STUDY OF KINETIC AND EQUILIBRIUM MODELS IN THE ADSORPTION OF ORANGE-FLAVORED SOFT DRINK ON PAPER TOWELS: A SEMIQUANTITATIVE MODEL

ESTUDO DE MODELO CINÉTICO E DE EQUILÍBRIO NA ADSORÇÃO DE REFRIGERANTE SABOR LARANJA EM PAPEL TOALHA: UM MODELO SEMIQUANTITATIVO

ESTUDIO DE MODELOS CINÉTICOS Y DE EQUILIBRIO EN LA ADSORCIÓN DE REFRESCO SABOR NARANJA EN PAPEL TOALLA: UN MODELO SEMICUANTITATIVO

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ABSTRACT

The understanding of concepts approached from a physicochemical perspective, whether kinetic or thermodynamic, in the classroom through calculations without conceptual visualization can represent, for most students, a major abstract paradigm. Thus, the possibility of using a common process in various activities—from its application in homemade filters to wastewater treatment—is highlighted. Adsorption was explored in the present study as a semi-analytical demonstrative approach for obtaining values of kinetic parameters present in the Arrhenius equation, as well as for determining classical thermodynamic quantities. The process occurred through the adsorption of orange-flavored soft drink (considering the beverage as a dye) onto paper towels, studying adsorption in the UV–Vis region of the electromagnetic spectrum. Initially, the absorption spectrum profile was verified, and a maximum value of 481 nm was found. Based on this result, dilutions were used to construct a calibration curve, observing a linear correlation. The equilibrium time, yield, and adsorption capacity at each determined time were studied under constant agitation of 150 rpm and variable temperatures of 25 °C, 35 °C, and 45 °C. The mechanisms studied were pseudo-first-order, pseudo-second-order, and the Elovich model, enabling the determination of kinetic parameters. Experiments carried out with different initial concentrations, monitored until equilibrium, allowed for the evaluation of the Langmuir and Freundlich models, together with thermodynamic data.

Keywords: Kinetic Chemical Adsorption. Isotherms.

RESUMO

A compreensão de conceitos tratados do ponto de vista físico-químicos, seja cinético ou termodinâmico, em sala de aula por cálculos sem a visualização conceitual pode ser, para a

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maior parte dos estudantes um grande paradigma abstrato. Assim, a possibilidade de utilizar um processo comum em diversas atividades, desde seu uso em filtros caseiros como em tratamento de efluentes. A adsorção foi explorada no presente trabalho como demonstrativo semi analítico na obtenção de valores de parâmetros cinéticos presentes na equação de Arrhenius assim como a obtenção de grandezas termodinâmicas clássicas. O processo ocorreu por meio da adsorção de refrigerante sabor laranja (considerando a bebida como corante) em papel toalha, estudando a adsorção na região UV-Vis do espectro eletromagnético. Inicialmente foi verificado o perfil do espectro de absorção, sendo encontrado o valor máximo de 481 nm, com esse resultado pôde ser aplicado, a partir de diluições, a construção de uma curva de calibração observando uma correlação linear. O tempo de equilíbrio, rendimento e capacidade de adsorção em cada tempo determinado, foram estudados sob agitação constante de 150 rpm e temperaturas variáveis 25 ºC, 35 ºC, e 45 ºC. Os mecanismos estudados foram: pseudo primeira ordem, pseudo segunda ordem e modelo Elovich, possibilitando encontrar parâmetros cinéticos. Experimentos realizados com concentrações iniciais diferentes, acompanhados até o equilíbrio, proporcionando avaliações dos modelos de Langmuir e Freundlich, juntamente com os dados termodinâmicos.

Palavras-chave: Adsorção Cinética Química. Isotermas.

RESUMEN

La comprensión de conceptos abordados desde un punto de vista fisicoquímico, ya sea cinético o termodinámico, en el aula mediante cálculos sin visualización conceptual puede representar, para la mayoría de los estudiantes, un gran paradigma abstracto. De este modo, se destaca la posibilidad de utilizar un proceso común en diversas actividades, desde su uso en filtros caseros hasta el tratamiento de efluentes. La adsorción fue explorada en el presente trabajo como un enfoque demostrativo semianalítico para la obtención de valores de parámetros cinéticos presentes en la ecuación de Arrhenius, así como para la determinación de magnitudes termodinámicas clásicas. El proceso se llevó a cabo mediante la adsorción de refresco sabor naranja (considerando la bebida como un colorante) en papel toalla, estudiando la adsorción en la región UV-Vis del espectro electromagnético. Inicialmente se verificó el perfil del espectro de absorción, encontrándose un valor máximo de 481 nm. Con este resultado, fue posible aplicar, a partir de diluciones, la construcción de una curva de calibración, observándose una correlación lineal. El tiempo de equilibrio, el rendimiento y la capacidad de adsorción en cada tiempo determinado fueron estudiados bajo agitación constante de 150 rpm y temperaturas variables de 25 °C, 35 °C y 45 °C. Los mecanismos estudiados fueron: pseudo-primer orden, pseudo-segundo orden y el modelo de Elovich, lo que permitió determinar parámetros cinéticos. Experimentos realizados con diferentes concentraciones iniciales, seguidos hasta el equilibrio, posibilitaron la evaluación de los modelos de Langmuir y Freundlich, junto con los datos termodinámicos.

Palabras clave: Adsorción Cinética Química. Isotermas.

1 INTRODUCTION

Adsorption processes are of great importance in the removal of contaminants, so it has environmental and industrial interest. The process occurs with the presence of two phases: a fluid (gaseous or liquid) in which the contaminant is present, the second a solid matrix that, by physical and chemical interactions, tends to incorporate the analyte. (HILLER CONNELL; KOZAR, 2017; JORDAN; BROEGA; MARTINS, 2019; PETERS; SIMAENS, 2020) (DABROWSKI, 2001; NASCIMENTO et al., 2014; SUBHI; AL-NAJAR; NOORI, 2022)

The process is widely studied in an attempt to search for new adsorbent materials or in an attempt to remove certain ions, complexes or dyes. Therefore, the interest in the technique generates environmental relevance. Many dyes promote organoleptic evaluation in humans, especially in the visual aspect in clothing dyes and other materials, for food dyes on taste and smell. (GEETHA; PRATHEEKSHA; SUBRAHMANYA, 2020; MOSTAFA; ABD EL-HAMID; AKL, 2023; SCHMITZ et al., 2021; SHEN et al., 2018; TAN et al., 2015a) (BRITO et al., 2021; LEHMKUHLER et al., 2023)

In the textile part, there is a major risk factor related to the disposal of waste in effluents, promoting the contamination of wastewater, imbalance in the biochemical oxygen demand (BOD), reducing the passage of light for the photosynthesis of primary beings in the aquatic food chain, contamination of groundwater and contamination of fish and waters with possible human use. (HILLER CONNELL; KOZAR, 2017; JORDAN; BROEGA; MARTINS, 2019; PETERS; SIMAENS, 2020)

In food, the dyes, although they have pleasant flavors and odors, are subject to possible health disorders such as allergies, hyperactivity, food poisoning, asthma, bronchitis, rhinitis, eczema and others. A color of great prominence is Tartrazine, one of those responsible for the orange color of foods such as carbonated drinks, cakes, powdered juices, energy drinks, mustard, colored breakfast cereals, meat and chicken broths and other wide variety of foods. (LEHMKUHLER et al., 2023) (KOMISSARCHIK; NYANIKOVA, 2014)

Adsorption has some characteristics and perspectives of obtaining kinetic and thermodynamic properties. The reduction in the concentration of dyes, from the laboratory aspect, is monitored by spectroscopy in the visible region of the electromagnetic spectrum (340-720nm). (LEVINE, 2014) (AHMADI; GHASEMPOUR; HASANZADEH, 2023; NASCIMENTO et al., 2014)

Seeking to promote the understanding of the adsorption process as a whole, the present work seeks to perform in a semiquantitative way the interpretation of the concepts of adsorption (equilibrium, kinetic and thermodynamic), using orange flavored soft drinks on paper towels as an absorptive model.

1.1 FOOD COLOURS

Substances from natural or synthetic sources that have pigments, being added to foods in small quantities, with or without nutritional value, exerting modifications in organoleptic properties capable of altering or not the state of physicochemical properties of industrial foods. (BRITO et al., 2021)

Artifices can cause damage to health, and their recommendation and use are practically supported by technological and commercial issues. Usually the visual aspect is highly accepted, in addition to promoting superior stability and uniformity when compared to the natural ones. However, they can still hide the low content of certain nutrients in industrial foods , (2023) (SANTOS, A. A. dos; CALIXTO, S. J. A. de O.; NEUMANN, K. R. da S.

