A Short Review on the Removal of Rhodamine B Dye Using Agricultural Waste-Based Adsorbents

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Authors’ contributions

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ABSTRACT

Among the various methods available for wastewater remediation, the adsorption technique remains one of the most preferred choices worldwide. This is because the process is effective and economically feasible. Over the years, the abundance and availability of agricultural waste products have encouraged their application in pure or modified forms as catalysts in environmental remediation. Such agricultural wastes are carbonaceous and lignocellulosic in nature with numerous surface functional groups which makes them favorable for use as adsorbents for the removal of various pollutants. Rhodamine B (RhB) dye is a common organic dye in industrial wastewaters but could be artificially removed via adsorption. In this article, the use of agricultural-waste based adsorbents for the removal of RhB dye from aqueous solution over the last decade have been reviewed.

Keywords: Adsorption; agricultural wastes; Rhodamine B dye; wastewater treatment.
1. INTRODUCTION

Rhodamine B (RhB) dye (Table 1) is a highly water soluble basic organic dye belonging to the xanthene class, which is chemically stable and non-biodegradable [1,2]. It is a list I inert ingredient and exists in three monomeric molecular forms of cation, zwitterion and lactone, but the lactone form has no contribution to the color [3,4]. RhB is widely used as textiles colorant, as stains in biochemical researches and as an illegal food additive [5,6]. It is also used for preparing paints, dye lasers, carbon papers, ball pens, stamp pad inks, dyeing mosquito coils, fireworks, crackers and in leather industry [7-10]. All these contributed towards detection of RhB dye in wastewaters.

### Table 1. Molecular structure and some physicochemical properties of RhB

| Property                     | RhB                      |
|------------------------------|--------------------------|
| Chemical formula             | C_{29}H_{27}O_{5}N_{3}Cl  |
| IUPAC name                   | [9-(2-carboxyphenyl)-6-diethylamino-3-xanthenylidene]-diethylammonium chloride |
| Class                        | Rhodamine                |
| C.I. Name                    | Basic violet 10          |
| Suggested Name               | Rhodamine B              |
| C.I. Number                  | 45170                    |
| CAS number                   | 81-88-9                  |
| λ_{max} (nm)                 | 554                      |
| Solubility                   | 5 g in 100 mL            |
| Ionization                   | Basic                    |
| Chemical structure           | ![Chemical structure](image) |

Over the past few decades, several physical, chemical and biological methods have been applied for the decolorization of dyes. Among the techniques employed, adsorption is widely accepted and has a high potential for the removal of dyes [11]. Other advantages of the adsorption process are; low cost, ease of operation, sludge free operation and recovery of sorbate [12]. One of the famous adsorbents used worldwide due to its high adsorption capacity, extended surface area and high degree of surface reactivity, is activated carbon. However, for some reasons such as high cost of purchase and regeneration after exhausting, the search for cheaper alternatives becomes imperative [13].

Non-product outputs from agriculture and agro-based industries such as rice husk, groundnut shell, coconut shell and egg shell generated during growing and processing of raw agricultural products are produced in huge amounts. This makes their disposal a major ecological and economic issue. In view of that, the conversion of such agricultural wastes to adsorbents could not only help reduce disposal problem but might also be a potential cheaper alternative to the commercially available adsorbents.

2. TOXICITY OF RhB DYE

RhB dye is carcinogenic, neurotoxic and causes skin, eyes, respiratory tract and gastrointestinal tract irritation, with developmental and reproductive toxicity in both humans and animals [1,14-16]. It also causes burning sensation, laryngitis, coughing, shortness of breath, wheezing, vomiting, nausea, chest pain, headache, subcutaneous tissue borne sarcoma and can form equivocal tumor [17-20]. It increases hypothalamic cell apoptosis and have
been reported to cause disruption of hormonal balance [21]. It may also cause teratogenic effects in humans, is toxic even in trace quantities and fatal in higher doses [22,23].

RhB also endangers aquatic life by inhibiting light penetration, leading to reduced photosynthesis [24]. It is also toxic to fish, as toxicity test reported LC50 of 83.9 mg/L for Cyprinodon variegatus [25]. The zooplankton and phytoplankton numbers were also found to be less in water polluted with RhB [23]. Through animal studies, RhB has been found to induce lymphoma and is poisonous to a fetus [18]. In fact, the toxicity of RhB towards human and animals has been proved experimentally [6].

