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Development of Advanced Oxidation Processes for Landfill Leachate Treatment in Muribeca (Pe-Brazil)

Desenvolvimento de processos oxidativos avançados para o tratamento de chorume de aterro na Muribeca (Pe-Brasil)

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Advanced oxidation processes (AOP's) stand out due to their high efficiency in the degradation of organic compounds with low operational cost. Through a 2^3 experimental design, variables such as pH, hydrogen peroxide, volume and reaction time were assessed. The response studied was the amount of degraded organic matter per mg/L of leachate. To optimize the process, the study sought to maximize the Chemical Oxygen Demand (COD) removal and Total Organic Carbon (TOC) reduction percentage. The white light reactor provided COD removal ranging from 5.3% to 46.6% and TOC reduction from 4.9% to 50.5%. The black light reactor provided COD removal ranging from 20.3% to 58% and TOC reduction from 27.3% to 46.5%. Finally, the sunlight reactor provided COD removal ranging from 21.3% to 77% and TOC reduction from 38.2% to 69.2%. A pseudo-first-order kinetic model was applied, allowing a satisfactory representation of the degradation process.

Keywords: AOP; Photo-Fenton; leachate

Processos oxidativos avançados (POA'S) se destacam devido a sua alta eficiência na degradação de compostos orgânicos com baixo custo operacional. Através de um planejamento experimental 2^3, as variáveis tais como o pH, o peróxido de hidrogénio, volume e tempo de reação foram avaliados. A resposta estudada foi a quantidade de matéria orgânica degradada por mg / L de chorume. Para otimizar o processo, o estudo procurou maximizar a remoção de Demanda Química de Oxigênio (DQO) e Carbono Orgânico Total (TOC) percentagem de redução. O reator de luz branca, desde remoção de DQO variando de 5,3% para 46,6% e redução de TOC de 4,9% para 50,5%. O reator de luz negra, desde remoção de DQO variando de 20,3% para 58% e redução de TOC de 27,3% para 46,5%. Finalmente, o reactor de luz solar, desde remoção de COD variando de 21,3% a 77% e redução de TOC a partir de 38,2% para 69,2%. Um modelo de pseudo-primeira ordem cinética foi aplicado, permitindo uma representação satisfatória do processo de degradação.

Palavras-chave: AOP; Foto-Fenton; o chorume

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1. INTRODUCTION

Landfill is a primary technique currently used in many countries for the disposal of urban solid waste. One of the major problems resulting from this waste management method is the generation of large amounts of leachate, which is a type of effluent with high content of dissolved organic matter[1].

Leachate is heavily polluted wastewater with a composition of complexes containing four groups of pollutants: dissolved organic matter, inorganic macro-compounds, heavy metals, and xenobiotic organic compounds[2]. The treatment of leachates is very complicated, expensive and usually requires applications of various processes due to their high load, complexity of chemical compounds and seasonally variable volume[3, 4].

The conventional treatment of landfill leachate can be classified into four major groups: (a) transfer of leachate: recycling and treatment combined with domestic wastewater[5, 6], (b) biodegradation: aerobic and anaerobic processes[7,8], (c) chemical and physical methods: chemical oxidation[9], Fenton's oxidative treatment[10], adsorption[11], chemical precipitation[12], coagulation/flocculation[11,13], sedimentation/flotation and air stripping[13,14], (d) membrane processes: nanofiltration[15,16], microfiltration[16], ultrafiltration[17] and reverse osmosis[18,19].

AOP's can achieve very high efficiency in the removal of organic matter from leachate, because hydroxyl radicals are extremely reactive and not selective when generated for the destruction of organic matter present in the solution[20]. The Fenton's oxidation is an example of AOP's with high efficiency and low cost[21, 22]. It is a mixture of H_2O_2 and iron salts which can generate hydroxyl radicals that degrade the organic matter present in the leachate at room temperature[23]. The radicals formed are capable of oxidizing a wide range of chemicals in the aquatic environment, and theoretically, all organic compounds containing hydrogen (R-H)[22]. The Fenton's procedure is effective to achieve not only an efficient oxidation of organic compounds, but also for their removal due to coagulation performed in the presence of iron salts[24]. In addition to conventional Fenton's reagent, which is catalyzed by ferrous ion (Fe^{2+}), Fenton reagents can be modified through catalysis with the application of zerovalent iron ions[25], be used alone or be applied in other technologies for leachate treatment[21].

