxyacetic acid (2,4,5-T) in a water environment using iron electrodes, which could be applied to treat environmental pollutions.

Degradation percentage can be calculated by the following equation

$$H(\%) = \frac{C_0 - C_t}{C_0} \times 100\% \tag{12}$$

where C_0 and C_t are the initial concentrations and the final concentrations of 2,4,5-T (mg/L), respectively.

■ PROCEDURES

Materials. The power source used in this study is a high-voltage DC. The voltage is regulated steplessly from 0 to 20 kV and the power capacity ranges from 0 to 30 kVA. The electrode material used in the reaction is made of Fe. The electrode diameter ($\varnothing$) is 3 mm, and the electrode length (l) is 250 mm. The iron electrode is molded by a low-viscosity epoxy system with a 17:1 ratio of resin and hardener from EpoFix. The iron electrode area that is in contact with water is 7.068 mm^2. The reaction vessel is made of heat-resistant glass and consists of two layers, the inside layer containing the reaction solution and the outside layer containing cooling water that is pumped cyclically. The reaction vessel has a valve to get the reaction solution below and a valve to release gases at the top. The reaction process is observed using a camera. The detailed description of the reaction diagram is given in Figure 1 in which full components are shown in Figure 1a and the experimental cell with anodic and cathodic plasma appearances is given in Figure 1b. 2,4,5-T herbicide was purchased from

Merck, Germany. The double-distilled water has an electrical conductivity of 1.3 μS cm^{-1} and pH of 7. 2,4,5-T is dissolved in double-distilled water to reach a concentration of 30 mg L^{-1}. *N,O*-Bis(trimethylsilyl)trifluoroacetamide (BSTFA) was used to silylate organic acids in solution and analyzed by a gas chromatography−mass spectrometry (GC−MS 6890-5975 Agilent).

Characterization. High-performance liquid chromatography (HPLC 1100 Agilent) was used to analyze the degradation of 2,4,5-T. The sample is injected directly into the HPLC system with the following analytical conditions: hypersil column C$_{18}$ (200 × 4 mm^2), λ = 285 nm, mobile phase with acetonitrile and H$_2$O = 70:30 (v.v^{-1}), flow rate = 0.5 mL min^{-1}, and pressure = 60 bar. GC−MS was used for the qualitative determination of intermediate products formed in the reaction solution from the 2,4,5-T decomposition. The analytical process with the water sample is extracted by diethyl ether and derived with BSTFA to form volatile compounds. After that, the sample is analyzed on the HP-5MS column with the following conditions: a scan parameter of 35 ÷ 550 amu, 1.3 mL min^{-1} of initial flow of He, 41 cm sec^{-1} average velocity, 70 keV of energy, 40 °C initial temperature in the temperature program with a 10 °C min^{-1} rate and 280 °C final temperature, a holding time of 5 min, and 1 μL of injection volume. The conductivity of the solution was adjusted by NaCl salt and measured by the HI 8733 Hanna Instrument, with the range from 0.0 ÷ 199.9 mS cm^{-1} and ±1% accuracy. The initial pH value was adjusted by H$_2$SO$_4$ 5 × 10^{-2} M or NaOH 2 × 10^{-2} M solutions and characterized by the pH meter (HI 8314, Hanna Instrument) with the pH range from 0 to 14 and ±0.01 accuracy. The amount of chemicals is weighed on a Shimadzu ATX 224 analytical balance with an accuracy of ±0.1 mg. The concentration of H$_2$O$_2$ was measured by UV−Vis instrument UH-5300, Hitachi. The chemical oxygen demand (COD) and total organic carbon (TOC) were determined by a chemical method and a TOC-5000A system, Shimadzu, respectively.

■ RESULTS AND DISCUSSION

The investigation of the formation of H$_2$O$_2$ generated by the discharging of the Fe electrode in a double-distilled water solution in the presence of plasma showed that the concentration of H$_2$O$_2$ increased over time, and the results are given in Table 1 and Figure 2. The discharge in water in

Table 1. Concentration of H$_2$O$_2$ Generated on the Fe Electrode

time (min)	10	20	30	40	50	60
$C_{H_2O_2}$ (mg/L)	0.014	0.027	0.037	0.040	0.042	0.043

the presence of plasma on the electrodes produced active species such as H$_2$O$_2$ and OH$^\bullet$ free radicals, as well as decreased the pH and increased the conductivity of the investigated solution. The use of the Fe electrode also generated Fe^{2+} ions that are dissolved due to the electrochemical reaction of anodic dissolution. Fe^{2+} ions reacting with H$_2$O$_2$ could be formed in the solution, leading to an enhanced formation of OH$^\bullet$ free radicals by the Fenton reaction. Therefore, this phenomenon could increase the degradation efficiency of the pollutant compounds.

