Emerging Materials and Technologies

Wastewater Treatment with the Fenton Process

Principles and Applications

Dominika Bury, Michał Jakubczak, Jan Bogacki, Piotr Marcinowski, and **Agnieszka Jastrzębska**

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Wastewater Treatment with the Fenton Process

The presence of refractory organic compounds in wastewater is a global problem. Advanced oxidation processes, in general, and the Fenton oxidation process are alternative technologies for wastewater and water treatment. This book gives an overview of Fenton process principles, explains the main factors influencing this technology, includes applications, kinetic and thermodynamic calculations and presents a strong overview on the heterogeneous catalytic approach. It demonstrates that the iron-based heterogeneous Fenton process, including nanoparticles, a new complex solution, is highly efficient, environmentally friendly and can be suitable for wastewater treatment and industrial wastewater.

FEATURES

- Describes in detail the heterogeneous Fenton process and process applications
- Analyzes the advantages and disadvantages of different catalysts available and their suitability to specific processes
- Provides economic analysis of the Fenton process in a ready-to-use package for industrial practitioners for adaptation into already existing industrially viable technologies
- Promotes a modern solution to the problem of degradation of hazardous compounds through ecological and environmentally friendly processes and the use of a catalyst that can be recycled
- Explains highly complex data in an understandable and reader-friendly way

Intended for professionals, researchers, upper-level undergraduate and graduate students in environmental engineering, materials science, chemistry, and those who work in wastewater management.

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Contents

1 Introduction

The presence of refractory organic compounds in wastewater is a many-year global problem. Despite the extensive development of various wastewater treatment technologies, they still fail to decompose potentially harmful and dangerous compounds. There is an essential need to develop novel solutions that are efficient and cheap. One of the effective methods to reduce pollutants is advanced oxidation processes (AOPs), consisting of effective radical generation, and deserves special attention. This study presents an insight into the used heterogeneous Fenton processes, which is one of the AOPs methods. The application of iron-based heterogeneous catalysts for wastewater treatment and pollution decomposition is promising, as demonstrated by many laboratory, field-based, pilot and full-scale studies. Further, fundamentals, primary applications, crucial process parameters, kinetics, and selected economic aspects were discussed.

The literature review has shown that the iron-based heterogeneous Fenton process can be suitable for wastewater treatment, including industrial wastewater characterized by complex composition. In addition, high efficiency and environmental friendliness indicate the potential for industrial application.

Clean and sufficient quality water is a crucial component of the environment and a resource in industry, agriculture, and human life. WHO and UNICEF noticed that in 2017 more than 785 million people did not have access to at least essential water services in the last their research published. Furthermore, more than 884 million people did not have safe water to drink. These problems affect especially poverty country with not enough developed systems of clean water or its complete absence. Access to safe drinking water is a basic human right, yet many people around the world do not have access to it. This lack of access is often due to poverty, inadequate infrastructure, and environmental degradation. Without safe drinking water, people may be forced to drink from contaminated sources, which can lead to waterborne illnesses such as diarrhea, cholera, and typhoid fever. These illnesses can be particularly dangerous for young children, pregnant women, and those with weakened immune systems. In addition to the health impacts, the lack of safe drinking water can also have economic and social consequences. Without access to safe water, people may have to spend a significant amount of time and energy collecting water, which can limit their ability to attend school or earn a living. Furthermore, the lack of safe water can contribute to conflict and displacement, as people may be forced to move in search of water. In developed countries, problems with clean water mainly generated pollutants from the transfer and agriculture or overexploitation of them. Water pollution is mostly caused by the discharge of untreated essential domestic and industrial wastewater. More than 400 million tons of pollutants are released into water supplies. Moreover, problems with water quality could be generated during accidents and failures in large companies [1–5].

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China, India, and some states in the USA and European countries are the places that generated the largest amounts of sewage (Figure 1.1). However, even treated wastewater is not free from pollutants, which are often present at the trace level. This is the effect of the high concentration of residents and the developed industry, including textiles, chemicals, and pharmaceuticals. The wastewater is often a by-product, generated, among others, when washing production lines. The presence of industrial contaminants in ground waters, surface waters, seawater, wastewater treatment plants, soils, and sludges was noticed by scientists from all world [6].