The common feature of all AOP's is the use of reactive free radicals, mainly hydroxyl radicals, which can be generated by various methods, including processes with H_2O_2 or ozone, or light-induced reactions[26].

The aim of this study was to evaluate the degradability of landfill leachate using advanced oxidation processes (AOP's) as a pretreatment step. The photo-Fenton system was studied using white light, black light and sunlight to evaluate the process. The specific objective was to optimize the operational condition of processes through the application of a 2^3 factorial design. Variables pH, H_2O_2 volume and reaction time were also studied. COD removal was quantified through batch tests for the treatment of natural effluents from the leachate using advanced oxidation processes.

2. MATERIAL AND METHODS

2.1 Chemicals and reagents

The leachate, or liquid wastewater studied in this work, comes from the Muribeca Landfill–municipality of Jaboatão dos Guararapes-PE-Brazil.

The collection of leachate samples was performed at the Decantation Lake-DL, located at the beginning of the Manure Treatment Plant–MTP, with random sampling at different points. The leachate was manually collected, filling the entire container with effluent, and packing it into thermal box.

The other reagents, except for the 53% hydrogen peroxide used in the organic matter removal process and in the analysis of substances in contact with the sample under study, were of analytical grade from *Merck S. A. Indústria Química*.

2.2 Leachate characterization

The leachate was characterized by determining the content of heavy metals. The leachate preparation and quantification of metals were performed according to methodology 3111-Flame Atomic Absorption Spectrometric from the Standard Methods[27].

The following parameters were evaluated: turbidity, pH (potentiometric method), alkalinity, TDS, VDS, FDS, phosphorus, COD, BOD, TSS, VSS, FSS, Oils and greases, SS, TS, VS, FS, Chloride, Total Nitrogen and ammonia Nitrogen, which were determined according to Standard Methods methodology[27]. Humic substances were removed from the medium according to method proposed by[28] using chemical precipitation under magnetic stirring at pH adjusted to 1.0 added of sulfuric acid 0.1 N. The precipitated solid was centrifuged and filtered through a millipore membrane of glass fiber GF, 47 mm. COD and TOC were determined in the filtrate obtained using a TOC analyzer Model TOC-V-CSH-SHIMADZU.

2.3 Factorial design

Initially, the liquid wastewater was acidified to pH 1 for 30 minutes with 0.1 M sulfuric acid for precipitation and subsequent removal of humic acids, which interfere in the COD analysis. Then, the leachate was filtered and submitted to the AOP process according to the factorial design shown in Table 1.

Table 1: Variables and levels studied in the 2^3 factorial design

Variables	Levels		
	Lower (-)	Intermediate (0)	Upper (+)
H_2O_2 (mL)	0.2	0.3	0.4
Time (h)	2	4	6
pH	3	4	5

Initially, iron was not added to the wastewater for the performance of AOP, since the sample showed 7.67 mg/L of the iron element. Then, 50 mL of wastewater were added to a 100 mL beaker, and following the experimental design used, the pH was adjusted with a sodium hydroxide solution 0.1 mol/L and hydrogen peroxide was added. Assays were performed in triplicate and the beakers were sealed and placed in black light and white light reactors or exposed to sunlight for the time established in the factorial design.

According to method proposed by[28], the iron concentration in the sample should be around 300 mg/L, so that there is removal of residual peroxide, and then through stoichiometry, 0.00075 mg $FeSO_4$. 7 H_2O/mL was added to the sample to obtain the expected concentration, leaving the sample to rest for 30 minutes. Then, the sample was filtered to determine the residual organic matter concentration by COD and TOC analysis.

2.4 Kinetic study

According to the results from the factorial design, degradation kinetics was carried out using 100 mL beakers containing 50 mL of leachate. From an initial solution at pH 8, sulfuric acid was added for the removal of humic substances, thus the solution pH was reduced to 1. Then, sodium hydroxide was added to raise the solution pH to 5. Also, 0.2 ml of hydrogen peroxide were added and the solutions were placed in a shaker IKA 130 CONTROL, according to degradation kinetics with times corresponding to 5, 15, 30, 45, 60, 120 and 240 minutes. When the kinetic time elapsed, 0.00075 mg of $FeSO_4$. 7 H_2O was added to the solution to obtain a Fe^{+2} concentration in the sample around 300 mg/L, as performed in the factorial design. After 30 minutes, the solution was filtered and submitted to the same COD and TOC analysis.

