

Article

Adsorption of the Pesticide (Divid Top) via the Use of Tea Leaf Residues

Yosef Othman Homeda*¹

1. Tikrit University, College of Education for Pure Sciences, Department of Chemistry, Iraq.

* Correspondence: yosef.a.homeda@tu.edu.iq

Abstract: The possibility of using ground tea leaves as an efficient, inexpensive, and environmentally friendly adsorbent to remove the pesticide Divid Top from aqueous environments has been studied, and the main conclusions can be summarized. The kinetic study was conducted in stages, and the thermal equilibrium study was carried out under different experimental conditions. in terms of contact time initial pesticide concentration, and the amount of adsorbent material. The study was conducted and showed good results. Adsorption was analyzed using pseudo-first-order and pseudo-second-order kinetic models. The study found that it follows the pseudo-second-order kinetic model better, which supports the hypothesis that the mechanism involved is chemical adsorption, involving binding forces between pesticide molecules and the active groups on the surface of tea leaves. The equilibrium data were represented using the Langmuir adsorption model, with a correlation coefficient ($R^2=0.9135$), indicating that the adsorption process occurs in the form of a monolayer over a surface with homogeneous energy. As for the efficiency and feasibility of the adsorption process, the maximum adsorption capacity ($q_m = 0.25 \text{ mg/g}$) according to the Langmuir model, and the separation factor value ($RL = 0.091$) confirmed that the adsorption process is highly preferred under the studied conditions. Thermodynamics: Thermodynamic calculations have proven that the adsorption process is endothermic due to the positive values of the enthalpy content (ΔH). The negative values of the Gibbs free energy (ΔG) at high temperatures revealed the spontaneity of the reaction and the increased removal efficiency with rising temperature.

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1. Introduction

Environmental pollution is considered one of the fundamental challenges of the modern era, especially in light of the technological and industrial expansion that has negatively altered natural systems [1]. The instabilities of dirtiness are not restricted to the deterioration of air and soil conditions; they pose a harmful danger to public health, as current environmental reports signify that pollutants cause the deaths of heaps of people occurring around the world [2]. In the circumstances of water contaminants, land pesticides are prominent as one of the ultimate complex and perilous compounds [3]. Despite their duty in saving crops and increasing output, their synthetic support leads to their draining into groundwater and accumulation in living tissues through the feeding relationships among organisms [1], [4]. Recent studies [5] have demonstrated that traditional analysis systems grant permission not answer to remove poison residues at reduced concentrations, making it necessary to approve more efficient and tenable

methods. Adsorption electronics is now considered the optimum resolution for water cleansing on account of its ease of movement and extreme competency to eliminate natural pollutants outside, making poisonous by-products [6], [7]. With the constant rise in the cost of monetary stimulated elements, research has shifted toward the "circular saving" through the reusing of land waste. In this framework, pioneering studies [8], [9] have justified that beverage leaf residues (*Camellia sinensis*) possess special properties as an adsorbent material, from an abundant surface field and an absorbent structure that allows for the productive capture of poison atoms [10]. The excellent talent of beverage residues is from their chemical arrangement, which is rich in hydrogen, lignin, and live functional groups to a degree hydroxyl and carboxyl, that comprise synthetic binding sites for pesticides through hydrogen bonds and electrostatic interactions [5]. Accordingly, this research aims to inspect the effectiveness of beverage residues in adsorbing [insert poison name present], as an environmentally companionable and cheap option that contributes to carrying out Sustainable Development Goals (SDGs 6), had connection with clean water [11].

2. Materials and Methods

2.1 Materials used: Chemicals

In this study, the poison Divid Top Tebuconazole ($C_{16}H_{22}ClN_3O$) made by Shanghai Agroriver Chemical Co., Ltd. was secondhand. Since the Tebuconazole pesticide used (Divid Top) contains a pink dye added by the manufacturer for visual distinction purposes, a wavelength of 555 nanometers was adopted to estimate the remaining concentrations after the adsorption process. This wavelength represents the maximum absorption of the pink dye chemically and volumetrically associated with the active substance, allowing for accurate tracking of the product's removal efficiency from the aqueous solution. It was used as a standard solution to prepare diluted solutions using distilled water for the current study [12].

