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Removal of Thallium (Tl+3) from Simulated Wastewater by Raw Rice Husks: Isotherms and Kinetics

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ABSTRACT

All feasible sources of affordable adsorbents should be investigated, as well as their practicality for the removal of heavy metals. As a result, this paper discusses the using inexpensive adsorbents made of agricultural waste, such as rice husk with a surface area of 17.5 m2/g with a maximum adsorption capacity of 2.099 mg/g for thallium via the adsorption process. The experimental data fitted to the Langmuir isotherm model and intra-particle diffusion model with (R2=0.9999). Thermodynamic studies show that the adsorption of thallium was exothermic. A Batch system has been used on thallium for studying the effect of pH, contact time, initial concentration, dose of adsorbents, and agitation speed and the results showed that optimum conditions to get the best removal percentage for thallium of 82.4083% were high pH value of 10, contact time 180min, dose of adsorbents 5g, C0 = 80mg/L, agitation speed 360 rpm. as a result of the research, rice husk can be effectively used as a low-cost option for metal ion removal.

Keywords: Thallium, Batch adsorption, Rice husk, Isotherm model, kinetics model.

Introduction

Thallium (Tl) is a rare but common element found in soils, plants, estuaries, and lakes in the natural environment to human health as well as a huge threat to the environment(Xiao *et al.*, 2012). Over the past few decades, thallium and its compounds have been used more frequently in specialized electronic research equipment, semiconductors, lasers, fiber (optical) glass production, scintilla graphic imaging, superconductivity, fireworks, pigments and dyes, and mineralogical separation processes (Kazantzis, 2000). The main anthropogenic sources of thallium include smelting waste, and industrial wastewaters Thallium is a serious threat to the ecological and human health. The major oxidation states of thallium are Tl(III). Because Tl(III) belongs to the IIIA group in the periodic table, its chemical characteristics are more similar to Al^{+3} Tl(III) has strong oxidizing capabilities and is transformed to a monovalent state slowly (Lin and Nriagu, 1999). As a result, Tl(III) is the most abundant thallium species in most natural habitats (Vink, 1993). Thallium is a dangerous element that has been called "the element cursed at birth" (Peter and Viraraghavan, 2005)**.** It is more harmful to animals than other heavy metals such as mercury, cadmium, lead, zinc, and copper. It is more acutely toxic to mammals than more common heavy metals, like Hg, Cd, Pb, Zn, and Cu (Lan and Lin, 2005). When Tl(III) ions are present in an aquatic environment, they are reported to be 34,000 times more hazardous than Cd(II) ions.Tl(III) is 50,000 times more poisonous than Tl(I) to the unicellular chlorophyte Chlorella, according to Ralph and Twiss(Twining, Twiss and Fisher, 2003).Ion exchange, adsorption, membrane filtration, and electrochemical treatment technologies, have been developed to remove heavy metal ions from wastewater. emissions from wastewater and coal-related solid waste (Zhang, 2014). Evaporation, filtration, reverse osmosis, electro deposition, and coagulation are all examples of chemical oxidation or reduction (Das, Vimala and Karthika, 2008). Solvent extraction, sedimentation, cementation, flocculation (Swathi *et al.*, 2014). Electrodialysis, ultrafiltration, photocatalysis, complexation, and foam floatation are some of the methods used to purify water (Gunatilake, 2015). Types of metals present in solutions and the cost of treating wastewater are the main parameters of determining which technology used for wastewater treatment. The process of removing thallium from aqueous solutions has been the subject of few investigations. The EPA has authorized ion exchange and activated alumina precipitation as effective procedures for eliminating thallium from drinking water. Hydroxide precipitation is an alternative approach, resin-based ion exchange (Horne, 1958), material adsorption (for example, activated carbon) (Arifi and Hanafi, 2011), ferrihydrite (Peter and Viraraghavan, 2005), sawdust (Memon, Memon and Solangi, 2008), biomass (Peter and Viraraghavan, 2008). Adsorption is the most often employed of these strategies due to its high removal effectiveness and ease of application. There are many applications for using adsorption to remove various contaminates. The adsorption technique is utilized to remove many organic or inorganic pollutants from wastewater like humic acid (Nsaifabbas and Abbas, 2014), phenol(Abbas *et al.*, 2019), phosphorus(Abbas, 2015), cyanide (Alalwan, Abbas and Alminshid, 2020), etc. Dyes also eliminated from wastewater by adsorption technique with high efficiency e.g., RG-19(Ghulam, Abbas and Sachit, 2020), methyl green(Alalwan *et al.*, 2021), and malachite green(Alwan *et al.*, 2021). The remediation by adsorption not limited for contaminants aqueous solutions, but also included removing heavy metals(Abbas *et al.*, 2021), and PO_4^{-2} ions from soil(Abbas, Al-Madhhachi and Esmael, 2019). The corroded sulfur ions and heavy metals found in the petroleum fractions such as heavy naphtha (Abbas and Alalwan, 2019), light naphtha(Abbas and Ibrahim, 2020), and nickel(Suha Anwer Ibrahim *et al.*, 2021), is also removed by adsorption technique. The heavy metals considered as one of most contaminants removing from water via adsorption such as zinc(Al-Hermizy *et al.*, 2022), manganese (Suha A Ibrahim *et al.*, 2021), antimony (Suha A Ibrahim *et al.*, 2021), lead (Abbas, Ali and Abbas, 2020), cadmium(Abbas and Nussrat, 2020), nickel(Maddodi *et al.*, 2020), thallium(Alalwan *et al.*, 2018), tin(Abbas and Abbas, 2013), bismuth (Abbas and Abbas, 2013), and magnesium (Nsaif and Saeed, 2013). The non-valuable materials are proved a suitable efficiency to use as an adsorbent media in addition to its low cost for instances water hyacinth (*Eichhornia crassipes*) (Abbas, Ali and Abbas, 2020), eggshells (S. A. K. Ali *et al.*, 2020). There is only one obstacle to this wonderful technology, which is the accumulation of waste, which is often toxic, after the completion of the treatment process. This requires additional effort and cost to dispose of this waste. The problem has been solved through the concept of zero residue level by converting this waste into useful and environmentally friendly materials. It was used as a rodenticide such as 2,4-D(Abd Ali *et al.*, 2018), ribavirin (Suha A Ibrahim *et al.*, 2021) and carbamazepine (Ibrahim *et al.*, 2020), where their effects were studied. The wastes were used as an additive to enhance concrete resistance (Abbas, Abdulkareem and Abbas, 2022), and to produce useful materials such as acetone (Abbas *et al.*, 2022) and nanoparticles (Alminshid *et al.*, 2021) as well as other applications for the transfer of beneficial substances within living bodies (S. T. Ali *et al.*, 2020). However, three objectives must be met for thallium removal by adsorption: Adsorbent reusability, efficient removal capabilities, and quick rate of absorption. Agriculture by-products have piqued the interest of researchers as adsorbent materials due to their low cost and lack of regeneration requirements Tl(III) removal has been tested on a variety of biomass sources Due to its high surface area and granular texture, rice husk has been found to have potential adsorption activity in the removal of heavy metals from wastewater (Qu *et al.*, 2018). Rice husk contains 50% cellulose, 25%–30% lignin (polar functional groups), and 15%–20% silica, which are the primary active sites for chemical reactions (Ummah *et al.*, 2015).In a batch mode, rice husk was investigated as a novel adsorbent material for extracting thallium from aqueous solutions. The effects of operating parameters such as the initial thallium concentration, the pH of the thallium solution, the mass of the adsorbent, agitation speed, contact time, and influent temperature are discussed in this study's adsorption kinetics and isotherms. Rice husk, has a promising sorption potential of thallium from wastewater industrially.

