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CONGO RED REMOVAL FROM AQUEOUS EFFLUENTS BY ADSORPTION ON CHERRY STONES ACTIVATED CARBON

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Abstract

Soluble dyes are intensively used in various industrial activities. They represent an important class of water pollutants from which they are rather difficult to remove. This paper focusses on the elimination of Congo Red (CR) ionic azo dye by adsorption on a low cost material. Cherry stones were used for activated carbon preparation by carbonization method. A temperature of 600 °C and a time of 4 h were found as adequate parameters for the physical activation and led to a final white powder with fine particles. The obtained product was put in contact with CR aqueous solutions having concentrations ranging between 200 mg/L and 1000 mg/L and pH values between 2 and 12 and the mixtures were magnetically agitated for periods of 10 to 180 minutes. The best results were recorded at acidic and neutral pH where the CR removal percentage was over 99 % while in alkaline environment the adsorption was negligible. The applied tests revealed that the process is well described by a pseudo-second-order kinetic model (with correlation coefficients close or equal to unity) and that Freundlich isotherm is suitable to insure a very good fit with the experimental data (with correlation coefficients values higher than 0.99).
The acquired results proved that cherry stones (a rarely used waste from food industry) can constitute an interesting and adequate alternative for manufacturing inexpensive materials possessing adsorption properties with high capacity for dye removal from aqueous solutions.

Key words: adsorption kinetics, cherry stones activated carbon, Congo Red dye, Freundlich isotherm

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1. Introduction

Soluble dyes coming from a wide variety of industrial activities (textile, paint, tanning, paper, cosmetic, food etc.) constitute an important class of pollutants (Basheer and Ali, 2018; Basheer, 2018) which is known as being difficult to remove from wastewater due to the high registered values of alkalinity, biological oxidation demand, chemical oxidation demand, total dissolved solids (Basheer, 2018; Srinivasan and Viraraghavan, 2010;) etc.

Biological, chemical and physical methods going from enzymatic degradation (Chacko and Subramaniam, 2011) or anaerobic-aerobic treatments (Manavi et al., 2017) to electrochemical destruction (Liang et al., 2018), electrocoagulation (Ali et al., 2013; Madi et al., 2019), ozonation (Mehrjouei et al., 2015), oxidation (Gagol et al., 2018; Kitinya et al., 2017), coagulation-flocculation (Gil Pavas et al., 2017; Sizykh et al., 2018), membrane filtration (Hosseini et al., 2019) or irradiation (He et al., 2016; Martin de Vidales et al., 2017) are applied for wastewater purification.

These methods are proved to be efficient in removing different dyes from aqueous effluents but they present also specific disadvantages. The biological treatments are not sufficient alone since they may not completely eliminate all dye particles.

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Moreover, they necessitate living microorganisms with length growth phase requiring particular conditions (nitrogen confined area, large reactors etc.) and their use can lead to undesirable by-products formation (methane, sludge, colorless toxic by-products). In terms of chemical dye removal methods, drawbacks such as high electrical energy demand, chemical reagents consumption, and generation of toxic secondary pollution can be cited (Katheresan et al., 2018).

As for the physical techniques, even though they can imply high financial costs and generate important quantities of sludge, due to their simplicity, efficiency and predictability (Yagub et al., 2014) and to the fact that they require the least amount of chemicals, they are the most commonly used methods for dye elimination adsorption being, by far, the best alternative among them (Ali et al., 2013; Ali et al., 2016). The process implies a mass transfer of a substance (called adsorbate) at the interface between a liquid and a solid phase (called adsorbent). It frequently takes place through physical sorption and, in rare cases, by chemical sorption. The materials employed for dye adsorption must possess a high surface area (Ali et al., 2018a; Ali et al. 2018b; Ali et al., 2019a; Nodeh et al., 2016) and an important porosity in order to insure a high diminishment of dye concentration from treated aqueous effluents. Adsorbents such as industrially obtained activated carbon, silica gel, zeolites, clay, alumina or nanomaterials (Ali, 2018; Ali et al., 2017; Ali et al., 2018c; Ali et al., 2019b; Natarajan et al., 2018) are often used to this purpose but, in the last years, low cost materials intensively replace them.