2.2 Method of Preparing Solutions A standard solution of the pesticide was prepared by dissolving it in distilled water at a concentration of 1000 ppm, from which several concentrations (150, 50, 75, 100, 125, 25 ppm) were prepared.

2.3 Preparation of the adsorbent material

A specific type of tea used in the Iraqi markets (Jihan type) was selected, and the exhausted tea leaves were prepared [13]. By heating 25 grams of tea leaves in one liter of drinking water at a temperature of 100 degrees Celsius for one hour, and then isolating the exhausted leaves to reheat them with the same amount of water and temperature for 20 minutes, twice. The drying is done using a home microwave oven (Panasonic, WF33G-NN) for 45 minutes at 800 watts and a wavelength of 2450 MHz. The dried leaves are left inside the oven for an hour until their temperature stabilizes, then they are collected in airtight plastic containers in preparation for grinding. The grinding process is done in batches to obtain the selected section, and a grinder of the type (Belgium, Benelux Intertest) was used. The ground leaves were separated using two sieves with mesh numbers (No Mesh) (230-120) from the Turkish company TEST & ANALiZ (ELĞi). No Mesh)(230-120), from the Turkish company TEST & ANALiZ (ELĞi). The product is collected in airtight plastic containers for later use in the adsorption process [14], [15].

2.4 Adsorption Experiments

1.2.4 Batch Adsorption Studies

The "batch adsorption" method was followed to study the efficiency of tea leaves in removing the pesticide tebuconazole, where the study included the evaluation of four key variables to ensure a precise understanding of the adsorption mechanism [16].

1. The effect of adsorbent dosage. The effect of adsorbent dosage

The experiments were attended utilizing variable amounts of tea leaf powder varying from (0.1 to 0.5 g) while keeping the resolution volume constant at (50 mL). This step aims to decide the optimum abundance that determines the maximum surface area and active sites for binding accompanying the pesticide particles [17].

2. Temperature Effect Temperature Effect

The system was calculated over a temperature range of (30°C to 50°C) accompanying a formal increase of 5 degrees Celsius for each calculation. This study helps in manipulating thermodynamic limits in the way that enthalpy (ΔH) and free energy (ΔG) are used to decide whether the response is endothermic or exothermic.

3. Effect of Contact Time: Effect of Contact Time

To estimate the adsorption action, the expulsion process was monitored over time periods varying from 10 minutes to 50 minutes. These dossiers help decide the time necessary to reach the physicochemical balance between the liquid chapter and the adsorbent material.

4. The effect of initial concentration. Effect of Initial Concentration

The beginning poison concentrations were transformed to judge the maximum loading volume of beverage leaves and to request adsorption isotherm models to a degree, Langmuir and the Freundlich.

5. For Separation and Spectroscopic Analysis

After developing the contact, come into sight each experiment, the resolution was separated from the beverage leaf atoms utilizing Whatman No. 1 permeate paper. 1) The clear solution withstood spectrophotometric calculation utilizing a UV-Vis tool at a wavelength of 555 nm, where the aggregation of the surplus poison was supposed to establish the intensity of the pink dye guide the poison [13].

3. Results and Discussion

3.1 The Effect of the Amount of Tea Leaves (Adsorbent Factor)

Different weights of tea leaves were used to determine the optimal removal weight for the tetrachlorazone pesticide at a constant concentration (75 ppm), as the absorbance obtained was converted into removal efficiency. By converting the absorbance data (Abs) into a percentage removal (%R) and adsorption capacity (qe), the percentage removal (R%) is calculated as shown below. In Table 1

$$R = \frac{Abs1 - Abs2}{Abs1} \times 100 \quad (1)$$

Table 1. Represents the removal percentage results at different weights of the adsorbent.

| No. | Wt gm | Abs 1 | Abs 2 | R % |
|-----|-------|-------|-------|-------|
| 1. | 0.1 | 1.069 | 0.869 | 18.7% |
| 2. | 0.2 | 1.069 | 0.475 | 55.6% |
| 3. | 0.3 | 1.069 | 0.360 | 66.3% |
| 4. | 0.4 | 1.069 | 0.233 | 78.2% |
| 5. | 0.5 | 1.069 | 0.188 | 82.4% |