Based on the work by[29], a kinetic model was tested to represent the experimental data regarding the concentration of organic substances in effluent (C) as a function of the reaction time (t). The phenomenological model can be expressed by the equations below, and equation (1) expresses a pseudo-first-order degradation model of the biodegradable fraction:

$$-\frac{dC_b}{dt} = k \cdot C_b \tag{1}$$

Assuming that the organic compounds in the leachate consist of biodegradable fractions, $C_{b,0,}$ and inert fractions, C_i, at instant t=0, we have:

$$C_0 = C_{b,0} + C_i \tag{2}$$

For $at > 0$, we have:

$$C = C_b + C_i \tag{3}$$

After integration and taking into consideration equations (1) and (2), equation (1) becomes:

$$C = (C_0 - C_i)e^{-kt} + C_i \tag{4}$$

or

$$\log(C - C_i) = \log(C_0 - C_i) - K_t \tag{5}$$

In previous equations: k is the constant in the pseudo-first-order equation, t is the reaction time (s), C is the concentration of organic compounds during the treatment operating cycle (mg O_2/L), $C_{o,b}$ is the concentration of biodegradable organic compounds in the leachate (mg O_2/L) at the initial time and C_0 is the concentration in the raw leachate (mg O_2/L) at time t=0.

3. RESULTS AND DICUSSION

3.1 Leachate characterization

The leachate characterization results are shown in Table 2.

Table 2: Leachate sample characterization

Analysis	Results
Turbidity (NTU)	9.1
pH	7.88
Alkalinity (mg/L de $CaCO_3$)	143.3
TDS (mg/L)	10.198.0
VDS (mg/L em CO)	2.186.0
FDS (mg/L em Mn)	8.012.0
Phosphorus (mg/L)	21.6
COD (mg O_2/L)	6.077.2
BOD (mg O_2/L)	136.0
TOC (mg/L)	1458,7
TSS (mg/L)	188.0
VS (mg/L)	184.0
FSS (mg/L)	4.0
Oils & grease (mg/L)	11.1
SS (ml/L)	0.25
TS (mg/L)	10.386.0
VS (mg/L)	2.370.0
FS (mg/L)	8.016.0
Chloride (mg/L)	20.095.5
Total Nitrogen (mg/L)	257.8
Ammonia nitrogen (mg/L)	153.6

The following metals were also obtained in the characterization of the material, with measurements shown in Table 3 below:

Table 3: Determination of heavy metals in the leachate

Parameters Analyzed	Concentrations (mg/L)	CONAMA Resolution N^o. 357 of March 17, 2005
Total iron (mg/L of Fe)	7.67	15.0 mg/L of Fe
Cooper (mg/L of Cu)	0.11	1.0 mg/L of Cu
Zinc (mg/L of Zn)	0.28	5.0 mg/l of Zn
Cadmium (mg/L of Cd)	ND	0.2 mg/L of Cd
Cobalt (mg/L of Co)	0.31	-
Manganese(mg/L of Mn)	0.18	1.0 mg/L of Mn
Plumb (mg/L of Pb)	0.24	0.5 mg/L of Pb
Nickel (mg/L of Ni)	0.47	2.0 mg/L of Nì
Chrome (mg/L of Cr)	ND	0.5 mg/L of Cr

Table 3 shows that all parameters analyzed are within specification of CONAMA Resolution N^o. 357/2005, especially regarding the presence of 7.67 mg/l of iron in the sample; thus, it is not necessary to add iron for the degradation reaction to occur using AOP.

3.2 Factorial design

The best results for the three types of radiation have higher reaction time (6 hours) and lower pH (3) in common, according to Table 4. In the black light assay, it was observed that the results were beyond expectations, since the results of the intermediate point should be within a range from the highest and lowest removal percentage.