3. REMOVAL OF RhB DYE USING VARIOUS AGRICULTURAL WASTE – BASED ADSORBENTS

Numerous agricultural waste – based adsorbents have over the past decade been used by different researchers for the removal of RhB dye from aqueous solution and will be discussed in the current section. Furthermore, the adsorption capacity of the sorbents has been summarized in Table 2.

Arivoli et al. [26] prepared activated carbon via acidic treatment of banana bark. The adsorbent was found to have high potential for the removal of RhB dye from aqueous solution via physisorption mechanism. Adsorption using the adsorbent was minimum at pH of 8 and maximum at pH of 6.5, however, its performance increased with increase in ionic strength and temperature and attains equilibrium in 40 min. Bhadusha and Ananthabaskaran [17] also reported the use of ZnCl2 activated wood apple carbon (ZAWAC) as adsorbent for the removal of RhB from wastewater. RhB was found to adsorb strongly on the surface of the activated carbon and the experimental data fitted well with the Langmuir isotherm and second order kinetics model.

Gad and El-Sayed [27] produced activated carbon from bagasse pith (BP) via chemical activation using H3PO4 (BPH) and KOH (BPK), followed by carbonization at 500°C. Both BPH and BPK were applied for the removal of RhB dye from aqueous solution, with maximum performance recorded at pH of 2.45 and equilibrium attained in 240 min. The adsorption of Rhodamine B dye by BPH was almost 10 folds time higher than that by BPK, and both processes followed the pseudo-second-order kinetics and Langmuir adsorption isotherm. The use of jute stick powder (JSP) for the removal of Rhodamine B dye from aqueous solution has been reported by Panda et al. [24]. Adsorption using JSP reaches equilibrium in 60 min and fitted well with the pseudo-second-order kinetics model and Freundlich adsorption isotherm. The removal was found to be favorable at around pH 7, while temperature had no significant impact on the process. Furthermore, Parab et al. [28] also made use of coconut pith (CP) and sugarcane fiber (SF) for the removal of RhB from aqueous solution. Although CP was found to have higher adsorption capacity for the removal of RhB compared to SF, however, both adsorbents gave a promising performance. The experimental data using both adsorbents fitted well with the second-order kinetics and the non-linear Freundlich adsorption isotherm.

The use of loquat seeds (LS) sorbent for the removal of RhB from aqueous solution using the conventional mechanical stirring method and a combined method of mechanical stirring and ultrasonic irradiation have been reported by Hamdaoui [10]. The LS was found to be efficient for the removal of RhB, and both rate and amount of RhB sorption by LS significantly improved in the presence of ultrasound. The sorption equilibrium data for the conventional and combined methods were best represented by the Langmuir and Radlich-Peterson isotherms. Apart from the LS sorbent, Hossain and Alam [29] reported the applicability of used black tea leaves (UBTL) for the removal of RhB from aqueous solution. Maximum performance was recorded at pH of 2, due to the zero-point charge pH of UBTL (pH_{zpc} = 4.2), indicating its adsorptive characteristics of negative species at low pH. The overall rate of RhB dye adsorption was found to be chemical reaction controlled.

Polygonum orientale Linn (PLAC) was also used by Wang et al. [30] to prepare low cost activated carbon via phosphoric acid activation. The produced PLAC was found to have a surface area of 1398 m²/g and exhibited high RhB dye uptake at acidic pH. The equilibrium time was found to be 240 min, and the experimental data fitted well with the pseudo-second-order kinetic model and the Langmuir adsorption isotherm. Thermodynamic study revealed that the adsorption process was spontaneous and endothermic. Furthermore, Leucaena leucocephala seed shell waste was also used by Karthikeyan and Rajendran [31] to prepare
activated carbon via carbonization with sulphuric acid, and subsequently used as adsorbent for the removal of RhB dye from aqueous solution. The material was found to have a BET surface area of 499.23 m$^2$/g, and 100 mg was able to remove 93.4% of 60 mg/L RhB, at 30°C and pH ~ 7, and in agitation time of 240 min. The whole process was endothermic and was best described by the pseudo-second-order kinetic model.

