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Integration of ultrasound-electro-Fenton processes for total organic carbon and inorganic carbon removal in pharmaceutical wastewater

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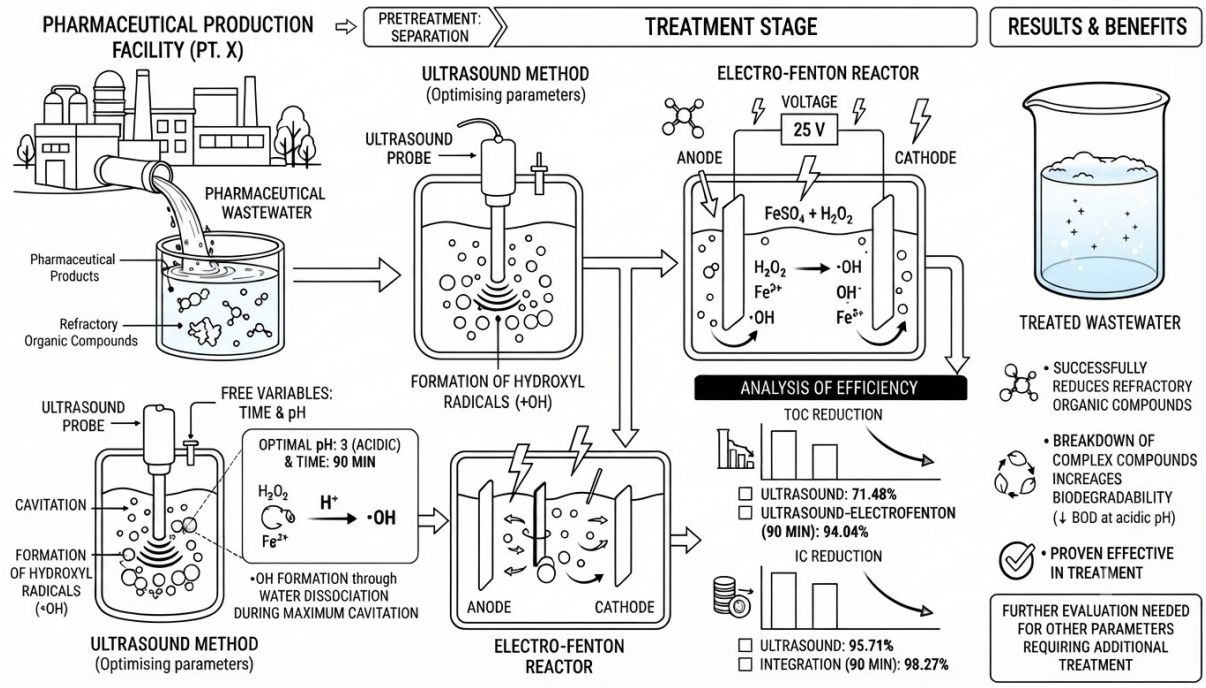
Abstract: Pharmaceutical are widely used by humans and are commonly discharged into the environment as waste after use. Pharmaceutical wastewater contains organic compounds that can be removed through the integration of ultrasound and electro-Fenton methods. This study aims to analyse the removal efficiency of total organic carbon (TOC) and inorganic carbon (IC) in pharmaceutical wastewater using an integrated ultrasound-electro-Fenton process. Pharmaceutical wastewater obtained from PT. X was used for laboratory experiments. The independent variables in this study consisted of treatment time and pH in the ultrasound process. The ultrasound–electro-Fenton process was conducted using variations in treatment time, $FeSO_4$ + H_2O_2 dosage, and a voltage of 25 V. The optimal treatment condition was achieved at 90 minutes of ultrasound exposure and pH 3 under acidic conditions. At acidic pH, hydroxyl radicals (•OH) are formed through water dissociation during maximum cavitation, resulting in a TOC reduction of 71.48% and an IC reduction of 95.71%. The integrated process successfully reduced TOC by 94.04%. IC was also reduced by 98.27% after 90 minutes of treatment. These results demonstrate that the integrated ultrasound–electro-Fenton process is effective in reducing refractory organic compounds in pharmaceutical wastewater. The degradation of complex compounds increased biodegradability, as indicated by a decrease in biochemical oxygen demand (BOD) under

acidic conditions. Further evaluation is needed for other parameters that may require additional treatment.

Keywords: biological oxygen demand (BOD), electro-Fenton process, inorganic carbon (IC), pharmaceutical wastewater, total organic carbon (TOC), ultrasound, ultrasound–electro-Fenton process.

Graphical abstract



1. INTRODUCTION

Pharmaceutical and personal care products (PPCPs) are an emerging environmental concern because they include various classes of chemicals, such as drugs and personal care products. PPCPs used by humans are generally discharged into the environment after use and eventually become waste; therefore, their presence in aquatic systems has become a major concern (Kim *et al*., 2022). The chemicals used in PPCPs are biologically active compounds designed to interact with specific mechanisms in the body. As a result, concerns have arisen regarding the potential impacts of active PPCPs in the environment on human health and ecosystems (Nazhifah, 2023). Several water quality parameters are affected by chemical characteristics, including organic substance parameters such as Total Organic Carbon (TOC), Chemical Oxygen Demand (COD), Biochemical Oxygen Demand (BOD), pH, and turbidity.

Pharmaceutical wastewater contains high concentrations of refractory organic compounds, with TOC concentrations ranging from 1,000 to 23,000 mg·dm^{-3} (Garcia-Rodriguez *et al*.,

2024). The main sources of TOC include residual active pharmaceutical ingredients (APIs), synthesis intermediates, and solvents (Anliker *et al*., 2020). High TOC concentrations can inhibit *Vibrio fischeri* luminescence and sludge respiration (Zhan *et al*., 2019).