Table 4: COD and TOC removal percentage

				% removal		% removal		% removal	
Assays	VARIABLES			White light	White light	Black light	Black light	Sun light	Sun light
	H_2O_2 (mL)	Time (h)	pH	COD	TOC	COD	TOC	COD	TOC
1	0.2	2	3	17.5	13.7	34.0	27.3	44.1	44.6
2	0.4	2	3	25.6	10.9	42.0	35.4	21.3	38.2
3	0.2	6	3	12.3	34.7	39.6	40.2	77.0	60.4
4	0.4	6	3	41.3	50.5	53.0	29.9	39.1	55.9
5	0.2	2	5	11.0	4.9	20.3	46.5	41.6	45.5
6	0.4	2	5	10.8	8.4	50.2	37.2	26.4	47.4
7	0.2	6	5	46.6	28.7	28.6	31.9	75.7	69.2
8	0.4	6	5	5.3	26.5	40.4	40.8	37.2	58.8
9	0.3	4	4	39.4	36.0	51.0	41.6	41.7	58.2
10	0.3	4	4	38.9	34.7	58.0	42.5	37.5	57.4
11	0.3	4	4	39.3	36.0	53.0	27.3	42.6	58.3

The following maximum COD removal percentage (46.6, 58.0, 77.0%) and maximum TOC reduction percentage (50.59, 46.5, 69.2%) for white, black and sunlight radiation were obtained, respectively. It was observed that sunlight showed the best results for both COD and TOC.

The Pareto diagrams of COD factorial design are shown in Figures 1, 2 and 3, while the Pareto diagrams of TOC are shown in Figures 4, 5 and 6.