In Brazil there are about eleven artificial dyes allowed, when compared to international legislation, the difference between the acceptance of dyes is noticeable, the American legislation has greater rigor in relation to the permission of the use of food dyes, as reported in Table 1 adapted from . (BRITO et al., 2021)

Table 1

List of food colourings permitted or not permitted by FDA and EFSA

Dye	FDA Permission	EFSA Permission
Tartrazine	Yes	Yes
Twilight Yellow	Yes	Yes
Azorrubin	No	Yes
Amaranth	No	Yes
Ponceau 4R	No	Yes
Erythrosine	Yes	Yes
Red 40	Yes	Yes
Blue Patent V	No	Yes
Indigotine Blue	Yes	Yes
Bright Blue	Yes	Yes
Fast green	Yes	No

Source: Brito et all., 2021 adapted.

From the legal aspect, there are regulatory bodies in several, the main ones being the Food and Drug Administration (FDA) in the United States; the European Food Safety (EFSA) in the European Union, in Brazil the National Health Surveillance Agency (ANVISA) is responsible. (BRITO et al., 2021)

1.2 ABSORPTION

Adsorptive processes are low cost, justifying their wide use in industrial processes. During the process, the analyte (adsorbent) interacts with the matrix (adsorbent). The matrix has superficial imperfections (active sites), sites with the ability to anchor the adsorbate. Whether through weak (physisorption) or strong (chemisorption) interactions, the process occurs until the saturation of the porous solid. (DABROWSKI, 2001; NASCIMENTO et al., 2014)

In general, the reaction can be described by Equation 1, occurring in three stages:

$$A_{(aq)} + B_{(s)} \rightarrow (AB)_{(s)} \qquad (1)$$

The steps described below are illustrated in Table 2.

Initially: there is an unwanted agent dissolved in the fluid (), the adsorbate with a known mass dissolved in a certain volume, then there is the initial concentration of the dye, which will be put in contact with the adsorbent ().$A_{aq}B_s$

Duration of the reaction: Part of the dye mass is transferred to the adsorbent providing a reduction in the concentration of the adsorbate in the solution and an increase in the adsorption ratio (.AB_s)

Table 2

Demonstrative of chemical equilibrium

$\boldsymbol{A_{(aq)} + B_{(s)} \rightarrow (AB)_{(s)}}$			
	Reagents		Products
Components	$\boldsymbol{A_{(aq)}}$	$\boldsymbol{B_{(s)}}$	$\boldsymbol{(AB)_{(s)}}$
Stage 1 t0	$C_0 = {}^{m_{corante}}/_{Volume_{solução}}$	$m_{adsorvente}$	--------------------
Step 2 tx	$-C$	Does not change	+m_{ads}
Step 3 Balance	$C_0 - C$	Does not change	$m_0 + m_{ads}$

Source: The author.

Equilibrium is reached, we have the analyte present in the two parts, the concentration in solution and the adsorbed mass, called the amount adsorbed per adsorbent mass (q), for each time of the process a qt value can be assigned. By looking at the table, it is possible to deduce the relationship represented by equation 2.

$$q_t = (C_0 - C)\frac{Volume_{solução}}{m_{adsorvente}} \qquad (2)$$

In order to find the percentage efficiency of the removal process (R), we have equation 3.

$$R = \frac{100(C_0 - C)}{C_0} \qquad (3)$$

1.3 ADSORPTION KINETICS

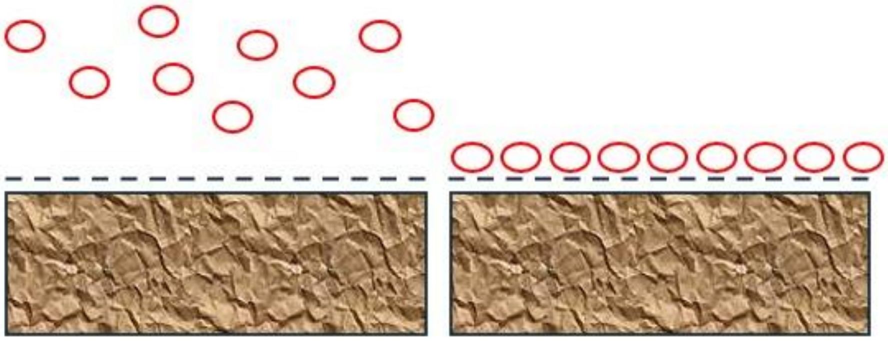
Every reaction that reaches equilibrium has a forward velocity equal to the inverse velocity occurring simultaneously. (HOUSECROFT; SHARPE, 2008; LEVINE, 2014; SKOOG; HOLLER; CROUCH, 2006)

To this end, the kinetics of adsorption is related to the speed of the diffusion transport of the substance and its movement. In addition, in adsorption kinetics there is the transport of the adsorbate and its binding to available sites of the adsorbent until it reaches equilibrium. This process describes the relationship of the interaction that occurs between the adsorbent and the adsorbate, which can be influenced by some factors such as: temperature, pH, concentration and property of the substance. Thus, the most common kinetic models are: pseudo-first-order, pseudo-second-order, and intraparticle diffusion.

Thus, as shown in Figure 1, which demonstrates the adsorption process. We have that, in this case, the molecules of atoms (red dots, Figure 1) of a certain substance (adsorbate) adhere to the surface of another substance that is called adsorbent.

Figure 1

Description of the adsorption process



Source: The author, 2024.

With the monitoring of adsorption at various times, it is possible to find the time when the concentration in solution no longer varies, the so-called equilibrium time. The first important factor to be determined is the order of reaction, a mathematical model in the quantitative aspect (capable of relating the speed and concentration of the reactants), qualitatively (indicates the kinetic reaction mechanism, forms of contact, presence of active sites, weak or strong interactions). (NASCIMENTO et al., 2014) (TAN et al., 2015b)

1.3.1 Pseudo-first-order kinetic mechanism

Qualitatively, it describes physical adsorptions, with the adsorption rate being proportional to the amount of active sites present in the adsorbent. Quantitatively, the model is represented by equation 4. (CHAIRAT et al., 2008; NASCIMENTO et al., 2014)

$$q_t = q_e(1 - e^{-k_1 t}) \qquad (4)$$

q_t represents the amount of adsorbate adsorbed at time t

q_e represents the amount of adsorbate adsorbed at equilibrium

k_1 represents the adsorption rate of the pseudo first-order model

1.3.2 Pseudo-second-order kinetic mechanism

Qualitatively it is related to chemical adsorptions between the solid matrix and analyte, in the process the adsorption rate dependent on the amount of solute superficially adsorbed on the adsorbent and how much can be adsorbed at equilibrium, quantitatively it is represented by equation 5. (CHAIRAT et al., 2008; NASCIMENTO et al., 2014)

$$q_t = q_e \frac{k_2 q_e t}{1+k_2 q_e t} \qquad (5)$$

q_t represents the amount of adsorbate adsorbed at time t

q_e represents the amount of adsorbate adsorbed at equilibrium

k_2 represents the adsorption velocity of the pseudo second-order model

1.3.3 Elovich kinetic model

The model tends to describe processes in which heterogeneous adsorbents are used. The matrix presents sites with different energies, also considers that the interactions between adsorbed particles and the desorption process do not significantly affect the adsorption kinetics. Equation 6 represents the model. (DABROWSKI, 2001; NASCIMENTO et al., 2014)

$$q_t = \frac{1}{\beta} \ln (\alpha \beta t) \qquad (6)$$

q_t represents the amount of adsorbate adsorbed at time t

α indicates the adsorption rate at the beginning of the process

β desorption constant

Some factors affect the kinetic relationship of processes, including the concentration of reagents, surface area of contact, temperature, pressure and use of catalyst. (HOUSECROFT; SHARPE, 2008; LEVINE, 2014)

In an attempt to understand such factors, the monitoring of the reaction at different temperatures allows the application of equation 7, developed by Arrhenius, making it possible to find two kinetic parameters. (LEVINE, 2014)

Pre-exponential factor (A): related to the effective shocks of the process, collision frequency.