II. MATERIALS AND METHODS/METHODOLOGY/EXPERIMENTAL PROCEDURE

A. Preparation of Rice Husks

Used as adsorbent for thallium, naturally collected date raw material from Al Mashkhab district rice mill in Al-Najaf Governorate, during rice production, large quantities of residual rice husks are obtained. A significant amount of RH was respectively washed 7-10 times with distilled water for takeoff impurities such as soluble and dried at 50°C in a hot air oven show Figure 1 for 2 h to get well completely dried sample and also remove residual moisture. At room temperature, rice husks are left to cool, the rice husks are stored in bottles clean and kept close for use.

Figure:1 Rice husk in the oven after washing.

B. Stock Solution

To avoid contamination by extra components that could be present in real wastewater, a simulated synthetic aqueous solution (SSAS) was created in the lab with various Tl^{+3} concentrations. To generate a 1000 mg/L thallium stock solution, 500 mg thallium nitrate $Tl(NO_3)_3.3H2O$ (Alfa Aesar, 99.5 percent) was dissolved in one litter of distilled water. Each experiment involved diluting Thallium stock solution to reach the needed concentrations of Thallium solutions. 0.1 N potassium hydroxide (KOH) (Alfa Aesar) and 0.1 N hydrochloric acid (HCl) (Sigma Aldrich) were added to the solution to achieve the desired pH value. to create a Thallium calibration curve used for Multiple solutions of different concentrations ranging from 5 to 75 mg/L. Absorbance at its wavelength of 335.1nm, was measured for each concentration using a UV-spectrophotometer, Figure 2 show linear relationship between absorbance and Thallium concentration was plotted to estimate unknown Thallium concentrations.

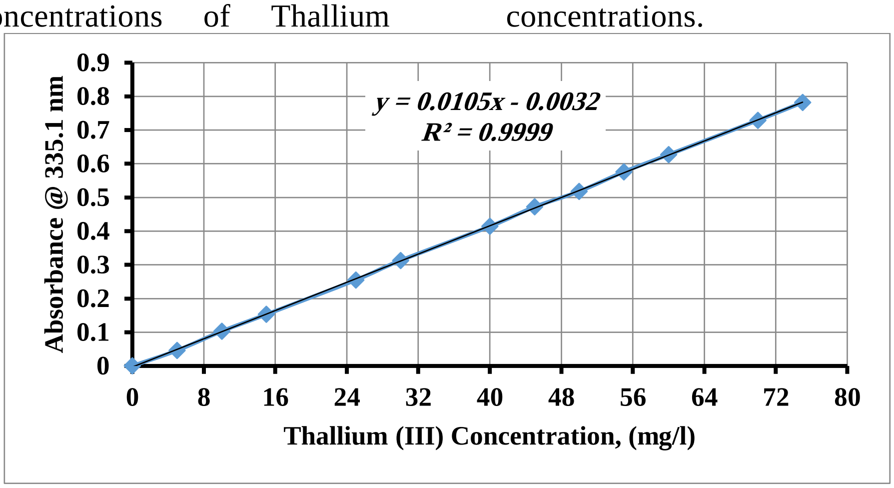


Figure :2 represents the calibration curve of Thallium.