Then we obtain the adsorption capacity (q_e) at equilibrium (the amount of adsorbate per unit mass of adsorbent) from the following equation, as in Table 2

$$q_e(\text{mg/g}) = \frac{(C_0 - C_e)V}{W} \quad (2)$$

Table 2. Represents the adsorption capacity at different weights of the adsorbent material.

| No. | W gm | Abs 2 | C _e (mg/L) | C ₀ -C _e (mg/L) | q _e (mg/g) |
|-----|------|-------|-----------------------|---------------------------------------|-----------------------|
| 1. | 0.1 | 0.869 | 60.9 | 14.1 | 7.05 |
| 2. | 0.2 | 0.475 | 33.3 | 41.7 | 10.43 |
| 3. | 0.3 | 0.360 | 22.2 | 49.8 | 8.30 |
| 4. | 0.4 | 0.233 | 16.3 | 58.7 | 7.34 |
| 5. | 0.5 | 0.188 | 13.2 | 61.8 | 6.18 |

By reviewing the results in Tables 1 and 2, we notice that the removal percentage continuously increases with the increase in weight, which is logical because increasing the adsorbent material increases the number of active sites available for pesticide adsorption, thereby enhancing its removal efficiency. As for the adsorption capacity, it reaches its peak at a weight of (0.2 grammes, which is 10.43 mg/g). It decreases after increasing the weight of the tea leaves, as shown in Table 2. When the weight of the tea leaves increases, the pesticide available at a concentration of (75 mg/L) is distributed over a larger number of adsorption sites. It cannot reach all sites at its maximum capacity, which reduces the adsorption capacity per gram of tea leaves. The best adsorption efficiency occurs at 0.2 grammes because it represents the highest adsorption capacity (10.43 mg/g) and a removal rate of 55.6% of the pesticide.

3.2 Contact Time (Rebound)

The effect of the contact time of the pesticide with tea leaf powder was studied, and upon observing Table 3, we notice an increase in the amount of pesticide adsorption with increasing time [18]. If the equation below was used.

$$C_t(\text{mg/L}) = C_0 \times \frac{\text{Abs } t}{\text{Abs } 1} \quad (3)$$

$$q_t(\text{mg/g}) = \frac{(C_0 - C_t) \times V}{W} \quad (4)$$

Table 3. Concentration and Capacitance after Contact Time.

| Tim mint | Abs 1 | Abs t | C _t (mg/L) | Q _t |
|----------|-------|-------|-----------------------|----------------|
| 10 | 1.084 | 0.445 | 30.77 | 11.06 |
| 20 | 1.084 | 0.295 | 20.37 | 13.66 |
| 30 | 1.084 | 0.265 | 18.32 | 14.17 |
| 40 | 1.084 | 0.326 | 22.53 | 13.12 |
| 50 | 1.084 | 0.204 | 14.11 | 15.22 |

After reviewing the results obtained in the table above, we note that at 40 minutes, there is a decrease in removal from 30 minutes, despite repeating the experiment in the practical part and obtaining the same value, which is rare in adsorption kinetics and may

be due to the unstable equilibrium state. The highest experimental capacity was obtained at 50 minutes, so this result will be used in the kinetic calculations.

3.2.1 False first-order models (PFO)

The Lagergren equation was used, which is the most commonly used equation when two different phases, solid and liquid, are adsorbed [19], as in Table 4.

$$\ln(qe - qt) = \ln qe - K1t \quad (5)$$

Returning to Table (4), we take the highest value for the capacity, which is ($q_{e \text{ exp}} = 15.22$).

Table 4. First-order results.

| T | q_t | $\ln(qe \text{ exp} - qt)$ |
|----|--------|----------------------------|
| 10 | 11.425 | 1.425 |
| 20 | 13.66 | 0.445 |
| 30 | 14.17 | 0.049 |
| 40 | 13.12 | 0.742 |
| 50 | 15.22 | 0 |

After the graph between $(\ln(qe \text{ exp} - qt), T)$ as in figure 1.

