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Use of strontium aluminate powders in the photocatalytic removal of dyes present in water

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Abstract. The removal of Iris No. 17 fabric dye in water using strontium aluminate powders doped with europium and dysprosium, and the radiation from an ultraviolet C lamp during photocatalysis, is reported. The degradation of the dye was studied by UV-Vis spectrophotometry, for two concentrations of strontium aluminate powder and five of the dye for one hour. The chemical composition and morphology of strontium aluminate powders was studied using X-ray scattered energy spectroscopy, X-ray fluorescence and scanning electron microscopy. The pH, total dissolved solids, and electrical conductivity of the water samples before and after treatment were measured using the Handy-Lab 680 multiparameter. It is evident in the morphology of the strontium aluminate powders a synthesis by reaction in solid state, and a chemical composition by weight of strontium oxide and small amounts of europium and dysprosium. The photocatalytic treatment reports an efficiency of 100% in the removal of the dye for the strontium aluminate powder concentration of 500 mg/L, while for 1000 mg/L an efficiency of 98% was found, likewise for a concentration of 60 mg/L and one hour of ultraviolet radiation a first order reaction kinetics in the degradation of the dye was evidenced.

1. Introduction

High concentration dyes that are discharged into surface waters come mainly from the printing industry and textile dyeing [1], the presence of these dyes in water bodies inhibits the insertion of sunlight, slowing photosynthetic activity and growth processes in biota. Their removal from water becomes difficult due to the complex structures of the dyes [2]. To counteract this environmental problem there are several treatment methodologies, among which are advanced oxidation technologies whose object is the degradation of organic dyes by light radiation which can occur basically through three mechanisms: (a) through a process of photolysis induced by energy from a visible radiation source; (b) through a photosensitization process in which visible radiation excites electrons in the π bond of the dye molecule. These electrons are injected into the conduction band of the semiconductor and then the dye is oxidized; (c) under the action of visible light, electrons are promoted from the valence band to the conduction band of the semiconductor through a conventional photocatalytic process to generate active sites for dye oxidation [3], This study applies the mechanism through the aggregation of photocatalytic substances to generate the breaking of the pollutant molecules in this case the direct blue dye 151 belonging to the azo family which are the most consumed and are characterized by the presence of an azo group ($-N = N -$) in the molecule that connects at least two aromatic rings. The azo group has 6 "mobile" (delocalized) electrons, which delocalize from the adjacent aromatic ring. All azo compounds are colored, but not all can be used as dyes [4].



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In this study, the treatment of colored waters with dark blue iris dye (direct blue 151) is performed using photocatalysis with ultraviolet light and strontium aluminate powders (SAP) doped with europium (Eu) and dysprosium (Dy) prepared with the hydrothermal method that consists of a heterogeneous reaction of synthesizing inorganic materials in aqueous media at above ambient temperature and pressure [5], contributing to advanced oxidation technologies in the removal of dyes in waters. For the above, water samples are prepared in different concentrations of dye and SAP following what is reported in the literature [6], the removal efficiency is determined as a function of the concentration of aluminate added and the exposure time to ultraviolet light by determining the color by absorbance through UV-Vis spectrophotometry for each of the samples prepared; additionally, parameters such as hydrogen potential (pH), electrical conductivity (EC) and total dissolved solids (TDS) are measured complementing the study. This work will contribute to technological innovations in the treatment of effluent water from dyeing industries as an alternative for improving water quality.

2. Materials and methods

For the development of the research, the stages presented in Figure 1 are established, which are described below to allow the understanding and continuity of development of the work; in turn, following what was reported in reference [7], the mixed factorial experimental design is used to define the samples to be developed in the laboratory

