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Original Research Article

Harnessing the power of agricultural waste peels: A renewable biomass for sustainable water purification

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ABSTRACT

The utilization of agricultural waste for sustainable water purification is an innovative and promising approach to addressing the global water crisis. Agricultural wastes, such as peels from fruits and vegetables, present a significant environmental challenge with the large quantities generated daily. By harnessing agricultural waste peels as a renewable biomass for water purification purposes, not only can we address the issue of waste management but also contribute to providing safe drinking water to communities in need while promoting sustainability. This study investigates the efficacy of orange peel, watermelon rind, pomegranate peel, and banana peel, in removing dyes (crystal violet and methyl orange) from water. Our objective was to determine the maximum adsorption capacities of these waste materials for each dye. The results revealed the following maximum amounts of crystal violet adsorbed by each waste material: banana peel (0.6836 mg/g), orange peel (0.8155 mg/g), pomegranate peel (0.3576 mg/g), and watermelon rind (0.6896 mg/g). Similarly, the maximum amounts of methyl orange adsorbed were as follows: banana peel (0.4727 mg/g), orange peel (0.4501 mg/g), pomegranate peel (0.4727 mg/g), and watermelon rind (0.3776 mg/g). These findings demonstrate the use of these agricultural waste materials as effective adsorbents for the removal of dyes from water.

1. Introduction

Dyes pose a significant threat to plants, aquatic life, and human beings. While they may be cost-effective to produce, certain dyes can have dangerous carcinogenic effects, including the development of cancer, allergies, and skin irritation [1]. These health risks arise from the ingestion and absorption of water contaminated with dyes. Moreover, dyes have an indirect impact on water by altering its color. This alteration reduces the transparency of the water, leading to a decline in light penetration and ultimately affecting photosynthesis. As a result, dissolved oxygen levels decrease, impacting both flora and fauna [2]. The presence of color compounds in industrial wastewater is particularly hazardous due to their intense color and poor biodegradability [3]. Even in small quantities, certain aniline dyes found in water can accumulate in fish organisms, altering the color of their meat and causing various diseases [5]. Additionally, these harmful dyes have a detrimental effect on fish plankton, with even a small amount posing a dangerous threat to algae, including methyl blue, and other species [6].

In many cases, if the concentration of dye is below 1 ppm, it will result in noticeable water discoloration. The primary source of pollution in textile wastewater is from dyeing and finishing processes [7]. The main pollutants include suspended solids, heat, color, acidity, basicity, and other inorganic contaminants [8].

There are several techniques available for removing dyes from wastewater. The most commonly used processes include chlorination, electrochemical processes, ozonation, membrane separation, coagulation, flocculation, and adsorption using various types of adsorbents [9]. Among these treatment technologies, one of the most popular methods is adsorption. In recent years, adsorption processes have proven to be highly effective in water and wastewater treatment across various industries. These techniques involve the use of natural or synthesized adsorbents to remove contaminants such as metals, dyes, and pharmaceutical products from the solution [10].

Adsorption has the advantage of producing no harmful residues and has the capacity to treat large volumes of water [11]. Additionally, adsorbents can be recycled multiple times for subsequent treatment processes. The development of adsorbents from various biomass wastes as replacements for commercial activated carbons further enhances the cost-effectiveness of the process. The application of waste organic materials, such as compounds extracted from peels, leaves, barks, and microbial biomasses (such as fungus bio-sorbents, bacterial biomasses, and green algal biomasses), is gaining increasing interest due to their effectiveness, low cost, and ecological friendliness as sorbents. The adsorption method is attractive and preferable, and carbonaceous adsorbents are



particularly suitable due to their high dye-binding capacity [10].

Several factors affect dye adsorption capacity, including initial dye concentration, pH, temperature, dosage of adsorbents and their type, contact time, etc [12]. For an efficient treatment process, the adsorbent should possess sufficient mechanical strength to withstand various wastewater conditions, have a high adsorption capacity and rapid adsorption rate, be effective against a variety of dyes or have selectivity for certain pollutants, and be easy to regenerate and reuse [13].

The adsorption process is a highly effective and cost-efficient method for removing dyes from aqueous solutions [14]. While activated carbon is commonly used as an adsorbent to separate dyes from industrial wastewater, its high cost limits its application on a large scale. However, experimental studies have shown that there are several readily available and inexpensive non-conventional adsorbents that can effectively remove dyes. Therefore, there is a growing importance in researching and identifying efficient and low-cost adsorbents derived from existing resources for dye removal [15].