In recent years, oxidation reactions have become a promising option for the treatment of organic compounds, one of which is the ultrasonic process. Ultrasound is based on the principle of ultrasonic irradiation, in which water is exposed to high-frequency waves, inducing the thermal decomposition of H_2O and generating radicals that oxidise and degrade dissolved contaminants (An *et a*l., 2024). The ultrasonic process has attracted significant attention due to its many advantages, including the production of •OH radicals and other oxygenated radicals, increased chemical reaction rates, ease of operation, reduction in solid particle size, minimal consumption of chemical reagents, and reduced sewage sludge formation (Anandan, Vinoth Kumar and Ashokkumar., 2020; Hassani *et al*., 2025). Ultrasound and Electro-Fenton processes are both included in Advanced Oxidation Process (AOP) technologies, which are currently widely used for wastewater treatment. The ultrasound process uses ultrasonic irradiation to induce the sonolysis of water, producing •H and •OH radicals. Meanwhile, the Electro-Fenton process involves the application of electrical voltage and the reaction between Fe^{2+} and H_2O_2. In both ultrasound and Electro-Fenton processes, several operational variables can be adjusted to achieve optimal pollutant removal efficiency.

In recent years, many studies have shown that AOPs are effective in degrading persistent and complex pollutant compounds. Hassani *et al*. (2025) conducted a detailed review of the integration of ultrasound with oxidants, including ultrasound-persulphate, ultrasound-hydrogen peroxide, ultrasound-ozone, ultrasound-chlorine, ultrasound-permanganate, ultrasound-periodate, and ultrasound-peracetic acid, for the removal of various organic pollutants. Pharmaceutical liquid residues, particularly diphenhydramine (DPH), have also been successfully degraded and mineralised using an integrated Electro-Fenton process with a Cu/C catalyst (Zhao *et al*., 2024). This pre-treatment allows wastewater to be treated more easily using biological processes. The removal of TOC using ultrasound is influenced by several factors, such as ultrasound frequency and power, initial TOC concentration, pH, reaction time, and the addition of other agents. pH is a critical factor in TOC removal from pharmaceutical wastewater using ultrasound-based treatment methods (Ghafoori *et al*., 2015; Patidar and Srivastava, 2021; Wei *et al*., 2016). Based on previous studies, research on the integration of ultrasound and Electro-Fenton processes for the simultaneous reduction of TOC

and Inorganic Carbon (IC) in pharmaceutical wastewater remains limited. This integrated approach is expected to enhance pollutant removal efficiency. Although previous studies have extensively explored ultrasound (US), Electro-Fenton (EF), and their hybrid systems for wastewater treatment, this study focuses on the simultaneous removal of TOC and IC from high-concentration pharmaceutical wastewater, a matrix that remains underexplored, particularly regarding IC behaviour during AOP treatment. Therefore, this study is positioned as a synergistic performance assessment and process validation using real pharmaceutical wastewater. This study aims to analyse the efficiency of TOC and IC removal in pharmaceutical wastewater using an integrated ultrasound–Electro-Fenton process.

2. MATERIALS AND METHODS

2.1 Study material

This research was conducted at the Environmental Laboratory, Department of Environmental Engineering, Universitas Diponegoro. Wastewater samples were obtained from one of the pharmaceutical industries, PT X, located in Semarang, Central Java, Indonesia. In the initial stage of the study, pharmaceutical wastewater was treated using an ultrasound process to determine the optimal operating time and pH. In the next stage, the pharmaceutical wastewater was treated using the electro-Fenton process.

2.2 Ultrasound

In this study, the wastewater treatment process was carried out using ultrasonic wave technology with a Digital Ultrasonic Cleaner (DIGITAL PRO, China). The device is designed to produce ultrasonic waves at a frequency of 40 kHz, which accelerate the degradation of contaminants in wastewater through an acoustic cavitation mechanism. This device has a capacity of 2 L and is equipped with two digital control panels: a temperature panel (°C) to monitor the wastewater temperature during the process and a time panel (minutes and seconds) to set the duration of ultrasonic exposure. The ultrasound device was calibrated before being used in the experiment. An experimental method was applied in this study to determine the optimal treatment time and pH. The characteristics of the wastewater are presented in **Table 1**.

Table 1. Characteristics of pharmaceutical wastewater used for laboratory experiments.

No.	Parameter	Concentration
1	pH	4.4
2	Conductivity	686 µS/cm
3	BOD	443.8 $mg \cdot dm^{-3}$
4	COD	4,551.67 $mg \cdot dm^{-3}$
5	TSS	91.5 $mg \cdot dm^{-3}$
6	Turbidity	468 NTU
7	Ammonia	0.096 $mg \cdot dm^{-3}$
8	Phosphate	0.658 $mg \cdot dm^{-3}$
9	Sulphate	59.2 $mg \cdot dm^{-3}$
10	Fe	0.01 $mg \cdot dm^{-3}$
11	TOC	2,841 $mg \cdot dm^{-3}$
12	IC	19.8 $mg \cdot dm^{-3}$

2.3 Experimental Setup

One liter of pharmaceutical wastewater was placed into the ultrasonic cleaner. In this study, four variations of ultrasound exposure time were used: 30, 60, 90, and 120 min. After the optimal exposure time was obtained, five pH variations were tested, namely pH 3, pH 5, pH 7, pH 9, and pH 12, to determine the optimal condition for the ultrasound method. A total of 500 cm^3 of sample was collected for parameter analysis in the laboratory. The optimal ultrasound condition was then applied in the Electro-Fenton stage. One liter of pharmaceutical wastewater that had been treated with ultrasound was further treated using the Electro-Fenton process by adding 15 g of $FeSO_4$ and 100 cm^3 of H_2O_2 to determine the optimal conditions for the Electro-Fenton method. This condition was based on previous research. The cathode and anode were immersed in the pharmaceutical wastewater and then connected to an AC power source. The voltage was maintained at 25 V, while the current was not adjusted. The Electro-Fenton process was carried out using three time variations: 30, 60, and 90 min. A total of 500 cm^3 of sample was collected for parameter analysis in the laboratory.

