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Environmentally friendly technology for treatment of pesticide and ammonia contaminated water with electrochemical plasma

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Abstract. Electrochemical plasma is formed by an electrochemical reaction with a high voltage direct current (DC) in a non-electrolyte solution such as distilled water to create a gaseous state on the electrodes. Under conditions of high voltage, strong electric field and suitable temperature rise due to the Joule-Lenz effect, an electrochemical plasma will appear and be noted by the glow on the electrode as well as the characteristic amperage of the reaction rate leading to a rapid increase in the electrochemical reaction. Plasma reactions produce substances with strong chemical reactivity such as H_2, H_2O_2, or OH* free radicals, etc. in the aquatic environment. Electrochemical plasma formation can be controlled by technological parameters such as voltage, the distance between electrodes, conductivity, and the metallic nature of electrodes, etc.

Using electrochemical plasma, it is possible to treat pollutants harmful to the aquatic environment such as 2,4-D; 2,4,5-T, and ammonia. In the process of treating polluted water by electrochemical plasma technology, chemicals and treatment materials are not used, so it is an environmentally friendly technology.

Keywords: electrochemical plasma, DC high voltage, free radicals, hazardous pollutants, environmentally friendly treatment.

Classification numbers: 2.4, 2.4.2, 3.7, 3.7.3.

1. INTRODUCTION

Water is the environment associated with the life and development of all things and people. In the process of living and producing, many substances are released into the water environment, including toxic substances that pollute the water environment such as pesticides and ammonia. They are durable toxins that are difficult to decompose, so they not only persist for a long time in the environment but also easily accumulate in the body, causing many dangerous diseases such as cancer.

Therefore, many methods of treating water pollutants by chemical, physical or biological technologies have been studied and applied to contribute to ensuring a friendly water environment for humans. Although each method has its advantages, each method also has limitations that affect its effectiveness and applicability, such as the use of materials and chemicals for treatment, which easily creates a polluting process, and secondary or improper treatment to form intermediate products that are also toxic or require a lot of time or a large area. To overcome the limitations stated, in recent years many studies have focused on using the plasma state, the fourth state of matter, which is an ionized environment with high reactive activation to treat environmental pollutants.

Unlike thermal plasma, which occurs at extremely high temperatures mainly in the universe, cold plasma with low temperature is interested in research and application in various fields of life. Electrochemical plasma can be generated by electrochemical reaction at normal temperature in non-electrolyte medium such as distilled water by high voltage direct current (DC) [1 - 3].

At the anodic electrode will occur a metal dissolution reaction or oxygen release:

$$Me \rightarrow Me^{n+} + ne \tag{1}$$

or

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

and on the cathodic electrode will form hydrogen gas:

$$2H^+ + 2e \rightarrow H_2\uparrow \tag{3}$$

or

$$2H_2O + 2e \rightarrow H_2\uparrow + 2OH^- . \tag{4}$$

On the other hand, water has a large resistance, so when there is a high voltage DC, the electricity will generate heat energy according to the Joule-Lenz law and the solution will gradually heat up: [4, 5]

$$Q = I^2Rt = UIt \tag{5}$$

The H_2 and O_2 gaseous environment generated by the electrochemical reactions on the electrodes with the power supply of high magnetic and electric fields along with increasing temperature will create favorable conditions for the occurrence of the gaseous plasma state [6]. Thus, the plasma formed by an electrochemical reaction at high voltage can be considered an electrochemical plasma, which is different from the plasma caused by arcing in a solution or on the surface of a gas in contact with the solution.

In the electrochemical plasma state, water will dissociate strongly on the anodic and cathodic electrodes according to the following reactions, respectively:

$$2H_2O \rightarrow 2H_2 + O_2\uparrow \tag{6}$$

or

$$6H_2O \rightarrow 4H_2\uparrow + O_2\uparrow + 2H_2O_2. \tag{7}$$

The amount of gas produced by the plasma dissociation of water according to (6) and (7) is more than the water electrolysis that is calculated by Faraday's law according to (2) and (4). That will further maintain and develop the plasma state on the electrodes.

