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Biodegradable natural carbohydrate polymeric sustainable adsorbents for efficient toxic dye removal from wastewater

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ABSTRACT

The toxic anionic dye of methyl orange encapsulation was studied using biodegradable natural carbohydrate polymeric adsorbents of rice flour (RF) and graham flour (GF). The adsorbents were characterized by several instrumentations to understand the functionality and potential use. The dye adsorption parameter was measured based on the solution acidity, contact time, initial concentration effect, competing anions affinity, bonding mechanism, maximum adsorption capacity and reuses with biodegradability. The solution acidity was exhibited the key factor, and the suitable pH 7.0 and 5.50 were selected for RF and GF adsorbent, respectively based on the efficiency. The competing ions were not adversely affected in the dye adsorption as defined by the stable bonding mechanism. The adsorption data were highly fitted with the Langmuir adsorption model with monolayer coverage. The determined maximum adsorption was 173.24 and 151.27 mg/g for RF and GF, respectively, which was comparable with the other forms of materials. The desorption data was promising as the RF and GF adsorbents were used several cycles and therefore, the biodegradable adsorbents are promising to use in the real sample treatment to clean up the contaminated water.

1. Introduction

Fresh water scarcity and resources are under immense in trouble due to the fast increasing of population with their daily living standards. Therefore, the pure water crisis is the most important problem, which is faced by the living word because of the industrial growth in textiles, tannery, plastics and food [1,2]. The scientists are assumed that millions of people will face drinking water problem by 2025 [3–6]. This also predicted that 65% people in the world will face tremendous water intake for daily lives. The water problem comes from the mismanagement and improper taking care of water supply and the human health is threatening [7]. In other words, water contamination induced from the large amount of industrial effluents without proper treatment before disposing to the water bodies and causes a serious threat to the entire human health [8,9]. Considering the several industrial effluents and waste, the textile industries are drawing extensive diligence to the scientists and water experts due to the huge amount of unwanted dye effluents into the water bodies to break the eco-system [10–12]. The dyes are nonbiodegradable and caused serious adverse effect to the eco-system [13–15]. When the dye effluents from the textile are released into the water, the dyes are impeding the invasion of sunlight into the water environment, adversely competed with the oxygen transfer and prohibited the re-oxygenation scope of the receiving water, and ultimately shrinkage the biological movement [16–20]. Therefore, contamination of drinking water by diverse toxic metal ions and dyes are inevitable and shows to severe toxicity [21]. Thousands of dyes are using in several industries and mainly the textile and most of them are highly toxic or even harmful to animals and humans, thus burdening a huge obstacle and immense impact on the purification of the world's water environment [22–24]. Then the removal of dyes from wastewater is the crucial need to save the human health in large-scale applications.

Based on the suitability and potentiality of the wastewater treatment, a variety of approaches are reported for water treatment [25].

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The promising techniques such as chemical precipitation, electrocoagulation, chemical degradation, photocatalysis, ion exchange, membrane separation, bio-removal, and adsorption [26–32]. However, many of them are faced with limitations due to the low adsorption capacity, slow adsorption kinetics, high cost, and difficult to use in field area. In order to address these problems, new materials are always needed to make the simplicity for water treatment process by developing functional group containing materials [33]. Over the last decades, adsorption process is indicated as the promising and sustainable approaches due to the simplicity, cost-effectiveness and potentiality in practical use stand point [34]. The nanomaterials-based adsorbents are used for water treatment for removing toxic metal ions. The adsorbents are fabricated by organic ligand or dye molecule anchoring to define the specificity [35]. Then the nanomaterials are promising candidate for toxic substance encapsulation from wastewater. The pore size, shape and composition of the silica monoliths are restrained for high amount of organic molecule coating. Therefore, these materials have potential uses in gas storage, separation and recovery, drug delivery, catalysis and sensors due to their tunable pore size, shape and composition of the material as controlled of the ordered channel porosity [36]. Apart from the synthesized materials, a variety of natural and biomaterials such as coal, natural polymeric starch, carbohydrate, cellulose, wood, rice husk, fly ash, activated carbon, cotton waste, clay and other biodegradable materials are indicated as effective adsorbent for the efficient encapsulation of dye from solution media [37–42].

In adsorption process using suitable adsorbent, two techniques such as batch and column are deployed to separate or adsorb the pollutants from water. The batch method is a simple method and it implements a simple way to define the parameters influencing adsorption method [43–46]. Column approach is also useful in deciding the amount of material needed for encapsulation of pollutants from waste solution [47]. In column approach, the adsorbent is constantly in contact with spanking fresh solution and the clogging materials were avoided based on the flow rate parameter. Columns have been attractive to have better uptake efficiency over batch in some cases [48]. In the present study, batch approach was implemented because of the nature of the adsorbent and process simplicity rather than column approaches [49].

