
Evaluation of potentially toxic metal biosorption kinetics Mn (II) in acai pit residues (*Euterpe Oleracea* Mart.)

Avaliação da cinética de bioabsorção do íon Mn (II) em resíduos de caroço de açaí (*Euterpe oleracea* Mart.) *in natura* e na forma de carvão ativado em HCl

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ABSTRACT

Contamination of ecosystems with industrial and population advances has caused numerous problems to the environment and human health, as inappropriate disposal of effluents contaminated by potentially toxic metals impairs soil and groundwater. A low-cost and efficient alternative for the removal of these metals is biosorption which can be performed through biosorbents such as the açaí pit residue (*Euterpe Oleracea* Mart.). This work evaluates the reuse of acai pit waste in fresh form and as chemically activated charcoal with HCl in Mn (II) ions biosorption. The reuse of acai residue as Mn (II) ions biosorbent has shown good biosorption capabilities, and modified biomass performed better performance in a shorter contact time than fresh (186.72 mg kg⁻¹ in 10 hours). The kinetic study according to PFO, PSO, ELOVICH, and intraparticle biomass diffusion models in fresh and chemically modified form has shown for both better adjustment to the pseudo-second-order model, indicating that the Mn (II) ion biosorption process occurs via chemisorption.

Keywords: Açaí waste; Charcoal; Biosorption; Kinetic study; PSO;

RESUMO

A contaminação dos ecossistemas com os avanços industriais e populacionais acarretaram em inúmeros problemas ao meio ambiente e a saúde humana, pois o descarte inadequado de efluentes contaminados por metais potencialmente tóxicos prejudica o solo e os lençóis freáticos. Uma alternativa de baixo custo e eficiente para a remoção destes metais é a bioabsorção que pode ser realizada através de bioabsorventes como o resíduo do caroço de açaí (*Euterpe oleracea* Mart.). Este trabalho tem como objetivo avaliar o reaproveitamento de resíduos do caroço do açaí na forma *in natura* e como carvão ativado quimicamente com HCl na bioabsorção de íons Mn (II). O reaproveitamento do resíduo de açaí como bioabsorvente de íons Mn (II) demonstrou ter boas capacidades de bioabsorção, sendo que a biomassa modificada apresentou

melhor desempenho em um menor tempo de contato do que a in natura (186,72 mg kg⁻¹ em 10 horas). O estudo cinético segundo os modelos de PFO, PSO, Elovich e Difusão intrapartícula da biomassa na forma in natura e modificada quimicamente demonstrou para ambas, melhor ajuste ao modelo de pseudo-segunda-ordem, indicando que o processo biossorbitivo de íons Mn (II) ocorre via quimiossorção.

Palavras-chave: Resíduo de açai; Carvão vegetal; Biossorção; Estudo cinético; PSO;

INTRODUCTION

One of the environmental problems currently faced is the pollution of water bodies and soil by potentially toxic metals (Othmani *et al.*, 2022). When they exceed tolerance levels, toxic metals harm the physiological systems of humans and other organisms, because they are not biodegradable and tend to accumulate in the living body, causing various diseases and disorders (Hammo *et al.*, 2021; Nascimento *et al.*, 2015; Nascimento e Oliveira, 2017). Among these potentially toxic metals, the Mn (II) ion stands out.

Traditional methods of physical-chemical treatment such as coagulation, flocculation, sedimentation, filtration, and ozonization correspond to a widely used alternative in the removal of wastewater metals from industrial processes such as galvanoplastic and metallurgical. However, these methods are costly and involve a long period of detention, which makes their implementation difficult (Spain, Plöhn e Funk, 2021; Villen-Guzman *et al.*, 2019).

Biosorption has been presented as a very effective and versatile method used to remove potentially toxic metals in an aqueous solution. Biosorption is considered a subcategory of adsorption, where the adsorbent is a biological matrix. Biosorption is a passive process in which metal ions capture is performed with inactive biomass with high detox efficiency of various effluents and biosorbent reuse (Othmani *et al.*, 2022; Rwiza *et al.*, 2018; Talukdar *et al.*, 2020).

