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The Present Advances in Electro -Fenton Technology for Pharmaceutical Treatment in Aquatic Environments

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Abstract

A highly efficient electrochemical advanced oxidation process is the electro-Fenton (EF) process. Over the past three decades, it has developed into a clean and effective method of treating wastewater. Conventional EF still has limitations for industrial-scale development, even though it has been established to be an effective method for the disintegration and mineralization of harmful and/or persistent organic chemicals. A major issue, water pollution affects ecosystems, human health, and economic growth in a variety of ways. Because of their possible eco-toxicity, pharmaceutical chemicals have been regarded as a developing water micro-pollutant in recent years. Micropollutants, usually referred to as emerging contaminants, includes synthetic and domestic items such as pharmaceuticals, personal care products, hormonal substances, and agricultural chemicals. Ecosystems and human health are in immediate danger by emerging concern contaminants (ECC) in the environment, demanding effective and creative treatment techniques. Because of its high mineralization rate and ecological compatibility, the electro-Fenton (EF) method, which is based on electrochemical advanced oxidation (EAOP), has been utilized largely for ECC treatment. With the purpose of giving a potent technique for treatment of wastewater contaminated with biorecalcitrant contaminants, this review article is meant to be complete, critical, and of general interest. It discusses the most recent developments and progress in the EF process.

Keywords: Eletro-Fenton, pharmaceutical pollutants, advanced oxidation processes, wastewater treatment, emerging contaminants, aqueous solution

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1. Introduction

Rapid urbanization, industrialization, and population increase leads to the unsustainable use of naturally occurring surface water supplies. The environment is currently under risk due to the growing number and variety of human-made pollutants that mix with natural water streams. Even a small amount (nano-gram to microgram per liter) of these pollutants may eventually build up in the food chain and represent a serious threat to human well-being, soil, and aquatic ecosystems due to their persistent chemical properties of cytotoxicity and intractability. Pharmaceutical chemicals have been considered as crucial worldwide and play a critical role in the treatment and prevention of diseases in both people and animals [1]. Because of their widespread consumption, they are becoming increasingly prevalent in the environment. These chemicals have been detected in concentrations ranging from g/L to mg/L in drinking water, groundwater, waste water, and sediment[2] . For the treatment of persistent organic pollutants, advanced oxidation processes (AOPs) have drawn a lot more attention lately. By mineralizing these substances, increasing the solution's ability to degrade, and reducing toxicity, AOPs are useful for eliminating them effectively [3] . The usefulness of Fenton reaction based AOPs in handling resistant organic compounds has been widely accepted. But the Fenton reaction's low ferrous regeneration rate meant that more ferrous ions were needed to effectively eliminate pollutants, which in turn led to more sludge generated [4].

Heterogeneous catalysts, chelating agents or the addition of external energy—such as electricity in the electro-Fenton (EF) process, UV light in the photo-Fenton process, and ultrasound in the sono-Fenton process—were used to partially overcome this limitation of the conventional Fenton process. Since its development in 2000, the EF approach has garnered significant trust from scientists because to its exceptional performance, ease of use, and low cost [5]. More researchers have been drawn to this issue since the EF method does not require H_2O_2, which makes handling easier H_2O_2 and less expensive. By decreasing dissolved oxygen by two electrons in an acidic environment, the EF process can produce at the H_2O_2 cathode surface. Fast cathodic reduction of ferric ions increases the process's activity through Fe2 regeneration, which eventually leads to the EF process's ideal ferrous ion requirement, which is in the range of a few mg L [6]. Various matrices of the environment were used for demonstrating the EF process's versatile uses. By oxidizing the arsenite to arsenate and eliminating ammoniacal nitrogen [7], the EF process is an alternate method for lowering the toxicity of arsenic in drinking water. The effectiveness of the EF process in treating complex wastewater and actual industrial discharges, such as mine water, textile wastewater, pharmaceutical was municipal wastewater treatment plant effluents[8] , mixed industrial wastewater.