Studying the dependence of the degradable efficiency of 2,4,5-T on time was carried out under the following

conditions: the distance between the two electrodes of 300 mm, the initial concentration of the 2,4,5-T solutions of 30 mg L^{-1}, the input voltage of 5 kV, the temperature of the solution of 30 °C, the initial electrical conductivity of 38.5 μS cm^{-1}, and the initial pH of the solution of about 6. The results show that the degradable efficiency achieved for 2,4,5-T was 36.97, 50.80, 61.83, and 71.17% for the reaction duration times of 30, 60, 90, and 120 min, respectively.

As such, it can be seen that the degradable efficiency increases over time, as shown in Figure 3a. For this reason, increasing the reaction time leads to more OH$^\bullet$ free radicals, and the amount of active species, such as H$_2$O$_2$, increases. Furthermore, the decomposition of 2,4,5-T also depends on the input voltage. The results indicate that the degradable efficiency achieved for 2,4,5-T was 14.3, 34.2, 55.10, 70.17, and 81.77%, which corresponded to the input voltages of 2, 4, 6, 8, and 10 kV, respectively. The reason for the increase in degradable efficiency was due to an increase in input voltage, thus leading to plasma formation. In addition, the area of the plasma that was formed was larger. Therefore, the formation of free radicals and active species was larger. This means that the oxidative ability was higher, according to formulas 1−10 that were cited earlier. Because of this, the degradable efficiency of the 2,4,5-T pollutants increases with voltage, as shown in Figure 3b.

Figure 3c illustrates the influence of electrode distance on the decomposition of the 2,4,5-T compound. The results show that when electric discharging at 5 kV input voltage with the initial concentration of 30 mg L^{-1}, the degradable efficiency of 2,4,5-T after 30 min was 45.56, 36.97, 28.56, 20.47, and 4.69%, which corresponded to the increasing electrode distance of 250, 300, 350, 400, and 500 mm, respectively. From the results, it can be seen that when the distance was larger, the degradable efficiency of 2,4,5-T was lower. The cause of the decrease in efficiency with the increase in the electrode distance could be the reduction in the kinetic energy of the electrons in the solution, the slower process of plasma formation, and the smaller area of the plasma formed. Therefore, the ability to form OH$^\bullet$ free radicals and active species (e.g., H$_2$O$_2$) was lower. Because of all these causes, the results led to a reduction in the degradable efficiency of 2,4,5-T when the distance between the two electrodes increased.