Agricultural waste materials have been extensively studied for their potential to remove various dyes from aqueous solutions under different operating conditions. These waste materials are particularly valuable due to their affordability, abundance, resilience, and ability to adsorb and eliminate dyes [16]. Organic peels, seeds, and leaves from agricultural waste have demonstrated remarkable efficacy in water contaminant removal. Furthermore, a wide range of materials have been utilized, prepared, or derived from different agricultural peel-based adsorbents. Watermelon, coconut shells, orange peel, banana peel, pomegranate peel, tea waste, and pistachio peel are among the most commonly used sources for dye removal [17]. The main components of agricultural waste, such as hemicellulose, lignin, lipids, proteins, simple sugars, water, hydrocarbon, and starch, contain various functional groups. These functional groups contribute to an increase in active sites and enhance the adsorption of pollutants [18]. Some studies have been conducted and reported previously on the use of agricultural waste peels for dye adsorption in water purification processes. The study conducted by Khatod [19] examined the efficacy of orange peel powder in adsorbing Methylene blue. It was found that the dye adsorption decreased as the concentration of dye increased. This decrease was attributed to the agglomeration of the biosorbent, resulting in less surface area available for the adsorption process. The effectiveness of orange peel in removing RB dye from water was tested in a study [20]. The maximum adsorption capacity was observed at a pH of 8-9, with an adsorbent dose of 50 g/L (particle size of 1200 μm), agitation speed of 160 rpm, and a contact time of 55 minutes. In a study conducted by Boumediene et al. [21], the efficacy of orange peel in removing MB was evaluated. Batch experiments were carried out under specific conditions (initial parameters: biosorbent mass = 1 g, V = 1 L of dyeing solution; T = 25 ± 1 °C, agitation rate of 400 rpm) to analyze the kinetics, isotherms, and thermodynamics of the process. Moubarak et al. [22] conducted a study on the adsorption of Methylene Blue onto banana peel powder. The study

examined the impact of particle size (d b 80 μm and 80 μm b d b 2 mm), solution temperature (22 and 50 °C), and the mass of biosorbents used. The results showed that the highest adsorption occurred with particle size d b 80 μm and 0.1 g of adsorbent mass. Interestingly, the temperature did not affect the amount of dye uptake. This research sheds light on the potential of banana peel powder as an effective adsorbent for removing Methylene Blue from solutions. Khalfaoui et al. [23] conducted a study on the removal of Methylene Blue (MB) using raw banana peels and activated banana peels treated with NaOH. The research revealed that the adsorption of the dye increased with higher pH levels (tested between 2 and 10) and initial dye concentrations (ranging from 5 to 100 mg/L) during equilibrium tests. Ahmad et al. [24] successfully converted pomegranate peel into activated carbon using microwave irradiation and KOH activation techniques. This activated carbon was then utilized for the adsorption of remazol brilliant blue reactive (RBBR) dye and achieved an impressive 94.36% dye removal efficiency at pH 2. In their study, Usman et al. [25] documented the adsorption of aniline blue dye on activated pomegranate peel. The Langmuir model revealed a maximum adsorption capacity of 27.322 mg/g at 30°C using 0.1 g of activated carbon derived from pomegranate peel. Ibrahim et al. [26] investigated the efficacy of watermelon rind in removing Congo red (CR) dye from wastewater, yielding promising results. Their findings indicated that the biosorption of CR into watermelon rind was primarily driven by physisorption. In their study, Shukla et al [27] documented the adsorption of methylene blue from an aqueous solution using watermelon rind. They reported a maximum adsorption capacity of 115.61 mg/g.

The primary objective of this study is to identify bioavailable and cost-effective adsorbents for the removal of dye compounds from industrial wastewater. Additionally, this research aims to present the physical and chemical adsorption processes, as well as the parameters that influence adsorption capacity. The study collects data on adsorption kinetics, isotherm models, thermodynamics, and adsorption capacity under various conditions. By doing so, it provides comprehensive and up-to-date information on the absorption of different colors from aqueous solutions using a wide range of adsorption techniques.