2.4 Analysis

The tested parameters included TOC, IC, pH, conductivity, COD, BOD, Total Suspended Solids (TSS), turbidity, and Fe. All parameters were analysed according to the standard methods for wastewater analysis (APHA, 2017). Fe was analysed using an atomic absorption spectrophotometer (AAS) VGP Series 210 (Buch Scientific, USA). TOC and IC were

analysed using a TOC analyzer (Shimadzu, Japan). COD analysis was performed using COD digestion equipment (Velp) and a UV-Vis spectrophotometer (Thermo Fisher Scientific, USA). All analyses were conducted in duplicate.

3. RESULTS AND DISCUSSION

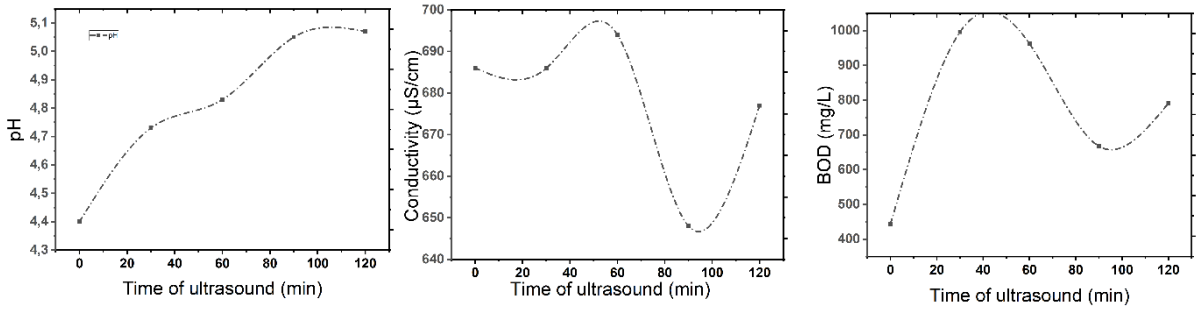
3.1 Ultrasound process for the removal of total organic carbon and inorganic carbon

Figure 1 and **Figure 2** show the decrease in TOC and IC concentrations in pharmaceutical wastewater after the ultrasound process. TOC is a key indicator of organic pollutants in clean water and wastewater; therefore, its removal is the main objective of various treatment processes (Hardyanti *et al*., 2024). Ultrasound treatment at pH 3 with an exposure time of 90 min achieved 71.48% TOC removal and 95.71% IC removal, primarily through acoustic cavitation and •OH radical formation. This ultrasound treatment produced a more oxidisable wastewater matrix, facilitating the high mineralisation observed in the integrated US–EF process. Although previous studies have identified an optimum pH near 4.5, the present system performed best at pH 3. This shift reflects the specific characteristics of the real pharmaceutical wastewater used in this study, as well as the fact that the ultrasound stage was designed and optimised as a separate pre-treatment unit. At pH 3, the dissociation of water during ultrasonic cavitation ($H_2O \rightarrow \bullet H + \bullet OH$) becomes more effective in producing hydroxyl radicals. In addition, this acidic condition creates favorable conditions for subsequent Fenton reactions, as iron (Fe^{2+}) speciation is more stable (Olabi, 2023). Therefore, the optimal pH is highly dependent on the specific wastewater matrix and treatment sequence. The pH 3 finding in this study remains consistent with the acidic range of pH 3–4, which is generally effective for Fenton-based processes. The decrease in TOC concentration is consistent with previous research stating that ultrasound can degrade organic compounds through pyrolysis and free radical reactions (Rayaroth *et al*., 2016b). The resulting cavitation effect triggers mechanical, thermal, and chemical responses that can be utilised in wastewater treatment processes (Wang *et al*., 2023). Ultrasound-based treatment utilises acoustic cavitation, which refers to the formation, growth, and collapse of bubbles in a liquid medium (Pétrier, Combet and Mason, 2007; Stucchi, Cerrato, G. and Bianchi, 2019). Bubble collapse produces localised hot spots with very high temperatures of up to 5,000 K and extreme pressures of up to 1,800 atm, which support the degradation of organic pollutants (Stucchi, Cerrato and Bianchi, 2019). The degradation rate of these organic compounds is generally

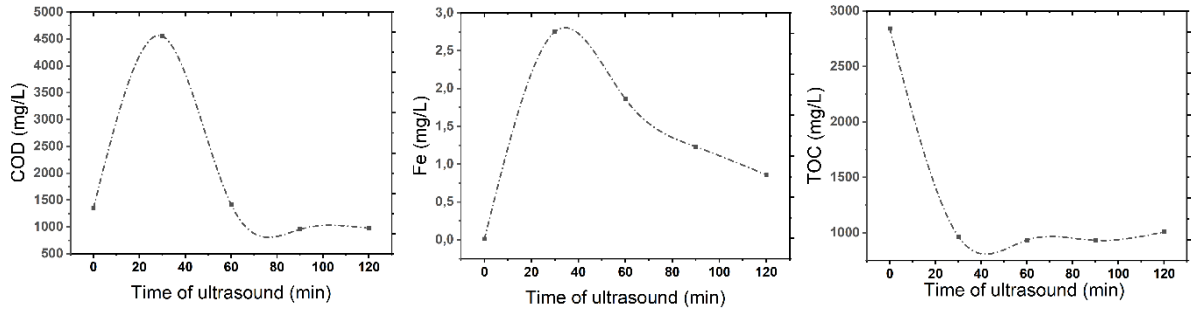
limited by the amount of hydroxyl radicals formed through the dissociation of water and oxygen during cavitation bubble collapse (Pétrier, Combet and Mason., 2007).

