RESEARCH ARTICLE

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Removal of Reactive Dyes (Green, Orange, and Yellow) from Aqueous Solutions by Peanut Shell Powder as a Natural Adsorbent

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A-B-S-T-R-A-C-T

Background & Aims of the Study: Textile dyes generally are made of synthetic, organic, and aromatic compounds that may be contain of some heavy metals in their structure. Complex structure and presence of these metals cause toxicity and may be mutagen, teratogen or carcinogen. This study has investigated the ability of peanut shell powder to removal of some reactive dyes (Green 19, Orange 16, and Yellow 14) from aqueous solutions.

Materials & Methods: The effects of contact time, initial concentration of reactive dyes, adsorbent dosage and pH have been reported. The applicability of Langmuir and Freundlich isotherm was tried for the system to completely understand the adsorption isotherm processes.

Results: Batch adsorption studies showed that the peanut shell powder was able to remove the reactive dyes from aqueous solutions in the concentration range 25 to 250 mg/L. The highest percent removal for the Green 19, Orange 16, and Yellow 14 dyes was 84.2%, 87.36% and 88.49%, respectively. The adsorption was favored with maximum adsorption at pH=2. Also the optimum adsorbent dose was obtained 0.4 g/100 mL. By increasing adsorbent dose and initial concentration, removal efficiency was increased considerably. The adsorption isotherm studies clearly indicated that the adsorptive behavior of dyes on peanut shell satisfies only the Freundlich with average R^2=0.926.

Conclusions: Based on findings, the peanut shell powder was found as a low cost, natural and abundant availability adsorbent to removal of reactive dyes from aqueous solution.

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Background

Textile industries are one of the most important sources to produce the various dyeing wastewaters. The concentration of used Azo dyes in textile industries ranges from 10 to 10000 ppm that depends on the dye quality and process of operation (1, 2). A huge quantity of reactive dyes (~30%) are wasted during dyeing process, that discharged into water stream without any effective treatment (3,4).

Textile dyes generally are made of synthetic, organic, and aromatic compounds that may be contain of some heavy metals in their structure. Complex structure and presence of heavy metals cause toxicity and may be mutagen, teratogen or carcinogen (5). Dyes can threaten human health such as skin dermatitis and respiratory system function effects (6).

Dyes can accumulate into the soil and water. Removal of dyes from wastewater effluents is considered as a major problem in textile industries. The Reactive Green 19 (RG-19), Reactive Yellow 14 (RY-14) and Reactive Orange 16 (RO-16) are the most frequent dyes in textile industries (7).

Many methods have been described in the literature for dye removal from wastewater that are including adsorption (*e.g.* active carbon), coagulation–flocculation, chemical oxidation (chlorination, ozonization, *etc.*), photodegradation (UV/H_2O_2, UV/TiO_2, *etc.*) enzymatic treatment and sonochemical processes (8-14).

Nevertheless, the effective and economical removal is an important problem yet. Adsorption process using activated carbon has the high removal efficiency but it is not effective on toxic conditions (15). In spite of it, adsorption process is the simplest, the quickest, the most efficient and economical alternative for removing phenol (16). Because the high cost of activated carbon and its regeneration, its application is limited in low-income developing countries (17,18).

In recent years agricultural and industrial wastes are using to removal of hazardous contaminants from water and wastewater (19). Some of these materials are tire debris, ash volatile, sludge, sawdust and coconut shells (20-22). Before peanut shell powder had been used for biosorption of two azo dyes, amaranth and sunset yellow (23). Peanut shell of agricultural waste is cheap and available by of the 3500 acre acreage in the Iran, annual production of 140 thousand tons of peanuts in the country. It can be as natural adsorbents of cheap and available can be used to remove dyes (24).

Aims of the study: The present study has proposed to use the peanut shell powder to removal of reactive dye Green 19 (RG-19), Yellow 14 (RY-14) and Orange 16 (RO-16) from synthetic aqueous. The effect of various parameters such as pH, coagulant dose and initial dye concentration are investigated.

