539 Egypt. J. Chem. Vol. 66 No. 11 pp 443 - 451 (2023)

Egyptian Journal of Chemistry
http://ejchem.journals.ekb.eg/

Modified Ultrasonic Irradiation reactor: Application on Produced Water Treatment

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Abstract

The world faces a huge lack of resources. Produced water is the most significant waste stream in the petroleum industry. Ultrasonic technology is a very promising technique to treat produced water in the hope of reusing it instead of wasting it. In this study, experiments were carried out using a bench-scale ultrasonic reactor for the oxidation of organic matter in produced water, which was collected from a natural gas processing field (a real sample). At atmospheric pressure and room temperature, ultrasonic waves (40 kHz) were applied in a batch system for 30, 60, and 90 minutes at pH values of 7.5, 5, and 2.5. The results showed that at optimum operating conditions at steady state case (pH 5, time 90 min) showed that the removal efficiency of COD was 55%. And by applying the coagulation process as a pretreatment by using ferrous sulphate as a coagulant before sonication, then applying ultrasonic at the optimum point, the experiment showed the COD removal efficiency was 70%

Keywords: Advanced Oxidation, Coagulation, COD Removal, Ultrasonic Irradiation, Sonication, , Wastewater

1. Introduction

The environment is greatly impacted by wastewater. Particularly because of its highly poisonous and caustic components and produced water, which is waste created by the oil and gas industry. Industrial wastewater discharges can occasionally have disastrous effects on both the environment and the human population. The lives of humans may be in danger from pathogens, germs, and viruses present in wastewater. Additionally, hazardous chemicals pose a serious threat to people. Depending on the field's geographic location, geological formation, and kind of hydrocarbon product being produced, Mineral salts, organic substances, inorganic metals, radioactive substances that occur naturally, chemical additives, and other byproducts may all be present in produced water [1,2].

Despite the fact that produced water from oil and gas wells is typically regarded as a large volume, high salinity waste stream, it has the potential to be used to balance out water needs and over-allocation of water supplies [1, 2]. Currently, conventional oil and gas operations in the United States reuse around 45 percent of the produced water created by onshore activities. This water is injected into formations to improve recovery. Using "water flooding" or "steam flooding," enhanced recovery techniques sustain pressure in the formation while assisting in sweeping more oil to the production well. These processes require both normally produced water and supplementary water [2 ,3]. A few specific applications for produced water include cattle irrigation, stream augmentation, and the irrigation of particular crops. Currently, less than 1% of produced water is repurposed in this manner. Further study may also make wider applications feasible and economical. Drought alleviation, fire prevention, dust control, irrigation of additional crops, irrigation of public access areas like golf courses and parks, industrial cooling or process water, mining, municipal water requirements, and recreational purposes are a few examples of potential applications. Since produced water typically needs to be carried farther and processed more thoroughly, beneficial reuse outside the oil and gas industry is typically less economically appealing than reuse within the business[4, 5].

It could be necessary to treat produced water in order to satisfy regulatory requirements for pre-

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Receive Date: 16 September 2022, Revise Date: 12 December 2022, Accept Date: 18 December 2022
DOI: 10.21608/EJCHEM.2022.163251.6989

disposal or to satisfy requirements for beneficial usage. Numerous types of treatment technologies have been developed to handle generated water since it may contain a wide range of contaminants and its concentrations can vary greatly [6]. In order to successfully remove a variety of contaminants that would not be eliminated by a single process, a good water treatment system typically consists of many distinct types of individual unit processes performed in sequence. We'll concentrate more on organic removal in this study.

According to their capacity for biological breakdown, the organic contaminants in wastewater can be classified into two groups: non-biodegradable and biodegradable. It is straightforward for the environment to breakdown organic contaminants with simple structures and good hydrophilicity. Bacteria, fungi, and algae may be able to break down these organic contaminants, including polysaccharides and methanol. However, several of them, such as acetone and methanol, could cause acute toxicity when present in wastewater in large quantities [4, 6].