C. Batch adsorption

A batch system was used to treat simulated wastewater. Using rice husks as an absorbent material, experiments were carried out at different laboratory temperatures using Pyrex Erlenmeyer flasks with a size of 100 cm^3. Each beaker contains 100 ml of the contaminated aqueous solution of the desired concentration, pH, and dosage of rice husk. The conical flasks were stirred for a certain time at the specified speed. After the contact period was over, samples were prepared for AAS analysis by filtration using 0.45µm Whitman filter paper, and the thallium ion concentration was determined using a calibration curve aid. Equations (1) and (2) were used to calculate the adsorption capacity and percentage of thallium ion removal by rice husk residues.

$$q_e = \frac{(C_o - C_e)V}{M} \quad (1)$$

$$R\% = \frac{(C_o - C_e)}{C_0} * 100 \quad (2)$$

D. Study of operation parameters

Many operation parameters were investigated to see how they affected the removal of TI by rice husk, including adsorbent dose, initial TI concentration, agitation speed, contact time, temperature, and pH values, which ranged from 0.5-5g, 1-100ppm, 100-500 rpm, 5-180 min, 25-50°C, and 1-10, respectively.

III. RESULTS AND DISCUSSION

A. Surface Area determination

The samples' specific surface areas were determined using the Brunauer-Emmett-Teller (BET) method. displays Table 1. with a surface area analyzer. BET tests were carried out in the Center of Petroleum Research and Development/ Catalysts Department (Ministry of oil/Iraq).

Table 1. shows the Surface area of the adsorbent.

Adsorbent	Rice Husk
Surface area (m^2 /g)	17.5

B. Effect of Various Factors on Rice Husk Adsorption

1) Effect of pH

One of the most important parameters regulating metal ion adsorption is the pH of the effluent. In the initial pH range of 1-10, the effect of pH on thallium adsorption was investigated. Figure 3 shows the relationship between the initial pH of the solution and the percentage removal of certain heavy metals The influence of pH on the adsorption features of thallium was substantially responsible for the variance in adsorption capacity in this pH range, indicating that the adsorbent's adsorption capacity was pH-dependent. The optimum pH was found to be about pH 10, with a percentage removal of 100% for thallium by rice husk. Due to electrostatic interaction between areas with the negative charge on the surface of adsorbent rice husk and of the heavy metals, thallium removal increased at higher pH values. As a result, the surface interaction of heavy metals took precedence. The number of hydrogen ions in the solution dropped as pH increased. Furthermore, competition for heavy metal ions decreased, resulting in more effective sorption. At pH values over 8, precipitation becomes dominant, or both ion exchange and aqueous metal hydroxide production become important mechanisms in the metal removal process, a situation that is not always ideal because metal precipitation can lead to a misunderstanding of the adsorption capacity. the result similar(Vieira *et al.*, 2014).

2) Effect of mixing speed

The effect of mixing speed, which ranged from 100 to 500 rpm, on the removal of thallium by rice husks was investigated Figure 4 shows that when the rotational speed reaches 360 rpm, the pollutant removal ratio increases to a maximum percentage, after which it drops. The increased mixing of thallium ions in the absorbing container could be one of the causes of this condition. These circumstances lead to one result repulsive forces dominate over sorption sites on the absorbent material's surface. and finally, reduce the attraction between the thallium ion and the rice husk Therefore, 360 rpm was chosen as the ideal speed for the following adsorption experiments.

3) Effect of Initial Concentration

In conditions of room temperature = 25C^0, thallium the influence of concentration on the adsorption rate was investigated. and 360 rpm shaking speed, as shown in Figure 5 In the range of 1 to 100 g/l, the influence of initial heavy metal concentration was investigated. The percentage of heavy metals removed reduced as the original concentration of heavy metals increased. The saturation of accessible active sites on rice husk over a particular concentration of heavy metals caused this result Similar results were reported by (Ahile *et al.*, 2017).

4) Effect of Adsorbent Dose

The dosage trial was carried out at room temperature (25^{o}C) with doses ranging from 0.5 to 5 g. The initial concentration of thallium ions in this investigation was set to 80 mg/L. The uptake of thallium rises as the dosage increases from 0.5 to 5g, as a result of the restricted number of adsorbing species present for a substantially larger number of accessible surface sites on the adsorbent at higher dosages, as seen in Figure 6 It is well known that at greater adsorbent doses, the availability of exchangeable sites from metal ion absorption increases. The optimum dose for thallium by rice husk was found to be 5g at a percentage elimination of 82.4083%. Because of the increased surface area and active sites, the percent removal of heavy metals by rice husk rose as the adsorbent dosage was raised**.** The result agrees (Vieira *et al.*, 2014).

