

Fenton Reagent for Organic Compound Removal in Wastewater

Vic Austen^{a,b}, Cindy Suyitno^{a,§}, Tesalonica Yakoba Priskadianti Ratu Gah^{a,§}, Philemon Sugiarta^{a,§}, Shella Permatasari Santoso^{a,c}, Felycia Edi Soetaredjo^{a,c*}, Kuncoro Foe^d, Artik Elisa Angkawidjaja^c, Yi-Hsu Ju^{c,e}, and Suryadi Ismadji^{a,c*}

^aDepartment of Chemical Engineering, Widya Mandala Surabaya Catholic University, Surabaya 60114, Indonesia

^bIntan Permata Hati, Raya Kedung Baruk No.112-114, Surabaya 60298, Indonesia

^cDepartment of Chemical Engineering, National Taiwan University of Science and Technology, Taipei 10607, Taiwan

^dFaculty of Pharmacy, Widya Mandala Surabaya Catholic University, Surabaya 60112, Indonesia

^eGraduate Institute of Applied Science, National Taiwan University of Science and Technology, Taipei 10607, Taiwan

[§]These authors contribute equally

*Corresponding Authors: felyciae@yahoo.com (Tlp. +62-31-3891264 ; Fax +62-31-3891267)

Abstract

The improper treatment of wastewater has cost humanity a large amount of access to clean water. Treating wastewater, by definition, means to remove pollutants, either physically or chemically. A chemical method of treating wastewater, the Fenton process, was deemed useful for the job. It includes a solution-based reaction that produces radicals to oxidize and break pollutants down. Variations of the Fenton process, each with their unique method, have been developed to increase the process's efficacy and efficiency further. Admittedly, however, the information on this subject is relatively few, when compared to other more recent methods of treatment. This paper aims to present and discuss a wide variety of information on the Fenton process and its derivatives, including Electro-Fenton, Sono-Fenton, and Photo-Fenton, among others.

Fenton process for a chemical method of treating wastewater includes a solution-based reaction that produces radicals to oxidize and break pollutants down. Variations of the Fenton process, each with their unique method, have been developed to increase the process's efficacy and efficiency further. This review aims to present and discuss a wide variety of information on the Fenton process and its derivatives, including Electro-Fenton, Sono-Fenton, and Photo-Fenton, among others.



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Authors Biography



Vic Austen is a senior high school student in Intan Permata Hati School Surabaya. He has been taken research internship in Process Laboroty, Department of Chemical Engineering, Faculty of Engineering, Widya Mandala Catholic University Surabaya. He is a brilliant high school students and able to conduct research in our research group.



Felycia Edi Soetaredjo is a professor in the Departement of Chemical Engineering, Faculty of Engineering, Widya Mandala Catholic University Surabaya. She was graduated from Taiwan Tech (National Taiwan University of Science and Technology). Currently, she is one of the vice dean in Faculty of Engineering.



Cindy Suyitno is third year student in the Department of Chemical Engineering, Faculty of Engineering, Widya Mandala Catholic University Surabaya. Currently, she is in Taiwan Tech University, Taiwan for joint degree program.



Kuncoro Foe is a senior lecturer in the Faculty of Pharmacy, Widya Mandala Catholic University Surabaya. He was gradutated from The University of Sydney, Australia. Currently, he is the Rector of Widya Mandala Catholic University Surabaya.



Tesalonica Yakoba Priskadianti Ratu Gah is third year student in the Department of Chemical Engineering, Faculty of Engineering, Widya Mandala Catholic University Surabaya. Currently, she is in Taiwan Tech University, Taiwan for joint degree program.



Artik Elisa Angkawijaya is an Assistant Professor in the Department of Chemical Engineering, Taiwan Tech (National Taiwan University of Science and Technology). She was graduated from Taiwan Tech and had some experiences as postdoc in Academia Sinica before joint Taiwan Tech as Assitant Professor.



Philemon Sugiart is third year student in the Department of Chemical Engineering, Faculty of Engineering, Widya Mandala Catholic University Surabaya. Currently, she is in Taiwan Tech University, Taiwan for joint degree program.



Yi-Hsu Ju is a honorary chair professor in Graduate Institute of Applied Science and Technology, Taiwan Tech (National Taiwan University of Science and Technology). He is an Chemical Engineer



Shella Permatasari Santoso is a lecturer in the Department of Chemical Engineering, Faculty of Engineering, Widya Mandala Catholic University Surabaya. She was graduated from Taiwan Tech (National Taiwan University of Science and Technology). Currently, she is deputy of Chemical Engineering Department.



Suryadi Ismadji is a professor in the Departement of Chemical Engineering, Faculty of Engineering, Widya Mandala Catholic University Surabaya. He was graduated from The University of Queensland, Australia. Currently, he is Dean in Faculty of Engineering.

