



Treatment of Industrial Wastewater of Variable Quality Using Ultrasound Irradiation

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Abstract

This paper investigated the use of ultrasound irradiation to treat real mixed industrial wastewater under various conditions, including single and dual frequencies, variable wastewater strengths, and different operating conditions, including flow rates/residence times. This work is important to evaluate the organics' removal efficiency and to identify operational control bottlenecks under actual wastewater conditions. The highest removal efficiencies were 69.5% and 31.9% for COD and TOC, respectively, for the high-strength wastewater, which were found to occur at 16 kHz frequencies and 500 ml/min flow rate. The removal efficiencies were slightly less in the case of medium-strength wastewater (66.7 and 25.3% for COD and TOC, respectively). They were found to occur at dual frequencies of 16/20 kHz and 1500 and 1000 ml/min, respectively. For the low-strength wastewater, the efficiencies reached 78.6 and 9.1% for COD and TOC, respectively, at the same frequency and flow rates as the medium-strength wastewater. These findings demonstrated the effectiveness of dual frequency in medium- to low-strength wastewater. Among the organics monitored, chloroform (CHCl₃), tetrachloroethene (C₂Cl₄), 1,4 dichlorobenzene (C₆H₄Cl₂), and dichloromethane (CH₂Cl₂) exhibited variable removal, and in some cases, the removal was found to be negative, indicating intermediary products as a result of incomplete oxidation of organics. Besides the frequency and flow rate, it was found that the concentration of metals and organics are mostly positive influencers on organics removal. At the same time, TDS and pH have mixed effects, but they positively influence organics' removal at higher flows in a few instances. Additionally, as the residence time decreased, the concentration of organics and the pH negatively influenced organics removal.

Keywords: Advanced Oxidation Processes; Cavitation, Dual Frequency; Organic Degradation; Ultrasound Technology; Wastewater Treatment.

1. Introduction

Industrial wastewater treatment is complex as it contains a diverse and wide variety of constituents [1, 2], which could pronouncedly impact human health and ecosystems [3] if not appropriately treated. Because of operational limitations, industrial wastewater treatment often necessitates combining treatment technologies, advanced treatment technologies, and requires post-processing [4]. Ultrasound technology (UST), a chemical-free advanced oxidation technology [5-7], is suitable for industrial wastewater treatment and promises added value in terms of its oxidative abilities [6, 8-13] and sludge reduction potential [14]. Therefore, ultrasound technology has the potential to circumvent some of the disadvantages of existing industrial wastewater treatment technologies when applied at full scale [15, 16].

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The underlying principle of UST is that when powerful ultrasound (16 kHz–100 MHz or higher) is applied to liquids, it can induce chemical changes due to acoustically generated cavitation [14]. The acoustic cavitation phenomenon causes bubbles' formation, growth, and collapse, accompanied by a generation of localized extremely high temperature, pressure, and reactive radical species ($H^\bullet$, $OH^\bullet$) via thermal dissociation of water and oxygen. These radicals could oxidize dissolved organic compounds while hydrogen peroxide (H_2O_2) is formed due to the radical recombination of $^\bullet OH$ and $^\bullet OOH$ in the outside circumference of the cavitation bubble [14, 17, 18]. Additionally, UST promotes hydrolysis, the rate-limiting stage for organics oxidation during wastewater treatment [8-13]. Fundamentally, ultrasound can destroy organics, including hard-to-degrade organics and bacterial cells [9-13], possibly without chemicals or biochemical agents. When used as a pre-treatment, a significant benefit of ultrasound technology besides the oxidation of organics is that it reduces chemical use and sludge where chemical treatment is used, improves volatile suspended solids (VSS) levels, maintains nutrient availability, and improves biodegradability [5, 6].

However, the cost of using UST for full-scale industrial wastewater treatment is a significant issue, as these costs are high, mainly due to intensive energy requirements [19]. For example, the costs for cleaning contaminated groundwater using acoustic cavitation are, in the order of magnitude, higher than those of air stripping or activated carbon processes [20]. However, more energy efficiency combined with operating at low frequencies could significantly improve the economy of UST [21, 22].

Many studies have investigated the use of UST to treat particular organics and even microplastics from water and wastewater at a laboratory scale [16, 22-25], but only a few studies have investigated its use for real industrial wastewater. However, Sponza & Oztekin [26] investigated the ultrasonic destruction of polycyclic aromatic hydrocarbons (PAHs) in real petrochemical industry wastewater, and it was found that the degradation of organic pollutants is difficult using ultrasound with low frequency alone. Gogate & Pandit [24] found that the pollutant molecules are degraded into smaller molecular weight compounds rather than completely mineralized. Therefore, effective removal of these pollutants was difficult. Other studies investigated different additives (such as NaCl, CCl_4 , ZnO, and Fe particles), multiple frequencies, and combinations with other oxidation processes [23, 27-29]. These techniques have been reported to be more efficient than single-frequency ultrasound and are one to two orders of magnitude lower in cost than UST alone.

These combinations' costs are also comparable to those of other advanced oxidation processes [23]. Also, Sivakumar et al. [30] have demonstrated that a combination of different frequencies produces more intense cavitation, as evidenced by the higher values of the pressure pulse generated at the end of the cavitation event as compared to the single-frequency operation, and hence, could yield higher organics removal. Thus, dual- or triple-frequency reactors could give similar results to single, very high frequencies, but with minimal problems of vessel and transducer erosion and energy requirements [31].

To summarize, it is important to stress that almost all published studies were conducted using synthetic solutions and may or may not give similar degradation rates when applied to real effluents containing various compounds [23]. It is also important to note that, in many instances, the advantages of UST outweigh its costs, and, therefore, it still has a potential use in industrial wastewater treatment. Also, optimizing UST use in wastewater treatment plants could lead to significant cost reduction [32], but this requires careful examination of its efficiency and real wastewater conditions. Furthermore, new advancements in the design of UST reactors have reduced the costs significantly [33].

Therefore, this research paper investigates using low and dual frequencies to treat real industrial wastewater utilizing a cylindrical design flow-through reactor. Testing on real industrial wastewater is essential as it typically contains a widely variable range of metals and salts, both of which positively enhance organic degradation, as discussed previously [23, 27-29]. Within this broad objective, the presence and fluctuation of selected organic compounds were monitored to get an insight, understand removal, and identify factors affecting the performance of UST. This paper also contributes to the scanty literature in this field.

Besides this introduction, Section 2 describes the methodology, including wastewater characteristics, ultrasound reactor setup, operating conditions, laboratory analysis, and data interpretation. Section 3 presents and discusses the overall organics removal performance of the sonication experiments, the removal of some specific organic compounds, and the effects of wastewater constituents on the ultrasound treatment performance. Section 4 summarizes the study findings while highlighting the key performance limitations of ultrasound of real industrial wastewater.

2. Research Methodology

A series of tasks have been performed to examine the potential use of dual and less energy-intensive ultrasound frequencies on variable-quality wastewater (Figure 1). This approach would enhance the knowledge of the limitations and benefits of this promising technology.