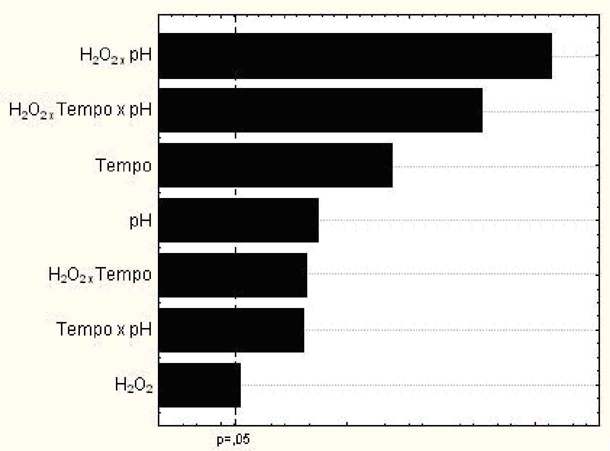

Figure 1: Pareto Diagram of COD using white radiation

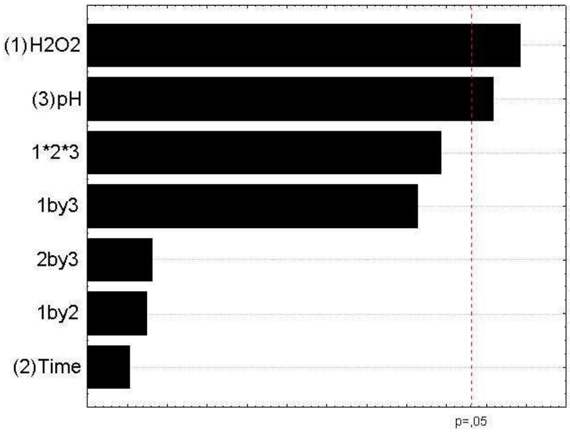

Figure 2: Pareto Diagram of COD using black radiation

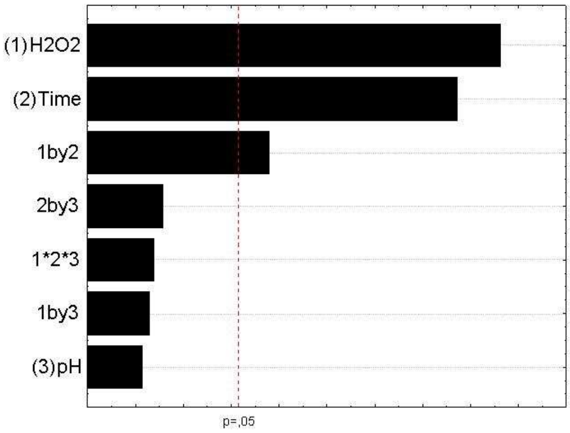

Figure 3: Pareto Diagram of COD using solar radiation

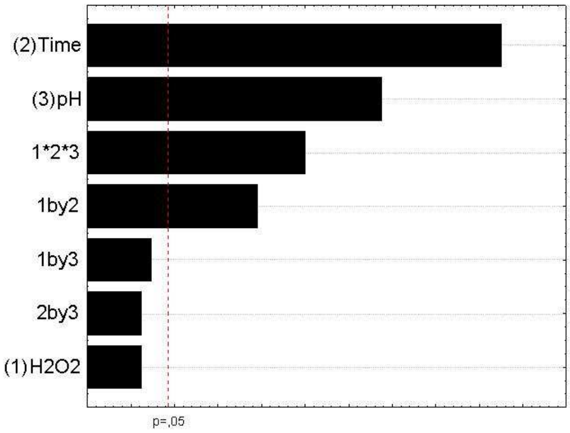

Figure 4: Pareto Diagram of TOC using white radiation

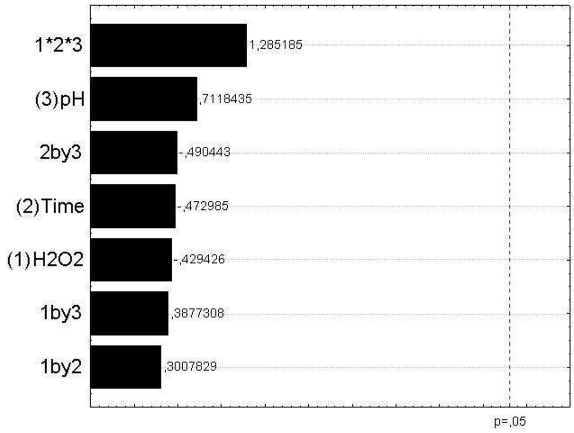

Figure 5: Pareto Diagram of TOC using black radiation

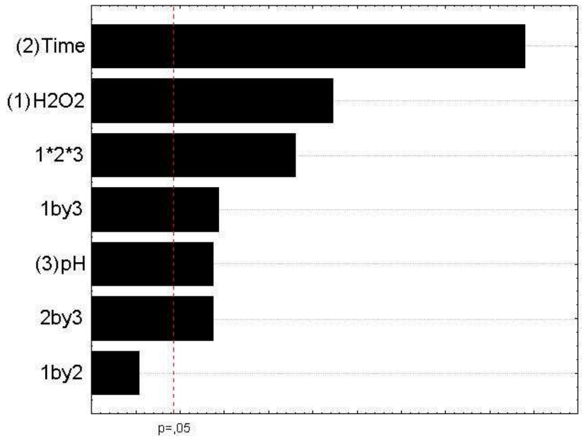

Figure 6: Pareto Diagram of TOC using solar radiation

The values that exceed the baseline, i.e., those corresponding to the confidence interval of 95% are statistically significant[30]. Pareto diagrams show that all factors studied for tests using white light, for COD, are statistically significant. The TOC tests show that time, pH and the interactions between H_2O_2-time-pH and H_2O_2-time are statistically significant, while that H_2O_2-pH and time-pH interactions, does not show statistical significance and H_2O_2 alone, also does not influence the process statistically.

The tests using black light, relative to COD, presented only the volume of hydrogen peroxide and pH as being statistically significant, while that in the TOC any factor presented statistical relevance.

For the tests using sunlight, relative to COD analyses, the volume of H_2O_2, the time and the H_2O_2-time interaction are statistically significant. The interactions between time-pH, H_2O_2-pH-time and H_2O_2-pH, does not have statistical significance, as well the pH. For the analysis of TOC the only value that does not have statistical significance is the interaction H_2O_2-time.

3.3 Kinetic modeling of the organic matter removal

Based on the 2^3 experimental design applied to this study, it was found that higher COD removal occurs in the sunlight reactor. Therefore, only the reaction that used this source of energy was applied to kinetic studies to obtain COD removal of at least 1265.2 mgO_2/L, corresponding to 79.18% of removal in a time of 240 minutes.