At the plasma state - the ionized state of matter, there will be atoms, molecules, and electrons at the same time - so the ionization reaction processes will continue [8 - 10] according to the following reactions:

$$H_2O + e^- \rightarrow H^* + OH^* + e^- \tag{8}$$

$$OH^* + OH^* \rightarrow H_2O_2 \tag{9}$$

$$2H_2O \rightarrow H_2O_2 + H_2\uparrow \tag{10}$$

The energy released from the glowing plasma (hv) also contributes to the radical formation reaction:

$$H_2O_2 + hv \rightarrow 2OH^* \tag{11}$$

Thus, the electrochemical plasma has created an environment with many strong reactive agents such as newly born atoms, H_2O_2 and especially OH* radicals with high oxidation potential values up to 1.8 V and 2.85 V, respectively [11 - 12] but has a very short lifetime $\approx 10^{-9}$s [13]. Therefore, it is necessary to catalyze the regular generation of OH* radicals. When using the Fe anodic electrode, the electrochemical reaction will generate Fe^{2+} ions and the plasma reaction will generate H_2O_2, so the ability to convert H_2O_2 into OH* radicals will be realized regularly according to the Fenton reaction [14]:

$$Fe^{2+} + H_2O_2 \rightarrow OH^* + OH^- + Fe^{3+} \tag{12}$$

$$Fe^{3+} + H_2O_2 \rightarrow H^+ + Fe^{2+} + 2OH^*. \tag{13}$$

2. EXPERIMENTAL METHODS

2.1. Reactor system

The high voltage electrochemical reactor system is presented in Figure 1, consisting of a 0 - 20 kV high voltage source, a double-layer glass reactor with Fe electrodes [2].

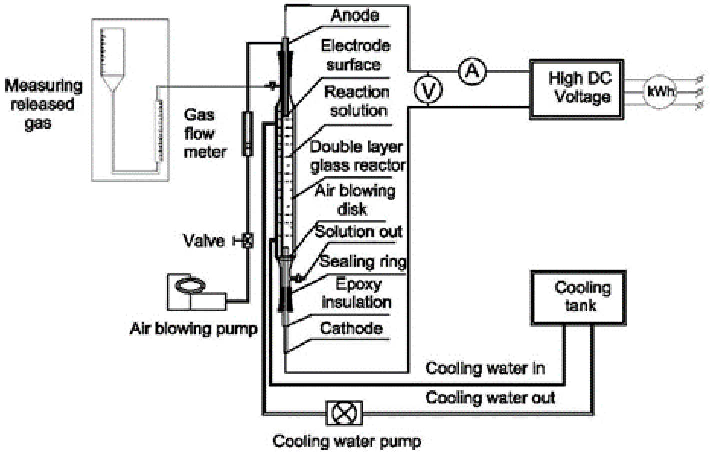


Figure 1. Diagram of electrochemical plasma generator system on iron electrode for wastewater treatment [2].

With this system, it is possible to operate and control the plasma generation process on the electrodes using various high voltage values, the distance between the anodic and cathodic electrodes as well as the reaction cooling rate [15]. The plasma appearance is recorded by a camera with the glow on the electrodes, concurrently by a strong increase in the amperage of the electrochemical reaction.

2.2. Analytical methods

The formation of H_2O_2 during plasma generation is determined by the reaction with titanyl ions to give a yellow complex [16] with the maximum absorption wavelength and is measured by UV-Vis spectroscopy.

$$Ti^{4+} + H_2O_2 + 2H_2O \rightarrow TiO_2.H_2O_2 + 4H^+ \tag{14}$$

Calibration curves were prepared with pure H_2O_2 from Sigma Aldrich at concentrations of 0.005 mg/L; 0.01 mg/L; 0.025 mg/L; 0.05 mg/L, followed by determining again by titration with $KMnO_4$ in H_2SO_4 solution. The optical density of the UV-Vis spectrum with the maximum absorption wavelength λ_{max} of 407 nm was determined on the device UH-5300, HITACHI at the Institute of Environmental Technology, Vietnam Academy of Sciences and Technology.

