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A Different Approach to the Removal of Organic Pesticide Waste: The Effect of Sono-Fenton and Ozone-H_2O_2 processes

Murat KIRANŞAN*

Department of Chemistry and Chemical Processing Technologies, Vocational School of Gumushane, University of Gumushane, 29100-Gümüşhane, Turkey.

--ABSTRACT--

Water is one of the vital natural resources for living on earth. However, speedy industrialization and urbanization cause the formation of great amounts of the wastewater containing various pollutants. Water pollution is a major problem worldwide. Intensive industrial and agricultural activities; toxic heavy metals, organic compounds, phenols, pesticides and different pollutants such as waste water and cause serious environmental pollution. Pesticides are toxic organic compounds used to protect plants, increase agricultural yields and prevent plant diseases. Organic pesticides are a general name for bioactive compounds that harm human health and ecosystems. Pesticides are compounds designed to be toxic to plant pests and sources of disease. As the world population grows, new and powerful pesticides are needed by researchers and manufacturers due to the need for more food. Although it has positive aspects such as increasing productivity in agricultural production, preventing diseases and product losses and obtaining quality products it poses a risk in terms of human health and environmental pollution. The main usage area of pesticides is agricultural activities. There are several factors that the effects of a pesticide in the environment, including chemical structure, physical properties, method of application, climate and agricultural conditions. During the application of the pesticides, some of them disappear by evaporation while some remain on the plant and soil surface. Pesticides remaining on the soil surface can pass into river and lake waters with the effect of precipitation. Organic pesticides are very difficult to remove from wastewater because they are strong pollutants and complex molecules. Classical treatment methods are not preferred for the removal of organic pesticides. Advanced oxidation processes are preferred for the removal of organic pesticides due to high efficiency and low cost. Therefore; ozonation (O_3), fenton (H_2O_2/Fe^{2+}) and sonication (US) processes are the preferred processes for the removal of organic pesticides among advanced oxidation processes. The ozone molecule (O_3) has a strong oxidizing property and is effectively used for the degradation of the organic pesticides. With the synergistic effect of hybrid sono-fenton and ozone-fenton processes, the removal efficiency of organic pesticides will increase further.

KEYWORDS; *Organic pesticides; Effect of fenton proces; Sono-fenton proces; Ozonation; Environmental risk*

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I. INTRODUCTION

In recent years, environmental pollution caused by factors such as the speedy increase in the human population and the acceleration of developments in the technology and industry has become a great problem worldwide. The main elements of environmental pollution are domestic and industrial wastes [1]. Due to the increasing environmental pollution, the importance of water for the ecological system and living life is increasing day by day. Organic and inorganic substances such as heavy metals, dyes, pesticides, pharmaceutical wastes are highly toxic compounds that contaminate water resources [2]. Pesticides are any substance or mixture used for the prevention, destruction and control of all kinds of harmful plant and animal species that may step in with agricultural production [3]. Food production is needed as a result of the speedy increase in the world population. Organic pesticides provide important yield increase in agriculture by controlling pests. At the same time, they provide important contributions to human health by preventing diseases caused by insects [4]. The fact that natural organic pesticides are expensive causes the excessive use of the synthetic pesticides to increase production efficiency in our country as well as in the whole world [5]. Pesticides classified as toxic substances are named as insecticides (against insects), herbicides (against weeds), fungicides (against fungi), acaricides (against bark and parasites), rodendicides (against rodents) and mollusides (against mollusks) [6].

Although the use of the pesticides benefits agricultural production, they are life-threatening to humans and other living organisms [7]. Today, organic pesticides released into the environment are harmful to human health and has been proven to a potential risk to other living things [8]. Although environmental pollution caused by pesticide wastes is tried to be solved in the developed and developing countries, humans and other living things cannot to be completely protected from the negative effects of pesticides [9]. The accession of pesticide wastes to water resources is controlled by different physicochemical factors such as soil texture, permeability, adsorption capacity and the solubility of the pesticide in water [10]. As a result, excessive use of pesticides used in agricultural production, pollution of water resources and soil is an important threat to the ecosystems of aquatic organisms [11]. Pesticides can easily get into surface waters with careless or unconscious pesticide applications and they can also enter or leak into groundwater by irrigation or rain [12]. Since pesticides with polar properties dissolve well in water, they easily reach groundwater resources without accumulating in the soil. Therefore, it increases the risk of the contamination of groundwater [13]. Groundwater is the most important source of drinking water, while groundwater contains more living things than surface water. Pesticides reach river beds and soil without being filtered polluting groundwater resources. Therefore, groundwater is undefended to pesticide hazard [14].

