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# Efficacy of Eosin Dye Removal by Peanut Shell Agrowaste Adsorbent

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**HIGHLIGHTS**

- Peanut shell waste is an alternative adsorbent for the removal dyes.
- Ultrastructural visualization of peanut shell is the fiber structure shell, multi-layers and many linkages among the fibers which could create to various pores with different sizes.
- The best eosin Y removal percentage had 87.7% in 25 g $L^{-1}$ of peanut shell at 30 minute.

**Abstract:** Peanut shell (PS) which is an excessive waste-product from agricultural processes, it can be recycled to a natural adsorbent for example it uses as removal dyes. Synthetic dye effluent without improperly discharged from industries to the river cause wastewater and damage to living organisms, especially, anionic dyes are difficult removed by conventional treatments such as biological, chemical, oxidation, and physical-filtration. However, an adsorption treatment is widely used for decolorization of dyes and give the best results for removal of various types of dissolved coloring materials. This research was used Eosin Y (EO) for the anionic model of dyeing wastewater and used PS for agrowaste adsorbent. The purpose of this study was investigated the efficiency adsorption of EO removal by PS. This efficiency adsorption was measured by different PS dosages, contact times, adsorbate concentration and equilibrium data. The results can be concluded that the PS had the efficiency adsorption of EO removal due to the equilibrium adsorption capacity ($q_e$) and the highest dose of PS were balanced to adsorption of dye. The highest EO removal percentage was found in 87.7%, the $q_e$ was 0.351 mg $g^{-1}$ and can adsorb from 10 mg $L^{-1}$ to 1.23 mg $L^{-1}$ in 25 g $L^{-1}$ of PS dose at 30 minutes. In addition, the PS structure was found in multi-layer and many porous which is suitable for adsorbent. The morphological examination of PS was shown before and after adsorption that not changed. Therefore, PS might be an alternative choice for removal dye, and be used for the recycle adsorbent agrowaste as a commercial product for adding their values.

**Keywords:** Adsorbent; Adsorption; Agrowaste; *Arachis hypogaea*; Eosin Y; Peanut shell.

## INTRODUCTION

The dyeing wastewater causes a big problem water pollution in worldwide. It is produced by many industries such as cosmetic, medical laboratory, paper, leather tanning and plastic [1], which textile manufacturing alone is the most release dyes into river due to it is not completely disposed of dyes during water treatment. It has been reported that the textile industry has the global annual production of dyes amounts to a million metric tons such as from the People's Republic of China and India, they are the largest textile industries in Asia [2]. The most of dye industries are wildly used synthetic dyes to present color to their products. Dyes are chemical compounds, and can be classified into three types according to the nuclear structures i.e., anionic dyes (acidic and reactive), non-ionic dyes (disperse), and cationic dyes (basic) [3]. Mostly, these dyes are dangerous to human health especially the workers who exposed in the work places such as lung eye, mouth and skin irritation [4]. They also have affected on living organisms and ecosystem such as disruption of plant growth or the heavy metal complex dyes are suffering the gills of fishes. However, the advantages of synthetic dyes, they have a wide range of colors for their products. In this study, Eosin Y (EO) was used for a model as anionic dye. EO is potentially problematic compounds from aqueous solution which can lead to wastewater on during several processes such as histological staining. This dye causes toxic, carcinogenic effects on aquatic biota including plants and animals and also humans. According to Sharma and coauthors [5] had reported EO may cause severe skin and eye irritation and while on ingestion that had also effected into vital organs. The inhalation of the dye can reduce the pulmonary gas exchange capacity [6] and the metabolites are also highly toxic and carcinogenic substances [7]. Therefore, it should be necessary to remove EO from aqueous solution.

