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Assisted Photodegradation of Polychlorobiphenyls for Ecologically Rational Elimination of Persistent Organics Pollutants

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Abstract Polychlorinated biphenyls (PCBs) are synthetic organic chlorinated compounds classified in persistent organic pollutants. They are very harmful for environment and human beings. Thus, international community (UNEP) has planned the complete elimination of PCBs for 2028 while there are no expansive, simple and friendly techniques for their elimination. The aim of this work is to investigate the easy and ecologically rational techniques for elimination of PCBs. So, the photodegradation of PCBs contained in Askarel formulation were conducted for this remediation. For an ecological optimization of this process, the photolysis of PCBs congeners takes place in aqueous solution (ethanol-water 1:10). The photonic effect related to the type and intensity luminous source (halogens lamps (1000 and 500W) and one LED lamp of 50W) and distance from the lamp to the reaction point (40cm /20cm) were first investigated. Also, the influence of solvent, pH and initial concentrations on PCBs photolysis were studied to choose the optimum condition of PCBs degradation. The addition of H_2O_2 as source of OH• radicals has boosted the chemical decomposition of PCBs. The degradation% under the optimum conditions (Lamp of 1000 W from 20 cm to reaction point, pH = 6.8, Ci = 10 ppm) after 3hours is 60% with direct photolysis and goes to 75% after addition of 5 mM of H_2O_2. The pseudo first order model describes well the kinetic of PCBs photodegradation under optimum conditions ($R^2 = 0.9628$). Finally, the performance of the UV-Visible/H_2O_2 process which has established the photoreactivity of PCBs via OH• radicals suggest the possible use of solar energy as luminous source, a guarantee of an ecologically rational elimination of POPs.

Keywords: *Askarel, PCBs photodegradation, photonic effect, UV-Visible/H_2O_2 process*

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1. Introduction

It is the worrying characteristics (persistence, bioaccumulation, ecotoxic activity teratogenic properties [1,2] of polychlorinated biphenyls that alerted the international community to classify them as persistent organic pollutants in 2001 [3,4]. PCBs are aromatic compounds which are thermally and chemically very stable and have good solubility in fats, oils and organic solvents and are explosion resistant. As the number of chlorine atoms contained increases, the state of aggregation changes as well as some properties such as the stability of the molecule in the environment [5]. They have been widely used in various fields of economic activities, particularly as a dielectric fluid in electrical transformers and capacitors, before being banned in the last 1980s [6]. Despite this, from 1.5 million tons of PCBs produced worldwide between 1929 and 1989 a large quantity of PCBs is disseminated in nature, or is still found in old electrical transformers in service or used [7]. PCBs molecules were first discovered in the environment in 1966 by Jensen [8]. Since that time, many authors have shown their presence almost everywhere in the environment, particularly in living organisms, where they tend to accumulate in adipose tissues [9]. Thus water, air and soil are contaminated, reaching living beings through foodstuffs of varied origin. Therefore, it is believed that approximately 90% of human beings are exposed to PCBs from their diets that contain animal products, seen that the hydrophobic and liposoluble characteristics make PCBs a bioaccumulative contaminant with a concentration of 8.10^{-12}g per gram of fresh weight (8 pg g^{-1}) authorized for human consumption [10]. In Africa and more particularly in Niger, it is mainly the electricity production and mining companies which are the biggest users and holders of PCBs waste [11,12], often stored in conditions that do not meet environmental standards. Furthermore, the United Nations Environment Program (UNEP) has set the complete elimination of PCBs for 2028 [13,14]. However, the available techniques of destruction are very expensive, and consume energy. They also present regularly health and environmental risks [11,15]. It is in this context that the advanced oxidation process (AOP), through assisted photocatalysis (UV-Vis/H_2O_2,) appeared to be a simple and effective technique of detoxification and elimination of liquid PCBs or objects contaminated with PCBs [12,16]. These AOP processes are the least expensive and more environmentally friendly to implement [17]. Given the complexity of photocatalysis where the photonic effect, the solvent effect, and the catalyst effect combine, it was first studied the effect of light intensity related to the type of radiation and the distance from the light source to the reactor. Also, the influences of solvent, pH, initial concentrations on PCBs photolysis were analyzed. The used of peroxide (H_2O_2) as oxidant was investigated, which allowed to establish the photoreactivity of OH• radicals on PCBs molecules degradation in aqueous solution. Finally, the photodegradation kinetics of PCBs congeners contained in the Askarel formulation, under optimum conditions was carried out.