Activation energy: energy required for the reaction to occur adsorption

$$k = Ae^{-E_a/_{RT}} \qquad (7)$$

R indicates the gas constant

T the temperature at which the process took place in Kelvin

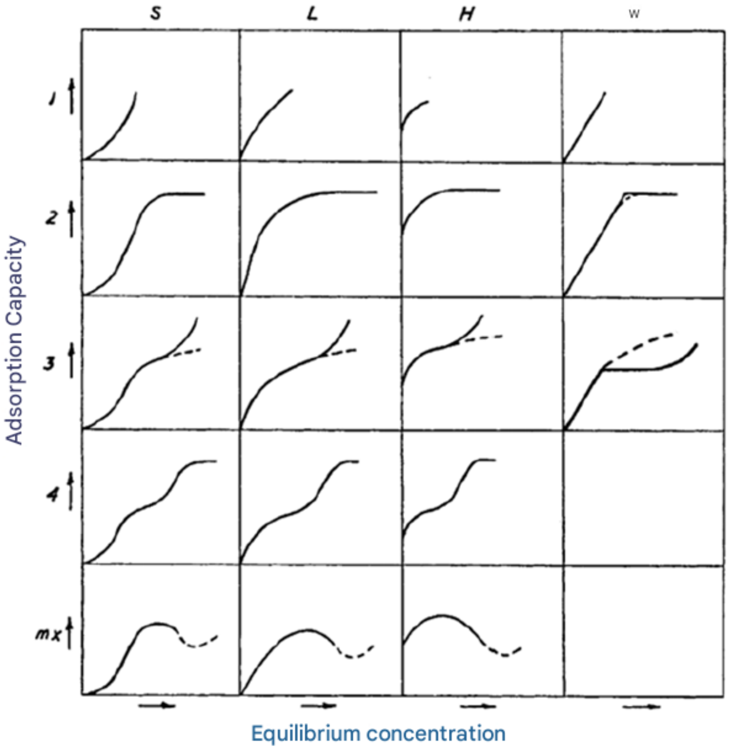
1.4 ADSORPTION ISOTHERMS

Isotherms are experiments carried out at constant temperature, in the case of the adsorptive process, they indicate the relationship between the amount of adsorbate retained in the matrix and the concentration remaining in the solution. The isotherms indicate information that can help to elucidate the adsorption mechanism (unlike the reaction mechanism predicted by the adsorption kinetics that aimed to relate the mechanism related to the speed of the process), it allows to predict the retention capacity of different adsorbents, the mass of pollutant that will be retained. Such information makes it possible to compare adsorption systems either by observing the matrix or the contaminant.

In processes involving solid-liquid adsorption, classified isotherms into four types as shown in Figure 2. (GILES et al., 1960)

Figure 2

Classical classification of isotherms



Source: Giles, 1960.

The presence of four columns (the types) and five rows indicating subclasses is observed.

Adsorptions that assume the S type:

They represent systems where:

The contaminant molecules are monofunctional.

The interactions between adsorbate and adsorbent must be intermolecular attraction, allowing vertical and regular interactions over the entire surface.

Possess competition for active sites between solvent and analyte molecules

For type L:

Adsorption occurs horizontally in layers.

The competition for the active sites of the matrix is less intense than the S-type isotherm.

Type H, similar to type L

Cases in which there is a high affinity between the matrix and the analyte, since in diluted solutions the adsorption occurs with a yield close to 100%, leaving in the fluid phase practically undetectable tractions.

Type C, processes in which there is a partition coefficient

Regions with different degrees of crystallinity promote the flexibility of the adsorbed species.

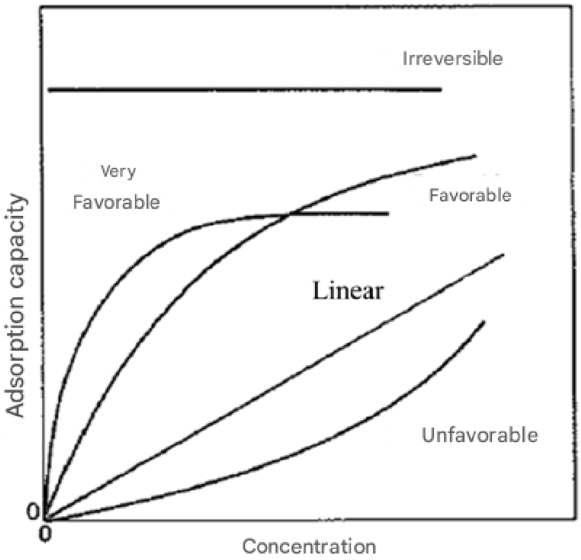
The interaction with the solute is greater than the interaction as an analyte.

The molecular geometry of the adsorbent can promote flexibility by altering the contact of the active sites with the adsorbate.

A simpler way of representing solid-liquid isotherms has been presented as shown in Figure 3. (MCCABE; SMITH; HARRIOTT, 1993)

Figure 3

Classification of isotherms



Source: Mccabe; Smith; Harriott, 1993.

The main issue represented in the classification is the behavior that exists, favorable adsorptions, whether very or only favorable, show an exponential growth with the region of maximum differentiating at the beginning of the cure, very favorable has a high capacity in low concentration and in the displacement of the maximum point. The unfavorable one also has an exponential growth, but it has a region of minimum, we also have the linear and the irreversible.

1.5 ISOTHERM MODELS

1.5.1 Langmuir

It considers uniform distribution of the adsorbate on the surface with monolayer formation, consequently the surface of the matrix must be energetically homogeneous, adsorbed particles do not interact with each other. Its expression is represented by equation 8. (AL-GHOUTI; DA'ANA, 2020; NASCIMENTO et al., 2014)

$$q = \frac{q_{max} K_L C_e}{1 + K_L C_e} \qquad (8)$$

q Amount of Adsorbed Solute per Unit Mass of Adsorbent

q_{max} Maximum adsorption capacity

K_L Adsorbate/adsorbent interaction constant

C_e Concentration of the adsorbate in solution

1.5.2 Freundlich

It can be adopted for models that have multilayers and heterogeneous characteristics, consequently the active sites are of different energies, causing different interactions between the adsorbent and the analyte. The model is interpreted with the help of equation 9. (AL-GHOUTI; DA'ANA, 2020; NASCIMENTO et al., 2014)

$$q = K_F (C_e)^{1/n} \qquad (9)$$

q Amount of Adsorbed Solute per Unit Mass of Adsorbent

K_F Freundlich adsorption capacity constant

C_e Equilibrium constant in solution

$1/n$ Constant related to system homogeneity

By monitoring the isotherms at different temperatures, it is possible to find the main thermodynamic factors, (thermodynamic constant). Using the fundamental equations of thermodynamics, equations 10, 11 and 12. .$\Delta H \Delta S \Delta G K$ (LEVINE, 2014; LIU; LEE, 2014; NOREEN et al., 2021; SHAHWAN, 2021)

$$\Delta G = -RT lnK \qquad (10)$$

$$\Delta G = \Delta H - \mathrm{T}\Delta S \qquad (11)$$

$$lnK = -\frac{\Delta H}{RT} + \frac{\Delta S}{R} \qquad (12)$$

2 OBJECTIVES

2.1 GENERAL OBJECTIVE

- To evaluate the adsorption of orange-flavored soda on paper as a model for future adsorption studies.

2.2 SPECIFIC OBJECTIVES

- Study the adsorption equilibrium by means of process throughput.
- Apply and compare models of reaction mechanisms.
- Calculate kinetic factors.
- Perform adsorption isotherm studies.
- Compare isotherm models.
- Estimate thermodynamic parameters.

3 MATERIALS AND METHODS

The experiments were carried out in the didactic laboratories of the UEMG unit on the campus of Ituiutaba, with the availability of a shaker table with temperature control and UV-Vis spectrophotometer.

- 8.3L Refrigerated Benchtop Shaker Incubator, Model IST-4075R
- UV-Vis Spectrophotometer Thermo Scientific Evolution One Series

The soft drink to be studied and the paper towels were purchased in the regional trade.

Initially, to perform the tests, the refrigerant gas was removed by means of a magnetic stirrer at room temperature for approximately 45 minutes.

