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Mineralization of Wastewater from the Teaching Hospital of Treichville by a Combination of Biological Treatment and Advanced Oxidation Processes

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

Biological treatment, due to its low installation cost, is widely used for wastewater treatment. However, this treatment remains ineffective for the oxidation of so-called emerging molecules. To solve this environmental problem, advanced oxidation processes (AOPs) combine with Biological treatment for rapid, efficient and cost-effective purification of wastewater. This combination used in this work, allowed a total mineralization of a real wastewater solution from the teaching hospital of Treichville named CHU of Treichville in Abidjan (CHUT), both in terms of organic and microbiological pollutants. Real wastewater from the CHUT underwent a Biological treatment for 28 days via the Zahn-Wellens methods which made it possible to have a reduction rate of the chemical oxygen demand of more than 90% of biologically active organic pollutants. The biologically treated

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wastewater was doped with ceftriaxone (CTX) to simulate a situation of wastewater containing a recalcitrant compound after Biological treatment. Subsequently, the doped solution underwent treatment with different AOPs (UV / H_2O_2, Fe^{2+} / H_2O_2 and UV / Fe^{2+} / H_2O_2). This combination resulted in a COD reduction rate of over to be higher 98% and total inactivation of microbiological germs.

Keywords: Zahn-wellens; advanced oxidation processes; wastewater; ceftriaxone.

1. INTRODUCTION

The treatment of hospital wastewater is of concern to all countries of the world today [1,2]. Studies have shown that hospital wastewater is biodegradable [3-6]. But wastewater treatment plants, which in their majority use biological treatment for the purification of hospital wastewater, are faced with a problem of total elimination of so-called emerging organic compounds [7] such as antibiotics and which are found in the receptacle medium [8; 9] after biological treatment (BT). This presence of antibiotics can lead to the proliferation of microorganisms that are bio-resistant [9-13] to their actions.

Indeed, antibiotics are widely prescribed in hospitals because of their effectiveness in the treatment of microbial infections [14,15]. This is the case of the teaching hospital of Treichville named CHU of Treichville (CHUT) in Abidjan where ceftriaxone (CTX) and amoxicillin (AMX) are often prescribed to patients after massive fluids and for whom the wastewater treatment plant does not work. The wastewater from this health facility is therefore discharged into the environment without any treatment. This constitutes a real danger for the population because this wastewater reaches the lagoon [16] where fishing activities are widely practiced. A general awareness is therefore necessary in order to propose effective methods of treating wastewater from the CHUT. In this sense, biological treatment methods such as the Zahn-Wellens (ZW) method have been used. However, biological methods prove ineffective in the face of non-biodegradable toxic compounds [17,18].

To resolve this inefficiency of BT in removing refractory compounds, an alternative has been found namely the combination of BT with advanced oxidation processes (AOPs) [19; 20]. This combination has been very successful in the treatment of dye wastewater [21-23], pulp mill wastewater [24], wastewater containing aromatic and pharmaceutical compounds [25,26].

Thus the objective of this work is to treat wastewater from the CHUT by a BT and AOPs coupling. ZW process will be used like BT, photochemistry, Fenton and photo-Fenton like POAs. ZW process will make it possible to eliminate biodegradable compounds and then the AOPs will make it possible to mineralize non-biodegradable compounds [27-29].

2. MATERIALS AND METHODS

2.1 Wastewater Parameters

Table 1 shows that the wastewater has a pH equal to 7.5. This shows that the wastewater of the teaching hospital is almost neutral. One observed that the temperature of the wastewater of the teaching hospital of Treichville is 29.9˚C.

From Table 1, one observes that the value of the chemical oxygen demand is 229 mgO_2/L and that of the biological oxygen demand is 96.19 mgO_2/L. In this investigation, the ratio COD/BOD_5 has been determined. A ratio inferior to three (ratio < 3) is characteristic of the presence of a great amount of biodegradable materials in the wastewater.