Acid activated mango leaf powder (MLP) has been used by Khan et al. [16] for the removal of RhB dye from aqueous solution. The optimal uptake occurred at pH of 6, as 25 g/L of MLP was able to remove 77% of 250 mg/L RhB dye solution within 45 min and 30°C temperature. Below pH of 6, the dye ions were able to enter the pore structure of the MLP surface, whereas at higher pH's beyond 6, the dimer formed was unable to enter the pores. The adsorption was best described by the Langmuir adsorption model and the kinetics fitted well with the pseudo-first-order kinetics model. In another study, Ahamed et al. [32] made use of activated carbon prepared via acid treatment of Azadirachta Indica bark (AIC), and used it as adsorbent for the removal of RhB. The material was found to have a pH$_{zpc}$ of 5.75 and the whole adsorption process was pH dependent, with minimum and maximum uptake recorded at pH of 7.5 and pH 3.0 – 6.0. Moreover, the amount of RhB adsorbed increased with increase in ionic strength and temperature, and the experimental data correlated well with the Langmuir and Freundlich adsorption isotherms.

The use of activated carbon prepared from rice husk having a specific surface area of 518 m$^2$/g for the removal of RhB from aqueous solution has been reported by Yadav et al. [33]. The percentage adsorption increases with increase in temperature, and equilibrium was established in 120 min. Furthermore, the total amount of RhB adsorbed per unit mass of the rice husk activated carbon was found to increase with increase in initial RhB concentration, through a process that was endothermic and controlled by entropy gain. Another low cost activated carbon (TPAC) was also prepared by Baseri et al. [34] via chemical impregnation of Thevetia peruviana wood with $H_3PO_4$ acid and used as adsorbent for the removal of RhB. The percentage removal of RhB using TPAC decreases from 97.33 to 87.12% with increase in initial dye concentration from 25 mg/L to 100 mg/L at 30°C. Kinetic studies revealed that, the adsorption of RhB onto TPAC followed the pseudo-second-order model, with multi-step intra-particle diffusion model. The sorption process was spontaneous, endothermic and fitted well with Langmuir isotherm model.

Waste fruit residues (FR) obtained from fruit juice processing industry were treated with phosphorous (V) oxy chloride and used as adsorbent (PFR) by Parimaladevi and Venkateswaran [35] for the removal of RhB from aqueous solution. The prepared PFR material was found to have a surface area of 310 m$^2$/g and a pH$_{zpc}$ of 5. Adsorption of RhB onto PFR involved chemisorption mechanism and was best described pseudo-second-order kinetics model. Furthermore, the adsorption was well described by the Langmuir isotherm and fairly by the Freundlich isotherm, in a process which was found to be spontaneous and endothermic. Nano-porous Jack fruit peel was also used by Jayarajan et al. [36] for the removal of RhB. The adsorption of RhB was found to increase with increase in adsorbent dosage, and maximum percentage removal of 25.3 g/L was obtained at RhB initial concentration of 100 mg/L using adsorbent dosage of 3.0 g/L.

Auta [37] also reported the use of activated carbon prepared using oil palm empty fruit bunch for the fixed bed adsorption studies of RhB. The experimental data was modelled using the Thomas model, and the result of the model was in good agreement with the experimental data. Another activated carbon was also produced by Venugopal and Kannan [23] via chemical activation of Prosopis juliflora seeds with sulfuric acid, for subsequent use as adsorbent for the removal of RhB from aqueous solution. The adsorption showed a linear increase with increase in concentration of the adsorbent and maximum removal was recorded at pH of 10. The experimental data was well defined by the pseudo-first-order kinetics and Langmuir monolayer adsorption isotherm model.

The use of sorbent prepared using cedar cone waste obtained from forests to remove RhB has been reported by Zamouche and Hamdaoui [38]. The sorption of RhB by CC was found to be pH dependent, and maximum performance was recorded at pH of 3, at an estimated contact time of 360 min. Ionic strength had no effect on the sorption of RhB by CC, and no strong competition between the sorbate and the solvent in occupying the sorbent sites was noticed.

The use of activated carbon produced from elephant dung for the removal of RhB has also
been reported by Theivarasu and Chandra [39]. The activated carbon was produced via treatment of the elephant dung with concentrated sulphuric acid, and was found to have a $pH_{zpc}$ of 3.2 and BET surface area of 23.13 m$^2$/g. Maximum RhB uptake was recorded at pH of 3, and equilibrium time was found to be 240 min. The experimental data shows that the sorption process followed the pseudo-second-order kinetic model and Langmuir adsorption isotherm. The adsorption process was endothermic and spontaneous. In another study, Ilayaraja et al. [40] also made use of chemical activation to prepare Maranta arundinacea carbon (MAC) for subsequent use as adsorbent for the removal of RhB from aqueous solution. The material was found to have a surface area of 22.43 m$^2$/g and an optimal 40 min removal time for RhB. Experimental data for the removal of RhB by MAC followed the pseudo-second-order and D-R adsorption isotherm model. The adsorption process was spontaneous and exothermic.