OH* free radicals generated in the plasma were determined by salicylic acid ($C_7H_6O_3$) to form 2,3-dihydroxybenzoic acids ($C_7H_6O_4$ - 2,3-DHB) and 2,5-dihydroxybenzoic acids ($C_7H_6O_4$ - 2, 5-DHB) [17]:

$$(HO)C_6H_4CO_2H + OH^* \begin{array}{l} \nearrow 2,3\text{-}(HO)_2C_6H_3CO_2H \\ \searrow 2,5\text{-}(HO)_2C_6H_3CO_2H \end{array} \qquad (15)$$

The total amount of products hydroxylated to 2,3-DHB and 2,5-DHB is equivalent to the number of OH* free radicals generated. The quantitative determination of these acids by UV-Vis molecular absorption spectroscopy [18] follows the principle of measuring the total absorbance (Abs_Σ) of the components from the absorbances of the individual compounds: salicylic acid (SA) with 2,3-DHB acid and 2,5-DHB acid:

$$Abs_\Sigma = Abs_{SA} + Abs_{2,3\text{-DHB}} + Abs_{2,5\text{-DHB}}. \qquad (16)$$

By measuring the absorbance of the solution at different wavelengths (Table 1), it is possible to determine the concentration of substances present in the solution, which is the quantification of OH* free radicals.

Table 1. Molecular absorption coefficients of SA, 2,3-DHB and 2,5-DHB.

λ, nm	**Molecular absorbance of the substance, ε, $M^{-1}cm^{-1}$**		
	SA	2,3-DHB	2,5-DHB
290	2434.70	1316.13	635.14
310	2591.04	2706.30	2402.49
330	216.97	1694.40	3238.56

Calibration curves were built over a linear range with salicylic acid and 2,3-DHB, 2,5-DHB acids from Sigma Aldrich with a purity of 99 %. Based on the change in maximum absorption wavelengths for salicylic acid at 290 - 303 nm, 2,3-DHB is 310 nm, 2.5-DHB is 329 - 330 nm on a UV-molecular absorption spectrophotometer (Vis UH-5300, HITACHI) at the Institute of Environmental Technology, Vietnam Academy of Science and Technology.

The degradation products of 2,4-D and 2,4,5-T upon electrochemical plasma treatment were determined by gas chromatography mass spectrometry (GC-MS). The reaction solution was extracted with diethyl ether solvent and converted to volatile derivatives with BSTFA at 70 ^oC for 30 minutes. The analysis was performed under the following conditions: HP-5MS column, sample injection volume 1 µL, scanning mode 35 - 550 amu, He gas flow 1.3 mL/min, He gas flow rate 41 cm/s, EI 70 keV, program temperature 40 ^oC, an increase of 10 ^oC/min to 280 ^oC, hold for 5 minutes. The intermediates were analyzed by static analysis method, through the NIST standard spectrum library it is possible to find the identification of these substances. The analytical instrument used is a gas chromatograph coupled to mass spectrometry (GC-MS 6890-5975, Agilent) at the Institute of New Technology, Military Institute of Science and Technology. The amount of CO_2 formed by the mineralization process when treating 2,4-D and 2,4,5-T by electrochemical plasma is determined by the weight of precipitated $CaCO_3$ [19]:

$$CO_2 + Ca\ OH)_2 \rightarrow CaCO_3\downarrow + H_2O \qquad (19)$$

Ammonium concentration was determined according to Vietnam Standard 6179-1:1996 [22], using a UV-Vis V-730 spectrometer from Japan's Jasco at 659 nm. Calibration curves were constructed with ammonium concentrations of 0.05; 0.1; 0.2; 0.5; 0.6; 0.8 and 1 mg/L, giving a linear function between ammonium concentration (c_{am}) and spectral height (h_{Abs}):