On the other hand, persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and dichloro-diphenyl-trichloroethane (DDT), are metabolised or otherwise eliminated very slowly. Although they are less hazardous and have a lower concentration in wastewater than soluble organic pollutants, they can persist for decades in sediment before being absorbed into the water supply and entering the food chain [7,8]. Numerous physicochemical techniques have been researched for the treatment of wastewater [10]. Adsorption, electrochemistry, sorption employing waste material degradation, and ozonation with degradation are some of these [8,9].

A revolutionary enhanced oxidation method called ultrasound irradiation has been developed in response to the increased demand for wastewater with fewer pollutants [10-13]. Ultrasonic irradiation of generated water is classified as a chemical oxidation process. The water is subjected to ultrasonic vibrations, which cause cavitation and the separation of OH radicals, which oxidise the organic components. Additionally, complicated organic compounds can be broken down into much simpler ones by ultrasonic waves, making them easier to oxidize. There are essentially three ways to introduce ultrasound into a system that reacts. First, by submerging the apparatus in a liquid that has been sonicated. Two: by immediately submerging an ultrasonic source within the reaction liquid. Third, by having a reactor with walls that vibrate ultrasonically [14,15].

When the Curies discovered the piezoelectric effect in 1880, it laid the groundwork for the current generation of ultrasound [16]. The first description of the cavitation phenomenon was published in 1895 [17]. Since the 1920s, ultrasonic microbe destruction has drawn a lot of attention. There are no byproducts produced by ultrasonic technology or additives added to the ultrasonic system. Therefore, this technology is not expected to cause any environmental issues. [13]. While many other processes suffer as the amount of suspended solids in the effluent increases, in the US, an increase in turbidity or suspended solids may actually increase efficiency [18].

A comprehensive study of the advantages of employing ultrasonic cavitation has been conducted over the past ten years. Although it has been demonstrated that the technique is a cutting-edge, efficient, and small-scale solution for treating water and wastewater, Given the form of microorganisms, the various types of pathogens in water and wastewater, high performance, and the lack of environmental concerns, ultrasonic technology has the following advantages: additional operating costs are associated with the maintenance and/or replacement of instruments that continue to be harmed by ultrasonic activity itself [13, 15].

Maleki A., et al. studied phenol degradation at various phenol concentrations under irradiation for 5 hours. For 300 minutes of sonication of a 100 mg/L phenol solution, only 13% phenol degradation was seen [19]. According to Naseri S., et al., the majority of COD removal was completed within the initial sonication time, and removal efficiency did not significantly improve over time. 130 kHz performs better at organics removal from secondary effluent than the lower frequency. 6 kHz efficiency. About 2.5 times as much H202 forms at 130 kHz as it did at 35 kHz. The elimination effectiveness of suspended COD was higher at 35 kHz compared to TCOD. The efficiency of removing all COD was estimated to be between 17 and 28%. It is easier to remove suspended COD than soluble chemical oxygen demand (SCOD) [20]. In eight distinct samples (200, 1000, 2000, 3500, 5500, 6500, 10000, and 17000 CFU/ml), Dehghani M., et al. studied the decrease of fungus in sewage using an ultrasound reactor. As the period of disinfection increases, the number of fungi decreases. The results demonstrated that fungus reduction is significantly impacted by lengthening the disinfection process. Additionally, fungi do not significantly decrease after less than 15 minutes of exposure to 42 kHz, but large levels of reduction are anticipated after longer times (99.92%). They hypothesised that USRT (Ultrasound Reactor Technology) operating at a frequency of 26 kHz could partially inactivate fungal cells. It can be demonstrated that experiments at 42 kHz are more successful than those conducted at lower frequencies [21]. In a laboratory setting, Mahvi A. evaluated the use of ultrasonic irradiation to control the algal population. The results showed that a brief exposure to ultrasonic irradiation collapsed algae gas vacuoles, which resulted in a loss of buoyancy and regulating ability and, as a result, localised the cells. 8.55, 35.22, 67.22, 90.67, and 100% of the algae population were eliminated after 30, 60, 90, 120, and 150 seconds of sonication, respectively [13].