2. Contaminants of Emerging Concern (ECCs)

Even at low concentrations, pollutants known as emergent chemical contaminants (ECCs) can negatively impact the human endocrine system. There are several anthropogenic sources that have been determined to have primary origins, including hospitals, municipal wastewater, and farming operations. Most of ECCs are organic in nature and contain producing micro-pollutants, which are trace organic compounds that are not subject to regulation [9]. Synthetic dyes, pharmaceutical waste, frequently utilized personal care products (PCPs), and endocrine-disrupting chemicals (EDCs) are among the categories of ECCs. In multiple sectors, synthetic dyes are mostly employed for coloring. The resulting hues are incredibly visible and can have unpleasant effects even at very low concentrations in water [10]. Numerous kinds of antibiotics, including prescription and over-the-counter medications, and distinct steroid hormones are instances of pharmaceutical waste (Figure 1).

3. Conventional method for the removal of pharmaceutical compounds from wastewater

3.1. Adsorption technology

The soluble chemical (adsorbate) is eliminated from a fluid by getting into contact with a solid surface (adsorbent) in the adsorption process [11]. This method offers a high probability of eliminating any leftover inorganic or organic compounds that may be present in wastewater. Adsorbent and adsorbate molecules interact through interactive forces; this can occur through chemisorption, which binds to the formation of a strong covalent bond because it includes the transfer or sharing of electrons, or physisorption, which is induced by a weak electrostatic interaction between the adsorbent and adsorbate. Instead of altering the structure of its adsorbent during an association, the connection is made through modifications in functional groups, hydrogen bonds, electrostatic contact, and the donation-accepting electron reaction. [12]. Adsorption method is regarded to be a powerful approach for treating pharmaceutical wastewater. Adsorption has several advantages over other technologies, such as cost-friendly, simplicity of use, and the potential for reuse of its adsorbents. Pharmaceutical are extracted from water using a number of organic and inorganic materials as adsorbents, including zeolites, clay, polymers, graphene, carbon nanotubes, and waste from agriculture (Figure 2) [13].

3.2. The electro-Fenton process

A key component of EAOPs used for wastewater treatment, in particular for ECs and pharmaceuticals, is EF technology. It makes use of the generation of powerful oxidizing agents, especially the highly reactive •OH, which is capable of attacking a number of organic pollutants. The Fenton catalysts used in the homogeneous EF process consist primarily of Fe^{2+} ions, which react with the H2O2 formed in situ. Equation (1) shows how the Fe^{2+} ions are transformed. •OH radicals are main oxidizing species that cause organic contaminants in wastewater to degrade as a result of this reaction. The regeneration of Fe^{2+} ions need to occur for this process to be effective. However, the quick buildup of ferric ions (Fe^{3+}) and the slow process of Fe^{2+} regeneration often decrease the effectiveness of homogenous EF. This mismatch causes insufficient H_2O_2 degradation and iron oxyhydroxide precipitation, which ultimately decreases efficiency [14].

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

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO^{2\bullet} + H^+ \quad (2)$$

The process's widespread use in actual wastewater treatment is limited by the need to maintain strong acidic conditions in order to attain optimal efficiency. This limitation adds complexity and expense to treatment process by requiring an extra neutralizing step to bring the pH of the wastewater below a safe level. Consequently, there are few large-scale practical uses for this technology. Heterogeneous EF technology, involving use of solid catalysts or integrated cathodes, has so recently been the subject of a lot of study. Its exceptional efficiency, flexibility, and sustainability with the environment are the reasons for this focus [15]. Enlargement of the operating pH range and the avoiding of iron sludge development are two main benefits of heterogeneous EF, which address limitations of the homogeneous EF method [14]. Fenton catalyst combines with water to generate •OH, which enables effective mineralization and pollution elimination. Electrogeneration of H_2O_2 through two-electron reduction of input air or oxygen is used to overcome security issues with H_2O_2 transport and storage. This method avoids the costs and risks related to organizing [16].

3.3. Iron Activated Cathode in a Heterogeneous Electro-Fenton Process

Three-dimensional electrodes are utilized in nano-engineering study to enhance the electro-Fenton system's transfer of mass and conductivity [17]. The primary surface of electrode as well as the electrode surface of particle are in which electrochemical reactions occur due to this novel electrochemical system's improved effective contact area and multiple micro-electrolysis cells. Iron and/or oxides of transition metals can be introduced for cathodic modification, which includes dual functionality as an iron supplier and a counter electrode. This is possible by immobilizing an iron source or by chemically altering carbonaceous material. Considering the high efficacy of these systems in reducing pollutants, their limited operating pH range and low current efficiency may prevent them being commonly utilized in water purification. Using Ti–PbO_2 and graphite felt as anode and cathode, respectively [17], investigated Fe–Ni foam particles on three-dimensional electrodes for elimination of NSAIDs. Compared this system to an analogous 2D electro-Fenton system, the removal percentage was almost doubled. Because Ni-foam electrode may activate molecular oxygen to form single oxygen, researchers attributed increased pace to larger concentration of radical species manufacture.

