

Amoxicillin oxidation by homogeneous and heterogeneous photocatalysis under solar irradiation

Abstract

In recent years, the presence of drug residues in water has become a major environmental issue. Among these pollutants is amoxicillin. Our aim is to propose effective methods for decontaminating wastewater containing amoxicillin. This study was carried out using the chemical oxygen demand method. The results showed that the rate of hydroxyl radical production from hydrogen peroxide and iron (II) ions increases in the presence of sunlight. Amoxicillin oxidation is optimal at a pH of 3 and a $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ ratio of 13.33. Amoxicillin degradation is faster at low concentrations than at high concentrations. The oxidation of amoxicillin by photo-Fenton results in degradation rates of up to 99%. A study of the adsorption of amoxicillin on copper oxides showed that amoxicillin adsorbs weakly to the amoxicillin surface, with an adsorption rate of 17%. However, in the presence of amoxicillin, hydrogen peroxide and sunlight, degradation rates of up to 99% were obtained.

Key words: Amoxicillin, photocatalysis, oxidation, sunlight.

1. Introduction

In an age of increasing industrialization and perpetual technical evolution, new products enter the market every day. The pharmaceutical industry is a case in point, constantly developing new molecules to meet the growing demands of patients and consumers [1, 2]. There is a huge diversity of drugs, consumed by the population at different frequencies [3, 4]. Consequently, in view of this extensive use of pharmaceutical products by the population, it may be suspected that these compounds could be released into the environment where anthropogenic activity is present. In fact, pharmaceutical compounds can be released into the environment via three different routes: wastewater discharge, sanitary landfill sites and agricultural use via excretion of drugs by animals [5-7]. However, wastewater discharge is the main route by which pharmaceuticals enter the environment [6, 8, 9]. Indeed, wastewater can be contaminated by pharmaceutical compounds due to excretion by the body of the compound at a certain percentage, via elimination in the toilet [10]. These contaminated wastewaters are conveyed by the sewage system to treatment plants, where they are treated before being discharged into the receiving environment. However, the volume and diversity of compounds found on the market complicate treatment efficiency. Indeed, some of these molecules are likely, because of their physico-chemical characteristics, to pass through the various stages of the treatment process. These compounds will then be released into the environment in trace quantities via the effluent. One of these persistent pollutants is amoxicillin (AMX). It is commonly found in wastewater treatment plant effluent, surface water and groundwater [11].

As a result, new treatment processes are needed to prevent further contamination of aquatic environments. Advanced oxidation processes ($\text{H}_2\text{O}_2/\text{Fe}^{2+}$, $\text{H}_2\text{O}_2/\text{O}_3$, O_3/UV , $\text{H}_2\text{O}_2/\text{UV}$, $\text{Fe}^{2+}/\text{H}_2\text{O}_2/\text{UV}$, TiO_2/UV) represent an interesting alternative [12-14]. In fact, they enable this drug to be mineralized through the in-situ creation of radicals, notably hydroxyl radicals (OH^\cdot). The latter are highly reactive species, capable of mineralizing most organic compounds [12, 15, 16]. Photocatalysis is an advanced oxidation process that combines light radiation with a photocatalyst. The principle of photocatalytic oxidation is based on the absorption of photons by the photocatalyst and the resulting separation of charges [17]. Our aim is to treat water polluted by amoxicillin using photocatalysis under solar irradiation.

2. Materials and methods

2.1. Amoxicillin analysis

The concentration of amoxicillin was determined by measuring the Chemical Oxygen Demand (COD). To measure COD, a 20 mL volume of each solution was taken and filtered successively through an ordinary FILTER-LAB filter to retain the sludge, then a second time and a third time through FILTER-LAB filters with a porosity of $0.45 \mu\text{m}$ and a diameter of 47mm. 2 mL of each solution are introduced into the HACH COD tubes and then heated in the digester at 150°C for 120 minutes.

Before each sampling, evaporation losses are compensated for.

The degradation rate at time t is determined using eq. 1.

$$D_t = \left[1 - \frac{C_t - C_B}{C_A - C_{BA}} \right] \times 100 \quad (1)$$

Where

D_t = percentage of degradation at time t ;

C_A = COD measured in the test suspension, measured after 3 h of incubation (mg/L);

C_t = COD measured in the test suspension at time t (mg/L);

C_B = COD measured in the control at time t (mg/L);

C_{BA} = COD measured in the control after 3 h of incubation (mg/L).

The same calculation is made with the reference solution.