5) Effect of Contact Time

Figure 7 shows the relationship between adsorption efficiency and contact time in terms of percentage thallium removal. The adsorption of TI(III) increased with increasing contact time whereas the percentage removal was 82.4083% at a contact time of 120 min. The maximum rate of adsorption. peaking at around 120 minutes for thallium, and then remaining nearly constant. Adsorption efficiency is improved. As time passes, accessible adsorption sites (surface functional groups) on the adsorbent surface are ascribed to rice husks The agree result (Vieira *et al.*, 2014).

6) Effect of Temperature

in the temperature range of 25-50°C, the effect of temperature on thallium removal on the rice husk adsorbent was examined. Temperature affects thermodynamic characteristics, indicating whether the adsorption is exothermic or endothermic. The adsorption of thallium reduced as the system's temperature was raised Figure 8 The decrease in surface activity was mostly attributable to the fact that heavy metals adsorption is an exothermic process for rice husk adsorbent Same findings are reported by (Abdel Halim, El-Ezaby and El-Gammal, 2019).

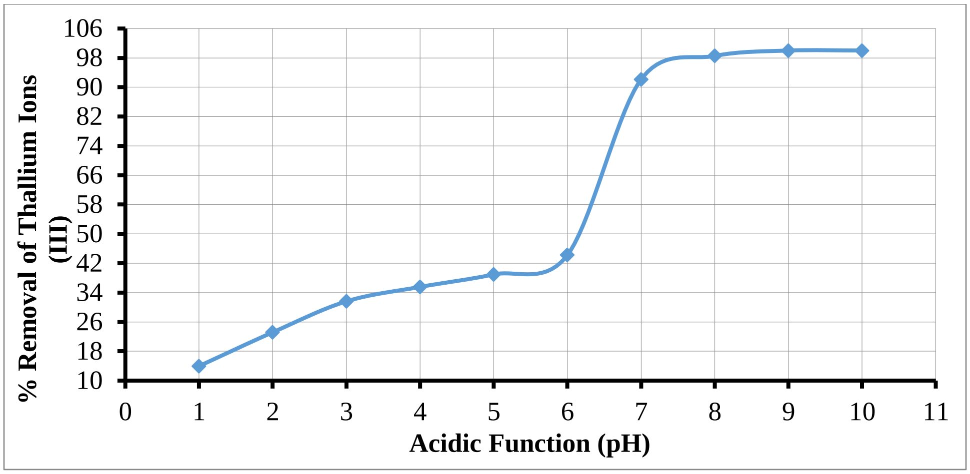


Figure:3 Effect of pH on removal percentage for TI^{+3}.

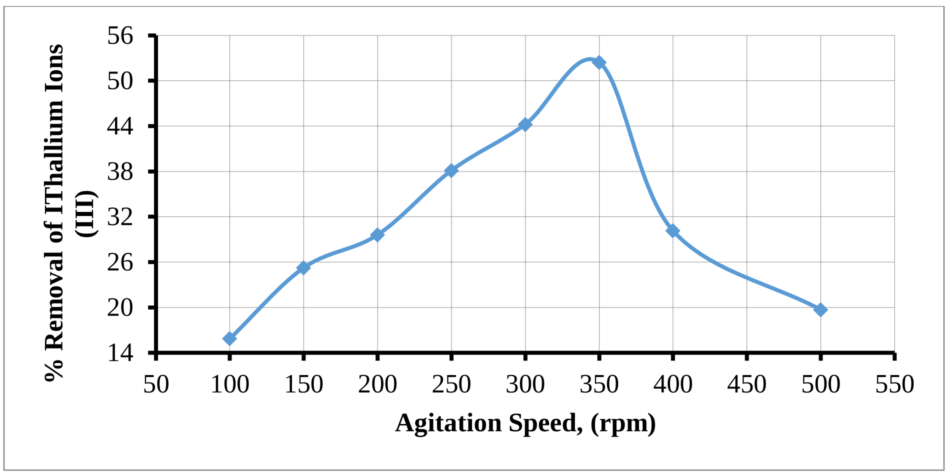


Figure:4 Effect of Agitation speed on removal percentage for TI^{+3}.

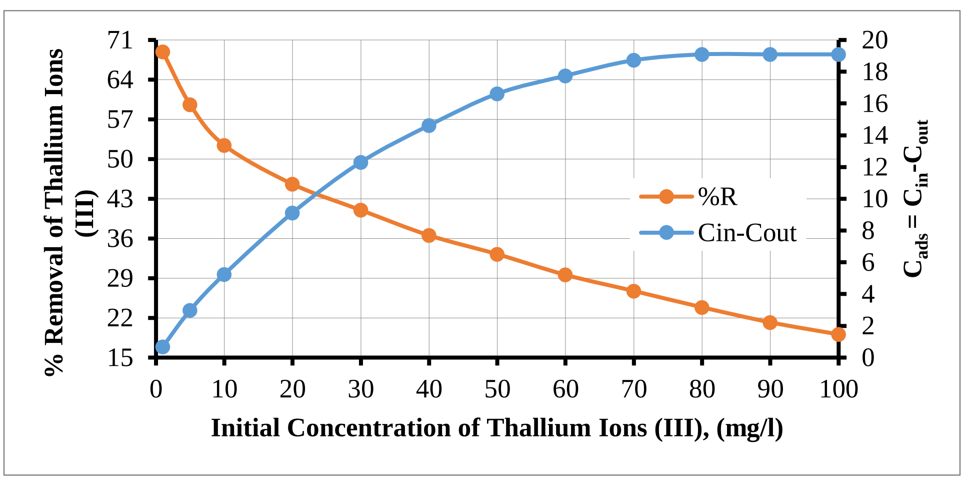


Figure:5 Effect of concentration on removal percentage for TI^{+3}.