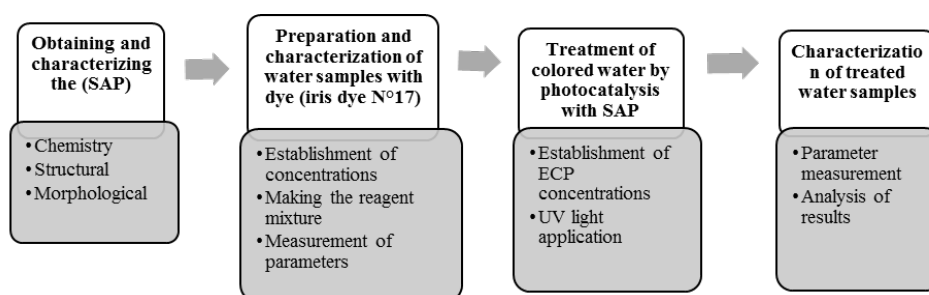


Figure 1. Structure of the methodological design.

2.1. Obtaining and characterization of the strontium aluminate powders

The aluminate strontium powders were supplied by the company Novacolor SAS[®], and in collaboration with the Universidad Pedagógica y Tecnológica de Colombia (UPTC), and Universidad Industrial de Santander (UIS), Colombia, information on the chemical, structural and morphological characterization was obtained.

2.2. Preparation and characterization of water samples with dye (iris dye No. 17)

Following what is reported by [6], each reactive mixture is homogenized for 10 minutes at 500 revolutions and the concentrations of dye to be worked are established for a volume of distilled water of 400 mL as shown in Table 1, the parameters (hydrogen potential pH, total suspended solids TDS, EC) are measured using the HandyLab 680 multiparameter equipment, to distilled water after dissolution of the dye. In Figure 2, an image of the water samples with the dyes for each concentration is presented before the photocatalytic process. In this the coloration intensifies as the amount of the dye increases.

Table 1. Water samples with dyes.

Samples	Concentration (mg/L)	Amount of dye (g)
M1	60	0.024
M2	80	0.032
M3	100	0.040
M4	150	0.060
M5	200	0.080

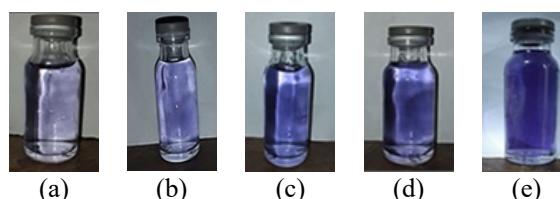


Figure 2. Water samples with dyes. (a) M1; (b) M2; (c) M3; (d) M4; (e) M5.

2.3. Treatment of colored water by photocatalysis with strontium aluminate powders

The dyed water is treated with the SAP concentrations described in Table 2. Applying UV light using a lamp with a wavelength of 255 nm and power of 15 W [8], this process is developed for times of 1 hour, 2 hours, 3 hours, 4 hours and 5 hours, respectively.

Table 2. Strontium aluminate powders concentrations.

Sample	Amount of SAP photocatalyst (g)	Concentration (ppm)
1	0.2	500
2	0.4	1000

2.4. Characterization of treated water samples

Absorbance by UV-Vis is determined using the GENESYS 10S UV-Vis spectrophotometer in the Environmental Quality laboratory for each treated sample; the dye removal efficiency is established and the results are analyzed and interpreted and reported in tables and graphs.

3. Results and discussion

Figure 3(a) shows the composition of the SAPs by XRF, the highest compositions present correspond to strontium oxide and aluminum with a concentration percentage in weigh of 61.7 and 37.3, respectively. In addition to aluminum and strontium, the SAPs contain europium oxide, and dysprosium oxide, these two are used as dopants and are the most important for our project, since they confer the photoluminescent capacity to the powders, which are due to the transition from $4f^65d^1-4f^7$ of the Eu^{2+} , presenting an excitation spectrum with a bandwidth in the range of 260 nm to 400 nm [9]

The morphology of the SAPs by SEM is presented in Figure 3(b), in which it is observed that the particles have irregular sizes and have different shapes, the irregular morphology of small, stacked particles composing larger particles is observed, which is typical of the solid-state synthesis that was used for the SAPs. In the particle size analysis, the ImageJ software was used, found the average particle value of 10.75 ± 5.36 with a minimum size of 1.72 μm and a maximum size of 30.63 μm .