There are researches showing, for example, that Psyllium stalks (Periyaraman et al., 2019) coffee wastes (Kyzas et al., 2012), orange peel (Annadurai et al., 2002), rice husk (Vadivelan and Kumar, 2005), *Pistia stratiote* aquatic macrophyte (Modenes et al., 2018) sewage sludge (Dhouadi and M'Henni, 2009), red mud (Gupta et al., 2004) or activated clay (Ho et al., 2001) can successfully adsorb dyes as Coomassie Brilliant Blue, Black Krom KJR, Basic Blue 30, Methylene Blue, Vat Red 10, Rhodamine B, Fast Green etc.

In this paper we report on the possibility to use by-products from food industry to adsorb dyes from liquid solutions. The experimental program was conducted on three different steps. The first one was focused on the preparation of low-cost activated carbon from dried, grounded cherry stones by physical process of carbonization at different temperatures and time periods. In the second step of the study, the obtained porous material was used as adsorbent for Congo Red (an anionic azo dye) elimination from aqueous solutions and the effect of pH, dye concentration and contact time was examined. At the end, adsorption kinetics and equilibrium sorption behavior of the adsorbents have been studied.

2. Material and methods

2.1. Reagents

Sodium chloride 0.1 N and hydrochloric acid 0.1 N used for the experiments were purchased from Chemical Company (Iasi, Romania).

2.2. Activated carbon preparation

The cherry stones, manually separated from fruits bought from a local market, were intensively washed with tap water to eliminate any residue or strange material, dried for one week at room temperature and crushed to powder with a coffee grinder. Aliquots of 10 g of this powder were placed in porcelain capsules which were introduced in a Caloris L1003 laboratory furnace (Caloris Group, Romania) at temperatures between 500 °C and 900 °C for periods between 4 h and 6 h. After calcination, the capsules were cooled in a desiccator. The obtained product (abbreviated as CS) was weighed and stored in closed recipients at room temperature. The process yield was calculated by multiplying with 100 the ratio between the quantity of the final product and the amount of grounded cherry stones.

2.3. Congo Red (CR)

The analytical purity CR dye powder (molecular structure in Fig. 1) was supplied by Sigma Aldrich (Merk, France). A stock solution (1 g/L) was prepared with distilled wat r. Dilutions to 20 mg/L were adjusted at five pH values (2, 4, 7, 10 and 12) by adding small volumes of hydrochloric acid 0.1 N or sodium chloride 0.1 N. pH measurements were made with a portable pH-meter (Hanna Instruments Service, Romania). The solutions were introduced in quartz cuvettes (pathlength 10 mm) and scanned in a Zuzi 4201 UV-VIS spectrophotometer (Auxilab, Spain). The recorded UV-VIS spectra are given in Fig. 2 and served for determination of maximum absorbance wavelengths which were: 570 nm for pH 2; 530 nm for pH 4 and 500 nm for pH 7, 10 and 12.

2.4. Adsorption experiments

In order to evaluate the adsorption of CR on the prepared cherry stones low cost adsorbent material, 0.1 g CS were put in contact with 20 mL of CR solutions having concentrations of 200, 400, 600, 800 and 1000 mg/L and different pH values (2, 4, 7, 10 and 12). The mixtures were magnetically stirred on a Nahita Blue 692 heating plate (Auxilab, Spain) at room temperature for 10, 20, 30, 40, 50, 60, 90, 120, 150 and 180 minutes. The adsorbent was than separated from the aqueous dye solution in a Nahita 2615/1 digital centrifuge (Auxilab, Spain) set at 3000 rpm for 5 minutes.