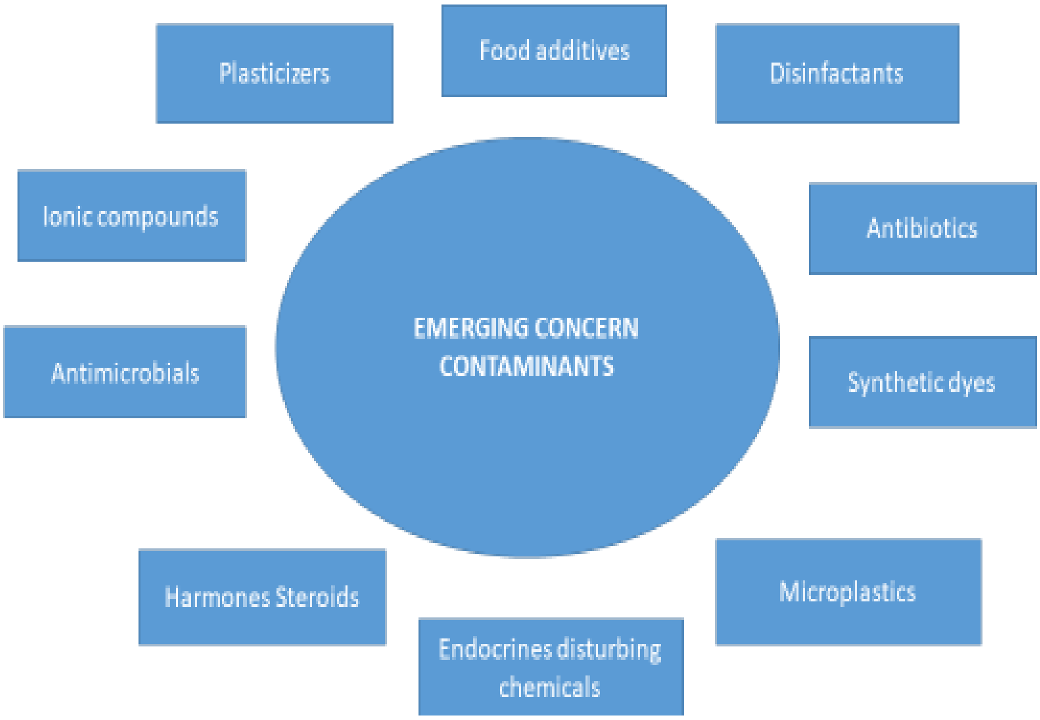

Figure 1: Types of contaminants of emerging concern

Table 1: Comparison of current techniques for removal of pharmaceutical compounds

Treatment processes	Advantages	Disadvantages
Adsorption process	By-product generation is limited or absent, and the design and operation are simple, convenient, highly efficient, environment friendly, and regenerating.	Several variables affect its effectiveness, insufficient adsorbent selectivity, and problems with disposal
Advance oxidation process	Most organic compounds can be converted into harmless or biodegradable products through wide. A quick and highly reactive process.	Large bandgap of semiconductors Extracting minerals from hazardous and resistant materials is not cost-effective. In addition, the manufacture of byproducts and the need of additional oxidant treatment
Hybride technology	It has been established that hybrid technology works as well as single procedures.	Pretreatment may be necessary for different kinds of wastewater samples, and combined procedures need for more treatment units, which raises the use of energy.
Membrane separation method	It includes nontoxic chemicals, has excellent mechanical and chemical stability, is appropriate for treating saltwater water as well as wastewater plant discharges, is suitable for a variety of pharmaceuticals, and needs less energy than previous techniques.	Difficulty disposal of membrane after usage, removal efficiency is affected by the toughness and endurance of the selected membrane. Membranes can be destroyed by pressure changes, have a large pH range, and are prone to clogging.
Process of biological treatment	When combined with additional treatment techniques, pharmaceuticals can be removed more efficiently. This is an easy process that limits the production of odors, lowers pathogens and lipids, allows for the use of a wider range of microorganism types for manufacturing, generates renewable energy, and has no adverse effects.	High expense, difficulties with persistence, and insufficient pharmacological elimination compared to alternate treatment approaches.