The degradable efficiency also depends on the initial concentration. The scale of the oxidative agent compared with the pollutant compounds was reduced due to the formation of OH$^\bullet$ and H$_2$O$_2$ under the same conditions. For this reason, the degradable efficiency was reduced, as shown in Figure 3d. The analytical results show that the degradable efficiency of 2,4,5-T at 30 mg L^{-1} concentration was 36.97, 50.80, 61.83, and 71.17%. Meanwhile, the degradable efficiency of 2,4,5-T at 50 mg L^{-1} was only 21.35, 39.46, 53.95, and 62.81%, corresponding to the electrolysis reaction time and plasma appearance time of 30, 60, 90, and 120 min, respectively. It is clear that the initial concentration of 2,4,5-T affects the treatment efficiency. When the initial concentration was high, the treatment efficiency decreased. In particular, the electric discharge process had a plasma appearance, simultaneously, the combining with process flowing air via the solution and the flow was about 500 mL min^{-1} to make increasing the degradable efficiency of 2,4,5-T, as shown in Figure 3e. The experimental results at 30, 60, 90, and 120 min indicated that when the 2,4,5-T concentration was 30 mg L^{-1}, the initial pH was 6, the initial electric
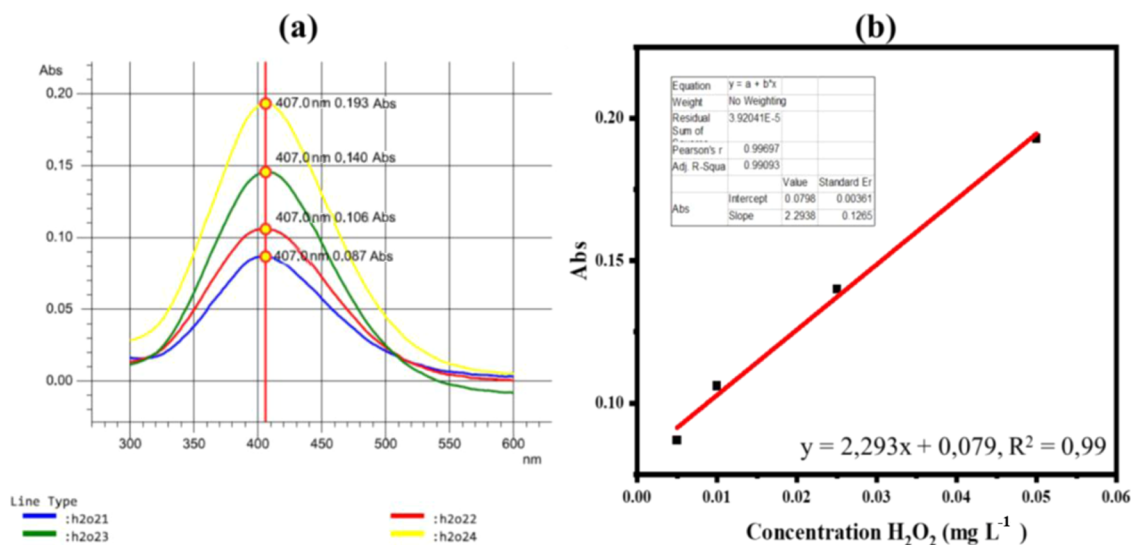
Figure 2. (a) UV−Vis spectrum of the titanyl ions and H_2O_2 complex and (b) quantitation standard curve of H_2O_2.

conductivity was 38.5 μS cm^{-1}, and the degradable efficiencies was 38.72, 67.52, 85.26, and 99.83%, respectively. It can clearly be seen that the degradable efficiency was the highest when the process had air flowing through the solution.

This can be explained by the fact that when air flows through the solution, it comes in contact with the plasma and therefore increases the concentration of ozone. Hence, ozone catalyzed the formation of free radicals OH$^\bullet$ by the reaction

$$3O_3 + H_2O \rightarrow 2OH^. + 4O_2 \tag{13}$$
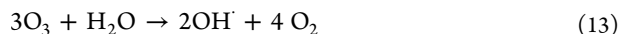
This led to a higher oxidative ability and an increased decomposition of the 2,4,5-T pollutant compound.[7,29] In addition, oxygen gas also reacted with phenol compounds, leading to a ring opening. In general, from the research results, it can be seen that when air flowed through the solution, the treatment process efficiency reached the highest. Furthermore, the intermediate products of the reaction were straight-chain organic acids. The oxidative process led to the opening of the benzene ring, and the next step is the oxidative process of the long-chain acids, which cuts them into shorter-chain acids. In the final step, these acids are oxidized and mineralized completely to generate the products CO_2 and H_2O. The electric discharging process of the reaction in the solution led to the formation of organic acids that increased the electrical conductivity, as shown in Figure 3f.

Figure 4a shows the concentration of the 2,4,5-T contaminant compound that was degraded depending on the duration of the reaction. Figure 4b shows the dependence on the time between the scale of the 2,4,5-T concentration in solution compared to the initial concentration (C_0), which was presented as the formula $\ln(C/C_0)$. From the graph in Figure 4a, it can be clearly seen that a hyperbolic curve characterizes the following differential equation of the 2,4,5-T decomposition kinetic reaction

$$v = \ dC/dt = k(C_0 - C) \tag{14}$$

Figure 4b describes the differential equation

$$\ln(C/C_0) = kt \tag{15}$$

which has a straight line corresponding to the model of the pseudo-first-order kinetic for the 2,4,5-T decomposition

process by the current of the high-voltage DC with plasma appearance on the iron electrode