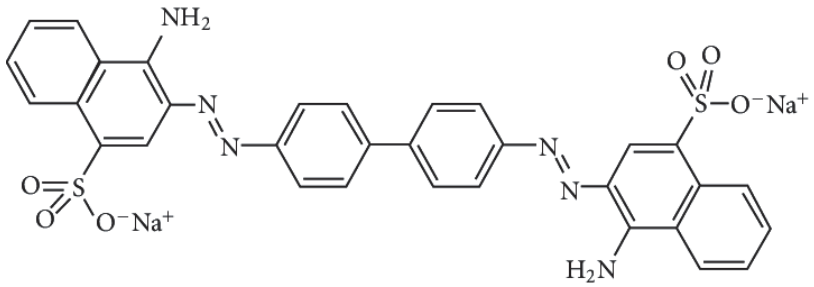


Fig. 1. Congo Red structure

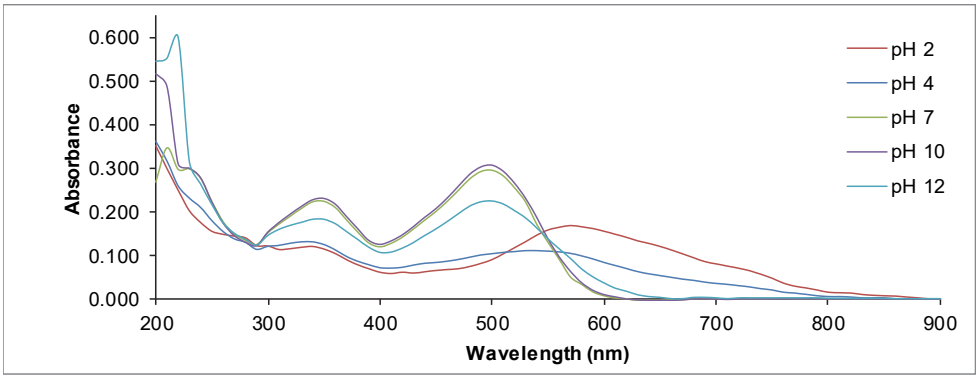


Fig. 2. Congo Red UV-VIS spectra at different pH values

The final residual concentration of each resulting supernatant was determined based on pH specific calibration curves.

2.5. Adsorption kinetics

The adsorption process mechanisms and dynamics were evaluated by calculating the dye uptake. Eq. (1) was employed to this purpose.

$$q_t = \frac{C_0 - C_t}{M} \cdot V \quad \text{(mg/g)} \qquad (1)$$

where q_t (mg/g, mg of CR per g of CS) is the sorption capacity at time t; C_0 and C_t are dye initial and time t concentrations (mg/L); V (L) is the CR solution volume and M (g) is the mass of CS adsorbent used in the experiment.

2.6. Adsorption isotherm determination

CR efficiency removal ($R_{\%}$) from tested solutions was calculated using the Eq. (2):

$$R_{\%} = \frac{C_0 - C}{C_0} \cdot 100 \qquad (2)$$

where C_0 (mg/L) and C (mg/L) are the CR initial and final concentrations respectively.

The amounts of CR adsorbed on CS were calculated using the following equation:

$$q_e = \frac{C_0 - C_e}{M} \cdot V \quad \text{(mg/g)} \qquad (3)$$

where q_e (mg of CR per g of CS) is the equilibrium adsorption capacity; C_0 and C_e are the initial and equilibrium dye solutions concentrations (mg/L); V (L) is the CR solution volume and M (g) is the adsorbent mass.

3. Results and discussions

3.1. Cherry stones activated carbon preparation

Cherry stones are a food waste with wide availability and high mechanical strength but in general abandoned or incinerated. Different researchers analyzed the possibility of using this waste to produce activated carbon by chemical or physical methods. For example, Angin (2014) studied the chemical activation process, which implies the presence of a reagent influencing the pyrolysis, and revealed as optimum conditions an impregnation ratio with zinc chloride of 3:1 and a carbonization temperature of 700 °C. Olivares-Marin et al. (2005, 2012) focused on the use of potassium hydroxide or of sulfuric acid as dehydration agents and of a temperature of 400 °C to 900 °C. Their resulted carbons possessed high specific surface area and porosity. Despite its good results, these methods generate undesirable wastes. That is why an increasing interest is accorded to the possibility of using the easiest available and the most environmental friendly technological processes of activated carbon production. Duran-Valle et al. (2006) evaluated the physical activation which consists in submitting CS to oxidizing gases (air, nitrogen, carbon dioxide etc.) at temperatures of 350 °C to 1000 °C. They applied heat