The average optimal conditions for TCOD elimination in this study were obtained through these earlier experiments. The following statement sums up our circumstances:

1. Degradation occurred faster in acidic conditions than in alkaline conditions (best pH range: 3 to 6).
2. The rate of ultrasonic deterioration was initially considerable but afterwards significantly decreased (average best time from 60 min to 90 min).
3. The ideal frequency range is 35 to 200 kHz.
4. The aeration system for more DO will help form more hydrogen peroxide (H_2O_2)(oxidant).
5. Power ultrasound is used as a chemical process (sonochemistry) where integrated coagulation and flocculation technology are used.

This paper aims to make a design for an ultrasonic irradiation reactor and study its effect on COD removal efficiency for the produced water. An ultrasonic cleaning bath is used at 3 different time intervals (30, 60, and 90 minutes) and at 3 different pH values (2.5, 5, and 7.5). In addition, the effect of coagulating water as a pretreatment before ultrasonic irradiation is being studied.

2. Materials and methods
2.1 Materials

A sample of wastewater from one of the northern Egyptian gas fields. The characteristics of the produced water sample and discharge limits in industrial areas according to Egyptian Environmental Law, decree 44/2000, are shown in Table (1). To alter pH, hydrochloric acid was utilized. Ferrous sulphate [Fe $(SO_4)_7H_2O$] acts as a coagulant. The purity of all compounds acquired from Sigma Aldrich was 98%.

Table 1 Main physical and chemical characteristics of produced water (real sample) and discharge limits [decree 44/2000]

Parameter	unit	influent	Consent standards (decree 44/2000)
pH	–	6.3	6-9.5 (range)
COD	mgO_2/L	8080	Less than 1100
BOD_5	mgO_2/L	4848	Less than 600
Total sulphides	mg/L	0.1	Less than 10
Total phosphorus	mg/L	12.4	Less than 25
Oil and grease	mg/L	79	Less than 100
TDS	mg/L	37 000	–
TSS	mg/L	429	Less than 800

COD, chemical oxygen demand; TDS, total dissolved solids. BOD_5, Biological Oxygen Demand; T.S.S, Total Suspended Solids

2.2 Transducer Design

The properties shown in table (2) have been employed with the piezoelectric ceramic transducer. The constant, uniform amplitude of this type makes it ideal for industrial use.

Table 2 Characteristics of Transducer

Ultrasonic power	80w
Frequency	40
Volt	AC 220-240 V 50Hz

The most recommended tank material is stainless steel type 304 (frequently referred to as 18-8 stainless) from the austenitic group. with a full capacity of 2.0 L and a size of 150*100*100 mm. First, a welding gun was used to fix the point nail on the stainless-steel tank. Each point's nail distance is about 5-8 cm. Use AB glue to evenly coat the stainless steel and transducer as shown in figure (1, A). Securing the transducer has been done with the hexagon tool to ensure there is no gap between the transducer and the stainless-steel tank, as shown in figure (1, B).