Therefore, it is essential to implement effective wastewater treatment processes that can remove a wide range of pollutants, including those that are present at trace levels. Furthermore, it is crucial to implement regulations and policies that promote the sustainable management of wastewater and the reduction of pollution at the source. This will require the cooperation of governments, industries, and individuals to ensure that the world's water resources are protected for future generations [7]. Industrial wastewater can be dangerous due to the presence of various pollutants that can be harmful to human health and the environment. These pollutants can include chemicals, heavy metals, pathogens, and organic compounds that can have adverse effects on aquatic life, soil, and vegetation. For example, some industrial wastewater contains toxic heavy metals such as lead, mercury, and cadmium, which can accumulate in the food chain and pose a serious threat to human health. Exposure to these heavy metals can result in neurological disorders, kidney damage, and cancer. Furthermore, industrial wastewater can contain organic compounds such as benzene, toluene, and trichloroethylene, which are harmful to human health even at low levels of exposure. These compounds can cause skin and eye irritation, and respiratory problems, and long-term exposure can lead to cancer.

In addition, industrial wastewater can contain pathogenic microorganisms such as bacteria, viruses, and parasites, which can cause waterborne diseases such as cholera, typhoid, and hepatitis A.

Moreover, the discharge of untreated or poorly treated industrial wastewater can have serious environmental consequences. For example, the presence of nutrients in wastewater can cause eutrophication, which can result in the depletion of oxygen in water bodies and the death of aquatic life.

The industrial compounds contaminate the high source of pharmaceuticals, hormones, consumer product chemicals, and other organic wastewater compounds. All of these solutions could be dangerous for the environment and people's health. Much of the compounds accumulate very well in the end, consequently, an increase in the concentration of pollutants in individual components of the environment. The highest threat is related to ones that possess toxic, mutagenic, and carcinogenic properties. They are usually difficult to remove and act as endocrine disruptors [8–11]. Interestingly, hormones presented in water could be linked to an increase in elevated female:male ratios, and their physiological alterations. Moreover, antibiotics could change the microbial structure and nitrogen biotransformation processes [7]. Pollution of wastewater origin can easily get into groundwater and contaminate drinking water resources [12,13]. What is especially important, such

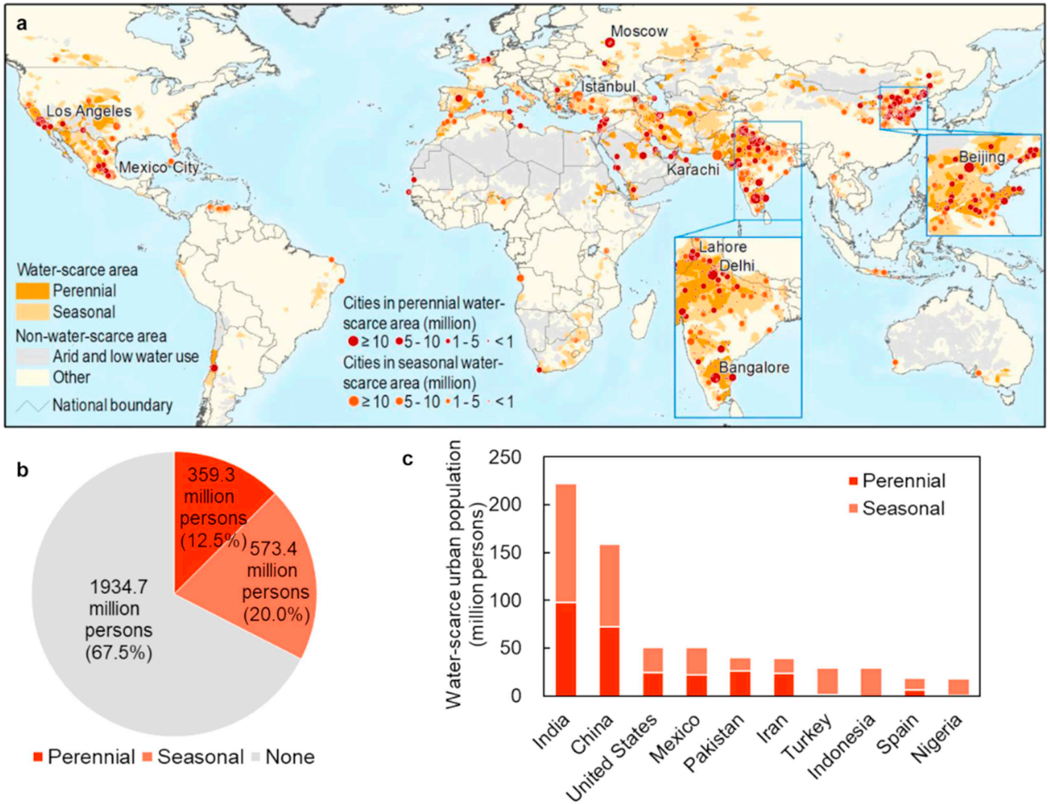

FIGURE 1.1 Current urban water scarcity.