2. Materials and Methods

2.1. Chemical Reagents and Materials

The Askarel formulation contained the PCBs congeners was gotten from an electrical transformer N°536 (as identified by TREDI®), stored at temporary PCBs waste storage site (Niamey, Niger) (Figure 1). The aqueous solution of Askarel was prepared in a mixture of distilled water and ethanol (≥99.8%, Sigma-Aldrich). The photodegradation of PCBs congeners and its kinetics were measured using a UV-Vis spectrophotometer (Thermo Fisher Scientific, Evolution 300 UV-Vis, Madison, USA). 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 H_2O_2 (30%) was purchased from Han Zhoo (China). The pH of the solutions was measured using a multimeter, Multi 3430 SET (Germany). The visible and UV radiation device is composed of two halogens lamps (1000 and 500W) and one LED lamp of 50W (Figure 2).

Figure 1. (**a**) used transformers and drums used for storing solid waste contaminated with pyralene (**b**) transformer oil samples with Askarel

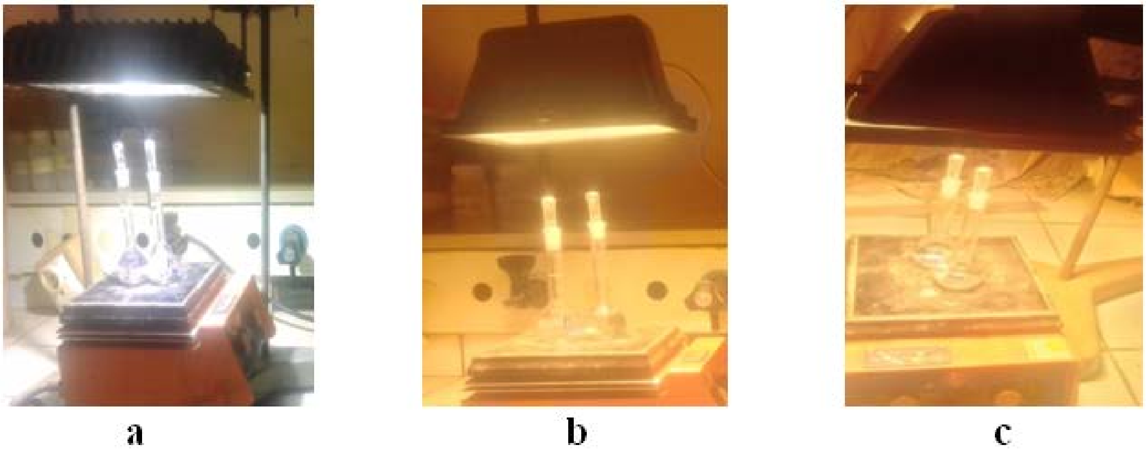


Figure 2. (**a**) LED lamp 50 W, (**b**) halogen Lampe 500 W, (**c**) halogen Lampe 1000 W

2.2. Experimental procedures

2.2.1. Askarel Solution Analysis

The Askarel commercial formulation is a mixture of several PCBs congeners which corresponds to that of Aroclor 1260 [18]. Its aqueous solutions (1:10, ethanol: water) were prepared from the diluted stock made by dissolving 0.5 ml of the concentrated sample from the electrical transformer N°536, in 50 ml of ethanol. The determination of its photodegradation and kinetics, was related to the concentration change according to the spectral analysis at regular time intervals. Thus, given that the density of askarel is 5.45 kg/gallon which corresponds to 1. 438kg.L^{-1} [19,9]. In view of PCBs molecules absorb between 195 and 300 nm, one spectral profile of PCBs congeners contained in askarel, were obtained from the absorption spectra of its aqueous solutions at different concentrations of 50, 40, 30, 20, 10, 5 and 2.5 ppm. using UV-Vis spectrophotometer. (Figure 3) [20,11].