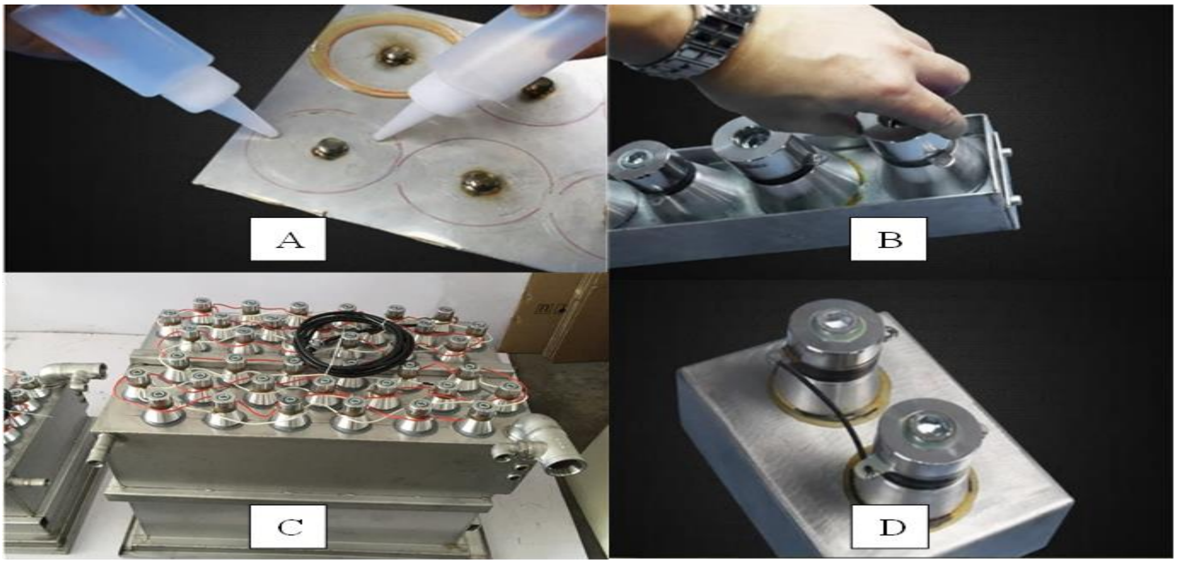

Figure (1) A) Fixing the point nail on stainless steel tank, B) Securing the transducer with the hexagon tool, C) Industry scale of ultrasonic reactor, D) Connecting the transduce in parallel

The positive and negative poles of the transducer must be accurately identified, as shown in figure (1, C), and the transducer must be linked in parallel. The drive generator may sustain damage if the transducer's positive and negative poles are switched around. Large numbers of transducers would be utilized on an industrial scale, and they should be connected as indicated in figure (1, D). The circuit was simulated with an output of 40 KHz for the purpose of designing the ultrasonic driver circuit, as illustrated in figure (2). We used a mechanical timer and a thermostat in our circuit to manage batch duration and temperature precisely. For the safety of the circuit and keeping in mind the cooling system for the circuit so it won't overheat, the circuit and the tank have finally been assembled in a stainless-steel box.