Different pH levels can significantly affect the efficiency of ultrasound degradation due to changes in chemical mechanisms, hydroxyl radical formation, and the overall acoustic cavitation process (Rayaroth, Aravind and Aravindakumar, 2016a; Gągol, Przyjazny and Boczkaj, 2018). Previous studies have shown that the optimal pH for TOC removal may vary. For example, a study using ultrasound-enhanced electrochemical treatment of pharmaceutical wastewater found that an initial pH of 4.5 was optimal for TOC removal (Patidar and Srivastava, 2021).

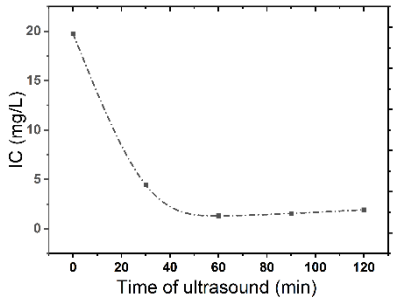
In conclusion, TOC degradation during ultrasound treatment is influenced by exposure time and pH within certain limits. IC, particularly in the context of pharmaceutical wastewater, refers to inorganic carbon species that may contribute to environmental impacts if not properly managed. Recalcitrant inorganic compounds are resistant to degradation and can persist in the environment, thereby posing ecological risks and inhibiting catabolic processes (Tiwari *et al*., 2019).



(a) (b) (c)

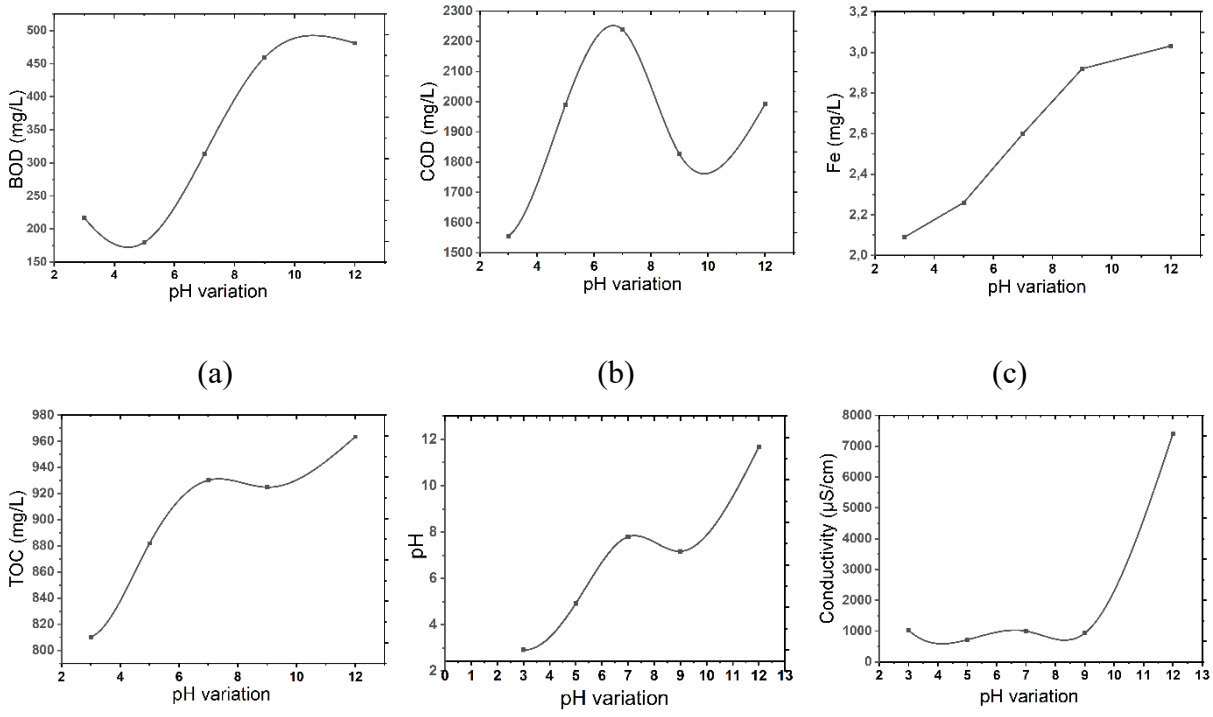


(d) (e) (f)



(g)

Figure 1. Changes in parameters: (a) pH, (b) conductivity, (c) BOD, (d) COD, (e) Fe, (f) TOC, and (g) IC due to time variations in pharmaceutical wastewater treatment using ultrasound.



(a) (b) (c)

(d) (e) (f)

Figure 2. Changes in parameters: (a) BOD, (b) COD, (c) Fe, (d) TOC, (e) pH, and (f) conductivity due to pH variations in pharmaceutical wastewater treatment using ultrasound.