The palm (*Euterpe Oleracea* Mart.), known as açazeiro, is typical of the Amazon region, presenting great economic, social, and cultural importance to the northern region of Brazil since the state of Pará stands out as the largest producer and consumer. The marketing potential of açazeiro products is mainly represented by palm heart, juice extracted from the fruit, as well as the great interest in research and investment of bioactive compounds, antioxidant capacity, and phenolic compounds, mainly anthocyanins present in this fruit (Silva *et al.*, 2019).

Acai fruit pulp configures as a functional product with consumption potential worldwide, but in the agro-industrial processing of the fruit, organic waste is generated, and about 80% of the total processed acai is transformed into waste. In Belém, the state capital of Pará, for example, the trade of acai pulp produces about 300 tons of organic waste per day. Most of this waste is composed of acai pits, which are inadequately arranged on the streets, watercourses, or landfills, and only a small part is used as organic fertilizer or local handicrafts (Cordeiro *et al.*, 2019; Farinas *et al.*, 2009). This work evaluates the reuse of acai pit waste in fresh form and as chemically activated charcoal with HCl in Mn (II) ions biosorption.

MATERIALS AND METHODS

Obtaining and preparation of biosorbent material

The açai fruit (*Euterpe Oleracea*) pit was purchased at open markets, in the municipality of Imperatriz-MA, and fruit processing factories for pulp production. After the acquisition of waste, they were taken to the Environmental Chemistry Research Laboratory (Uemasul - Campus Imperatriz-MA), then washed in running water for debris removal. They were later brushed with polypropylene bristles and washed with deionized water. Açai pits were fragmented and conditioned in porcelain capsules where they were placed to dry in the greenhouse at $60\text{ }^{\circ}\text{C} \pm 5\text{ }^{\circ}\text{C}$ for approximately 24 hours. Then they were ground in the mill and later passed on sieves to obtain particles with average diameters from 2 to 4 mm.

Preparation of chemically modified charcoal with HCl

After 24 hours of greenhouse drying, a portion of açai pits was subjected to the pyrolysis process, they were packed in a large aluminum tray, and burned in an oven at a temperature of $550\text{ }^{\circ}\text{C}$. The burning of pits lasted 5 hours and the process was maintained until all pits acquired the visual characteristics of activated charcoal. After burning it was left for 15 minutes in contact with air for cooling. A portion of charcoal was activated with an acid solution of (HCl 0.1 mol L^{-1}) for 24 hours at room temperature. They were later washed with deionized water, and buffer solution (pH 5.0) and placed to dry the room temperature. Then the fractions were left in the vacuum desiccator until the biosorption experiments (Souza, De *et al.*, 2008).

Potentially toxic metal biosorption kinetics Mn (II) in fresh açai pit residues and the form of HCl activated charcoal.

The removal capacity at any time (min), represented by q_t in (mg kg^{-1}) was determined by equation 1 (Nascimento, Oliveira and Leite, 2019). The tests were performed with two repetitions using 10 mg of biosorbent, and the data obtained were plotted with the test version of the Learning Edition Origin2022b® software.

$$qt = \frac{(C_i - C_t)V}{m} \quad (\text{Equation 1})$$

Where:

C_i = initial concentration of the solution, (mg L^{-1});

C_t = final concentration at any time t, (mg L^{-1});

q_t = biosorption capacity at any time t, (mg kg^{-1});

m = Biosorbent mass, (kg);

V = volume of synthetic aqueous solution with the metal species under study, (L);

To better understand the mechanism of manganese biosorption by acai biomass in fresh form and HCl-activated charcoal, the data obtained from the biosorption kinetics test were applied to mathematical models of adsorption kinetics, from the Linearized pseudo-first-order (PFO), Eq. 2, pseudo-second-order (PSO), Eq. 3, intra-particle diffusion, Eq. 4 and Elovich, Eq. 5 (Elovich, S.Y., Larinov, 1962; Fawzy, 2020; Guo e Wang, 2019; Ho e McKay, 1998; Lagergren, 1898; Nascimento, R.F.; Lima, A.C.A., Vidal, C.B.; Melo, D.Q.; Raulino, 2014; Weber, W.J.; Morris, 1962).