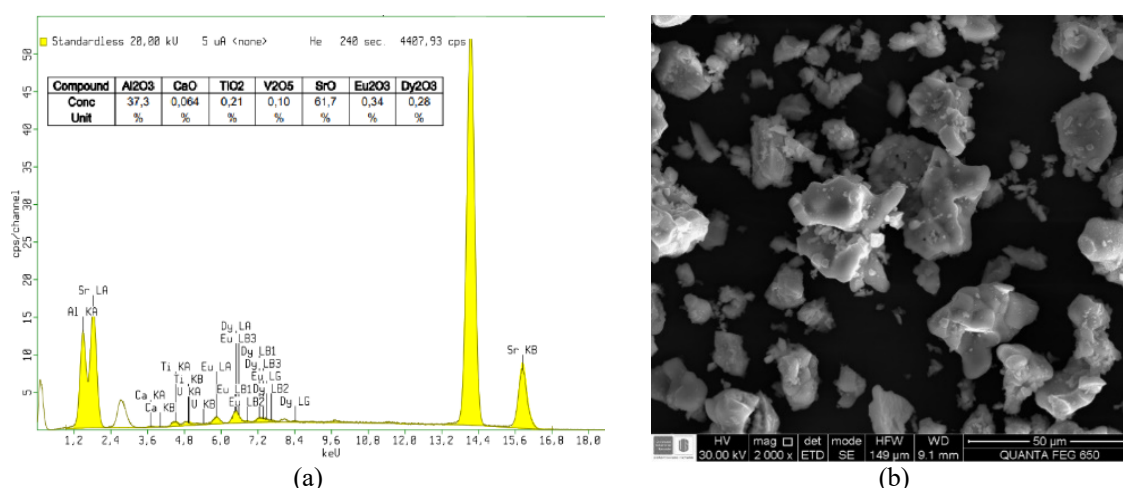


Figure 3. (a) X-ray fluorescence spectrum of strontium aluminate (SAP); (b) image of SEM to 2000X of SAP.

The EDS spectrum and the microanalysis are presented in Figure 4, where the presence of the elements corresponding to Al, Sr, Dy, Eu is appreciated, which confirms what was found by XRF. The measurements of pH, TDS, and EC of the water samples with the colorants before starting the photocatalytic process are presented in Table 3, from these it is observed that as the concentration of the dye increases, the pH decreases which can contribute to effective removal, considering that the catalyst

is more effective in media close to an acidic pH [10]. But both the TDS and the EC increase this is due to the fact that the water acquired impurities, due to increased dye particles [11].

In Figure 5(a), the UV-Vis absorption spectrum for sample M3 is presented, in which it is observed that the maximum absorbance is presented for $\lambda = 561$ nm, the spectra for the other samples are not presented because they are similar. The calibration curve between the absorbances and the concentration carried out for this wavelength is presented in Figure 5(b), which is carried out to determine the efficiency in the degradation of the dye for each established time.

To calculate the percentage of degradation of the dye in the sample using UV-Vis spectrophotometry, the Equation (1) was used, where A_0 corresponds to the absorbance before the photocatalytic process and A_f to the value found after the treatment, both measurements made for $\lambda = 561$ nm, as it was where the maximum absorbance was found (see Figure 5(a)).

$$\% \text{ remotion} = \frac{A_0 - A_f}{A_0} * 100. \quad (1)$$

The photocatalytic degradation of the dye for the studied times was very efficient according to the calculation of the removal efficiency percentage [12], it was found that for the SAP concentration of 0.2 g (500 mg/L) in the sample with the highest concentration of dye ($M5 = 200$ mg/L), for the first hour there is an efficiency in the removal of the dye of 80.8% which increased as the treatment time elapsed, reaching 100% for 5 hours, the above is presented in Table 4.