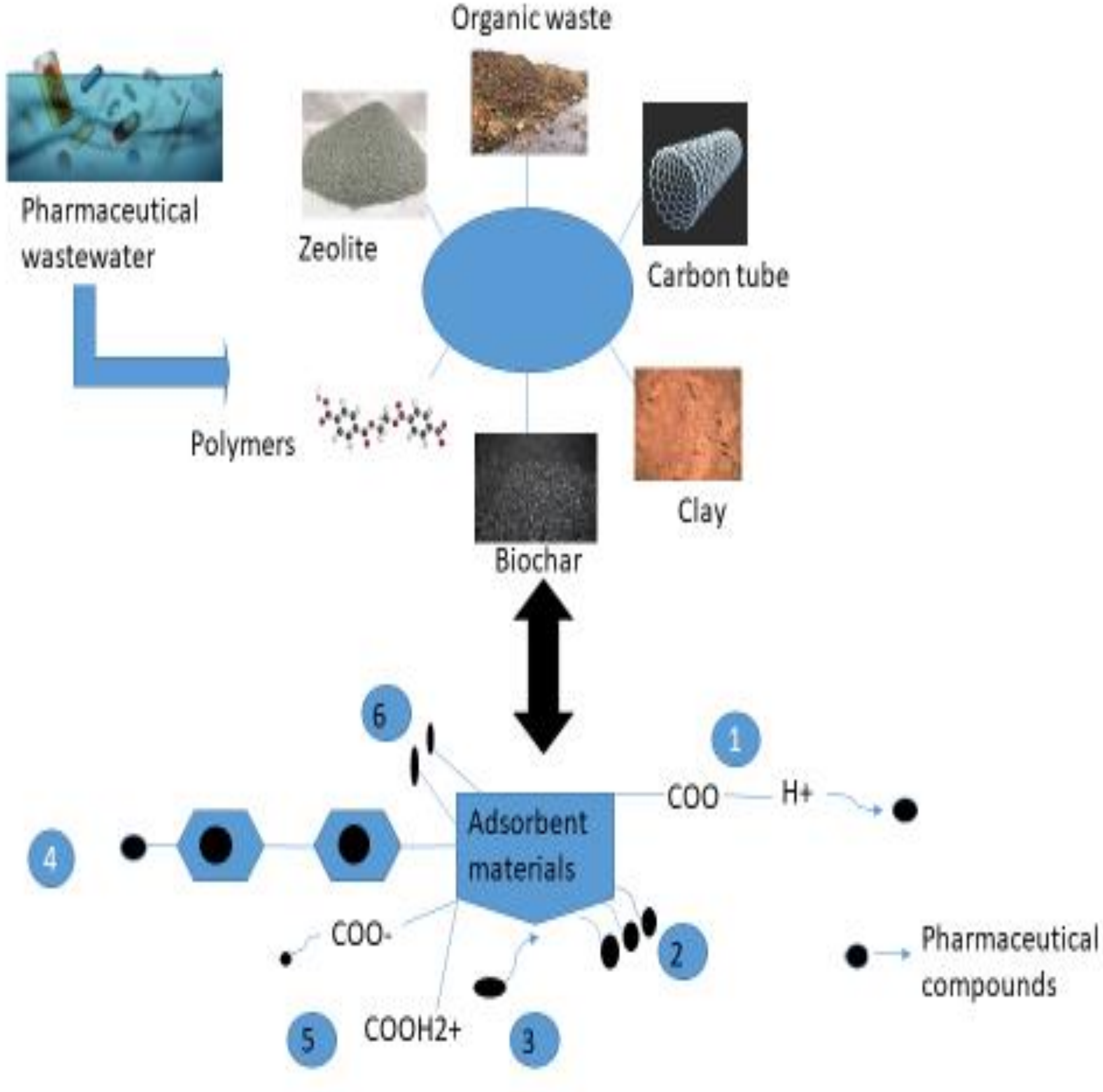

Figure 2: Adsorption mechanism for removal of pharmaceutical substance from water