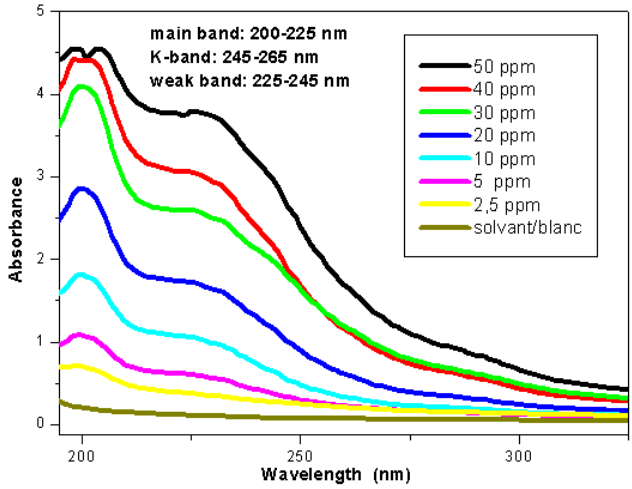


Figure 3. Additive absorption spectra of PCBs congeners contained in Askarel formulation

2.2.2. Photoirradiation process and photodegradation kinetics

Spectrophotometric analysis is a quantitative analysis based on the principle of absorption of light by the substance to be analyzed. This absorbance which is proportional to the concentration and the relationship between the both, is given by the Beer-Lambert law below:

$$A = \log\left(\frac{I_0}{I_1}\right) = \varepsilon l C \text{ , using } \frac{A_i}{A_0} = \frac{\varepsilon l C_i}{\varepsilon l C_0} = \frac{C_i}{C_0}$$

Then the reading absorbance (A) allowed to calculate the corresponding concentrations from the following formula:

$$c_i = \frac{Ai \times Co}{A0}$$

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

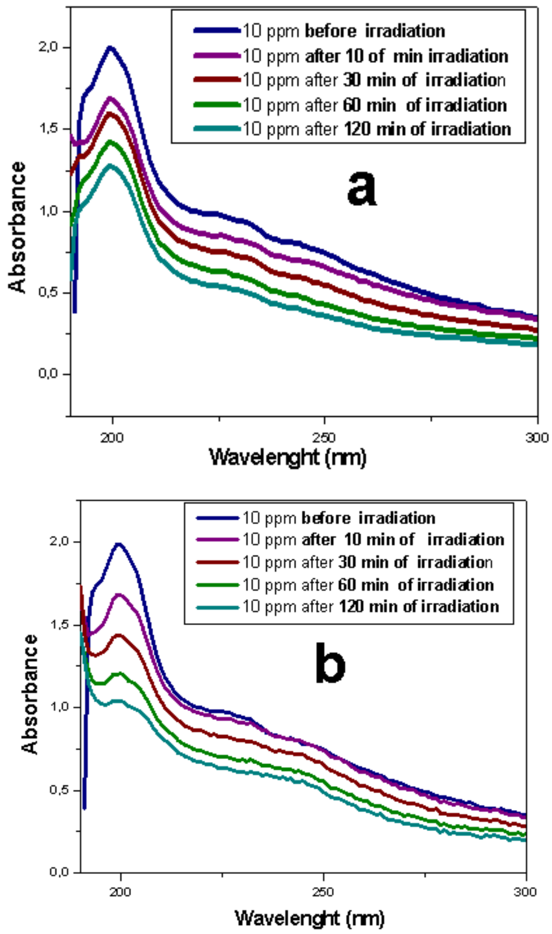
Ai: absorbance at time t,

C0:is the initial concentration.

The Askarel solutions to be irradiated are placed on a magnetic plate at a distance of 40 or 20 cm from the lamp in order to study the radiation intensity parameter. In order to obtain radiation similar to sun light (approximately 1200 $W.m^{-2}$) the 1000 W lamp was placed 20 cm from the reaction point. The influence of the solvent on photolysis was studied by varying the proportion of ethanol: water (1:10, 1.5:10, 2:10) and methanol-water (1:10, 1.5:10, 2: 10). For each experiment the evolution of the temperature was controlled to limit the thermal effect on the boiling temperature of the solvent. The influence of the initial concentration was studied by driving the photodegradation at different concentrations of 10, 20, 30 and 40 ppm. All experiments have been done at pH 6.8 ± 0.1 which is the pH without adjustment giving the best spectral signature of the Askarel mixture in aqueous solution. The kinetics study of the photodegradation of PCBs was based on the model applied to the photodegradation of biphenyl [12]. The influence of hydrogen peroxide on the photodegradation of PCBs was studied by adding the same volume of H_2O_2 at different concentrations to obtain four solutions in 30 ppm concentration of PCBs, with H_2O_2 in 5, 10 and 15 mM of concentration.

