

Photo degradation of biphenyl in prior to the ecological elimination of polychlorinated biphenyls “PCBs”

Oumarou Tidjani Malam Mani¹

Département de chimie, faculté des Sciences et Techniques, Université Dan Dicko Dan koulodo de Maradi/Niger

Rabani Adamou²

Département de chimie, faculté des sciences et Techniques, Université Abdou Moumouni de Niamey/Niger

Abstract: Polychlorinated biphenyls (PCBs) are part of persistent organic pollutants which are very harmful for environment and human beings. The international community envisaged the complete elimination of PCBs for 2028. However, the techniques available of destruction are very expensive because of consuming energy, with environmental and health risks. To offset these difficulties, we investigate a simple and effective method for elimination of PCBs in liquid phase. This technique is based on catalyzed photodégradation. Because biphenyl is the major products of PCBs dechlorination, the photodegradation of biphenyl were studied in prior to that of PCBs. The ultraviolet radiations UVA, UVB and visible radiations were used to investigate the effect of type of radiation on the direct photolysis. The effect of oxidant and photocatalyst were determined respectively by UV/H₂O₂ and UV/Fe₂O₃ processes. The highest degradation in direct photolysis were obtained under UVA+UVB + Visible (73.21%) follow-up by UVB (72%). On the other hand, UVB/H₂O₂ process was the greatest (75.24%), while the addition of Fe₂O₃ did not produce the expected effect of a photocatalytic. Finally, this performance of these processes, (UV, Vis/H₂O₂) allowed suggesting the use of solar radiation for an ecologically rational elimination of PCBs.

Keywords: photocdegradation, , PCBs , photonique effect , Biphenyl, H₂O₂.

Date of Submission: 14-10-2022

Date of Acceptance: 30-10-2022

I. Introduction

Polychlorinated biphenyls are chlorinated organic compounds, which are among the most important and dangerous pollutants of the twentieth centuries [1]. Also called PCBs, they are still in the twenty-first century (decades after their commercial production), a major global concern because of their thermal stability. Despite the cessation of their production since the 1980s. PCBs are still used, mainly as dielectric fluids in transformers, capacitors and radiators or as lubricants in turbines and pumps [2] The word global PCB production is around 2 million tons, with 30% dispersed in the environment and 70% stored and/or in use [3,4]

Over the word, especially in Africa, it is mainly the electricity production and mining companies which are the biggest users and holders of PCB waste. This is the case in Niger where a large quantity of waste has been stored at Nigerien electricity company (NIGELEC) and mining companies (SOMAIR and old COMINAK) [5]

Furthermore, the worrying characteristics of PCBs (persistence, bioaccumulation, ecotoxic activity) alerted the international community to classify PCBs on the list of persistent organic pollutants "POPs" in 2001 during the Stockholm Convention [6] Therefore, it has been established their complete elimination by the United Nations Environment Program (UNEP), for 2028. However, the available destruction techniques are very expensive, energy consuming, often causing health and environmental risks. ([7,8] Also, their combustion around 1100°C releases carcinogenic substances (dioxins and furans); while their incineration is done at temperatures between 3000 and 15000°C, and the alkaline reduction is explosive [9]. To offset these reactional and mechanical difficulties, we are investigating a simple and effective method for elimination of liquid PCBs using the catalyzed photodegradation techniques. Thus, in prior to the photodegradation of PCBs, the assisted photodegradation of biphenyl was studied. Because photocatalysis is a complex process, involving the cumulative effects of photons, solvent, oxidizing agent and the photocatalyst. [10,11], the effect of different types of irradiation (UVA, UVB, and Visible) on the photodegradation of biphenyl was first investigated. Also, the direct biphenyl photolysis in different solvents allowed to appreciate the role of solvent on the analytical

quality of spectrum and the by percentage of photodegradation. The effect of oxidant on photolysis was studied by adding hydrogen peroxide (H₂O₂). The addition of ferric dioxide (Fe₂O₃) did not confirm the expected beneficial effect of a photocatalyst.