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Introduction

The abundance of liquid water on Earth's surface is one of the main reasons life on earth exists. Covering 71% of its surface area, water itself can be found virtually anywhere on the Earth. However, clean water is scarce due to pollution of the freshwater sources by either industrialization or simply negligent humans. Considering the amount of wastewater the industrialized world creates, it is alarming that most of it (about 80%) is left untreated and disposed of as is to the environment [1]. Unfortunately, our drinkable water sources are finite: we have access to less than 1% of the freshwater on Earth, the remainder being trapped in and under the glacial ice sheets of the Arctic and the Antarctic. If things continue as is, the clean water crisis now will only pale in comparison to the projected crisis in 2050, when worldwide freshwater demand is anticipated to be one-third higher than it is now [1]. Even though water is an essential element in sustaining human life, humans are still less aware of the diminishing availability of clean water. Excessive use of freshwater for various industrial activities will ultimately damage the environment.

The use of water in various industrial processes will produce wastewater that should be treated before being discharged back to the environment. Currently, several wastewater treatment technologies are available, with membrane filtration, ultraviolet irradiation, chemical oxidation, and the use of microorganisms to digest organic contaminants being among them. One of the available processes is Fenton oxidation (Figure 1), with said process capable of considerably eliminating organic recalcitrant and toxic compounds, and improving organic compounds' biodegradability. The quality of the leachate post-Fenton treatment is

significantly improved, in terms of color, odor, and organic content (or lack thereof).

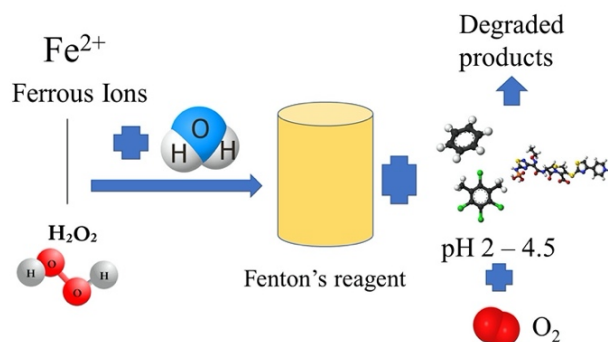


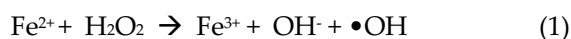
Figure 1. Traditional Fenton oxidation.

Table 1. Current progress on Fenton reactions for the removal of organic compounds.

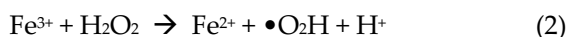
Process	Emphasis of study	Ref.
Fenton	The factors affecting the efficiency of the Fenton process in treating various organic substances	[2]
Photo-Electro Fenton	The recent development of EAOPs (electrochemical advanced oxidation processes)	[3]
Heterogeneous Fenton oxidation	Discuss the role of heterogeneous Fenton oxidation for the treatment of hazardous landfill leachate	[4]
Fenton/Photo-Fenton	One of the paper emphasizes the degradation of sulfamethoxazole using various methods such as Fenton and Photo-Fenton process	[5]
Fenton	Comparison of several processes for the treatment of wastewater containing triclosan	[6]
Electro Fenton and bio-electro Fenton	This review focusses on the cost-effectiveness of the removal of pharmaceutical compounds using electro Fenton and bio-electro Fenton processes	[7]
Fenton, electro-Fenton, photo-Fenton	The roles of Fenton processes on the degradation of pharmaceutical contaminants	[8]
Photo-Fenton	Environmental and medical applications of Photo-Fenton	[9]
Electro-Fenton	The review focuses on the innovative application of electro-Fenton for degradation of hazardous waste	[10]

Several reviews on the Fenton process are available in the literature (Table 1). These review papers discuss various aspects of the Fenton process in treating wastewater containing various organic compounds.

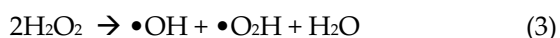
According to Babuponnusami and Muthukumar [2], Fenton oxidation is defined as using an aqueous solution containing both hydrogen peroxide and Iron(II) ions to oxidize both organic and inorganic compounds in wastewater. The oxidative reaction of Fenton's reagent can be written as follows:



The ferric ions then undergo another Fenton-like reaction; it will be reduced by the excess of H_2O_2 , regenerating the Iron(II) ions and producing hydroxyl radicals.



Therefore the total reaction equation, found by eliminating spectator ions and disregarding the regenerated Iron(II) ions, can be written as follows:



There is room for modification in the Fenton process, which includes, but is not limited to the utilization of ultrasound (Sono-Fenton process – SF) [8,11–14], anodic oxidation (Electro-Fenton Process – EF) [15–18], using UV light and adding ferric or ferrous oxalate ions (Photo-Fenton process –PF) [19–23], utilizing both ultrasound and ultraviolet light (Sono-Photo-Fenton process – SPF) [24–26], utilizing both ultrasound and anodic oxidation (Sono-Electro-Fenton process – SEF) [27], and utilizing a combination of electrochemical and photochemical properties of UV radiation (Photo-Electro-Fenton process – PEF) [2]. Other notable modifications have been developed, such as Solar-Photo-Electro-Fenton process (SPEF) [28], Peroxi Coagulation (PC), Photo-Peroxi Coagulation (PPC), Photo-Electro Catalysis (PEC), Ferred Fenton process, and Electrochemical Peroxidation (ECP) [29].