2.2. Reagents

Amoxicillin was purchased from a pharmacy in Abidjan (Ivory Coast). The semi-developed formula of amoxicillin is given in Fig. 1. Hydrogen peroxide (H_2O_2) and copper oxide were supplied by Expertise Chimique and SCHARLAU respectively. Ferrous sulfate (FeSO_4), sodium hydroxide (NaOH) and sulfuric acid (H_2SO_4) were supplied by Fluka.

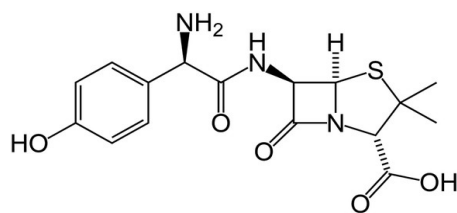


Figure 1: Semi-developed formula of amoxicillin

2.3 Amoxicillin degradation tests

2.3.1. Degradation of amoxicillin by the Fenton process

Several series of 500 mL reaction mixtures are prepared with solutions of amoxicillin, hydrogen peroxide, ferrous ions and sulfuric acid at 25°C. One of the following parameters is varied: amoxicillin concentration, hydrogen peroxide concentration, Fe²⁺ concentration and pH, while the other parameters are fixed.

2.3.2. Amoxicillin degradation by the Photo-Fenton process

We used the optimum conditions obtained with the fenton process, and the experiments were carried out in the presence of sunlight. We set the hydrogen peroxide concentration at 200 mg/L, the ferrous ion concentration at 15 mg/L and the pH at 3, varying the amoxicillin concentration (5 mg/L; 10 mg/L; 15 mg/L and 20 mg/L).

2.3.3. Amoxicillin photodegradation in the presence of copper oxide

In this section, the adsorption of amoxicillin on copper oxides was studied by contacting 1 g/L copper oxide with 5 mg/L amoxicillin at pH=3. Next, the photocatalytic degradation of amoxicillin was studied by setting the hydrogen peroxide concentration at 200 mg/L, the copper oxide concentration at 1 g/L and the pH at 3, varying the amoxicillin concentration (5 mg/L; 10 mg/L; 15 mg/L and 20 mg/L).

3. Results and Discussion

3.1 Study of amoxicillin oxidation by the Fenton process

3.1.1. Study of the pH influence

Amoxicillin degradation was carried out in solutions at different pH values. Fe²⁺, amoxicillin and H₂O₂ concentrations were kept constant. The results obtained are shown in **Fig. 2**. In general, a rapid increase in the degradation rate is observed in the first 20 min of the experiment, followed by a slow increase until 30 min, when a maximum is reached. We also note that the degradation rate increases as the pH rises from 2 (58%) to 3 (79.5%), only to fall again as the pH rises above 3.

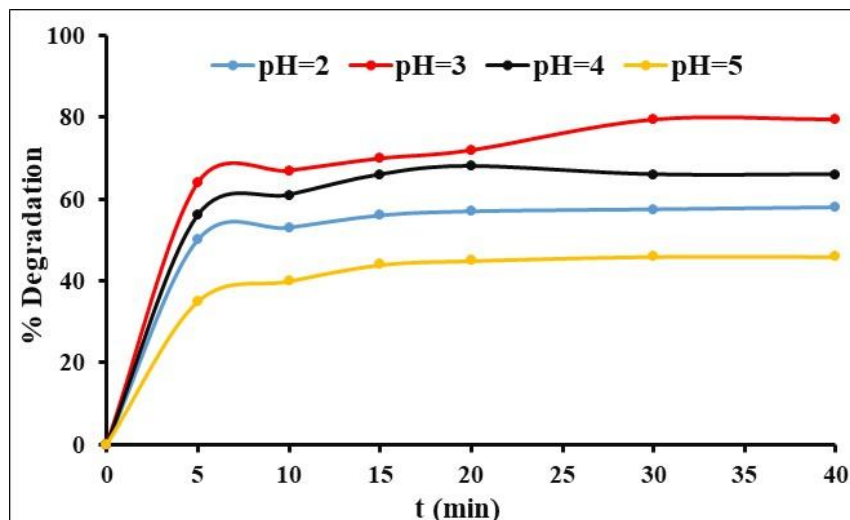
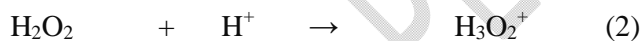


Figure 2: Effect of pH on amoxicillin degradation as a function of time, [amoxicillin] = 5 mg/L, $[\text{Fe}^{2+}] = 15 \text{ mg/L}$, $[\text{H}_2\text{O}_2] = 100 \text{ mg/L}$, $T=25^\circ\text{C}$

This work shows that the best pH for amoxicillin degradation is pH = 3. At very acidic pH, H_2O_2 becomes highly unstable following the formation of oxonium ion (H_3O_2^+) by solvating a proton (Eq. 2). This acidic form of electrophilic hydrogen peroxide severely restricts its reactivity with ferrous iron. This greatly reduces the number of hydroxyl radicals generated by the decomposition of H_2O_2 by Fe^{2+} . Moreover, low pH levels favor the consumption of hydroxyl radicals by hydrogen peroxide. At $\text{pH} < 2.5$, ferrous iron is essentially in the form of an aqueous complex $[\text{Fe}(\text{H}_2\text{O})_6]^{2+}$ in equilibrium with its conjugated form [12, 18].