Water hyacinth plant stem was also used by El-Wakil et al. [41] to prepare activated carbon (ACS) via chemical activation using H$_3$PO$_4$. The performance of ACS towards removing RhB was high at normal pH and the experimental data fitted well with the Langmuir isotherm model. Kinetic studies revealed that the adsorption process followed the pseudo-second-order model and the involvement of intra-particle diffusion was also confirmed. The adsorption process was spontaneous and exothermic in nature. In another study, Inyinbor et al. [42] reported the use of cysteine modified Raphia hookerie (CMRH) as adsorbent for the removal of RhB from aqueous solution. CMRH had a $pH_{zpc}$ value of about 6 and maximum RhB adsorption was recorded at pH of 3. The adsorption capacity of CMRH decreases from 85 to 69% with increase in temperature from 303 to 323 K. The experimental data for the adsorption of RhB by CMRH followed the Freundlich adsorption isotherm and pseudo-second-order kinetics model.

Ashoka leaf powder (ALP) was also used by Gupta et al. [43] for the removal of RhB from aqueous solution. The adsorption was found to be spontaneous, feasible and exothermic in nature, in a process that was well described using the pseudo-second-order kinetic model and Freundlich isotherm. Shah et al. [10] made use of adsorbent prepared by pretreatment of walnut shell (WS) for the removal of RhB from aqueous solution. The removal process was pH dependent and best performance was achieved at pH 3. The experimental data fitted well with the pseudo-second-order kinetic and Langmuir isotherm model. Adsorption process of RhB using WS was endothermic and spontaneous.

Animal bone meal prepared by El Haddad et al. [44] was also used as adsorbent for the removal of RhB from aqueous solution. Removal of RhB using animal bone meal was rapid, as equilibrium was attained in 60 min. Thermodynamic studies conducted revealed that, the adsorption of RhB using animal bone meal is spontaneous and endothermic in nature. In a different study, Aliabadi et al. [45] reported the use of almond shell (Prunus dulcis) biosorbent for the removal of RhB from aqueous solution. The process was pH dependent and maximum performance was recorded at pH of 2. The RhB uptake process by the adsorbent was rapid within the first 15 min and 2 h of contact was enough to attain equilibrium. The percentage removal of RhB decreases from 92.2 to 75.7% with increase in initial concentration from 5 to 200 mg/L and experimental data fitted well with the Freundlich isotherm model.

An agricultural biowaste, water chestnut peel (WCPW) has been applied by Khan et al. [46] for the removal of RhB from aqueous solution. The WCPW was found to have a $pH_{zpc}$ of 5.8 and maximum RhB adsorption occurred at pH 9.0. The experimental data for the removal of RhB using WCPW fitted well with the pseudo-second-order kinetic, Freundlich and Halsey isotherm models. The adsorption process was spontaneous and endothermic. Pigeon dropping has also been used by Kaur and Kaur [7] for the removal of RhB from aqueous solution. The experimental data fitted well with the pseudo-second-order kinetic and Freundlich isotherm models. The adsorption of RhB using pigeon dropping was found to be exothermic and spontaneous.

Sugarcane bagasse of different surface areas were used by Zhang et al. [47] for the adsorption of RhB, although the gain in amount of RhB removed by increasing the surface area from 0.57 m$^2$/g to 1.81 m$^2$/g was small. The bagasse had a $pH_{zpc}$ of about 5, however, the adsorption of RhB by the bagasse was to some extent independent of pH. The experimental data followed the pseudo-second-order kinetics and Freundlich isotherm model. The adsorption process was exothermic and spontaneous. In a different study, Cao et al. [48] made use of raw
corn stalk (CS) to produced citric acid modified corn stalk (CCS). The chemical modification resulted in the introduction of carboxylic groups into the raw corn stalk. The adsorption capacity of the CCS was 203.0 mg g\(^{-1}\) while that of CS was 58.71 mg g\(^{-1}\). The experimental data fitted well with the Langmuir adsorption isotherm and pseudo-second-order kinetics model.

