Journal of Chemical Health Risks

www.jchr.org

JCHR (2024) 14(1), 3349-3357 | ISSN:2251-6727

Advanced Processes for Pharma Wastewater Treatment: A Mini Review

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(Received: 26 November 2023 Revised: 21 December Accepted: 6 March 2024)

KEYWORDS
Chemical Oxygen Demand, Environment, Pharmaceuticals, Cavitation, Wastewater

ABSTRACT:
Water is a limited resource that is essential for long-term development. In recent years, industrialization has exponentially increased; the pharmaceutical industry has highly profitable growth while also contributing to environmental damage. Many Pharmaceutical industry discharges wastewater into the surrounding containing complex persistent organic compounds which are highly harmful to aquatic life and people's health. Ultrasonic Cavitation is one of the methods that are used to treat wastewater and reduce the number of harmful substances that are released into the water bodies. In order to attain maximum Chemical Oxygen Demand reduction, present review demonstrates a combination of ultrasonic cavitation method using different oxidizing agents is a practical choice to decrease the hazards from wastewater discharged through pharmaceutical companies.

1. Introduction

One of today's major worldwide challenges is environmental destruction [1]. One of the main issues for developing nations like India is industrial wastewater and other hazardous releases from companies. Industrial waste water is the primary cause of one-third of India's maritime pollution and natural water body contamination [2]. Currently, large quantities of pharmaceutical compounds are utilized to prevent and treat illnesses in both humans and animals, resulting in the production of large volumes of wastewater from pharmaceutical industries [3]. The use of ultrasonic waves is one of the creative techniques that has been utilized to enhance the water treatment process. Extensive treatment methods are needed to reduce the level of pollutants to the specified disposal limits since the wastewater generated in the bulk pharmaceuticals manufacturers is complicated. As a result of the large chemical and total oxygen demand of produced effluent, a single treatment system or a combination of conventional processes may not be enough to fulfil the advised discharge limits [4,18]. Manufacturers of new products, particularly antibiotics, have significantly magnified waste treatment and disposal issues. In order to treat patients effectively, it is therefore of utmost importance to establish innovative treatment facilities. The discharge of many chemicals into the environment, such as antibiotics, contraceptives, antiseptics, antivirals, antipyretics, and analgesics, is increasing pollution rates daily Figure 1. In pharmaceutical wastewater, which is adverse for the environment, includes drugs like Acetaminophen, Chemical Oxygen Demand, Aspirin, Caffeine, Amoxicillin, etc. The primary elements of complex pharmaceutical wastewater are the bacterial noxiousness, large salt and carbon-based material concentrations which are tough for biodegradation. Despite several treatments, there are still significant amounts of suspended particles and dissolved organic debris. To balance environmental safety, many procedures are used to treat pharmaceutical wastewater, including

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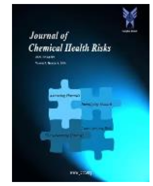
JCHR (2024) 14(1), 3349-3357 | ISSN:2251-6727



coagulation, sedimentation, membrane separation, Advanced Oxidation Processes, active carbon adsorption, and biological treatment, from our finding we come to know that acoustic Cavitation is the best technique to treat pharmaceutical waste water [5,19].

2. Wastewater

In today's economy, it is essential to treat wastewater to decrease pollution in order to preserve water resources. The wastewaters from this industry are generally strong and may contain toxic pollutants [9]. To eliminate water contamination and solve the issue of water disposal, several techniques and conventional procedures can be used in order to achieve a balance of clean water; there are a variety of steps that must be taken, such as mechanical purification, aeration, and the removal of microbes. During manufacturing, the pharmaceutical industry uses a variety of reactants, solvents, and solid water, then the reactant discharge to effluent water containing bulk medicines that are harmful to the environment, people, and mostly aquatic life [8].