One of the most effective and widely-used modifications of the Fenton Process is the EF. In this modification, a specific electrochemical reaction is involved in the continuous generation of H_2O_2 , with either an oxygen atmosphere or air being fed in the cathode, with an addition of iron catalysts to produce hydroxyl radicals via Fenton's oxidation reaction [29]. Some advantages of the EF process include:

- A lower overall safety hazard for the process, attributed to the fact that this process indirectly forms hydrogen peroxide, instead of directly handling the radical compound.
- More control in degradation kinetics, and a significantly higher degradation rate of the hazardous compounds due to the constant supply of ferrous ions in the cathode, made possible by constant regeneration of said ferrous ions.
- Less sludge is formed as a byproduct of the reaction.
- A lower operation cost, given that the reaction is run at its optimum parameters.

The essential parameters in conducting Fenton's process, including any derivative or modification, are the reaction pH, the concentration of Fenton's reagent, dissolved oxygen level, effluent pH, the mode of addition and multi-stage treatment, the temperature used when conducting the recycling of iron sludge, and the presence of UV irradiation [30]. The optimal pH of classical, EF and PF process generally lies between 2-4.5. The dosage of the reagent affects both the efficiency of the removal rate of organic compounds and operating costs. A higher concentration of the reagent will result in a higher percentage of organic compound removal, but with an excess of it, the increment will gradually become marginal, and sludge floatation may occur. In terms of Dissolved Oxygen (DO), the usage of saturated air (or oxygen) has been proven to better perform treatment compared to N_2 - and helium-saturated conditions. The pH of the effluent may also affect the removal rate of hazardous material for wastewater treatment. Said optimum pH lies in the range of 2-9 for coagulation after the oxidation. The multiple steps in Fenton treatment are conducted to improve the removal of Chemical Oxygen Demand (COD) in wastewater. Another way to increase COD removal, while simultaneously increasing the coagulated sludge's settling velocity, reducing sludge production, and decreasing coagulant consumption is to recycle the iron sludge from the process, and using it in the coagulation step before the Fenton's process. Temperature differences slightly increase the removal of COD in leachate treatment. However, with high temperatures, the increase in the removal rate of COD becomes marginal due to the negative effect that temperature has on the decomposition of H_2O_2 . The last parameter, the presence of UV radiation, may improve the reduction of ferric ions. However, a study [31] has drawn the opposite

conclusion concerning the effect of UV irradiation on the reduction of ferric ions.

By this point, a large number of studies have been conducted by numerous individuals to find the optimum parameter and method in wastewater treatment via Fenton's process. Most of the studies apply the EF Process. In comparison, the derivatives of said process, namely the PEF Process and SEF are rarely discussed. This study aims to crystallize the most important aspects, including the mechanism, efficacy, and important significant result-altering factors of said rarely discussed the method to increase further the ease of access for those who may want to develop this field in the future.

Electro-Fenton (EF) Process

EF Process (Figure 2) was developed as an alternative method to the conventional Fenton reaction, albeit at a lower cost. In electrochemistry, H_2O_2 can be generated by the reduction of oxygen in the cathode (4), which in turn reacts with ferrous ions and pollutants (denoted as R) (equations 5-7). The reaction mechanism can be written as follows:

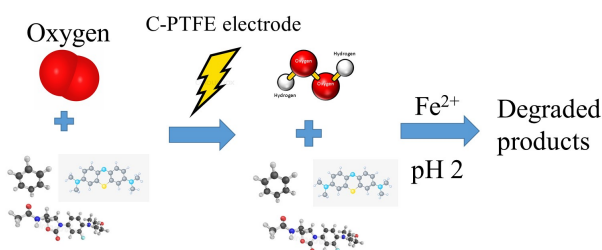
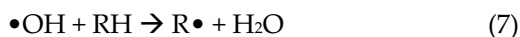
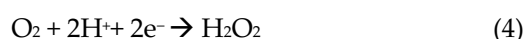


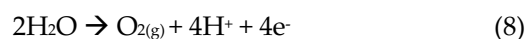
Figure 2. EF process.

So, instead of directly using the H_2O_2 solution, which can be dangerous at high quantities and concentration as the reactant, the EF Process uses H_2O_2 generated in situ in controlled amounts. To set off the Fenton reaction, the only step necessary is to add a ferrous catalyst. This method has been applied to remove organic pollutants in water, such as antibiotics [17], petrochemicals [32], aromatic benzene-derived compounds [33,34], dyes [15,35–38], and other organic pollutants [39–41]. Selections of the

experimental results of some of the studies on this topic, along with their operational parameters, are summarized in Table 2.