Materials & Methods

Preparation of Peanut Shell Powder: Peanut seeds were collected around the vicinity of Zahedan city (located in the southeastern Iran). After preparing peanut shell, it was washed with distilled water, dried at 80°C in a hot air oven for 24 h, ground and then sieved with the size 80 mesh ASTM (*i.e.*, American Society for Testing and Materials). The powder was placed in an airtight container for further use.

Dye solution preparation: All used chemicals obtained from Merck Company (Germany). For RG-19, RY-14 and RO-16 Stock solutions preparation, weighted the purified grade chemicals and dissolved in deionized water without pH adjustment. The chemical structure and some of the physicochemical properties of the used dyes are given in Table 1.

Adsorption experiments: At first equilibrium time according to formula (1) determined, and then the major effective variables on adsorption processes were studied. These parameters were including pH, adsorbents dose, contact time and initial dye concentration.

The effect of these variables on the removal efficiency of used dyes examined. Experiments were carried out by shaking the adsorption mixture (120 rpm) at various predetermined intervals and the samples took the supernatants. Then it filtered with 0.45 µm, Watman No. 1, and was analyzed for dye content using an UV-

VIS spectrophotometer (Shimadzu, Tokyo, Japan; Model 1601).

To pH adjusting 2 to 12 were used 1 M H_2So_4 and/or 1 M NaOH. Amount of pH was measured using a pH-meter (model E520, Metrohm Herisau, Switzerland). The employed ranges for various parameters are given in Table 2. Finally, the efficiency of dye (RG-19, RY-14 and RO-16) removal calculated as follows:

(1) $\text{Removal\%} = \frac{(C_i - C_e)}{C_i} \times 100$

(2) $qe = \frac{(C_i - C_e)V}{W}$

Where:

C_i is the initial concentration (mg L^{-1});

C_e is the equilibrium dye concentration (mg L^{-1});

qe is the amount of solute adsorbed at equilibrium per unit weight of adsorbent (mg/g);

V is the volume of dye solution (L); W is the weight of the adsorbent (g).

Table 1) Characteristics of the used dyes

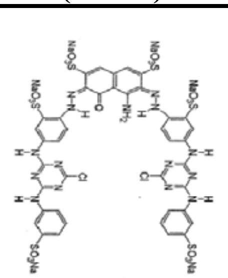
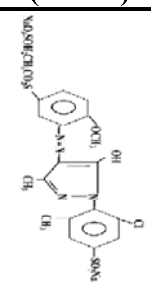
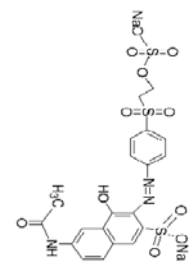
Characteristic	Reactive Orange-16 (RO-16)	Reactive Yellow-14 (RY-14)	Reactive Green-19 (RG-19)
Chemical Structure			
Molecular Formula	$C_{20}H_{17}N_3Na_2O_{11}S_3$	$C_{20}H_{19}ClN_4Na_2O_{11}S_3$	$C_{40}H_{23}Cl_2N_{15}Na_6O_{19}S_6$
Color Index Name	Reactive Orange-16	Reactive Yellow-14	Reactive Green-19
Molecular Weight	g/mol2022.58	669 g/mol	1418.93 g/mol
λ_{max}	496 nm	410 nm	630 nm

Table 2) The ranges of experimental parameters

pH	Adsorbents Dose (g/100 mL)	Dye Concentration, mg L^{-1}	Contact Time (min)	Mixing (rpm)
2-12	0.05, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.8, 0.9	25, 50, 100, 150, 200, 250	30, 45, 60, 75, 90, 105, 120, 135, 150, 180	120

Results

Figure 1 shows the effect of pH on RG-19, RY-14 and RO-16 dye removal from synthetic solutions.