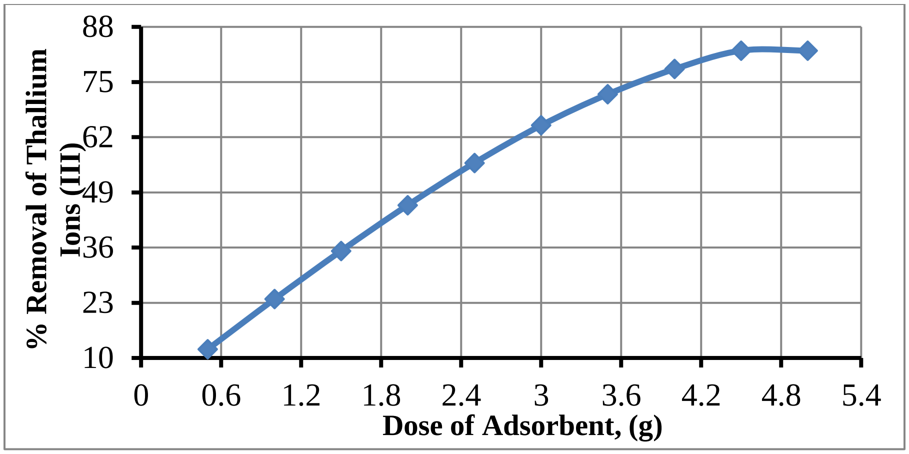


Figure:6 Effect of dosage on removal percentage for TI^{+3}.

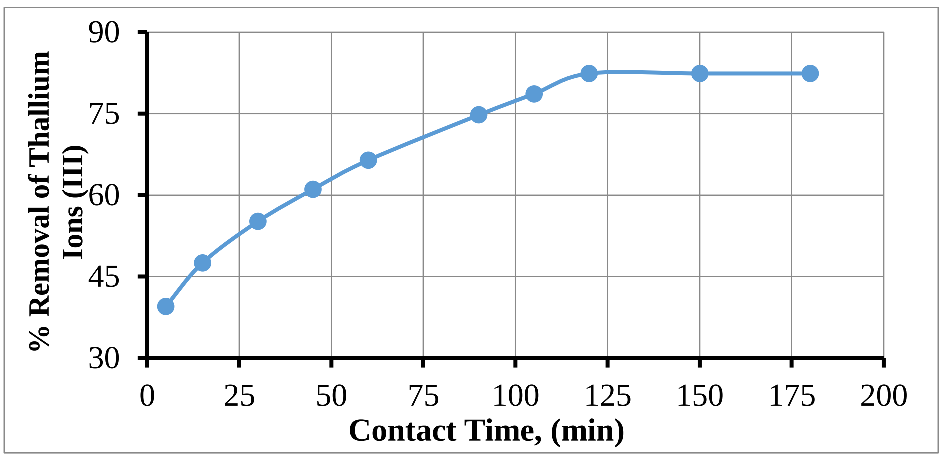


Figure:7 Effect of contact time on removal percentage for TI^{+3}.

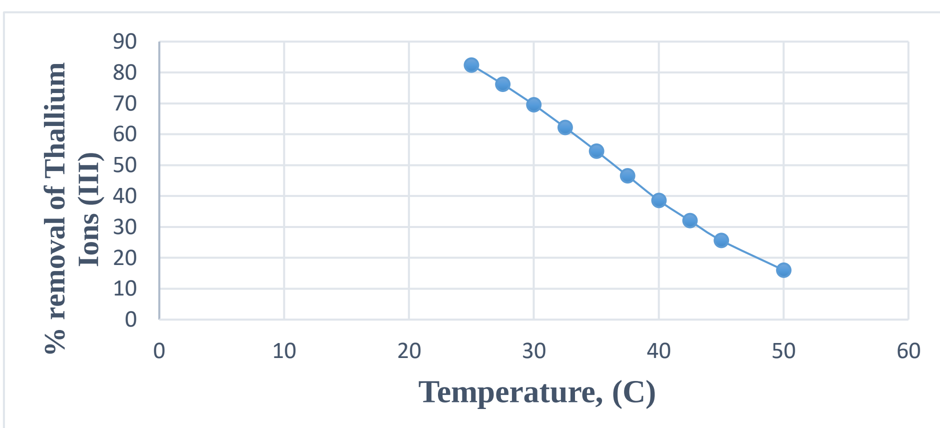


Figure:8 Effect of Temperature on removal percentage for TI^{+3}.