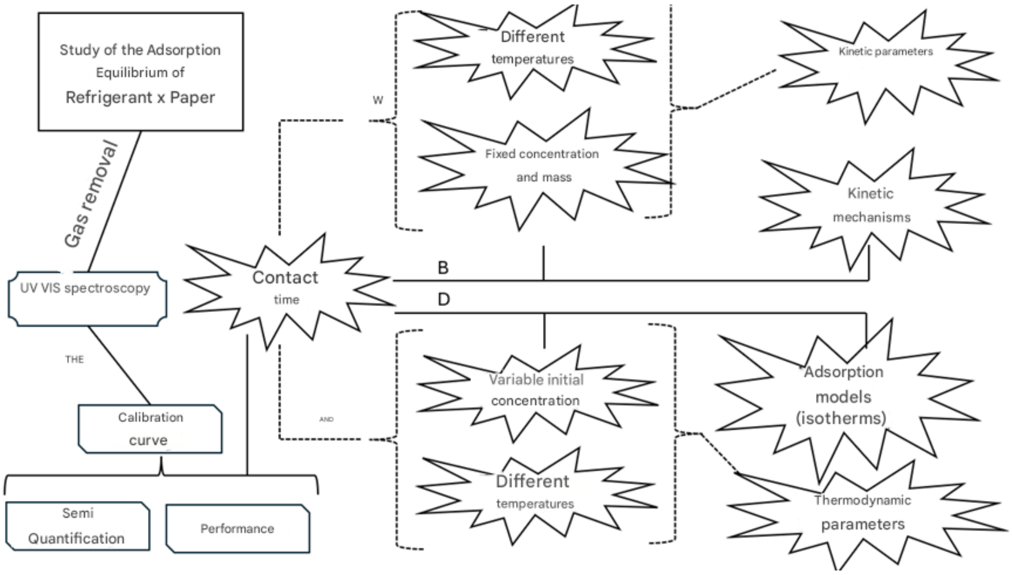
The measurements performed in the spectrophotometer were scans in the range between 400-750nm with velocity rate and 1nm/s.

In the tests proposed in this work, all soft drinks were considered as a dye without distinction of their composition, considering that the present work is a semi-quantitative model. The working solutions were prepared in distilled water considering the mass of refrigerant and not its volume, obtaining concentrations in $g_{refrigerante}/L_{solução}$.

The methodology consisted of 5 stages, named from "A" to "E", described below and represented in a didactic way in Figure 4.

Figure 4

Experimental Script



Source: The author.

In order to find a way to quantify the concentration of the refrigerant in solution, an initial spectrum was performed in a solution with a concentration of 391g/L, later for the highest value of maximum wavelength dilutions were read in the same The dilutions of the initial solution were prepared for the following $(\lambda_{max})\lambda_{max}$.351.9; 312.8; 293.25; 234.6; 195.5; 97.75; 39.1 g/L, thus, it was possible to obtain the calibration curve (step A).

In the following tests, the concentration of the initial solution () was 391g/L, with aliquots of 100mL and stirring speed of 150rpm. C_0

For the study of the adsorption time, the variation of the concentration of the solution by UV-Vis spectroscopy at a controlled temperature of 25ºC was studied at intervals of 2; 5; 7; 10; 30; 60; 90 minutes, with paper towel dough (adsorbent) of approximately 0.600g. Obtaining the yield and a semi-quantitative analysis of the adsorptive process and mechanism (end of step A). In addition to the kinetic mechanism at this temperature (step B).

To study the kinetic parameters, studies of adsorption time variation in the previous conditions were carried out for two other temperatures, 35º, 45ºC. Allowing to analyze the kinetic mechanism (step B), and from the mechanism find the kinetic parameters (step C).

In order to study adsorption models, an experiment was carried out with a controlled temperature of 25ºC, fixed mass of adsorbent 600mg, varying the initial concentration of the contact solution (391, 351.9, 312.8, 293.25, 234.6, 195.5, 97.75, 39.1 g/L), following the concentration of the solution at the end of the 90 minutes. The obtaining of adsorption isotherms or adsorption models was possible (step D), repeating the experiment for two other temperatures (35ºC and 45ºC), the three isotherms obtained generate the studies of the thermodynamic variables (step E).

4 RESULTS AND DISCUSSIONS

4.1 STEP A

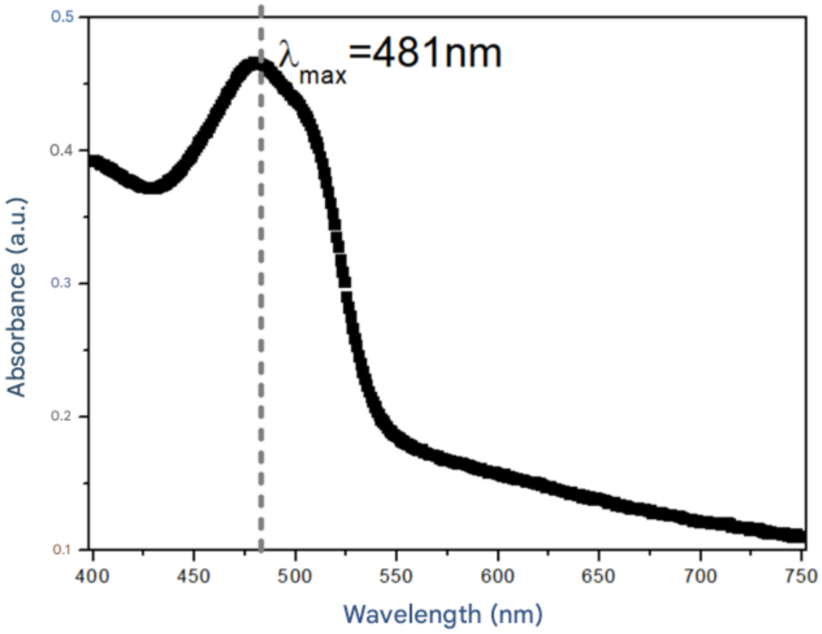
The results obtained enabled the initial qualitative study of the analyte under study, obtaining an analytical quantification curve that is fundamental in the rest of the study.

4.1.1 Evaluation of soft drinks in the uv-vis region

The initial evaluation was performed with a scan in the region between 400-750nm and dilution of 1.5mL/3mL in the quartz cuvette, in order to know the behavior of the refrigerant in the visible region of the spectrum, as represented by Figure 5. A broadband between 425 and 555nm was found with a maximum absorbance of 481nm. The presence of this broadband may indicate various electronic transitions in the region or electronic relocation. In carbonated beverages there is not a single type of dye, probably a mixture of at least two main dyes: twilight yellow (food yellow 3, yellow 5, sunset yellow) and tartrazine, in the literature, several studies with food dyes are carried out in the region between 426nm and 430nm suggesting that the value found is reasonable for the mixture of dyes, which have maximum absorption respectively at 480nm and 425nm, with the possibility of overlapping bands, as indicated by . (SKOOG; HOLLER; CROUCH, 2006) λ_{max} (DOTTO et al., 2011; SAHNOUN; BOUTAHALA, 2018) *DOS SANTOS; DEMIATE; NAGATA, 2010*

Figure 5

Physical characterization by spectroscopy in the UV-Vis region of the analyte



Source: The author.

4.1.2 Calibration Curve

With the knowledge of the behavior of the refrigerant in the visible region of the spectrum, it is possible to make a calibration curve, considering the absorbances in the value of the maximum wavelength, the values are present in Table 3. With the results obtained, it is possible to plot the calibration curve, represented in Figure 6.

Table 3

Relationship between concentration and absorbance for curve construction of calibration

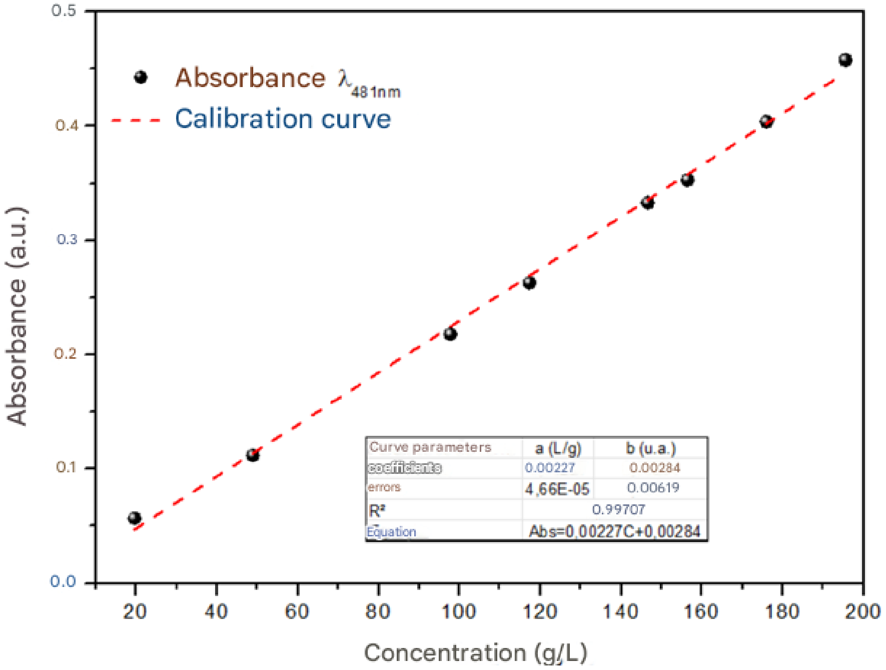
Concentration	Absorbance (481nm)
195,5	0,458
175,95	0,404
156,4	0,353
146,625	0,333
117,3	0,263
97,75	0,218
48,875	0,112
19,55	0,057

Source: The author.