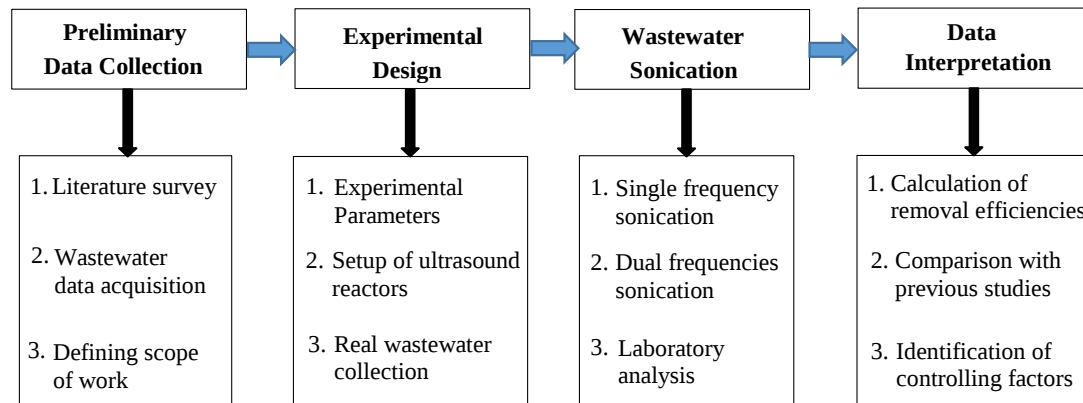


Figure 1. Schematic of research workflow

2.1. Wastewater Sonication Experiments

Sonication experiments were conducted using a laboratory-scale dual-frequency (model M/N: DRC-4-DPP, Advanced Sonic Processing Systems, USA, internal dimensions 0.14"/25 ml cell, 16 and 20 kHz) flow-through ultrasound reactor (Figure 2) and a batch ultrasound bath (model Elmasonic P30, Elma, Germany, internal dimensions 240 × 137 × 100 mm, 37 and 80 kHz). The ultrasound frequency and wastewater detention times were varied during the experiments.

The wastewater (a mix of food and beverage, petrochemical, and slaughterhouse wastewater) was obtained from an industrial wastewater treatment plant in Kuwait and classified as high, medium, and low strength (Table 1). This classification is adopted here for the sake of analyzing the results only. Samples of wastewater were collected before and after sonication to determine removal.

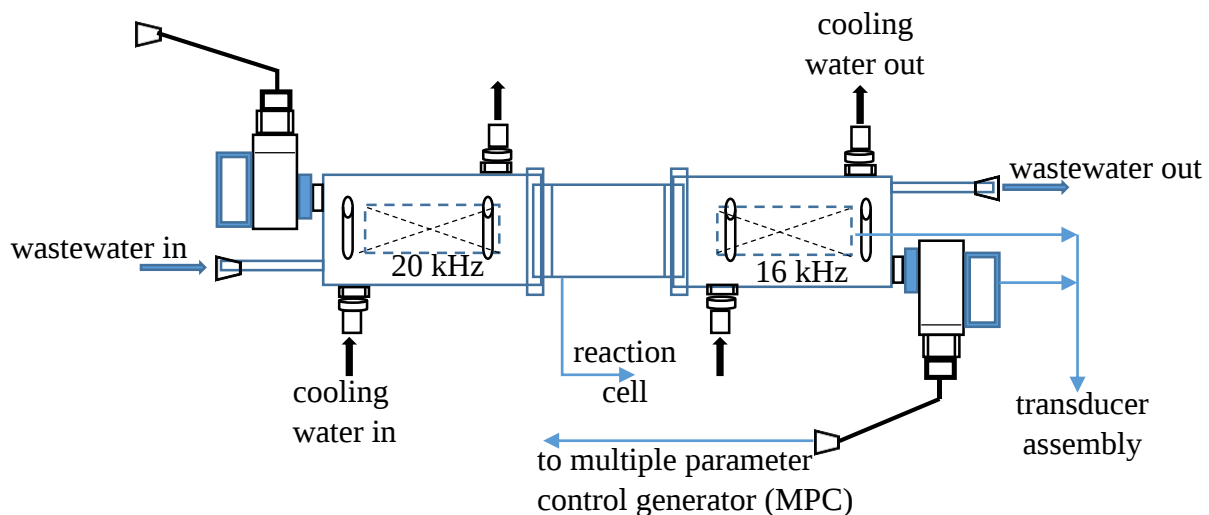


Figure 2. Schematic of the dual-frequency ultrasound flow-through reactor

Sonication of wastewater was conducted using the flow-through reactor at 16, 20, and 16/20 kHz dual frequencies. Flow rates were 500, 750, 1000, 1500, and 2280 ml/min. The average characteristics of all types of wastewater are shown in Table 1.

To compare the oxidation potential of the flow-through reactor operating at low and dual frequencies compared to higher frequencies, the wastewater was sonicated at 37 and 80 kHz using the ultrasound bath. The sonication duration was selected to correspond to the detention time of the flow-through reactor to enable comparison of the results.