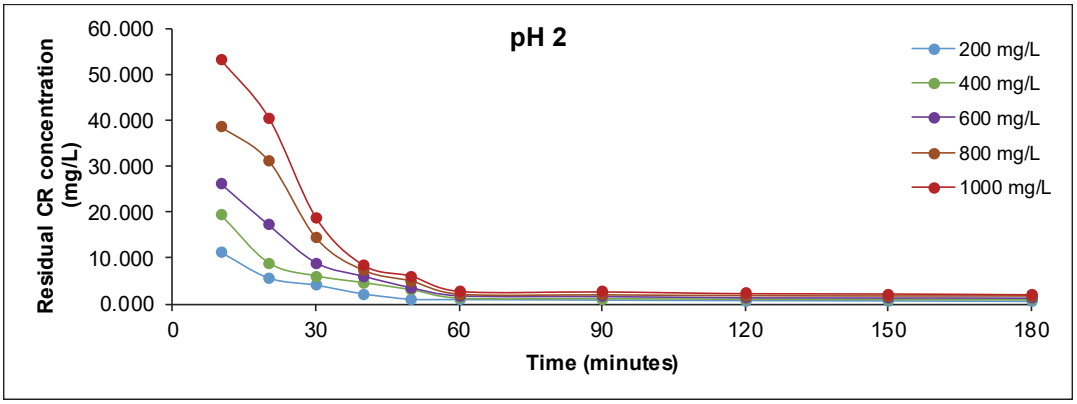
treatments going from 400 °C to 900 °C in nitrogen atmosphere and they concluded that the obtained products are characterized by a low ash content and an important fixed carbon amount being essentially porous materials. Nowicki et al. (2015) submitted the cherry stones to pyrolysis at 500 °C and 800 °C and, after a cooling down period in argon, to a physical activation step at 800 °C under carbon dioxide. Good adsorption properties towards organic (methylene blue, iodine) and inorganic (nitrogen dioxide, hydrogen sulfide) pollutants were registered.

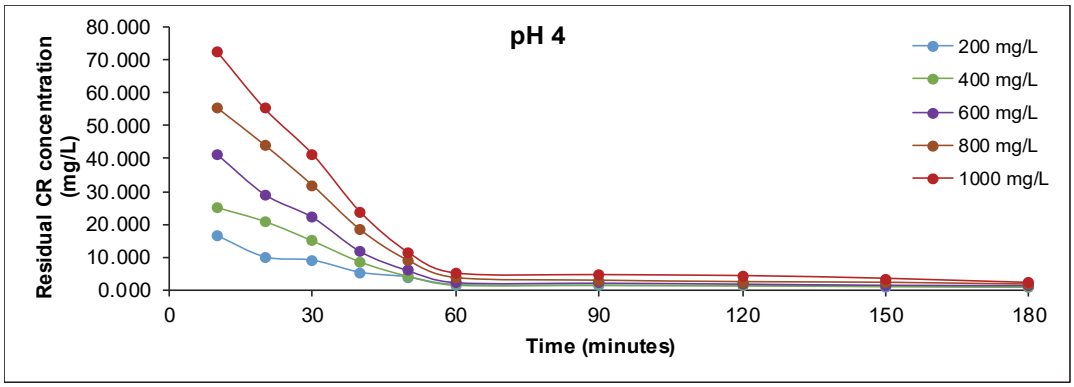
In view of the above, our experiments were directed on the preparation of activated carbon by direct calcination without any preliminary treatment. 600 °C and 240 minutes were considered as adequate and used afterwards. In this case the process yield was of 1.2 %. At a temperature of 500 °C applied for 5 h, the product contained black particles of residual unburned organic material. When higher temperatures (*e. g.* 900 °C) and longer periods (*e. g.* 6 h) were employed the yield was substantially reduced. The evolution from the initial cherry stones to the activated carbon is illustrated in Fig. 3.

3.2. Effect of adsorption parameters

One of the most important parameters affecting the CR adsorption is represented by the pH of dye aqueous solutions. The process has good results at low values (Fig. 4). The color removal is over 99 % at pH 2, 4 and 7 (for a starting concentration of 1000 mg/L). At pH 10 and 12 the removal percentage decreases drastically being of only 14.95 % and 5.99 % respectively. The observations recorded are explained by the fact that at alkaline pH, the HSO_3^- groups of CR turn into $–SO_3^-Na^+$ radicals while the CS has more negative charges which has undesirable effects on the adsorption process due to electrostatic repulsion.