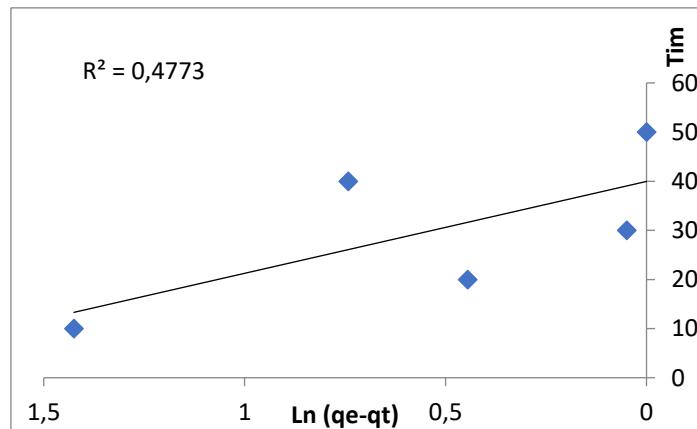


Figure 1. Graph first-order results.

Through the graph, we observe a value of (0.4773 R²), which is considered low, and the deviation of the results we obtained from the first rank.

3.2.2 False second-order PSO model

The pseudo-second-order adsorption equation also relies on the value of the solid phase capacity, as this equation can explain the adsorption behaviour, since its results are generally closer to reality. The second-order equation is represented as follows. As in Table 5.

$$t \setminus qt = \frac{1}{K2q2} \quad (6)$$

$$Ct(mg \setminus L) = Co \times \frac{\text{Abs } t}{\text{Abs } 1}$$

$$t \setminus k = 1 \setminus K2q2 + 1 \setminus qe * t \quad (7)$$

Table 5. False Second Order.

| T mint | Qe | t/q |
|--------|-------|-------|
| 10 | 11.06 | 0.904 |
| 20 | 13.66 | 1.464 |
| 30 | 14.17 | 2.117 |
| 40 | 13.12 | 3.049 |
| 50 | 15.22 | 3.285 |

After plotting the graph between $(t, t/q)$, the value $(0.97779=R^2)$ was obtained. Compared to the value we obtained in the first order, we notice a significant difference between the values, as in the second order, the value was very high. This indicates that the second-order false model explains the majority of the variance in the experimental adsorption data. Values close to 1 are a good indicator that the model is perfectly suitable. This high value confirms that the pseudo-second-order model is the most suitable model to describe the kinetics of this adsorption process. The results indicate that the mechanism controlling the adsorption rate is most likely chemisorption, which involves the sharing or exchange of electrons between the adsorbent and the adsorbate.

3.3 Effect of Temperature

Different temperatures were used in the adsorption process to determine whether the adsorption is spontaneous, endothermic, or exothermic. Where the weight of the tea leaves was 0.2g, the pesticide concentration was 75ppm, the volume was 50ml, the time was 30 minutes, and the absorbance before was 1.33. 1- Calculation of adsorption capacity at equilibrium. After using different thermal gradients, the two equations below are used.

$$Ce(mg/L) = C_0 \times \frac{\text{Abs te}}{\text{Abs nt}} \quad (8)$$

$$qe(mg/g) = \frac{(C_0 - Ce)V}{W} \quad (9)$$

To obtain the adsorption capacity and residual pesticide concentration after each adsorption cycle, as shown in the table below. As in Table 6.

Table 6. Effect of Temperature on Adsorption.

| T C° | Abs nt | Ce(mg/L) | C ₀ - C _e | qe(mg/g) |
|------|--------|----------|---------------------------------|----------|
| 30 | 0.287 | 16.19 | 58.81 | 14.70 |
| 35 | 0.277 | 15.63 | 59.37 | 14.84 |
| 40 | 0.173 | 15.40 | 59.60 | 14.90 |
| 45 | 0.177 | 9.98 | 65.02 | 16.26 |
| 50 | 0.097 | 5.47 | 69.53 | 17.38 |

When the temperature rises, we observe an increase in the adsorption capacity, and we notice that the adsorption efficiency increases with the rise in temperature. This proves that the adsorption of tea waste for the pesticide is an endothermic process, meaning that thermal energy activates the reaction and improves its efficiency [20].