One of the parameters to be considered when conducting an EF experiment is the cathode material selection. Sirés *et al.* [42] provided an experimental result of antimicrobial triclosan and triclocarban degradation using EF, and found Pt/carbon felt electrode as the best choice, in terms of the generation of the hydroxyl radicals, which serve as the primary oxidant source in the Fenton oxidation method. While some researchers prefer three-dimensional electrodes, such as gas diffusion cathodes (GDE) to reach a higher level of current density, oxygen solubility, and mass transport, Lei *et al.* [43] proposed an electrode design that consists of graphite chips coated with carbon black and polytetrafluoroethylene (C-PTFE) instead of carbon cloth to prevent gas bubbles and electrolyte outflow to the gas chamber.

Yuan *et al.* [44] suggested a novel modification that changed the method of H_2O_2 generation. This suggestion is based on the fact that H_2O_2 can be produced as a result of the reaction between H_2 gas and O_2 gas (equation 10). H_2 gas can be produced by the reduction of hydrogen ions in the cathode (equation 9), while O_2 gas can be produced as a result of the oxidation of water in the anode (equation 8). The total reaction can be written as follows:



As the acidic and basic compounds are formed in the cell, the adjustment of pH can easily be achieved. The results of their experiment on EF-assisted Rhodamine B (RhB) degradation shows that the H_2O_2 formation was dependent on acidic pH, and the RhB decay was optimum at pH 3 and 4, while the one catalyzed with Fe^{2+} was optimum at pH 2. The optimum current density was 50 mA. Although the amount of H_2O_2 produced by this method is less than the gas diffusion cathode method, this method costs less to operate, and its accumulated H_2O_2 concentration is higher compared to other methods, such as graphite electrodes, CNT, and carbon fiber cathode [45].

Table 2. Summary of EF process on various wastewater treatments.

Pollutant	Cathode-Anode	Current Intensity/ Density	pH	Catalyst	Result	Ref.
Orange II	Activated carbon fiber	100 mA	3	0.3 mM Fe ²⁺	96.7% removal of Orange II	[15]
Sulfamethazine	Carbon black – polytetrafluoroethylene modified graphite	50 - 300 mA	2, 3, 5, 7, and 9	0.3 mM Fe ²⁺	Combination Fe ²⁺ /H ₂ O ₂ , UV/H ₂ O ₂ gave more efficient mineralization (> 83.5%)	[17]
Phenol	graphite felt modified by carbon black and PTFE (cathode), and perforated dimensionally stable anode (DSA)	100 mA	3 - 10	0.3 mM Fe ²⁺	High efficiency of phenol removal was observed at pH 5 to 8	[33]
Benzene, toluene, and <i>p</i> -xylene (BTX)	Carbon and nickel (anode)	400, 600, 900 mA	3	5 mg/L Fe ²⁺	Degradation of BTX > 85% within 10 min	[34]
Bordeaux Red (E123)	Platinum (anode), and carbon felt (cathode)	50 – 250 mA	3	8.5 g/L Fe ²⁺	Complete removal of color was achieved within 4 hours	[35]
Amaranth	Commercial iron plate (anode), and commercial carbon plate (cathode)	0 – 0.2 mA/cm ²	3, 5, 7, and 12	Fe ²⁺	pH 3 was the optimum for degradation of Amaranth	[36]
Metanil yellow	Platinum plate (anode), and graphite (cathode)	6.6 mA	2.5	Fe ²⁺ , Co ²⁺ , Mn ²⁺ , Ni ²⁺ , and Ce ³⁺	Ni ²⁺ was a very effective catalyst for degradation of Metanil yellow	[37]
Triclosan	MOF-derived hierarchical Mn/Fe@PC modified cathode	0 - 100 mA	2, 3, 5, 7, and 8	Mn/Fe@PC	Direct reduction of cathode increasing the triclosan degradation	[39]
<i>p</i> -nitrophenol	Dimensionally stable anode (DSA), and carbon black modified graphite felt (cathode)	10, 15, 25, and 50 mA	3 - 9	Pre-magnetized Fe ⁰	Pre-magnetized Fe ⁰ could improve the degradation of <i>p</i> -nitrophenol	[40]

A potential modification to reduce production cost has been investigated by Kishimoto *et al.* [46] by reusing iron sludge for EF. This method was considered suitable for use with the batch separation model with Fe²⁺/HOCl as the Fenton-like reagents. Another study has also investigated the use of this iron-rich sludge as a coagulant in the Fenton process pretreatment [47].

The influence of electrical current on the removal efficiency of an organic compound, *p*-nitrophenol has been studied by Tian *et al.* [40]. In general, at a certain range of electrical current, by increasing the current, the rate of degradation of *p*-nitrophenol increased remarkably. By increasing the current, the rate of generation of hydrogen peroxide also increased, and leading to the higher formation of •OH. With the increase of •OH concentration in the

system, the degradation of *p*-nitrophenol also increased.