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (\text{Equation 2})$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (\text{Equation 3})$$

$$q_t = k_i t^{1/2} + C_i \quad (\text{Equation 4})$$

$$q_t = \frac{1}{\beta \ln(\alpha\beta)} + \frac{1}{\beta \ln t} \quad (\text{Equation 5})$$

Where:

q_e : Quantity adsorbed by the adsorbent kilogram in equilibrium, (mg kg^{-1});

q_t : Quantity adsorbed by the adsorbent kilogram at time t , (mg kg^{-1});

k_1 : Pseudo-first-order adsorption rate constant (min^{-1});

k_2 : constant in the pseudo-second-order adsorption rate, ($\text{kg mg}^{-1}\text{min}^{-1}$);

K_i : intra-particle diffusion coefficient ($\text{mg kg}^{-1}\text{min}^{-1/2}$);

C_i : Constant related to diffusion resistance, (mg kg^{-1});

α : initial adsorption rate, ($\text{kg mg}^{-1} \text{min}$);

β : constant associated with the extent of surface cover (kg mg^{-1});

T : Time, (min);

RESULTS AND DISCUSSION

In natura acai residue

In figures (1a - 1d) the kinetic profile of manganese ion is represented through the biomass of the in natura acai residue. The analyzed biosorbent has been shown to have good biosorption capacity for the Mn (II) species (Figure 1a). From 2 hours of contact biosorption capacity increased, reaching the value of $186.72 \text{ mg kg}^{-1}$ at the time of 10 hours of contact, after this time, the biomass surface was saturated and again presented biosorption-free sites in time

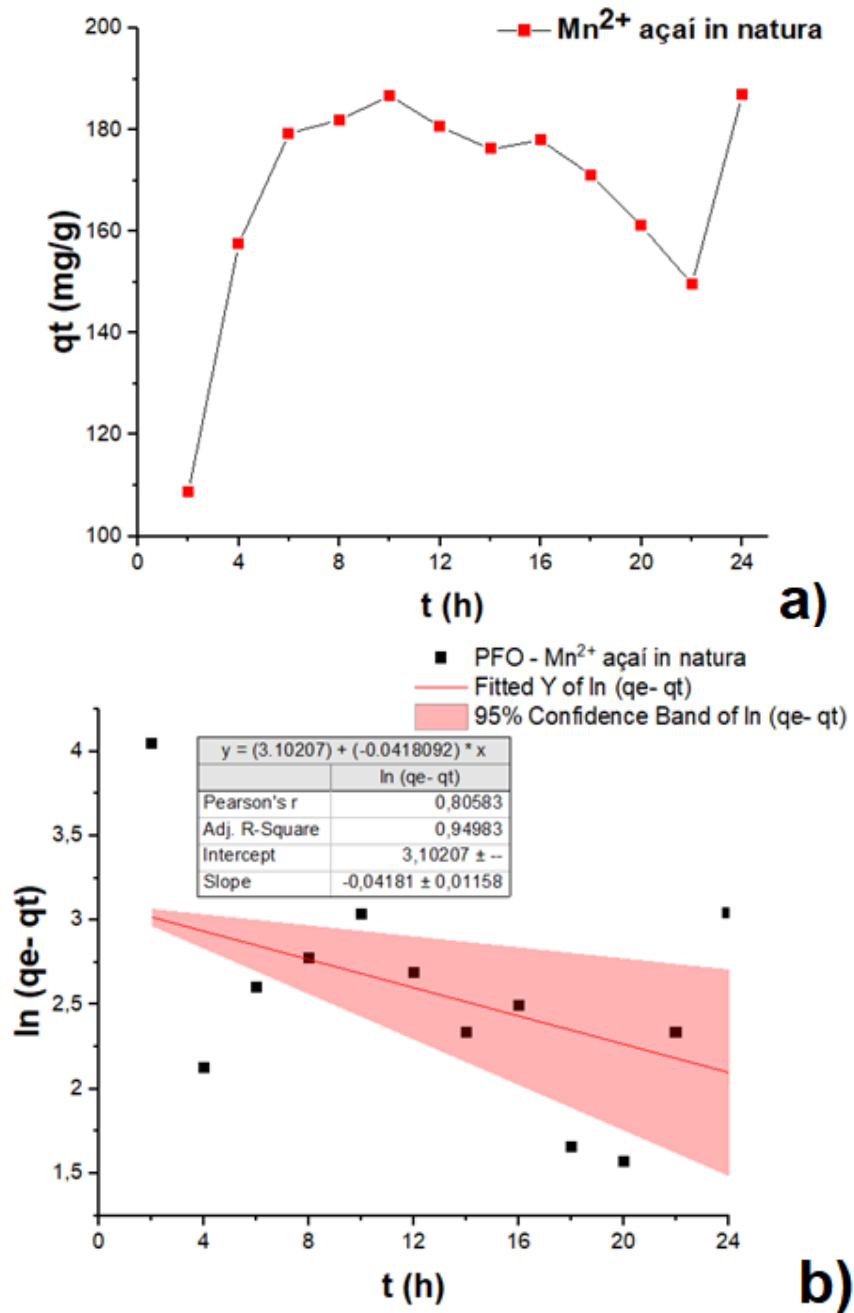
24 hours demonstrating a capacity of $186.87 \text{ mg kg}^{-1}$. This behavior shows that the analyzed biosorption process can be divided into two phases. In the first phase there were many free active sites on the surface of the biosorbent, while during the second phase, these filled sites exerted a strong repulsion to the free metal ions in solution, thus making their biosorption difficult (Nascimento *et al.*, 2021; Xu *et al.*, 2020; Zhang *et al.*, 2018).