2. Materials and methods

2.1 Materials used

All chemicals were purchased from commercial vendors and used in their original form. Crystal violet and methyl orange were obtained from Himedia Industries Pvt. Ltd., located in Mumbai. Methyl alcohol was purchased from Merck Lifesciences Pvt. Ltd., Mumbai. The peels of oranges, pomegranates, bananas, and watermelons were obtained from their respective fruits.

2.2 Methods

Preparation of biosorbent powder: Four samples were collected from agricultural waste for color removal: orange peel, watermelon rind, banana peel, and pomegranate peel. The samples were washed several times with distilled water to remove adhered impurities. Subsequently, these samples were dried in a hot air oven at 50°C, powdered using a mixer

grinder, and utilized without the need for additional purification. Pomegranate peel powder is abbreviated as PG, orange peel powder as OP, banana peel powder as BP, and watermelon rind powder as WR. Images of powdered samples are given in Figure 1.

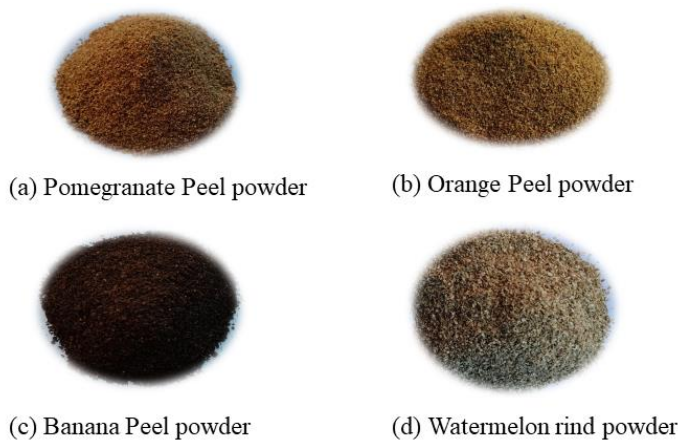


Figure 1: Various biosorbents collected for the study in powdered form.

Morphological analysis: The morphology of the powdered samples was analyzed using a Field Emission Scanning Electron Microscope (FESEM). Specifically, a Carl Zeiss Sigma scanning electron microscope from Germany was employed for this purpose, operating at a voltage of 5kV. The samples were scanned at a magnification of 25kX. Prior to the microscopic analysis, a thin layer of gold was applied to the samples under vacuum conditions.

Dye adsorption studies: To assess the adsorption mechanism of dye on adsorbent systems, two kinetic models, namely the pseudo-first-order and pseudo-second-order models, have been utilized. Two main types of adsorption isotherm models, namely the Freundlich [29] and Langmuir, have been widely employed to comprehend the nature and mechanism of the adsorption process. The thermodynamic parameters, such as the changes in standard free energy (ΔG°), enthalpy (ΔH°), and entropy (ΔS°) associated with the adsorption process, were also calculated.

3. Results and discussion

3.1 Morphological analysis

FESEM images of the samples are shown in Figure 2. The morphology of the samples was found to possess a granular

nature indicating the increased surface area on the bioadsorbents. This granular nature will enhance the adsorption efficiency providing more surface area for the adsorbate molecules to occupy.

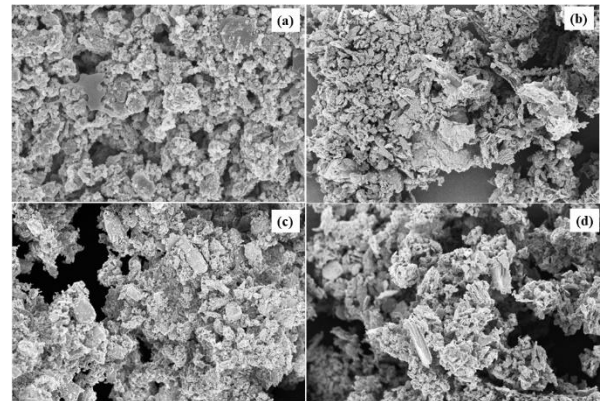


Figure 2: FESEM images of powders of (a) OP, (b) PG, (c) WR, and (d) BP.

3.2 Adsorption of crystal violet (VC) and methyl orange (MO) onto different biosorbents

The prepared bio-adsorbents were examined for their ability to adsorb the cationic dye, crystal violet, and anionic dye methyl orange from an aqueous solution.