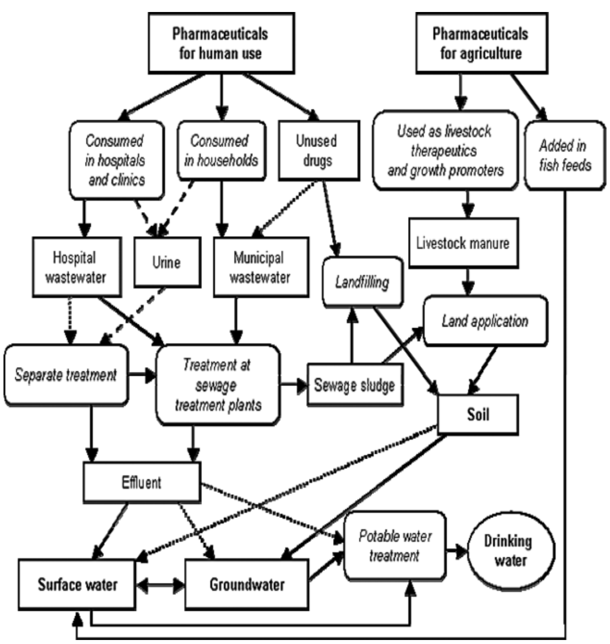


Figure 1: Ways of contamination of Pharmaceuticals (Ikehata et al., 2006)

3. Occurrence of Pharmaceuticals in wastewater effluents

Many pharmaceuticals have been found and detected in the marine ecosystem all over the world thanks to the use of sophisticated measuring technology [31,32]. Pharmaceuticals wind up in soil, surface waters, and finally land and drinking water after passing into the sewage system and through the influent of wastewater treatment plants, where they are excreted by humans or animals [33]. Pharmaceuticals reach the environment through a variety of routes. The average percentage of occurrence of different pharmaceutical compounds in wastewater is shown if figure 2. Pharmaceutical manufacturing facilities, wastewater treatment plants (WWTPs), hospitals, landfills, and even burial grounds are among the most common causes of pollution [34].

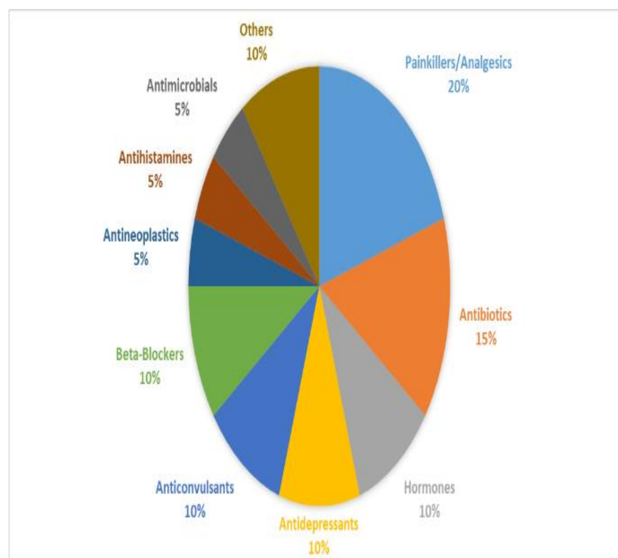


Figure 2: Occurrence of pharmaceuticals in wastewater

Human excretion of active pharmaceutical ingredients (APIs) conjugated to polar molecules which on cleavage released the initial API into the atmosphere [35]. The frequently observed pharmaceutical compounds in wastewater and their concentrations are shown in table 1.