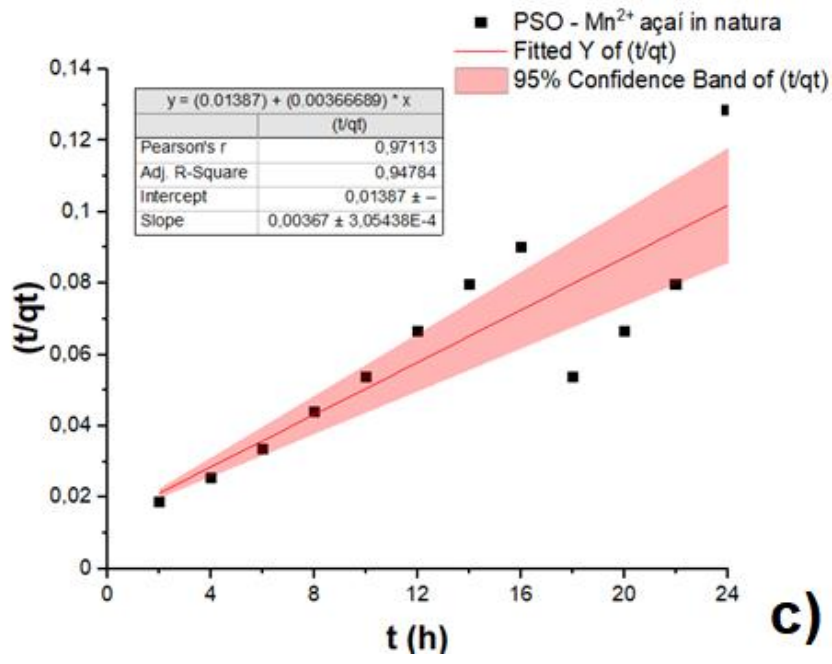
In figures (1b to 1d) the kinetic profiles are represented according to the model's pseudo-first-order, pseudo-second-order, intraparticle diffusion, and Elovich. The analysis of the graphs showed that manganese biosorption using fresh acai residue has better adjustment for the pseudo-second-order kinetic model (PSO) with 0.95 correlation coefficient and calculated capacity ($q_e \text{ cal}$) of $272, 48 \text{ mg kg}^{-1}$ (Table 1). This model assumes that the adsorption process occurs via a chemical bond between adsorbate and adsorbent, through free valences of species involved, therefore, via chemisorption (Ho, Y.S.; McKay, 1999; Nascimento *et al.*, 2021).

The evaluation of the use of the *Caryocar Coriaceum* Wittm bark, a fruit known as Pequi, as a possible biosorbent for copper removal of aqueous solutions, showed that this biomass has a better adjustment to the pseudo-second-order model, which produced a coefficient of Linear Regression of 0.999 (Menezes *et al.*, 2021).