2.2 Zanh-Wellens Test

The Zanh-Wellens test is the method used in this work to remove substances biodegradable by microorganisms for 28 days. The equipment used for this test consists of 2 liter vials carefully washed and sterilized in the oven at 105 ° C for 30 minutes, magnetic stirrers, a magnetic bar, air diffusers (HX-406A) and activated sludge. A mixture containing the test substance, mineral nutrients and a relatively large proportion of aqueous activated sludge is stirred and aerated at 20-25 ° C in the dark or in diffused light for up to 28 days (photo 1). For this work, three different solutions were prepared:Solution 1 is a real wastewater solution from CHUT containing 1.2 g / L of activated sludge and nutrients. Some parameters of the wastewater are given in Table 1.

-Solution 2 is the reference solution containing 1 g / L of sodium acetate, 1.2 g / L of activated sludge and nutrients

-Solution 3 is the control solution containing 1.2 g / L of activated sludge, distilled water and nutrients.

The nutrient solution was prepared according to the methodology based on the Organisation for Economic Co-operation and Development (OECD) ZW test protocol according to its guidelines 302B and 301F. The experimental conditions of the ZW test are presented in Table 2. The experimental setup is presented by the Fig.1.

The degradation rate T (%) of biodegradable organic matter determined at a given time t for 28 days was calculated from the following equation:

$$T(\%) = \left[1 - \frac{C_t - C_1}{C_2 - C_3}\right] \times 100$$

T (%) = Degradation rate of biodegradable organic matter at time t
C_t = COD value of the test or reference solution at time t (mgO_2 / L)
C_2 = COD value of the test or reference solution after 3 h ± 30 min of incubation (mgO_2 / L)
C_1 = COD value measured in the reference solution at time t (mgO_2 / L).
C_3 = COD value measured in the control solution after 3 h ± 30 min of incubation (mgO_2 / L).

Table 1. Physico-chemical parameters of wastewater

Parameters	Values
Potential hydrogen, pH	7.5
Temperature (°C)	29.9
Disolved oxygen (mgO_2/L)	2.09
COD (mgO_2/L)	229
BOD_5 (mgO_2/L)	96.19
COD/BOD_5 ratio	2.38

Table 2. Experimental conditions of the ZW method

Parameters	Standards (OECD 301-302)	This experience (real wastewater from CHUT)
Chemical oxygen demand (mgO_2/L)	100 - 1000	229
Disolved oxygen (mgO_2/L)	>2	4.5 ± 0.1
Activated sludge (g/L)	0.2 – 1	1.3 ± 0.2
Potential hydrogen, pH	6.5 - 8	6.5 - 8
Temperature (°C)	20 - 25	25 -28
Experimental volume (L)	2- 5	2

Fig. 1. Biological treatment device using the ZW method

For the Zahn-Wellens test, the COD and the BOD_5 were determined. The samples examined were aerated for 28 days at a temperature of 25 °C. The process of organic substances degradation was survival from the COD of the samples determined on days 1, 2, 3, 4, 5, 6, 7, 11, 13, 19, 21, 26 and 28 of the experiment. The dissolved oxygen and the pH of the medium were monitored by a dissolved oxygen probe and a pH meter, respectively. The COD was determined using pre-dosed COD tubes of the HACH product. To determine the value of this parameter, 2 mL of sample are taken and filtered and introduced into a COD tube of the HACH product and then heated in a digester at 150 ° C for 120 minutes. After cooling, the COD value is read directly on the DR6000 spectrophotometer (HACH) at a wavelength of 620 nm. For zeroing, a reference solution was used by adding to a COD tube, 2 ml of distilled water. BOD_5 was determined by the "manometric" method using the HACH VELP SCIENTIFICA (HACH) oxitop. This method was developed by Caldwell and Langelier [30]. The principle is based on the measurement of the decrease in pressure linked to the consumption of dissolved oxygen by microorganisms during the oxidation of organic compound. From the change in pressure measured by a manometer, the value of BOD_5 is automatically given. In this work, it was determined before the start of the experiment and after the Zahn-Wellens test.