The values of the angular and linear coefficients are 0.00227L/g and 0.00284, with an adjustment coefficient R2 of 0.99707, a value close to those found by DOTTO et al., 2011 , for calibration curves of twilight yellow dyes and tartrazine at concentrations of 100mg/L.

Figure 6

Refrigerant solution calibration curve



Source: The author.

The equation that correlates the absorbance values obtained with the concentration is represented by the expression: $C = (Abs - 0{,}00284)/0{,}0227$, used for the following determinations of the study.

4.1.3 Study of equilibrium time and performance

The adsorption profile of the refrigerant solution, studied at three temperatures (25ºC; 35ºC; 45ºC), to verify the time it takes for the system to reach equilibrium. Under the following experimental conditions: solution volume () 0.1L; initial concentration () 391g/L; adsorbent mass () 0.600g; 150rpm rotation. VC_0m

The monitoring of the concentration reduction was performed by reading in a spectrophotometer with a wavelength of 481nm. The monitoring is described in Table 4.

Table 4

Monitoring of adsorption over time

Temperature	Time (minutes)	Concentration (g/L)	Yield (%)	Adsorbed Grease

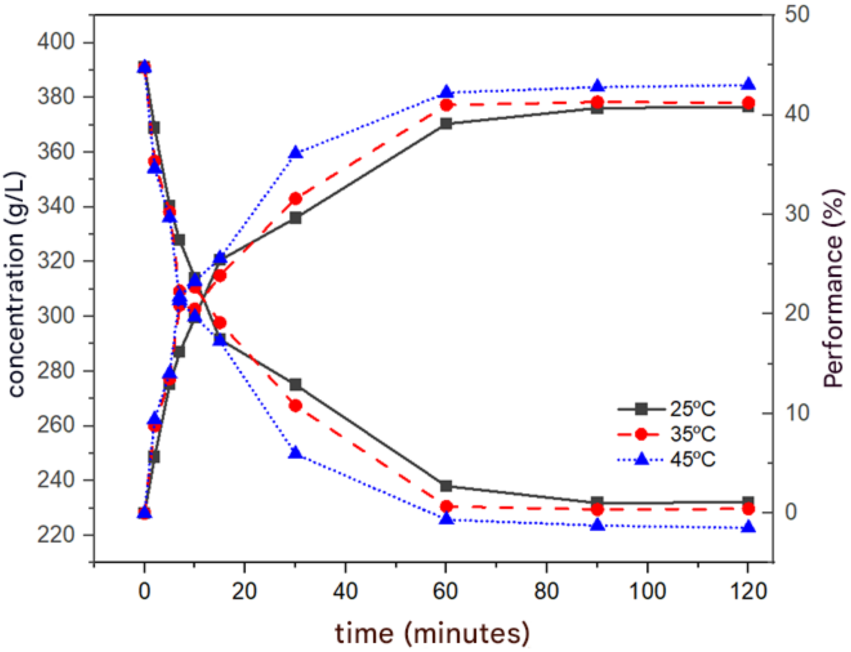
				(Grefrigerante/Gadsorbent)
25ºC	2	368,94	5,64	3,68
	5	340,51	12,91	8,42
	7	327,82	16,16	10,53
	10	314,20	19,64	12,80
	15	291,65	25,41	16,56
	30	275,08	29,64	19,32
	60	238,07	39,11	25,48
	90	231,90	40,69	26,51
	120	232,21	40,80	26,58
	2	356,72	8,77	5,71
35ºC	5	338,16	13,51	8,81
	7	309,27	20,90	13,62
	10	310,79	20,51	13,37
	15	297,75	23,85	15,54
	30	267,44	31,60	20,59
	60	230,67	41,00	26,72
	90	229,50	41,30	26,91
	120	229,82	41,22	26,86
	2	354,13	9,43	6,14
	5	336,16	14,03	9,14
45ºC	7	305,98	21,75	14,17
	10	299,87	23,31	15,19
	15	290,94	25,59	16,68
	30	249,82	36,11	23,53
	60	225,86	42,24	27,52
	90	223,62	42,80	27,88
	120	22,87	43,01	28,02

Source: The author.

The yield and adsorbed concentration values were obtained by applying Equations 2 and 3. For all temperatures, the monitored time intervals between 2 minutes and 120, finding the equilibrium time, in 90 minutes. Time in which the variation in removal or adsorbed concentration tends to no longer vary significantly, as shown in Table 4. It is possible to plot the equilibrium path, the relationship of the reduction of the concentration of reactants, together with the yield Figure 7.

Figure 7

Evaluation of the reduction in the concentration of the soft drink in the solution and evaluation of the percentage of removal



Source: The author.

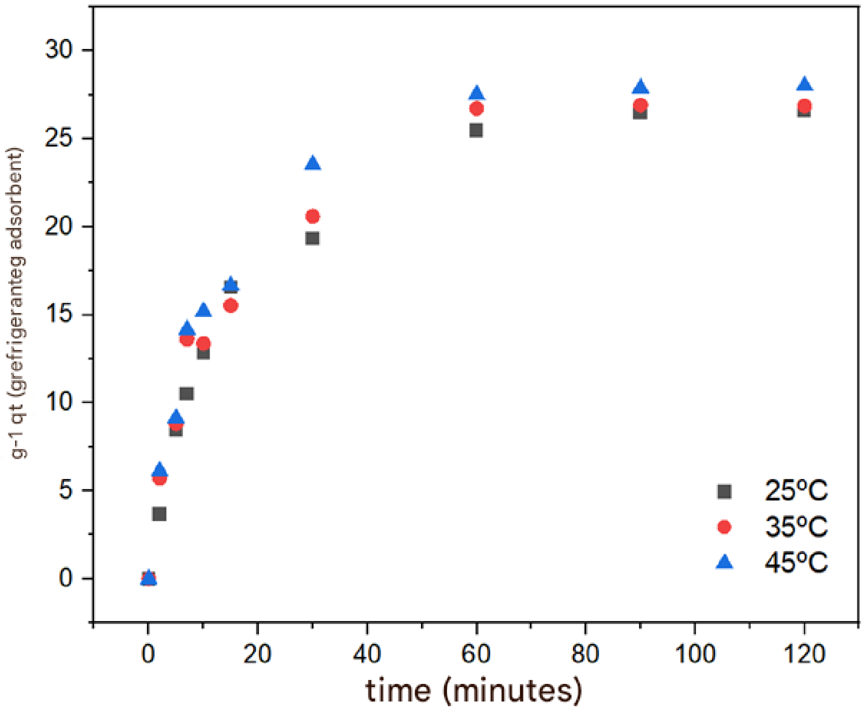
The decrease in the concentration of the refrigerant in solution occurs as expected, since the adsorption process occurs, having at all times analyzed the greatest reduction to the temperature of 45ºC, since higher temperatures promote greater molecular agitation, favoring molecular shocks and consequently greater speed. and the highest yield occurs for the experiment carried out at a temperature of 45ºC.

As for the yield, in the order of 40%, the studied process presents similarity to yields described for other adsorbents for removal of similar dyes. Studying the adsorption of Tartrazine, they found for 90 minutes of adsorption at pH 7 and temperature of 23ºC, values between 22 and 34% of removal, using eggshell, kaolin (kaolin), zeolite (zeolite) and rice husk (rice husk). While KHADER; MOHAMMED; ALBAYATI, 2021 SAHNOUN; BOUTAHALA, 2018 , studying chitosan modified with polyaniline obtained a yield in the order of 40% of tartrazine removal under the conditions of pH 7.2; initial concentration of 25mg/L; initial volume 25mL; 10mg of adsorbent at 25ºC. Removal values very close to those reported.