3. Results and Discussion

3.1. Influence of Light Flow on PCBs Degradation



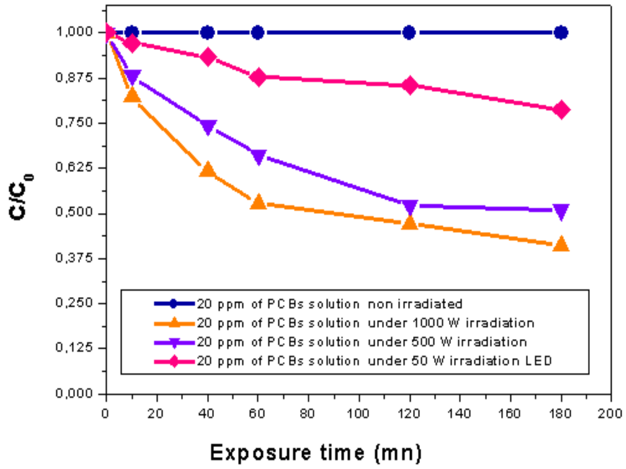
(**a**) 40 cm from the light source; (**b**) 20 cm from the light source (pH = 6.7 ± 5, 1000 W

Figure 4. Spectral tracking of photodegradation of PCBs contained in Askarel during 2 hours of irradiation

To initiate a photolysis reaction, it's required irradiation by light flux. It has been established by several authors [21,22,23] that the speed of the reaction is proportional to the illumination. The intensity of the photon flux which reaches the point of the reaction is related to the nature and type of the light source, and the remoteness from this source (according to different artificial sources and types of lamps), or to the positioning from the sun. Thus, for the same light source (1000 W lamp), according to how the source is placed, from 40 or 20 cm to the reaction medium, the speed and the quantity of PCBs photodegradation are not the same (Figure 4a and Figure 4b.) So, when the lamp is placed 40 cm from the reaction medium the irradiance measured at the reaction point is 470 ± 5 $W.m^{-2}$, while it reached 1300 ± 10 $W.m^{-2}$ when the same source is placed to 20 cm. In this context, the rise on temperature could have an influence on the photonic effect. After 2 hours of irradiation the temperature increased from 27 ± 1 to 65 ± 2°C in the reaction medium when the lamp was placed 20 cm from the reaction point; and from 27 ± 1 to 45 ± 0.5°C, when the lamp was placed at 40 cm from the reaction point. To minimize the thermal effect, after each sampling, the solution is placed outside the irradiation field to lower the temperature. Thus, the sample collected is cooled before analysis.

3.2. Effect of Light Intensity Depending on the Light Source

To assess the effect of light intensity according to the light source, a photodegradation of 25 mL of solution (20 ppm) of PCBs congeners was carried out under irradiation of respectively LED (50 W) and halogen lamps. (500 W and 1000 W). The curves describing the kinetics of these different degradation are shown in Figure 5.



(C_0 = 20 ppm ; pH = 6.8,T_{1000W} = 69.3 °C , T_{500W} = 52.3 °C T_{50W} = 34.1°C)

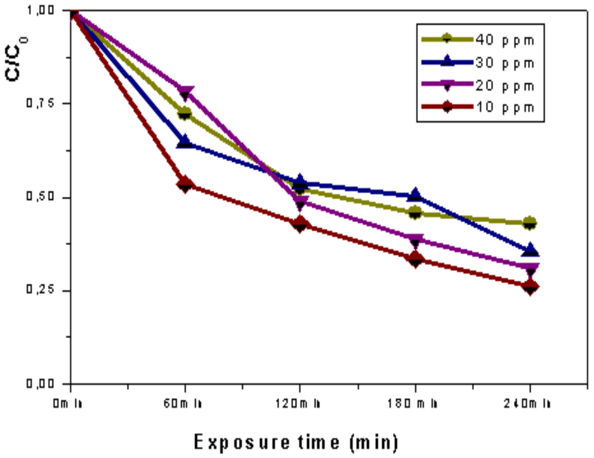
Figure 5. Energy flow effect on the PCBs photodegradation

After 3 hours of irradiation, it's observed that the best degradation is obtained under halogen 1000 W radiation (58.87 ± 3% degradation) followed by 500 W (49.13 ± 2% degradation). Under 50 W LED lamp, the degradation rate is only 21.19 ± 1%.