Using the Visual Fortran 95 software, the experimental COD's were calculated for the kinetic study. This rapid and first degradation should be related to the dissolved and initially pre-treated organic matter, for the removal of humic substances (humic acids), which interfere in the physicochemical analyses such as COD and TOC, according to[28].

To determine the model parameter k, the results of the kinetic study with COD control by adopting equation (5) were used. The adjustment was performed by linear regression in Fortran language. The k value was estimated to be 6.026 10^{-03} min^{-1}, with a relative error of 8.3%, indicating a good fit between proposed model and experimental data.

From Figure 7, a linear regression coefficient equal to 0.924 could be observed through the intersection formed by experimental and calculated COD data, confirming the good fit of experimental and calculated data.

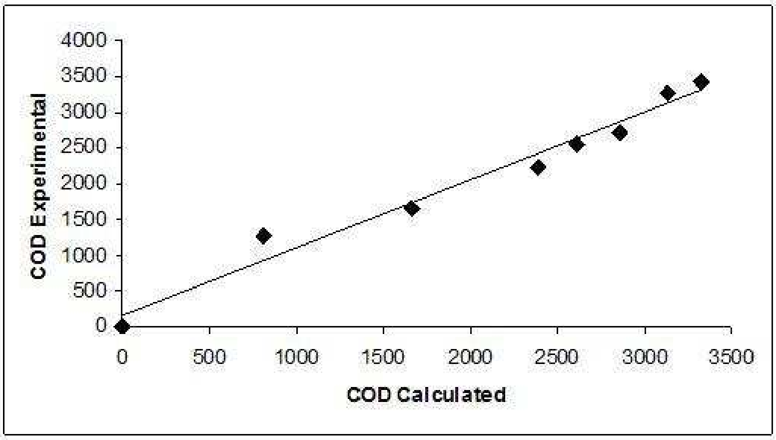

Figure 7: $COD_{exp.}$ *x* $COD_{Calculated}$

In the studies by[31], the authors studied the physical and oxidative removal of organic compounds using Fenton treatment in municipal landfill leachate. This researcher used tests to determine the functions of oxidation and coagulation in the removal of the mature organic leachate content, in which Fenton treatment is present. Working with the following conditions: initial pH 3, mole fraction $[H_2O_2]/[Fe^{2+]}$ = 3 and $[H_2O_2]$ = 240 mM, and six dosage stages, the authors obtained 61% COD removal, while this study obtained COD removal percentage of 77.0% in studies conducted in sunlight reactor.

4. CONCLUSION

Through the factorial design, it was found that the COD removal in the white light reactor showed all statistically significant effects, being the time of exposure to the reactor the most significant effect. The TOC analysis in the white light reactor showed the same characteristics as the COD analysis, and all effects were statistically significant, being the time of exposure to the reactor the most significant effect. This reactor provided COD removal percentage ranging from 5.3% to 46.6% and TOC reduction ranging from 4.9% to 50.5%.

Also through the factorial design, the black light reactor showed, among the main effects, that the hydrogen peroxide concentration was the only statistically significant variable for the COD analysis, while for the TOC analysis, it was found that among the main effects (pH, H_2O_2 concentration and time), only pH was statistically significant. This reactor provided COD removal percentage ranging from 20.3% to 58% and TOC reduction ranging from 38.2% to 69.2%.

Also in the factorial design, the sunlight reactor showed, among the main effects, that the hydrogen peroxide concentration and exposure time were statistically significant, while for the TOC analysis, it was found that the main factors were statistically significant. This reactor provided COD removal percentage ranging from 21.3% to 77% and TOC reduction ranging from 38.2% to 69.2%.

The best COD removal was 77% in the sunlight reactor, which showed pH 3 with 0.2 ml of H_2O_2 and an exposure time of 6 hours. The best TOC removal was also obtained in the sunlight reactor, 69.2%, but this time the pH was 5, 0.2 ml of H_2O_2 and exposure time of 6 hours.

The pseudo-first-order kinetic model proposed satisfactorily represented the experimental results of organic matter degradation in function of time, with average relative error of 8.3%.

The COD removal percentage of 77% and TOC reduction of 69.2% gave the method technical feasibility to treat leachate, since it is a pre-treatment that should be complemented with a biological treatment.

5. ACKNOWLEDGMENTS

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