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JCHR (2024) 14(1), 3349-3357 | ISSN:2251-6727

Table 1: Different pharmaceuticals in wastewater with their concentration

(Gomez et. al., Vieno et. al., Al-Rifai et al., 2007)

Sr. No.	Drug	Examples	Conc.
1	Central Nervous Drugs	Caffeine	3.2-11.44 µg/L
2	Analgesic	Acetaminophen	10-23.33 µg/L
		Ibuprofen	0.49-990 µg/L
		Carbamazepine	0.1-1.68 µg/L
3	Antibiotics	Sulfamethoxazole	0.02-0.58 µg/L
		Ofloxacin	6-52 ng/L
		Ciprofloxacin	6-60 ng/L
4	Cardiovascular Drugs	Metoprolol	10-390 ng/L
		Propranolol	0.05 µg/L
		Clofibric Acid	0.47-170 µg/L
5	Absorbable Organic halogen compounds	Iomeprol	1.6 µg/L
		Iopromide	0.026-7.5 µg/L

4. Cavitation as an Advanced Technology for treatment

Cavitation is the process of creating micro bubbles that continuously burst and then collapse over brief periods of time. It is the process that results in exceptionally favourable conditions for the degradation or eradication of contaminants. Pollutant degradation is a current issue that must be effectively addressed to achieve necessity for a healthier and harmless ambient atmosphere [6].

Categorization of Cavitation

A) *Acoustic Cavitation*: Using ultrasound, pressure changes in wastewater cause cavitation. This method typically uses ultrasound at a frequency of 16 to 100 MHz ultrasonic waves are used to cause the deterioration of the pollutants. Sonochemistry is the name given to the process since it combines chemistry and ultrasonic technology [7].

B) *Hydro-dynamic Cavitation:* It has caused through pressure changes in waste-water that are brought on by varying system velocity. The orifice meter, venturimeter, and other devices frequently exhibit these phenomena [8].

C) *Cavitation through Optic*: High intensity light photons that continuously break the composition of waste-water to produce cavitation. It uses short pulsed lasers to focus in a liquid solution, which causes impurities to degrade [8].

D) *Particle Cavitation*: To create Cavitation, an elementary particle beam is used. It helps rubber become more durable [8].

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5. Pharmaceutical wastewater

Several different forms of pharmaceutical medications used to treat humans and animals are becoming environmental pollutants. Different types of medications, including antibiotics, contraceptives, antiseptics, antivirals, antipyretics, and analgesics, are released into the surroundings on a daily basis and increasing pollution rates [10]. In pharmaceutical wastewater, which is hazardous to the environment, are drugs like Acetaminophen, Chemical Oxygen Demand, Aspirin, Caffeine, Amoxicillin, etc. The average usage of different pharmaceutical compounds on day to day life is shown in figure 3. The primary components of complex pharmaceutical wastewater are the microbial toxicity, high salt content, and high concentration of organic matter, all of which are difficult to biodegrade. Despite different treatments, there are still significant amounts of suspended solids and dissolved organic matter. To balance environmental safety, several procedures are used to treat pharmaceutical wastewater, such as coagulation, sedimentation, membrane separation, Advanced Oxidation Processes, active carbon adsorption, biological treatment, etc. [11].

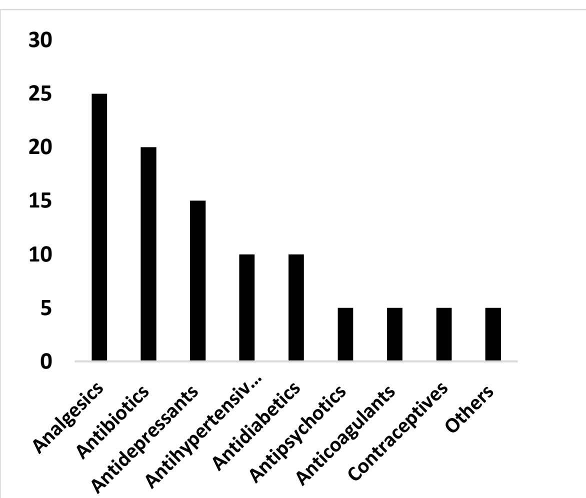

Figure 3: Usage of pharmaceutical compounds in different forms

In the 1980s, Advanced Oxidation Processes were first suggested as a means of purifying drinking water [12]. Both hydroxyl radicals and sulphate radicals, which are potent oxidizing agents, have the ability to quickly breakdown organic contaminants in wastewater, turning them into a less hazardous or even non-toxic end product in the process. Cavities are formed in the solution during ultrasonication, and when they collide or explode, a large quantity of energy is released that is used to break apart the complex chemical component. The degradation of organic pollutant is significantly affected by pH of the solution. Degradation was found to be more effective in acidic medium (pH 3.0-5.6) than in basic medium (pH 9.5-12.0). The Anaerobic Contact Process rate at pH 3.0 or 5.6 is 80% more than at pH 12.0. According to this study the best result is found at low pH range as the substrate is in non-ionized state [13].