Table 1. Characteristics of industrial wastewater used in the experiments

Parameter	Units	High Strength		Medium Strength		Low Strength	
		Ave	STD	Ave	STD	Ave	STD
Temperature	°C	28.9	5.2	28.9	5.2	28.8	5.0
Dissolved Oxygen	mg/l	3.1	1.8	4.2	1.2	3.4	1.4
pH	-	6.7	1.5	6.9	1.5	7.7	1.6
Electrical Conductivity	mS/cm	2.2	0.4	2.5	0.6	3.9	2.9
Biochemical Oxygen Demand (BOD ₅)	mg/l	1387.2	691.3	823.9	257.8	31.8	14.9
Chemical Oxygen Demand	mg/l	2702.7	1728.4	1355.7	420.8	51.7	24.5
Total Organic Carbon (TOC)	mg/l	263.2	144.4	290.2	113.6	21.5	10.6
Total Suspended Solids (TSS)	mg/l	892.5	886.0	210.7	225.3	33.6	16.3
Total Dissolved Solids (TDS)	mg/l	1263.2	258.6	1426.1	348.9	1558.6	438.8
Volatile Suspended Solids (VSS)	mg/l	606.4	638.4	159.6	210.3	22.4	9.8
Total Phosphate (TPO ₄ P)	mg/l	27.8	23.5	7.1	4.9	2.2	1.4
Ammonia (NH ₃)	mg/l	58.0	20.1	57.3	18.9	2.4	5.5
Nitrate (NO ₃)	mg/l	1.1	2.0	0.6	0.4	15.6	25.5
Nitrite (NO ₂)	mg/l	1.0	1.2	2.7	10.8	1.9	7.9
Total Nitrogen (TN)	mg/l	99.1	30.9	112.4	161.5	31.6	17.7
Phenols	mg/l	0.3	0.1	0.4	0.08	0.1	0.1
Fluoride (F ⁻)	mg/l	22.1	41.5	26.5	57.3	25.7	50.2
Sulfide (S ₂ ⁻)	mg/l	0.4	0.2	0.1	0.1	0.1	0.2
Total Oil & Grease (O&G)	mg/l	56.7	32.0	14.33	12.2	0.5	1.3
Total Petroleum Hydrocarbons (TPH)	mg/l	37.4	72.2	7.74	9.9	3.5	2.4
Aluminium (Al)	mg/l	64.7	5.5	18.3	0.7	3.4	0.8
Arsenic (As)	mg/l	5.3	3.2	6.5	5.6	5.7	2.9
Barium (Ba)	mg/l	79.1	68.6	34.7	37.9	26.7	44.2
Boron (B)	mg/l	80.8	42.8	107.6	79.99	103.6	76.5
Cadmium (Cd)	mg/l	3.0	6.7	2.2	2.1	0.7	0.8
Chromium (Cr)	mg/l	47.6	46.8	11.5	9.5	4.1	4.1
Nickel (Ni)	mg/l	103.4	350.1	26.3	24.7	10.2	5.7
Mercury (Hg)	mg/l	4.2	3.9	1.8	1.9	1.3	1.9
Cobalt (Co)	mg/l	4.4	5.2	8.5	9.1	1.6	1.7
Iron (Fe)	mg/l	35.9	42.1	31.2	40.1	21.6	20.2
Antimony (Sb)	mg/l	7.4	5.7	8.1	6.4	6.1	5.8
Copper (Cu)	mg/l	189.7	183.1	63.2	68.1	30.9	39.6
Manganese (Mn)	mg/l	442.5	351.1	974.1	727.3	197.3	120.8
Zinc (Zn)	mg/l	107.8	116.4	25.3	62.39	12.7	9.2
Lead (Pb)	mg/l	103.2	165.1	20.4	30.9	16.7	14.8
Benzene (C ₆ H ₆)	mg/l	0.4	0.3	0.3	0.2	BDL	BDL
Toluene (C ₇ H ₈)	mg/l	2.0	0.7	3.4	1.7	BDL	BDL
Ethyl Benzene (C ₈ H ₁₀)	mg/l	BDL	BDL	BDL	BDL	BDL	BDL
o-Xylene [(CH ₃) ₂ C ₆ H ₄]	mg/l	3.5	2.4	3.3	2.1	0.1	0.0
m-Xylene [(CH ₃) ₂ C ₆ H ₄]	mg/l	2.5	1.7	2.2	1.4	BDL	BDL
p-Xylene [(CH ₃) ₂ C ₆ H ₄]	mg/l	0.8	0.4	1.6	1.1	0.1	0.0
Dichloromethane (CH ₂ Cl ₂)	mg/l	0.4	0.3	0.4	0.1	0.53	0.2
Tetrachloroethene (C ₂ Cl ₄)	mg/l	7.2	4.2	5.1	2.4	1.64	3.7
Chloroform (CHCl ₃)	mg/l	1.1	0.8	0.5	0.3	1.6	1.46
1, 3, 5 -Trimethylbenzene (C ₉ H ₁₂)	mg/l	1.2	0.7	1.0	0.6	BDL	BDL
1, 4 Dichlorobenzene (C ₆ H ₄ Cl ₂)	mg/l	10.6	7.2	10.8	7.3	0.8	0.3

BDL: Below detection limits

2.2. Laboratory Analysis

Major parameters were measured before and after the ultrasound treatment, including temperature, pH, electrical conductivity (EC), dissolved oxygen (DO), chemical oxygen demand (COD), total organic carbon (TOC), total suspended solids (TSS), total dissolved solids (TDS), total nitrogen (TN), total phosphorous (TP), volatile suspended solids (VSS), cadmium, chromium, nickel, mercury, cobalt, iron, antimony, copper, manganese, zinc, lead, boron, barium, arsenic, aluminum, sulfide, fluoride, oil and grease, total petroleum hydrocarbon (TPH), BTEX compounds, and phenol.

The analysis was conducted using standard procedures outlined in the Standard Methods for Water and Wastewater Examination [34]. Laboratory analysis was performed at SRP laboratories at KISR's Water Research Center (WRC), which are ISO 9001:2015 certified. All the necessary equipment was calibrated and inspected, and quality was assured routinely. Blanks and duplicate samples were analyzed to ensure quality per the recommended instructions for the equipment's QC. Calibration periodicity is different for each parameter's examination following the requirements of the applied method. The quality of the analytical results was ensured by monitoring proper QC values during the analysis as described in the reference method [34].

2.3. Statistical Analysis

Pearson correlation matrix of removal efficiency and selected wastewater parameters was generated to identify any statistically significant correlations among the dependent (removal rates) and independent variables (concentrations) using the Statistical Package for the Social Sciences (SPSS) v. 19.0 statistical software, according to the previously described methods [35, 36].

3. Results and Discussion

The results of organics removal/oxidation using UST are discussed with a focus on the overall organics removal (COD and TOC), the removal of specific organics, and understanding the influence of wastewater characteristics on organics removal/oxidation using UST. The specific organics such as BTEX, dichloromethane, tetrachloroethene, chloroform, 1,3,5-trimethyl benzene, and 1,4-dichlorobenzene selected to get a better understanding of the organics' removal and any transformation, generation, or intermediate products of organic compounds during the sonication process [37] and because of the profound effects these compounds may inflict on the environment and health [38, 39].

3.1. Overall Organics Removal

The removal efficiencies were calculated for different residence times and frequencies of 16, 20, and dual 16/20 kHz for COD and TOC for COD and TOC and the three classes of wastewater. The removal efficiencies are shown in Figures 3-5. For the high-strength wastewater (Figure 3), the removal efficiency of COD reached 69.5%, while the TOC removal was a minute 31.9%, which were the highest removals that occurred at 16 kHz and 500 ml/min. The removal of COD may have been hampered by the destruction of VSS (48.0%), which may partly convert to organics upon disintegration and reaction with the radicals [40, 41].

For the medium-strength wastewater (Figure 4), the organics removal reached 66.7, and 25.3% for COD and TOC, respectively. However, this removal occurred at 16/20 kHz for COD but at 20 kHz for TOC. The corresponding flow rates were 500 ml/min for COD and 2280 ml/min for TOC. Here, the dual frequency (16/20 kHz) performed better than single frequencies (16 and 20 kHz) in high-strength wastewater, probably due to the influence of the wastewater characteristics as reported in previous studies [8, 24, 30, 42].

The efficiency of the low-strength wastewater (Figure 5) did not improve over the medium-strength wastewater, reaching 78.6% and 9.1% for COD and TOC, respectively. These efficiencies occurred at a dual frequency of 16/20 kHz and 1500 and 1000 ml/min, respectively. Once again, the dilute nature of the wastewater has led to better dual-frequency performance than single frequencies. However, the efficiencies were generally low compared to the high and medium-strength wastewater [8, 24, 30, 42].