II. Materials And Methods

Chemical Reagents and Materials

Biphenyl (≥99%), was purchased from Merck Schuchardt OHG (Germany). Its solution was prepared in a mixture of distilled water and ethanol (99+% Wagtech International LTD). The hydrochloric acid HCl (37%) and the sodium hydroxide pellets which were used to adjust the pH of the solutions, were respectively from Prolabo (made in CE) and HANG Zhoo fine chemical industry, CO LTD (China). Hydrogen peroxide H₂O₂ (30%) was purchased from Han Zhoo (China) and ferric oxide Fe₂O₃ from Labosi (Paris, France). The pH of the solutions was measured using a multimeter, Multi 3430 SET (Germany) The photodegradation of biphenyl and its kinetic were measured using a UV-Vis spectrophotometer ((Thermo Fisher Scientific, Evolution 300 UV-Vis, Madison, USA) The UV radiation device is composed of UVA [320 – 400 nm] and UVB [290 - 320 nm]) provided respectively by five UVA lamps (Hitachi, FL 8BLB) and five UVB lamps (Luzchem Research INC). The visible radiation, is obtained using five mercury lamps (Sylvania 5058, Hg F8T5/ C10, P= 8W, Mexico)



Figure 1. :photo irradiation Dispositive

Experimental procedures

Preparation of biphenyl solutions

Since biphenyl has very low solubility in water (6.17 mg.L⁻¹) [12], a stock solution (1g.L⁻¹) was prepared in a water-ethanol mixture (10:90, V/V). From this stock, other more diluted solutions (100 ppm) are prepared in different solvents in order to test the effect of the solvent. Then, the prepared solutions are enveloped with aluminum foil and put in the refrigerator to be protected from any radiation.

Photoirradiation procedure

The photo-irradiation consisted of preliminary tests. by direct photolysis of the biphenyl solution (100 mg.L⁻¹), in different solvents, using UVA irradiation. So, samples of these different solutions, contained in a test tube (section 1 cm², volume 3 mL), were placed on an optical bench, 20 cm to the lamps and irradiated for 2 hours. The absorbance spectra of each solution before and after irradiation, allowed to deduce the quantity of biphenyl photodegraded according to the solvent used. Thus, the percentage of degradation can be calculated by:

$$d\% = \frac{A_0 - A_t}{A_0} \times 100\% \quad (1)$$

The same procedure was followed to study different parameters (addition of H₂O₂ as oxidant, addition of Fe₂O₃ as photo catalyst, pH of irradiating solution, initial concentration, irradiation source) on the biphenyl photolysis.

Kinetic of biphenyl photodegradation

The kinetic of photodegradation of biphenyl was followed, using three different sources of irradiation, and adding some substances (H₂O₂, Fe₂O₃) at different pH in order to compare the intensity of radiation on the photolysis one hand, and the effects of pH, oxidant and photocatalyst in the other hand. The solution (100 mL of concentration 100 mg.L⁻¹) to be irradiated, contained in a 150 mL beaker, is placed on an optical bench at 10 cm from the lamps. At regular times (every 10 min during 1 hour, and every 20 min during 2 hours), some amount of sample is taken for uv-spectrophometric analysis. Then the reading absorbance (A) allowed to calculate the corresponding concentrations from the following formula (2): obtained from Beer-Lambert law: $A = \log\left(\frac{I_0}{I_t}\right) = \epsilon l C$ using $\frac{A_t}{A_0} = \frac{\epsilon l C_t}{\epsilon l C_0} = \frac{C_t}{C_0}$

$$\frac{A_t}{A_0} = \frac{\epsilon l C_t}{\epsilon l C_0} = \frac{C_t}{C_0}$$

$$C_t = \frac{A_t \times C_0}{A_0} \quad (2)$$

Where A₀ is the absorbance (t = 0 min),

A_t:absorbance at time t,

C₀:is the initial concentration.

The photooxidation of biphenyl followed the second order reaction [13,14]with this kinetic model:

$$\frac{d[biph]}{dt} = -k_{biph,OH}[biph][OH^-]. \quad (3)$$

Because it is difficult to determine $[OH^-]$.concentration, the rate constant K is experimentally determined

$$\text{from: } \frac{[biph]}{[biph]_0} = e^{-(K_{biph}) \times t}$$

where $(K_{biph}) = K_{biph,OH} \times [OH^-]$ and it is given in min^{-1}

III. Results And Discussion

Influence of the solvent on the photolysis of biphenyl

The analysis of UV-Vis spectrophotometry of biphenyl solutions (100 mgL^{-1}), prepared in different solvents, led to study the effect of the solvent on the absorption spectrum (figure 2), and on the photolysis rate of biphenyl (figure 3). Thus, the different spectra on Figure 2, show that the solvent, depending on its type (polar, apolar, protic, aprotic), can have a bathochromic or hypochromic effect on the spectrum of the dissolved substance [15]. Also, It is observed in figure 3 that, for the same irradiation time, under the same conditions, the percentage of degradation depended on the type of solvent. This, show that the choice of solvent is strategic, not only for its influence on the photolysis, also for the quality of the analysis. The highest percentage of degradation is obtained with diethyl ether (81.5%). To improve an ecologically rational degradation, a water-ethanol mixture was preferred as the solvent for the rest of the work.