Photo-Electro Fenton (PEF) Process

With the help of electromagnetic radiation, such as UV-radiation or solar light, the Electro-Fenton process can be enhanced due to the increase of the electric current that can be absorbed into the system for the wastewater treatment process [48]. PEF can more efficiently remove organic compounds, relative to EF. In short, the PEF process improves the mineralization process, but on the other hand, it may add some cost to the process [49]. Several comparatively similar studies report that PEF has higher efficiency relative to EF when running with the same operational parameters, for the removal of, among others, benzene ring, drugs, antibiotics, herbicides, antimicrobial agents, and dyes in water treatment [50–58]. However, an observation by Deuna *et al.* [58] has found that a comparative study of acetaminophen degradation by EF and PEF process using a double cathode resulted in marginally different COD and acetaminophen removal, but the latter is more energy saving.

The results of the experiment conducted by Pajootan *et al.* [48] concluded that dyes (Acid Red 14 and Acid Blue 92) in colored wastewater could be treated by using the Electro Fenton process with CNT coated graphite as the cathode. The advantages of said material are the fact that it is accessible, low electrical resistance, is chemically inert, low cost, and has a significant potential over hydrogen evolution. The process used a constant intensity of current (0.14 to 0.30 A) and was assisted via UV-irradiation and the addition of TiO₂ to remove more than 90% of the dye in the wastewater. It has been observed that higher current density resulted in higher H₂O₂ formation, which led to higher removal efficiency. However, with the higher initial concentration of the dye, the increase of the removal efficiency declined. Efficiency can be enhanced by adding Iron(II) ions to the reaction mixture. However, an excess of said ions would result in progressively less increases. The most suitable electrolyte to be used was NaCl due to the active chlorine formation that can electro-oxidize the dyes. Kinetic studies show that the reaction best follows the kinetics of the pseudo-second-order.

Other innovations of this method include one that utilizes solar power. The sun's rays are rich in a UV light so that it can be used for a more economical source of UV light. SPEF process has been utilized to remove cresols [59], azo dye [60], and ibuprofen [61].

All comparisons with the EF process have shown that the SPEF Process is superior in terms of efficacy. However, the decoloration rate of the dyes via the two processes were similar. These results are attributed to the higher oxidation power in SPEF, in part due to photolysis and photodecomposition of ferric complexes that cannot be done by hydroxyl radicals. Therefore, SPEF can be applied as an energy-efficient method of wastewater treatment. Also, Skoumal *et al.* [61] reported that a higher degree of mineralization was achieved in the degradation of ibuprofen in various Fenton methods (such as EF, PEF, and SPEF) by utilizing a BDD anode instead of a Pt anode because the BDD anode has higher oxidizing power than a Pt anode when bonded with the hydroxyl radicals.

Sono-Electro Fenton (SEF) Process

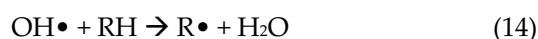
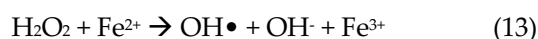
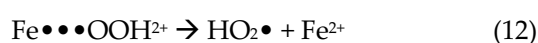
Şahinkaya [62] reported on the subject of the usage of SEF in textile wastewater treatment and found that it achieved better degradation of the pollutant (C.I. Reactive Black 5) and COD removal compared to EF due to a larger amount of oxidizing agent formed in SEF. The optimal parameter was reported to be similar to EF process. Mehrdad *et al.* [63] stated in his experimental observation that the SF process had higher efficiency of methylene blue removal compared to sonification and classical Fenton process under the same operational conditions due to collapsing cavitation bubbles in aqueous solution and a larger amount of hydroxyl radicals generated. This observation can be applied to the field of SEF. With an ultrasound wave power of 15 W, it increases the removal rate of Orange G, but in an additive effect, rather than the synergistic effect in the SF process [64]. Furthermore, it affects EF positively due to the increment of mass transport and activation of the electrode via the impurity layer removal at the electrode surface. It is proven in Oturan *et al.* [65] observation that the optimum condition for OH radicals formation for the degradation of herbicides 2,4-D and DNOC was under a 20 W ultrasound; a higher power ultrasound would not improve the result of the degradation, rather, excessively strong ultrasound diminishes the production of the radicals instead. However, in the degradation of the azo dye AB, due to its relatively weak and degradable bond, SEF is deemed unsuitable and is thought to be highly cost-inefficient.

Comparative studies found that the optimum pH to be around 3, and a current density of 100-250 mA as the optimal parameters. Initial Fe²⁺ and

additional initial H_2O_2 concentration can act as a catalyst. But so far, there is still a very limited number of studies conducted to find the optimum parameter and usage of this modification of Electro-Fenton, which has a significant potential for application in the field of wastewater treatment.