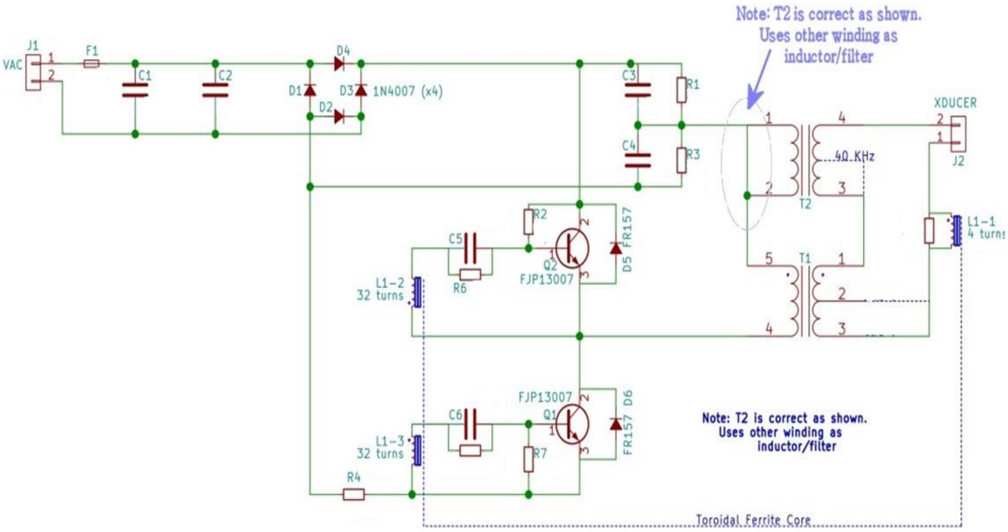

Figure (2) Ultrasonic driver circuit simulation

2.3 Experimental Work

The process's ultrasonic equipment is shown in Figure 3. First, using hydrochloric acid, the pH metre needed to be adjusted to the correct pH. For the aeration system, the air compressor and ultrasonic cleaning bath were connected. A 500 cc sample of petroleum was added to the ultrasonic cleaning solution. Then, as illustrated in Figure(4, A), the batch system was started for 30 minutes at room temperature. A sample was collected to use the titration test to measure COD removal. The influences on this process were investigated. The time is set to 30, 60, and 90 minutes, and the PH is set to (7.5 - 5 - 2.5).

Figure (3) The apparatus required for ultrasonic process

The experiment was repeated for a waste water sample by coagulation as a pretreatment method using Ferrous sulphate [Fe $(SO_4)_7H_2O$] with doses of 80 mg/l at 350 rpm mixing speed as shown in figure (4, B). The sample was settled for 150 min. before ultrasonic step. pH was determined using a pH-meter type Schott Greate CG820. COD for the waste water sample was determined using the Spectrophotometer; Direct Reading; Dr/2000; Hach, and the reactor with model HI839800 Reactor; Hanna instruments.

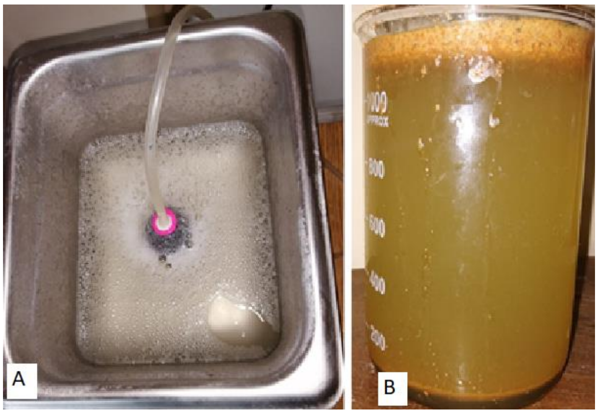

Figure (4): A) produced water (real sample) in ultrasonic cleaning bath with aeration system, B) The produced water (real sample) after coagulation

Figure (5) shows the stages of the treatment process for the produced water. The process begins with the addition of the coagulant, Ferrous sulphate [Fe $(SO_4)_7H_2O$], with doses of 80 mg/l, then adding Flocculant material (poly- electrolyte), so that the efficiency of the treatment process is improved. The Effect of this process on COD removal% was studied. The water resulting from the treatment process is collected in settling tank from there to sand filter. Finally, the disposed of treated produced water occur in accordance with the Environmental Sanitation Law

3. Result and discusion

3.1 The Reaction Mechanism

The CF-Ultrasonic processes (coagulation, flocculation) generated -OH species with strong oxidation power. The subsequent responses provide an explanation for this:

$$\mathbf{2H2O} \rightarrow \mathbf{2OH} + \mathbf{2H^+} + \mathbf{2}e^- \qquad \text{Equation (1)}$$

$$\mathbf{OH^-} \rightarrow \cdot\,\mathbf{OH} + \mathbf{e^-} \qquad \text{Equation (2)}$$

$$\mathbf{H2O} \rightarrow \cdot\,\mathbf{H} + \cdot\,\mathbf{OH} \qquad \text{Equation (3)}$$

The free radicals in OH are powerful oxidants. They are able to remove a variety of organic and inorganic contaminants. As a result, the COD and turbidity contents will decrease.