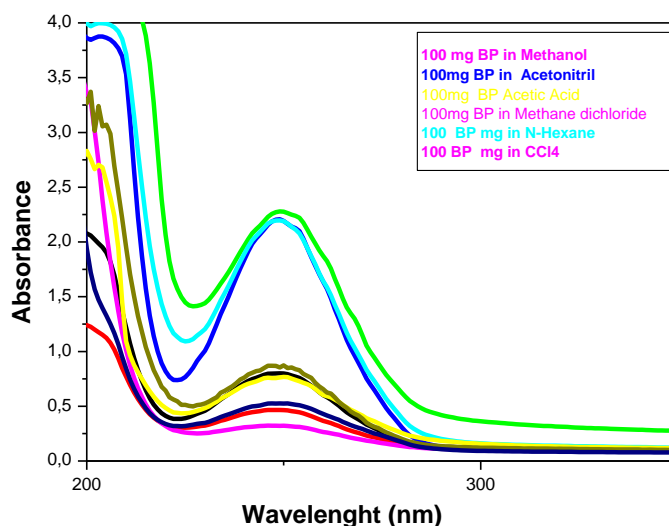


Figure 2: UV absorption spectrum of biphenyl in different solvents (pH = 6,8±0,1)

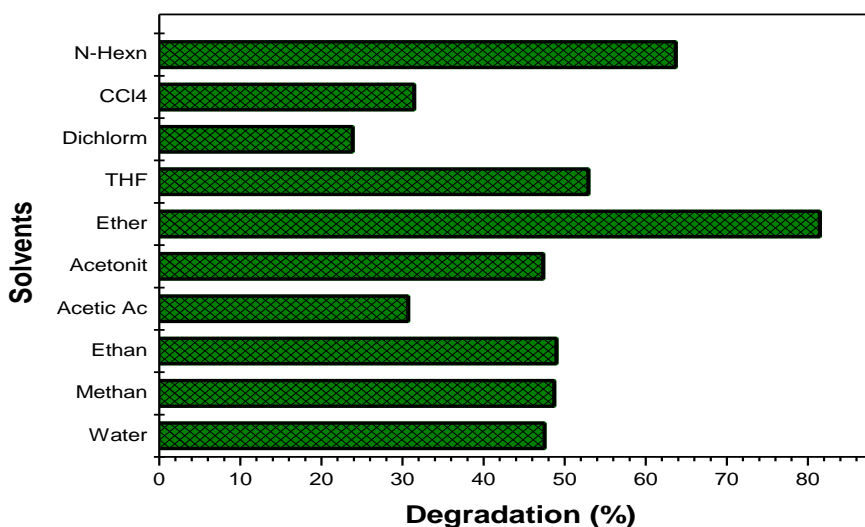


Figure 3 :Photolysis of biphenyl versus solvents after 2 hours, Under UVA irradiation (Experimental conditions: pH=6.8±0.1; T= 30±0.5)

Influence of H₂O₂ on biphenyl photolysis

In photocatalysis, the addition of hydrogen peroxide as an oxidant is an important parameter for efficient treatment [16]. To investigate the influence of H₂O₂ on direct photolysis, the solution of H₂O₂ (5.10⁻³M) was added, at different volumetric proportions (H₂O₂ [1:9], [2:8], [3 :7] , [4:6] , [5:5]) in the way to obtain a total volume (10 mL) of biphenyl solution to be irradiated. It is observed in Figure 4 that the best photodegradation rate (69.72%) was reached with the addition of H₂O₂with the proportion [2:8]; after one hour of irradiation under UVA. So, this addition of H₂O₂ [2:8] means a concentration of 1 mM in the irradiated solution. In fact, the H₂O₂/UV process is one of the advanced oxidation processes, which is essentially based on the production of free radicals such as hydroxyls (OH•) having a great oxidizing powers. [17] The major mechanism for the production of hydroxyl radicals in the UV/H₂O₂ process is the photolysis of hydrogen peroxide [18]. This would suggest that increasing the concentration of H₂O₂ would increase the rate of degradation. But is not the case, since at high concentrations, hydrogen peroxide acts as a hydroxyl radical scavenger too; when it absorbs all the available light [19]. Therefore, OH• radicals are generated by homolytic cleavage of the O-O bond of H₂O₂ , and would attack the benzene rings, thus reacting with the C-C double bonds [17,19]