The biodegradable materials are growing attention due to the existing of all kind biological properties. Activated carbon, chitosan, carbohydrate and starch polymeric material are used for diverse uses especially the toxic dyes removal in aqueous media. Chitosan is a natural polysaccharide and deacetylated in the form of chitin [50–52]. In addition, chitosan has excellent biological properties such as good coating ability, good adhesion, high water permeability, nontoxicity and biocompatibility due to the presence of amino and hydroxyl functional groups [53]. However, several drawbacks including the infectious diseases are caused by pathogenic bacteria worldwide and the drug resistance of the pathogenic microorganisms is a serious concern for next generation drug development [54]. Moreover, the biodegradability property is not existed and make them difficult in real sample treatment. Therefore, natural polymer-based carbohydrate starch materials are promising concerning the efficiency, cost-effectiveness, and biodegradability. The natural carbohydrate starch is very bounteous, low-cost, and available in everywhere in the globe. From this stand point, the natural carbohydrate-based adsorbent was selected in this study to remove the toxic dye from waste solution.

In this study, we focused on a novel green accessible accession to reach in helpful find out for wastewater treatment and specially dye removal in effective way. The natural carbohydrate polymer based biodegradable adsorbent of rice flour (RF) and graham flour (GF) was chosen for successive anionic dye of methyl orange (MO) removal under optimum condition. The selected bio-adsorbents were characterized using FTIR spectroscopy, SEM and others techniques. Detailed adsorption study of a reactive dye of MO by the RF and GF was performed and experimental influencing various parameters such as pH, time, concentration, kinetics, isotherms and reuses of the adsorbents was measured systematically. At the end, the possible mechanism was presented for effectiveness of the biodegradable adsorbents.

2. Materials and methods

2.1. Materials

All materials and chemicals were of analytical grade and used as purchased without further purification in all experimental work. The rice flour (RF) and graham flour (GF) were collected from local market. The RF and GF were washed with water and then dried before using the dye adsorption operations. The methyl orange (MO) was purchased from Sigma–Aldrich Company Ltd. USA and other chemicals were procured from Wako Pure Chemicals, Osaka, Japan. For pH solution adjustments in adsorption experiment, the dilute HCl and NaOH was used as required. All the experiments were carried out under optimum conditions.

2.2. Instrumentations

The SEM analysis was carried out on Hitachi S-4300. Before insertion into the chamber the powder substrates were fixed on a SEM stage using carbon tapes. The Pt films were deposited on substrates at room temperature by using an ion-sputter. The distance between the target and the substrate was 5.0 cm. The Ar working pressure, the power supply (100 W) and the deposition rate were kept constant throughout these investigations. In addition, to better record the SEM images of samples, the SEM micrographs were operated at 20 keV.

The functional group of the biodegradable adsorbents were measured by the FTIR spectra (IR-470, Shimadzu, Japan) using KBr pellet technique. Small angle powder X-ray diffraction (XRD) patterns were measured by using 18 kW diffractometer (Bruker D8 Advance) with monochromated CuKα radiation and with scattering reflections recorded for 2θ angles between 0.1 Å and 6.5 Å corresponding to d-spacing. First, the powder samples were ground and spread on a sample holder. The XRD data were not plotted due to the Figures limitation in this publication media. The absorbance spectrum was measured by UV–Vis–NIR spectrophotometer (Shimadzu, 3700).

2.3. Dye adsorption by the RF and GF adsorbents and reuses

In the adsorption system, two biodegradable adsorbents of RF and GF were added into the MO dye solution and adjusted at specific pH values by adding HCl or NaOH in 20 mL solutions. The solid adsorbents of RF and GF were separated by filtration system and MO concentration in before and after adsorption operation was analyzed by UV–Vis-NIR. The MO adsorption was determined according to the following equations:

$$\text{Mass balance } q_e = (C_0 - C_f)\, V/M \text{ (mg/g)} \quad (1)$$

and

$$\text{Pb(II) ion adsorption efficiency } Re = (C_0 - C_f)\, 100/C_0 \text{ (\%)} \quad (2)$$

where V is the volume of the aqueous solution (L), and M is the weight of the RF and GF (g), C_0 and C_f are the initial and final concentrations of MO dye in solutions, respectively.

To define the equilibrium contact time, 10.0 mg/L MO amount of fixed volume solution was used in each fraction, and the RF and GF adsorbents amount was fixed at 20 mg in each case. After 20 min interval, the solid material was separated and filtrate was determined by the UV–Vis-NIR. The highest adsorption amount was defined based on the different initial concentrations of MO, and each sample was determined by the UV–Vis-NIR.

In desorption operations, first 30 mL of 2.0 mM MO solution was adsorbed on the 50 mg RF and GF adsorbents and then desorption