$$c_{am} = 1.0201 h_{Abs} - 0.0001 \text{ with } R^2 = 0.9968. \qquad (20)$$

3. RESULTS AND DISCUSSION

3.1. Generation of electrochemical plasma

When performing the electrochemical reaction at high voltage DC, the anodic dissolution reaction will occur and H2 will be drained on the cathode as reported in [20]. With the reaction time, the ambient temperature gradually increased and glowing plasma appeared on the electrodes corresponding to the increased reaction current. Figure 2 presents the electrochemical reactions of cavitation (Figure 2a), the appearance of yellow-orange luminescence at the cathode (lower Figure 2b) and blue (upper Figure 2b), and the dependence of the reaction current on time at different voltages (Figure 2c). The distances between the anode and cathode are different (Figure 2d) and the initial temperature of the circulating cooling water is different (Figure 2e).

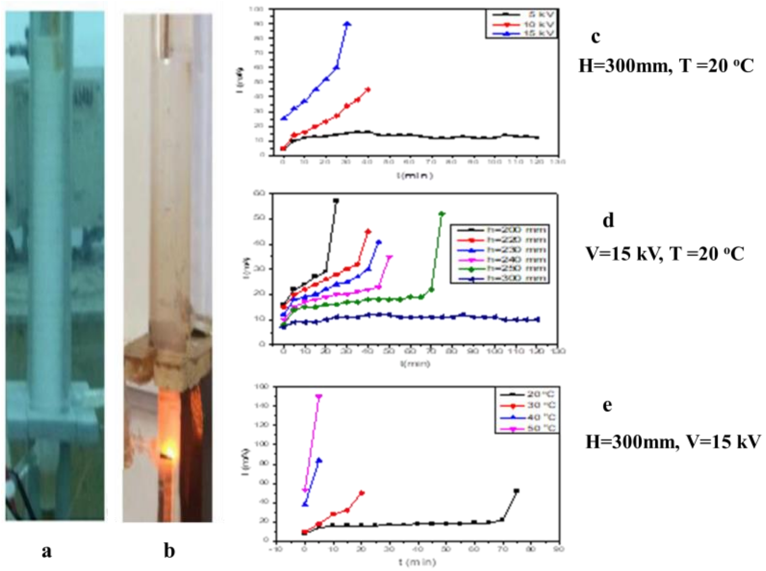


Figure 2. Gas-generating high-voltage DC reaction (a), plasma generation (b) and *I-t* curves at different voltages (c), anode-cathode distance (d) as well as ambient temperature (e).

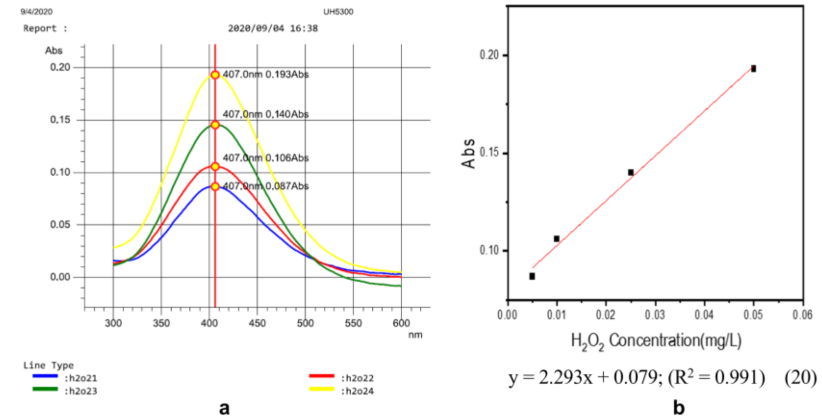
From Figures 2c, d and e it can be observed that the electrochemical plasma appearance was faster when applying higher voltage for the electrochemical reaction, the shorter distance between the anode and the cathode and the initial temperature of the water accelerated the time to reach the plasma appearance temperature as soon as possible. With a voltage of 5 kV, the distance between the cathodic and anodic electrodes of 300 mm and the initial temperature of the cooling water of 20 ^{o}C, the reaction time of more than 70 minutes still does not appear in electrochemical plasma. Thus, the electrochemical plasma appearance time can be controlled and is controlled by adjusting the following parameters: voltage, the distance between the anodic and cathodic electrodes and the initial temperature of the cooling water.