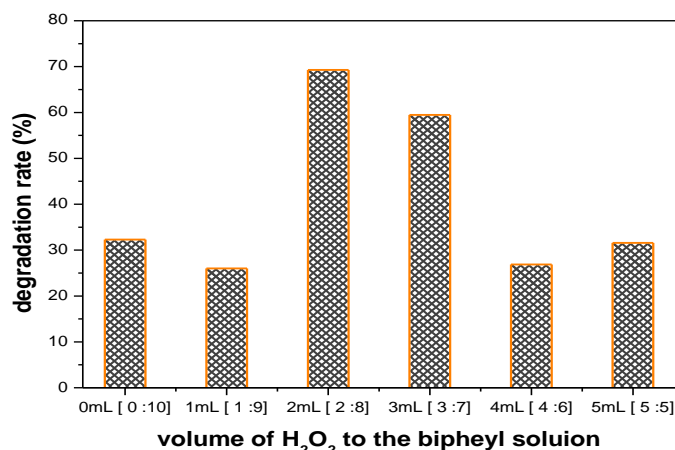


Figure 4: Effect of H₂O₂ on the biphenyl photodegradation (experimental conditions : pH = 6,8 ±0.5.; T= 30°C , under UVA radiation, exposure time 60 minutes)

Effect of pH on the photodegradation of biphenyl

After 1 hour of irradiation under UVA, the best photodegradation performance (68.81%) was observed for a pH = 7 (figure 5). These results are better than those of the oxidation of biphenyl by the Fenton process observed by Li [14, 24], where after 2 hours of reaction 48% of the biphenyl remains. However, at pH=3 and at pH= 11 the degradation rates are respectively 51.16 and 46.7%.these results suggest that the optimal pH of biphenyl photolysis is near to the neutral.

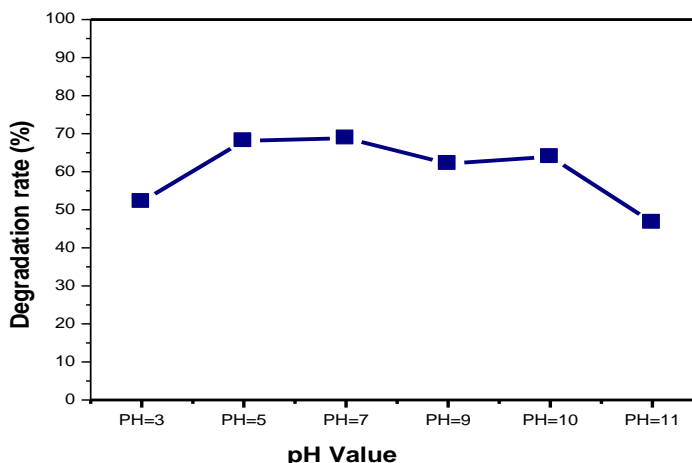


Figure 5 : pH effect on the biphenyl photolysis (experimental conditions: Ci = 100 ppm, T= 30 ± 0.5°C; Exposure time: 1hour)

Influence of the type of radiation

To investigate the effect of the type of radiation, the photoirradiation was carried out with different UV lamps: (UVA [320 – 400 nm]; UVB [290 - 320 nm]) and visible lamps (8W power mercury lamps). The results in Figure 6 show that after five hours of radiation, the best photodegradation rate (73.82%) was obtained with the mix radiation (UVA + UVB + Visible) when the UVB radiation gave 72.02% of destruction .and the lowest one (65.03%) was attained under the UVA The performance of mixed radiation (UVA + UVB + Visible) on the degradation of biphenyl suggests a possible use of solar energy, then reinforcing the practice of ecologically photo-elimination of biphenyl; so of PCBs. [25]

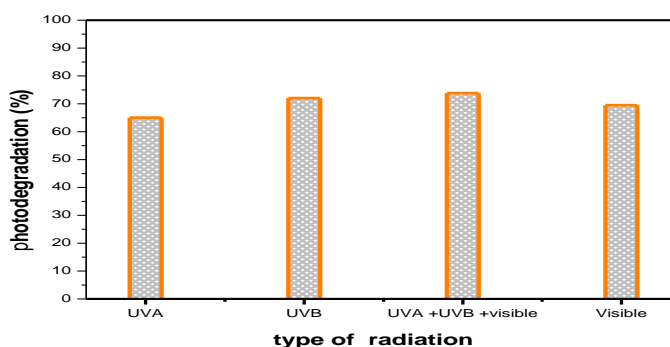


Figure 6: Influence of the radiation type on biphenyl photodegradation (experimental conditions: pH= 7± 0.5 , T= 25±0.5°C, C₀ = 100 mgL⁻¹ Exposure time: 5hour)