Similar removal efficiencies have been reported for COD [10, 27, 29, 43], but most important is that the dual frequency only dominated at low concentrations, indicating that the use of UST for the final destruction of organics is promising for treatment effluents. For example, Patidar & Srivastava [43] used a combination of low-frequency

ultrasound and nanomaterial additives for the oxidation of organics in cosmetic industry wastewater, which contains dilute concentrations of hard-to-degrade organics similar to those in industrial wastewater and achieved the COD removal of 60.2 to 82.4%. Although lower and under different conditions, this study's ranges fall within the previously published results [10, 27, 29, 43]. However, it must be noted that nanomaterials' oxidative properties have increased their removal efficiency.

In general, the effectiveness of ultrasound for organics' destruction depends on the frequency, residence time, and flow conditions (or mixing conditions) [21, 37, 41]. These dependencies are evident from Figures 3-5. Nonetheless, some anomalies will be further investigated.

Furthermore, it has been reported that ultrasound efficiency in organics destruction has an optimum range since it depends on many factors such as the frequency, uniformity of ultrasound waves, physicochemical properties of the liquid (such as vapor pressure, surface tension, viscosity, and presence of impurities and gases), rate constant for sonochemical reactions (higher at lower concentrations), oxygen levels, and the presence of catalysts (TiO_2 , NiSO_4 , CCl_4 , $\text{Fe}_2\text{Fe}_3\text{O}_4$, CuSO_4 and also salts such as NaCl enhances the extent of degradation), temperature (lower temperatures enhance degradation), pH, dual frequencies (could produce intense cavitation) [24, 30, 42].

In Figures 3 to 5, many optima and inflection points could be observed due to the variable wastewater characteristics, and therefore, only maxima have been discussed within the range of operational parameters considered in this study.

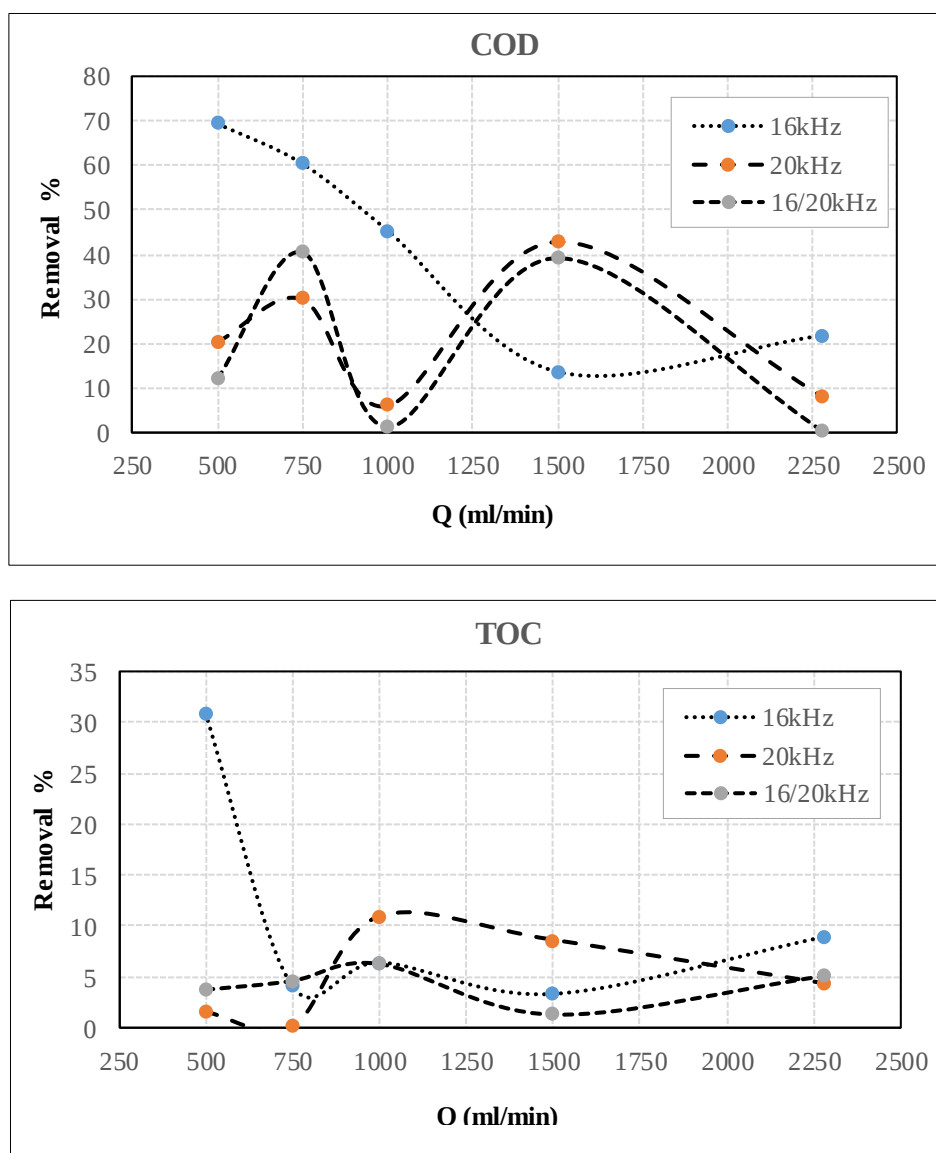


Figure 3. High-strength wastewater sonication in the flow-through ultrasound reactor