Kinetically, the $[\text{Fe}(\text{OH})(\text{H}_2\text{O})_5]^+$ is much more reactive with hydrogen peroxide than the $[\text{Fe}(\text{H}_2\text{O})_6]^{2+}$. Thus, the speed of the initial step in the mechanism of H_2O_2 decomposition by Fe^{2+} increases with pH in the range $1 < \text{pH} < 3$. Consequently, for $\text{pH} < 2.5$, the Fenton reaction generates fewer HO^\cdot radicals, which experimentally translates into a decrease in the degradation rate of organic compounds.



These particles aggregate to form a precipitate from $\text{Fe}(\text{OH})_3$ [12, 19], and their concentration increases with pH. Following precipitation, species concentrations restrict the ability to produce hydroxyl radicals from the Fenton reaction (Eq. 5). Regeneration of Fe^{2+} from precipitated Fe^{3+} is very slow and represents the limiting kinetic step. Moreover, at $\text{pH} > 4$, hydrogen peroxide is unstable, decomposing into O_2 and H_2O and losing its oxidation capacity [12]. Based on the above, the pH favorable to amoxicillin oxidation is 3. This pH was chosen for further work.

The gradual decrease in the abatement rate with initial concentration could be explained by competitive reactions between amoxicillin molecules and those of intermediates formed during the Fenton oxidation process [18]. The amoxicillin molecules and intermediates formed will compete to react with HO[•] radicals. This increases the competitive effect with the initial concentration of amoxicillin [18].

3.1.2. Study of the influence of the initial amoxicillin concentration

The quantity of material to be degraded is one of the factors determining the efficiency of the treatment process. Thus, various initial concentrations of amoxicillin were studied by fixing the values in concentration of H₂O₂, Fe²⁺ and pH. The results obtained are presented in **Fig. 3**, which shows that degradation efficiency decreases with increasing initial amoxicillin concentration. Degradation rates of 79.5%, 72%, 63% and 56% are observed for 5 mg/L, 10 mg/L, 15 mg/L and 20 mg/L respectively. This is in line with the results reported in the literature [20-22]. Thus, a high rate of degradation is observed for low concentrations of amoxicillin.

The gradual decrease in the abatement rate with initial concentration could be explained by competitive reactions between amoxicillin molecules and those of intermediates formed during the Fenton oxidation process [18]. The amoxicillin molecules and intermediates formed will compete to react with HO[•] radicals. This increases the competitive effect with the initial concentration of amoxicillin [18].

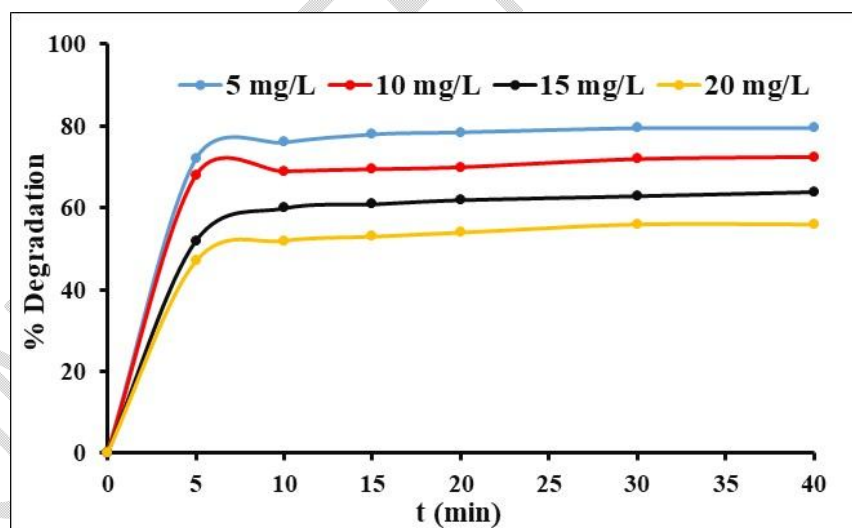


Figure 3: Effect of amoxicillin concentration on its degradation as a function of time [Fe²⁺] = 15 mg/L, [H₂O₂] = 100 mg/L, pH=3, T=25°C