3.2 Ultrasound–electro-Fenton integration for the removal of total organic carbon and inorganic carbon

The integrated ultrasound–Electro-Fenton treatment was carried out under the optimal pH condition obtained from the ultrasound process, namely pH 3. The time variations were 30,

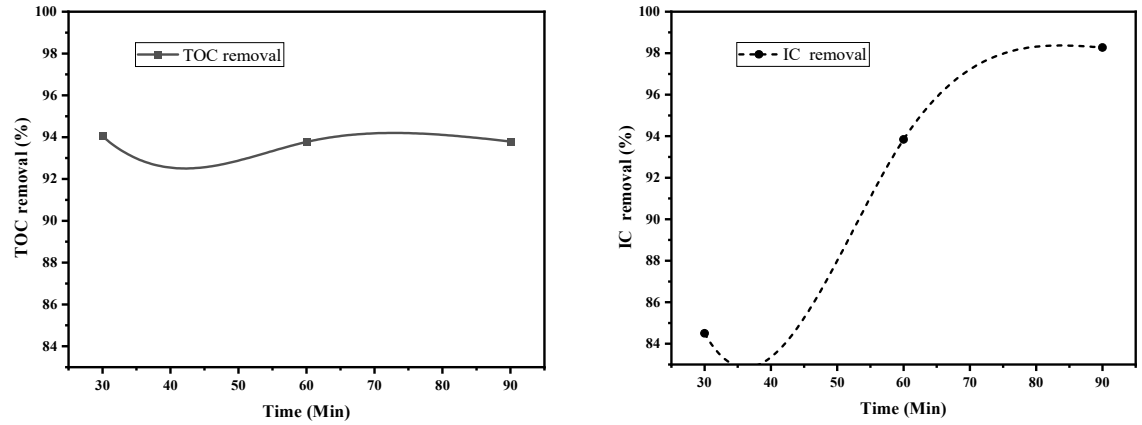
60, and 90 min. The $FeSO_4$ dose was 15 g, and the volume of H_2O_2 solution (30%, v/v) was 100 cm^3. The results of TOC and IC removal are shown in **Figure 3**. This integrated treatment successfully reduced TOC from the initial concentration of 2,841 $mg \cdot dm^{-3}$ to 169.2 $mg \cdot dm^{-3}$ after 30 min, corresponding to 94.04% TOC removal. IC removal reached 98.27% after 90 min. These results indicate that the integrated ultrasound–Electro-Fenton process is effective in reducing refractory organic compounds in pharmaceutical wastewater. The performance data show that US–EF achieved 94.04% TOC removal, far exceeding ultrasound pre-treatment alone, which achieved 71.48% removal. This result also suggests that EF treatment alone applied directly to raw wastewater may not achieve similarly high efficiency within the same treatment time.

The integrated ultrasound–Electro-Fenton (US–EF) process demonstrates strong performance due to the synergistic effects of sonolysis and Fenton reactions. The Fenton reaction ($Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + \bullet OH + OH^-$) produces hydroxyl radicals, while ultrasound enhances oxidation efficiency by promoting additional radical formation. In addition to increasing •OH production, ultrasound improves mass transfer, prevents electrode fouling, and physically breaks down complex organic molecular structures into intermediates that are more easily oxidised during the EF stage (Liu, Yu and Liu, 2023 Baştürk, 2024). The ultrasound–Electro-Fenton process enhances the mineralisation of organic pollutants during wastewater treatment (Hassani *et al.*, 2022). Electro-Fenton produces •OH, a strong oxidant, as shown in Equation (1), while ultrasound further enhances •OH production through acoustic cavitation and sonochemical reactions (Zhang et al., 2020; Bose *et al*., 2021; Zore *et al.*, 2021). Ultrasonic waves disrupt the structure of complex organic molecules, making them more susceptible to oxidation. Hydroxyl radicals oxidise organic compounds, including recalcitrant compounds, into CO_2 and H_2O. Ultrasonic waves also help prevent electrode fouling, allowing reaction efficiency to be maintained (Kapuralage *et al*., 2025).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + \bullet OH + OH^- \quad \text{..... (1)}$$

Carbon dioxide (CO_2), carbonic acid (H_2CO_3), carbonate (CO_3^{2-}), and bicarbonate (HCO_3^-) are IC species that may be present in pharmaceutical wastewater. These compounds can originate from pharmaceutical raw materials, acid–base neutralisation, and chemical reactions during drug synthesis (Luo *et al*., 2019). Ultrasonic cavitation produces microbubbles that absorb CO_2 and subsequently release it into the gas phase (Shanmugam, Balraj and Babarao,

2025). According to Ying *et al*. (2017)ultrasound increases the conversion of IC to CO_2 by 30–50% compared with conventional methods.



(a) (b)

Figure 3. Removal of (a) TOC and (b) IC from pharmaceutical wastewater using integrated ultrasound–Electro-Fenton treatment.

3.3 pH and Conductivity During the Pharmaceutical Wastewater Ultrasound Process

The acidity level of pharmaceutical wastewater is a critical parameter that significantly influences both the efficiency of the treatment process and the final quality of the treated effluent (Huang *et al.*, 2012; Ahmad *et al*., 2017). Therefore, maintaining an appropriate pH is essential to ensure the effectiveness of pharmaceutical wastewater treatment. After ultrasound treatment with time and pH variations, the pH value of the wastewater is expected to meet the quality standard of 6–9.

The initial pH value of the pharmaceutical wastewater sample was 4.4. The best pH value was obtained at the 120-min time variation, reaching 5.07, while the pH 7 variation resulted in a pH value of 7.8. Meanwhile, the lowest pH value was obtained at the 30-min time variation, reaching 4.73, whereas the pH 12 variation resulted in the highest pH value of 11.68. Ultrasound treatment can cause pH changes in pharmaceutical wastewater, mainly due to acoustic cavitation and sonochemical reactions (Zhang *et al*., 2020; Bose *et al*., 2021; Zore *et al*., 2021). The high energy generated inside cavitation bubbles can initiate and accelerate chemical reactions. In aqueous solutions, sonochemical reactions may lead to the formation or consumption of acidic and alkaline compounds, thereby causing changes in pH (Bose *et al*., 2021; Ma *et al*., 2021). In addition, ultrasound treatment can break down water molecules into H^+ and OH^- ions, which directly affects pH (Jiang, Xiao and Fu, 2023). Longer exposure

times generally promote greater pollutant degradation, which may result in more significant pH changes depending on the by-products formed (Li, Liu and Li, 2016; Suherman et al., 2020; Rosińska, 2021). The initial pH of the wastewater also affects the effectiveness of the treatment process and the direction of pH change (Wei *et al*., 2016). Therefore, the pH of wastewater after ultrasound treatment is influenced by both the initial pH and the ultrasound exposure time. In pharmaceutical wastewater, increased conductivity may originate from various sources, including raw materials, solvents, and by-products generated during pharmaceutical manufacturing processes (Dawood, Aziz and Ismael, 2023; Vasudha and Laiju, 2024).