Raw dika nut (DN) was modified via acidic treatment by Inyinbor et al. [49] to produce acidic – treated dika nut (AND). The formed AND was found to have a \(p_{\text{H}_{\text{zpc}}}\) of 6.60 and optimum performance was recorded at pH of 3, with removal efficiency of 97.05%. The experimental data fitted well with the Langmuir adsorption isotherm, pseudo-second-order kinetics and multi-linear plot was observed for intraparticle diffusion studies. The adsorption process was spontaneous and feasible. In a different study, Inyinbor et al. [50] again made use of Raphia hookerie (RH) fruit epicarp for the removal of RhB dye from aqueous solution. The uptake was rapid between pH of 2 and 3 but decreased gradually above pH 3.2 g/L of RH was able to remove 88.88% of 100 mg/L Rh B dye within 60 min and at 26°C. The adsorption process was found to fit the Freundlich isotherm and pseudo-second-order kinetics model.

The waste product of medicinal plant (Jatoba bark) processing was applied by Demarchi et al. [51] for the removal of RhB from aqueous solution in a batch system. Jatoba bark was found to have a \(p_{\text{H}_{\text{zpc}}}\) of 6.25 and a specific surface area of 124.25 m\(^2\)/g. The performance of Jatoba bark towards RhB adsorption was found to be pH dependent, as increase in pH from 2 to 10 resulted in increase in RhB adsorption from 20 to 35 mg/g. The experimental data fitted well with the pseudo-second-order kinetics and the Langmuir-Freundlich isotherm model. The adsorption process was spontaneous and endothermic. Chieng et al. [52] also collected peat from Brunei Darussalam and made use of it as adsorbent for the removal of RhB from aqueous medium. The adsorption capacity of peat was found to increase with increase in temperature and no pH adjustment was needed throughout the adsorption process. The experimental data fitted well with the pseudo-second-order kinetics and the Sips isotherm model. The adsorption process was feasible, spontaneous and endothermic in nature.

Li [53] made use of pummelo peel obtained from a local fruit market of Changchun in China, as adsorbent for the removal of RhB from aqueous solution. The surface of pummelo peel was negative, and the effective adsorption capacity of RhB decreased significantly with increase in pH. The experimental data fitted well with the pseudo-second-order kinetics and the Langmuir isotherm model. The production of activated carbon using seed husk of Moringa oleifera has been reported by Harouna et al. [54], for subsequent use as adsorbent for the removal of RhB from aqueous solution. During application, adsorption equilibrium was established in 40 min and maximum performance was recorded at pH 7. The experimental data fitted well with the pseudo-second-order kinetics and the Langmuir isotherm model.

The use of low-cost adsorbent obtained from waste seeds of Aleurites moluccana (WAM) as for the removal of RhB from aqueous solution has been reported by Postai et al. [55]. The \(p_{\text{H}_{\text{zpc}}}\) of WAM was found to be 5.84 and increase in pH from 3 to 9 was accompanied by an approximately three-fold increase in amount of dye adsorbed. The experimental data fitted well with the pseudo-second-order kinetics and the Sips isotherm. The adsorption of RhB using WAM was spontaneous and endothermic. Lim et al. [18] also reported the use of the skin of a locally abundant fruit, Artocarpus odoratissimus, otherwise known as Tarap skin (TS) for the removal of RhB from aqueous solution. TS showed a strong removal ability towards RhB at its ambient pH of 4.15, and optimum contact time was found to be 210 min. Thermodynamic studies revealed that the adsorption process is endothermic and spontaneous in nature. The experimental data fitted well with the Langmuir isotherm.

Kooh et al. [56] reported the use of another low-cost adsorbent Casuarina equisetifolia needle (CEN) as adsorbent for the removal of RhB from aqueous solution. The material had a \(p_{\text{H}_{\text{zpc}}}\) of 4.4 and maximum performance was recorded at pH 2.9. Adsorption of RhB by CEN was endothermic in nature and the experimental data fitted well with the pseudo-second-order kinetics and Langmuir isotherm model. In a different study, Dahri et al. [57] made use of Casuarina equisetifolia cone (CEC) as an adsorbent for the removal of RhB from aqueous solution. Maximum adsorption was recorded at pH of 2, although adsorption of RhB onto CEC was independent of the electrostatic interactions. The experimental data fitted well with the pseudo-second-order kinetics and Langmuir isotherm.
model, in a process that was thermodynamically favorable, feasible and endothermic.