Biphenyl photodegradation kinetics

The major mechanism of biphenyl photodegradation proceeds by oxidation: It started by hydroxylation with formation of hydroxybiphenyl, followed by the opening of benzene ring, giving carboxylic acids and Carbon dioxide CO₂. [14,26].

Kinetics according to the type of radiation

The different values of the rate constants (Table I) and the graph related to the type of irradiation (Figure 7: exposure time versus concentration), show the exclusive effectiveness of the photonic effect on the degradation of biphenyl. Thus, from UVA radiation (k = 0.0056 min⁻¹) to UVB radiation (k = 0.01436 min⁻¹) the rate constant was multiplied by 2.5, while with the mixture UVA+ UVB+ Visible (K=0.0146

min⁻¹), the rate constant is 2.6 times greater than the constant of UVA radiation use. Thus, these values of the different constants reflect the same photonic effects depending to the nature of radiation. [27]

Tableau I: Rate constant (K) and linear coefficient of regression(R²) for Photodgradation kinetics of biphenyl proceeding under types of radiation

Type of radiation	Rate Constantes K(min ⁻¹)	linear coefficient of regression R ²
UVA	0.0046	0.9338
UVB	0.01436	0.9503
Visible	0.0104	0.973
UVA+UVB +Visible	0.0146	0.9243

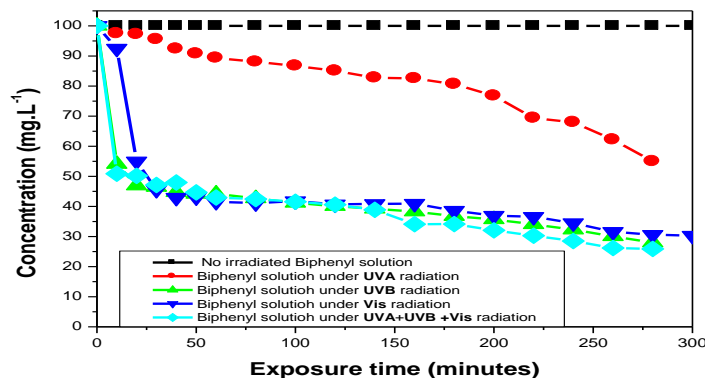


Figure 7 : Photodgradation kinetics of biphenyl proceeding under types of radiation (experimental conditions: pH= 7± 0.5 , T= 25±0.5°C, C₀ = 100 mgL⁻¹ Exposure time: 5hour)

Kinetics of assisted photodegradation (UVA/H₂O₂; UVB/H₂O₂; UVA+UVB+Visible/H₂O₂)

The results registered in Table II and presented in figure 8 allowed to compare the rates of photodegradation according to the rate constants K(min⁻¹) and the linear coefficient of regression R² related to the three processes [28]. Thus it is noted that the rate constant of biphenyl degradation by direct photolysis (K= 0.0046 mi¹) in table I, under UVA has been multiplied by 2 when H₂O₂ is added (UVA/H₂O₂ process K = 0,089 min⁻¹) in table II. But for the other two processes (UVB/H₂O₂; UVA+UVB+Visible/H₂O₂) when H₂O₂ is added the constants have remained substantially the same. With the best degradation rate for the UVB/H₂O₂ process.

Tableau II : Rate constant (K) and linear coefficient of regression(R²) for Photodgradation kinetics of biphenyl proceeding under UV, Vis /H₂O₂ processes

UV,Vis / H ₂ O ₂ processes	Rate Constante K(min ⁻¹)	Linear Coefficient of regression R ²
UVA/H ₂ O ₂	0.00897	0.9012
UVB/H ₂ O ₂	0.01412	0.8953
Visible/H ₂ O ₂	0.01316	0.8451
(UVA+UVB +Vis) /H ₂ O ₂	0.009324	0.9773