Fig. 3. Cherry stones activated carbon (A – dried cherry stones; B – grounded cherry stones; C – activated carbon)





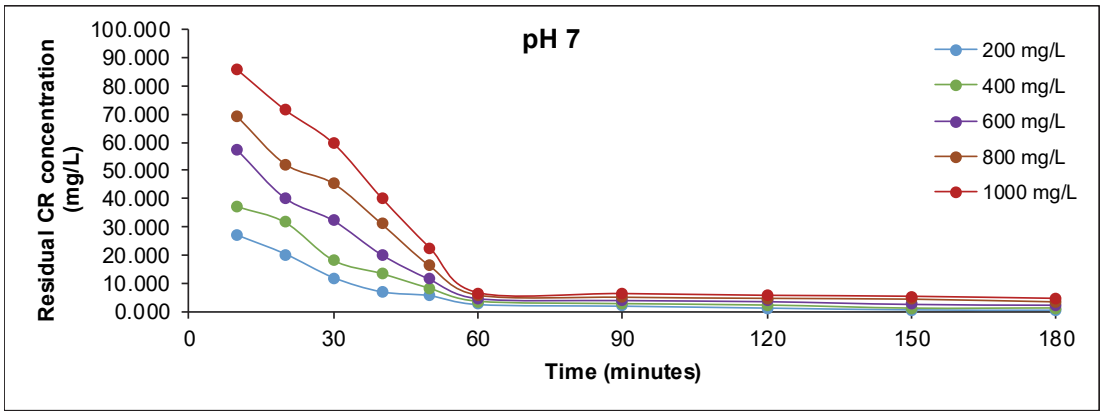


Fig. 4. Residual CR concentration depending at studied adsorption parameters

Moreover, in this case, a CR high concentration increases the ion strength reducing also the CS adsorption capacity. At acidic pH, CR is dissociated to polar groups and the adsorbent has an increased protonation at its surface which favors the interactions between the CS and HSO_3^- groups and enhances the adsorption. These findings are similar with those reported by other studies focused on CR elimination from aqueous effluents by using various adsorbents such as carbonized leonardite (Ausavasukhi et al., 2016), biowaste materials (Kaur et al., 2013), or activated carbon obtained from wood sawdust (Cheng et al., 2015) or with electropositive amine modification (Tian et al., 2018).

Dye initial concentration is another parameter which influences the adsorption. The removal percentage decreased with the increase of CR initial concentration. This indicates that the available active sites of CS are not abundant enough for the adsorption of all the amount of CR existing in the treated aqueous solutions. The process is even more difficult in alkaline environment and high CR concentrations the registered results being far from satisfactory.

In terms of contact time, one can note (also from Fig. 4) that the adsorption takes place systematically in the first 60 minutes after that the CR retained on the adsorbent surface being rather insignificant. Taking into consideration the obtained results, in the experienced conditions, it is recommended to conduct the removal of CR by adsorption on cherry stones activated carbon at pH values between 2 and 7, for at least 1 h.

3.3. Adsorption kinetics

As specified earlier, the contact time has a great impact on CR adsorption. Two recurrently employed kinetic models (pseudo 1st order and pseudo 2nd order) were tested to establish which is more appropriate to describe the studied process. If the pseudo 1st order kinetic was not capable to define the acquired data, very good results were recorded for the pseudo 2nd order kinetic model. Its equation is illustrated by the Eq. (4):

$$\frac{t}{q_t} = \frac{1}{k \cdot q_e^2} + \frac{t}{q_e} \qquad (4)$$

where: k (g/mg·min) is the pseudo-2nd order rate constant, q_e is CR quantity retained by CS at equilibrium and q_t is CR amount adsorbed at time t.

k and q_e values were established from the slopes and intercepts obtained by figuring t/q_t against t at different CR concentrations and are shown in Table 1. The correlation coefficients which are very close or even equal to unity reveal that there were no significant differences between the experimental collected q_e and those determined from the obtained equations. Therefore, it can be considered that the pseudo-2nd order kinetic model successfully describes the adsorption process. This conclusion is sustained also by Li et al. (2016) which concentrated their studies on the CR retention on an activated carbon prepared from a Mexican herb.

3.4. Adsorption isotherms

The adsorption equilibrium can be defined by numerous isotherm models providing information on adsorbent – adsorbate relation. Among them, Langmuir and Freundlich ones are regularly employed. The first is based on the assumption that there are no conjoint interactions between the adsorbed molecules the process occurring only as monolayer adsorption on the active sites of the adsorbent material (Azizian et al., 2018; Hu et al., 2018). The hypothesis of the second mentioned model sustains that the sorption happens initially on the stronger binding sites existing on the adsorbent heterogeneous surface and that the retention diminishes gradually until all the sites are occupied by the adsorbed molecules (Brizi Neris et al., 2019; Putro et al., 2017; Syafiuddin et al., 2018).