3.1.3. Study of the influence of initial Fe²⁺ concentration

Fig. 4 shows that the rate of amoxicillin degradation varies with Fe²⁺ concentration. The rate of degradation increases from 5 mg/L to 15 mg/L Fe²⁺. Then, above 15 mg/L, it decreases. Above 15 mg/L, Fe²⁺ are engaged in a secondary reaction, consuming hydroxyl radicals, and the degradation of amoxicillin decreases. The final result is that amoxicillin

degradation is greatest at 15 mg/L Fe²⁺, with a degradation rate of 79.5%. Above 15 mg/L, amoxicillin degradation decreases with increasing iron II concentration. When the Fe²⁺ concentration is very high, parasitic reactions such as eq. 7 occur. These reactions compete with the degradation reaction of organic compounds (Eq. 6). This reduces the oxidation of organic compounds [12].

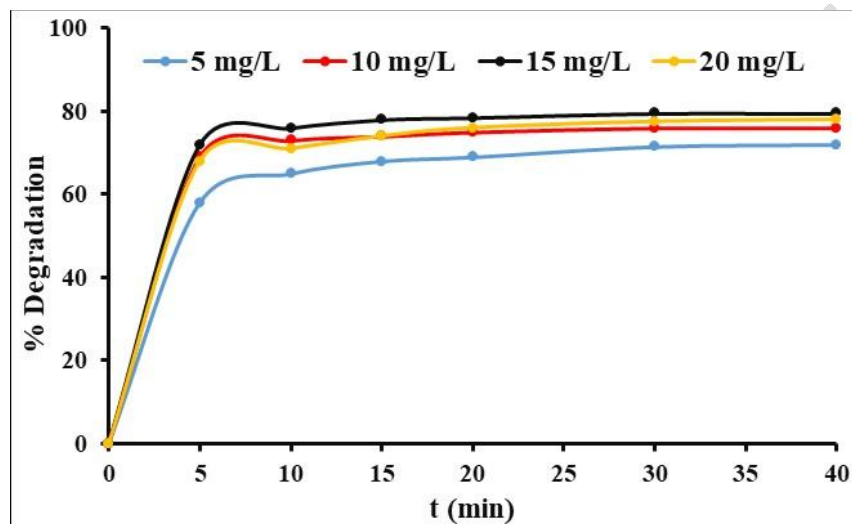
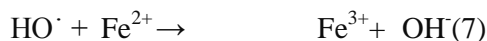


Fig. 4: Effect of Fe²⁺ concentration on amoxicillin degradation as a function of time, [amoxicillin] = 5 mg/L, [H₂O₂] = 100 mg/L, pH=3, T=25°C

According to eq. 8, Fe²⁺ react with hydrogen peroxide to give Fe³⁺. So if the concentration of Fe²⁺ is high, the quantity of Fe³⁺ produced will be high. However, Fe³⁺ decomposes hydrogen peroxide according to eq. 9. The decrease in the rate of degradation with an excess of Fe²⁺ is also linked to the decomposition of H₂O₂ by the Fe³⁺ produced [12].



3.1.4 Study of the influence of initial H₂O₂ concentration

The initial H₂O₂ concentration was studied. The results obtained are shown in Fig. 5, where it can be seen that the degradation rate increases for initial H₂O₂ concentrations ranging from 50 mg/L to 200 mg/L. Amoxicillin's degradation rate rises from 73 to 85.5%. Above 200 mg/L, the amoxicillin degradation rate decreases. At 250 mg/L, it drops to 82%. This shows that 200 mg/L is the optimum H₂O₂ concentration for amoxicillin degradation under the conditions studied. Increasing the H₂O₂ concentration causes an increase in the amount of OH[·] and consequently it increases the rate of amoxicillin degradation. However, too high a concentration of peroxide causes hydroxyl radical scavenging due to excess hydrogen

peroxide forming hydroperoxyl radicals ($\text{HO}_2\cdot$) and causes a decrease in the degradation rate of the organic compound [21, 23].