Untreated *Azolla pinnata* (AP) obtained from Brunei Darussalam was used by Koooh et al. [25] for the removal of RbB from aqueous solution. The material was found to have a pH<sub>zpc</sub> of 6.6 and maximum adsorption was recorded at pH 3.6. The experimental data fitted well with the pseudo-second-order kinetics and Langmuir isotherm model. The adsorption process was spontaneous, endothermic and physisorption-dominant in nature. El Haddad et al. [58] made use of Animal Bone Meal (ABM) as a low-cost adsorbent for the removal of RbB from aqueous solution. The material was found to have a pH<sub>zpc</sub> of 8.4, and adsorption equilibrium was rapidly attained after 60 min of contact time. The experimental data fitted well with the Langmuir isotherm and the adsorption process was a physical reaction, spontaneous and endothermic.

Huang et al. [59] made use of *Lythrum salicaria* L (LS) obtained from Shandong China to produce activated carbon (LSAC). LSAC was found to have a pH<sub>zpc</sub> of 5.94 and maximum RbB dye adsorption was recorded at pH of 3. The adsorption was dependent on contact time and equilibrium was attained in 480 min. The experimental data fitted well with the pseudo-second-order kinetics and Langmuir isotherm model. In a different study, Koooh et al. [60] reported the use of jackfruit seeds (JS) as low-cost adsorbent for the removal of RbB from aqueous solution. The material was found to have pH<sub>zpc</sub> of 3.75 and higher dye adsorption was recorded at pH values above pH<sub>zpc</sub>. The experimental data fitted well with the pseudo-second-order kinetics and the Langmuir isotherm model. Thermodynamic studies revealed that the adsorption process was feasible and endothermic in nature.

Adsorbents produced using palm kernel shell were also used by Yong and Zaini [61] for the removal of RbB from aqueous solution. The adsorption of RbB dye onto the adsorbent was not surface area dependent. The experimental data agreed well with the pseudo-first-order kinetics and Redlich – Peterson model. Üner et al. [62] also produced activated carbon (OAC) via chemical activation of okra wastes using zinc chloride. The OAC produced had a surface area of 1044 m<sup>2</sup>/g and a pH<sub>zpc</sub> value of 7.3. The adsorption of RbB by OAC was positively affected by the increase in pH and adsorption temperature. The experimental data fitted well with the Langmuir isotherm and pseudo-second-order kinetic model. The process was feasible, spontaneous, physical and endothermic in nature.

Acid activated *moringa oleifera* seed pod (MOSPAC) was used by Bello et al. [63] for the removal of RbB from aqueous solution. The material was found to have a pH<sub>zpc</sub> value of 3.05 and optimum adsorption was recorded at pH 3.01. The experimental data fitted well with the Langmuir isotherm and pseudo-second-order kinetic model. Thermodynamic study revealed that, the adsorption process is spontaneous, feasible and endothermic. In a different study, Inyinbor et al. [64] made use of urea modified *Raphia hookerii* (UMRH) as adsorbents for the removal of RbB from aqueous solution. UMRH had a pH<sub>zpc</sub> of 7.60 and optimum adsorption was recorded at pH 3. The experimental data fitted well with the pseudo-second-order kinetics and Elovich model. The adsorption process was spontaneous, feasible and endothermic in nature.

Microwave-activated rice husk ash (ARHA) was also used by Suc and Kim Chi [65] for the removal of RbB from aqueous solution. ARHA had a BET surface area of 61.48 m<sup>2</sup>/g and a pH<sub>zpc</sub> value of 7.45. Highest percentage removal of RbB using ARHA was achieved at pH 5.5 and 200 min of contact time. Thermodynamic studies revealed that the adsorption process is favorable, endothermic and spontaneous. The experimental data fitted well with the pseudo-second-order kinetic and Freundlich isotherm model. Hong et al. [66] also produced LS-AC-5 and MS-AC-5 activated carbons via chemical activation fruit-biomass wastes of longan seeds and mangosteen skins using KOH. MS-AC-5 had a BET surface area of 2960.56 m<sup>2</sup>/g, while LS-AC-5 had a BET surface area of 2728.98 m<sup>2</sup>/g. Both MS-AC-5 and LS-AC-5 were used as adsorbents for the removal of RbB from aqueous solution. However, due to the higher surface area of MS-AC-5, its adsorption capacity was much higher than that of LS-AC-5. The experimental data using both adsorbents fitted well with the Langmuir isotherm model.