The use of empty bunches of date fruit (DPEFB) as a biosorbent was examined for the removal of hexavalent chromium ions (Cr VI) from synthetic effluents. Kinetic modeling showed that the Cr (VI) ion biosorption process by DPEFB obeyed the pseudo-second-order model more than the pseudo-first-order models order and intraparticle diffusion (Rambabu *et al.*, 2020).

Figure 1. Effect of contact time: a) on the biosorption capacity; b) Pseudo-first-order kinetic plot, c) Pseudo-second-order kinetic plot, d) Intraparticle diffusion kinetic plot, and e) Elovich kinetic plot for the biosorption of manganese ions by açai in natura biomass.





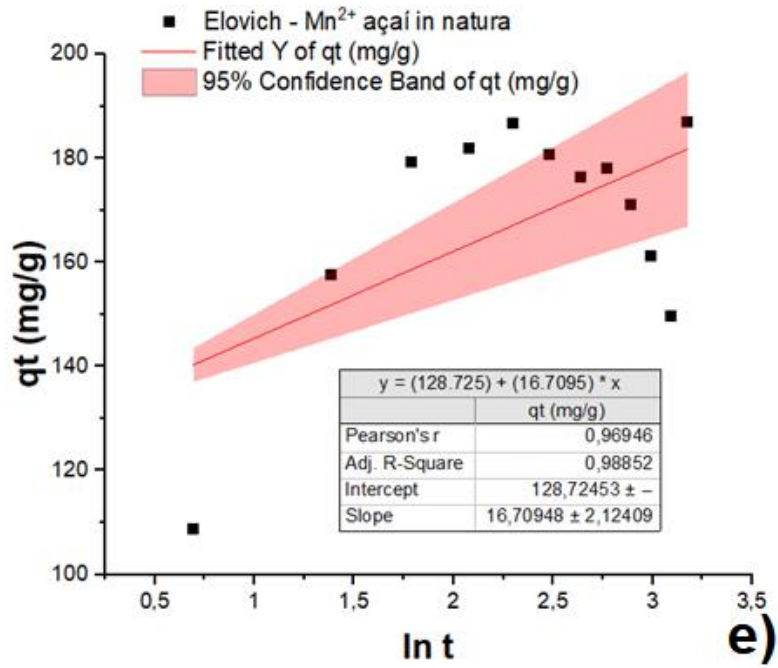
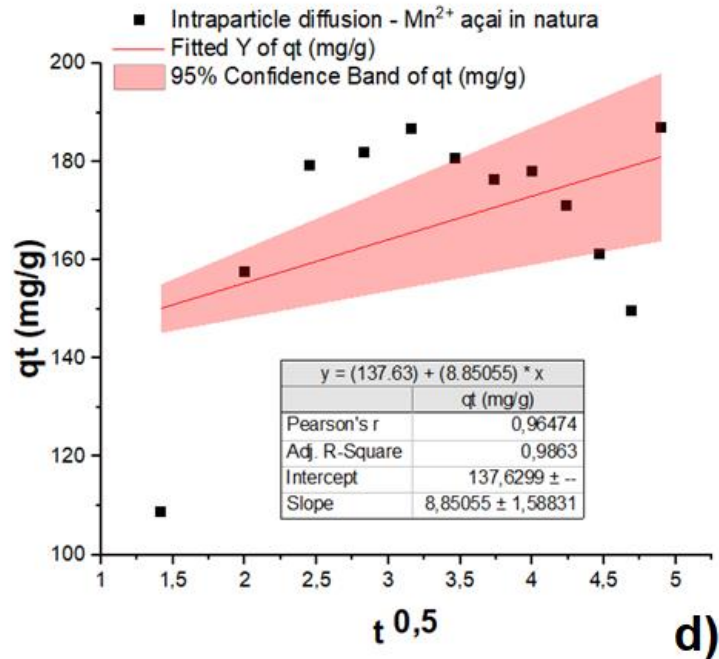


Table 1. PFO constants, PSO, intraparticle diffusion, and kinetic models of Elovich for manganese ion removal using in natura acai.

