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REMOVAL OF AMARANTH (AR) ONTO PUFFED RICE IN AQUEOUS SOLUTION

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ABSTRACT

Water pollution is very common term we use, now a days water pollution occur due to untreated water disposal from various industries especially textile and dying industries use huge amount of water and their untreated effluent causes the water pollution. Adsorption is well known method for such water treatment by using different adsorbents. Scientist are still trying to find new adsorbent for adsorption of dye from aqueous solution. This study shows Puffed Rice can be used as an effective adsorbent to remove Amaranth Dye from aqueous solution. The adsorption process followed the second order kinetics and endothermic in nature. The process was feasible too. All experimental data showed that Puffed Rice is wonderful adsorbent to remove Amaranth dye from aqueous solution.

KeyWords

Activation Energy, Adsorption, Adsorbent, Amaranth Dye, Thermodynamic, Isotherm, Langmuir, Puffed Rice

1 INTRODUCTION

Industrialization is a precondition for rapid economic development for every country that's why industry is growing sector. Industry help us to our economic boost but on the other hand it is creating some serious problems too. Now a days industrial untreated effluents are one of the major causes of environmental pollution especially effluents from textile and dyeing industries contain highly colored with a large amount of unused organic solid [1] which are polluting water bodies. This contaminated water is harmful to aquatic life or to human beings by mutagenic and carcinogenic effect. [2]. Different types of dyes are available such as anionic, cationic, and non-ionic [3]. Anionic dyes are highly soluble in water they are commercially available in the forms sodium salts. Different techniques like precipitation, ion exchange, chemical membrane filtration, coagulation, electrochemical, oxidation, removal [4-6], etc. are available for the removal of dyes from wastewater. Among all these methods adsorption is the superior one in terms of initial cost and ease operation. Advantages of this technique are its low generation of residues and the possibility of its adsorbent being recycled and reused [7]. Many material are used as adsorbent but researcher are still trying to find out new one [8-13] as low cost adsorbent material.

In this study puffed rice is introduced as adsorbent to adsorb amaranth from aqueous solution. The influence of pHs, solution temperature, concentration effect, ionic strength effect, desorption and reuse were studied in batch mode. Various models were tested to investigate the removal kinetics and equilibrium removal behavior.

2 EXPERIMENTAL

2.1 MATERIALS

Puffed rice was purchased from local market and was used without further purification although it was grained to make powder. No other chemical or physical treatments were used prior to removal experiments. The amaranth (AR) was obtained from Sigma-Aldrich Germany and was used without purification. The chemical structure of AR is shown in Fig: 1. All other reagents and solvents were commercially available and highest grade of purity; hence they were used without purification. Deionized water was prepared by passing distilled water through a deionizing column (Branstead, Syboron Corporation, Boston. USA).

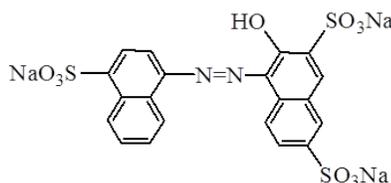


Figure: 1. Chemical structure of Amaranth (AR)

2.2 FTIR SPECTRA

The Fourier transformation infrared (FTIR) spectra of amaranth (AR) and puffed rice before and after removal of amaranth (AR) were recorded in the frequency range 400-4000cm⁻¹ using FTIR spectrometer (IR Prestige-21 FTIR Spectrophotometer, Simadzu, Japan).

2.3 BATCH REMOVAL EXPERIMENTS

A series of batch experiments were performed to study removal of AR (44.76 μmole/L) solution with the pH ranging from 2 to 8. The concentration of AR in the supernatant was determined by spectrophotometric method. λ_{max} value of AR was 521nm. The λ_{max} (521nm) of AR was found to be constant at the pH ranging from 2 to 8. The amounts of AR adsorbed onto Puffed rice q_t (μmole/g) at any time t and q_e (μmole/g) at equilibrium were determined from the following relationships:

$$qt = V(C_0 - C_t)/m \quad (1)$$

and

$$qe = V(C_0 - C_e)/m \quad (2)$$

where C₀ (μ mole/L), C_t (μ mole/L), C_e (μ mole/L), are the liquid-phase concentrations of AR at initial, at any time t, and equilibrium, respectively; V (L) is the volume of AR solution and m (g) is the amount of dry Puffed rice power used. The equilibrium removal of AR onto Puffed rice in aqueous solution were also performed at temperatures 35, 40 and 45 °C. Desorption was also studied by producing complex of Puffed rice and AR and 0.1g of the complex was taken into 25 mL of 0.1 mole/L of NaOH solution and then performed batch removal experiments for 180 min. Reuse process were also performed to study the feasibility of Puffed rice using as a good and commercially viable low cost reusable adsorbent.

3 RESULTS AND DISCUSSION

3.1 POINT OF ZERO CHARGE (PHPZC) MEASUREMENTS

Point zero charge determination is important to know the surface charge of an adsorbent [14]. The pHPZC of puffed rice estimated to be 6.

3.2 FTIR ANALYSIS

FTIR ANALYSIS OF AMRANTH

In the FTIR spectra of Amaranth (AR) broad intense band at 3444.87 cm^{-1} attributed to hydrogen bonded -OH vibration [15]. The in-plane OH deformation vibration appears in the IR spectra as strong band at 1496.76 cm^{-1} corresponding to in-plane-bending vibration of OH. Peaks in the region $520\text{-}702\text{ cm}^{-1}$ for associated OH [16]. The medium strong band at 740.67 cm^{-1} in the IR spectra corresponds to out-of-plane bending mode of hydroxyl vibrations. The band occurring between 1435.04 cm^{-1} and 1339.60 cm^{-1} corresponds to the stretching mode of an azo-compound [17]. C-N stretching vibrations of azo-compounds appear in the $1197.79\text{-}1139.93\text{ cm}^{-1}$ region (IR) [18]. Asymmetric vibrations of SO_3^- group of sulfonic acid salts usually occur in the IR at $1250\text{-}1140\text{ cm}^{-1}$. The band due to the symmetric stretching vibration is sharper and occurs at $1130\text{-}1080\text{ cm}^{-1}$. SO_3^- symmetric deformation modes give strong bands in the $550\text{-}660\text{ cm}^{-1}$ region (IR) [19]. Naphthalene ring stretching vibrations are expected in the region $1580\text{-}1300\text{ cm}^{-1}$ whereas naphthalene CH bending modes give rise to bands in the region $1230\text{-}970\text{ cm}^{-1}$ [20]. These vibrations are mixed with in-plane-bending vibration of OH group, coupled with C-N stretching vibration or with SO_3^- vibrations. Peaks at 1197.79 cm^{-1} and 1041.56 cm^{-1} attributed for S=O stretching as sulfonate salts.

FTIR ANALYSIS OF PUFFED RICE (BEFORE REMOVAL)

The peaks at 3422.89 cm^{-1} for surface -OH and -NH stretching, peak at 2924.28 cm^{-1} for aliphatic C-H stretching, peak at 1653.93 cm^{-1} for unsaturated group like alkene, peak at 932.86 cm^{-1} for glycosidic linkage. In fact the FTIR of puffed rice is similar to FTIR of carbohydrates (Figure is not shown).

FTIR ANALYSIS OF PUFFED RICE (AFTER REMOVAL)

In the FTIR spectra of Puffed rice (before removal) peaks at 3422.89 cm^{-1} for surface -OH stretching shifted to 3429.43 cm^{-1} , peak at 2926.01 cm^{-1} for aliphatic C-H stretching, peak at 1658.78 cm^{-1} for unsaturated group like alkene, sharp peak at 1197.79 cm^{-1} in amaranth for S=O stretching is absent in the FTIR spectra of puffed rice after removal (Figure is not shown).

FTIR analysis shows that there is some sort of interaction between -OH groups of puffed rice and -SO_3^- group of amaranth has occurred.