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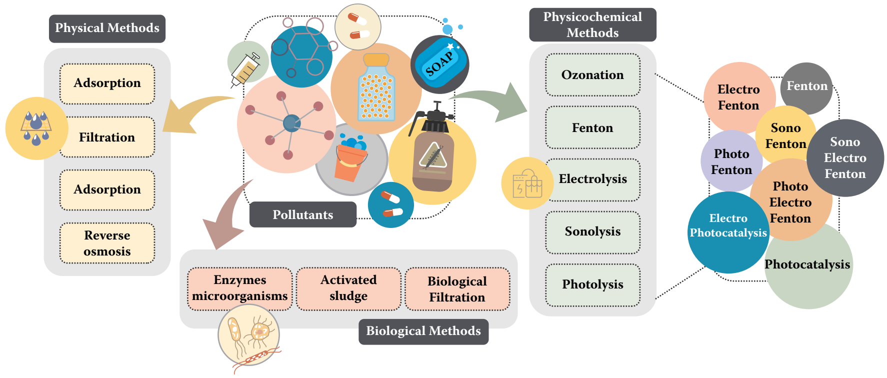

FIGURE 1.2 Kinds of wastewater treatment.

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effects appear even in low concentrations. Currently, the basic actions to protect the environment are:

- •. Wastewater treatment to remove pollutants,
- •. Decreasing the generation of wastewater,
- •. Reusing/closing circuits [14].

The most popular and common method is wastewater treatment. The basic methods of wastewater treatment are mechanical, biological, chemical, and mixed, as shown in Figure 1.2 [15]. Other methods, such as decreasing the generation of wastewater are less effective, while reusing/closing circuits cost lots of money.

Due to the usually high content of organic compounds in industrial wastewater, methods such as:

- Coagulation – is a process based on the coagulation of oppositely charged compounds, negative colloids contained in the wastewater (after previous destabilization) and positively charged aluminum or Fe^{3+}, which are in the form of inorganic coagulants (PAC, PIX) [16–18].
- Electrocoagulation – is a process that uses metallic electrodes. Scientists used them to remove organic compounds and suspensions from treated wastewater. The electrode base, located on the anode, is built from the metal part and is used to generate the metal ions M^{n+}. The metallic ions formed at the anode may undergo hydrolysis or form polymeric compounds with adsorption properties.
- Flotation – is the process of separating the fats and flocs present in the wastewater in a flotation device (open separation tank). The process takes place under the influence of dissolved air. Compounds contained in sewage are mixed with fine air bubbles with water and air (a mixture is formed). While mixing, air bubbles stick to the fluff and raise it to the surface. Too heavy substances in the wastewater do not flow out with the bubbles; they settle and then are discharged through the drainage system [17,19].

- Activated carbon adsorption – is a process based on the electron donor-acceptor complex mechanism. These processes reals on pollutants of the adsorbate act as the electron acceptors and the basic sites on the carbon surface serve as the donors. Moreover, scientists noticed the – dispersion interactions and the solvent effects during the adsorption process [20,21].

However, these methods used alone are not effective enough. Treatment installation is required, consisting of more than one unit process. On the other hand, each method has its drawbacks. For example, the filtration requires maintenance of the filter in a proper condition due to pores clogging, which affects less flow velocity, and sorption surface [22]. A by-product of sedimentation is sludge, and the process itself is used to complement a proper treatment process. Additionally, sludge needs to be removed after the process. During the coagulation, it is also necessary to introduce additional chemical compounds, coagulants, and flocculants, which increase the salinity of the wastewater [18,23]. The poor removal of fine emulsions characterizes the adsorption process [24]. In the case of biological wastewater treatment, one should consider the high sensitivity to the content and pH of the toxic compound of the wastewater and the conditions that the process should meet [25]. Most of the methods mentioned above have their weaknesses and drawbacks, which may significantly hinder their usage in industrial wastewater treatment. A growing amount of wastewater encourages to development of new treatment methods and processes [26].