Kinetics models	Parameters	Value
Experimental data	q_e (exp), (mg kg ⁻¹)	165,88
PFO	q_e (cal), (mg kg ⁻¹)	1,13
	k_1 (min ⁻¹)	0,04
	R^2	0,95
PSO	q_e (cal), (mg kg ⁻¹)	272,48
	k_2 (kg mg ⁻¹ min ⁻¹)	1,87E-07
	R^2	0,95
Intraparticle diffusion	K_i (mg g ⁻¹ min ^{-0,5})	8,85
	C_i (mg kg ⁻¹)	138
	R^2	0,98
Elovich	α (kg mg ⁻¹ min)	2,04
	β (kg mg ⁻¹)	0,06
	R^2	0,98

Acai residues in the form of charcoal treated with HCl

In the figures (2a – 2d) the kinetic profile of the manganese ion is represented through activated charcoal with acai residue biomass HCl. Chemically modified with HCl has been shown to have good biosorption capacity of the Mn (II) species, as well as fresh biomass (Figure 2a). However, biomass modification influenced biosorption kinetics, with the maximum capacity found at 8 hours (182.78 mg kg⁻¹).

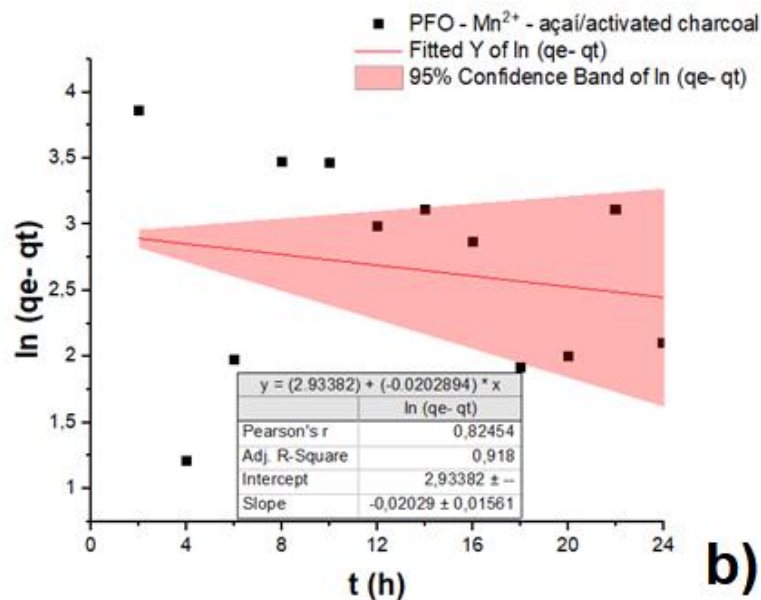
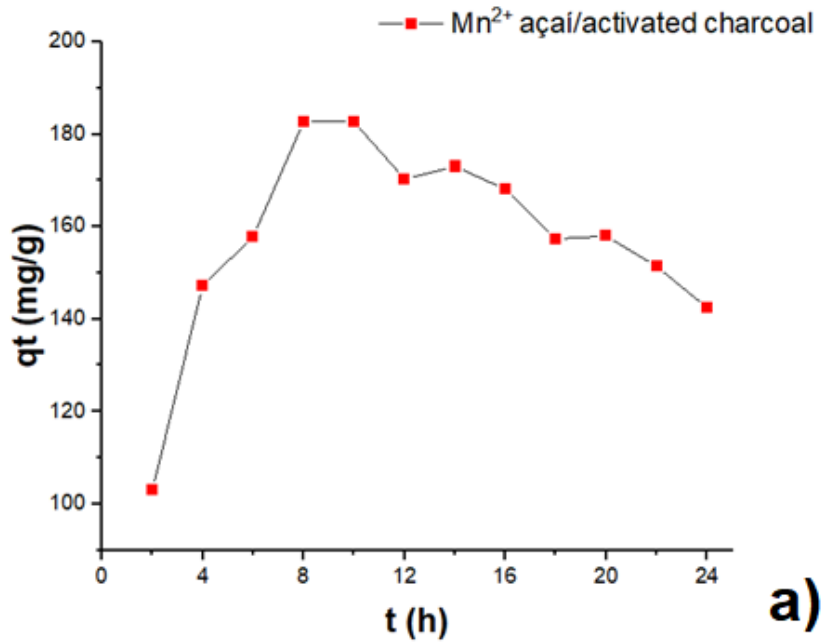
In the figures (2b to 2d) the kinetic profiles are represented according to the model's pseudo-first-order, pseudo-second-order order, intraparticle diffusion, and Elovich. The analysis of the graphs showed that manganese biosorption using activated charcoal showed that the best adjustment was for the kinetic model of pseudo-second-order (PSO), even behavior presented by fresh biomass. In the (PSO) model the biomass analyzed presented a 0.99 correlation coefficient and calculated capacity (q_e cal) of 150.16 mg kg⁻¹ (Table 2), very close to the experimental (150.62 mg kg⁻¹) This indicates that this biomass presented better adjustment to the PSO model than in natura, indicating that chemical activation did not influence the type of biosorption process (Ho, Y.S.; McKay, 1999; Nascimento *et al.*, 2021).