The effects of sorbent dosage on the removal ratio of dyes and contact time effects on adsorption processes were shown in figure 2 and figure 3, respectively.

The influence of initial dye concentration on adsorption percentages of dyes was studied. The results are shown in figure 4.

The results of the isotherm adsorption were shown in Table 3. Freundlich and Langmuir isotherm were studied.

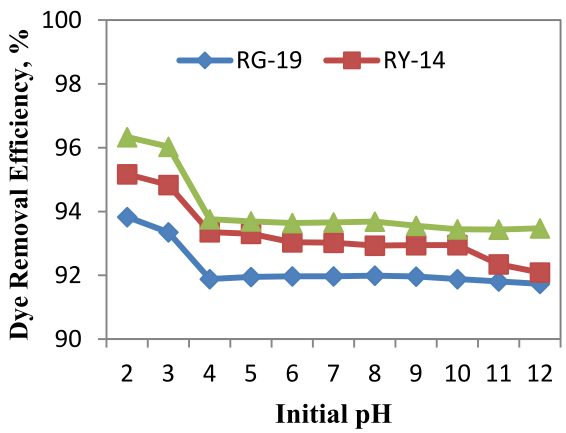


Figure 1) Effect of pH on RG-19, RY-14, and RO-16 dye removal from synthetic solutions (contact time 60 min, Adsorbent dose 0.5 g/100 mL, dye concentration 50 mg L^{-1} and agitation speed 120 rpm).

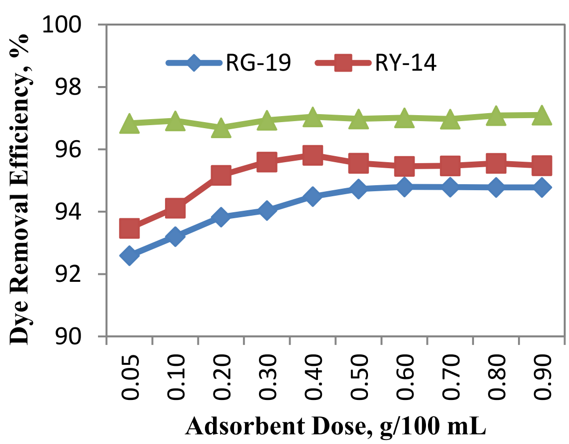


Figure 2) Effect of adsorbent dose on RG-19, RY-14m and RO-16 removal from synthetic solutions (contact time 60 min, dye concentration 50 mg L^{-1}, optimum pH 2 and agitation speed 120 rpm).

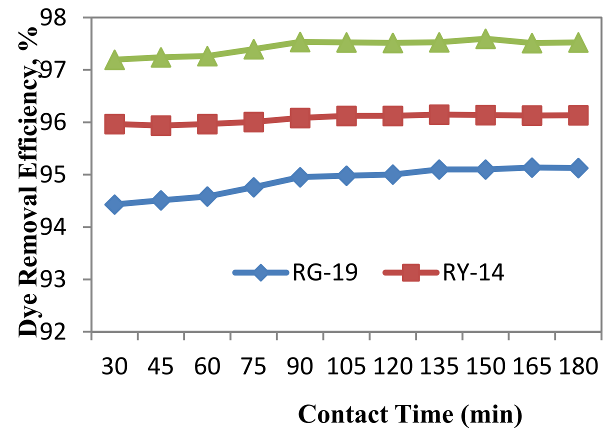


Figure 3) Effect of contact time on dye removal from synthetic solutions (Initial concentration of dye 50 mg L^{-1}, pH 2, adsorbent dose 0.4 g/100 mL and agitation speed 120 rpm).