3.2. Formation of OH* radical

When the plasma state is formed on the Fe electrode according to reactions (7) and (10), H_2O_2 (hydrogen peroxide) will be generated and determined by UV-Vis spectroscopy at the maximum absorption wavelength λ_{max} of 407 nm with the standard curve as shown in Figure 3, giving the results as presented in Table 2, which shows that the H_2O_2 content increases with increasing the lifetime of the electrochemical plasma on the electrode.

Table 2. H_2O_2 content formed with plasma lifetime on Fe electrode.

t, 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



$$y = 2.293x + 0.079; (R^2 = 0.991) \quad (20)$$

Figure 3. UV-Vis absorbance spectrum of $TiO_2.H_2O_2$ complex (a) and H_2O_2 standard curve (b).

Also, in the plasma state on the Fe electrode [20], OH* free radicals were formed according to reactions (8), (11), (12), (13) which were determined through the content of 2,3-DHB and 2,5-DHB by UV-Vis spectroscopy, the maximum absorption wavelengths for salicylic acid was 290 nm, 2,3-DHB was 310 nm and 2,5-DHB was 330 nm (Figure 4). Calibration curves were built over a linear range with the concentrations of salicylic acid and 2,3-DHB in the range of 1×10^{-4} M to 1.0×10^{-3} M and with the 2,5-DHB acid concentration in the range of 6×10^{-5}M to 5×10^{-4}M.

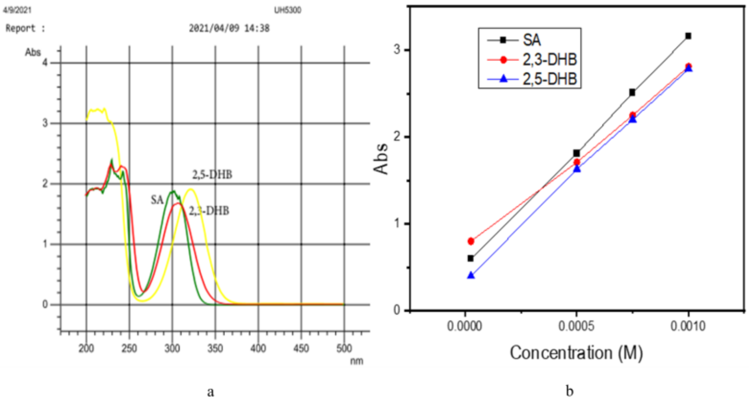


Figure 4. UV-Vis absorption spectrum (a) and calibration curves (b) of SA; 2,3-DHB; 2,5-DHB.

The standard curve of the optical absorbance of each concentration-dependent substance is determined by the following formula:

$$Abs_{290nm} = 2632.80[SA] \pm 33.42 + 0.52 \pm 0.02\ (R^2 = 0.999) \quad (21)$$

$$Abs_{310nm} = 2055.13[2,3\text{-}DHB] \pm 58.23 + 0.72 \pm 0.03\ (R^2 = 0.997) \quad (22)$$

$$Abs_{330nm} = 2599.10[2,5\text{-}DHB] \pm 115.99 + 0.23 \pm 0.07\ (R^2 = 0.994). \quad (23)$$

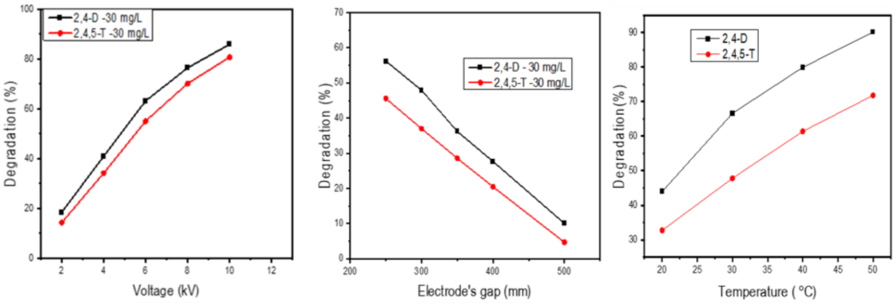
The results after solving the equation to determine the concentration of substances in the solution mixture through measuring the optical absorbance at the above wavelengths showed that the 2,3-DHB acid concentration achieved after plasma formation was 2.1×10^{-4} M, while the obtained acid concentration of 2,5-DHB was 9.5×10^{-5} M. This indicated the ability to react with OH* radicals to form 2,3-DHB with a higher concentration compared to that of 2.5-DHB produced. Thus, the total concentration of OH* free radicals obtained after 30 and 60 minutes was 3.0×10^{-4} M and 3.7×10^{-4} M, respectively.