3.2 Effect of Time on COD Removal Efficiency

As shown in figure (6), Using this method at a wavelength of 40 kilohertz produces the best removal efficiency after 90 minutes, which is 40.59%, and at 120 minutes, the COD removal efficiency remained constant, according to the experiment's results on the relationship between the COD removal efficiency and the treatment period. While adopting another innovation, an "electrochemical cell," the steady state COD removal efficiency for an actual sample of generated water achieved 66.52% within one to two hours [4] . So, these results using the ultrasonic technique is more efficient than other methods.

3.3 Effect of pH on COD Removal Efficiency

Figure (7) illustrates the results of the study on the influence of pH on COD removal effectiveness for an actual sample of generated water using ultrasonic irradiation at 40 KHz. For economic considerations, the best efficiency is 54.45% at 90 min. and pH 2.5–5. The oxidation reaction that results from the use of this technology and the oxygen added during the air pumping process causes the Fenton reaction, which is the best operating condition for it at pH in an acidic medium. The higher the COD removal efficiency, the lower the pH. This is because the Fenton reaction is formed for the oxidation of organic materials, and the

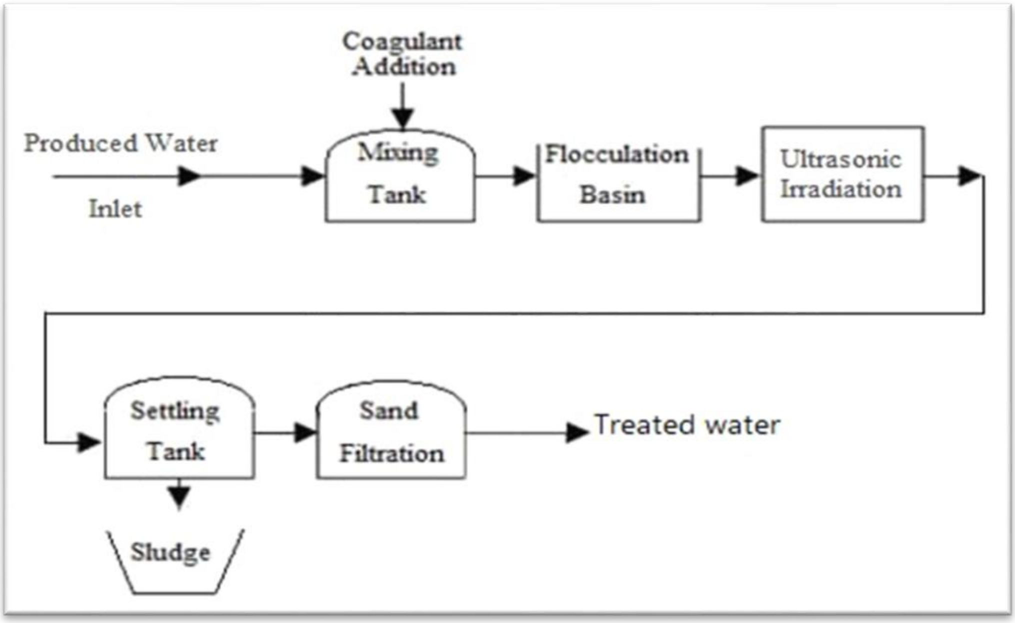

Figure (5): Scheme of produced water treatment with ultrasonic irradiation with coagulation