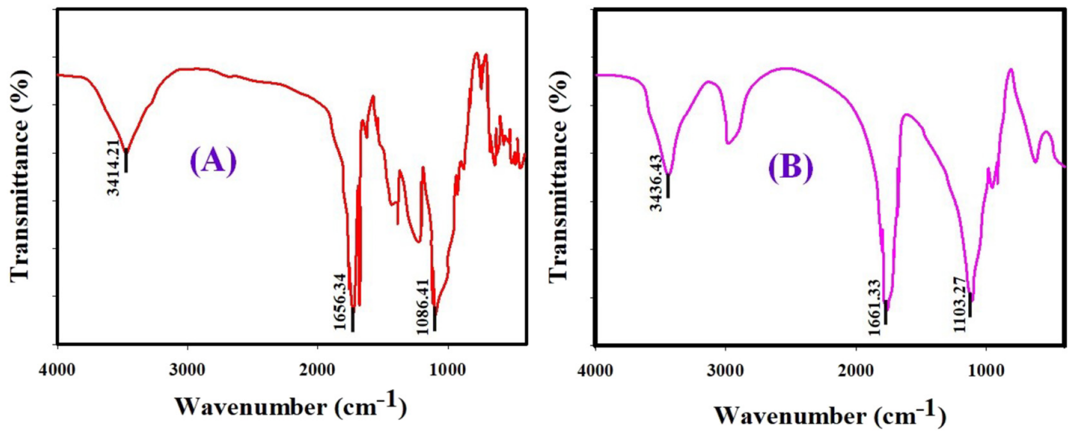


Fig. 1. Determination of functional groups of the rice flour (RF) and graham flour (GF) by the FT-IR spectra at optimum measurement conditions.

experiments were performed using different eluents. The MO dye adsorbed onto RF and GF was washed with deionized water several times and transferred into a 50 mL beaker. The washing solution was also analyzed by UV–Vis-NIR. The ethanol solution was used as appropriate eluent and then the mixture was stirred for 15 min. The concentration of MO released from the RF and GF adsorbents into aqueous phase was analyzed by the UV–Vis-NIR. Then the RF and GF adsorbents were reused several cycles to investigate the reusability after rinsing with water.

All experimental demonstrations were duplicated to confirm the extracted data in this study. The highest variation for each adsorption operation was 3.0%.

3. Results and discussion

3.1. Characterization of RF and GF adsorbents

The functional group of the RF and GF adsorbents were evaluated by the FTIR and data are shown in Fig. 1. The specific O—H stretching peak define by the absorption band of H-bond was shown at 3414 and 3436 cm^{-1} in RF and GF, respectively. The water molecule was connected and it was defined at the of 1656 and 1661 cm^{-1} in RF and GF adsorbent, respectively. In addition, the coupling bond of C—O, C—C and O—H stretching bond in the peak area at 1086 and 1103 cm^{-1} in RF and GF, respectively [55]. This finding confirmed the active functionality of the RF and GF adsorbent with the crystallinity and moisture content for taking up the MO at optimum condition through the interaction of active sites adsorbent surfaces.

The particle size and porosity were measured by the SEM images as shown in Fig. 2. The SEM images of RF exhibited irregular particles shape with granular morphology as judged from Fig. 2(A, B). On the other hands, the SEM images of GF were exhibited stone like morphology with the accumulation of grains [56], those were existed as irregular channel as judged from Fig. 2(C, D). However, the inorganic materials are exhibited large particles morphology with textural properties for high accumulation of organic substances in diverse uses [57].

3.2. Dye adsorptions

3.2.1. Effect of pH

In adsorption operation, solution pH is key the important parameter to control the high uptake to define the optimum condition [58].

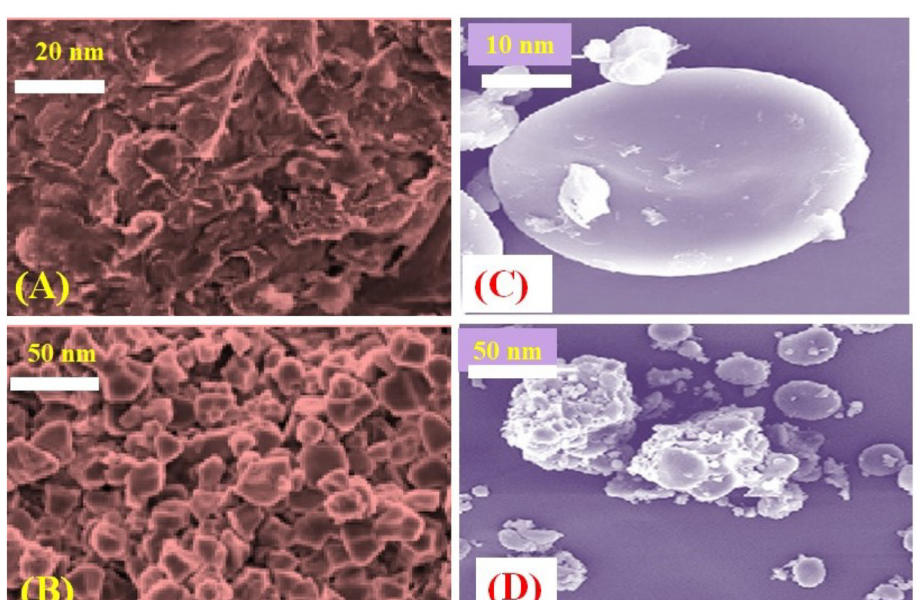


Fig. 2. The SEM images of the (A and B) rice flour (RF) and (C and D) graham flour (GF) with different scaling to understand the particles exhibition.