2.3 Advanced Oxidation Processes (AOPs)

2.3.1 Equipment

The equipment for the treatment using AOPs includes a monochromatic UV-C lamp (λ = 254 nm), a magnetic stirrer, a magnetic bar, a pH meter, a water bath and a quartz tube to protect the lamp and a double-walled quartz system to cool the lamp during the experiment. This whole device was placed in a dark room.

2.3.2 Chemicals

The chemicals used during this experiment are H_2O_2 (Fluka® analytical), H_2SO_4 (Fluka® analytical), $FeSO_4$ (Panreac) and NaOH (Panreac). The ceftriaxone (CTX) used is from LDP TORLAN.

2.3.3 The combination Zahn-Wellens-AOPs

After 28 days of treatment by the ZW method, the COD of the wastewater solution was determined. The value of this parameter being low, a simulation of the presence of antibiotic after biological treatment was developed. To do this, after treatment by the ZW process, the solution obtained was filtered to retain the sludge. The filtrate obtained was doped with ceftriaxone (CTX). For each AOP, 250 mL of the doped solution was used with a concentration of 0.206 M hydrogen peroxide (H_2O_2). A monochromatic UVC lamp (λ = 254 nm) was used for the photochemical (PP) and Photo-Fenton (PPF) processes. For the Fenton (PF) and PPF processes, the ratio $[Fe^{2+}] / [H_2O_2] = 2$ and the pH of the reaction medium was maintained between 2.9 and 3.1. For each process, the electrolysis was carried out for 10 hours with monitoring of the COD which was done in 2 hour intervals. The COD was determined using COD tubes already pre-dosed with the HACH product. To determine this parameter, a quantity of 2 mL of wastewater sample is taken and introduced into a COD tube of the HACH product. The whole is heated in a digester at 150 ° C for 120 minutes. After cooling, the COD value is read directly using the DR / 6000 spectrophotometer of the HACH product at a wavelength of 620 nm.

3. RESULTS AND DISCUSSION

3.1 Biological Treatment (Zahn-Wellens test)

The COD reduction rate of the different solutions during the ZW test is given in Fig. 2. This Figure indicates after 3 hours of treatment, an evolution of the COD reduction rate from 0 % to 67 % for the reference solution and from 0 % to 40 % for the real wastewater solution. Subsequently, a rapid increase in this rate was observed for both the reference solution and the real wastewater during the first two days. After that, the COD abatement rate kept this growth from the second day until the seventh day for the reference solution and reached a value of 99% which remains constant until the 28th day. The same observation was made for real wastewater where the COD reduction rate experienced this upward evolution until the 19th day to reach 95% and subsequently decrease to reach 92% on the 28th day of the experience.

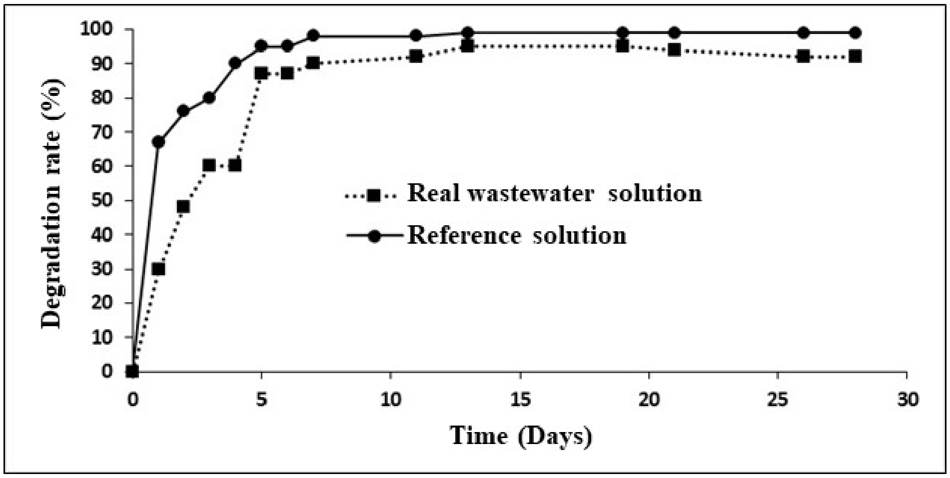

Fig. 2. Rate of degradation of wastewater as a function of time by the ZW; pH = 6.5-8; T = 25 ° C; [activated sludge] = 1.2 g / L; [CH_3COONa] = 1.2 g / L.