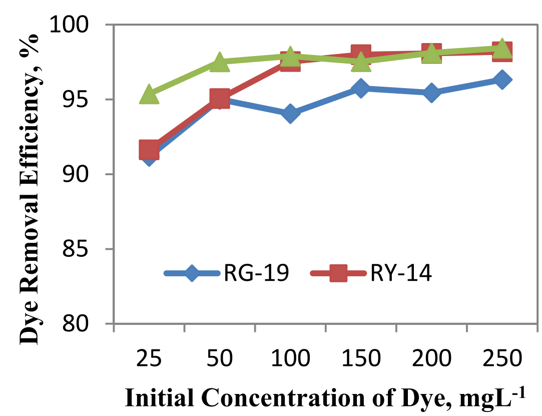


Figure 4) Effect of Initial concentration of dye on dye removal from synthetic solutions (Contact time 90 mg L^{-1}, pH 2, adsorbent dose=0.4 g/100 mL, agitation speed 120 rpm).

Table 3) Characteristics of adsorption isotherms

Isotherms	Freundlich			Langmuir	
Type of dye	1/n	K_f	R^2	B	R^2
RO-16	1/0.54	1.51	0.91	0.538	0.81
RY-14	1/0.21	1.601	0.93	0.574	0.87
RG-19	1/0.38	2.24	0.94	0.774	0.85

Discussion

Effect of initial pH: Solution pH is one of the most critical parameters in the adsorption process and pollutants removal from aqueous solutions (14,25,26). The effects of initial pH on adsorption percentages of dyes were researched over a range of pH values from 2 to 12. As elucidated in figure 3, for all three dyes, the maximum dye removal ratios were at the initial pH 2.The results showed that an increase in pH, the removal efficiency of dye had decreased. The best removal percentage obtained at pH 2, were 96/33, 96/17 and 93/83% for dyes RO-16, RY-14 and RG-19, respectively.

Obtained results at optimum pH values of adsorbent are in good agreement with the values given in literatures such as walnut shell, sawdust, clay, bentonite, native strains (22, 25).

Effect of sorbent dosage: Sorbent dosage has been considered to determine the optimum condition for the performance of adsorption. The removal efficiency of dyes changed with an increase peanut powder dosage from 0.05 to 0.9 g/100 mL. The adsorption ratios of dyes increased from 92.6 to 94.8%, from 93.5 to 95.5%, and from 96.8 to 97.1% in RG-19, RY-14 and RO-16 dyes, respectively. So, the peanut hull mass of 0.4 g/100 mL was chosen for subsequent experiments. The removal efficiency of dyes increased with increasing adsorbent mass which can be attributed to increased surface area and availability of more adsorption sites (27).

Effect of contact time on dye removal: Contact time can be effective on adsorption processes. For a fixed concentration of reactive dyes and a fixed adsorbent mass, the retention reactive dyes increased with increasing contact time. Figure 3 shows that the adsorption rate initially increased rapidly, and that the optimal removal efficiencies were reached 90 min: 97.5%, 96% and 95% for RO-16, RY-14 and RG-19, respectively. In the other studies, similar results were presented (20,22).

Effect of Initial concentration: As shown in figure 4, when the dye concentration was increased from 25 to 250 mg/L, the percentages of dyes adsorption increased from 95.4 to 98.4% in RO-16, from 91.8 to 98.2% in RY-14 and from 91.2 to 98.3% in RG-19. So the equilibrium sorption capacity of the biomass increased with a rise in the initial dyes concentration that this increase in adsorption capacity may be due to the higher adsorption rate and the utilization of all available active sites for adsorption at higher dyes concentration (26). In fact, because of the fixed amount of adsorbent and absorbed positions available, by increasing the initial concentration of dye, removal is increased too. Increasing of adsorption capacity occur as a result of the increased mass transfer (26).

Adsorption isotherms (Langmuir and Freundlich): The adsorption isotherms reveal the specific relation between the concentration of the adsorbate and its adsorption degree onto adsorbent surface at a constant temperature and they are fundamentally important in the design of sorption systems (28).