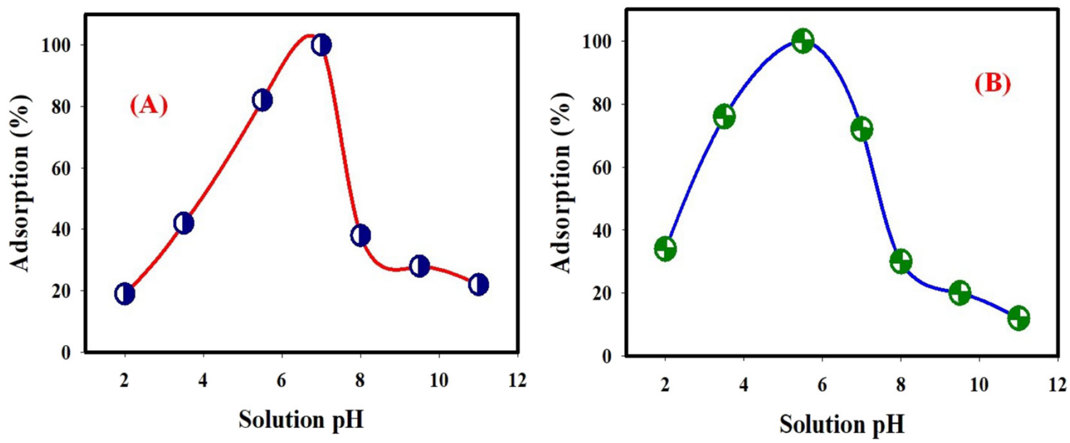


Fig. 3. Effect of solution pH for the determination of methyl orange adsorption by the (A) rice flour (RF) and (B) graham flour (GF) at optimum experimental protocol as indicated in experimental section.

Therefore, the pH solution has significant effects on the anionic dye of MO adsorption by the RF and GF adsorbent to measure the influence of interaction [17,56,59]. In this connection, the experiments were carried out to measure effect of pH on the removal ratio of MO by RF and WF biodegradable adsorbents. The solution MO concentration was 10 mg/L, whereas the amount of RF and GF was 20 mg in each fraction. The pH was ranged from 1.0 to 11.0 to define the ionization effect in the adsorption operation. The mixed solutions of MO, RF and WF were stirred for 30 min and the solid adsorbents were separated and measured the MO removal efficiency by UV–Vis-NIR. The results are shown in Fig. 3. The data revealed that the removal results of MO by RF and GF were highly affected by the solution pH. It was clarified that at each pH, the addition of RF and GF made the MO solution decolorized effectively and RF and GF were changed the color at optimum level. At low pH region, the uptake of MO was low in both of adsorbents. However, increasing the solution pH, the MO removal efficiency was increased by the both adsorbents. The high amount of MO removal efficiency was at pH 7.0 and 5.50 by the RF and GF, respectively. Over pH 7.0, the MO removal efficiency was decreased as the hydroxyl group competition as expected as judged from Fig. 3. However, within a relatively wide pH range, RF and GF effectively removed MO dye molecule from the aqueous solution in wide pH ranges. It is therefore suggested that at each pH, MO dye had an interaction with RF and GF to form a complex with a color different from that of the MO solution for effectively taken up under the conditions. Based on the high uptake efficiency, the pH 7.0 and 5.50 was selected for MO adsorption by the RF and GF, respectively to define the other experimental parameters.

3.2.2. Effect of contact time

The adsorption kinetic determination by the adsorbent material in wastewater treatment is crucial due to the major insight internal mechanism for reaction kinetics [60]. Therefore, sufficient contact time between MO and biodegradable adsorbent of RF and GF for equilibrium adsorption was evaluated. For the evaluation of contact time effect, 30 mg of the RF and GF were added with constant initial concentration of MO at pH 7.0 and 5.50, respectively for different intervals time of stirring. The MO adsorption onto the RF and GF with the function of time is shown in Fig. 4. The results clarified that MO adsorption was increased with increasing the contact time by the both adsorbents as judged from Fig. 4. The similar trend is also reported by the functional ligand based diverse materials [61]. The MO adsorption efficiency was high at beginning and then gradually increases to reach the plateau by the RF and GF adsorbent. This is expected that the functional groups were abundant and high amount of pollutant was anchored onto the adsorbent surface [62]. The maximum amount of removal efficiency was observed by 220 and 260 min for RF and GF, respectively. Then the 5 h was selected in the determination of maximum adsorption capacity in the case of RF and GF adsorbents. This is noted that ligand based composite materials and ion-exchange resins and fibrous adsorbents are exhibited high kinetic performance comparatively to the carbohydrate biodegradable materials [63].