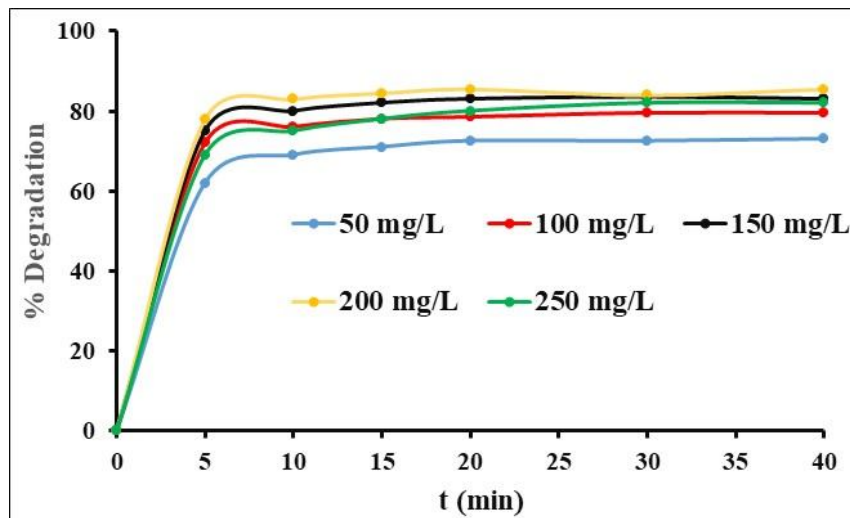


Fig. 5: Effect of H_2O_2 concentration on amoxicillin degradation as a function of time, $[\text{amoxicillin}] = 5 \text{ mg/L}$, $[\text{Fe}^{2+}] = 15 \text{ mg/L}$, $\text{pH}=3$, $T=25^\circ\text{C}$

$[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ ratio is a very important parameter in the Fenton process. This ratio was calculated from the results shown in Figure 5. The results obtained are presented in Table II. According to the results in Table I, degradation of the organic compound is optimal when the ratio $[\text{H}_2\text{O}_2] / [\text{Fe}^{2+}]$ is equal to 13.33.

Table I: Effect of $[\text{H}_2\text{O}_2] / [\text{Fe}^{2+}]$ ratio on amoxicillin degradation, $[\text{amoxicillin}] = 5 \text{ mg/L}$, $\text{pH}=3$, $T=25^\circ$.

$[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ ratio	% degradation
3.33	73
6.67	79.5
10	83
13.33	85.5
16.67	82

3.2 Study of amoxicillin oxidation using the solar photo Fenton process

The influence of sunlight on the Fenton reaction was studied. The results obtained are shown in **Fig. 6**. This process is based on the Fenton reaction coupled with UV-visible irradiation. **Fig.6** shows degradation rates ranging from 84% to 99%. These results are significantly better than those obtained in the absence of sunlight. During the implementation of this process, $\text{HO}\cdot$ radicals are produced by various routes:

- direct reactions of H_2O_2 with Fe^{2+} introduced into the solution at the start of the reaction (eq. 8) or formed by photoreduction of Fe^{3+} during the reaction (eq. 11).



- H_2O_2 photolysis



The effectiveness of photo-Fenton treatment depends essentially on the concentrations of Fe^{2+} and H_2O_2 , and of course on light intensity. The production of hydroxyl radicals by these different pathways explains the improved rate of amoxicillin degradation.

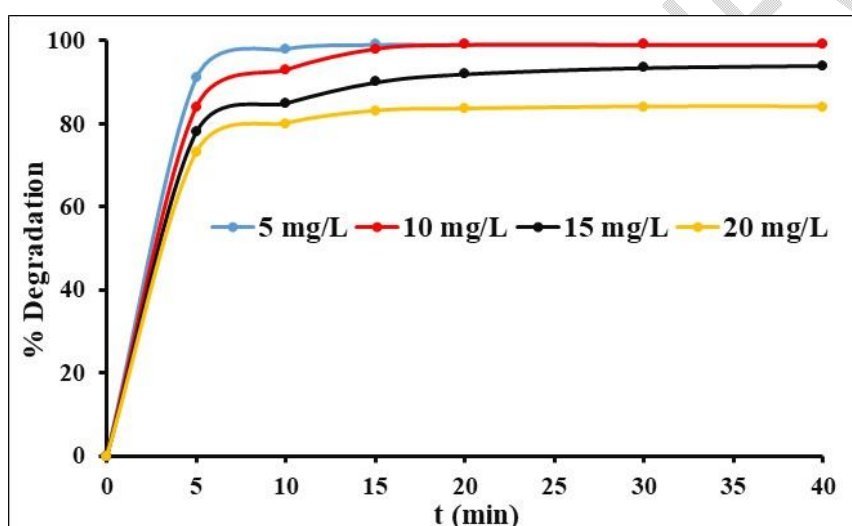


Fig.6: Sunlight degradation of different amoxicillin concentrations as a function of time, [amoxicillin] = 5 mg/L, $[\text{Fe}^{2+}] = 15 \text{ mg/L}$, $[\text{H}_2\text{O}_2] = 200 \text{ mg/L}$, $\text{pH}=3$, $T = 39 \pm 2^\circ\text{C}$