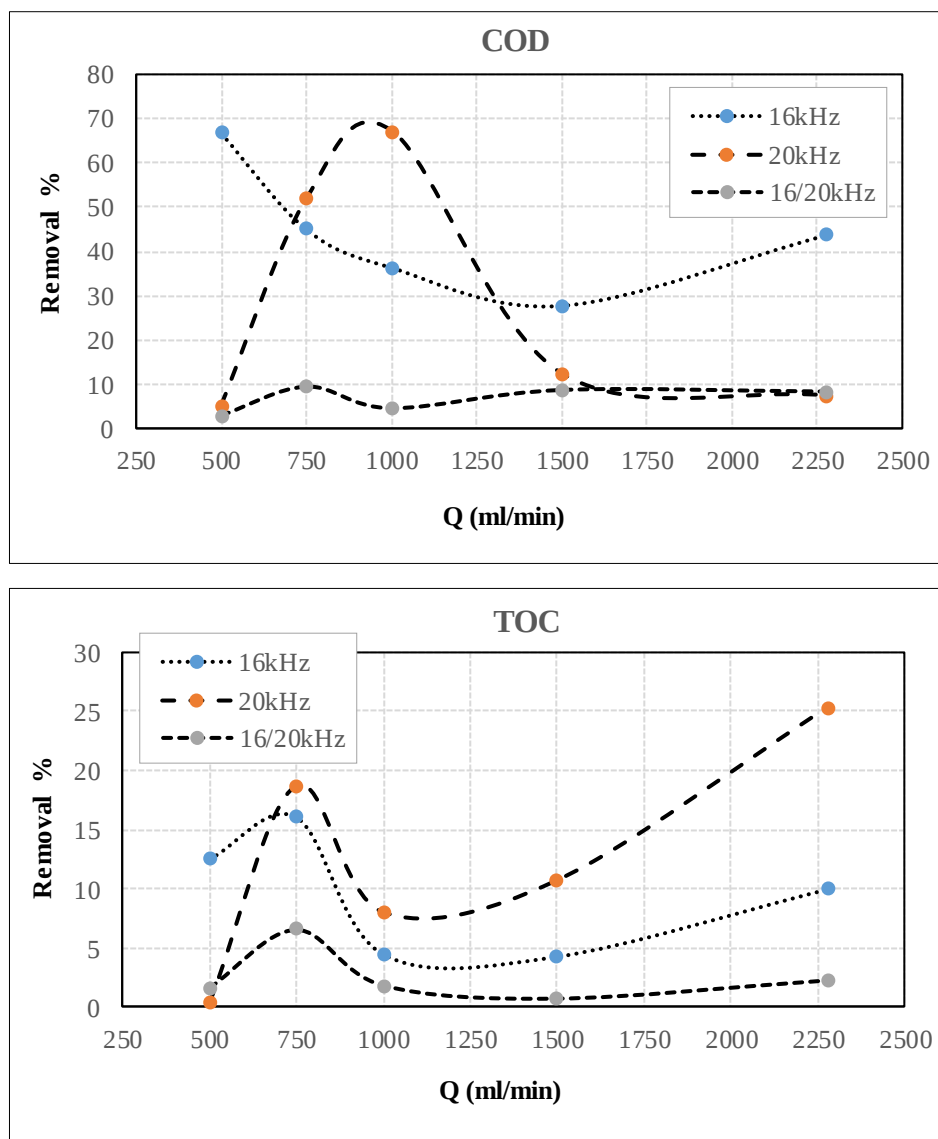


Figure 4. Medium-strength wastewater sonication in the flow-through ultrasound reactor

In the case of high-strength wastewater (Figure 3), adequate mixing and high frequencies seem to be the most appropriate conditions for using ultrasound for COD and TOC removal for medium-strength wastewater the higher the frequencies and the lesser the mixing could maximize COD and TOC COD removal, while for the low strength wastewater, it seems that lower frequency would achieve the highest removal with even lower mixing conditions. A pattern could be noticed depending on the strength of the wastewater, which is that for wastewater at high strength, the removal of the organic depends mainly on the high frequency, while for medium strength wastewater, more mixing is required. This pattern could even explain that for low-strength wastewater since the concentrations of organics are low, the even lower frequency is appropriate with lower frequencies or radical generation, as reported by Ghosh & Sahu [21] and Yang et al. [22].

While COD is relatively easier to degrade compared to TOC, as demonstrated by the results (Figures 3-5), vigorous mixing is required in the case of high and medium-strength wastewater. In the case of low-strength wastewater, maximum TOC removal could be achieved under the same conditions as maximum COD removal (Figure 5).

Furthermore, the ultrasound bath was used to test 37 and 80 kHz for low-strength wastewater regarding the removal of TOC. The comparison of the results with different frequencies is shown in Figure 6. TOC removal reached 9.3 and 7.8% at 37 and 80 kHz, respectively, and at higher residence times (> 1 hr), while a comparable removal at 16 and 16/20 kHz was achieved at 30 min residence time (Figure 6). In this context, the advantage of using low dual frequencies is evident. Figure 6 also demonstrates the complex nature of TOC degradation using ultrasound. However, while it is clear from Figure 5 that better TOC removal requires higher frequencies and more residence time, time, and energy could be saved by using dual frequencies. Again, the observation that an optimum exists, as has been previously reported by Ang et al. [16], could be seen in Figure 5.

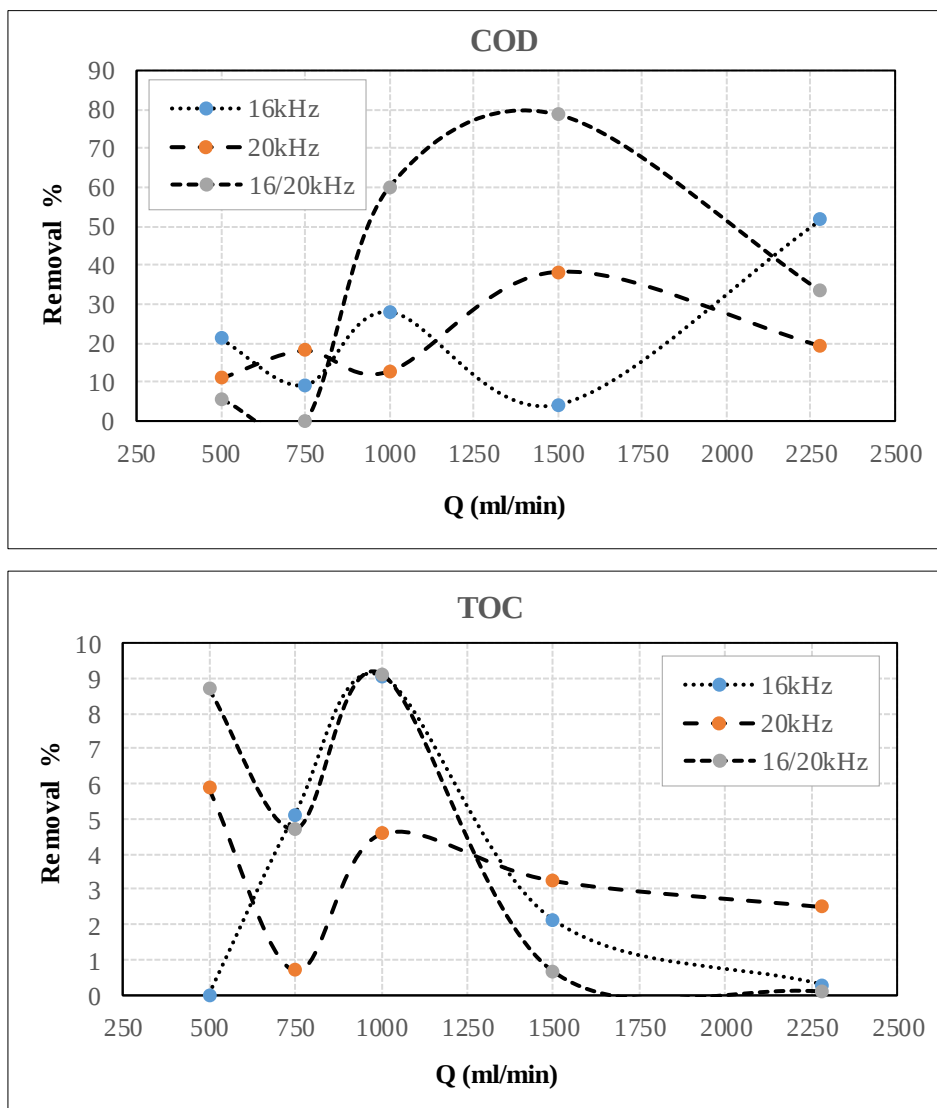


Figure 5. Low-strength wastewater sonication in the flow-through ultrasound reactor