1.1 Methods Used in the Removal of Pesticides from Waste Water

Today, the detection of many pesticide residues in the natural waters and wastewater creates a serious concern for life. Despite the low concentrations of pollutants they contain, the development of the appropriate techniques for the detection and removal of all pollutants, including pesticides in water has become an important process [15]. Many techniques such as precipitation, photocatalytic degradation, chemical oxidation, membrane filtration, ion exchange, biological treatment and adsorption are used to remove organic pesticides from wastewater. Among these techniques, the adsorption process has come to the fore as a highly efficient, low cost, simple and easily applicable technique [16].

For the removal of pesticides from water, methods such as the photocatalytic oxidation [17], biological degradation by the photo-fenton process [18], chemical/electrochemical oxidation [19], aerobic oxidation [20], nanofiltration membranes [21], ozonation [22] are preferred. Another remarkable process among advanced oxidation processes is the sonolytic and sonocatalytic processes [23]. Hybrid processes are preferred for the removal of organic pesticides from wastewater. Among these processes, the preferred sono-fenton and ozone-H_2O_2 processes.

II. METHODS

2.1 Proces of Fenton

Fenton reactions were discovered in 1894 during the research of Henry John Horstman Fenton [24]. It was found by Haber and Weis in 1934 that the active oxidizer in the Fenton reaction is hydroxyl (OH•) radicals [25]. The Fenton reaction is based on the production of hydroxyl radicals (OH•) with high oxidation potential as a result of the reaction of H_2O_2 and Fe^{2+} ions in acidic aqueous solution [26]. The Fenton mechanism mainly occurs in the absence of the organic compounds with the following reactions (Eq. (1-7)) [27].

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^{\bullet} + OH^{-} \quad (1)$$
$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2^{\bullet} + H^{+} \quad (2)$$
$$OH^{\bullet} + H_2O_2 \rightarrow HO_2^{\bullet} + H_2O \quad (3)$$
$$OH^{\bullet} + Fe^{2+} \rightarrow Fe^{3+} + OH^{-} \quad (4)$$
$$Fe^{3+} + HO_2 \rightarrow Fe^{2+} + O_2H^{+} \quad (5)$$
$$Fe^{2+} + HO_2 + H^{+} \rightarrow Fe^{3+} + H_2O_2 \quad (6)$$
$$2HO_2 \rightarrow H_2O_2 + O_2 \quad (7)$$

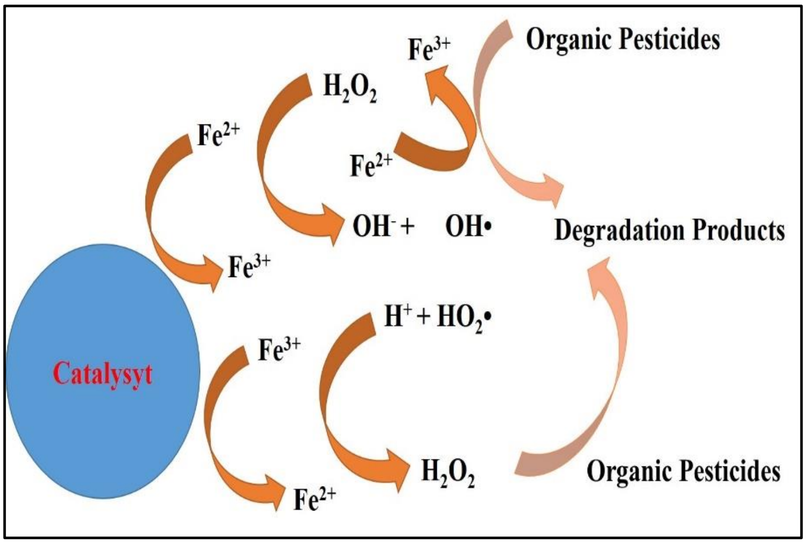

Fig. 1 Schematic representation of the fenton oxidation process [27].