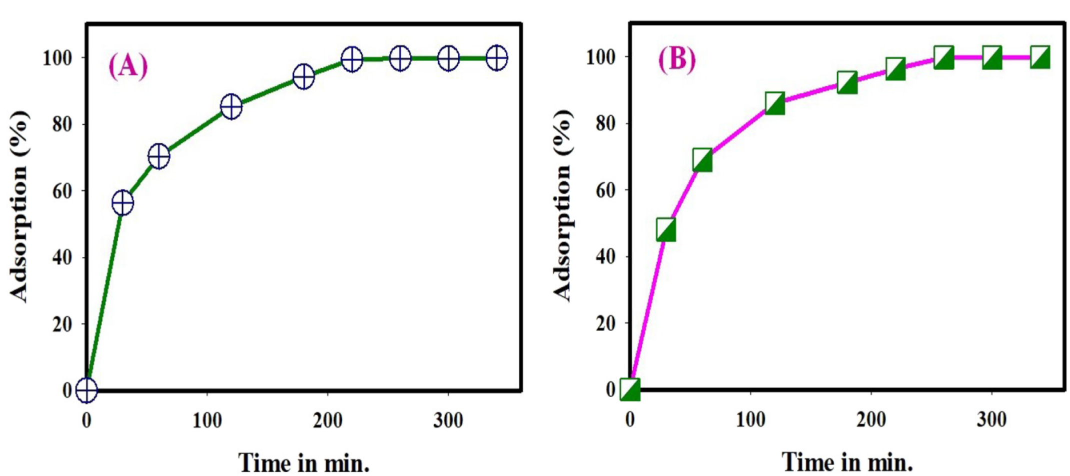


Fig. 4. Effect of contact time for the evaluation of equilibrium methyl orange dye adsorption by the (A) rice flour (RF) and (B) graham flour (GF), where the initial concentration was fixed.

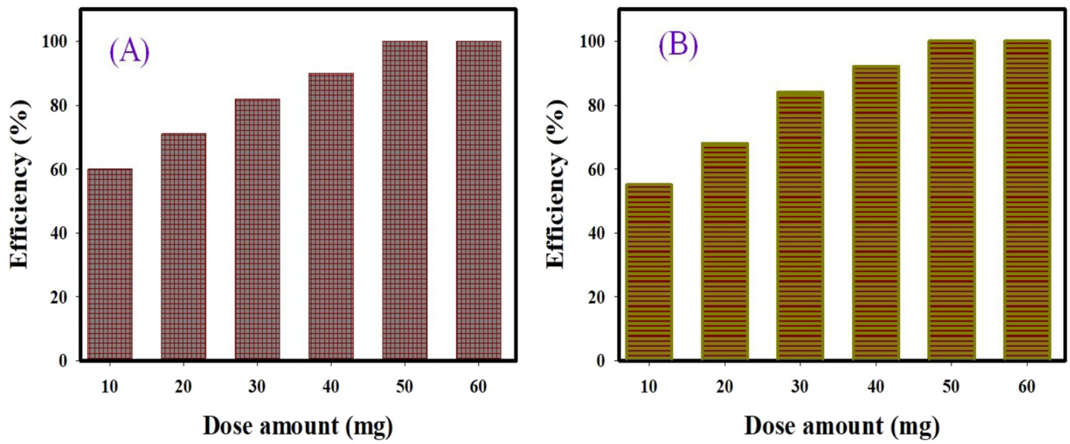


Fig. 5. Effect of adsorbent amount for the evaluation of maximum methyl orange dye adsorption by the (A) rice flour (RF) and (B) graham flour (GF), where the initial concentration and solution volume were fixed.

3.2.3. Effect of adsorbent amount

The effect of adsorbent amount on MO removal efficiency was evaluated by adding various amounts of RF and GF when the initial concentration was fixed at optimum pH conditions. With different adsorbent amount, the MO removal efficiency are shown in Fig. 5. The date clarified that the MO removal efficiency was increased with increasing the amount of adsorbent of RF and GF as judged from Fig. 5. The maximum MO removal efficiency was observed with 50 mg when the initial MO concentration was fixed at 20 mg/L. This is usual case that the dye removal percentage would remain the same or enhance with the increase of the adsorbent amount [57,64]. Several researchers also reported that pollutants adsorption efficiency was decreased with increasing the amount of adsorbent [65,66]. This is unusual because the optimum pH condition was not evaluated. Therefore, the optimum pH solution is the key factor in the adsorption process with the diverse functional materials [67].

3.2.4. Effect of initial concentration and adsorption isotherms

The dye removal performance by the biodegradable adsorbents of RF and GF was assessed by adsorption amount and removal percentage in the condition of various MO initial concentrations. As the initial concentrations of MO dye getting higher, the removal amount was enhanced by the both RF and GF adsorbent because of the driving force to overcome the mass transfer resistance between the bulk liquid phases to the solid material phase, and RF and GF adsorbent put up more considerable potential for the MO dye removal quantity [68,69]. Fig. 6 shows the effect of initial MO concentration effect on the adsorption by RF and GF adsorbent and various initial MO concentrations was varied from low to high where the other experimental conditions were fixed. It was found that the adsorption of MO dye was high in the initial low concentration region and then increased gradually to reach in equilibrium state for the determination of maximum adsorption capacity as judged from Fig. 6.