The above results have demonstrated that the electrochemical plasma generation process on the Fe electrode will create reactive substances, H_2O_2 and OH* free radicals. Together with the anodic dissolution of the reaction on the iron electrodes, Fenton reactions (11) and (12) will contribute to the conversion of H_2O_2 into OH* free radicals.

3.3. Environmental treatment efficiency of electrochemical plasma

Thus, when creating plasma on the Fe electrode, it will ensure the creation of an environment with high reactive agents such as H_2O_2 and OH* radicals. For this reason, it can be used to decompose pollutants that is difficult to degrade in the water environment such as 2,4-D and 2,4,5-T aromatic compounds.

Figure 5 presents the treatment efficiency of 2,4-D and 2,4,5-T at a concentration of 30 mg/L for 60 minutes on the Fe electrode with electrochemical plasma depending on voltage U (a), the distance between the anodic and the cathodic electrodes H (b), and the initial temperature of the cooling water T (c).



a: at H = 300 mm and T = 20 °C b: at U = 5 kV and T = 20 °C c: at U = 5 kV and H = 300 mm

Figure 5. Dependence of the treatment efficiency of 2,4-D and 2,4,5-T on a) voltage U, b) distance between anode and cathode H, and c) initial cooling water temperature T.

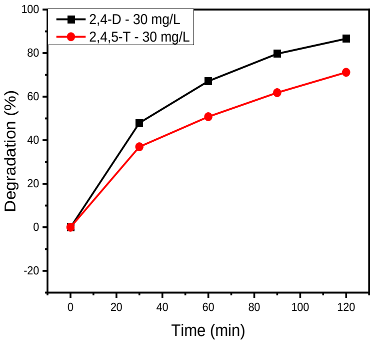


Figure 6. Dependence of the decomposition efficiency of 2,4-D and 2,4,5-T, concentration of 30 mg/L at U = 15 kV, H = 200 mm, T = 30 °C on the reaction time of electrochemical plasma.

It can be seen from Figure 5 that the treatment efficiency of 2,4-D and 2,4,5-T increased with increasing the reaction voltage as well as increasing the initial temperature of cooling water, but the treatment efficiency decreased when increasing the distance between the anodic and cathodic electrodes. These results showed the role of the electrochemical plasma appearance as well as the reaction time in the ionized environment of the electrochemical plasma. Figure 5 also shows that the treatment efficiency of 2,4-D was higher than that of 2,4,5-T, demonstrating that the reactivity of 2,4-D with plasma-derived substances as well as with free radicals was easier than that of 2,4,5-T.

Figure 6 shows the dependence of the treatment efficiency of 2,4-D and 2,4,5-T at the concentration of 30 mg/L on the Fe electrode at a voltage of 15 kV, electrode distance of 200 mm and initial cooling water temperature of 30 ^oC at the time of reaction. The results from Figure 6 showed that after 60 minutes, the processing efficiency of 2,4-D and 2,4,5-T was 67.13 % and 50.80 %, respectively; but when increasing to 120 minutes, the performance was 86.16 % and 71.17 %, respectively. The degradation of 2,4,5-T was always lower than 2.4-D.