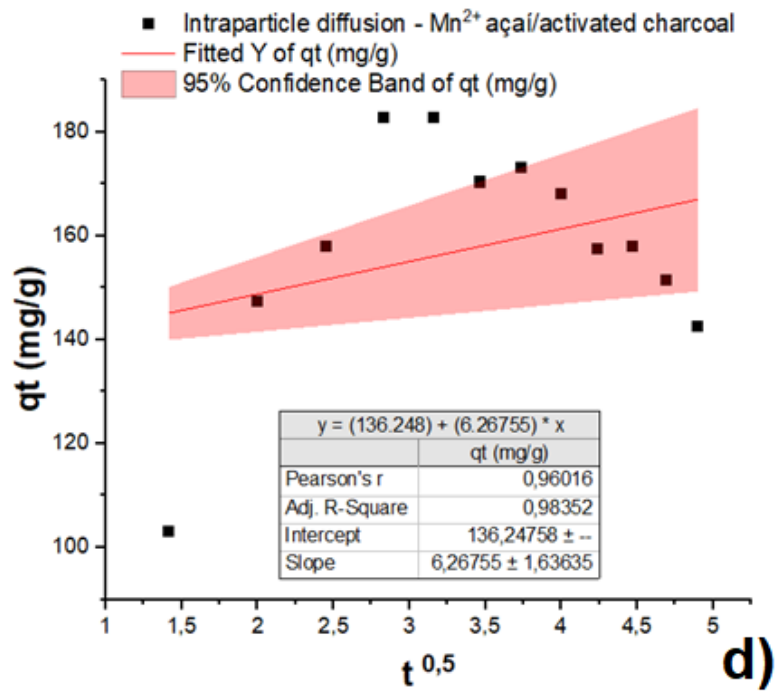
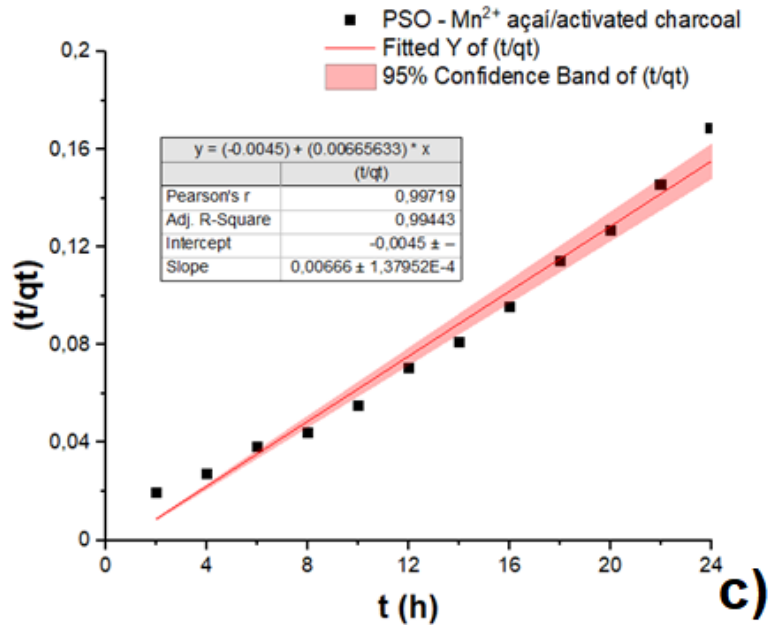
Chemical modifications in biosorbents can significantly improve biosorption capacities. Regarding kinetic modeling, biosorption processes using chemically modified biosorbents often adjust to the pseudo-second-order model (Syeda *et al.*, 2022).