The adsorption isotherm studies clearly indicated that the adsorptive behavior of dyes on peanut shell satisfies only the Freundlich isotherm with average R^2=0.926 (table 3). The Freundlich model can be applied for non-ideal sorption on heterogeneous surfaces and multilayer sorption. Results of Tanyidizi study on Reactive Black 5 removal with peanut hull, showed that the Langmuir model exhibited fits better to the adsorption data than the Freundlich model (29).

Conclusions: Results of this study showed that peanut shell powder could remove RG-19, RY-14 and RO-16 dyes from aqueous solution effectively. The optimal pH for favorable adsorption of dyes was 2 and removal percentage were 96.3%, 98.2%, and 98.4% for RG-19, RY-14 and RO-16, respectively.

The adsorption equilibriums were reached at about 90 min. Also the optimum adsorbent dose was obtained 0.4 g/100 mL. By increasing adsorbent dose and initial concentration, removal efficiency was increased considerably. The isothermal data fitted the Freundlich model. Based on finding, the peanut shell powder was found as a low cost, natural and abundant availability adsorbent to removal of reactive dyes from aqueous solution.

Footnotes

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Conflict of Interest:

The authors declare no conflict of interest.

References

1. García-Montaño J, Torrades F, García-Hortal JA, Domènec X, Peral J. Combining photo-Fenton process with aerobic sequencing batch reactor for commercial hetero-bireactive dye removal. Appl Catal B Environ 2006;67(1):86-92.

2. Shu HY, Huang CR. Degradation of commercial ago dyes in water using ozonation and UV enhanced ozonation process. Chemosphere 1995;31(8):3813-25.

3. Clarke EA, Anliker R. Organic dyes and pigments. In The Handbook of Environmental Chemistry. Berlin: Springer-Verlag; 1980;3(part A):181-215.

4. Riu J, Schönsee I, Barceló D. Determination of sulfonated azo dyes in water and wastewater. TrAC Trends Anal Chem1997;16(7):405-19.

5. Venkatamohan S, Mamatha VVS, Karthikeyan J. Removal of colour from acid and direct dyes by adsorption onto silica fumes. Fresenius Envion Bull 1998;7(1):51-8.

6. da Silveira Neta JJ, Moreira GC, da Silva CJ, Reis C, Reis EL. Use of polyurethane foams for the removal of the Direct Red 80 and Reactive Blue 21 dyes in aqueous medium. Desalination 2011;281:55–60.

7. Paul J, Naik DB, Sabharwal S. High energy induced decoloration and mineralization of reactive red 120 dye in aqueous solution:a steady state and pulse radiolysis study. Radiat Phys Chem. 2010;79(7):770-6.

8. Merzouk B, Gourich B, Madani K, Vial Ch, Sekki A. Removal of a disperse red dye from synthetic wastewater by chemical coagulation and continuous electrocoagulation. A comparative study. Desalination 2011;272(1-3):246-53.

9. Gholami Borujeni F, Mahvi AH, Naseri S, Faramarzi MA, Nabizadeh R, Alimohammadi M. Application of immobilized horseradish peroxidase for removal and detoxification of azo dye from aqueous solution. Res J Chem Environ 2011;15:217-22.

10. Gholami-Borujeni F, Mahvi AH, Nasseri S, Faramarzi MA, Nabizadeh R, Alimohammadi M. Enzymatic treatment and detoxification of acid orange 7 from textile wastewater. Appl Biochem Biotechnol 2011;165(5-6):1274-84.

11. Dehghani MH, Mesdaghinia AR, Nasseri S, Mahvi AH, Azam K. Application of SCR technology for degradation of reactive yellow dye in aqueous solution. Water Qual Res J Can 2008;43(2/3):183-7.

12. Mahvi AH, Ghanbarian M, Nasseri S, Khairi A. Mineralization and discoloration of textile wastewater by TiO_2 nanoparticles. Desalination 2009;239(1-3):309-16.

13. Maleki A, Mahvi A, Ebrahimi R, Zandsalimi Y. Study of photochemical and sonochemical processes efficiency for degradation of dyes in aqueous solution. Korean J Chem Eng. 2010;27(6):1805-10.