The analysis of intermediate products formed during the decomposition process of 2,4-D and 2,4,5-T using GC-MS equipment at the beginning time showed the presence of acetic acid and aromatic compounds with ^-OH group such as phenol; 1-chlorobenzene; 2,3-dichlorophenol; 3,4-dichlorophenol; and 2,4-dichlorophenol. This proves that 2,4-D and 2,4,5-T have been oxidized by OH* free radicals to acetic acid and chlorophenols compounds. Analyzing at a longer time also found long-chain unsaturated organic acids including pentanoic acid; 2-butenoic acid; 2-propenoic acid, and short-chain organic acids such as acetic acid; formic acid; oxalic acid; succinic acid; propanoic acid; quinones and hydroxyquinones. This shows that OH* free radicals further oxidized chlorophenol compounds to hydroxyquinone and then hydroxyquinone is further oxidized by OH* free radicals to quinone, which continues to be oxidized, resulting in its ring being opened to straight-chain organic acids. [21] The organic acids experience further oxidation and are fragmented into shorter-chain organic acids. Eventually, these organic acids undergo complete oxidation to CO_2 and H_2O. The emission of CO_2 gas takes place during the electrochemical plasma treatment process. The weight of the emitted CO_2 was measured and is presented in Table 2. Therefore, it is evident that the electrochemical plasma environment facilitates the full conversion of 2,4-D and 2,4,5-T into CO_2 and H_2O through the oxidation process.

Table 3. Values of CO_2, COD and TOC content determined when treating 30 mg/L 2,4-D and 2,4,5-T by electrochemical plasma at U = 15 kV, H = 200 mm, T = 30 ^oC at t = 60 and 120 minutes.

***t*, min**	**60**		**120**	
	2,4-D	2,4,5-T	2,4-D	2,4,5-T
m_{CO_2}, mg/L	10.8	8.9	21.5	17.6
H, %	26.7	24.1	59.8	55.4
COD, mg/L	15.9	12.4	2.1	4.1
H, %	85.0	78.2	98.0	92.8
TOC, mg/L	3.7	2.9	2.1	1.8
H, %	39.3	36.9	65.6	60.8

Table 3 presents the amount of CO_2, COD and TOC determined from the treatment process of 2,4-D and 2,4,5-T at a concentration of 30 mg/L by electrochemical plasma at a voltage of 15 kV, the distance between anodic and cathodic electrodes was 200 mm and the initial cooling water temperature was 30 ^oC with the reaction time of 60 and 120 min. The results from Table 2 showed that with increasing the reaction time from 60 to 120 min, the CO_2 production increased nearly 2 times while the COD decreased approximately 8 times with 2,4-D and 3 times with 2,4,5- T. Thus, the number of organic pollutants via the COD parameter was greatly reduced and the efficiency achieved was very high, up to 98 % with 2,4-D and 92.8 % with 2,4,5-T. However, the treatment efficiency of carbon compounds through CO_2 and TOC parameters decreased less and only more than double when the treatment time increased from 60 minutes to 120 minutes. The treatment value in TOC for both 2,4-D and 2,4,5-T at 60 minutes was only 1.5 times higher than for CO_2 but at 120 minutes the difference was only 1.1 times. This proves that the longer the treatment time with electrochemical plasma, the more TOC is converted into CO_2.

Table 4 presents important parameters according to Vietnam standard 40:2021 issued by the Ministry of Natural Resources and Environment (MNRE) after treatment of 2,4-D and 2,4,5-T with electrochemical plasma as well as the power consumption for the treatment process.

Table 4. Values of pollution parameters after treatment of 2,4-D and 2,4,5-T solutions.