Figure 8 shows the relationship between the adsorbed mass and the time, with the profiles having exponential growths, because the mass retained by the adsorbent increases over the contact time, it is possible to state that the equilibrium time for the adsorption process occurs from 90 minutes onwards. Time also reported for other adsorbent in the removal of tartrazine (KHADER; MOHAMMED; ALBAYATI, 2021) .

Figure 8

Adsorption kinetics of soft drink on paper towels



Source: The author.

4.2 STEP B

The studies of kinetic mechanisms represent the type of interaction between the solid phase and the liquid phase, three models were studied, from the result of the models it is possible to find the kinetic parameters of the process.

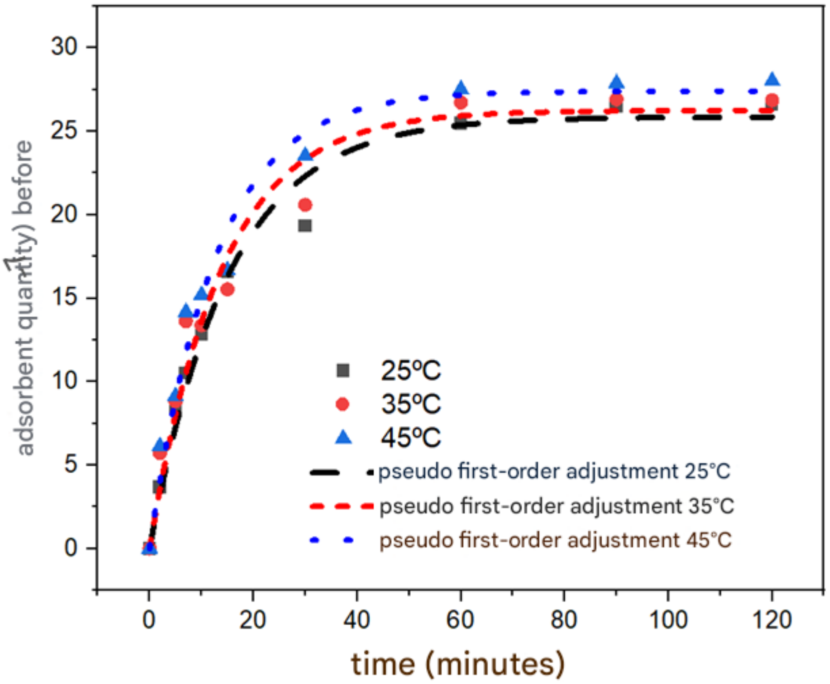
4.2.1 Pseudo-first-order mechanism

Evaluating the possibility that the active sites are proportional to the adsorption rate, the experimental data of the adsorption capacity at the three temperatures studied were adjusted to the model of Equation 4, represented by Figure 9.

The adjustments are very close to the experimental points, with the time of 30 minutes for the three temperatures having the greatest visual deviation, the highest adsorption of the dye at the temperature of 45ºC.

Figure 9

Tuning to the pseudo-first-order kinetic mechanism



Source: The author.

Table 5 verifies the parameters and for the pseudo first-order mechanism.$q_e k_1$

Table 5

Pseudo-first-order engine parameters

Pseudo 1st order	q_e	k_1	**R²**
25ºC	25,856	0,06624	0,9827
35ºC	26,254	0,07311	0,9613
45ºC	27,409	0,07962	0,9755

Source: The author.

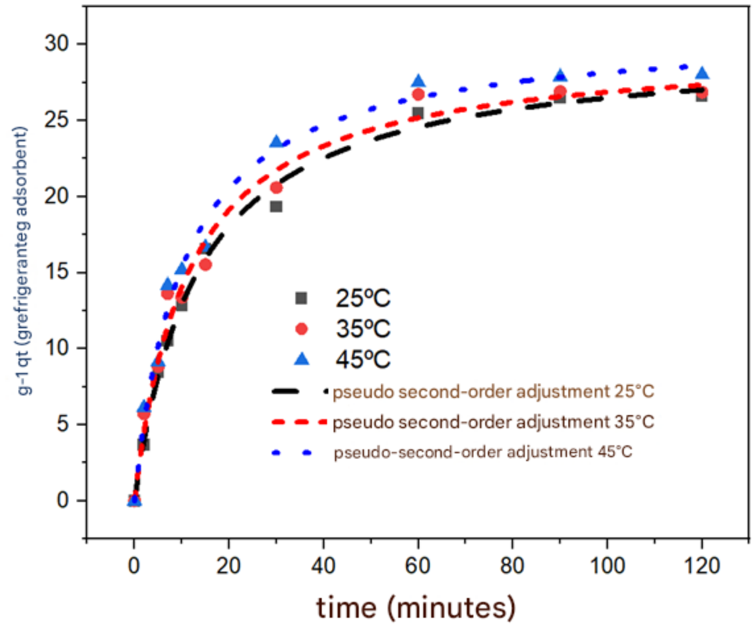
The values found indicate a good fit to the mechanism, compared to other studies, according to a review published by . The values of very divergent as values are slightly divergent from other studies in the literature for dyes similar to other adsorbents AMAKU et al., 2024 $q_e k$ (DAI et al., 2021; LIMA et al., 2019; RECK et al., 2018; YU et al., 2016) .

4.2.2 Mechanism of pseudo-followed order

With the possibility that adsorption can occur both on the surface electrostatically (physical process) and through chemical interaction, the pseudo-second-order model was studied, using Equation 5, as shown in Figure 10.

Figure 10

Tuning to the pseudo-second-order kinetic mechanism



Source: The author.

As with the pseudo-first-order mechanism, the adjustments to the pseudo-second-order mechanism are in accordance with the experimental data.

Table 6 presents the values of the constants for the pseudo-second-order mechanism.

Table 6

Pseudo-second-order engine parameters

Pseudo 2nd order	q_e	k_2	**R²**
25ºC	30,002	0,00222	0,994
35ºC	29,897	0,00279	0,982
45ºC	31,040	0,00325	0,981

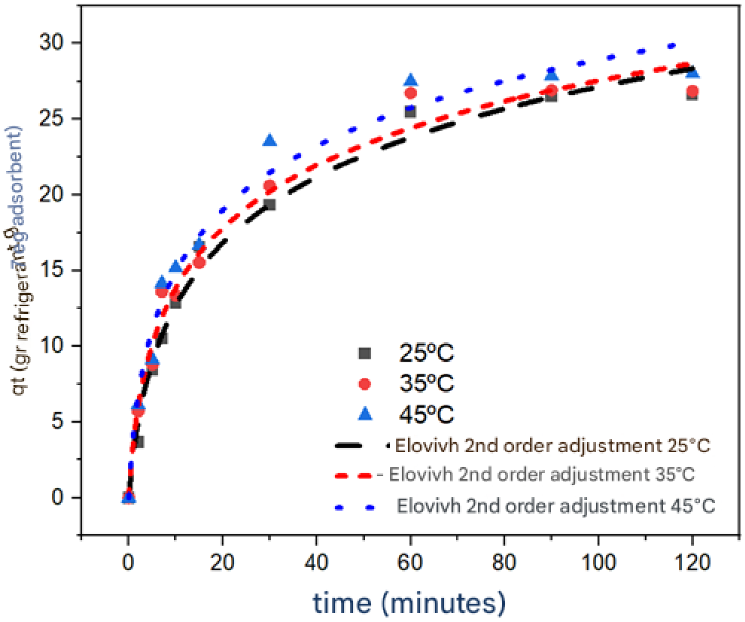
The R² values are higher than those observed for the pseudo-first-order mechanism, so the pseudo-second-order mechanism should be the preferred one for the process studied, a fact that is in agreement with other studies in the literature on works for Tartrazine adsorption and yellow 03. The parameters of and , present values with divergence and approximation to other studies, respectively. (DAI et al., 2021; LIMA et al., 2019) (YU et al., 2016) $q_e k$ (AMAKU et al., 2024)

4.2.3 Elovich mechanism

The last mechanism to be compared with the experimental data was Elovich's, which indicates a greater predominance of chemical interactions. The comparison is present in Figure 11, and the adjustments follow Equation 6, the results are presented.