Possibility for some degree of ultrasonic-induced germicidal effect on faecal coliforms in water. The faecal coliform kill was found to be more when the sonication time was increased. During Sono-oxidation of humic acids, these compounds were completely broken down in 60 minutes, whereas only 40% of Total Organic Carbon was reduced. Suspended Chemical Oxygen Demand is converted into Soluble Chemical Oxygen Demand during sonication process [14]. The integrated physicochemical methods for industrial wastewater treatment were found more impressive. Fenton-based methods were economically favourable for industrial wastewater treatment according to their studies [15]. The degrading efficiency of complex organic compounds is more when there is use of combination of Fenton and heterogeneous photocatalysts than traditional method of Fenton only. This combination can aid in converting Ferric ions (Fe^{3+} to Fe^{2+}) and improving the use of Ferric ion Fe^{2+} [16].

The transducer with a specified resonance frequency that can range from 20 kHz to 10 MHz and is powered by an amplifier often produces ultrasound. Due to this maximum formation of radical species take place, which favours the degradation of medicines, the highest degradations are seen between 200 and 400 kHz [17]. In Researched, hybrid treatment technique that is combination of Cavitation and Advanced Oxidation Processes were used for treatment of wastewater containing pharmaceutical compounds. The initial Chemical Oxygen Demand of this effluent ranged from 1000-1200 mg/l. Among the combinations for

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JCHR (2024) 14(1), 3349-3357 | ISSN:2251-6727

Acoustic Cavitation and Advanced Oxidation Processes as Acoustic Cavitation /Hydrogen Peroxide, Acoustic Cavitation/Ferrous (II)/Hydrogen Peroxide, Acoustic Cavitation/Potassium persulphate and Acoustic Cavitation / Ozone, the maximum Chemical Oxygen Demand reduction was observed as 83.53% for Acoustic Cavitation/Ferrous (II) / Hydrogen Peroxide. In this combination the Ferrous (II) to Hydrogen Peroxide ratio was 1:5. The ultrasonic power, frequency, pH and time period of sonication was 125 W, 22 kHz, 3, and 60 min respectively. Among the combinations for hydrodynamic Cavitation and Advanced Oxidation Processes as Hydrodynamic Cavitation/ Hydrogen Peroxide, Hydrodynamic Cavitation/Fe (II)/ Hydrogen Peroxide, Hydrodynamic Cavitation/Potassium persulphate, and Hydrodynamic Cavitation/Ozone, the maximum Chemical Oxygen Demand reduction Table 2 was found to be 87.4% for Hydrodynamic Cavitation/Iron (II)/ Hydrogen Peroxide. The system pH and inlet pressure were 3 and 4 bar respectively [18]. The Carried-out permutations and combinations of processes for treatment of real pharmaceutical waste water having different initial level of Chemical Oxygen Demand. For this they used a sonicator setup of power 200 W, horn tip 2.1 cm and frequency 22 kHz. The time period of sonication was 120 min. The ozone flowrate was 400 mg/h, Copper Oxide catalyst amount was 0.3 gm/lit as well for Fenton's reagent the ration of Iron (II) sulfate: Hydrogen Peroxide was 3:5 [19].