The schematic representation of the fenton oxidation process is shown in Fig. 1. The Fenton process is a promising advanced oxidation process applied by the directly adding a mixture of H_2O_2 and Fe^{2+} salts to wastewater. The most important feature of the fenton process is that it is used for the treatment of both organic and inorganic compounds [28]. The Fenton process has double treatment properties due to the hybrid effect of oxidation and coagulation processes. Fenton oxidation has wide application in the treatment of the non-biodegradable wastewater in AOPs processes [29]. Fenton oxidation process is applied for the oxidation of the organic compounds with hydroxyl radicals with high oxidizing power produced by the reaction between H_2O_2 and Fe^{2+}. It has been reported that toxic compounds in wastewater such as the phenol, pesticide and herbicide can be removal by using fenton reaction [30].

2.2 Proces of Sonolytic

Sonolytic and sonocatalytic oxidation is based on the principle of removal and degradation of the organic pollutants in water with the help of ultrasound [31]. Ultrasound used in the sonolytic applications is defined as sound higher than 16 kHz (16000 cycles/s) which is outside the upper limits that the human ear can hear. During the cycle of the ultrasound wave, the water molecules separate and combine.Bubbles formed as a result of the ultrasonic cavitation reach critical sizes after a few cycles. The bubbles formed have a stable lifetime of less than 2 microseconds and a degradation time of less than 100 ns [32]. Ultrasound is shown as “)))” in reactions and it provides the separation of water molecules and forms reactive oxygen derivatives according to the reaction steps seen in the equations below (Eq. (8-14)) [33].

$$H_2O +))) \rightarrow \bullet OH + \bullet H \quad (8)$$

$$\bullet OH + \bullet OH \rightarrow H_2O + O\bullet \quad (9)$$

$$\bullet OH + H_2O \rightarrow H_2O_2 + H\bullet \quad (10)$$

$$H\bullet + \bullet OH \rightarrow H_2O \quad (11)$$

$$H\bullet + H\bullet \rightarrow H_2 \quad (12)$$

$$O\bullet + O\bullet \rightarrow O_2 \quad (13)$$

$$O\bullet + H_2O \rightarrow 2\ \bullet OH \quad (14)$$

As can be seen in sonolytic reactions, atomic oxygen, atomic hydrogen, hydroxyl, hydroperoxyl radicals and hydrogen peroxide are formed as a result of ultrasonic cavitation. The stability of these radicals is low and their willingness to reaction is high. Radicals oxidize the pollutant molecules by the reacting with the organic pollutant molecules in the solution ambiance [34].

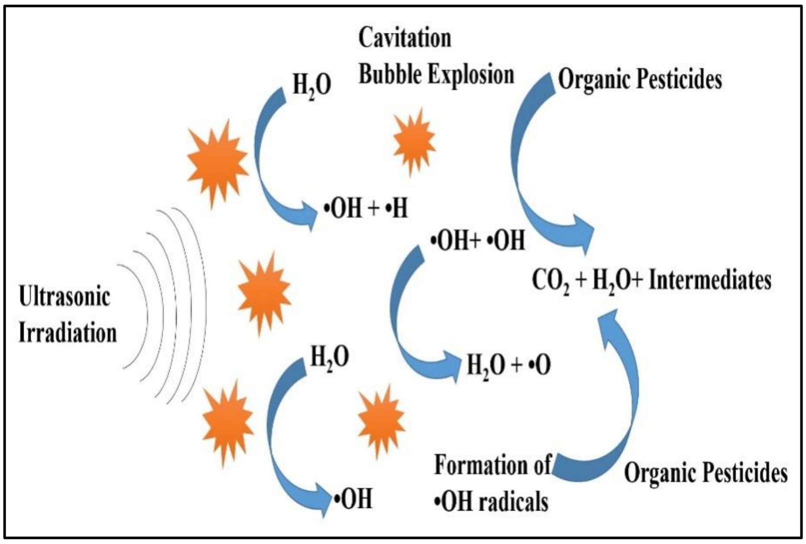

Fig. 2 Schematic representation of the sonolytic oxidation process [35].

The schematic representation of the sonolytic oxidation process is shown in Fig. 2. There are many factors that affect the sonolytic and sonocatalytic cavitation and therefore the amount of the reagents formed as a result of ultrasound. Sonolytic and sonocatalytic reactions are affected by solution temperature, dissolved gases and their species, ambient pressure and frequency of ultrasound [35]. Ambient temperature is another parameter that affects ultrasonic cavitation. The effect of ambient temperature on ultrasonic cavitation is to provide more bubbles as a result of the solvent evaporation more easily with the increase in the vapor pressure.It has been reported that increasing the temperature decreases the sonochemical effect [36].