$$y = ax + b \tag{16}$$

For 2,4,5-T, the value of the slope coefficient was calculated as

$$a_{2,4,5-T} = -0.01007 \tag{17}$$

The value of the intercept coefficient was calculated as

$$b_{2,4,5-T} = -0.0844 \tag{18}$$

The value of R square correlation coefficient was calculated as

$$R^2_{2,4,5-T} = 0.974 \tag{19}$$

The reaction rate constant was calculated as

$$k_{2,4,5-T} = 0.010 \ \text{min}^{-1} \tag{20}$$

Research on the kinetics of the decomposition process of 2,4,5-T compounds showed that the process depended on the initial concentration. The process with concentrations of 30 and 50 mg L^{-1} showed that the reaction rate decreased at high concentrations, as shown in Figure 4c. The kinetics of the decomposition process of 2,4-D also depended on the solution conductivity. The change in the conductivity of the solution (pH 6) by adding NaCl salt to adjust the conductivity reached the values of 300, 600, and 900 μS cm^{-1}, respectively. The experimental results indicated that the higher the electrical conductivity, the slower the decomposition reaction rate of 2,4,5-T. The reason for this phenomenon is that the Cl$^-$ anion quenched and reduced the formation of free radicals OH$^\bullet$ by the following mechanism

$$OH^. + Cl^- \rightarrow Cl^. + OH^- \tag{21}$$

$$Cl^. + Cl^. \rightarrow Cl_2 \tag{22}$$

$$Cl_2 + H_2O \rightarrow HCl + HClO \tag{23}$$

$$2HClO + H_2O_2 \rightarrow 2Cl^- + 4O + 4H^+ \tag{24}$$
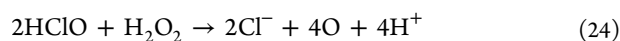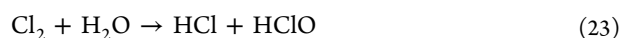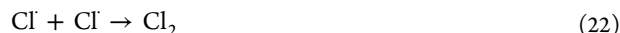
Some studies in the literature also mention the ability to inhibit the degradation in the presence of Cl$^-$ anions in a defined conductivity range. On the other hand, when the conductivity increases, the electric field decreases, affecting the ionization
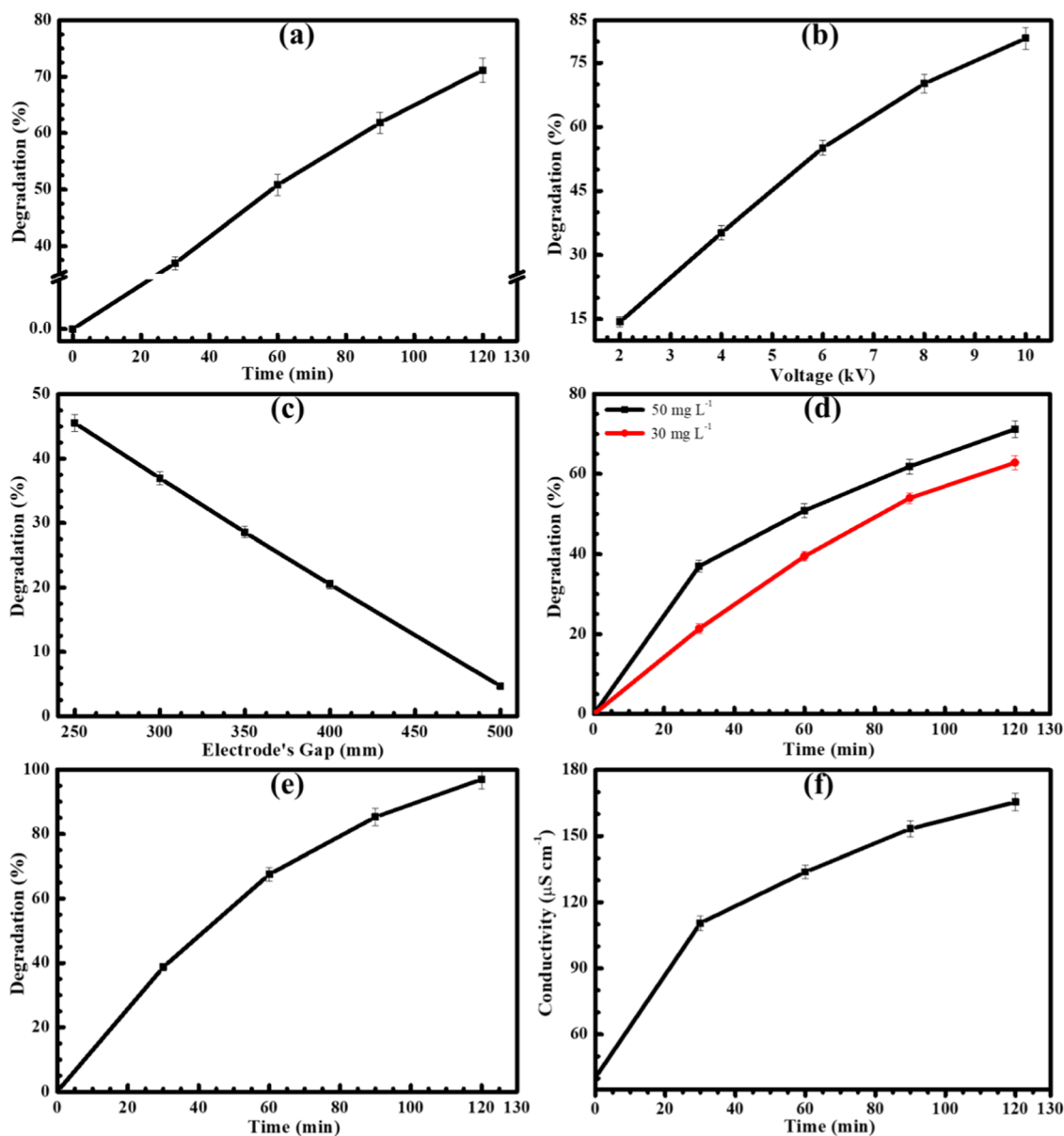
Figure 3. Effect of (a) reaction time and (b) voltage on the degradable efficiency of 2,4,5-T with an initial concentration of 30 mg L^{-1} and an electrode gap of 300 mm; (c) electrode distance and (d) initial concentration on the degradable efficiency of 2,4,5-T at 5 kV input voltage and electrode distance of 300 mm; and the flowing air through the solution on (e) 2,4,5-T degradable efficiency. (f) Change in the electrical conductivity of the treated solution with the initial pH of 6 and 38.5 μS cm^{-1} electrical conductivity.