C. Adsorption Isotherm study

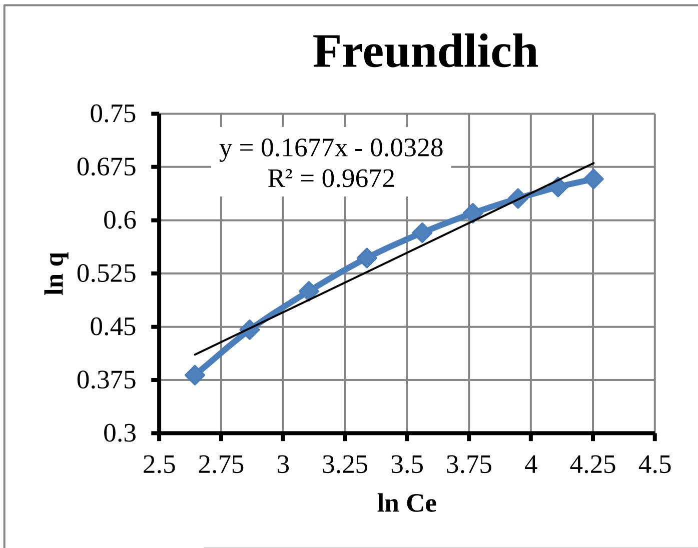
Adsorption isotherms are described as a relationship between the concentration of the adsorbate in the liquid and the amount of adsorbate absorbed by the unit mass of adsorbent at a fixed temperature. There may not be a simple expression capable of explaining the equilibrium relation between the adsorbate in a liquid phase and the adsorbate in the solid phase due to the complexity of a liquid-phase adsorption on microporous substances. A brief description of various adsorption isotherm models is given below, including Langmuir**,** Freundlich, Temkin.

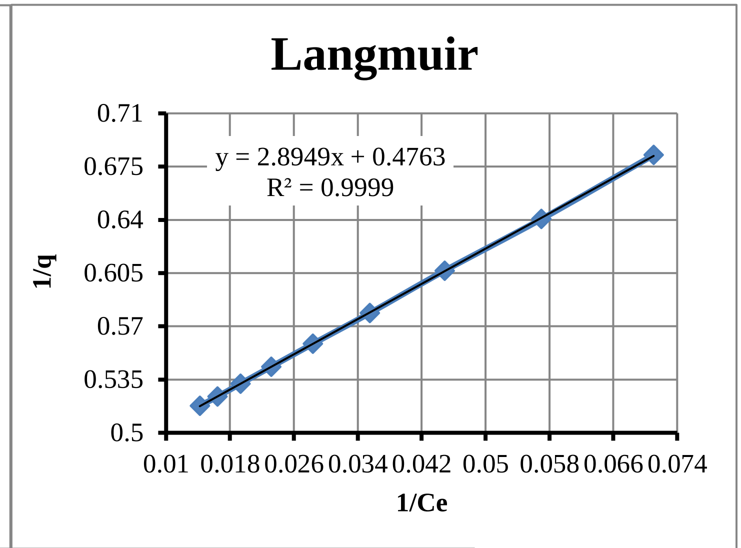
Isotherm data was linearized using the Langmuir equation and plotted between 1/q and 1/Ce , log C_{out} VS log q for Freundlich model and ln C_{out} VS q for Temkin model. the Langmuir isotherm model was shown to be the best fit to the experimental data (R^2 = 0.9999) for thallium adsorption on rice husk compared to the Freundlich and Temkin isotherm (R^2 ranged from **0.9672** to **0.979**) illustrated in Table.2 and Figure 9 below.

Langmuir constant qm, which is a measure of monolayer adsorption capacity, was found to be 2.099. For thallium by rice husk, the Langmuir constant b, which represents adsorption energy, was 0.1645. This indicates that the surface of the rice husk is homogeneous and that adsorption takes place in a monolayer. Similar results were reported by (Abdel Halim, El-Ezaby and El-Gammal, 2019).

Table2.Isotherm parameter for thallium adsorption onto rice husk.

Model	Parameter	Rice husk
Langmuir Isotherm	q_m (mg/g)	2.099
	K_L (l/mg)	0.1645
	R_L	0.0706
	R^2	0.9999
Freundlich Isotherm	K_F ($mg^{1-1/n}L^{1/n}/g$)	0.9677
	n	5.963
	R^2	0.9672
Temkin Isotherm	K_T (L/g)	13.459
	B_T	8652.07
	R^2	0.979