Activated carbon (ZAC) prepared via chemical activation of *Ziziphus jujuba* seed using zinc chloride has been used by Sivarajan and Shanmugapriya [67] for the removal of RbB from aqueous solution. Maximum and minimum percentage removal of RbB using ZAC was recorded at pH 4 and 7. Thermodynamic studies revealed that the adsorption process is spontaneous and endothermic in nature.
Table 2. Reported adsorption capacities (mg/g) of various agricultural wastes-based adsorbents

| Adsorbent                                           | Maximum adsorption capacity (mg/g) | References  |
|-----------------------------------------------------|------------------------------------|-------------|
| Banana bark carbon                                  | 34.19                              | [26]        |
| Jute stick powder                                   | 87.7                               | [24]        |
| Bagasse pith activated carbon                       | 263.85                             | [27]        |
| Coir pith                                           | 55.54                              | [28]        |
| Sugarcane fiber                                     | 15.98                              | [28]        |
| *Polygonon orientale* Linn                          | 556                                | [30]        |
| *Leucaena leucocephala* seed shell waste activated carbon | 49.48                            | [31]        |
| Waste fruit residues (FR)                           | 34.48                              | [35]        |
| *Azadirachta Indica* bark (AIC)                     | 45.47                              | [32]        |
| Rice husk activated carbon                          | 212.76                             | [33]        |
| Nano-porous Jack fruit peel                         | 4.36                               | [36]        |
| Elephant dung                                       | 18.57                              | [39]        |
| Loquat seeds (LS)                                   | 410.05                             | [1]         |
| Mango leaf powder                                   | 3.31                               | [16]        |
| Used black tea leaves (UBTL)                        | 53.2                               | [29]        |
| Ashoka leaf powder (ALP)                            | 66.6                               | [43]        |
| Oil palm empty fruits bunch activated carbon        | 69.86                              | [37]        |
| Cedar cone (CC)                                     | 4.55                               | [38]        |
| Animal bone meal                                    | 64.95                              | [44]        |
| *Thevetia peruviana* activated carbon (TPAC)        | 174.23                             | [34]        |
| Almond shell (*Prunus dulcis*) biosorbent           | 33.22                              | [45]        |
| ZnCl₂ activated wood apple carbon (ZAWAC)           | 46.7                               | [17]        |
| Jatoba bark                                         | 211.5                              | [51]        |
| Water chestnut peel waste (WCPW)                    | 3.0                                | [46]        |
| Pigeon dropping                                     | 8.55                               | [7]         |
| *Maranta arundinacea* carbon                       | 88.4                               | [40]        |
| Walnut shells (WS)                                  | 2.292                              | [10]        |
| Sugarcane bagasse                                   | 65.5                               | [47]        |
| Water hyacinth plant stem activated carbon (ACS)    | 280.2                              | [41]        |
| Cysteine Modified *Raphia hooker* (CMRH)            | 357.14                             | [42]        |
| Corn stalk modified with citric acid (CCS)          | 203.0                              | [48]        |
| *Prosopis juliflora* seeds activated carbon         | 9.8                                | [23]        |
| Acid-treated dika nut (AND)                         | 232                                | [49]        |
| Peat                                                | 201.19                             | [52]        |
| Pummelo peel                                        | 133                                | [53]        |
| Activated carbon from seed husk of *Moringa oleifera* | 16.07                             | [54]        |
| *Raphia hooker* fruit epicarp                       | 666.67                             | [50]        |
| *Casuarina equisetifolia* needle (CEN)              | 82.34                              | [56]        |
| *Casuarina equisetifolia* cone (CEC)                | 49.5                               | [57]        |
| *Azolla pinnata* (AP)                               | 199.7                              | [25]        |
| Animal Bone Meal (ABM)                              | 64.95                              | [58]        |
| *Lythrum salicaria* L. activated carbon (LSAC)      | 370.37                             | [59]        |
| Jackfruit seeds (JS)                                | 26                                 | [60]        |
| Palm kernel shell adsorbent                         | 18.1                               | [61]        |
| Waste seeds of *Aleurites moluccana* (WAM)          | 117                                | [55]        |
| Acid activated *moringa oleifera* seed pod (MOSPAC) | 1250                               | [63]        |
| Urea modified *Raphia hooker* (UMRH)                | 434.78                             | [64]        |
| Okra wastes activated carbon (OAC)                  | 321.5                              | [62]        |
| Tarap skin (TS)                                     | 131                                | [18]        |
| Microwave-activated rice husk ash (ARHA)            | 21.89                              | [65]        |
| Longan seeds activated carbon (LS-AC-5)             | 117.65                             | [66]        |
| Mangosteen skins activated carbon (MS-AC-5)         | 1265.82                            | [66]        |
Artocarpus odoratissimus (Tarap) leaves (TL) were used by Zaidi et al. [68] for the removal of RhB from aqueous solution. Maximum adsorption capacity using TL was recorded at pH 3 and showed resilience when tested in a wide range of pH. The removal of RhB by TL in the presence of NaCl, NaNO\textsubscript{3} or KCl did not show any significant effect. The experimental data fitted well to the Langmuir isotherm and pseudo-second-order kinetics model. In a different study, Postai and Rodrigues [69] reported the use of waste of the fruits of Eugenia umbelliflora (FEU) as adsorbent for the removal of RhB from aqueous solution. Raw untreated FEU was named FEU-UT, while FEU subjected to maceration treatment was named FEU-T. The pzc values for FEU-UT and FEU-T were found to be 5.33 and 4.42. The experimental data for the adsorption of RhB by both FEU-UT and FEU-T followed the pseudo-first-order kinetics and the Sips isotherm model, and the process was exothermic and spontaneous. However, FEU-UT was more effective in removing RhB compared to FEU-T. This was due to the decrease in adsorption sites due to the extraction process of FEU-T.