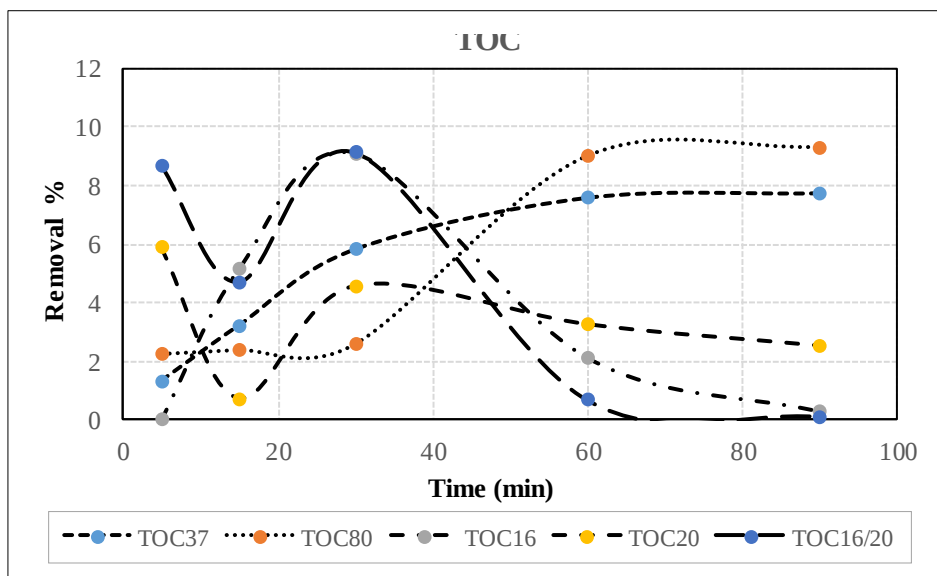


Figure 6. TOC removal under different ultrasound operating conditions

Nonetheless, operating the sono-electrochemical system at an ultrasonic power exceeding an upper limit will reduce pollutant removal efficiency [16]. Several reports have shown an optimal ultrasonic power value (which is case-specific) where the degradation performance peaks before declining [44, 45]. A few explanations have been given to describe this observation, which will be explored in the following sections.

3.2. Specific Organics Removal During Sonication of Low-Strength Wastewater

Although BTEXs were detected in raw wastewater at higher concentrations, the effluent contained zero levels. However, other VOCs, such as chloroform (CHCl_3), tetrachloroethene (C_2Cl_4), and 1, 4 dichlorobenzene ($\text{C}_6\text{H}_4\text{Cl}_2$) were detected at higher levels in the effluent. In a few instances, dichloromethane (CH_2Cl_2) was also detected in effluents. The removal efficiency of these four compounds under different ultrasound testing conditions is reported in Tables 2 and 3.

As can be seen from the flow-through experiments (Table 2), the removal efficiency of four compounds tends to decrease at higher flow rates or becomes negative due to releasing these compounds from bubbles. However, the highest efficiency of dichloromethane was achieved under 500 ml/min and at a dual frequency of 16/20 kHz, and when the flow increased to 750 ml/min, the efficiency was achieved at 16 kHz. This frequency shift may lead to an inverse proportionality between the ultrasound energy and water flow energy. An important observation is the positive removal of chloroform under all conditions.

Conceivably, the removal of dichloromethane and 1, 4 dichlorobenzene could be achieved at reasonable efficiencies compared to chloroform and tetrachloroethene, for which the removal efficiency was marginal in the flow-through reactor. Upon using higher frequencies, the removal efficiency of 1, 4 dichlorobenzene improved to 46.7% at 37 kHz with 5 min residence time. The longer residence times did not achieve better efficiencies.

Table 2. Removal Efficiency of Various Organics in a Flow-through Reactor

Residence Time (min)	Compound	Removal Efficiency %				
		16 kHz	20 kHz	16/20 kHz	37 kHz	80 kHz
5	Dichloromethane (CH_2Cl_2)	NCD	NCD	86.6	NCD	NCD
	Tetrachloroethene (C_2Cl_4)	17.1	17.1	22.9	-115.6	56.4
	Chloroform (CHCl_3)	6.9	4.5	0.0	NCD	NCD
	1, 4 Dichlorobenzene ($\text{C}_6\text{H}_4\text{Cl}_2$)	-60	-41.7	45.2	46.7	11.5
15	Dichloromethane (CH_2Cl_2)	78.3	NCD	50.0	NCD	NCD
	Tetrachloroethene (C_2Cl_4)	13.3	-14.9	-381.3	76.1	-291.7
	Chloroform (CHCl_3)	4.9	NCD	NCD	NCD	NCD
	1, 4 Dichlorobenzene ($\text{C}_6\text{H}_4\text{Cl}_2$)	56.7	66.7	-275	21.05263	-15.4
30	Dichloromethane (CH_2Cl_2)	NCD	NCD	NCD	NCD	NCD
	Tetrachloroethene (C_2Cl_4)	1	25.3	-17.9	12.5	68
	Chloroform (CHCl_3)	-0.7	-0.1	NCD	NCD	NCD
	1, 4 Dichlorobenzene ($\text{C}_6\text{H}_4\text{Cl}_2$)	76.5	-30.0	-44.4	-25.0	-33.3
60	Dichloromethane (CH_2Cl_2)	NCD	NCD	NCD	NCD	NCD
	Tetrachloroethene (C_2Cl_4)	-10.6	-10.3	74.5	78.3	73.8
	Chloroform (CHCl_3)	4.3	0.9	NCD	NCD	NCD
	1, 4 Dichlorobenzene ($\text{C}_6\text{H}_4\text{Cl}_2$)	14.3	-15.4	56.3	38.5	14.23
90	Dichloromethane (CH_2Cl_2)	NCD	NCD	NCD	NCD	NCD
	Tetrachloroethene (C_2Cl_4)	5.2	3.3	-16.4	NCD	NCD
	Chloroform (CHCl_3)	3.3	2.0	NCD	NCD	NCD
	1, 4 Dichlorobenzene ($\text{C}_6\text{H}_4\text{Cl}_2$)	-4.0	-5.0	3.0	NCD	NCD

NCD: No change in concentrations detected.

As evident from Table 3, some VOCs have negative removal efficiencies, which can be attributed to chemical transformations due to the oxidation of these VOCs or other organics in the effluent water, as confirmed by Gujar et al. [46].

Ayyildiz et al. [10] found that for less volatile compounds ($H_v < 0.1$), Henry's constant exerts a positive influence on the sonochemical degradation, but its effect is not continuous, while for highly volatile compounds ($H_v > 1$), the impact of Henry's constant on the degradation is marginal. In this study [10], they were able to remove 60 to 75% of trichloroethylene (TCE) and over 90% removal of ethylene dibromide (EDB) from groundwater, which is similar to the low-strength wastewater in this study.

In summary, using UST for raw wastewater could eliminate the need for the primary treatment stage, as the UST effluent is similar to the primary treatment effluents. In addition, the use of UST on aerated wastewater improves biodegradability and biomass levels due to the transformation of organics and degradation of flocs [47]. Finally, using UST for effluent treatment could further reduce the concentration of some hard-to-degrade organics, such as VOCs, including BTEXs. These findings shall be taken with the caveat that UST operating conditions must be optimized to remove specific contaminants.