3.3 EFFECT OF PH ON THE REMOVAL PROCESS

The effect of initial pH of amaranth solution onto puffed rice was studied by varying the initial pH (2-8) under constant temperature 30°C . The aqueous solution of dyes having concentration $44.76\text{ }\mu\text{mol/L}$ were treated by 0.1g of puffed rice for 240 minutes. The pH was maintained with the help of 0.1 M HCl and 0.1M NaOH solutions. pH variation had a significant effect on the removal of amaranth by puffed rice. After 240 min. the equilibrium sorption capacity (q_e) was found to be $11.13\text{-}0.79\text{ }\mu\text{mol/g}$ at pH 2-8 (Table-1). It was observed that the initial rate of removal (h) decreases significantly with increasing solution pH. Fig: 4 shows that the highest amaranth removal was detected in pH2. The removal of amaranth is more at lower pH, because in the acidic medium there is an increase of the density positive charge of surface of the puffed rice which would then attract the negatively charged amaranth molecule thereby increasing the removal. The same behavior was observed in many cases [21-22]. Percentage of removal decreases with increase in pH.

3.4 EFFECTS OF INITIAL DYE CONCENTRATION ON THE REMOVAL PROCESS

AR removal at different initial concentration ($44.76\text{ - }1312.50\text{ }\mu\text{mol/L}$). It was found that as the initial dye concentration was increased relative to a fixed sorbent dosage, the extent of removal increased. The equilibrium dye removal (q_e) increases with the increasing initial amaranth concentration. Experimental values were listed in Table- 1. The rapid uptake of dye particles at the beginning is due to the availability of the positively charged surface of puffed rice. Percentage of removal decreases with increase in concentration.

3.5 EFFECT OF SALT ON DYE REMOVAL

The electrostatic attraction is a significant mechanism, as showed by the results where at high ionic strength, the increased amount of KCl can help to render the surface of the puffed rice is not easily available to amaranth molecule hence decreasing the removal of amaranth onto puffed rice. We know from the Gouy-Chapman theory about surface chemistry, when solid adsorbent is in contact with adsorbate species in solution, they are bound to be surrounded by an electrical diffused double layer, the thickness of which is profoundly expanded by the presence of electrolyte. The effects of ionic concentration on the removal kinetics are observed under various ionic strength (0.01M , 0.05M , 0.10M , 0.15M and 0.20M) of KCl solution and other fixed operating conditions (concentration

31.86 μ mole/L, and at pH-2 of the dye solution). The results are shown in Table-1; it indicates that the higher the KCl concentration is, the lower the removal capacity. This agrees with the prediction of the mechanism of electrostatic interactions [23]. Percentage of removal decreases with increase in ionic strength of dye solution.

3.6 EFFECT OF TEMPERATURE ON THE REMOVAL PROCESS

Temperature is an important parameter for a removal process. Removal of amaranth onto puffed rice was investigated at four different temperatures: 30, 35, 40 and 45 °C. It was found that the temperature had effect on the removal process. When the temperature is raised from 30°C to 45°C, the removal AR dye by removal onto Puffed rice increased from 11.47 to 11.52 μ mole/g by increasing the temperature of the solution from 303 to 318K, indicating that the process is endothermic. This may be attributed to increased penetration of reactive dyes inside microspores at higher temperatures or the creation of new active sites. The temperature change the equilibrium capacity (q_e) of AR at 30, 35, 40 and 45°C at the initial concentration of amaranth was 46.44 μ mole/L. experimental values were listed in Table-1. It indicates the removal process endothermic in nature. Similar behavior observed in the case of removal of reactive yellow 84 onto hydroxyapatite in aqueous solution [24]. Percentage of removal increases with increase in temperature.

Table-1 Comparison of calculated and experimental q_e values and kinetic parameters for the AR adsorption onto puffed rice at various pHs, initial concentrations of AR, ionic strength and temperatures.