No	**Parameter**	**Value after treatment**		**Unit**	**Vietnam standard 40:2021/MNRE**	
		2,4-D	2,4,5-T		A	B
1	pH	5.7	5.5	-	6 - 9	5.5 - 9
2	COD	6.8	8.9	mg/L	75	150
3	Fe^{2+}	3.1	4.5	mg/L	1	5
4	Cl-	7.6	10.7	mg/L	500	1000
5	TSS	14.2	15.8	mg/L	50	100
6	2,4-D	< 0.005	-	mg/L	0.05	0.1
7	2,4,5-T	-	< 0.05	mg/L	0.05	0.1
8	Y, Energy	22.46	22.46	mg/kWh		

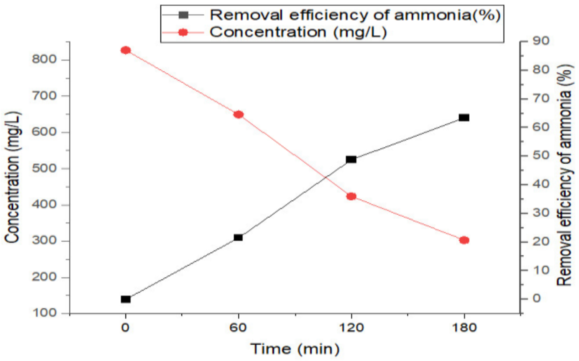


Figure 7. Dependence of ammonium concentration and efficiency of electrochemical plasma treatment with iron electrode at 0.5 kV on treatment time.

Figure 7 shows the results of ammonium treatment in leachate from Nam Son waste treatment plant (Phu Dien, Ha Noi).

The results presented in Table 3 demonstrate that pollutants such as 2,4-D and 2,4,5-T, which are challenging to treat using conventional methods, can achieve significant improvements when employing the electrochemical plasma method. Specifically, after treating 30 mg/L solutions of 2,4-D and 2,4,5-T for 120 minutes, the important parameters reached level A of Vietnam standard 40:2011/MNRE. Notably, this treatment process solely relies on electricity and does not involve the use of chemicals or materials. The electricity consumption for removing 22.46 mg of pollutants is merely 1 kWh. Therefore, the plasma method can be regarded as an environmentally friendly approach for treating pollution.

Based on Figure 7, it is evident that the ammonium content in the leachate decreased from 827 mg/L to 303 mg/L after a 3-hour electrochemical plasma treatment using an iron electrode at 0.5 kV. This treatment resulted in an improved treatment efficiency of over 60 %, which is comparable to the reported effectiveness of oxidizing agents by Xianping Luo *et al*. [23].

4. CONCLUSIONS

In a non-electrolyte environment such as distilled water, the electrochemical reaction still occurs when using high voltage DC with the anode process: $Me \rightarrow Me^{n+} + ne$ and $2H_2O \rightarrow O_2 + 4H^+$, simultaneously with the cathode process: $2H_2O \rightarrow H_2 + 2OH^-$. The amount of H_2 and O_2 gas produced by the electrochemical reaction on the electrodes in a strong electric and magnetic field along with the gradual increase in temperature to an appropriate value will cause plasma appearance, the ionized state of material. The feature of the electrochemical plasma is the glow on the electrodes and the characteristic electric currents, which represent a spike in the rate of the electrochemical reaction.

In the electrochemical plasma state, water will be strongly electrolyzed on the cathode: $2H_2O \rightarrow 2H_2 + O_2$ and on the anode: $6H_2O \rightarrow 4H_2 + H_2O_2 + O_2$ producing many times more gas than calculated by Faraday's equation. Besides, H_2O_2 was determined to increase gradually with electrochemical plasma generation over time, reaching 0.043 mg/L at 60 min, reactive free radicals such as OH* were also identified and reached the value of $3.7.10^{-4}$ M. The application of the Fe anodic electrode will contribute to the generation of more OH* free radicals due to the Fenton reaction.

Using an electrochemical plasma state with an ionized environment of H_2O_2 and OH* free radicals to treat water pollution with hazardous aromatic compounds in the structure such as 2,4-D or 2,4,5-T has been determined to be a strongly oxidized process to form long chain organic acids and finally to CO_2 and H_2O. The leachate treatment process of Nam Son waste treatment plant (Phu Dien, Ha Noi) also achieved an efficiency of more than 60 % after 3 hours. The treatment of hazardous waste with electrochemical plasma was highly efficient, without the use chemicals or materials. Therefore, it can be considered as an environmentally friendly technology.

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Declaration of competing interest: We declare that we have no competing interests related to the content of this article.

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