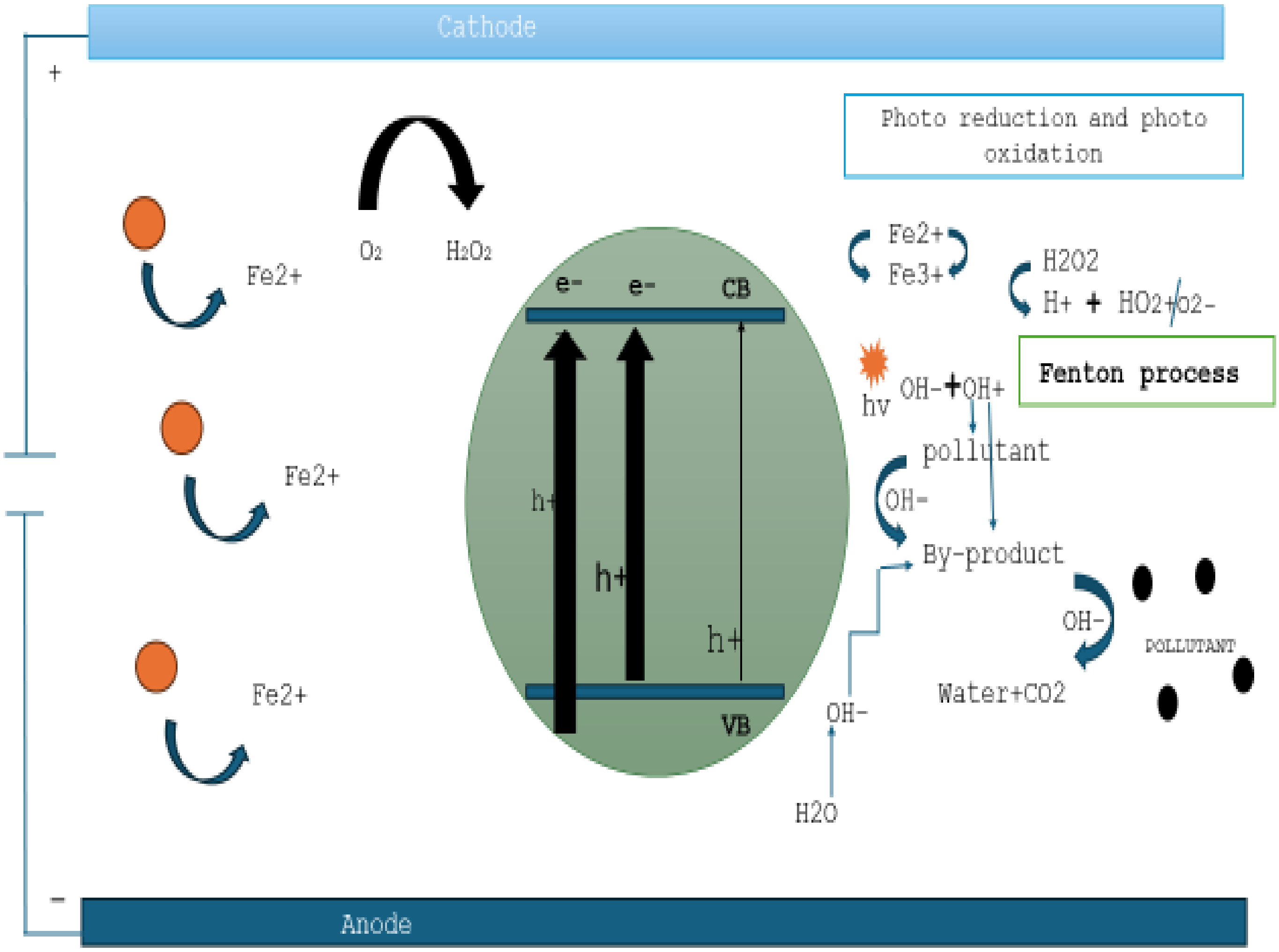

Figure 3: Process of photoelectric Fenton process

Recently, it has been a lot to explore in development of functionalized iron cathodic material including carbon aerogels, graphite felt, carbon felt, and active carbon. Specifically [18], examined the electrocatalytic activity of carbon felt modified with mesoporous $MnXCo_3$-XO_4 nanomaterials as zelectro-Fenton cathode. Increased transport of electrons and redox reactivity led to higher rates of ciprofloxacin decomposition. This related to development of a more accessible active site and the combined effect of embedded manganese and cobalt within the porous matrix.