3.3 Photocatalytic oxidation of amoxicillin in the presence of copper oxide

3.3.1. Study of amoxicillin adsorption on copper oxides

Before studying the photocatalytic oxidation of amoxicillin in the presence of copper oxide, it is necessary to study the adsorption of amoxicillin on the surface of copper oxides. Adsorption is a highly effective separation technique, widely used in wastewater treatment due to its low cost, simple design and ease of use [26]. The extent of this technique depends on the nature of the adsorbate (molecular weight, molecular structure, polarity, solution concentration, etc.) and the adsorbent (particle size, specific surface area, surface charge, etc.).

Thus, the adsorption of amoxicillin on copper oxides was studied. In **fig.7**, the rate of amoxicillin elimination was recorded for 1 g/L CuOx , 5 mg/L amoxicillin and $\text{pH} = 3$. This figure shows low adsorption (17%) of amoxicillin on the copper oxide surface after a contact

time of 45 min. Thereafter, the adsorption rate remains constant. This shows that equilibrium is reached after 45 min [27, 28].



Fig.7: Adsorption rate of amoxicillin on copper oxides as a function of time, $[\text{CuO}_x] = 1$ g/L, $[\text{amoxicillin}] = 5$ mg/L, $\text{pH}=3$, $T = 25^\circ\text{C}$

3.3.2. Amoxicillin oxidation in a copper oxide suspension in the presence of sunlight

The solar photocatalytic oxidation of amoxicillin was studied in the presence of copper oxide. Prior to photocatalytic oxidation, amoxicillin adsorption was carried out by contacting amoxicillin and copper oxides in the dark for 45 min (equilibrium time). The results obtained are shown in **Fig.8**. According to this figure, the efficiency of degradation decreases as the initial concentration of amoxicillin increases. Almost complete degradation (99%) is observed after 20 min for 5 mg/L amoxicillin. A degradation rate of 99% is also reached after 40 min of solar irradiation for an amoxicillin concentration of 10 mg/L. At higher concentrations (15 mg/L and 20 mg/L), degradation is relatively slow, with rates of 90% and 81% after 40 min for 15 mg/L and 20 mg/L respectively. Indeed, the higher the concentration of the solution, the opaquer it becomes and the lower the penetration of solar rays. The decrease in the degradation rate as the concentration increases would appear to be due to the increasing formation of a screen by the pollutant, making it virtually impossible for light to penetrate the solution.

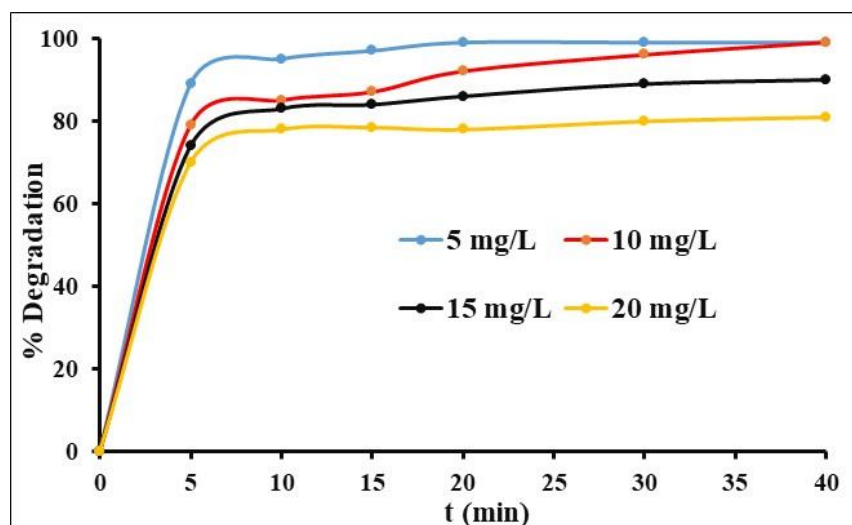


Fig. 8: Solar photocatalytic degradation of different concentrations of amoxicillin in the presence of copper oxide as a function of time, $[\text{CuO}_x] = 1 \text{ g/L}$, $[\text{amoxicillin}] = 5 \text{ mg/L}$, $[\text{H}_2\text{O}_2] = 200 \text{ mg/L}$, $\text{pH}=3$, $T = 40 \pm 1^\circ\text{C}$