To determine the mechanistic parameters associated with MO dye adsorption by the RF and GF adsorbents, this study measured the relationships between adsorbed MO and its residual in aqueous solution at adsorption equilibrium by using Langmuir adsorption model. The Langmuir adsorption isotherm is the best known of all isotherms describing adsorption operation based on the material morphology. The linear form of the Langmuir isotherm equations is represented by the following equation:

$$C_e/q_e = 1/(K_L q_m) + (1/q_m)C_e \text{ (linear form)} \tag{3}$$

where Ce is the concentration of MO dye in the solution at equilibrium in mg/L, qe is the amount of MO dye adsorbed at equilibrium in mg/g, q_m is theoretical maximum adsorption capacity of MO in mg/g, and K_L is Langmuir equilibrium constant. Fig. 6 (inset) shows the linear adsorption isotherm obtained using the Langmuir model, and the Langmuir

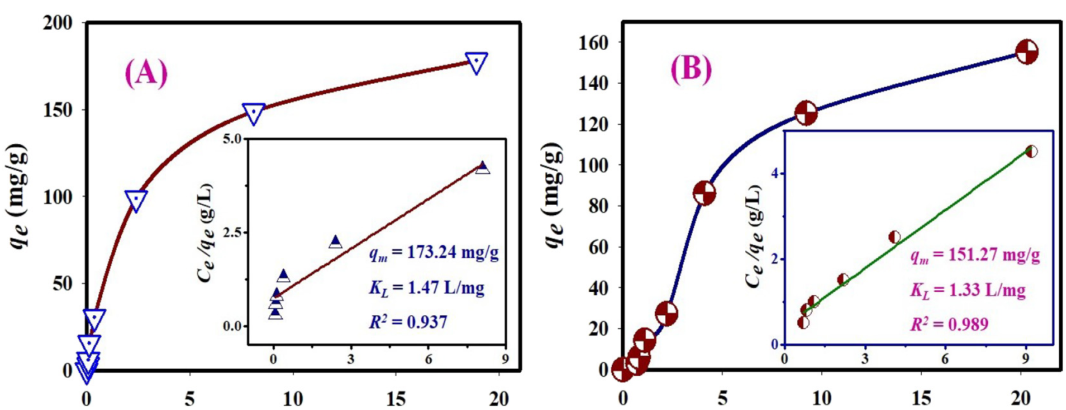


Fig. 6. Determination of maximum dye adsorption capacity with the variations of initial MO concentration by the (A) rice flour (RF) and (B) graham flour (GF) according to the data plotted with the Langmuir isotherm model.

Table 1
Comparison of dye adsorption capacities using different forms of adsorbent materials.

Used adsorbents	Name of dye	Capacity (mg/g)	Ref.
PABA@AC	Malachite green	66.87	[49e]
Fe-BDC MOF	Methylene blue	8.65	[15]
Pine leaves	Methylene blue	126.58	[73]
Citric Acid Modified Sawdust	Methylene blue	111.46	[65]
Cellulose surface	Reactive red dye	78.00	[51]
Mango seed	Orange 16	52.5	[43]
Rice flour (RF)	Methyl orange	173.24	This study
Graham flour (GF)	Methyl orange	151.27	This study

isotherm model parameters including the q_m values of conjugate material. The q_m values of MO adsorption on RF and GF adsorbents indicate better adsorption, which was 173.24 and 151.27 mg/g, respectively. The bioadsorbents in this study were significantly increased the adsorption capacity due to the surface functional group, interlayers, and edges of the adsorbent surfaces. The coefficient (R^2) for Langmuir model was indicated the suitable monolayer coverage by the RF and GF adsorbents. The experimental results shown that the adsorption data are fitted well with Langmuir isotherms, suggesting the monolayer coverage on the adsorbent surface as expected naturally. The maximum adsorption capacity of RF and GF were not the best among those reported in previous studies. However, the maximum adsorption capacity is not the only performance indicator rather than the focused more on the dye removal sensitivity and potential waste sample treatment. Obviously, the RF adsorbent showed the slightly higher adsorption capacity compared with GF adsorbent as judged from data in Fig. 6. However, this is not clear of the specific reasons as the consisted of the functional group content. In the case of nanomaterials, the highly porous materials are possessing large specific surface area based on the surface homogeneous morphology. Therefore, the high amount of organic substances is accumulated as expected [70].

According to reported previous deployed adsorbent materials in Table 1, there are many materials applied in different types of dye removal from aqueous solution. This is noted that they can be divided into three categories based on their compositions, including natural materials, synthetic compounds and modified adsorbents. Due to their significantly different physical and chemical characteristics, the different kinds of functional materials are not comparable due to the similar functionality effect [47,57]. However, for the same kind of adsorbent materials, the proposed natural carbohydrate polymeric adsorbent in this study, it is meaningful to compare their adsorption performance to understand their potential application in real contaminated wastewater treatment.

3.2.5. Effect of foreign anions

The adsorption of anionic dye on the biodegradable and inorganic-organic composite adsorbent mainly depends on the electrostatic interaction based on the complexation and ion exchange mechanism [25,59]. Then the similar ionic radii or similar valance of ionic molecules are competed due to the valance factor in the complexation and co-valent bonding mechanism [71–73]. Therefore, the coexisting anions may interfere with the MO anionic dye for the adsorption sites on the adsorbent of RF and GF. The chloride, nitrate and sulphate were selected as the coexisting anions to the point of anionic MO dye, and with different amounts of these anions, the MO dye adsorption efficiency were carried out by the RF and GF adsorbent and the data are shown presented in Fig. 7. It can be found that the coexisting anions were not interfered in the MO dye adsorption by the RF and GF adsorbent even the presence of high amount of divalent sulphate anion was negligibly interfered as judged from Fig. 7. This is probably the reason of anion substance of MO molecules rather than anion of chloride, nitrate or sulphate. However, the real cause is not clear at this stage but in can be assumed the binding mechanism. No doubt, the results of MO dye adsorption on the RF and GF mainly depends on the electrostatic interaction. Besides the charge competition, the slight inhibition capacity caused by sulphate might be also resulted in the different valance state with the sulphate and MO dye molecule.