state of the charged particles in the system. Due to the above reasons, the 2,4,5-T decomposition reaction rate decreases when the conductivity increases by the addition of NaCl salt, as described in Figure 4d. The pH values of 3, 6, 9, and 11 corresponded to the electric conductivity of 1.19 mS cm^{-1}, 38.5, 45.1, and 73.2 μS cm^{-1}. Research of the decomposition kinetics of 2,4,5-T at the initial pH of 3, 6, 9, and 11 showed that the decomposition rate of 2,4,5-T changed significantly depending on the pH value. The decomposition rate at pH 6 was faster than that at pH 3, as shown in Figure 4e. The cause of this phenomenon is that the concentration of H$^+$ ions is high in a low-pH environment, leading to the recombination between the free radicals OH$^\bullet$ and the H$^+$ ions, which occurred according to the reaction

$$OH^- + H^+ + e \rightarrow H_2O \tag{25}$$

or the reaction

$$OH^- + SO_4^{2-} + H^+ \rightarrow SO_4^{-\bullet} + H_2O \tag{26}$$
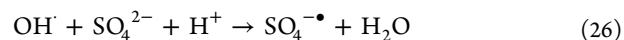
The oxidation potential of SO$_4^{-\bullet}$ free radicals was lower than that of the OH$^\bullet$ free radicals. For these reasons, the reduced density of OH$^\bullet$ free radicals leads to a decrease in the 2,4,5-T degradation efficiency when the pH value was low. In contrast, in the alkaline environment, the OH$^-$ ion content was high. The alkaline environment was more advantageous than the acidic environment for the decomposition of 2,4,5-T. However, at high pH, the ability to decompose 2,4,5-T also decreased due to the increasing electrical conductivity, which reduced the electric field for the ionization process. In addition, the discharge process in water produces H$_2$O$_2$ that in an alkaline environment, the ability to form OH$^\bullet$ free radicals is reduced, leading to the decrease of the degradation
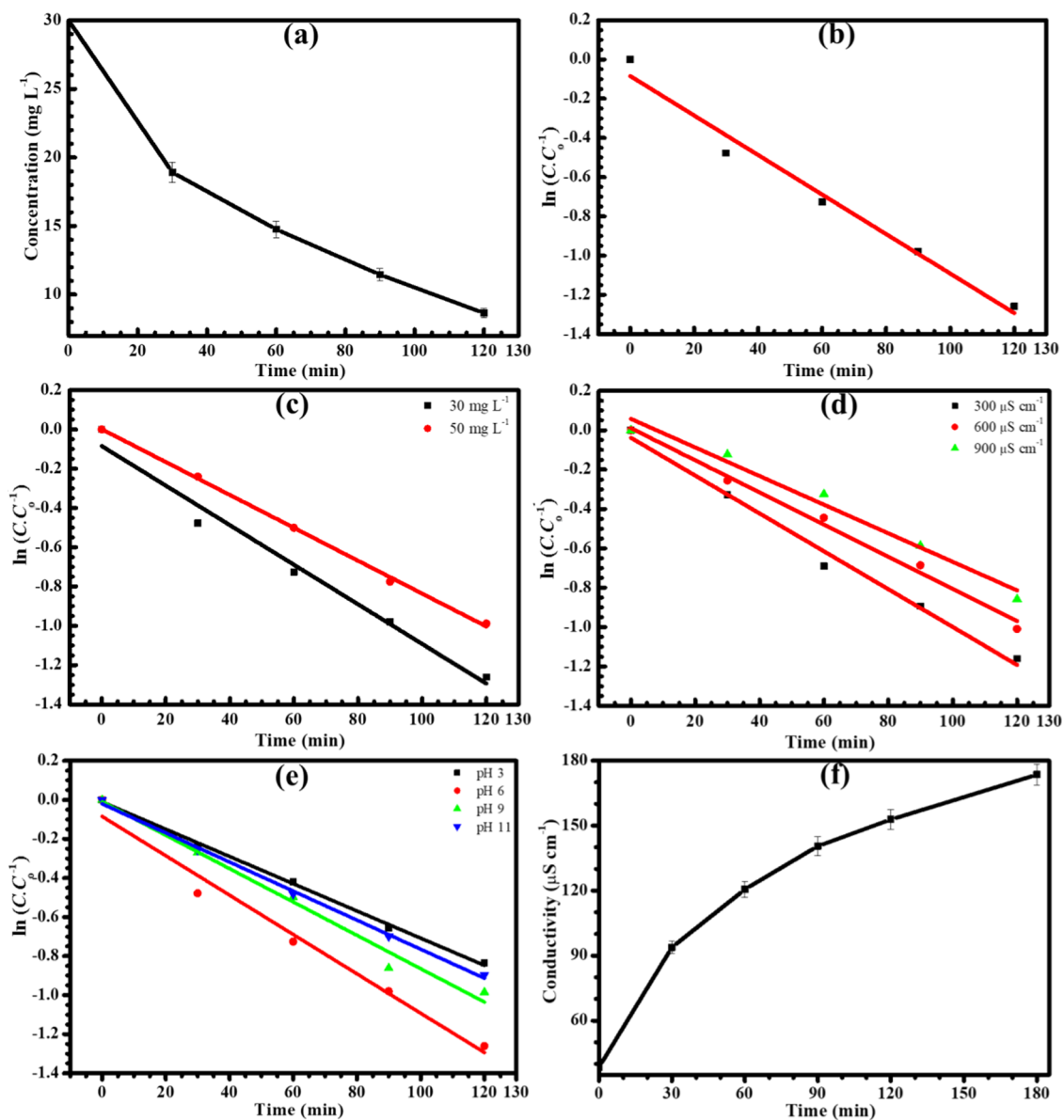
Figure 4. (a) Concentration of 2,4,5-T and (b) ratio $\ln(C/C_o)$ depend on the reaction time, input voltage of 5 kV, electrode gap of 300 mm, and at 30 °C. Influence of (c) initial concentration and (d) electrical conductivity on degradable efficiency of 2,4,5-T. (e) Influence of pH on the decomposition rate of 2,4,5-T. (f) Change in the electrical conductivity of the solution during the treatment process.