3.3 Kinetics of adsorption

These studies aimed to find the optimal kinetic model for the adsorption process of CV and MO. The pseudo-first-order and pseudo-second-order kinetic models were carefully assessed.

The values of Q_e and k_1 are derived from the linear plot of $\log(Q_e - Q_t)$ versus time, which represents the Pseudo-first order kinetics. Similarly, the values of Q_e , k_2 , and h are obtained from the plot of t/Q_t versus t , representing the Pseudo-second order kinetics. These values are comprehensively presented in Table 1 for CV and Table 2 for MO.

Based on analysis, it has been determined that the adsorption process adheres to a pseudo-first-order model. This conclusion is drawn due to the highest value of the regression coefficient (R^2) obtained in this model. Furthermore, the calculated Q_e value closely aligns with the experimental value. Hence, we can confidently state that the adsorption of both CV and MO on all the bio adsorbents strictly follows pseudo-first-order kinetics, rather than pseudo-second-order kinetics.

Table 1: Kinetic parameters for the adsorption of CV using various biosorbents

Adsorbent	1 st order			2 nd order			
	Q_e (mg/g)	k_1 (min ⁻¹)	R^2	Q_e (mg/g)	k_2 (g/mg/min)	h (mg/g/min)	R^2
BP	0.6836	0.0118	0.9988	0.8843	0.0232	0.0182	0.9957
OP	0.8155	0.0112	0.9931	0.9869	0.0150	0.0146	0.9858
PG	0.3576	0.0091	0.9937	0.4713	0.0285	0.0063	0.9928
WR	0.6896	0.0096	0.9564	0.8223	0.0173	0.0117	0.9492

Volume: 50 mL; Co: 5 mg/L; Weight of adsorbent: 250 mg; pH: 7.0; Temperature: 30°C

Table 2: Kinetic parameter for the adsorption of MO using various biosorbents

Adsorbent	1 st order			2 nd order			
	Q_e (mg/g)	k_1 (min ⁻¹)	R^2	Q_e (mg/g)	k_2 (g/mg/min)	H (mg/g/min)	R^2
BP	0.4727	0.0078	0.9813	2.8488	0.0002	0.0017	0.0117
OP	0.4501	0.0086	0.9974	0.5918	0.0203	0.0071	0.9930
PG	0.4727	0.0079	0.9813	0.6065	0.0123	0.0045	0.9499
WR	0.3776	0.0067	0.9729	0.7255	0.0035	0.0019	0.1174

Volume: 50 mL; Co: 5 mg/L; Weight of adsorbent: 250 mg; pH: 7.0; Temperature: 30°C

3.4 Adsorption isotherm studies

Two primary models for adsorption isotherms, specifically the Freundlich and Langmuir models, have been utilized to examine the adsorption process. The Freundlich model proposes that the adsorption capacity relies on the equilibrium concentration of the dye and occurs on non-uniform surfaces. In contrast, the Langmuir model suggests that the maximum adsorption happens when a saturated monolayer of the adsorbate forms on the adsorbent's surface [31]. The isotherm parameters for CV can be found in Table 3 while those for MO are listed in Table 4. These tables provide

evidence of a strong correlation between the adsorption process and the Freundlich adsorption isotherm model, suggesting a multilayer adsorption process. The irregular R_L values that do not fit between 0 and 1 indicate that the adsorption is unfavourable with regards to the Langmuir isotherm model suggesting a multilayer adsorption process. The driving force behind this adsorption is the Van der Waals force of attraction between the adsorbent and adsorbate molecules. Consequently, it can be inferred that the adsorption does not occur through the formation of a chemical bond between the adsorbent and adsorbate [30].

Table 3: Isotherm parameters for the adsorption of CV using various biosorbents

Adsorbent	Freundlich			Langmuir			
	K_F	n	R^2	Q_m (mg/g)	K_L	R_L	R^2
BP	0.3906	1.0060	0.9940	73.4549	-0.0052	1.0553	0.0157
OP	0.2372	1.4290	0.9973	1.7230	0.1489	0.4017	0.9081
PG	0.1517	0.9996	0.9926	27.0081	-0.0055	1.0583	0.0318
WR	0.6009	1.0054	0.9964	40.9788	0.0150	0.8698	0.0970