4. Conclusion

This study showed that the mixture of hydrogen peroxide and iron (II) ions produces hydroxyl radicals responsible for the degradation of amoxicillin. According to the results obtained, for maximum oxidation of amoxicillin, a pH equal to 3 and a $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ ratio equal to 13.33 are required. The oxidation reaction of amoxicillin by the Fenton reaction is a rapid one, occurring in 30 min with a degradation rate of up to 85.5%. During the Fenton reaction, Fe^{2+} are transformed into Fe^{3+} , which decompose hydrogen peroxide. However, in the presence of sunlight, Fe^{2+} are regenerated from Fe^{3+} and hydrogen peroxide is photolyzed, increasing the rate of amoxicillin degradation. Solar photo-Fenton thus improves the Fenton yield, with a degradation rate of 99%. Characterization of the copper oxide powder by XRD showed the presence of Cu_2O and CuO . A study of the adsorption of amoxicillin on these copper oxides showed that amoxicillin adsorbs weakly to the oxide surface, with an adsorption rate of 17%. However, in the presence of hydrogen peroxide and sunlight, degradation rates of up to 99% were obtained.

REFERENCES

- [1] Beatriz GT, Fernando A (2023) The Pharmaceutical Industry in 2022: An Analysis of FDA Drug Approvals from the Perspective of Molecules. *Molecules* 28(3): 1038.
- [2] Diniz LF, Tenorio JC, Ribeiro C, Carvalho Jr PS (2023) Structural aspects, solid-state properties, and solubility performance of pharmaceutical sertraline-based organic salts. *Journal of Molecular Structure* 1273: 134293.

- [3] Wu D, Li M (2023) Current State and Challenges of Physiologically Based Biopharmaceutics Modeling (PBBM) in Oral Drug Product Development. *Pharm Res* 40: 321–336.
- [4] Larabi IA, Ghish A, Kintz P, Marillier M, Fabresse N, Pelletier R, Adeline Knapp, Ameline A, Willeman T, Barguil Y, Aknouche F, Mathieu O, Chèze M, Lelong-Boulouard V, Matheux A, Le Carpentier EC, Brunet B, Gambier N, Edel Y, Cartiser N, Dumestre-Toulet V, Cohen S, Lelièvre B, Gaulier J-M, Alvarez J-C, Péliissier A-L (2023) A national study through interlaboratory collaboration under the auspices of the French Society of Analytical Toxicology (SFTA). *Toxicologie Analytique et Clinique* 35(3) : 175-197
- [5] Castellano-Hinojosa A, Gallardo-Altamirano MJ, González-López J, González-Martínez A (2023) Anticancer drugs in wastewater and natural environments: A review on their occurrence, environmental persistence, treatment, and ecological risks. *Journal of Hazardous Materials* 447: 130818.
- [6] Sadia SP, Berté M, Loba EMH, Appia FTA, Gnamba CQ-M, Ibrahima S, Lassiné O (2016) Assessment of the Physicochemical and Microbiological Parameters of a Teaching Hospital's Wastewaters in Abidjan in Côte d'Ivoire. *Journal of Water Resource and Protection* 8: 1251-1265.
- [7] Sadia SP, Gnamba CQ-M, Kambiré O, Konan KM, Berté M, Koffi KS, Kouadio KE, Kimou KJ, Pohan LAG, Ouattara L (2023) Principal component analysis of physico-chemical parameters of wastewater from the University Hospital Center of Treichville in Côte d'Ivoire. *J. Mater. Environ. Sci.* 14(7): 826-837
- [8] Kouadio KE, Kambiré O, Koffi KS, Ouattara L (2021) Electrochemical oxidation of paracetamol on boron-doped diamond electrode: analytical performance and paracetamol degradation. *Journal of Electrochemical Science and Engineering*, 11(2), 71-86
- [9] Kimou KJ, Kambiré O, Koffi KS, Kouadio KE, Koné S, Ouattara L (2021) Electrooxidation of Iohexol in Its Commercial Formulation Omnipaque on Boron Doped Diamond Electrode. *International Research Journal of Pure & Applied Chemistry* 22(11): 29-41
- [10] Zhang Y, Guo L, Hoffmann MR (2023) Ozone-and Hydroxyl Radical-Mediated Oxidation of Pharmaceutical Compounds Using Ni-Doped Sb–SnO₂ Anodes: Degradation Kinetics and Transformation Products. *ACS ES&T Engineering* 3 (3): 335-348.
- [11] Sadia SP, Berte M, Ouattara L (2020) Removal of antibiotics containing simulated wastewater by biological-photochemical process. *Rev. Ivoir. Sci. Technol.* 35 (2020) 111 - 120