Figure 5. Change in (a) chemical oxygen demand (COD) and (b) total organic carbon (TOC) over time during the treatment process of 2,4,5-T.

efficiency. The research results also showed that the decomposition rate at pH of 9 was greater than at pH of 11. The decomposition rate in the alkaline environment pH of 9 was greater than in the acidic environment pH of 3. The discharge process in water also decreased the pH and increased the conductivity due to the plasma species produced in water. The cause for this change was the formation of H^+ ions according to the reaction

$$H_2O \rightarrow OH^. + H^+ + e- \tag{27}$$

In addition, the decomposition process of the organic compound of 2,4,5-T changed and increased the electrical conductivity in the solution because there was the formation of organic acids, as given in Figure 4f.

The COD index reflected the milligrams of oxygen required to oxidize the organic substances in the solution. The high COD index indicated that there were large amounts of organic compounds in the solution. In other words, the COD index reflected the degree of conversion and the decomposition of organic compounds in solution. The COD value also reflected the pollution level of the 2,4,5-T compound after the treatment process. The COD value before treatment of 2,4,5-T was 57 mg L^{-1}. After the treatment time of 30, 60, 90, and 120 min, the COD value of the 2,4,5-T solution decreased to only 30.2, 12.4, 8.9, and 4.1 mg.L^{-1}, respectively, as shown in Figure 5a. Additionally, the TOC measurement method allowed us to determine the level of carbon conversion in solution to CO_2. This method was more accurate than the COD method. The analysis results showed that the organic carbon content after 120 min decreased to 60.8% for 2,4,5-T, as shown in Figure 5b.

The 2,4,5-T pollutant decomposition reaction was performed under the following conditions: 2,4,5-T concentration of 30 mg L^{-1}; electrode distance of 300 mm, and an input voltage of 5 kV. The intermediate products after the decomposition process are extracted and transformed into a volatile compound form by silylating with BSTFA. After that, the GC−MS analysis was performed. The analyzed results before and after 120 min of 5 kV input voltage with an electrode gap of 300 mm, reaction temperature of 30 °C, and flowing air through the solution are given in Figure S1. Figure S2 presents the GC−MS results after 120 min of degradation of 5 kV input voltage, an electrode gap of 300 mm, a reaction temperature of 30 °C, and application with flowing air through the solution. Furthermore, the high-performance liquid chromatography results of the 2,4,5-trichlorophenoxyacetic (2,4,5-T) compound (30 mg L^{-1}) before and after 120 min of 5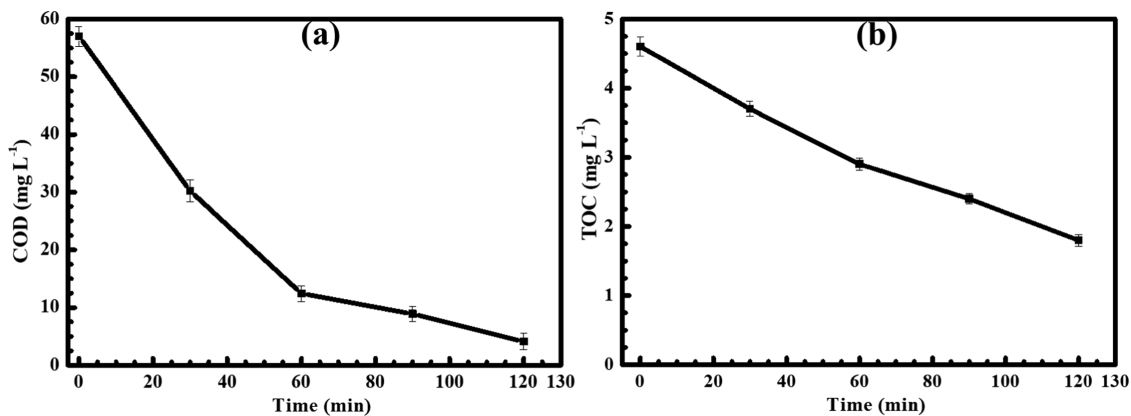 kV input voltage, electrode gap of 300 mm, reaction temperature of 30 °C, and flowing air through solution are also given in Figure S3. The analytical results showed that 2,4,5-T decomposed into intermediate organic compounds, as 2,4,5-TCP, 2,4-DCP, 3,4-DCP, and 2,3-DCP. These chlorophenol compounds are oxidized by OH• free radicals to reduce the chlorine atom in the aromatic ring and continue to create hydroquinone. Hydroquinone is oxidized, and the ring opens into straight-chain organic acids. These organic acids have different chain lengths and can be oxidized to carboxylic acid at one or both ends of the carbon chain. Hexanoic acid has the longest carbon chain, and formic acid has the shortest carbon chain. Finally, these organic acids are oxidized and mineralized to carbon dioxide and water. From the analytical results mentioned above, it can be assumed that the decomposition process of 2,4,5-T is according to the following mechanism in Scheme 1. The comparison of the decomposition ability of