A direction that seems particularly promising is the development of AOPs. The reaction is driven due to the generation of molecules with unpaired valence electrons, so-called highly reactive radicals. Radical addition is the attachment of a radical to a neutral molecule, during which the multiple bonds are broken, resulting in a new product with one unpaired electron [27]. The reaction consists of taking hydrogen atoms from C–H, N–H, or O–H. In the next stage, OH bonds transform into unsaturated C=C bonds or aromatic rings [28]. In AOP, the typically generated radical is the hydroxyl one – $^{\bullet}OH$, one of the strongest known oxidants [29].

The advantages of the process include:

- Lack of specialized conditions and devices necessary to carry out the reaction – “room conditions”,
- The reagents for carrying out the process are widely available, thus making the process inexpensive,
- Efficiency and effectiveness of converting organic pollutants into simple non-toxic compounds,
- In the case of heterogeneous catalysts (iron minerals), there is a possibility of reusing the material – saving raw material and activities conducive to environmental protection and reduction of process costs [30].

AOPs differ from each other in radicals generated, the reagents used or required conditions, or other process parameters. The advantages of AOPs include the use of catalysts and oxidants, which are environmentally friendly, and the high efficiency in the oxidation of organic compounds. AOPs are an excellent alternative to hazardous oxidation processes and toxic inorganic oxidants such as permanganates or chlorine. Due to their high efficiency and effectiveness, the AOPs are readily used in the treatment of water and wastewater (including from various industries, including textiles or food). Moreover, it is popular to remove pollutants, during soil and sediment reclamation, municipal sediment conditioning, and volatile organic compounds removal [29].

AOPs processes are divided into:

- Ozone-based AOPs: This type of AOP involves the use of ozone gas (O_3) to generate hydroxyl radicals. Ozone is a strong oxidizing agent that can break down many organic and inorganic pollutants in water. Examples of ozone-based AOPs include ozone alone (O_3), ozone combined with hydrogen peroxide (O_3/H_2O_2), and ozone combined with ultraviolet light (O_3/UV).
- Hydrogen peroxide-based AOPs: This type of AOP involves the use of hydrogen peroxide (H_2O_2) to generate hydroxyl radicals. Hydrogen peroxide is a relatively weak oxidizing agent that requires a catalyst or another oxidizing agent (such as ozone or UV light) to generate hydroxyl radicals. Examples of hydrogen peroxide-based AOPs include Fenton's reagent (Fe^{2+}/H_2O_2) and photo-Fenton's reagent (Fe^{2+}/H_2O_2/UV).
- UV-based AOPs: This type of AOP involves the use of ultraviolet (UV) light to generate hydroxyl radicals. UV light can break down some organic pollutants, but it is not strong enough to break down many inorganic pollutants. Therefore, UV-based AOPs are often combined with other oxidizing agents such as hydrogen peroxide or ozone. Examples of UV-based AOPs include UV alone (UV), UV combined with hydrogen peroxide (UV/H_2O_2), and UV combined with ozone (UV/O_3).
- Electrochemical AOPs: This type of AOP involves the use of an electric current to generate hydroxyl radicals. Electrochemical AOPs can be used to treat a wide range of pollutants, including those that are difficult to treat using other AOPs. Examples of electrochemical AOPs include electro-Fenton (Fe^{2+}/H_2O_2/DC), anodic oxidation (AO), and cathodic reduction (CR) [31].

Another breakdown of AOPs includes:

- Chemical oxidation *vs.* photochemical oxidation: AOPs can be divided into chemical oxidation and photochemical oxidation processes. Chemical oxidation processes use chemical oxidants such as ozone, hydrogen peroxide, or persulfate to generate hydroxyl radicals. Photochemical oxidation processes use UV light irradiation or other light sources to generate hydroxyl radicals.
- Homogeneous *vs.* heterogeneous AOPs: Homogeneous AOPs have the catalyst in the same phase as the solution, whereas heterogeneous AOPs have the catalyst in a different phase. Examples of homogeneous AOPs include Fenton's reagent, photo-Fenton, and the TiO_2-based photocatalytic

process. Examples of heterogeneous AOPs include photocatalysis using immobilized catalysts, such as TiO_2 on glass plates or ceramic beads.