Table 1. The pseudo-second-order kinetic parameters for CR removal by CS

Concentration [mg/L]	*Linear equation coefficients*		*Calculated q_e [mg/g]*	*k [g/mg min]*	*R^2*
	a	*b*			
pH 2					
200	0.02498	0.01223	40.032	0.051	1.00000
400	0.01249	0.00542	80.064	0.029	1.00000
600	0.00832	0.00354	120.192	0.020	1.00000
800	0.00624	0.00315	160.256	0.012	0.99999
1000	0.00499	0.00264	200.401	0.009	0.99999
pH 4					
200	0.02492	0.02671	40.128	0.023	0.99998
400	0.01246	0.01092	80.257	0.014	0.99998
600	0.00830	0.00708	120.482	0.010	0.99998
800	0.00622	0.00595	160.772	0.007	0.99998
1000	0.00498	0.00492	200.803	0.005	0.99998
pH 7					
200	0.02477	0.04714	40.371	0.013	0.99997
400	0.01243	0.01734	80.451	0.009	0.99997
600	0.00830	0.01093	120.482	0.006	0.99997
800	0.00622	0.00827	160.772	0.005	0.99996
1000	0.00498	0.00700	200.803	0.004	0.99994

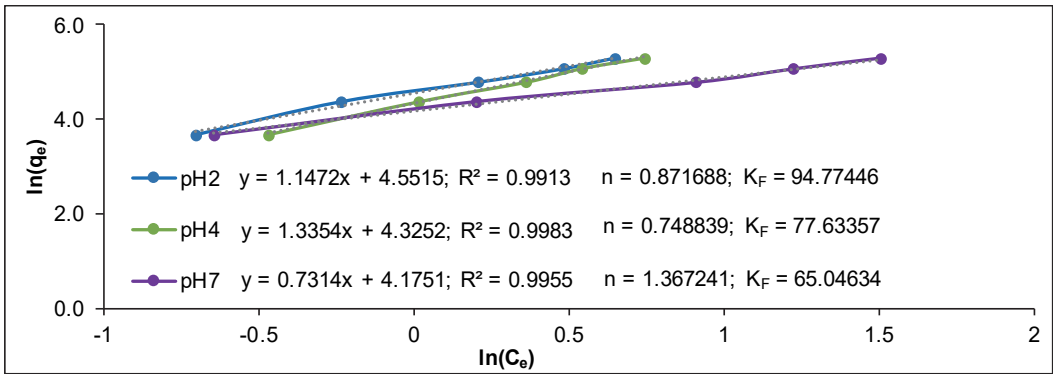


Fig. 5. Freundlich isotherm model of CR adsorption on CS

Our experimental data do not follow the Langmuir model even though there are studies such as that published by Belhachemi and Addoun (2012) showing this type of isotherm along with that of Redlich-Peterson as being highly adequate. Instead, they accurately fit the Freundlich isotherm in which equation's (Eq. 5) q_e is the amount of CR (mg) adsorbed on CS (g), C_e (mg/L) is the dye equilibrium concentration and K_F and n are constants.

$$\log q_e = \log K_F + \frac{1}{n}\log C_e \qquad (5)$$

Linear graphical representation of log(qe) versus log(Ce) (Fig. 5) served to calculate K_F and n constants their registered values indicating an encouraging adsorption. Comparable conclusions were emphasized also by other researches (Mane and Vijay Babu, 2013; Kannan and Meenakshisundaram, 2002).

4. Conclusions

The possibility of using cherry stones for activated carbon preparation by carbonization method was investigated in this study. The obtained product possesses good adsorption properties its abilities to retain pollutants being tested for the removal of Congo Red from aqueous solutions.

The experiments carried out show that, in acidic and neutral media, over 99 % of the existing dye was eliminated after 180 minutes and that a pH above 7 it is not recommended. The conducted research allowed us to conclude that the adsorption process follows a pseudo-second-order kinetics and that it fits with a very high confidence degree the Freundlich isotherm model.

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