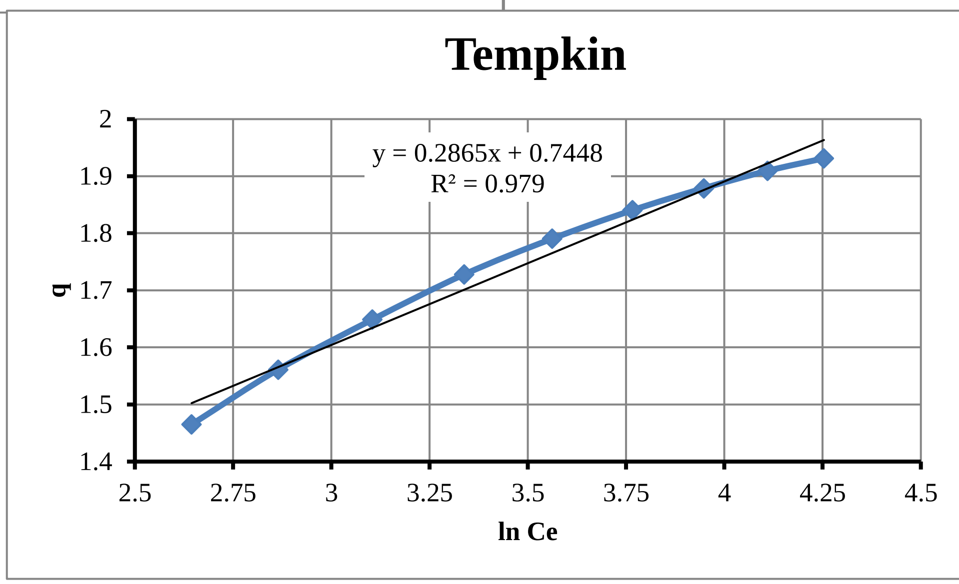


Figure :9 Adsorption isotherm model; Langmuir, Freundlich and Temkin for TI^{+3}.

D. Kinetic Study

Adsorption kinetics explains reaction pathways and the duration it takes to reach equilibrium. All experimental data were fitted into three kinetic model Intra-particle diffusion, Pseudo first order and Pseudo second order models. A straight line representing pseudo first-order kinetics for the elimination of thallium using rice husk was plotted as ln (q_e-q_t) versus t, for pseudo second-order kinetics by plotting t/qt vs t, for Intraparticle Diffusion Model by plotting q vs t.

The intraparticle diffusion model was shown to be the best fit to the experimental data (R^2 = 0.9999) for thallium adsorption on rice husk compared to the pseudo first-order and pseudo second-order isotherm (R^2 ranged from0.9722 to0.9857) illustrated in Table.3 and Figure 10 below the values of intraparticle diffusion constants, C and K, the calculated intraparticle diffusion coefficient is 0.0872 m^2 /sec for thallium on rice husk Similar results were reported by (Abdel Halim, El-Ezaby and El-Gammal, 2019).

Table 3. Kinetic parameter model for TI^{+3}adsorption on RH.

	Pseudo first order model		Pseudo second order model		Intra-particle diffusion model
q_e	15.335	q_e	1.574	K_I	0.5046
K_1	$-2.2222*10^{-6}$	K_2	0.0437	C	0.0872 m^2 /sec
R^2	0.9722	R^2	0.9857	R^2	0.9999