- Traditional *vs.* novel AOPs: Traditional AOPs have been in use for several decades, while novel AOPs are relatively new technologies that have been developed in recent years. Examples of traditional AOPs include Fenton's reagent, ozonation, and photocatalysis. Examples of novel AOPs include plasma-activated water, ultrasound, and sonication.
- Ozone-based *vs.* hydrogen peroxide-based AOPs: Ozone and hydrogen peroxide are two common oxidants used in AOPs. Ozone-based AOPs generate hydroxyl radicals through the reaction of ozone with water, whereas hydrogen peroxide-based AOPs generate hydroxyl radicals through the reaction of hydrogen peroxide with iron (Fe^{2+}) or light.
- Thermal *vs.* non-thermal AOPs: Thermal AOPs rely on heat to generate hydroxyl radicals, whereas non-thermal AOPs do not. Examples of thermal AOPs include wet air oxidation (WAO) and supercritical water oxidation (SCWO). Non-thermal AOPs include photocatalysis, Fenton's reagent, and ozonation [31,32].

Scientists used several methods to generate radicals, as shown in Figure 1.3, especially homogeneous, heterogeneous Fenton process, photo-Fenton or photocatalysis, and reaction with ozone [30].

The most popular of these methods is the Fenton process, which utilized chemical catalysts (including Fe^{2+}, TiO_2 ions). In the classical Fenton process, the hydroxyl radical ($^{\bullet}OH$) can be generated from the reaction between aqueous ferrous ions and hydrogen peroxide (H_2O_2). In the next step, hydrogen peroxide destroys refractory and toxic organic pollutants in wastewater. The conventional Fenton process realizes

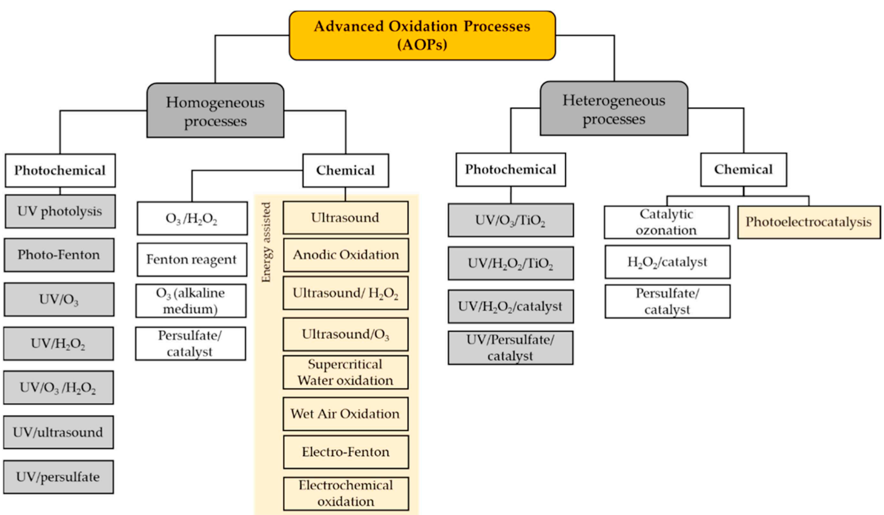

FIGURE 1.3 Types of advanced oxidation processes.

Source: https://www.mdpi.com/2073–4441/11/2/205#.

this by including acid regulation, catalyst mixing, oxidation reaction, neutralization, and solid-liquid separation.