Fenton-like

While the Fenton reaction uses Fe^{2+} ions reacting with H_2O_2 to form oxidizing agent hydroxyl radicals, the ferric ions produced can also react with hydrogen peroxides to reform ferrous ions (equations 11–14):



Fe^{3+} salts are preferred over Fe^{2+} salts due to its lower cost. Shaobin Wang [66] in his 2008 comparative study between Fenton and Fenton-like reaction in dye decoloration concluded that both Fenton-like reaction, either with Fe^{2+} or Fe^{3+} , under the same operating condition (pH, temperature, initial concentration of reactants and catalyst) resulted in faster degradation rate than conventional Fenton oxidation, yet still only achieves a similar degree of degradation for both methods after 80 minutes. This is due to the lower oxidation capability of HO_2 radicals, relative to HO radicals. The kinetics of Fenton reaction follows pseudo-first order, while the Fenton-like reactions best follow the kinetics of a first-order reaction. The system depends on pH, initial Fe^{3+} concentration, and initial H_2O_2 concentrations. However, temperature itself affects the results minimally when tested on a large range (15-45 °C).

Other researchers, such as Rodríguez-Narváez *et al.* [67] studied conventional Fenton reactions as well as Fenton-like reactions for use in L-proline degradation, and found that the Fenton-like systems produced better results. Both Fenton-like reactions, homogenous and heterogenous, gave a higher L-proline removal rate compared to conventional Fenton reaction with the same operating conditions. The Fenton-like reaction did not differ much under different pH (3 and 7) or radiation ($\lambda = 365 \text{ nm}$, 12 W, 28.3 μ Einstein/min). Therefore, neutral pH can be used to reduce effluent costs. Also, Wang *et al.* [68] had used HNO_3 -modified coal fly ash (HFA) to perform an oxidation reaction in the treatment of p-

Nitrophenol pollutants, in which it is proven to be effective, stable, and reusable up to 9 runs of application with retaining removal percentage of > 91% after 9 runs. Increasing the temperature of the system does increase the efficacy of the treatment, but it is also imperative that the cost of said higher temperature be taken into account.

Photo Fenton (PF) Process

Ghaly *et al.* [69] defined the Photo-Fenton process as a reaction between H_2O_2 , UV radiation, and $\text{Fe}(\text{II})$ ion, in which the Iron(II) ions act as a photocatalyst and the H_2O_2 acts as the oxidizing agent (Figure 3). This process added H_2O_2 with $\text{Fe}(\text{II})$ ion using a UV light until degradation products produced. The advantages of this process are the fact that it is efficient, cheap, more efficient in terms of hydroxyl radicals produced per energy spent, low operational investment costs, and less energy consumed overall. The hydroxyl radicals produced are capable of oxidizing even the most chemically resistant organic molecules into forms easier to process.

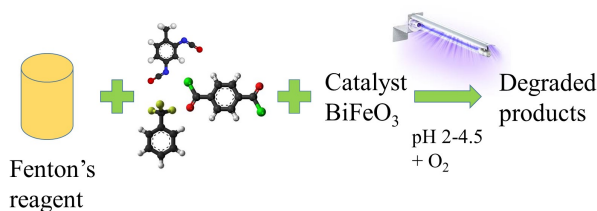


Figure 3. PF process.

In a study by An *et al.* [70], the process was used to remove organic pollutants with the aid of in situ surface-modified BiFeO_3 as a catalyst. The BiFeO_3 used were prepared by mixing several reactants such as iron nitrate, bismuth nitrate, 2-methoxyethanol, HNO_3 , citric acid, and ethylene glycol until it forms a sol. After that, the sol was heated until it becomes a viscous, brown resin-like material. As a result, a powder can be extracted by drying and used. This process is called the sol-gel method. These BiFeO_3 nanoparticles were then further modified with NTA (nitrilotriacetic acid) and EDTA (ethylenediaminetetraacetic acid) to form BiFeO_3 -NTA and BiFeO_3 -EDTA nanoparticles, respectively. Both nanoparticles also affect the PF process by increasing catalyst load and the number of active sites on the catalyst's surface. Furthermore, the BiFeO_3 nanoparticles have been proven to be able to decompose organic compounds in pollutants.

In a study done by Rodriguez *et al.* [71], PF process has been proven to decrease biorecalcitrant

concentration in wastewater from the textile industry. Via the Zahn-Wallens test, the wastewater was tested for biocompatibility. Wastewater treatment was done by extracting the wastewater, then react the raw wastewater with unacclimated municipal sludge. The pollutants were tested and found to be non-biodegradable. The authors also experimented to find the optimum operating parameters for the PF process. After a few trials, the authors concluded that the best condition so far is 1.43 mmol/l Fe^{3+} and 441.2 mmol/l H_2O_2 in solution, and the temperature is kept constant at 60 °C.