In this study, conductivity increased after the samples were treated under different pH variations. The lowest increase in conductivity occurred at the pH 5 variation, reaching 719 µS/cm. The highest conductivity values were observed at the 60-min time variation, reaching 693.54 µS/cm, and at the pH 12 variation, reaching 7,410 µS/cm. The effect of ultrasound treatment on wastewater conductivity is associated with several mechanisms. Ultrasound irradiation can directly influence the conductivity of wastewater by promoting pollutant degradation. The degradation of pollutants can alter the ionic composition of wastewater, thereby affecting its conductivity. Ultrasound treatment causes cavitation, which can break down complex organic molecules into simpler ionic species and consequently increase the conductivity of the solution. Longer exposure times may intensify cavitation, leading to greater ion release and increased conductivity (Wei-chu, Ri-fu and Tai-qiu, 2011; Kesari *et al*., 2011).

3.4 Biochemical oxygen demand and chemical oxygen demand during the pharmaceutical wastewater ultrasound process

BOD is a water quality parameter used to assess the amount of biodegradable organic matter in water and wastewater (Lin *et al*., 2022). BOD measures the amount of oxygen consumed during the decomposition of organic substances. Based on the results of the ultrasound time variation treatment, the lowest increase in BOD concentration occurred at 90 min, increasing from 443.8 $mg \cdot dm^{-3}$ to 667.2 $mg \cdot dm^{-3}$. Meanwhile, the highest increase occurred at 30 min, increasing from 443.8 $mg \cdot dm^{-3}$ to 996.2 $mg \cdot dm^{-3}$. In the pH variation treatment, the most optimal BOD concentration was obtained at pH 5, with a final concentration of 180 $mg \cdot dm^{-3}$ and a removal efficiency of 59.4%. The lowest removal was obtained at pH 12, where the BOD concentration increased from 443.8 $mg \cdot dm^{-3}$ to 481

mg·dm^{-3}. Based on the results of this study, the initial BOD/COD ratio was 443.8/4,551.67 = 0.10. After ultrasound treatment for 90 min, the BOD/COD ratio increased to 667.2/3,231.69 = 0.21. Thus, although the ratio improved, it remained below 0.5, and the absolute BOD and COD values still exceeded the discharge limits for municipal sewer networks. Therefore, the proposed US–EF process is intended as a pre-treatment to enhance biodegradability rather than as a standalone treatment for direct discharge. This finding is in line with previous research, which reported that ultrasound can break down complex compounds into simpler molecules that are more easily degraded by microorganisms, thereby increasing BOD concentrations (Patidar and Srivastava, 2021). Ultrasound can improve wastewater biodegradability by increasing the BOD/COD ratio, making the wastewater more suitable for biological treatment (Andrio *et al*., 2019). Exposure to ultrasonic waves for 30 min has been shown to increase COD reduction by breaking down complex organic compounds into simpler compounds (Sangave and Pandit, 2006). The formation and efficiency of ultrasonic cavitation, as well as the type and reactivity of free radicals produced during sonochemical degradation, are strongly influenced by pH. Under acidic conditions, hydroxyl radical formation is more optimal, thereby increasing the degradation of organic pollutants (Wei *et al*., 2016). pH variations can also affect the surface tension of the liquid, which further influences the formation, size, and stability of cavitation bubbles and consequently affects the effectiveness of pollutant degradation (Wang *et al*., 2023).

High COD levels in pharmaceutical wastewater are often caused by active pharmaceutical ingredients (APIs) and other organic contaminants that are resistant to biodegradation (Verdini *et al*., 2024). In the ultrasound time variation treatment, the highest COD removal efficiency was obtained at 90 min, with a removal efficiency of 29%. After the optimum ultrasound time of 90 min was obtained, pH variation treatment was performed. In the pH variation treatment, the lowest increase in COD concentration occurred at pH 3, increasing from 1,351.66 mg·dm^{-3} to 1,555 mg·dm^{-3}. Meanwhile, the highest increase occurred at pH 7, where COD increased from 1,351.66 mg·dm^{-3} to 2,238.33 mg·dm^{-3}. The increase in COD at 30 min is thought to occur due to partial sonolysis, which breaks down complex organic compounds into intermediate compounds, such as carboxylic acids, that are more easily oxidised during COD analysis (Abdelhay *et al*., 2022). Because COD was analysed using unfiltered samples, the initial increase may also include organic particles released from suspended solids that were previously less bioavailable or not easily oxidised. In contrast, TOC was analysed using filtered samples; therefore, it only reflects the transformation and

mineralisation of dissolved organic carbon, which began to decrease. In addition, ultrasonic energy may release previously bound organic compounds, thereby increasing the measurable COD load. The downward trend in COD after longer exposure times, particularly at 60 and 90 min, confirms that these intermediate compounds were eventually further degraded through a gradual oxidation mechanism.