In a homogeneous AOPs, the catalyst and the wastewater or solution to be treated are in the same phase, usually in the liquid phase. In this type of AOPs, the catalyst is typically a metal ion or a combination of metal ions that are used to generate highly reactive hydroxyl radicals that can degrade or mineralize various types of organic and inorganic pollutants in wastewater. One of the most widely used homogeneous AOPs is the Fenton reaction, which involves the reaction of hydrogen peroxide with ferrous ions (Fe^{2+}) to produce hydroxyl radicals. The Fenton reaction is typically carried out at acidic pH conditions, which favors the formation of Fe^{2+} ions. The hydroxyl radicals generated in the Fenton reaction can oxidize a wide range of organic compounds, including aromatic compounds, dyes, and pharmaceuticals. Another example of a homogeneous AOPs is the photo-Fenton reaction, which combines the Fenton reaction with UV light to generate even more hydroxyl radicals. In this process, UV light is used to excite the Fe^{2+} ions, which react with hydrogen peroxide to generate hydroxyl radicals. Homogeneous AOPs have several advantages, including high reaction rates, high efficiency, and the ability to treat a wide range of pollutants. However, they also have some limitations, such as the high cost of the metal catalysts, the requirement for low pH conditions, and the potential for the formation of sludge or by-products that may require further treatment. Additionally, homogeneous AOPs are not effective for treating pollutants that are present in high concentrations or in complex matrices. The homogenous Fenton is defined as the Fenton reaction in which iron salts are used as a catalyst. However, these processes are characterized by many disadvantages. Among them, we distinguish the generation of ferric hydroxide sludge at pH values above 4.0. The sludge has to be removed, which generated additional costs in the process. Moreover, the catalyst is difficult to recycle and reuse. Other problems with the method are high energy consumption and limitation of the operating pH range [33–37].

In a heterogeneous Fenton process, the catalyst and the wastewater or solution to be treated are in different phases. Typically, the catalyst is immobilized on a solid support, such as a porous ceramic or metal oxide material. The wastewater is then passed over or through the catalyst, allowing the catalyst to generate highly reactive hydroxyl radicals that can degrade or mineralize various types of organic and inorganic pollutants. The immobilized catalyst used in heterogeneous Fenton processes is typically based on iron or other transition metals, such as titanium, manganese, or copper. The catalyst can be prepared by impregnation, precipitation, or other methods that involve depositing the metal ions onto the support material. The catalyst can also be modified with various agents, such as carbon, to enhance its reactivity and stability.

Heterogeneous Fenton processes have several advantages over homogeneous Fenton processes. One of the main advantages is that they allow for the reuse of the catalyst, reducing the overall cost of the process. Heterogeneous Fenton processes also have a wider pH range of operation, which makes them more versatile for treating different types of wastewater. Additionally, they do not require the addition of acid to lower the pH, which can result in reduced sludge formation and by-product formation [38,39].

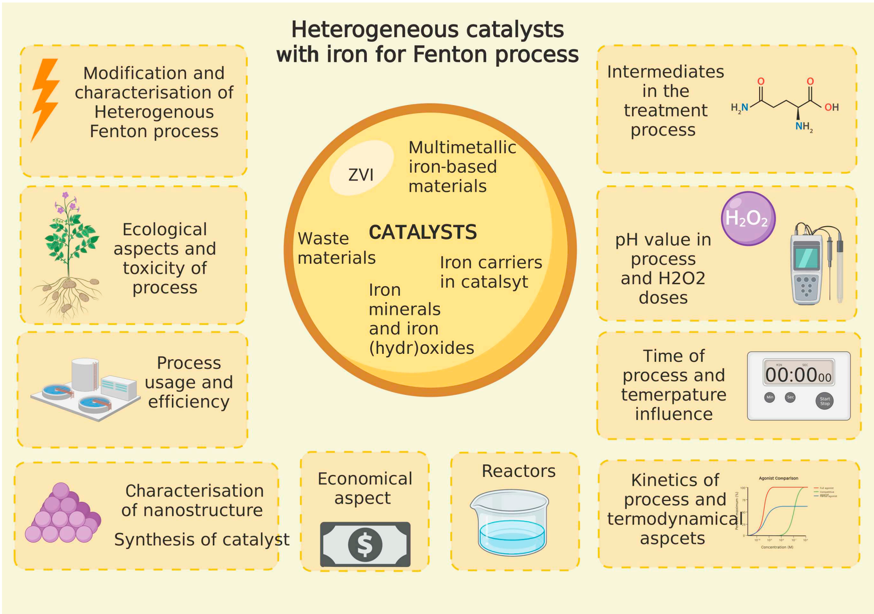

FIGURE 1.4 Summary of the content of the book.

Source: Own work.

Therefore, scientists more often use the heterogeneous Fenton process to degrade wastewater. This study aims to provide brief and summarized information about heterogeneous Fenton processes usage. The secondary aim of the article is to summarize the current state of knowledge regarding the conditions and parameters of the process (Figure 1.4).

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Introduction

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Application and Efficiency of the Process

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Economical Aspects and Hydrogen Production

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