14. Mahvi AH, Heibati B, Yari AR, Vaezi N. Efficiency of Reactive Black 5 dye removals and determination of Isotherm Models in aqueous solution by use of activated carbon made of walnut wood. Res J Chem Environ 2012;16(3):26-30.

15. Pajooheshfar SP, Saeedi M. Adsorptive removal of phenol from contaminated water and wastewater by activated carbon, almond, and walnut shells charcoal. Water Environ Res 2009;81(6):641-8.

16. Rodrigues LA, da Silva MLCP, Alvarez-Mendes MO, Coutinho AR, Thim, GP. Phenol removal from aqueous solution by activated carbon produced from avocado kernel seeds. Chem Eng J 2011;174(1):49-57.

17. Rasoulifard MH, Taheri Qazvini N, Farhangnia E, Heidari A, Doust Mohamadi SMM. [Removal of direct yellow 9 and reactive orange 122 from contaminated water using Chitosan as a polymeric bioadsorbent by adsorption process]. J Color Sci Technol 2010;4(1):17-23. (Full Text in Persian)

18. Asgari G, Sidmohammadi A, Ebrahimi A, Gholami Z, Hosseinzadeh E. [Study on phenol removing by using modified zolite (Clinoptilolite) with FeCl3 from aqueous solutions]. J Health Syst Res 2010;89:848-57. (Full Text in Persian)

19. Ahmadi Moghadam M, Amiri H. [Investigation of TOC removal from industrial wastewaters using electrocoagulation process]. Iran J Health Environ 2010;3(2):185-94. (Full Text in Persian)

20. Daraei H, Manshouri M, Yazdanbakhsh AR. [Removal of Phenol from Aqueous Solution Using Ostrich Feathers Ash]. J Mazand Univ Med Sci 2010;20(79): 81-7 (Full Text in Persian).

21. Shokouhi R, Ebrahimzadeh L, Rahmani AR, Ebrahimi SJAD, Samarghandi MR. [Comparison of the advanced oxidation processes in phenol degradation in laboratory scale]. Water Wastewater 2010;20(4(72));30-5. (Full Text in Persian)

22. Lin SH, Juang RS. Adsorption of phenol and its derivative from water using synthetic resins and low-cost natural adsorbents: A review. J Environ manage 2009;90(3):1336–49.

23. Yang C, KE L, Gong R, Liu H, Sun Y. [Utilization of powdered peanut hull as biosorbent for removal of azo dyes from aqueous solution]. J Biol 2005;2:16. (Full Text in Chinese)

24. Rasekh H, Safarzadeh Vishkaei MN, Asghari J. [Response of yield and qualitative characteristics of peanut (Arachis hypogaea L.) to planting pattern and plant density in Guilan province]. J Agric Sci 2006;12(2):387-96. (Full Text in Persian)

25. Shokoohi R, Vatanpoor V, Zarrabi M, Vatani A. Adsorption of Acid Red 18 (AR18) by Activated Carbon from Poplar Wood - A Kinetic and Equilibrium Study. E J Chem 2010;7(1):65-72.

26. Nagda GK, Diwan AM, Ghole VS. Potential of Tendu leaf refuse for phenol removal in aqueous systems. Appl Ecol Environ Res 2007;5(2):1-9.

27. Rahman IA, Saad B. Utilization of Guava seed as a source of activated carbon for removal of methylene blue from aqueous solution. Malays J Chem 2003;5(1):8-14.

28. Rafatullah M, Sulaiman O, Hashim R, Ahmad A. Adsorption of copper(II), chromium(III), nickel(II) and lead(II) ions from aqueous solutions by meranti sawdust. J Hazard Mater 2009;170(2-3):969–77.

29. Tanyildizi MŞ, Modeling of adsorption isotherms and kinetics of reactive dye from aqueous solution by peanut hull. Chem Eng J 2011;168(3):1234-40.