Figure 11

Adjustment to Elovich's kinetic mechanism



Source: The author.

The mechanism visually is the one with the least adjustment, being confirmed by the values of R² in Table 7. No studies were found in the literature presenting this mechanism for the adsorption of dyes similar to those that may compose the orange-flavored soft drink.

Table 7

Kinetic parameters of the Elovich model

Elovich model	**α**	**β**	**R²**
25ºC	3,795	0,1493	0,985
35ºC	5,112	0,1560	0,980
45ºC	6,067	0,1614	0,977

Source: The author.

Regarding kinetics, it is possible to infer that the pseudo-second-order model is the most favorable, as reported in the literature. While the pseudo-first-order model is also reported in the literature with values slightly lower than the pseudo-second-order model, as verified in the study. On the other hand, the Elovich model, although it presents good similarity (although inferior to the others), has practically no reports in the literature.

q_eValues of the adsorption constants do not have similar values in the literature, most likely due to the way the adsorbent was studied, solid sheet of paper in the present work and granules or smaller pieces in practically all the studies verified.

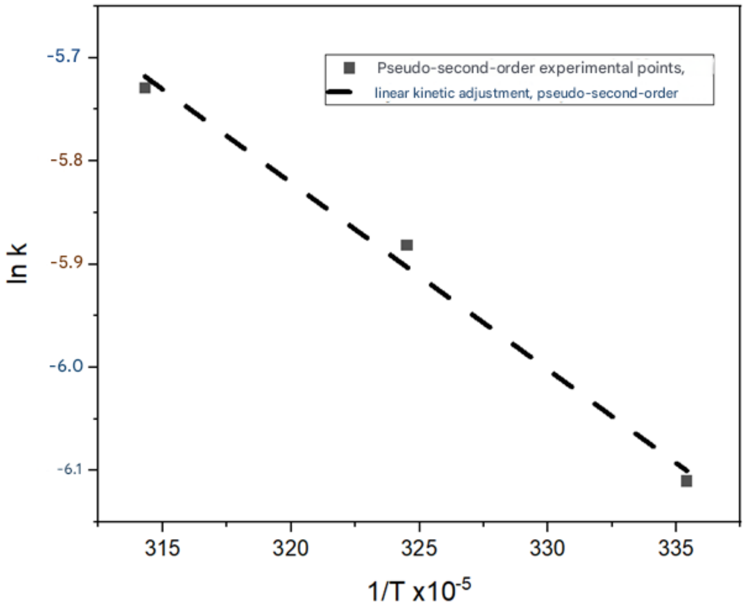
Values of the velocity constants are not so far apart because the process has yields compatible with those of other works.

4.3 PART C

From the three adsorption curves and the values of k or ,$\beta lnk\ x\ ^1/_T$ described by the mechanisms, the correlation was constructed, making it possible to evaluate the kinetic parameters described in the Arrhenius equation. The representation of the correlation information as well as their linear adjustments are described in Figure 12 in Table 8.

Figure 12

Relationship between ln x 1/T



Source: The author.

The relation describes a decreasing line, indicating that the increase in temperature the value of the velocity constant also increases, the correlation adjustment R² was 0.981, indicating a good fit of the relation with its linearization.

Table 8

Kinetic parameters for the pseudo-second-order mechanism

K	lnk	T	1/TX10-5	lnA	A	EA/R	Ea	R²
0,00222	-6,110	298,15	335,4	0,975	0,975	1810,97	217,809	0,981
0,00279	-5,882	308,15	324,5					
0,00325	-5,729	318,15	314,3					

Source: The author.

4.4 PART D

Seeking to know the adsorption mechanisms, experiments with solutions of variable initial concentrations of soft drink were placed in contact with paper towels (0.600g mass) under 150rpm agitation at controlled temperature, for 90 minutes, allowing to calculate the amounts of dye adsorbed at equilibrium and the amount present in solution using Equation 2. The results are presented in Table 9.

Table 9

Adsorption Balance

Temperature	Initial concentration	Ce (90 minutes)	Qe (90 minutes)
25ºC	391	184,54537	36,75667
	351,9	153,68811	35,57248
	312,8	115,80852	33,81004
	293,25	103,41028	33,29054
	234,6	53,22878	30,61118
	195,5	30,97797	27,85204
35ºC	391	178,20264	34,77629
	351,9	151,02496	33,46802
	312,8	122,59618	32,1725
	293,25	107,67695	31,01154
	234,6	64,32893	28,04201
	195,5	47,41263	24,3085
45ºC	391	223,18062	25,90601
	351,9	197,30984	25,85985
	312,8	172,28781	23,64331
	293,25	160,54038	21,94273
	234,6	120,90455	18,68761
	195,5	89,70455	14,79271

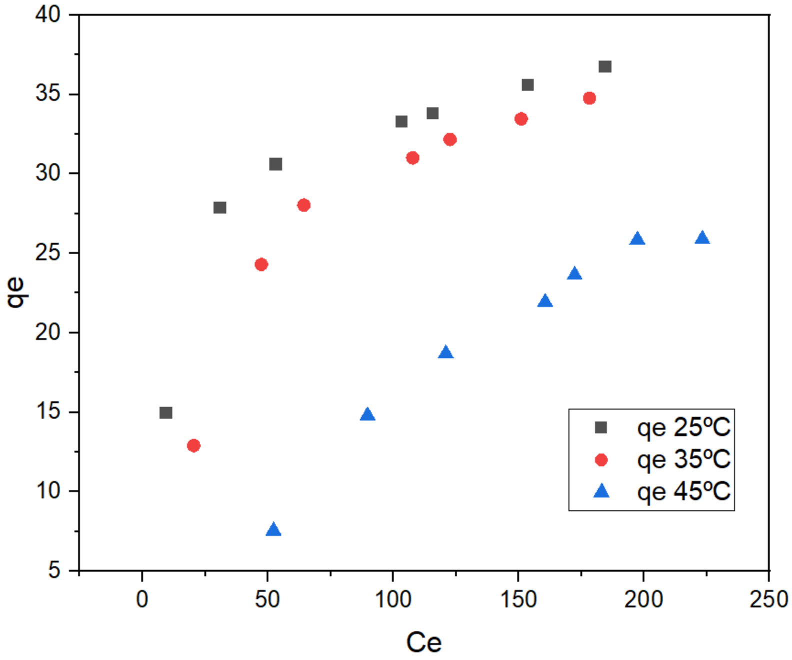
Source: The author.

The tabulated data can be plotted through the ratio $C_e x q_e$, providing the adsorption isotherms, Figure 13 presents three isotherms, one for each temperature, and it is observed that the amount adsorbed qe is greater for the temperature of 25º C.

The isotherms are similar to the classical classification of the L2 type, indicating horizontal adsorptions in layers, also classified as favorable.

Figure 13

Adsorption Isotherms



Source: The author.

4.4.1 Langmuir isotherms

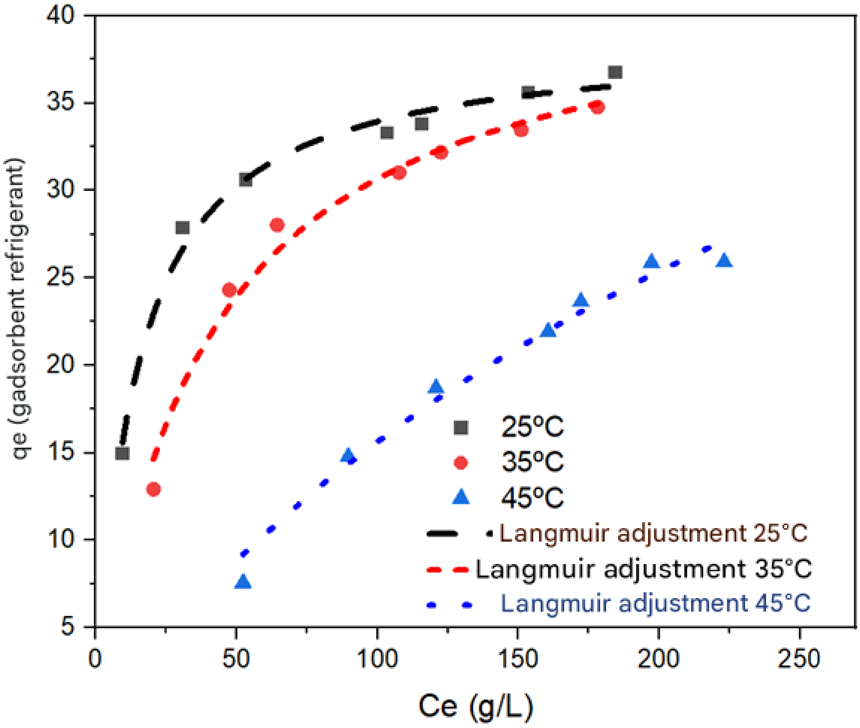
The typical model of chemisorption, Figure 14, provides the adjustment of the experimental points with Equation 8. The parameters are presented in Table 11.