On the other hand comparing the previous results (see Figure 6) with those obtained for this sample (M5) treated with a SAP concentration of 0.4 g, it can be seen that at one hour of treatment the removal efficiencies are very similar, but for the time between 1 hour to 4 hours, the efficiency was better for the treatment with 0.2 g of SAP, which is due to the fact that possibly to higher concentration of the powders of the SAP, these act as a barrier to incident UV irradiation and prevent them from reaching some particles of dye [13], it is also appreciated that after the fourth hour the efficiencies are practically the same, the above is seen in Figure 6, in which the absorbances as a function of time for the degradation of the dye were plotted using 0.2 g and 0.4 g of the catalyst (SAP). An image of the samples before and after the treatment for each time is presented in Figure 7(a), from which the efficiency of the photocatalytic activity of the SAPs for the degradation of the iris blue dye No. 17 is evidenced.

On the other hand, it was found that the degradation of M4 achieved 100% effectiveness from the third hour of removal with the SAP concentration of 0.2 g, while M3 achieved an efficiency of 100% at the fifth hour using 0.2 g of the catalyst, likewise for M2 it was observed that from 2 hours it reached 100% effectiveness with 0.2 g of SAP; finally, for sample M1, the maximum dye removal was obtained from the first hour of removal with both the SAP concentration of 0.2 g and 0.4 g (see Figure 7(b)). In general, the effectiveness of the treatment for the removal of the dye was when 0.2 g of SAP was used.

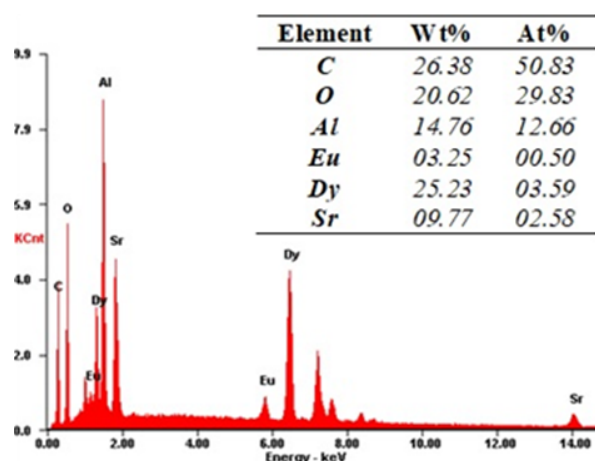


Figure 4. EDS spectra and microanalysis of SAPs.

Table 3. pH, TDS, EC of water samples with dyes.

Samples	pH	TDS (ppm)	EC ($\mu\text{S}/\text{cm}$)
M1	7.8	178	242
M2	7.5	246	246
M3	7.3	306	282
M4	7.2	378	385
M5	7.1	426	410

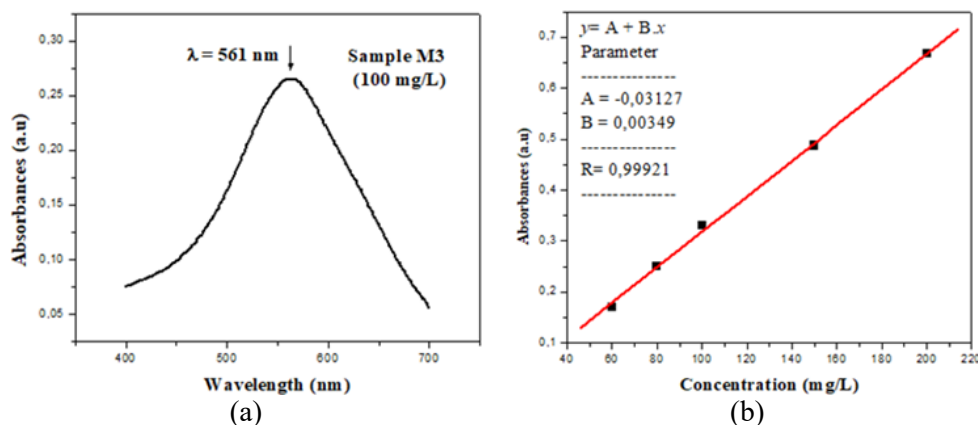


Figure 5. (a) UV-Vis spectrum for sample M3; (b) calibration curve absorbance vs. dye concentration for $\lambda = 561$ nm.