In fact, at this distance (20 cm) from the reaction medium, the irradiance at the reaction point is 1300 ±10 Wm^{-2} for the 1000 W lamp and 800 ± 5 Wm^{-2} for 500 W and 4 ± 1 Wm^{-2} for the 50 W LED. As we see, the speed of photodegradation is a function of the irradiance and therefore of the energy flow supplied by the source. Also, it should be noted that at this distance from the reactor, the powers supplied can have a thermal effect on photodegradation. This thermic influence of energy flow was observed by Cao and his collaborators [24].

3.3. Influence of Solvent, pH and Initial Concentrations on PCBs Photolysis

After several tests at different ethanol-water proportions, the proportion (1:10) was chosen as the solvent which gave the best photodegradation performance of PCBs in aqueous solution. Considering the influence of pH on the spectral analysis of askarel analysis all experiments have been done at pH 6.8 ± 0.1 which is the pH without adjustment giving the best spectral signature of the Askarel mixture in aqueous solution [11]. From the graph describing the influence of the initial concentration in Figure 6, it's observed that the rate of photodegradation decreases with the increase in the initial concentration. The same phenomenon has been observed by several authors on the photodegradation of PCBs molecules [25,26,27]. This is in agreement with Einstein's law "one photon, one molecule" in the photochemical reaction principle [28,29].

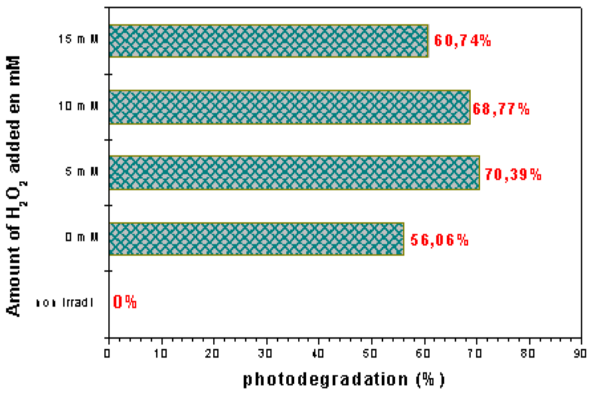


(pH = 6.8 ± 0,1, P = 1000 W, $T_{1000 W}$ = 69.3°C)

Figure 6. Influence of the initial concentration on the photodegradation of PCBs

3.4. Influence of Hydrogen Peroxide on the Photodegradation of PCBs

The addition of H_2O_2 in photochemical reaction is essential for the production of hydroxyl radicals; and this has been experimented by several authors in remedial photodegradation process. The influence of hydrogen peroxide on the photodegradation of PCBs was studied by adding the same volume of H_2O_2 at different concentrations to obtain four solutions in 30 ppm concentration of PCBs, with H_2O_2 in 5, 10 and 15 mM. concentration. Thus, it's observed in Figure 7 that with concentrations of 5 and 10 Mm of H_2O_2 added, the degradation rates are respectively 70.39% and 68.70%. But when the concentration of H_2O_2 increases to 15 mM this rate drops to 60.74%. Overall, the addition of H_2O_2 had a stimulating effect on the photodegradation of PCBs because under the same conditions the degradation rate under direct photolysis is only 50.06%. These results can be explained by the same effect as the high concentration of hydrogen peroxide on the photolysis of biphenyls studied previously [12,30]. This would suggest that increasing the concentration of H_2O_2 acts as a hydroxyl radical scavenger, when the overflow of H_2O_2 absorbs all the available light. In fact, OH• radicals are generated by homolytic cleavage of the O-O bond of radiated H_2O_2 molecules. This performance of the UV-Vis/H_2O_2 process is confirmed by the comparative kinetics of photodegradation of 10 ppm for 3 hours (Figure 8).