Table 2: Representation of percentage reduction in Chemical Oxygen Demand [19]

Advanced Process ↓	**% COD reduction** →	**Samples used (ppm)**			
		A (75 x 10^2 ppm)	**B (65 x 10^2 ppm)**	**C (30 x 10^2 ppm)**	**D (5 x 10^2 ppm)**
Only US		9%	11%	13%	14%
US and H_2O_2		29%	31%	33%	36%
US and O_3		30%	33%	35%	37%
US and $H_2O_2 + O_3$		62%	64%	63%	66%
US and Sono-Fenton		67%	69%	70%	73%
US and CuO + O_3		86%	89%	90%	92%

In a nutshell, the combination processes of ultrasonic + hydrogen peroxide + ozone reduced chemical oxygen demand (COD) by 73%, whereas the operation of ultrasonic, ozone and copper oxide catalyst reduced chemical oxygen demand by a high of 92% [19]. The Maximum degradation of carbamazepine concentrations for ozone dose range of 3.5-17.0 mg/L and Hydrogen Peroxide (10 mg/l) was achieved 76% by ozone alone and 62% under peroxone treatment, with ofloxacin and Ibuprofen achieving 31% and 41% degradation by ozone alone and under peroxone respectively [20]. The frequency range between of 28 to 1000 kHz the degradation of pharmaceutical compound was most effective at 580 kHz. According to this research at a relative high frequency, the number of Cavitation occurring in solution would be higher, allowing a large fraction of hydroxide ion to be

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ejected to the bulk liquid thus increasing the degradation efficiency from 28 to 580 kHz. Also, the optimum concentration of Hydrogen Peroxide used during sonication was found to be 5 mm, high temperature and acidic conditions were found favorable for effective degradation degree [21].

The treated wastewater from herbal pharmaceutical industries containing COD, BOD, and SS within 21.96-26.00 gm/lit, 12.2-15.66 gm/lit and 5.46-7.37 gm/lit respectively. This water was treated in 3 states as physicochemical treatment, secondary biological activated sludge process and then by tertiary Fenton's oxidation process. For Fenton's oxidation process the effluent received had gone through activated sludge process. The parametric values COD= 0.896-0.944 gm/lit, BOD=0.156-0.174 gm/lit and SS=0.066-0.074 gm/lit after Activated Sludge Process. This effluent was then treated by Fenton's oxidation process. The average optimum removals of organics in terms of COD, BOD, SS, and TOC were found to be 0.138 gm/lit, 0.02 gm/lit, 0.021 gm/lit, and 0.098 gm/lit at pH equal to 3.5, contact reaction time of sixty minutes with the dose of ferrous sulphate is 0.2 gm/lit, and dose for hydrogen peroxide of 6.0 ml/lit, respectively [22]. Treated industrial pharmaceutical wastewater with ultrasound cavitation + persulfate combination having ultrasound frequency was 20 kHz used in this study. The optimum parameters pH=5, amplitude intensity=20% and oxidant dosage=0.1gm/lit were observed with 39.5% COD has been removed in 60 minutes that is fixed processing time [23]. for a few decades, emerging contaminants like pharmaceuticals have been the focus of attention on a global scale. Ciprofloxacin is a frequently used medication that can be discovered in rivers, wastewater treatment facilities, and hospital effluents. The viability of CIP degradation by high-frequency ultrasonic process is explored in this paper, and sonolysis with hydrogen Peroxide and Sono-Fenton are assessed. The essential amounts of ferrous-ions with hydrogen peroxide were optimized using response surface approach. The Sono-Fenton method produced the best results, causing a complete pharmacological degradation within 15 min and a mineralization of more than 60% after 60 minutes [24].

The cavitation technique has been used to explore the breakdown of pharmaceutical wastewater. This study looked at the impact of acoustic cavitation over various time periods ranging from 0 to 90 minutes. A high frequency probe with a 20 kHz capacity was employed in acoustic cavitation, and the reactor's volume was 1 liter. The maximum Chemical Oxygen Demand elimination achieved using acoustic Cavitation was 75.53% in 75 minutes [25]. The most effective method is to use "total" metrics like Chemical Oxygen Demand and Biological Oxygen Demand combined to regulate real wastewater treatment processes. It enables the biodegradability to be calculated and controlled as a ratio of these two characteristics.