The isotherms indicate that the increase in temperature, the curve tends to lower adsorption capacity and higher concentration in Ce solution, a fact also verified by studying poly(trimethyl allyl ammonium chloride) nanoparticles grafted with chitosan in Fe (YU et al., 2016) $_3O_4$ to adsorb the yellow food coloring 03.(RECK et al., 2018)

Results similar to those present at maximum adsorptive capacity in the Langmuir isotherm model for tartrazine adsorption were observed by ; 31.10 mg/g using Activated carbon of babassu coconut at pH 2, 12 hours and 25ºC and 35.72mg/g using Activated bone carbon at pH 2, 10 hours and 25ºC.

Figure 14

Isotherms fitted to the Langmuir model.



Source: The author.

Table 10

Parameters of Langmuir's constants

Langmuir	K_L	Q_{max}	R^2
25ºC	0,07145	38,705	0,987
35ºC	0,01863	42,638	0,977
45ºC	0,00303	67,251	0,974

Source: The author.

4.4.2 Freundlich isotherms

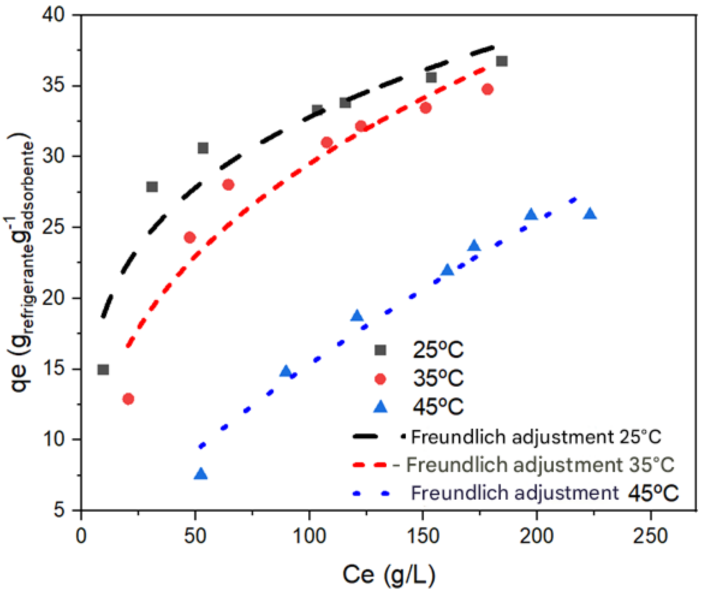
In the possibility of evaluating the experimental data as a system of heterogeneous characteristics, Figure 15 presents the fit of the isotherms to the Freundlich model, using Equation 9, and Table 11 the adjusted parameters.

The description of the Freundlich adsorption model determines a heterogeneous surface, the lower the values of 1/n (closer to zero) the more heterogeneous the surface tends to be, similar values of 1/n are reported for adsorption of the twilight yellow dye in biopolymers are described by (NAGARPITA et al., 2017) LIMA et al., 2019 .

The R^2 correlation values were high, a fact that indicates that the model may be competing with Langmuir's, despite having a lower correlation.

Figure 15

Isotherms fitted to the Freundlich model



Source: The author.

Table 11

Parameters of Freundlich constants

Freundlich	K_F	n	**1/n**	**R²**
25ºC	11,023	4,223	0,237	0,887
35ºC	4,9521	2,782	0,359	0,898
45ºC	0,530	1,366	0,732	0,958

Source: The author.

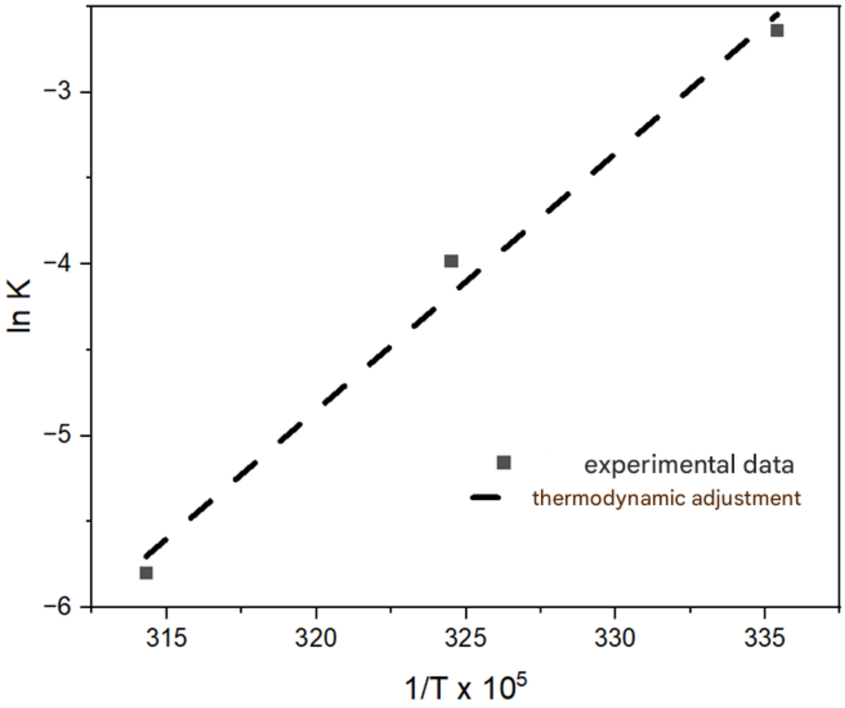
As discussed earlier, Langmuir's model is predominant for the adsorption of soft drink on paper towels, although both models have had ewashed-up settings.

4.5 PART E

All physicochemical processes have thermodynamic parameters that can be calculated (ΔG ΔH ΔS)or ΔG can be calculated for each temperature through Equation 10, the other parameters are obtained by the relation of the slope and linear coefficient of the ratio. $lnKx\ {}^{1}/_{T}$Based on the Langmuir isotherm model reported in the present study, Figure 16 shows the adjustment of the relationship to obtain the thermodynamic and Table 12 presents the values.

Figure 16

Relationship between lnK x 1/T



Source: The author.

Table 12

Thermodynamic Data

Temperature	Langmuir Model	ΔG(J/K)	ΔH(KJ/mol)	ΔS(KJ/Kmol)
T	lnk		124,38	0,438
298,15	-2,639	-6,54		
308,15	-3,983	-10,2		
318,15	-5,799	-15,3		

Source: The author.

Positive values for entropy, , indicate an increase in the degree of freedom of the studied process.ΔS

The Gibbs free energy variation increases in modulus, indicating that the system demonstrates an increase in the degrees of freedom at the adsorption boundary (solid-liquid), a fact also verified in other works (GUPTA; JAIN; VARSHNEY, 2007; JAIN; GUPTA; SIKARWAR, 2010; KHADER; MOHAMMED; ALBAYATI, 2021)

The values between 2.1 and 20.9 KJ/mol indicate weak adsorptions and 20.9 and 418.4 KJ/mol strong adsorptions, in the present work it is proven a value of variation in the enthalpy that indicates strong adsorptions, differently from what was observed in other works, since our analyte is not only dye, we can infer that the sugar present in the soft drink can favor chemisorption.ΔH (HOSSEINI et al., 2011; SAHNOUN; BOUTAHALA, 2018)

5 CONCLUSIONS

- The experimental model was successful for application as an adsorption model.
- The best kinetic mechanism for the equilibrium curves was the pseudo-second-order mechanism, showing similarity to the best model for adsorption of dyes that can compose the soft drink (tartrazine, twilight yellow, yellow 03, yellow 05...).
- The experiment allows the calculation of kinetic parameters.
- The most suitable isotherm was that of Langmuir.
- The work made it possible to calculate the thermodynamic parameters.
- The predicted model has a greater tendency to the chemisorption process, proven through adsorption kinetics as well as isothermal experiments.

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