These results are consistent with previous research showing that ultrasound can break down particle aggregates and flocs in wastewater, thereby increasing the surface area available for chemical reactions. This dispersion allows more efficient oxidation of pollutants (Obukhova and Korolev, 2023). Therefore, ultrasound is often combined with other wastewater treatment methods to improve COD removal efficiency (Chandak, Ghosh and Gogate, 2020). In addition, wastewater pH plays an important role in COD removal efficiency. Lower pH values generally enhance COD removal during internal electrolysis (Sun *et al*., 2011). This is supported by a previous study on the treatment of pharmaceutical wastewater from bulk medicine production, which found that an initial pH of 4.5, combined with ultrasonic power, resulted in the highest COD removal efficiency (Patidar and Srivastava, 2021). This finding is also consistent with the present study, in which ultrasound treatment under acidic pH conditions produced the highest removal efficiency. However, an undesirable result was observed during ultrasound treatment with pH variations: none of the tested pH variations produced a COD concentration lower than the initial COD concentration, resulting in negative COD removal efficiency. In other words, ultrasound exposure time and pH variation influence COD removal effectiveness within a specific range, but inappropriate pH conditions may reduce treatment performance.

3.5 Total suspended solids and turbidity during the pharmaceutical wastewater ultrasound process

In the context of pharmaceutical wastewater, TSS may include residual medicinal raw materials, by-products from the production process, and other chemical substances (Shah and Shah, 2020). TSS measurement is essential for monitoring, evaluating, and determining the effectiveness of wastewater treatment processes (Katare *et al*., 2023). In this study, TSS was analysed using the gravimetric method.

The expected result after ultrasound treatment with time and pH variations was a decrease in TSS concentration. The initial TSS concentration was 91.5 $mg \cdot dm^{-3}$. In the ultrasound time variation treatment, the lowest increase in TSS concentration occurred at 30 min, increasing

from 91.5 mg·dm^{-3} to 122 mg·dm^{-3}. Meanwhile, the highest increase occurred at 120 min, increasing from 91.5 mg·dm^{-3} to 268 mg·dm^{-3}, corresponding to a 192% increase from the initial value. After the best ultrasound time variation, namely 30 min, was obtained, pH variation treatment was performed. In the pH variation treatment, the highest removal efficiency was found at pH 12, with a removal efficiency of 41%. Meanwhile, the lowest removal efficiency was observed at pH 3, where TSS increased from 91.5 mg·dm^{-3} to 186 mg·dm^{-3}. The increase in TSS observed in this study was likely due to the dispersion and breakdown of particles. Intense and prolonged ultrasonic cavitation energy can disperse fine agglomerates into smaller and more stable suspended particles, making them measurable in TSS analysis. In addition, changes in pH conditions and partial oxidation processes may induce the precipitation of dissolved compounds, thereby increasing the amount of suspended solids. These results remain consistent with the literature because the effects of ultrasound are highly dependent on operating conditions. Prolonged exposure or certain pH conditions can increase colloidal dispersion, as observed at pH 3, in contrast to the commonly reported decreasing trend under optimal conditions or in the presence of coagulants.

The results of the ultrasound time variation treatment are not consistent with previous studies, which reported that ultrasound can reduce TSS in wastewater by breaking down large particles into smaller particles, thereby facilitating separation processes such as sedimentation or filtration (Liu, Gao and Lu, 2021). A graph depicting changes in TSS and VSS during ultrasonic treatment also showed a decrease in TSS concentration over time; for example, TSS decreased from approximately 90 mg·dm^{-3} to 30 mg·dm^{-3} after 60 h of treatment (Amabilis-Sosa *et al*., 2018). Meanwhile, the results of the pH variation treatment are consistent with previous research stating that pH can affect the solubility of various chemical compounds in water, including compounds that contribute to TSS. For example, under low pH conditions, some minerals and metals may dissolve more easily, which can increase TSS. In contrast, under high pH conditions, some compounds may precipitate or form particles, thereby contributing to TSS reduction (Rifai and Widiarti, 2024). Therefore, it can be concluded that ultrasound exposure time does not always lead to TSS reduction, whereas pH variation affects TSS quality. Acidic pH may increase TSS, while more alkaline conditions may decrease TSS due to changes in solubility and precipitation behaviour. Turbidity is also an important parameter in wastewater treatment. Turbidity removal is important because suspended particles that cause turbidity can interfere with subsequent treatment processes. Ultrasound, through the cavitation mechanism, plays an important role in this process (Wang

et al., 2023).

The most optimal turbidity removal was obtained at 90 min, resulting in a turbidity value of 415 NTU and a removal efficiency of 11.3%. In the pH variation treatment, the best result was obtained at pH 12, with a turbidity value of 85.5 NTU and a removal efficiency of 81.7%. Meanwhile, the lowest turbidity removal was observed at 120 min, with a turbidity value of 434 NTU, and at pH 3, with a turbidity value of 637 NTU. These results are in line with previous research stating that increased exposure time to ultrasonic radiation can improve the degradation of organic pollutants and reduce turbidity (Majeed and Abdullah, 2018). Ultrasound produces cavitation bubbles that collapse rapidly and release high energy locally. This process supports the decomposition of suspended particles and organic matter that cause turbidity (Afreen and Upadhyayula, 2021; Wang *et al*., 2023). Longer ultrasound exposure increases the occurrence of cavitation, thereby increasing the possibility of particle breakdown and turbidity removal (Barati *et al*., 2007; Xu and Bigelow, 2011). However, there are limitations to the exposure time required to achieve optimal turbidity reduction, which can be further analysed using response surface modelling. Meanwhile, the optimal pH for ultrasound treatment depends on several factors, such as the specific contaminants present, the combination with other AOP methods, and the type of catalyst used (Moreira *et al*., 2016; Park, Lee and Jin, 2024; Roslan *et al*., 2024). The pH value affects the surface charge of colloidal particles, which influences their stability and tendency to aggregate. pH adjustment can improve coagulation and flocculation, making turbidity easier to remove (Mohammed, 2015). In other words, ultrasound exposure time and pH influence the effectiveness of turbidity removal within a specific treatment range.