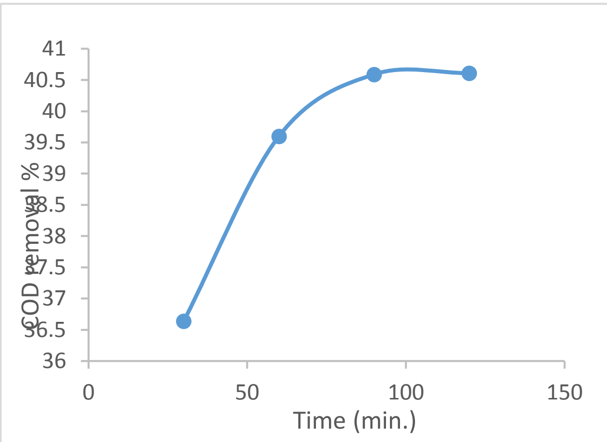

Fig. (6) COD removal efficiency% at 40 kHz

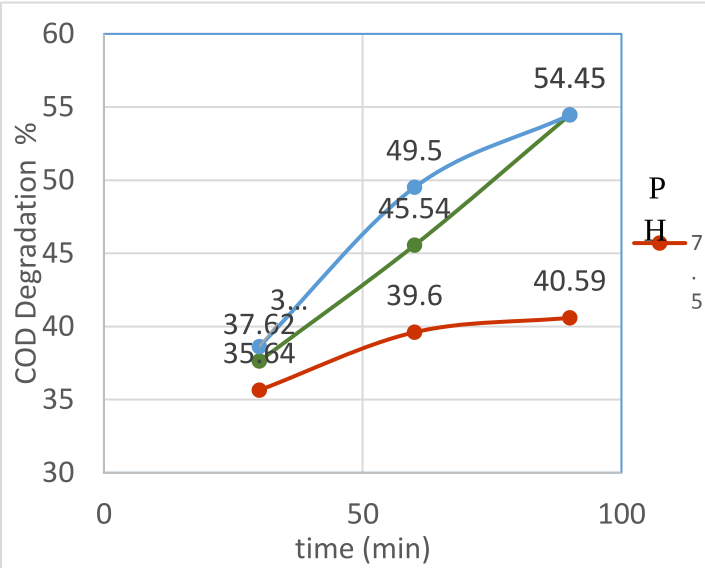

Fig (7). Cod Removal Efficiency At Different Ph Values At 40 Khz

best operating condition for it is at pH in an acidic medium. The greatest choice in this condition is pH 5, as it won't require a lot of caustic soda to treat it. In this regard, pH, hydrogen peroxide, and iron concentrations derived from the first-stage coagulation process all affect how effective the process is. Because of the chemistry of iron, the pH must be acidic for the Fenton reaction to take place without the iron precipitating as hydroxide [22] .

3.4 Effect of Coagulation Pretreatment on COD Removal Efficiency

The effectiveness of COD removal was examined using figure 8 to examine the impact of the coagulation process as a pretreatment method. An optimal condition ultrasonic irradiation procedure was used to treat a sample from the coagulation process (pH 2.5 and 90 min for sonication). It was found that COD removal efficiency after the coagulation process is higher than COD removal efficiency without the coagulation process. The best efficiency is 70.29% with the coagulation process at 90 minutes and pH 2.5. The coagulation process, which reduces suspended materials and contaminants as well as some inorganic and organic components by manipulating the electrostatic charges of particles suspended in water, increased the effectiveness of COD removal before the treatment procedure utilising ultrasonic irradiation. In order to destabilise the charges on particles, colloids, or oily materials in suspension, this procedure inserts small, highly charged molecules into water. As a result, the organic and inorganic loads are reduced, increasing the effectiveness of the treatment process [23].

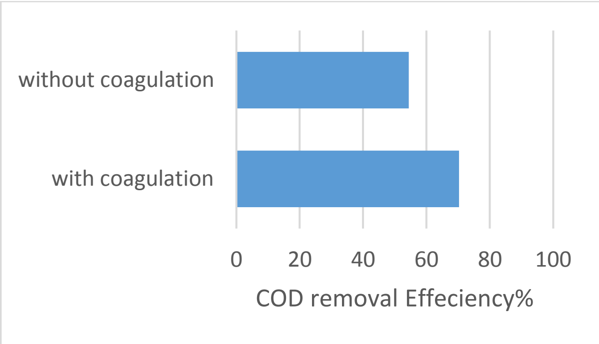

Figure (8) COD optimum removal efficiency with and without coagulation pretreatment