2.3 Ozonation Process

Ozone molecule (O_3) is a uncolored gas at room temperature with a distinctive odor. Ozone molecule (O_3) is the most effective germicidal and deodorizer known. O_3, formed by ultraviolet rays and electric arcs that occur during lightning, protects the environment of the earth and living things against the radiation effect of the sun [37]. The ozone molecule (O_3) is a very strong oxidizing agent consisting of a single bond and a double bond. Ozone has two resonance structures that can be transform into each other. Molecular ozone reaction as a dipole substance as an electrophile or nucleophile it has a high oxidizing ability to degradation organic pesticides [38].

The degradation of the ozone molecule be formed according to the circular chain process. The process starts by the addition of a base such as OH^- or by the introduction of H_2O_2 or photolysis of O_3. The most important feature of the chain degradation mechanism is the formation of the hydroxyl radicals ($OH^•$). Hydroxyl radicals ($OH^•$) are a very reactive species that can reaction with almost all organic substances [39]. The schematic representation of the ozonation oxidation process is shown in Fig. 3.

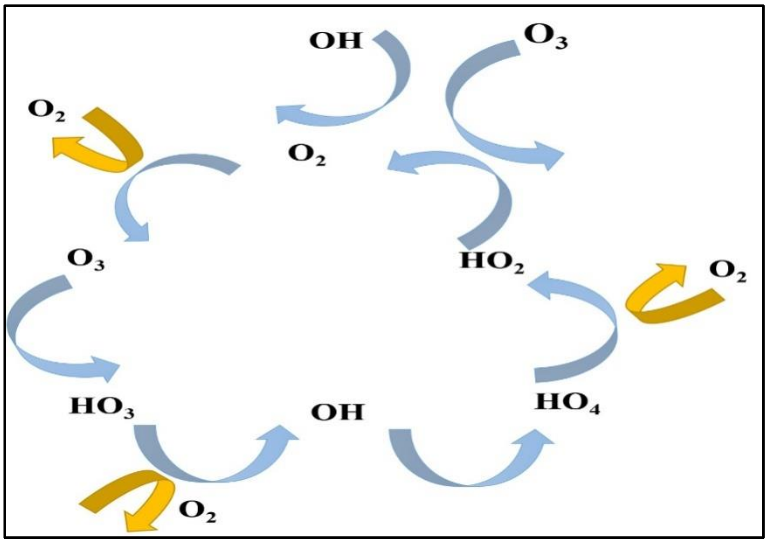

Fig. 3 Schematic representation of the ozonation oxidation process [39].

The reactions that be formed as a result of the ozonation process are as follows (Eq. (15-19)) [39].

$$O_3 + OH^- \rightarrow HO_2^{\bullet} + O_2^{-\bullet} \quad (15)$$

$$HO_2^{\bullet} \rightarrow O_2^{-\bullet} + H^+ \quad (16)$$

$$O_3 + O_2^{-\bullet} \rightarrow O_3^{-\bullet} + O_2 \quad (17)$$

$$O_3 + H^+ \rightarrow HO_3^{\bullet} \quad (18)$$

$$HO_3^{\bullet} \rightarrow OH + O_2 \quad (19)$$

III. RESULTS AND DISCUSSION

3.1 Sono-Fenton Processes (US/H_2O_2/Fe^{2+})

For the degradation of organic pesticides, the sono-fenton (US/H_2O_2/Fe^{2+}) process continues with a synergistic effect reaction. In this process, a synergistic effect occurs between the fenton reagent and the speedy degradation of ultrasound Fe-OOH^{2+} to Fe^{2+} and OOH• (Eqs. (20)) [40].

$$Fe\text{-}OOH^{2+} +))) \rightarrow Fe^{2+} + OOH\bullet \quad (20)$$

Fe^{2+} ion creates a cyclic mechanism as a result of reaction with H_2O_2 to form active radicals. The synergistic effect of Fenton's reagent with ultrasound not only increases the reaction rate of Fe-OOH^{2+} to Fe^{2+}, but also increases radical production by reactions between Fe^{2+}/H_2O_2 or Fe^{3+}/H_2O (Eqs. (21-23)) [41].