3.2.6. Possible mechanism of dye adsorption

In the understanding of bonding mechanism, the adsorption process is clearly present the nature of selectivity and sensitivity [74–76]. The functional groups associated in the RF and GF adsorbent were measured based on the FTIR measurement. In addition, the mechanism of toxic dye adsorption and organic compound anchoring by the functional adsorbent materials may be clarified by considering the solution acidity, degree of dissociation of dye species and surface functionality presenting the charges of the functional adsorbent materials [77,78]. From the

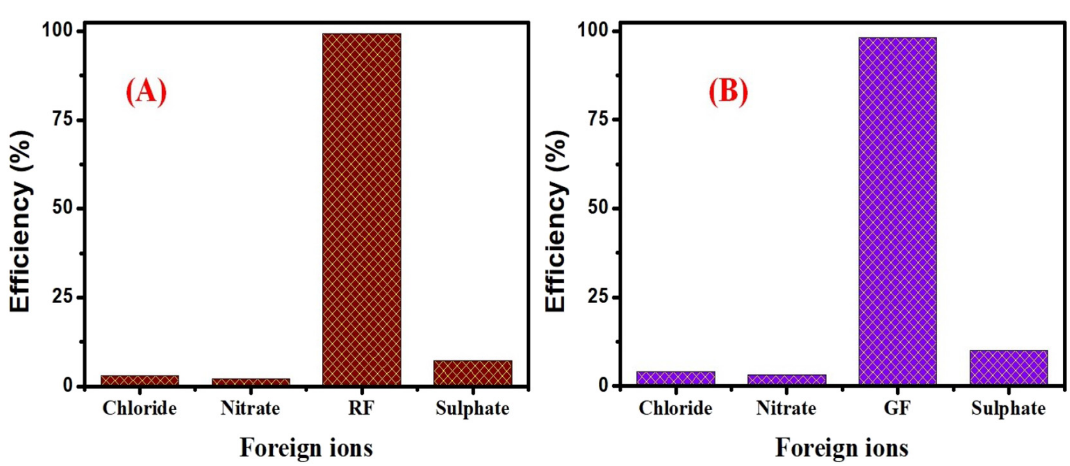
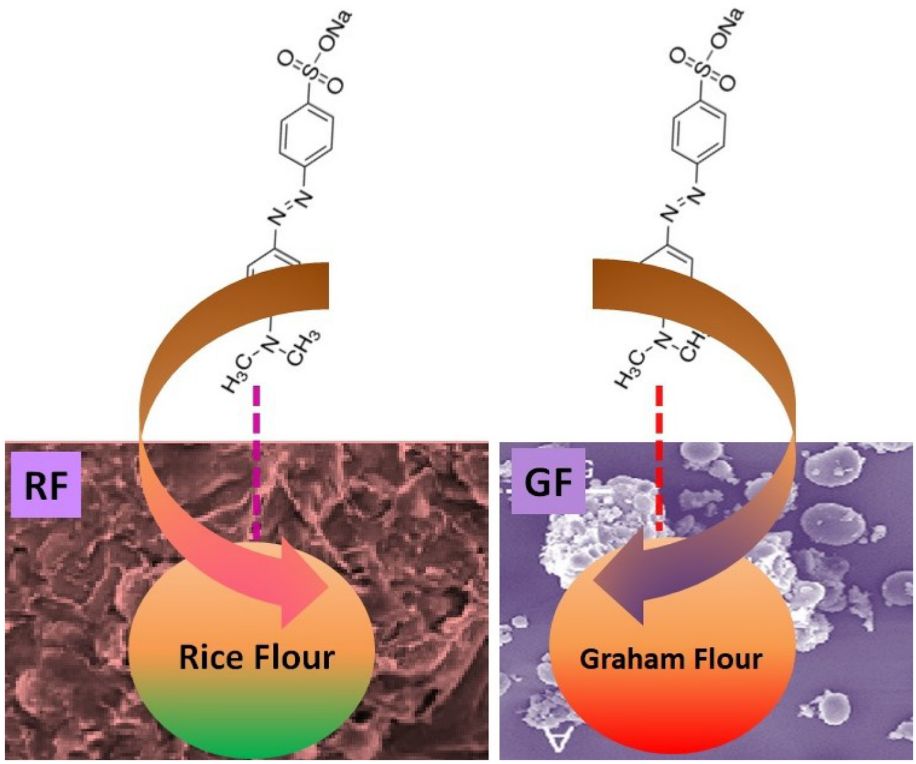


Fig. 7. Effect of coexisting anions on MO adsorption onto (A) rice flour (RF) and graham flour (GF), where the competing ion amount was higher than the MO.