In the ZW test, the reference solution is very important because it makes it possible to check the correct functioning of the treatment process as well as the quality of the activated sludge [31]. Quality sludge completely eliminates biologically active pollutants [32]. The reference solution used is sodium acetate as recommended by OECD guideline 301F. According to this guideline, the ZW test is considered valid if the COD reduction rate of sodium acetate reaches approximately 68% in 14 days of experience [31]. In this work, the reference solution reached a COD reduction rate of 99% after 14 days of incubation (Fig. 2). This result shows the good degradation power of the activated sludge used [32]. For the real wastewater, the Figure shows a COD abatement rate of 95% after 15 days of incubation. This result shows that the real wastewater from the CHUT used for this experiment contains biodegradable molecules [32]. This rate then decreased to 92 % on the 28th day corresponding to the endogenous phase, manifested by an absence of substrate or death of the microorganisms. The organic pollutants contained are degraded by microorganisms including a microflora of bacteria and a microfauna of animals, protozoa and metazoa close to the worms contained in the activated sludge [33]. In the presence of dioxygen (O_2), the microorganisms responsible for purification will develop by using the inorganic pollutants (phosphorus and nitrogen) contained in the real wastewater, as a substrate necessary for the production of vital energy to degrade these organic pollutants. During this degradation, these biologically active organic pollutants are transformed into water (H_2O) and carbon dioxide (CO_2) [32], with the synthesis of new living cells (biomass). The 95 % COD abatement rate achieved after 28 days (Table 3) of BT shows that 5 % of organic pollutants in real wastewater from CHUT are said to be biologically inactive.

3.2 Zahn-Wellens and AOPs Process Coupling

After 28 days of BT, the real wastewater was filtered through ordinary wire to retain the solid sludge. The COD of the real wastewater after the Z.W. process being very low, it was doped with CTX to simulate a situation of the presence of non-biodegradable compounds after BT. The COD and BOD_5 values of the doped wastewater solution gave 790 mgO_2 / L and 190 mgO_2 / L, respectively. After doping, the doped wastewater suffered degradation by the various AOPs. The results are reported in Table 4.

Analysis of this table shows that the ZW-AOPs coupling gave a COD abatement rate of 100% for the photochemical process and photo-Fenton process. While this rate is 98.78% for the Fenton process. This degradation is due to the highly reactive and non-selective hydroxyl radicals [34; 35] produced in the reaction medium during the decomposition of the H_2O_2 molecule [36].

Table 3. COD abatement rate after ZW method

Parameters	COD abatement rate
Reference solution	100 %
Real wastewater solution	95 %

Table 4. Characteristics of the doped wastewater after different AOPs

Parameters	Photochemistry (UV/H_2O_2)	Fenton (Fe^{2+}/H_2O_2)	Photo-Fenton ($UV/Fe^{2+}/H_2O_2$)
COD (mgO_2/L)	0	16	0
BOD_5 (mgO_2/L)	-	10.67	-
Rapport COD/BOD_5	-	1.5	-
COD degradation rate (%)	100	98.78	100

For the photochemical process, a H_2O_2 molecule absorbs photons, causing its electronic excitation and the breaking of the O-O bond to form two hydroxyl radicals according to the main reaction described by equation 1 [37]:

$$H_2O_2 + h\nu \rightarrow 2OH^{\bullet} \quad (1)$$

The Fenton process is done in the dark. Thus, hydroxyl radicals are produced by the combination of a molecule of hydrogen peroxide (H_2O_2) and a ferrous ion (Fe^{2+}) called the Fenton reagent. The formation of hydroxyl radicals takes place mainly according to Equation 2 [38]:

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

As for the Photo-Fenton process, the production of hydroxyl radicals is based on the redox reaction between Fe^{2+} and H_2O_2 with the photocatalytic regeneration of Fe^{2+} thanks to UV irradiation reflected by equation 3 and equation 4 [39].

$$Fe^{3+} + H_2O_2 + h\nu \rightarrow Fe(OH)^{2+} + H^{+} \quad (3)$$

$$Fe(OH)^{2+} + h\nu \rightarrow Fe^{2+} + OH^{\bullet} \quad (4)$$

The BT-AOPs combination completely mineralized the real wastewater from CHUT. During the treatment of real wastewater, bacterial inactivation of microbiological germs was investigated. These are total coliforms, Salmonella, faecal streptococci and Pseudominas Aeruginosa. The results obtained are shown in Table 5.

Analysis of Table 5, shows that the ZW-AOPs coupling inactivated bacteria contained in the real wastewater from CHUT. At the total coliforms level, their concentration went from 1.7 10^2 CFU / mL before BT to 12 CFU / mL after the ZW-Photochemistry coupling, i.e. an inactivation rate of 92.29%, from 1.7 10^2 CFU / mL before BT at 10 UFC / mL after ZW-Fenton coupling, i.e. an inactivation rate of 94.12% and 1.7 10^2 UFC / mL before BT at 0 UFC / mL after ZW-Photo-Fenton coupling, i.e. a rate of 100% inactivation. As regards the Pseudominas Aeruginosa, their concentration went from 10 CFU / mL before BT to 0 CFU / mL after the ZW-POAs coupling, ie an inactivation rate of 100%.

For faecal streptococci, the number fell from 1570 10^2 CFU / mL before BT to 0 CFU / mL after ZW-POAs, i.e. an inactivation rate of 100%. The observed inactivation of microorganisms during treatment with the various AOPs is believed to be due to the action of hydroxyl radicals produced during the decomposition of hydrogen peroxide. In fact, the $OH^{\bullet}$ produced in situ participate in the disinfection of water. The action of $OH^{\bullet}$ is similar to the action of some disinfectants such as chlorine used to rid water of impurities [40].

Table 5. Monitoring of microorganisms during the treatment of real wastewater from the CHUT by the BT and POAs coupling

Germs	Real wastewater before BT	Real wastewater after BT	Real wastewater after AOPs		
			Photochemistry	Fenton	Photo-Fenton
total coliforms (UFC/mL)	1.7 10^2	60	12	10	0
Salmonella (UFC/mL)	Absent	Absent	Absent	Absent	Absent
faecal streptococci (UFC/mL)	1570 10^2	0	0	0	0
Pseudominas Aeruginosa (UFC/mL)	10	14	0	0	0

4. CONCLUSION

BT remains the most widely used treatment means to purify hospitals wastewater. However, it does not allow total elimination of so-called emerging organic pollutants and certain microbiological germs. But this work has shown that BT-AOPs coupling can solve this problem even if the so-called emerging compounds remain insensitive to the BT stage. ZW-AOPs coupling resulted in a 100% COD reduction rate for actual wastewater and total inactivation of several microbiological germs. Thus, the BT-AOPs coupling remains a very good alternative for a total purification of wastewater.

At the end of this study, we recommend that the CHUT authorities set up a coupled BT-AOPs system for the wastewater treatment before discharging it into the waters of the Ebrié lagoon. A first treatment will be carried out by the ZW method. Then a second treatment with AOPs will be carried out if the biological treatment fails to completely eliminate the organic pollutants.

DISCLAIMER

The products used for this research are commonly and predominantly use products in our area of research and country. There is absolutely no conflict of interest between the authors and producers of the products because we do not intend to use these products as an avenue for any litigation but for the advancement of knowledge. Also, the research was not funded by the producing company rather it was funded by personal efforts of the authors.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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