Scheme 1. Decomposition Process of 2,4,5-Trichlorophenoxyacetic Acid

2,4,5-T with some other results shows that the decomposition mechanism and the formation of intermediate compounds are carried out by OH• free radicals, and the degradation efficiency was almost the same according to the 2,4-D and phenol degradation efficiency.[25,34] Moreover, the high-voltage DC electrochemical engineering with plasma appearance on the iron electrode showed that the degradation ability of 2,4,5-T at the initial concentration of 30 mg L^{-1} achieved after 120 min was 71.17%, which was better than that of phenol at the initial concentration of 40 mg L^{-1} with 46% degradation in the same time of 120 min by pulsed high-voltage discharge technology.[18] In addition, the decomposition ability of 2,6-dichlorophenol at the initial concentration of 20 mg L^{-1} reached 95% by coaxial

dielectric barrier discharge technology.[16] The TOC value that showed mineralization ability of 2,4,5-T achieved after 120 min was 60.8% compared with the mineralization ability of azo dyes at the initial concentration of 24 mg L^{-1} by bipolar pulsed barrier discharge technology (15%).[35,36] Therefore, it can be seen that the high-voltage DC electrochemical engineering with plasma appearance on the iron electrode could be a good candidate for high degradation efficiency.

CONCLUSIONS

The high-voltage electric discharge process occurs on the iron electrode that was immersed in an aqueous solution, causing plasma formation and creating free radicals. In particular, the OH$^\bullet$ free radicals with a high oxidation potential caused an oxidative and degradable process of the 2,4,5-T pollutant organic compound. The oxidation process depended on several factors, including the electric discharge time, input voltage, initial concentration, distance between two electrodes, and the process of flowing air through the solution. The experimental results also showed that the treatment ability of 2,4,5-T reached up to 99.83% when discharging electric with plasma appearance and combining it with the process of flowing air through the solution. The kinetic 2,4,5-T oxidative reaction was determined to obey the model of the pseudo-first-order with the reaction rate constant $k_{2,4,5-T} = 0.010$ min^{-1}.

The analysis of the intermediate products by GC–MS showed that the degradable reaction and oxidative process of 2,4,5-T were performed by OH$^\bullet$ free radicals. The intermediate products were straight-chain organic acids, which increased the electrical conductivity of the reaction solution. The oxidation process always occurred, leading to the cutting of long-chain organic acids into short-chain organic acids until the final product was carbon dioxide and water. Therefore, it can be seen that the high-voltage electrochemical reaction with the electric discharge and the combination of using an iron electrode with plasma on the electrode could almost completely decompose the 2,4,5-T pollutant compound in a water environment, which was difficult to treat by other methods.

ASSOCIATED CONTENT

ⓈI Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c03487.

Information about the gas chromatography–mass spectrometry result of the 2,4,5-trichlorophenoxyacetic (2,4,5-T) compound (30 mg L^{-1}) before and after 120 min of 5 kV input voltage, electrode gap of 300 mm, reaction temperature of 30 °C, application with flowing air through the solution; gas chromatography–mass spectrometry result of the 2,4,5-trichlorophenoxyacetic (2,4,5-T) compound after 120 min of degradation of 5 kV input voltage, electrode gap of 300 mm, reaction temperature of 30 °C, and application with flowing air through the solution; and high-performance liquid chromatography results of the 2,4,5-trichlorophenoxyacetic (2,4,5-T) compound (30 mg L^{-1}) before and after 120 min of 5 kV input voltage, electrode gap of 300 mm, reaction temperature of 30 °C, and application with flowing air through the solution (PDF)

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Notes

The authors declare no competing financial interest.

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