3.4 Calculation of Gibbs Free Energy (ΔS , ΔH , ΔG)

The Gibbs free energy is calculated using the following equation. As in Table 7.

$$\Delta G^\circ = -R T \ln K_c \quad (10)$$

Table 7. Free Energy Calculation.

| $\Delta G^\circ(\text{kJ/mol})$ | $\ln K_c$ | $K_c (\text{L/g})$ | $q_e(\text{mg/g})$ | $C_e(\text{mg/L})$ | Temp (K) |
|---------------------------------|-----------|--------------------|--------------------|--------------------|----------|
| 0.24 | -0.096 | 0.908 | 14.70 | 16.19 | 303 |
| 0.13 | -0.052 | 0.949 | 14.84 | 15.63 | 308 |
| 0.08 | -0.032 | 0.968 | 14.90 | 15.40 | 313 |
| -1.29 | 0.488 | 1.629 | 16.26 | 9.98 | 318 |
| -3.10 | 1.156 | 3.177 | 17.38 | 5.47 | 323 |

Upon inspecting the results in the table above, we notice that the principles of Gibbs free energy at hotnesses (30, 35, 40) are definite, signifying that the backlash is non-willing and demands separation energy. As for the hotness (45, 50), we find that the principles of the Gibbs free energy are negative, that confirms that the process is spontaneous and prefers larger hotnesses. The enthalpy principles were too premeditated, and the advantage was certain (16.666 J/mol), while the deterioration profit was likewise beneficial (0.03 J/mol), which is compatible with the results in the table above, as the backlash enhances impulsive and endothermic accompanying growing hotness.

3.5 Langmuir Modell

This model assumes that the adsorption sites have uniform energy, meaning that adsorption occurs in an orderly and equal manner across the surface, and there is no transfer or exchange of sites by the adsorbed materials on the surfaces. The linear Langmuir equation can be represented by the following equation [21]. As in Table 8.

$$\frac{Ce}{qe} = \frac{1}{q_{\max} \text{KL}} + \frac{1}{q_{\max}} Ce \dots \dots \dots (11)$$

Table 8. Calculation of the Langmuir Model.

| C M | qe | C\qe |
|-----|-------|--------|
| 25 | 0.177 | 141.24 |
| 50 | 0.181 | 276.24 |
| 75 | 0.265 | 268.82 |
| 100 | 0.265 | 377.36 |
| 125 | 0.231 | 541.13 |

After using the results we obtained in the table below and the graph as in Figure 2,

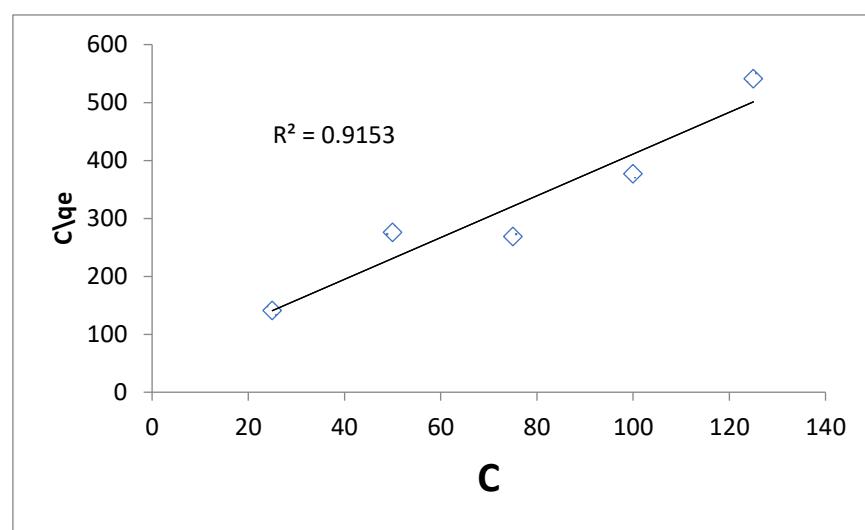


Figure 2. Calculation of the Langmuir Model.