In biosorption processes, kinetic studies are intended to describe reaction rate control mechanisms, such as the progress of chemical reaction processes, as well as mass transfer. Kinetic models can mathematically describe the solute binding rate on the biosorbent surface.

The most commonly used models are often those of pseudo-first-order and pseudo-second-order. Where they assume that the sorption rate is proportional to the number of free sites on the surface of the biosorbent at the proper power (first or second), (Syeda *et al.*, 2022).

Figure 2. Effect of contact time: a) on the biosorption capacity; b) Pseudo-first-order kinetic plot, c) Pseudo-second-order kinetic plot, d) Intraparticle diffusion kinetic plot, and e) Elovich kinetic plot for the biosorption of manganese ions by açai/activated charcoal.





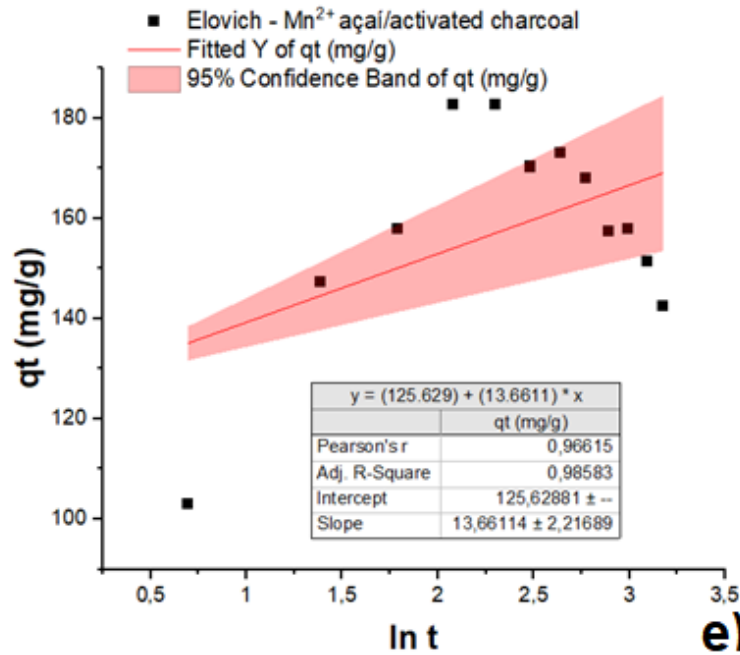


Table 2. PFO constants, PSO, intraparticle diffusion, and kinetic models of Elovich for manganese ion removal using açai in the form of charcoal.

Kinetics models	Parameters	Value
Experimental data	q_e (exp), (mg g ⁻¹)	150,62
PFO	q_e (cal), (mg g ⁻¹)	1,08
	k_1 (min ⁻¹)	0,02
	R^2	0,92
PSO	q_e (cal), (mg g ⁻¹)	150,15
	k_2 (g mg ⁻¹ min ⁻¹)	-2,00E-07
	R^2	0,99
Intraparticle diffusion	K_i (mg g ⁻¹ min ^{-0,5})	6,27
	C_i (mg g ⁻¹)	136,25
	R^2	0,98
Elovich	α (g mg ⁻¹ min ¹)	2,22
	β (g mg ⁻¹)	0,073
	R^2	0,99

CONCLUSIONS

The kinetic study of the potential of açai pit waste (*Euterpe oleracea* Mart.) evaluated in fresh form and with physicochemical modification (HCl activated charcoal) have shown that biomass is a potentially toxic metal Mn (II) promising biosorbent. Data application to kinetic biosorption models pointed to better adjustment to the pseudo-second-order model suggesting, therefore, the process of metal-biomass interaction is via chemisorption.

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