- [12] Kambiré O, Abollé A, Kouakou AR, Koffi AE, Koffi KS, Kouadio KE, Kimou KJ, Koné S, Ouattara L (2022) Oxidation of rhodamine B using the Fenton process: optimization, kinetics and inorganic ions studies. *Journal de la Société Ouest-Africaine de Chimie* 051 : 45 – 55.
- [13] Zhaobo W, Ying C, Chen W, Rui G, Junhua Y, Hangzhou Z (2023) Optimizing the performance of Fe-based metal-organic frameworks in photo-Fenton processes: Mechanisms, strategies and prospects. *Chemosphere* 339: 139673.
- [14] Fuhang X, Cui L, Mingming Z, Dengsheng M, Ling L, Shiyu L, Xuerong Z, Huchuan Y, Neng W, Mengyi X, Lei Q, Huan Y (2023) Graphite carbon nitride coupled with high-dispersed iron (II) phthalocyanine for efficient oxytetracycline degradation under photo-Fenton process: Performance and mechanism. *Separation and Purification Technology* 308: 122829
- [15] Kouakou KE, Pohan LAG, Ollo K, Lassiné O (2021). Oxidation of paracetamol by combination of sonochemistry and electrochemistry on boron-doped diamond electrode. *RAMReS Sciences des Structures et de la Matière* 4: 1-16.
- [16] Sadia SP, Kambiré O, Gnamba CQ-M, Pohan LAG, Berté M, Ouattara L (2021) Mineralization of Wastewater from the Teaching Hospital of Treichville by a Combination of Biological Treatment and Advanced Oxidation Processes. *Asian Journal of Chemical Sciences* 10 (2): 1-10.
- [17] Pohan A, Goure-Doubi H, Kouyate A, Nasir M, Visa M, Ouattara L (2019) Hydrothermal Sol-gel TiO₂ Nanoparticles fixed to Clay and its Photocatalytic Application for the Degradation of Methyl Orange. *Mediterranean Journal of Chemistry* 9(2): 125-132
- [18] Hu Q, Zhang C, Wang Z, Chen Y, Mao K, Zhang X, Xiong Y, Zhu M (2008) Photodegradation of methyl tert-butyl ether (MTBE) by UV/H₂O₂ and UV/TiO₂. *Journal of Hazardous Materials* 154(1–3):795-803
- [19] Hug SJ, Leupin O (2003) Iron catalyzed oxidation of arsenic (III) by oxygen and by hydrogen peroxide: pH dependent formation of oxidants in the Fenton reaction. *Envir. Sci. Technol.* 37: 2734-2742.
- [20] Vajnhandl S, Le Marechal AM (2007) Case study of the sonochemical decolouration of textile azo dye Reactive Black 5. *J. Hazard. Mater.* 141 (1): 329–335.
- [21] Behnajady MA, Modirshahla N, Tabrizi SB, Molanee S (2008) Ultrasonic degradation of Rhodamine B in aqueous solution: Influence of operational parameters. *J. Hazard. Mater.* 152 (1): 381–386.
- [22] Wang X, Yao Z, Wang J, Guo W, Li G (2008) Degradation of reactive brilliant red in aqueous solution by ultrasonic cavitation. *Ultrason. Sonochem.* 15 (1): 43–48,

- [23] Nidheesh PV, Gandhimathi R, Ramesh ST (2013) Degradation of dyes from aqueous solution by Fenton processes: a review. *Environ. Sci. Pollut. Res.* 20 (4): 2099-2132.
- [24] Kambiré O, Chia YPA, Kouakou YU, Yeo F, Lassiné O (2023) Photocatalytic degradation of methyl orange in an aqueous solution in presence of copper oxide. *Journal of Chemical, Biological and Physical Sciences. Section A* 13(4): 401-412
- [25] Daoud I, Mesmoudi M, Ghalem, S (2013) MM/QM study: Interactions of copper(II) and mercury(II) with food dyes in aqueous solutions. *International Journal of Chemical and Analytical Science* 4 (2): 49-56.
- [26] Zaviska F, Drogui P, Mercier G, Blais J-F (2009) Procédés d'oxydation avancée dans le traitement des eaux et des effluents industriels : Application à la dégradation des polluants réfractaires. *Revue des sciences de l'eau* 22(4) : 535-564.
- [27] Kambiré O, Kouakou YU, Kouyaté A, Sadia SP, Kouadio KE, Kimou KJ, Koné S (2021) Removal of rhodamine B from aqueous solution by adsorption on corn cobs activated carbon. *Mediterranean Journal of Chemistry* 11(3): 271-281
- [28] Kouakou YU, Kambire O, Zran VE-S. Properties of magnetic carbon based on *ricinodendron heudelotii* and application in removal of methylene blue. *Int. J. Adv. Res.* 10(11): 440-453