3.4. The Photoelectric-Fenton Process

Integrating the heterogeneous electronic Fenton process with additional methods can enhance its efficiency. Simultaneous alternative and consecutive treatments, as well as hybrid processes incorporated into systems, have been presented; numerous cases report the complete elimination of pollutants from wastewater [19]. By simultaneous the solution to visible light, the photoelectro-Fenton (PEF) method is a "upgrade" to electro-Fenton [20]. When photons are absorbed, their power can be used to drive photodecarboxylation, resulting in ferrous iron, and photo-reduce $[Fe(OH)]^{2+}$ in an acidic atmosphere [21]. A few studies investigated into possible use of PEF at circumneutral pH; the majority of PEF research has been carried out in acidic environments [22], examined on the effectiveness of a Fe-based 2D metal–organic framework (MOF) catalyst for the removal of bezafibrate in sulphate-based medium at pH 5.5 using a single-compartment glass cell upon Xe lamp irradiation, reporting 92% removal—an increase in removal in comparison with control experiments. Bezafibrate, bisphenol A, fluoxetine, and naproxen are amongst emerging pollutants that were spiked with urban wastewater in studies conducted at neutral pH. Results indicated good efficacy for removal, with 93, 85, and 78% removal reported for naproxen, fluoxetine, and bisphenol A, respectively, after 90 minutes of treatment. Scientists reached conclusion that the existence of organic matter, high turbidity, and higher initial pH all lead to the decrease of the bezafibrate treatment (Figure 3) [23].

4. Comparison of current techniques for removal of pharmaceutical compounds

Every wastewater treatment method has advantages as well as disadvantages. Several drugs can be efficiently eliminated from pharmaceutical effluent through adsorption [24]. Although it still has several drawbacks such as its low selectivity as an adsorption. On the other end, biological treatments are more common as they can cause complete destruction. However, it was considered a lengthy procedure, and most of its methods have been costly [25]. Technology of membranes may experience membrane fouling problems, although it has high physical & chemical stabilities. Most organic substances might be broken down by complex oxidation processes into safe or biodegradable compounds, however this procedure could not be economical [26]. Table 1 below enumerates advantages versus drawbacks of some existing pharmaceutical material removal techniques.

5. Conclusions

Investigation describes several modern methods for purifying water sources that may contain considerable amounts of pharmaceuticals that people or animals use daily. Some studies employed adsorption approach, a strong tool for removing pharmaceutical wastewater EF process is ideal for wastewater that contains ECs and PCs. Because of its high efficiency, excellent rates, and low cost particularly in concentrating pharmaceutical treatments EF method suggested for direct usage at hospital or pharmaceutical plant release sites. Its production of non-selective hydroxyl radicals makes it easier for various pollutants to break down, contributing to almost entire mineralization Resistant ECCs can be successfully and sustainably treated with heterogeneous electro-Fenton (HEF) methods. Heterogeneous catalysts quicken up Fenton's reactions as they are more responsive throughout a higher pH range.

6. Prospects for future

The efficiency of the EF technique when treating wastewater contaminants with ECs and PCs has been determined throughout many applications. However, it is imperative to address pollution and immediately filter wastewater away from research labs. These steps are necessary to protect environment and public health from serious threats. Additionally, there is presently no literature on scaling up EF process for EC treatment, and research has focused mainly on developing new wastewater treatment protocols to remove pharmaceuticals from synthetic wastewater for phase conditions. From greatest hurdles for deploying EF technology are iron leaching and the resultant production of iron sludge. But Iron sludge creation can be significantly reduced by integrating the Fenton process with biological systems using solid catalysts rather than $Fe2+$ to promote the Fenton reaction on the solid catalyst's surface.
The adverse impacts of ferrous sludge formation may also be reduced by using nZVI powder. Combining EF process with other AOPs is essential for treating highly resistant wastewater with low BOD/COD ratios. Furthermore, wide use of EF technology relies on economic evaluations. Future studies should examine technical and functional features of reactors, including design, hydrodynamics, flow regime, and primary treatment effectiveness. By considering cost-saving techniques, as employing affordable reagents & electrochemical materials, maximizing production and consumption of •OH radicals, and reducing sludge residues, operating expenses might be reduced even more, providing EF a viable alternative to wastewater treatment.

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