3.6 ~~Fe~~ Iron Content Change During the Pharmaceutical Wastewater Ultrasound Process

Iron (Fe) in wastewater refers to the presence of Fe ions or Fe-containing compounds in wastewater (Wu, Vymazal and Brix, 2019). After the optimal ultrasound exposure time was obtained, five pH variations were tested, namely pH 3, pH 5, pH 7, pH 9, and pH 12. Based on the results of the ultrasound time variation treatment, the lowest increase in Fe concentration occurred at 90 min, increasing from 0.01 $mg \cdot dm^{-3}$ to 1.228 $mg \cdot dm^{-3}$. Meanwhile, the highest increase occurred at 30 min, increasing from 0.01 $mg \cdot dm^{-3}$ to 2.747 $mg \cdot dm^{-3}$. The increase in Fe concentration during ultrasound treatment without the addition of a catalyst was likely caused by cavitation-induced equipment corrosion or the release of bound iron from the wastewater matrix. This unexpected finding requires further

investigation. To minimise corrosion, shorter treatment times and polarity reversal (PR) may be applied. PR can help dislodge insulating deposits, while shorter operating times can reduce overall metal dissolution and energy consumption (Yasri *et al*., 2022). In the pH variation treatment, the lowest increase in Fe concentration was obtained at pH 3, increasing from 0.01 $mg \cdot dm^{-3}$ to 2.087 $mg \cdot dm^{-3}$. Meanwhile, the highest increase occurred at pH 12, increasing from 0.01 $mg \cdot dm^{-3}$ to 3.032 $mg \cdot dm^{-3}$. This finding is consistent with previous research stating that ultrasound efficiency is highly dependent on pH and ultrasound exposure time. Optimal conditions often occur under acidic pH, which favors hydroxyl radical formation (Saghafinia, Emadian and Vossoughi, 2011; Abdili *et al.*, 2017). Hydroxyl radicals are produced from collapsing cavitation bubbles generated during ultrasound treatment. These bubbles create localised extreme temperatures and pressures, which can influence Fe transformation through oxidation–reduction reactions involving Fe^{2+} and Fe^{3+} (Kosman, 2013; Ahmed and Mohamed, 2024). Thus, it can be concluded that pH and ultrasound exposure time can affect Fe concentration in pharmaceutical wastewater.

4. CONCLUSIONS

Based on the results of this study, the best operational condition for reducing TOC in pharmaceutical wastewater using ultrasound treatment was obtained at pH 3, with a TOC removal efficiency of 71.48%. Under acidic conditions, •OH formation through water dissociation during cavitation was enhanced, resulting in the degradation of recalcitrant compounds, as indicated by 71.48% TOC removal and 95.71% IC removal. The integrated ultrasound–Electro-Fenton process successfully reduced TOC by 94.04%. IC was also successfully reduced by 98.27% at 90 min. These results indicate that ultrasound–Electro-Fenton integration is effective in reducing refractory organic compounds in pharmaceutical wastewater. Ultrasound contributes to the degradation of organic pollutants in pharmaceutical wastewater through mechanical cavitation and hydroxyl radical formation. Meanwhile, the Electro-Fenton process produces •OH as a strong oxidant, while ultrasound enhances •OH production through acoustic cavitation and sonochemical reactions. Turbidity decreased significantly by 81.7% at pH 12 due to the destabilisation of colloidal particles by ultrasonic waves. The increase in the BOD/COD ratio indicates the breakdown of complex pollutants into simpler compounds that are more readily biodegradable. Acidic pH conditions, particularly pH 3–5, were more effective for the degradation of organic aggregates than alkaline conditions. The breakdown of complex compounds increased biodegradability, as

indicated by the decrease in BOD under acidic pH conditions. Finally, the treated effluent still requires post-treatment, such as activated sludge treatment, pH neutralisation, or filtration, to meet municipal sewer discharge standards for BOD, COD, TSS, and pH 6–9. The main implication of this study is that ultrasound–electro-Fenton integration has potential as a partial treatment solution for pharmaceutical wastewater. However, further integration with other treatment methods, energy optimisation, and economic–environmental feasibility validation are required. The main challenge for industrial-scale applications is the high energy consumption of ultrasound; therefore, optimisation of technical parameters, reactor design, and operational costs should be the focus of future studies.

ABBREVIATIONS

AOP	Advanced Oxidation Process
API	Active Pharmaceutical Ingredient
BOD	Biochemical Oxygen Demand
COD	Chemical Oxygen Demand
DPH	Diphenhydramine
EF	Electro-Fenton
IC	Inorganic Carbon
•OH	Hydroxyl Radical
PPCP	Pharmaceutical and Personal Care Products
TOC	Total Organic Carbon
TSS	Total Suspended Solids
US	Ultrasound
US-EF	Ultrasound-Electro Fenton Integration
Fe^{2+}	Ferrous Ion
Fe^{3+}	Ferric Ion
H_2O_2	Hydrogen Peroxide
kV	Kilovolt (used for voltage)
kHz	Kilohertz (ultrasonic frequency)
NTU	Nephelometric Turbidity Unit
µS/cm	Microsiemens per centimeter
$mg \cdot dm^{-3}$	milligrams per cubic decimeter
min	Minute
V	Volt

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CONFLICT OF INTERESTS

All authors declare that they have no conflict of interest.

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