Parameter	Pseudo-first-order kinetic				Pseudo-second-order kinetic				Elovich kinetic model		
	$q_e(\text{exp.})$ ($\mu\text{mol/g}$)	k1 (min^{-1})	$q_e(\text{cal.})$ ($\mu\text{mol/g}$)	R2	k2 ($\text{g}/\mu\text{mol}/\text{min}$)	$q_e(\text{cal.})$ ($\mu\text{mol/g}$)	h ($\mu\text{mol/g}/\text{min}$)	R2	α ($\mu\text{mole/g}/\text{min}$)	β (g/mole)	R2
pH	Initial AR Concentration 44.76 $\mu\text{mol/L}$										
2	11.13	0.0530	1.27	0.734	0.1609	11.16	20.04	0.999	4.57E+07	2.06	0.701
3	11.07	0.0339	1.00	0.728	0.1332	11.11	16.45	0.999	2.88E+05	1.56	0.737
4	10.93	0.0392	1.24	0.796	0.1319	10.89	15.65	0.999	3.47E+04	1.41	0.827
5	1.54	0.0150	3.66	0.901	0.2901	1.54	0.69	0.999	1.66E+05	12.92	0.958
6	1.13	0.0193	4.46	0.965	0.3789	1.14	0.49	0.999	5.75E+04	16.84	0.962
7	0.85	0.0189	3.80	0.643	1.1741	0.85	0.29	0.999	1.51E+04	21.41	0.982
8	0.79	0.0276	3.97	0.925	0.4003	0.80	0.26	0.989	7.59E+00	11.61	0.940
Initial AR Concentration ($\mu\text{mole/L}$)(pH-2)											
44.76	11.15	0.0309	0.94	0.758	0.1422	11.19	17.79	0.999	4.80E+06	1.82	0.730
163.46	26.44	0.0622	7.70	0.955	0.0270	26.67	19.19	0.999	3.93E+04	0.55	0.908
615.38	31.25	0.0719	9.60	0.872	0.0250	31.25	24.39	0.999	7.82E+03	0.40	0.774
865.38	36.06	0.0352	5.90	0.791	0.0275	36.36	36.36	0.999	1.59E+06	0.50	0.873
1312.50	36.06	0.0306	4.03	0.600	0.1814	36.23	68.49	0.999	5.80E+07	0.60	0.776
Ionic Strength (mole/L), Initial AR Concentration 31.86 $\mu\text{mol/L}$, (pH-2)											
0.001	7.87	0.0325	3.29	0.690	0.4627	7.88	28.74	0.999	3.80E+13	4.72	0.722
0.05	7.83	0.0281	1.90	0.874	0.4099	7.85	25.25	0.999	3.24E+08	3.16	0.705
0.10	7.77	0.0281	1.90	0.874	0.4099	7.81	10.19	0.999	1.20E+07	2.81	0.887
0.15	7.52	0.0221	1.20	0.930	0.0974	7.53	5.52	0.999	3.72E+07	3.13	0.975
0.20	7.51	0.0999	1.28	0.850	0.0999	7.42	5.50	0.999	2.87E+05	2.41	0.968
Temperature ($^{\circ}\text{C}$), Initial AR Concentration 46.44 $\mu\text{mol/L}$, (pH-2)											
30	11.47	0.0306	1.30	0.757	0.1773	11.49	23.42	0.999	6.61E+09	2.43	0.789
35	11.49	0.0276	1.85	0.662	0.2444	11.51	32.36	0.999	3.72E+11	2.78	0.696
40	11.50	0.0272	1.93	0.662	0.2545	11.52	33.78	0.999	6.31E+12	3.04	0.705
45	11.52	0.0290	2.41	0.654	0.3234	11.52	42.92	0.999	1.38E+15	3.52	0.729

3.7 RATE CONSTANT STUDIES

In order to study the kinetics of the removal process, pseudo first-order and pseudo second-order and Elovich kinetics models were used. It is found that the experimental data fitted well to the second-order kinetics than that for the pseudo-first-order kinetics model (Table-1). Similar result obtained for the removal of amaranth from aqueous solutions by bottom ash and de-oiled soya [25].

3.8 ADSORPTION MECHANISM

Corresponding rate constants, the kinetic data were further used by the intraparticle and film diffusion models to determine the rate controlling steps. Weber and Moris's intraparticle model and film diffusion model equation proposed by McKay were used. The experimental results show none of the linear plots at any concentration pass through the origin (Figures are not shown) which means both intraparticle diffusion and film diffusion are the rate limiting steps of the removal of AR onto Puffed rice.