3.3. Effects of Wastewater Constituents on Organics Removal

Hydroxyl radicals (produced by high-power and low-frequency ultrasound) in combination with sulfate and chlorine radicals can change harmful pollutant matters into intermediate substances, which can react with other oxidizing agents or these radicals and become mineralized as CO_2 , H_2O , and other inorganic minerals [16, 48]. However, in many studies, the enhancement of organics' degradation efficiency is attributed to producing more hydroxyl radicals due to higher frequencies, which produce more frequent and intense bubble collapse. However, according to Patidar & Srivastava [43, 49], there appears to be an optimal electrolyte concentration, above which the effectiveness of oxidation of pollutants is impaired [50]. For example, it was found that once the concentration of Na_2SO_4 exceeded 1.5 g/L, the degradation of organic contaminants in cosmetic wastewater started to decline [49]. The explanation is that at higher concentrations, sulfate and hydroxyl radicals might combine to produce persulfate, which in turn also contributes to the scavenging of hydroxyl radicals to produce other non-reactive compounds [43]. Similar trends were reported by Thokchom et al. [51] in that higher chloride concentration was held to promote the generation of substantial amounts of unwanted chlorine species (i.e., ClO_2^- , ClO_3^- and ClO_4^-), which consumed hydroxyl radicals to generate species with lower oxidizing potentials, thus reducing the degradation efficiency.

In the previous section, it was demonstrated that for high-strength wastewater, the removal efficiency of COD reached a maximum of 69.5%; the TOC removal was a minute 30.9% and was found to occur at 16 kHz frequencies and 500 ml/min flow rate. Figure 7 shows the reduction in TDS and metals from the high-strength wastewater after sonication at 16 kHz and different flow rates. This reduction indicates that much of the ultrasound energy is consumed in other reactions, which explains the low removal efficiency of organics [16, 24, 30, 52]. Apparently, metals are complexed at varying percentages and different flow rates, reducing their concentrations, especially at 2280 ml/min. The reason could be related to the mixing conditions leading to chemical reactions, which may complex the metals to different chemical forms.

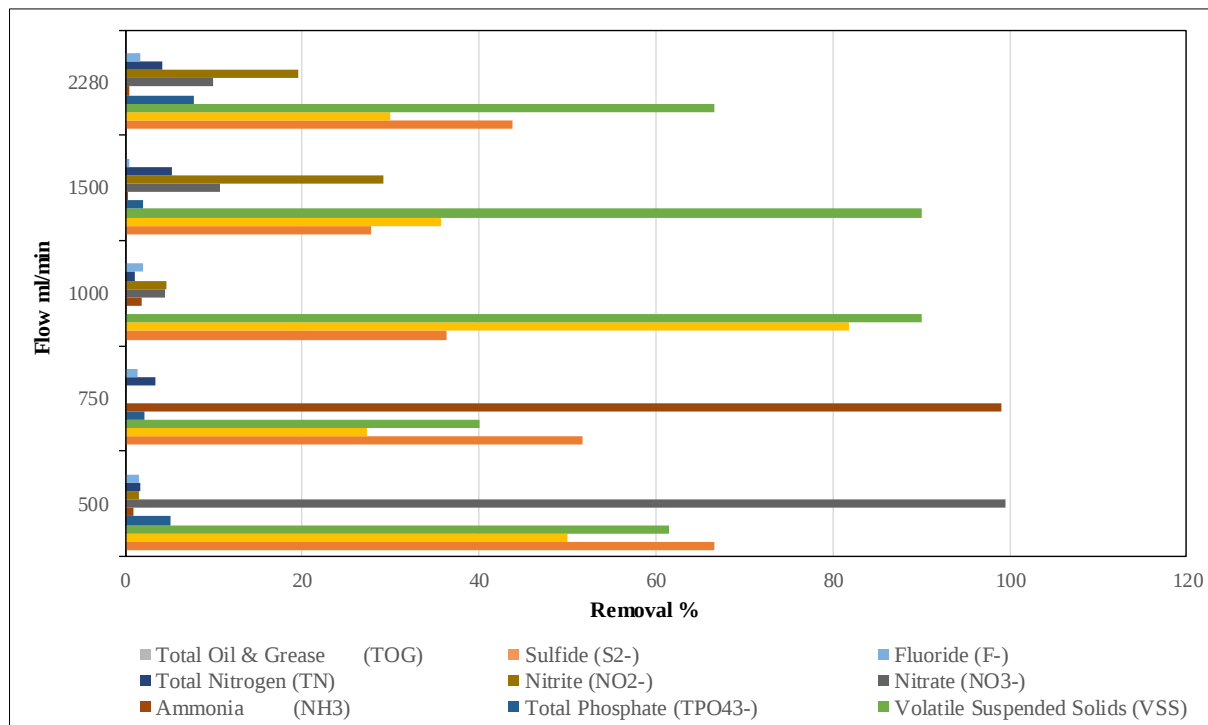


Figure 7. Reduction in total dissolved solids and metals from high-strength wastewater at 16kHz and different flow rates

Figure 8 shows the TDS results of the sonicated high-strength wastewater at a frequency of 16 kHz at different flow rates. The removal efficiency was significant except for fluoride, sulfides, and oil and grease (O&G). In particular, TSS was reduced by 3.9%, and TDS was reduced by 81.0%. This reduction in solids was attributed to the disintegration of TSS, which increased TDS concentration, resulting from the mechanical energy produced by the ultrasound waves [19, 24].