In a paper written by Ayodele *et al.* [72], phosphoric acid modified kaolin supported ferric-oxalate catalyst was synthesized for the purpose of removing phenol in wastewater by the PF process. The catalyst for this process was derived from raw kaolin (RK) clay, further modified with phosphoric acid by condensation. The resulting clay is called Phosphoric Acid Modified Kaolin (AMK). After AMK was successfully synthesized, it was dried in an oven and ground into a powder. This powder was then mixed with oxalic acid and iron hydroxide so to enable catalyst action. Thus, it is called as Phosphoric Acid Modified Kaolin Catalyst (AMCK). The authors investigated the catalytic activity of the synthesized AMCK by studying the action of the AMCK-assisted Photo-Fenton process in the removal of phenol in wastewater. To quantify the percentage removal of the phenol in wastewater, a UV-Vis spectrophotometer was used at 272.5 nm before and after the treatment.

A possible modification of the PF process was done by adding an iron-modified montmorillonite clay catalyst in the process. This idea was the core of Leon *et al.*'s study [73]. In this study, raw montmorillonite clay was dried and sifted through a filter that has a pore size of 250 μm . The ones that made through the said filter were used as the raw materials for the catalyst. Then, an aqueous solution of $[\text{Fe}_3(\text{OCOCH}_3)_7\text{OH}\cdot 2\text{H}_2\text{O}]\text{NO}_3$ was mixed with the fine clay, forming the catalyst Fe-PILC. The resulting solution was then filtered to extract the Fe-PILC, which was then washed with deionized water. To get the finished product, the resulting wet powder was dried at 60 °C under a calcine atmosphere. This catalyst's efficiency was investigated by testing the difference in the result of a Fe-PILC assisted batch reaction and an unassisted process. The catalyst was then found to increase the amount of Iron(II) and Iron(III) ions in the solution, explaining the high catalytic activity it exhibits in the PF process.

Sono-Fenton (SF) Process

In a journal by Liu *et al.* [74], a group of processes called the Advanced Oxidation Processes (AOPs) was deemed to be noteworthy instruments that should be developed regularly. The Fenton process, along with all of its derivatives and modifications, belongs to this group of processes. However, the overall data in the field of AOPs are minimal. The most pressing issue against the widespread use of AOPs is the lack of methods to pick the most profitable AOP for a given toxin. Thus, for AOPs to become a mainstay of the wastewater treatment process, future research must further study the reaction mechanisms, possible viability improvements, and optimizations for AOPs.

In Babu *et al.* paper [75], the author reported on the effect of ultrasound on AOPs. The ultrasound was utilized to decrease the concentrations of organic pollutants in wastewater that came from industrial, military, or commercial activities. There are many processes that constitute the ultrasound-assisted AOP group, including but not limited to sonolysis, Sono-Ozone process, Sono-Photo Catalysis, SF process, and the SPF process. Most of the time, ultrasound was integrated into one or more AOPs. Ultrasound treatment by itself is generally inaccessible in a biological setting since there is not many a condition whose treatment must resort to the admission of sonolysis (ultrasound treatment). The efficiency and accessibility of this treatment will be improved from here on out. For example, the inclusion of oxidants, such as hydrogen peroxide and/or potassium persulfate.

Another possible improvement is by blending two or more AOPs via ultrasonic assistance. This redesign makes it so that treatment does not require expensive external oxidizing agents, but it will call for redesigns of the apparatus for the execution of this combination of (or hybrid) AOPs. Curiously, however, hybrid AOPs exhibits a synergistic effect divergent from individual AOP structures. In short, no doubts are cast about the worth of ultrasound as a potent and essential tool in the developing field of AOPs.

In-Situ Oxidation Fenton (ISCO-Fenton) Process

One modification of the Fenton process is the addition of a transition metal catalyst to decompose hydrogen peroxide into hydrogen radicals homolytically (Figure 4). This modified Fenton

process is called the In-Situ Chemical Oxidation Fenton (ISCO-Fenton) process. The operating parameters of ISCO-Fenton are different from the conventional Fenton process, in the regard that it used a neutral environment, and the fact that the iron source used can be ore or an iron-containing salt [76].

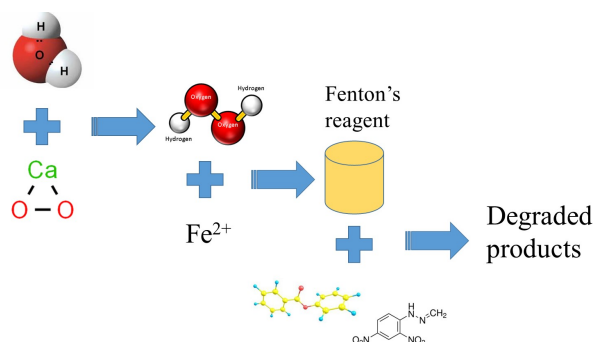


Figure 4. ISCO-Fenton process.