Scheme 1. Possible bonding mechanism between RF and GF adsorbent and MO dye molecules at optimum conditions.

hydroxyl availability in the RF and GF adsorbent, the anionic MO dye might be associated as hydrogen and covalent bonding as pustulated in Scheme 1. Based on the bonding ability, the RF and GF adsorbent were able to selectively adsorbed the anionic MO dye with high efficiency (Fig. 7).

3.3. Reuse and biodegradability

The desorption is an important criterion in adsorption process to make the adsorbent cost effective and promising and real sample treatment [79,80]. Therefore, the desorption of MO dye from the RF and GF was evaluated based on the elution process. The desorption of MO dye or elution experiments were evaluated with the ethanol from the stand point of regeneration ability. By the using of this eluent, the RF and GF adsorbents was simultaneously regenerated into the initial form and able reuses for several cycles without significant deterioration of the RF and GF adsorbent functionality as the data were depicted in Fig. 8. Then the reusability was evaluated of each adsorption–desorption cycles for several times, and the adsorption efficiency in each cycle was compared with the 1st cycles adsorption ability of the RF and GF adsorbent as reported elsewhere [61,70]. As the RF and GF adsorbents are collected naturally and exhibited the biodegradable properties based on the carbohydrate characteristics as indicated in the preceding sections. This reusability studies clarified that the RF and GF adsorbent can be used in many cycles without lose the ability to perform in its original adsorption affinity to the MO anionic dye removal. Therefore, RF and GF are low-cost, selective and sensitive, recyclable and promising biodegradable adsorbent for sustainable adsorption of anionic dye removal to clean up the contaminated water.

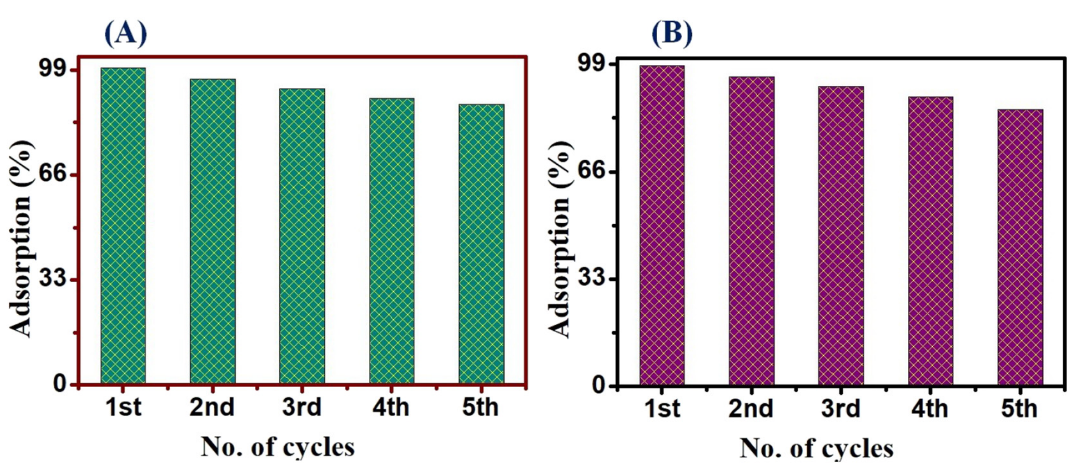


Fig. 8. The reusability performances after successful elution of MO with ethanol in successive several cycles of (A) rice flour (RF) and (B) graham flour (GF).

4. Conclusions

To remove pollutants from wastewater with efficient manner at the wide range level will lead to new frontiers in technology and natural carbohydrate polymeric materials, and tunable and sensitive materials are highly desired. Therefore, the natural carbohydrate polymeric materials of rice flour (RF) and graham flour (GF) was deployed to remove the anionic dye of methyl orange from contaminated water. The RF and GF adsorbents in this study were exhibited interesting selective behavior that permits accurate, specific removal of toxic dye with high efficiency according to the optimum experimental conditions. The solution pH was highly affected due to the nature of adsorbent charge functionality. The influence of each affecting factor and of their interactions was evaluated at optimum experimental procedure. The kinetic data revealed that the RF and GF adsorbents were exhibited slow kinetic performance compared to the functional nanomaterials and ion-exchange fibrous adsorbents. The adsorption data were well fitted to the Langmuir isotherms and exhibited high adsorption capacity of 173.24 and 151.27 mg/g of RF and GF adsorbent, respectively with monolayer coverage. The foreign anions of chloride, nitrate and sulphate were not interfered with the dye adsorption operation by the RF and GF adsorbents. Moreover, the adsorbed dye on the RF and GF was completely desorbed with ethanol and regenerated into the initial form for next removal operation without significant lose in functionality in several cycles. According to the data presentation with sensitivity, selectivity and reuses, the RF and GF natural carbohydrate polymeric adsorbents could be utilized to remove the toxic anionic dye in large-scale application as well as clean up wastewater to safe the human health.

CRediT authorship contribution statement

All authors were agreed to include their names based on the contribution. M.M.H., M.N.H. and M.A.S. were performed the experiments and analyzed the data. M.M.I. and J.I. were participated in the discussion. M.R.A. was designed the work, analyzed the data, wrote and revised the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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