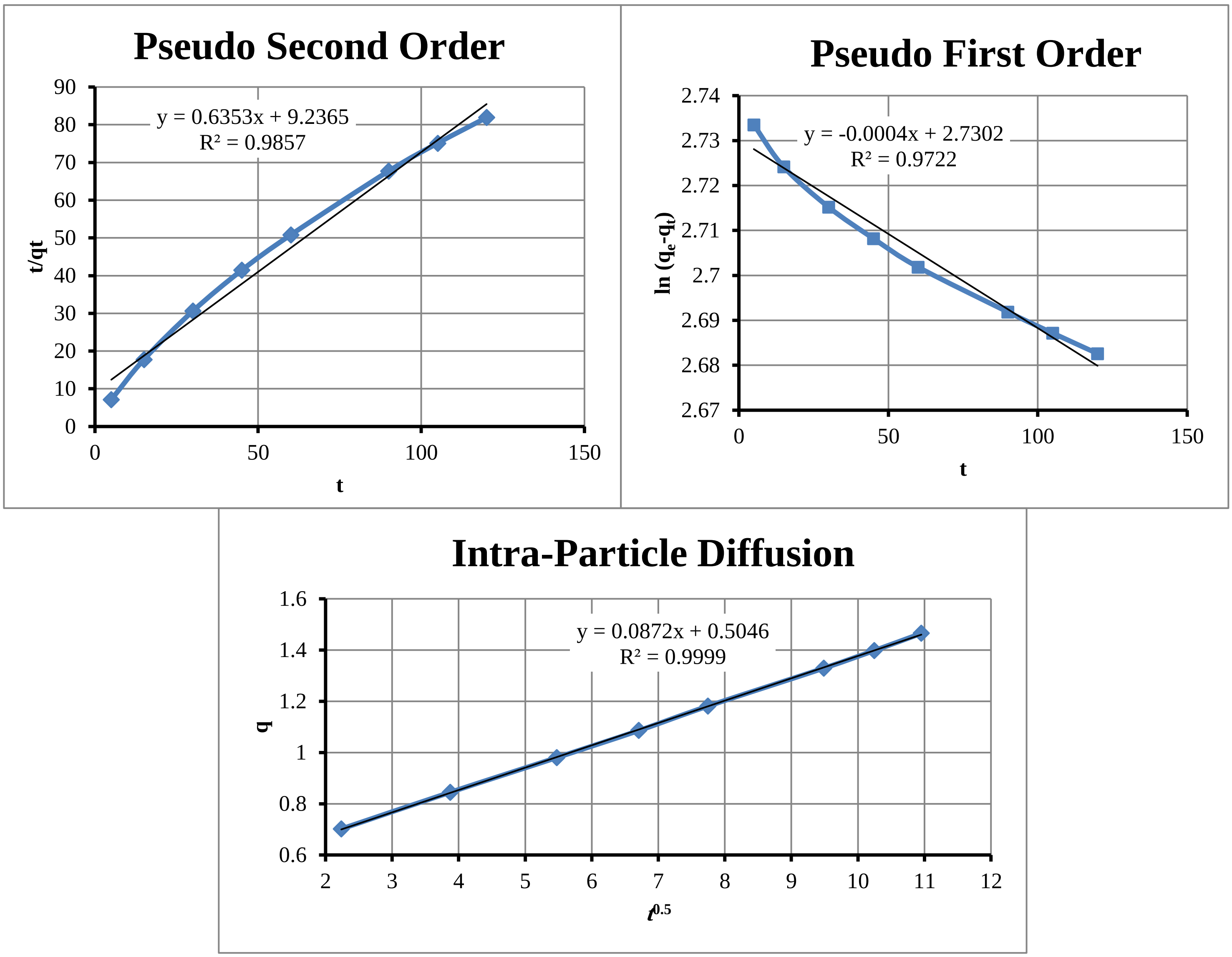


Figure :10 Kinetic study models of TI^{+3}adsorption on rice husk

E. Thermodynamic Studies for Rice Husk

the plot of ln K versus 1/T the slope and intercept were used to derive the values of (ΔH) and (ΔS), and the sorption Gibbs free energy change (ΔG) was calculated. Table.4 and Figure 11 below show the results of thallium adsorption onto rice husk. Free energy, entropy, and enthalpy must all be addressed when determining whether a process will occur spontaneously in any adsorption process. The measurement of ΔH proved the exothermic character of thallium. The increase in ΔG with temperature was attributed to the spontaneous nature of the adsorption process, which was more beneficial at low temperatures. Similar results were reported by (Abdel Halim, El-Ezaby and El-Gammal, 2019).

Table 4. Thermodynamic parameters of TI^{+3}adsorption on rice husk.

T(K)	ΔH (J/mol)	ΔS (J/mol.K)	ΔG (kJ/mol)
298.15			-17.46950119
300.65			-16.76167020
303.15			-16.05383922
305.65			-15.34600823
308.15	-101885.4249	-283.1323955	-14.63817724
310.65			-13.93034625
313.15			-13.22251526
315.65			-12.51468427
318.15			-11.80685328
323.15			-10.39119131

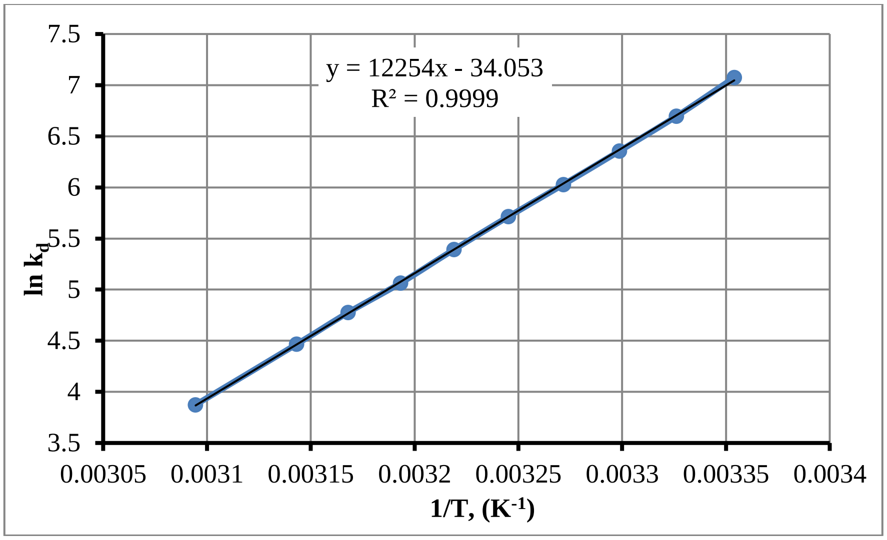


Figure :11Thermodynamic parameters for $TI^{+3}$by rice husk.

IV. CONCLUSION

Rice husks are a type of agricultural waste this product has excellent adsorption properties. The adsorption of thallium by rice husks was found to be highly dependent on pH, rice husk dosage, and contact time. the optimum sorption occurring pH 10, $C_{=}$80mg/l agitation speed 360 rpm, contact time 180 min, adsorbent dosage 5g. the Langmuir isotherm model was shown to be the best fit for heavy metals at R^2 = 0.9999. The Intraparticle Diffusion model best described the adsorption process, according to kinetic studies. The current study found that rice husk can be employed as an adsorbent for wastewater treatment, with removal rates of up to 82.4083% for TI^{+3}. Furthermore, because the raw material is inexpensive, it is a cost-effective technique.

AUTHOR CONTRIBUTIONS

Z.A.Gadooa: Conceptualization, Methodology, Writing – original draft, Writing – review & editing. **M.N.Abbas:** Supervision, Writing – original draft, Writing – review & editing.

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