Instead of Total Organic Carbon, Chemical Oxygen Demand is a better metric for controlling the total load of contaminants [26]. The treatments for the pharmaceutical waste-water in this investigation, a combination strategy of ozone-based AOPs and adsorption by activated char was used. Although hydroxyl radicals, which are a non-selective stronger oxidant than ozone, were produced in situ as a result of the injection of Hydrogen Peroxide, ozone is a selective oxidant. The majority of the pharmaceuticals found in the pharmaceutical industry's effluent were antipsychotics, painkillers, and anticancer medications. At pH 5–11, the peroxone procedure reduced Chemical Oxygen Demand with an efficiency of 75–88.5% in 3 hours. The Chemical Oxygen Demand was further decreased by adsorption by activated char to 85.4-92.7% for pH 5-11 in 2.5 hours. During ozonation, all other water quality metrics were drastically reduced (removed by more than 73% in total) [27]. A number of Advanced Oxidation Processes, including ionizing radiation, photocatalytic oxidation, electrochemical oxidation, ozonation or catalytic ozonation, and Fenton or Fenton-like reaction occurs good results and included information on their principles, characteristics, key influencing factors, and photocatalytic oxidation applications [28]. To be deemed genuine for its goal, drugs' benefits in treating humans and animals must balance their negative environmental effects. However, there is much to be desired when observing the speed at which pharmaceutical waste contamination is dispersing over the world. Even

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while everyone engaged is trying their best to alleviate the problem, nothing has been done to focus on the future trend of pollution brought on by pharmaceutical waste. This is partially a result of the absence of well-established laws, regulations, sources, handling procedures, testing standards, and threats that we face right now and in the near future [29]. The Ultrasonic and Hydrodynamic Cavitation are able to activate persulfate and hydrogen Peroxide to produce sulphate, peroxymonosulfate, or hydroxyl radicals, hence significantly improving mass transfer, catalytic conversions, and the formation of bulk radicals. The removal efficiencies of Sulfadiazine with the HC/Persulfate/HydrogenPeroxide/ Iron (III) sulphite and Ultrasonic/Persulfate/Iron (II) oxide processes were 81.0% and > 95% in 60 min, respectively. With the Ultrasonic /Hydrogen Peroxide /Iron Oxide and Ultrasonic / Carbon dioxide + processes, respectively, total reductions of 93.6% Tetracycline and 85% were accomplished in 60 min. The Ozone mass transfer and catalysis were improved, resulting in a nearly two-fold increase in the Tetracycline removal rate constant with Ultrasonic / Ozone /goethite over ozonation alone (0.174 min). Additionally, the Ultrasonic / Ozone /Copper Oxide method was able to remove more than 85% of the Chemical Oxygen Demand from all pharmaceutical industrial effluents (500–7500 mg/L Chemical Oxygen Demand) [30].

Conclusion

It is necessary to treat the pharmaceutical wastewater to eliminate the persistent complex organic compound present in them as they hazardous to aquatic life and adversely affect human health. For this treatment integrated physicochemical methods approach is found to be impressive. The degradation was found to be maximum in acidic medium pH 3.0 to 5.6. Thus, we can say acidic medium is favourable for the treatment of wastewater. The best reaction time was 60 minutes to 120 minutes for the treatment. Ultrasonic Cavitation was carried out at different frequencies, significant results were found at 22 kHz. From all the combination of Ultrasonic Cavitation with different additives, maximum reduction in Chemical Oxygen Demand was found with Ultrasonic / Copper Oxide + Ozone combination. Ultrasonic, Ozone and Copper Oxide catalyst reduced Chemical Oxygen Demand by 92%. By combination of Ultrasonic Cavitation with addition of different oxidizing agent remarkable reduction in Chemical Oxygen Demand of pharmaceutical wastewater is found.

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