The use of pretreated agricultural wastes including orange peels, pomelo peels and passion-fruit peels as adsorbents for the removal of RhB from aqueous solution have been reported by Nhung et al. [70]. Both materials were found to be effective with removal rates reaching 80 – 99%. The order of the adsorption capacity was orange peels > passion-fruit peels > pomelo peels. The experimental data fitted well with the Freundlich isotherm model. In a different study, the cobs of Zea mays (RZM) were used by Olyuyori et al. [71] for the removal of RhB from aqueous solution. RZM was found to have a pzc value of 6, and maximum adsorption of RhB using RZM was recorded at pH 3. The experimental data fitted well with the pseudo-second-order kinetics and Freundlich isotherm model.

Acid modified banana peels were used by Oyekanmi et al. [72] for the removal of RhB from aqueous solution. Optimized performance was recorded at pH 2, within contact time of 60 min, using 0.2 g/L dose. The acidic functional groups such as the carboxylic groups and the hydroxyl functional groups on the surface of the banana peel were predicted as being responsible for the removal of positively charged RhB molecules. The experimental data fitted well with the pseudo-second-order kinetics and Langmuir isotherm model. In a different study, Bello et al. [3] also reported the use modified activated carbon prepared from locust bean husk using orthophosphoric acid, as adsorbent for the removal of RhB from aqueous solution. The modified material was found to have a pzc value of 2.87, and maximum adsorption was recorded at pH 3.1. The experimental data was best described by pseudo-second-order kinetics and Langmuir isotherm model. The adsorption process was endothermic, spontaneous and feasible in nature.

4. CONCLUSION

Various agricultural waste-based adsorbents reported over the last one decade for their effectiveness towards removal of RhB dye from aqueous solution have been reviewed. The interest in such materials is usually because they are easily available, renewable, relatively cheap or even free of charge. Although some researchers have subjected the adsorbents to certain pretreatments aimed at improving their adsorption capability or even prepared activated carbon using the agricultural wastes, however the performance recorded using the pristine agricultural waste-based adsorbents was still remarkable. The maximum adsorption capacity

| Adsorbent | Maximum adsorption capacity (mg/g) | References |
|-----------|-----------------------------------|------------|
| Ziziphus jujuba seed activated carbon (ZAC) | 142.9 | [67] |
| Artocarpus odoratissimus (Tarap) leaves (TL) | 104.96 | [68] |
| Untreated waste of the fruits of Eugenia umbelliflora (FEU-UT) | 161.1 | [69] |
| Treated waste of the fruits of Eugenia umbelliflora (FEU-T) | 106.4 | [69] |
| Passion-fruit-peels | 51.81 | [70] |
| Orange peels | 312.5 | [70] |
| Pomelo peels | 38.31 | [70] |
| Cobs of Zea mays (RZM) | 500 | [71] |
| Acid modified banana peels | 9.5220 | [72] |
| Acid activated locust bean husk pod (ALBP) | 1111.1 | [3] |
was found to vary between 3 – 1265.82 mg/g. With such significant performance, it is hoped that agricultural waste-based adsorbents will replace commercial adsorbents in real wastewater remediation.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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