(C0 = 30 ppm, pH = 6.8±1, P = 1000 W, T= 69.3 °C)

Figure 7. Influence of hydrogen peroxide (H_2O_2) on the photolysis of PCBs

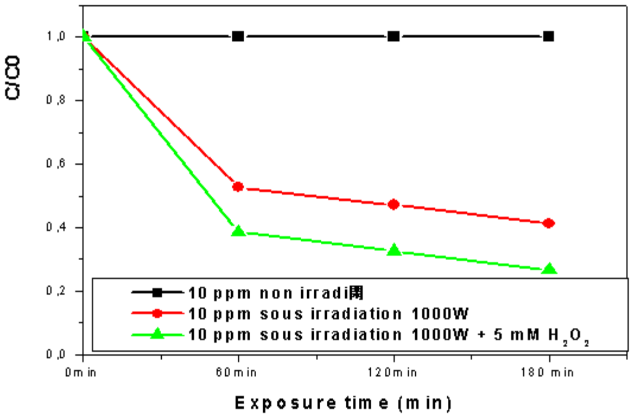


Figure 8. Kinetics of direct PCBs photolysis versus assisted photolysis by H_2O_2 (C0 = 10 ppm, pH = 6.8, P = 1000 W,T = 69.3 °C)

3.5. Photodegradation Kinetics of PCBs

It has been reported by several authors [31,32,33] that the degradation of PCBs obeys the law of first order kinetics. In Figure 9, the graph ln (C0/Ct) = f(t) at different concentrations, allowed to determine the parameters such as kinetic constants Kapp (min^{-1}), linear regression coefficients R^2 and the half-life time $t_{1/2}$.

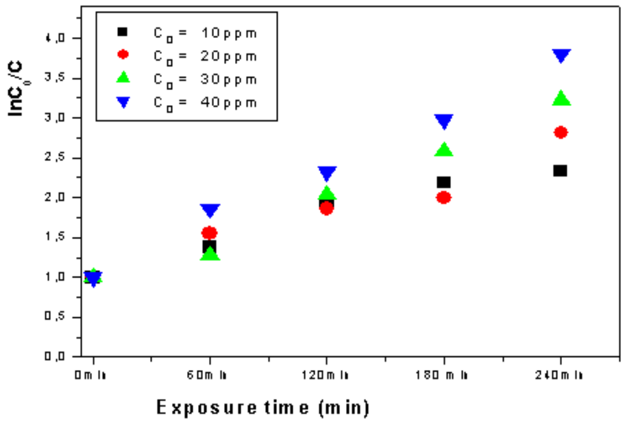


Figure 9. Linear regression lines describing the first order kinetics of PCBs photodegradation

The following table presents two values of R^2 (0.9628 and 0.9616) for the respective concentrations 10 and 20 ppm which confirm the existence of a predominate first order model.

Therefore, the photo-oxidation reactions of PCBs congeners in an aqueous medium is very complex because of the presence of several types of substrates (PCB congeners at different degrees of chlorination); other kinetic models can govern these degradation reactions, what explains the R^2 value (0.785) when the initial concentration increases from 10 to 30 ppm.

Table 1. Kinetic parameters corresponding to the photodegradation of PCBs at different initial concentrations

Initial concentration	K (min^{-1})	$t_{1/2}$ (min)	R^2
10 ppm	0.775	0.894	0.9628
20 ppm	0.657	1.055	0.9616
30 ppm	0.576	1.203	0.741

4. Conclusion

Finally, the performance of the UV-Vis/H_2O_2 process have proven the photoreactivity of PCBs related to OH• radicals, when after addition of 5 mM of H_2O_2, the degradation% under the optimum conditions (Lamp of 1000 W from 20 cm to reaction point, pH = 6.8, Ci = 10 ppm, T = 69.3 °C) during 3 hours, passed from 60% with direct photolysis to 75%. It's established also that the photonic effect, depending on the type of irradiation, remains the driving force of photocatalysis. Though these results showed a relatively slow degradation of PCBs, the optimization of operating conditions can conduct degradation of PCBs until mineralization. However, the performance of direct photolysis under 1000 W irradiation, once its radiation concentrated and the thermal effect regulated, allows to admit the possible use of solar energy as luminous source, which is a guarantee of an ecologically rational elimination of waste or objects contaminated by PCBs.

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