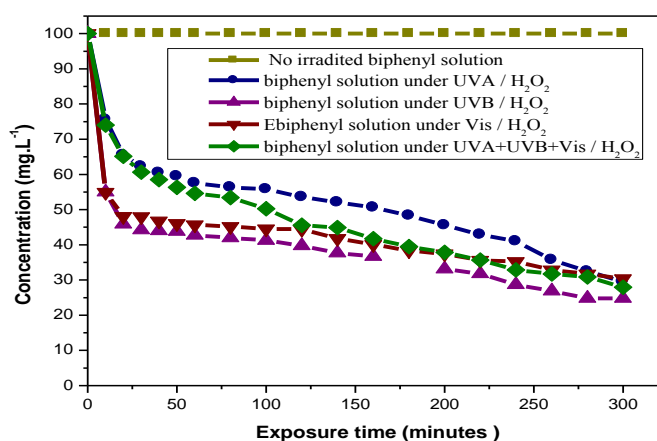
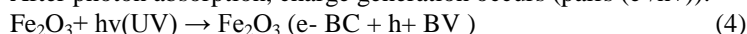


Figure 8 : Photodgradation kinetics of biphenyl proceeding under UV, Vis / H₂O₂(experimental conditions: pH= 7± 0.5, T= 25±0.5°C, C₀ = 100 mgL⁻¹ Exposure time: 5hour)

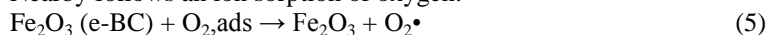
Influence of a photocatalyst (Fe₂O₃)

To investigate the effect of the photo catalyst, the preliminary tests were carried out with different initial concentrations of the photocatalyst Fe₂O₃ (50; 75; 100; 125; 150 and 200 mg L⁻¹). These outcomes allowed to retain 100 mgL⁻¹ as the optimal concentration of the photocatalyst. Indeed, Fe₂O₃ is a semiconductor (gap = 2.91ev) of oxidant type ([21 ,29]The promising mechanism of photocatalysis follows the classical scheme[22].

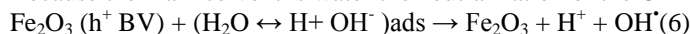
After photon absorption, charge generation occurs (pairs (e-/h+)):



Nearby follows an ion sorption of oxygen:



Because the main solvent is water the neutralization of the OH⁻ ions into the OH[•] radicals by h⁺ BV took place:



Finally; Biphenyl oxidation occurs through successive attacks by OH[•] radicals, or a direct reaction with the positive hole (h⁺) [23]. Thus, from figure 9, it was shown that the addition of the photocatalyst did not produce the expected effect since the three processes gave substantially the same result. This could be explained by the mismatch between the semiconductor (Fe₂O₃) and the type of irradiation received. [30]

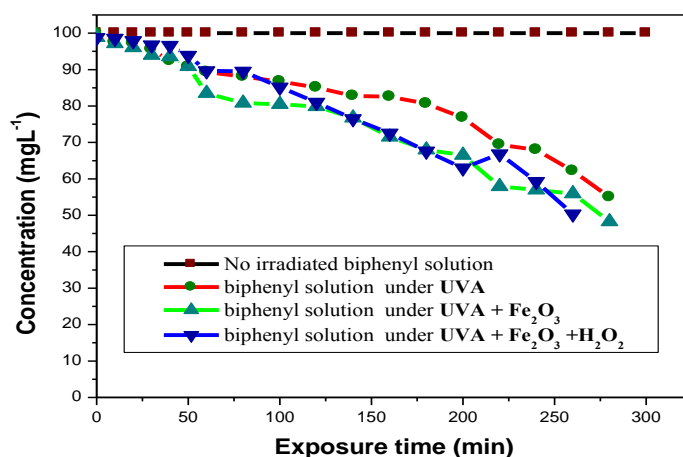


Figure 9:Photodgradation kinetics of biphenyl proceeding under UVA, Fe₂O₃ , H₂O₂ (experimental conditions: pH= 7± 0.5, T= 25±0.5°C, C₀ = 100 mgL⁻¹,exposure time: 5 hour)

Conclusion :

The results of this study have established the evidence of photoreactivity of biphenyl through OH[•] radicals, under the performance of the (UV, Vis / H₂O₂) processes. Also it has been proven that the photonic effect, depending on the type of irradiation, remains the driving force of photocatalysis. However these results

showed a relatively slow degradation of biphenyl, the optimization of the operational conditions, can led to a degradation of PCBs until mineralization, after dechlorination or directly. Finally, the performance of direct photolysis under visible irradiation and UVA+UVB+Visible, allowed suggesting the use of solar radiation, promising an ecologically rational elimination of PCBs.

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