3.9 ACTIVATION PARAMETERS OF THE REMOVAL PROCESS

The activation energy (E_a) for the removal AR onto Puffed rice were calculated by using Arrhenius equation. The value of E_a from the slopes of the plots are 29.58 kJ/mole for AR dye. Which indicating removal process might be physical type. Similar removal type was found in case of the removal of anionic dye from aqueous solution by kernel fiber [26]. To calculate the enthalpy of activation (ΔH^\ddagger), entropy of activation (ΔS^\ddagger) and free energy of activation (ΔG^\ddagger) the Eyring equation (3s) was used.

$$\ln(k_2/T) = -\Delta H^\ddagger/R(1/T) + \ln(k_B/h_p) + \Delta S^\ddagger \quad (3)$$

The slope and y-intercept of the plot $\ln(K_2/T)$ versus $1/T$ (Figure is not shown). The value of ΔH^\ddagger was found to be 27.00 kJ/mole, positive value of ΔH^\ddagger indicates endothermic nature of the removal process. The value of ΔS^\ddagger was estimated to be -55.12 J/mole K, which reflects no significant change occurs in the internal structure of the adsorbent material during removal. Negative value of ΔS^\ddagger is familiar in removal. The positive value of ΔG^\ddagger (not shown) indicated the presence of an energy barrier in the removal process.

3.10 ADSORPTION ISOTHERMS

Adsorption isotherm is generally used to describe the equilibrium relationship between sorbent and sorbate. Three isotherm models such as Tempkin [27] Freundlich [28] and Langmuir [29] were used to describe the equilibrium characteristic of removal. Linear forms of the mentioned three models were used to determine values of the various constants.

Equilibrium removal capacity (q_e) of Puffed rice versus equilibrium concentration of dye in aqueous phase (C_e) (Figure is not shown). It shows removal capacity increases with increasing temperature. This observation agrees with the temperature effect. The rise in removal capacity is due to the increase in collision frequency between adsorbent and adsorbate, which results in the enhanced removal onto the surface of the adsorbent. Values were listed in the Table-2.

Experimental RL values are listed in Table-4 which shows type of removal is favorable. K_T ($\mu\text{mol/L}$) is the Tempkin isotherm constant, b (J/mol) is a constant related to heat of removal. R (8.314J/mol K) is an ideal gas constant and T is a absolute temperature (K). K_F ($(\mu\text{mol/g})(\mu\text{mol/L})^{1/n}$) and n are Freundlich isotherm constant indicating the capacity and intensity of the removal respectively. The values of K_L and a_L were computed from the slopes and y-intercepts of different linearized plots of (C_e/q_e) versus C_e representing temperature (Fig-15). The data shows that the Langmuir removal isotherm provides a good description of data for AR dye over the whole temperature range studied in single systems since R^2 values from Langmuir isotherms (Table-2) are always greater than that of Freundlich isotherms (Table-2) and Tempkin isotherm (Table-2). Thus, it can be concluded that monolayer removal is occurred in this study. The equilibrium constants or binding constants (a_L) obtained from Langmuir isotherms are used to calculate the thermodynamic parameters for the removal process.

Table: 2 Tempkin, Freundlich, and Langmuir isotherm constants at different temperatures and thermodynamic parameters for the adsorption of AR onto puffed rice from aqueous solution at pH 2.

Isotherms	Parameters			
	30	35	40	45
Tempkin				
Temperature (oC)	30	35	40	45
K_T ($\mu\text{mole/L}$)	169.82	87.10	67.61	85.11
b_T (J/mole)	0.0011	0.0013	0.0014	0.0015
R^2	0.973	0.979	0.997	0.998
Freundlich				
$K_F ((\mu\text{mol/g})(\mu\text{mol/L})^{-1/n})$	14.79	15.14	15.49	16.60
b_F (J/mole)	0.12	0.14	0.15	0.15
n	8.30	7.34	6.87	6.86
R^2	0.961	0.969	0.976	0.973
Langmuir				
K_L (L/g)	0.96	1.32	1.88	2.04
a_L ($\mu\text{mole/L}$)	0.026	0.033	0.045	0.045
q_m ($\mu\text{mole/g}$)	36.36	39.53	42.02	44.84
R_L	0.0005	0.0004	0.0003	0.0002
R^2	0.999	0.999	0.999	0.999
Thermodynamic Parameters				
Temperature (K)	303	308	313	318
ΔG (kJ/mole)	-25.64	-26.68	-27.86	-28.36
ΔH (kJ/mole)		30.98		
ΔS (J/mole/K)		187.16		
R^2		0.922		