3.6 Freundlich Model

This model assumes that the adsorption sites are unequal in their energies, allowing for multilayer adsorption [22]. The equation of this model can be represented as follows in Table 9:

$$\ln q_e = \ln K_f + \frac{1}{n} * \ln C_e \dots \dots \dots (12)$$

Table 9. Frendlich Model.

| Log q_e | log C_e | C_e/q_e |
|-----------|-----------|-----------|
| -0.752 | 1.398 | 141.24 |
| -0.742 | 1.699 | 276.24 |
| -0.554 | 1.875 | 268.82 |
| -0.577 | 2 | 377.36 |
| -0.636 | 2.097 | 541.13 |

The results obtained from the table above were used.

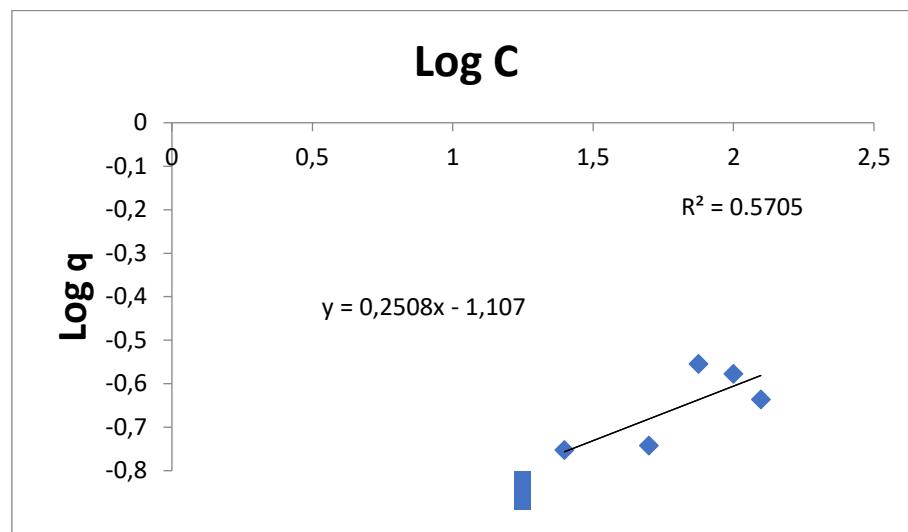


Figure 3. Freundlich Adsorption Diagram.

After comparing the values using the Langmuir method, the values were ($R^2 = 0.9153$), while in the Freundlich model, the value was ($R^2 = 0.5705$). Observing the results of the difference between the two methods indicates that the adsorption process. The final calculations were obtained using the Langmuir model, as shown below. As in Table 10

Table 10. Adsorption constants of the Langmuir.

| Parameter | Value | Significance |
|-----------|--------|---------------------------------------|
| R2 | 0.9135 | Model accuracy constant |
| qm | 0.25 | Single-layer capacity |
| KL | 0.08 | Adsorption energy |
| RL | 0.091 | (0 < RL < 1) The process is preferred |

The application of the model showed that the Freundlich constant value is very small, which suggests that multilayer adsorption is unlikely to occur extensively over the

adsorbed surface. Between the Langmuir and the Freundlich models, adsorption tends to be monolayered following the Langmuir model. As the process has proven to be highly efficient.

4. Conclusion

The results of the current study demonstrated the efficiency of using ground tea leaves waste as a low-cost and environmentally friendly adsorbent for the removal of the pesticide Tebuconazole from aqueous solutions. The main findings can be summarised as follows:

Equilibrium model: The study confirmed that the adsorption process follows the Langmuir model with a correlation coefficient ($R^2 = 0.9135$), indicating that adsorption occurs in the form of a monolayer over a surface with homogeneous energy. **Removal efficiency:** The maximum adsorption capacity (q_m) was approximately 0.25 mg/g, and the separation factor value ($R_L = 0.091$) indicated that the adsorption process is highly favoured chemically. **Chemical Kinetics:** The results indicate that the adsorption process follows the pseudo-second-order (PSO) model better, suggesting that the dominant mechanism is chemisorption, which involves the exchange or sharing of electrons between pesticide molecules and functional groups on the surface of tea leaves. **Thermodynamics:** Calculations have proven that the reaction is endothermic based on positive ΔH values, and the negative Gibbs free energy (ΔG) values at high temperatures confirm the spontaneity of the process and its increased efficiency with rising temperature.

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