$$Fe^{2+} + H_2O_2 +))) \rightarrow Fe^{3+} + OH\bullet + OH^- \quad (21)$$

$$Fe^{3+} + H_2O +))) \rightarrow Fe^{2+} + OH\bullet + H^+ \quad (22)$$

$$Fe^0 + 2\,H_2O +))) \rightarrow Fe^{2+} + 2\,OH\bullet + H_2 \quad (23)$$

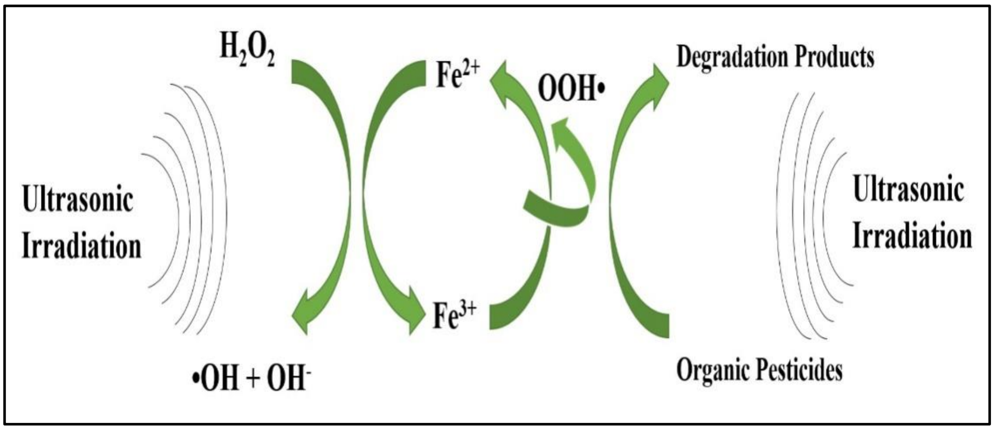

Fig. 4 Schematic representation of the sono-fenton oxidation process [41].

The schematic representation of the sono-fenton oxidation process is shown in Fig. 4. Different complex reactions occur as a result of the sono-fenton process applied for the degradation of the organic pesticides. More hydroxyl (OH•) radicals are formed when the sono-fenton process is applied to organic pesticides than when ultrasound and Fenton processes are the applied alone. It can be said that the degradation efficiency increases as a result of thermal division oxidation reactions in ultrasonic cavitation bubbles [42].

3.2 Ozonation-H_2O_2 processes (O_3/H_2O_2)

In recent years, hydrogen peroxide (H_2O_2) has been widely used as an oxidant to removal hazardous pollutants from different industrial wastewaters. The hybrid O_3/H_2O_2 system provides with high efficiency removal of organic pesticides. Hydrogen peroxide accelerates the decomposition of the ozone molecule (O_3) to form hydroxyl radicals by the electron transfer mechanism represented by the following equations (Eqs. (24-26)) [43].

$$O_3 + H_2O_2 \rightarrow 2\,OH^{\bullet} + 3O_2 \quad (24)$$

$$H_2O_2 \rightarrow HO_2^- + H^+ \quad (25)$$

$$HO_2^- + O_3 \rightarrow HO_2^{\bullet} + O_3^{\bullet -} \quad (26)$$

The Ozone/Hydrogen peroxide (O_3/H_2O_2) process is an O_3/H_2O_2 process known as peroxide which involves the oxidation of the pollutant by direct and indirect ozone reaction mechanism. Hydrogen peroxide reaction with ozone when present as an anion, HO_2^-. The reaction rate depends on the initial ozone/hydrogen peroxide concentration [44]. H_2O_2 has strong pollutant degradation, even with a small amount of ozone. Hydrogen peroxide not only increases the reaction rate, but also improves the wastewater quality [45].

IV. CONCLUSION

Advanced oxidation processes are preferred for the removal of the pollutants with complex molecules and difficult to decompose such as organic pesticides. Hybrid-AOPs the highest yields in the removal of organic pesticides from wastewater from advanced oxidation processes. Intensive hydroxyl radical production as a result of the synergistic effect of the hybrid processes ensures effective removal of organic pesticides a wide pH range. Therefore, the use of highly developed hybrid processes for efficient mass transfer makes the technology sustainable.

The removal of the pesticide wastes in the water ambiance by fenton processes is both an economical and an effective process. Fenton and fenton-like processes are that increase the production of hydroxyl radicals. The positive effects of the sonolytic process are due to acoustic cavitation, that is the formation growth and collapse of bubbles in the liquid. Sono-fenton ($US/H_2O_2/Fe^{2+}$) oxidation studies disclose that as the ultrasonic power energy increases, the amount of degradation of the pesticide wastes in the pollutant environment increases. The reason the O_3/H_2O_2 hybrid process is effective in the removal organic pesticides is the high oxidation potential of the ozone molecule.

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