3.11 THERMODYNAMIC PARAMETERS

Thermodynamic parameters are very important to understand the removal process. Van't Hoff equation is used to calculate the values of thermodynamic parameters (ΔH , ΔS and ΔG). The values of ΔH , ΔS were calculated from the slope and y-intercept of Van't Hoff plot of $\ln a_L$ vs. $1/T$ (Fig: 2). The values of ΔH and ΔS are presented in Table-2. The results show that the changes in enthalpy, ΔH for the removal of AR by Puffed rice were 30.98 kJ/mol. Which means the removal process is physisorption [30]. A positive enthalpy change, ΔH suggests that the removal of AR by Puffed rice is endothermic in nature which is supported by the increasing removal of the dye with the increasing temperature.

The negative values of Gibb's free energy changes (ΔG) (Table: 2) indicate that the removal process is spontaneous. The positive value of ΔS indicates that the randomness increases at the solid-solution interface during the removal of AR onto the Puffed rice.

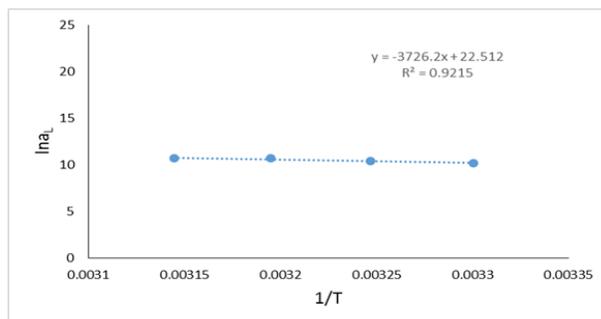


Figure: 2 Activation parameters

3.12 DESORPTION STUDIES

Desorption studies help to understand the nature of removal and recycling of the spent adsorbent and the dye. Certain concentration of dye solution was agitated with certain amount of the adsorbent powder for the equilibrium time after shaking the supernatant dye solution was discarded and the adsorbent was separated and allow to wash with deionized water and then dried. For desorption study 0.1g of that was taken in to a shaking bottle with 25ml of 0.1M NaOH solution and batch removal process was followed. If the Absorbed dye can be desorbed using neutral pH water, then the attachment of dye on the adsorbent is by weak bonds. If sulphuric acid or alkaline water can desorbs the dye, then the removal is by ion exchange. If an organic acid like acetic acid can desorbs the dye, then the dye is held by the adsorbent through chemisorption [31]. AR desorbs in alkaline solution which suggests the ion exchange mechanism involved in the removal. Desorption study also confirmed that the removal process was physisorption.

3.13 REUSE

This step is performed to test the feasibility of Puffed rice as a reusable adsorbent. For reuse study 0.1g of one time used (after desorption) puffed rice was taken in to a shaking bottle with 25ml of 1312.50 μ m/L of dye solution for 300 min, also maintained pH2 and batch removal process was followed. It is found that Puffed rice can be used as reusable adsorbent.

4 CONCLUSION

Present study shows that puffed rice is a good reusable adsorbent for removal of amaranth from aqueous solution. FTIR analysis shows that -OH groups of puffed rice and -SO₃⁻ group of amaranth is mainly involved in the removal process. Temperature effect shows the removal process is endothermic type. The removal process is involved here is physical type in nature and spontaneous. Obtained data were well fitted with pseudo-second order kinetics and can be well described by Langmuir isotherm. The equilibrium monolayer capacity of puffed rice for AR was 36.36 μ mol/g. RL values shows that the process is favorable.

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