Table 4. Percentages of dye degradation for sample M5, using 0.2 g of SAP.

Time (h)	Absorbance (a.u.) $\lambda = 561$ nm	Degradation of dye (mg/L)
0	0.463	0
1	0.089	80.8
2	0.025	94.6
3	0.005	98.9
4	0.003	99.4
5	0	100

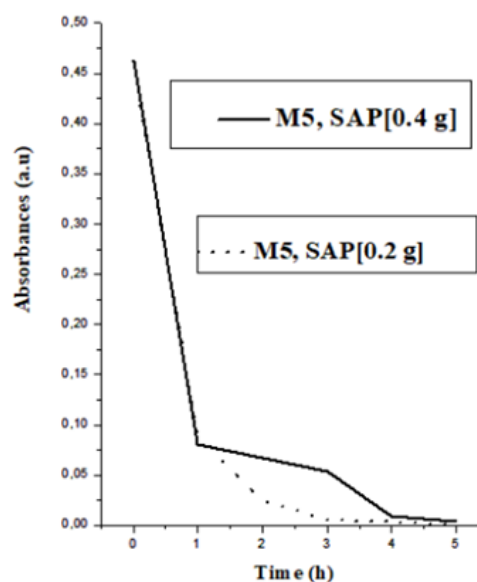


Figure 6. Absorbances vs. time, for sample M5, using 0.2 g and 0.4 g of SAP.

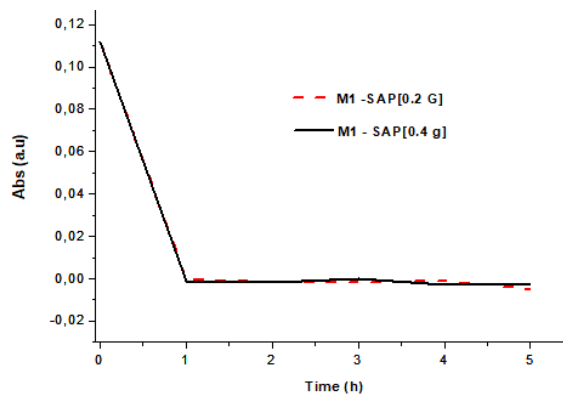
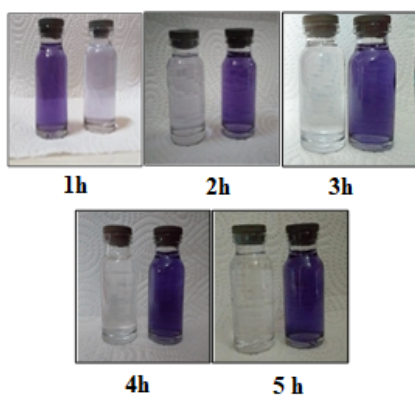


Figure 7. Treatment efficiency. (a) image of sample M5 before and after of photocatalytic degradation using 0.2 g of SAP; (b) absorbances vs. time, for sample M1, using 0.2 g and 0.4 g of SAP.

4. Conclusions

The monoclinic structure of strontium aluminate doped with europium and dysprosium shows a high removal effect when treating water dyed with direct blue dye 151 throughout the treatment, due to the influence of its chemical, morphological, and structural characteristics, allowing it to absorb the organic pollutant with a removal effectiveness of over 95%.

The results were efficient working the treatment in basic medium with hydrogen potential between the range of 7-9, due to the permanent incidence of the lamp on the water to be treated, which allows the easy adhesion of the pollutant to the catalyst improving the removal process, in turn it can be deduced that the removal was more effective for the treatment carried out with strontium aluminate in concentration of 500 mg/L since with this concentration better removal percentages were obtained.

The excess of the photocatalyst used decreases the reaction speed, lowering in turn the removal efficiency, being minimal the amount needed to remove the contaminants, generating a good cost-benefit ratio since the amount and market cost of the catalyst used is low in comparison to the cost of titanium dioxide catalyst mostly used and studied for water treatment, showing that our method can be innovated and applied with high removal efficiencies and low costs.

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