Northup and Cassidy [76] observed the oxidation of Perchloroethylene using a modified Fenton process that involves Calcium Peroxide (CaO₂) as the substitute for hydrogen peroxide solution. CaO₂ was chosen as the peroxide agent due to its efficiency when applied in in-situ chemical oxidation because H₂O₂ decomposes rapidly in soil. Furthermore, CaO₂ in solution is found to produce more H₂O₂ molecules than the same mass of liquid H₂O₂ because CaO₂ has a much higher oxidant efficiency when releasing H₂O₂ upon dissolution. After all, the dissolution of CaO₂ in aqueous solution results in less volatilization and greater oxidation of perchloroethylene than liquid H₂O₂ [76].

Watts *et al.* [77] observed the action of the modified Fenton's reagent in degrading tetrachloromethane in dense non-aqueous phase liquid (DNAPL). Tetrachloromethane is a compound that is unable to be oxidized by hydroxyl radicals. However, it can be broken down by the modified Fenton's reagent. Fenton systems have extremely reactive oxidants, but they are not reactive if the pollutants are sorbed contaminants. Also, they are probably too short-lived to degrade DNAPLs. In this paper, tetrachloromethane was broken down by a combination of Iron(III) and pyrolusite. Pyrolusite, a metal oxide, was used as metal oxides are considered natural catalysts for ISCOs [77].

Combination Fenton Oxidation and Subcritical Water Processes

In one of our industrial projects, we have utilized the Fenton oxidation and its modified

processes to treat wastewater from the painting department of audio-video and home appliances factory located in Central Java, Indonesia. The wastewater from the company contains a high concentration of UV-resistance dyes (a mixture of various organic chemicals such as 4-methyl-2-pentanone, propylene glycol methyl ether acetate, ethylene glycol phenyl ether, toluene, trimethylolpropane triacrylate, *etc.*). The concentration of the UV-resistance dyes in wastewater was 8000 ppm. Using the traditional Fenton oxidation process (varying the dose of Fenton's reagent), the maximum removal efficiency was 74%. However, at low to moderate concentration of dyes (< 2000 ppm), the excellent removal efficiency (> 99%) was obtained.

Subcritical water is known as a sustainable reaction medium. Subcritical water is also a unique reaction medium; it acts as the catalyst during the process. Owing to its unique characteristic, we employed this sustainable reaction medium to degrade UV-resistant dyes in the wastewater. The subcritical water oxidation process was conducted at a range temperature of 120 to 240 °C and the pressure of 40 to 50 bar. The maximum removal efficiency (64%) was achieved at 200 °C and 40 bar. A combination of Fenton oxidation and subcritical water processes was also conducted to degrade the UV-resistant dyes. This combination process could reduce the concentration of UV-resistant dyes from 8000 ppm to 78 ppm.

Conclusions and Future Perspective

As the advanced oxidation treatment process, the Fenton oxidation process and its modified form could degrade a wide range of organic pollutants. Some studies discussed in this review paper have proven that the AOPs have a very potential application for the real industrial wastewater treatment process with one condition if it is economically feasible.

A large number of studies on the degradation of the various organic hazardous pollutants using Fenton oxidation and its modified forms have been conducted in the last three decades. These technologies have proven to be useful for the degradation of effluents from various industries such as pharmaceuticals, textiles, food, pulp, and paper, *etc.* [78]. The successful implementation of the Fenton oxidation technologies depends on several process parameters such as the temperature, the concentration of hydrogen peroxide as well as the

catalyst, and the pH of the system. Even these technologies successfully applied for the treatment of industrial effluent in the lab and pilot plant scale; however, the implementation of Fenton oxidation and its modified forms in the real wastewater treatment plant is still scarce. The main drawbacks of using Fenton oxidation and its modified forms in the industrial scale wastewater treatments are:

- The cost of hydrogen peroxide is high, which is not economically feasible for wastewater treatment.
- The catalyst used in the Fenton process is the homogeneous catalyst which is required further separation process before it can be discharged to the environment.
- Most of the heterogeneous catalysts studied are also expensive, and their reusability also low.

More extensive studies still required before these advanced oxidation processes can be implemented in large scale of industrial wastewater treatments. More studies on the development of heterogeneous catalysts using low cost and natural material should be conducted in order to make these processes feasible economically in industrial-scale applications.

Authors contribution

Author and coauthors have contributed in writing this review article. The topic of review has been a part of our research group work. The topic was conveyed by Felycia Edi Soetaredjo and Suryadi Ismadji. Vic Austen, Cindy Suyitno, Tesalonica Yakoba Priskadianti Ratu Gah, and Philemon Sugiarta are our students who wrote the first draft. Shella Permatasari Santoso, Artik Elisa Angkawijaya and Kuncoro Foe contributed on the review of current progress on Fenton reaction for organic compound removal. Prof. Yi Hsu Ju contributed on the discussion of review format and proofreading.

Conflict of Interest

There is no conflict of interest to declare.

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