Volume: 50 mL; Weight of adsorbent: 250 mg; pH: 7.0; Temperature: 30°C

Table 4: Isotherm parameters for the adsorption of MO using various biosorbents

Adsorbent	Freundlich			Langmuir			
	K_F	n	R^2	Q_m (mg/g)	K_L	R_L	R^2
BP	0.0134	1.5193	0.9507	0.1002	0.1332	0.4288	0.9446
OP	0.1170	2.7669	0.9947	0.3109	0.4631	0.1776	0.9909
PG	0.0879	1.3765	0.9987	0.8976	0.0959	0.5105	0.9506
WR	0.03629	1.0277	0.9607	1.9457	0.0196	0.8362	0.1424

Volume: 50 mL; Weight of adsorbent: 250 mg; pH: 7.0; Temperature: 30°C

3.5 Thermodynamics of adsorption

The adsorption process was conducted at five distinct temperatures (303 K, 308 K, 313 K, 318 K, and 323 K) to examine the adsorption properties under varying temperature conditions. To evaluate the thermodynamic performance of the adsorption process, a graph depicting the correlation between $\ln K_c$ and $1/T$ has been generated.

Thermodynamic parameters, including ΔG , ΔH , and ΔS , have been computed from the plot and are provided in Table 5 for CV and Table 6 for MO. The remarkably positive ΔG

value indicates that the adsorption process is non-spontaneous across all temperatures investigated. Moreover, the positive ΔS value suggests a degree of disorder during the adsorption process, implying that the adsorbate molecules move randomly and occupy different adsorbent sites. Also, the positive value of ΔH suggests an endothermic reaction that uses temperature during the adsorption process. These findings shed light on the thermodynamic aspects of the adsorption phenomenon, highlighting its feasibility and the dynamic nature of the adsorbate-adsorbent interaction.

Table 5: Thermodynamic parameters for the adsorption of CV using various biosorbent

Adsorbent	Thermodynamic parameters		
	ΔG (kJ/mol)	ΔH (kJ/mol)	ΔS (kJ/mol/K)
BP	2.8346	39.0937	0.1158
OP	3.5481	51.2447	0.1524
PG	4.9113	44.9493	0.1279
WP	2.4291	57.7280	0.1767

Volume: 50 mL; Weight of adsorbent: 250 mg; pH: 7.0; Co: 5 mg/L

Table 6: Thermodynamic parameters for the adsorption of MO using various biosorbents

Adsorbent	Thermodynamic parameters		
	ΔG (kJ/mol)	ΔH (kJ/mol)	ΔS (kJ/mol/K)
BP	8.1140	91.4288	0.2662
OP	6.5965	88.6701	0.2622
PG	8.8170	79.3860	0.2255
WP	11.4026	142.7951	0.4198

4. Conclusions

The presence of dyes in wastewater poses a significant threat to both environmental conservation and human health. Certain dyes are known to be toxic, mutagenic, and even carcinogenic. To address this issue, researchers have explored the use of low-cost adsorbents derived from agricultural waste materials to remove these dyes from aqueous solutions. Remarkably, agricultural waste materials such as orange peel, watermelon rind, pomegranate peel, and banana peel have demonstrated exceptional efficacy in eliminating dyes from water. Furthermore, it has been observed that the efficiency of dye removal increases with higher dosages of adsorbents. This can be attributed to the greater availability of sorption sites for the adsorption of dye molecules, which increases as the dose of adsorbent is increased. To determine the most suitable kinetic model, all the materials were subjected to rigorous testing. The results revealed that the adsorption process adheres to the pseudo-first-order kinetic model. Additionally, it was found that the adsorption process reaches saturation after 420 minutes. Isothermal studies further indicated that the adsorption process follows the Freundlich isotherm model, suggesting multilayer adsorption facilitated by weak forces of attraction between the dyes and adsorbent material. Moreover, the temperature-dependent adsorption behaviour demonstrated that the process is favourable across all tested temperatures. The maximum amounts of CV adsorbed by banana peel, orange peel, pomegranate peel, and watermelon rind were determined to be 0.6836 mg/g, 0.8155 mg/g, 0.3576 mg/g, and 0.6896 mg/g, respectively. Similarly, the maximum amounts of MO adsorbed by banana peel, orange peel, pomegranate peel, and watermelon rind were found to be 0.4727 mg/g, 0.4501 mg/g, 0.4727 mg/g, and 0.3776 mg/g. The removal efficiency of each adsorbent for both CV and MO were observed to be more than 90% in 6hrs.

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