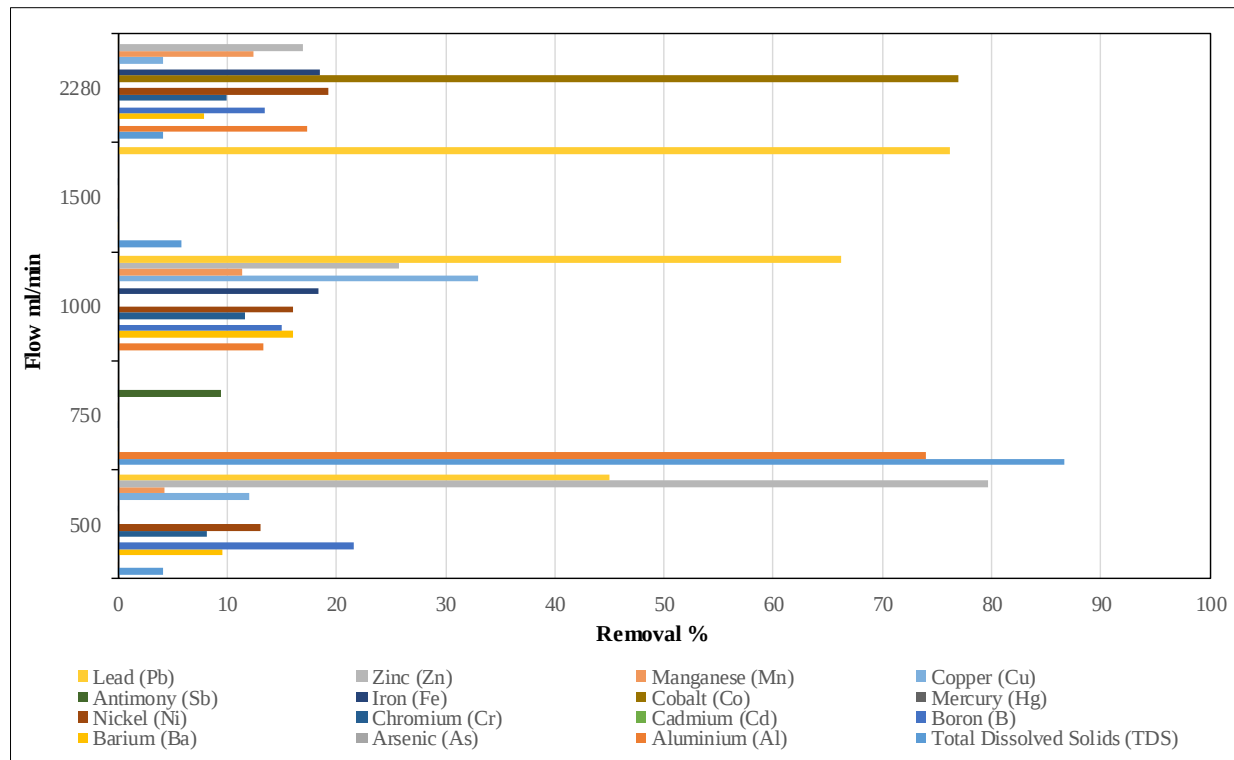


Figure 8. Removal of selected parameters from high-strength wastewater under 16 kHz at different flow rates

It was reported by Gujar et al. [4] that at 20 kHz, a COD reduction of 86% was associated with a TDS reduction of 47%. They recommended a combination with Fenton to achieve a 92% COD reduction. In addition, the conversion of TSS to COD has been reported in their study [46]. Therefore, these changes are indeed associated with the removal efficiency of organics, such as COD and TOC, and will be explored further using Pearson's correlation analysis [35, 36] performed on the organic removal rates and wastewater constituents as well as the operating conditions. The results are reported in Table 3.

Table 3. Pearson's Correlation of COD and TOC removal with other constituents at different conditions

Parameter	Frequency	Pearson's Correlation	Residence Time				
			1	2	3	4	5
COD	16	+ve	COD, M	COD, M	COD, M	M	TDS, PH
		-ve	TDS, PH	TDS, PH	TDS	pH	COD, M
	20	+ve	COD, M	M	-	-	pH
		-ve	pH, TDS	pH	pH	pH, TDS	COD, M
	16/20	+ve	COD, M	COD, M	pH, TDS	pH, TDS	pH, TDS
		-ve	TDS, pH	TDS, pH	COD, M	COD, M	COD, M
TOC	16	+ve	TOC, M	TOC	pH, TDS	TOC, M	TOC, M
		-ve	pH, TDS	pH	TOC, M	pH	pH
	20	+ve	pH, TDS	TOC	TOC, M	TOC, M	TOC
		-ve	TOC, M	pH	pH, TDS	pH, TDS	pH
	16/20	+ve	TOC, M	TOC, M	pH, TDS	pH, TDS	pH, TDS
		-ve	pH, TDS	pH, TDS	TOC, M	TOC, M	TOC, M

Table 3 demonstrates that the initial concentration of metals (M) and organics are mostly positive influencers, while TDS and pH have mixed effects. Also, as the flow increases, the concentration of organics, along with the pH, switches

to a negative influence. Finally, while TDS and pH tend to have adverse effects, they are positive influencers in limited instances at higher flows. These observations are valid irrespective of the frequencies used. These observations confirm earlier results [8, 23, 28, 40].

In general, ultrasound has been recommended as pre-treatment [47, 53]. However, while some studies used ultrasound for disinfection only (tertiary stage) [54-56], others recommended use as a tertiary treatment for the removal of biological and organic pollutants as well [57]. The findings of this paper revealed a critical observation; although the removal of organics is not the sole objective in the primary treatment of wastewater treatment plants and while the main aim of the chemical treatment at the primary stage is to remove metals and TDS as they are not removed efficiently in the biological treatment stage, another added value of UST (in addition to chemical-use reduction) when used at primary stage (on raw wastewater) is the possible elimination of the need for chemical treatment. Therefore, a strategy would be to identify the best scenario under which the maximum metal and TDS removal occurs depending on pH, metal content, TDS, and organics load.

4. Conclusion

This paper evaluated the usage of UST for high-, medium-, and low-strength industrial wastewater. In general, UST could eliminate the need for the primary treatment stage for high-strength wastewater, as the UST effluent is similar to the effluents of the primary and secondary treatment altogether in terms of COD. Using UST for effluent treatment could further reduce the concentration of some hard-to-degrade organics, including VOCs and BTEXs. Note that this conclusion shall be taken with the caveat that the operating conditions of UST need to be optimized to remove specific contaminants. Additionally, as evident from the results, adequate mixing and high frequencies seem to be the most appropriate conditions for using ultrasound for COD and TOC removal from high-strength wastewater; higher frequencies and less mixing could maximize COD and TOC removal from medium-strength wastewater, while for low-strength wastewater, it seems that lower dual frequencies would achieve the highest removal with even lower mixing conditions.

In general, it was found that in the case of high-strength wastewater, the removal of the organics depends mainly on the frequency. In contrast, it depends on mixing conditions for medium- to low-strength wastewater. However, as the COD is relatively easier to degrade compared to TOC, as demonstrated by the results, it appears that it requires rigorous mixing to remove it from high- and medium-strength wastewater, while parable TOC removal could only be achieved under the same frequency and flow conditions as COD in the case of low-strength wastewater. Finally, it was found that the concentrations of metals and organics are mostly positive influencers on organics removal, while TDS and pH have mixed effects. At higher flows, the concentration of organics switches to a negative influence on organics removal together with the pH. While TDS and pH tend to have adverse effects, they are positive influencers in limited instances at higher flows. The main finding is that dual frequency (16/20 kHz) achieved similar TOC removal as higher frequencies (37 and 80 kHz) in a shorter time (30 min compared to 1 hr in the case of higher frequencies).

5. Declarations

5.1. Author Contributions

Conceptualization, M.A.; methodology, M.A., M.Kh., and R.A.; software, M.A. and M.Kh.; validation, M.A. and M.Kh.; formal analysis, M.A. and R.A.; investigation, M.A.; resources, M.A. and H.A.; data curation, H.A.; writing—original draft preparation, M.A.; writing—review and editing, M.A. and M.Kh.; visualization, M.A., M.Kh., and R.A.; supervision, M.A.; project administration, M.A.; funding acquisition, M.A. All authors have read and agreed to the published version of the manuscript.

5.2. Data Availability Statement

The data presented in this study are available on request from the corresponding author.

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5.4. Conflicts of Interest

The authors declare no conflict of interest.

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Appendix I: Plates of the Ultrasound Testing Equipment



Plate A1. Ultrasound testing bench



Plate A2. Ultrasound flow-through reactor



Plate A3. Ultrasound flow-through reactor dual frequency transducers