3.5 Comparison of produced water treatment using Ultrasonic techqniue and other electrochemical cells

Table 7 summarises the results that we obtained from the treatment of a real sample of produced water using ultrasonic irradiation technology with a coagulation process and compares them with other techniques of treatment, such as using electrochemical cells with different methods. The treatment time using ultrasonic irradiation technology is much better than the treatment that was carried out in Rajkumar & Palanivelu (2004) [24] and Yavuz et al. (2010) [25], despite the fact that the samples used for the treatment in this previous research were synthetic samples. On the contrary, with the research in Omar E. et al. (2018) [4], where the treatment time is one and a half times less than the treatment time using ultrasonic irradiation technology, the concentration of COD in our real sample is 65% higher than the concentration of COD in the real sample used in this previous research. When compared to Wahbi Al-Ameri, et al. (2022) [26], the treatment duration is reduced for a sample with a low COD concentration.

According to COD removal efficiency, It was achieved at 70% using ultrasonic irradiation technology for a real sample of petroleum factories, which contain a very high concentration of COD (8080), compared to Rajkumar & Palanivelu (2004) [24]. Comparing the previous survey [25] (for the synthesis sample) and our results (for the real sample), COD Removal Efficiency% have the same. Our results is considered better in terms of COD removal efficiency than the previous survey [4] with 3.5%. When compared to Wahbi Al-Ameri, et al. (2022) [26], the COD removal efficiency for their sample is better than ours due to the fact that the concentration of COD in our sample is very high for the sample used in this research. Therefore, the use of ultrasonic irradiation technology with the coagulation process is much better compared to other previous research in terms of COD removal efficiency and treatment time. It is worth noting that there is no use of any chemicals in this method.

4. Conclusion

- The world faces a huge lack of resources, so we need multiple ways to maximise the use of produced water after treatment, which is the main source is oil fields.
- Experiments were carried out using a bench-scale ultrasonic reactor, which is the new innovation for the oxidation of organic matter in produced water, which was collected from a natural gas processing field (real sample).
- The use of ultrasonic technology has many advantages, the most important of which is reducing the use of chemicals used for the treatment process and thus reducing the formation of sedimentary solids (sludge), which requires safe disposal processes as well as high costs for the safe disposal process.
- The optimum operating conditions for our bench scale experiment were achieved by applying ultrasonic waves (40 kHz) in a batch system at a time of 1 hour, pH value of 5 , at atmospheric pressure and room temperature .
- The best removal efficiency for COD using the ultrasonic reactor at the optimal operating conditions for one stage was 55%, and the total removal efficiency for COD rose to 70% using a pretreatment stage by the coagulation method.
- The treatment method using the ultrasonic reactor helped in treating the produced water (real sample) with high concentrations of 8080 mg/liter to reach the concentrations required (low concentration) by environmental law for safe disposal or reuse again.

Techqniue Used	Treatment Time (hr)	System	COD Removal Efficiency%	Reference
Electrochemical cell used Ti/TiO2RuO2IrO2 oil refinery	20	Batch	92	Rajkumar & Palanivelu (2004) [22]
Electrochemical cell used Ru-MMO as electrode (EO process) petroleum refinery	3.5	Batch	70	Yavuz et al.(2010) [23]
Electrochemical cell using Graphite granules anode and flow-by technique	1	Continues	66.52	Omar E. et al. (2018) [19]
using graphite electrodes	1.33	Batch	98	Wahbi Al-Ameri, et al (2022) [24]
Ultrasonic irradiation method with coagulation step as a pretreatment process	1.5	Batch	70	Our experiment results

Table 7 Comparison Of Produced Water Treatment Using Ultrasonic Techqniue And Other Electrochemical Cells

5. Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

6. Formatting of funding sources

No Funding

7. Acknowledgments

We appreciate the help from all the practical helpers who helped us achieve these results, as well as the hard work of the Canal Higher Institute for Engineering and Technology students in suez who collaborated with the authors.

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