Technology of wastewater treatment can be divided into three categories based on characteristic and structural properties [8, 9]. First, it is the biological treatment that used microorganism such as bacteria, nematodes, or small organisms to break down organic wastes. Second, it is the chemical treatment that used chlorine, ozone, hydrogen peroxide or etc. Last, it is the physical treatment that used sedimentation, flotation, filtration, separation or adsorption [10]. Among these processes, the adsorption is a popular treatment that used for supporting any treatments to dye effluent manufacturing [11-14]. The dye effluents generated from rubber, plastic, leather and textile processing plants contain different types of synthetic dyes, such as malachite green, methyl violet, azure dye, indigo carmine, methylene blue, eosin and rhodamine [15]. This adsorption process will separate the substances of solution or contaminants, and bind to outer surfaces of solid materials that call adsorbent [16]. Some of the low cost adsorbent used for adsorption are obtained from the domestic, agricultural or industrial sources [17,18]. Our research group is focusing in the natural adsorbents from agricultural waste such as activated carbon, peanut shell, egg shell, banana peel and orange peel. Activated carbon can be produced from wood or coconut which it has been reported that can adsorbed methylene blue from aqueous solution [9]. Pumice and walnut activated carbon can removed reactive black from aqueous solution [12]. The peanut shell (PS) is eco-friendly environment that used in separation of oil and water [19]. Aloe vera adsorbent was removal of hazardous crystal violet [20]. Egg shell is also natural adsorbent for removal of fluoride from aqueous solution [21]. Banana peel is used to adsorption of rhodamine dye and heavy metal from aqueous solution [22]. Moreover, orange peel is also used for adsorption rhodamine dye from aqueous solution [17]. The commercial activated carbon is a popular absorbent for the elimination of variety of dyes [23]. However, it has high costly and can lead to global warming from burning process. The dust from activated carbon can risk to respiratory system. More interesting, PS is a numerous solid waste from agriculture after it is harvested in farm. PS has high volume and low density, therefore, it may be an alternative choice for color removal [24]. In addition, it is inexpensive, easy find in location, and include for recycling of the agricultural waste or agro-waste and want to more adding values [25].

The aim of this study was to use the agrowaste peanut shell as a natural adsorbent and investigated the efficiency adsorption of eosin Y from aqueous solutions. The parameters of adsorption and equilibrium data were studied via various contact times, adsorbent dosages and adsorbate concentrations. In addition, the morphological examination of peanut shell was observed by Field Emission Scanning Electron Microscopes (FE-SEM).

## MATERIAL AND METHODS

### Adsorbent preparation

Peanut was bought from a local market in Bangkok, Thailand. The preparation of peanut shell agrowaste methodology was modified from Arunlertaree and coauthors [26] and Massie and coauthors [27]. First, the peanut was washed thoroughly with tap water to minimize its chromaticity and remove any dirty. Next, it was boiled in 100ºC for 30 min, then removed the shell. The adsorbent as PS was dried in the hot air oven at 65ºC for 24 hours. The dried PS was grinded by blender machine to fine powder, then PS powder was sieved through to 2 mm diameter and stored in glass container.

### Adsorbate preparation

Eosin Y (EO), C20H6Br4Na2O5 (Sigma-Aldrich, USA) was used as model for anionic dye in this study. The EO was molecular weight of 647.89052 g $mol^{-1}$ [28-30]. The stock dye solution was prepared by dissolving 0.1 g of dye powder in 1.0 L of distilled water to give a concentration of 100 mg $L^{-1}$ and diluting when necessary. The 5 ml of 10 mg $L^{-1}$ of dye solution was measured for optical density using UV-visible spectrophotometer wavelength from 400 to 800 nm for range finding and the definitive finding wavelength. The test was done triplicate. The maximal wavelength ($\lambda_{max}$) was recorded and used in this study. The 0, 2, 4, 6, 8 and 10 mg $L^{-1}$ of EO were measured optical density at maximum wavelength for the standard curve.

### Ultrastructural visualization of adsorbent

Morphological examination of PS was performed by using Field Emission Scanning Electron Microscopes (SU8010 FE-SEM, Hitachi, Japan). The PS was mounted on a circular disc-like structure or stub at size 2 inches. Specific carbon tape was used to attach with PS to the surface of the stub, and was coated on platinum-palladium alloy (Pt-Pd) by using ion sputter machine (E102, Hitachi, Japan). The ion sputter operated at 0.1 Pascal Torr, 20 mA for 2.30 minute. The microscopic analysis was operated at 10 kV and 10 µA. Image analysis was used Hitachi SU8000 series SEM software to determine the size of the adsorbent.

### Adsorption of peanut shell

The adsorbent dosages of dried PS powder were 5, 25, and 50 g $L^{-1}$, and the adsorbate EO dose was fixed at 10 mg $L^{-1}$. They were shaken at speed 140 rpm by Orbit Shaker (VRN-480 Gemmy, Taiwan) for 30, 60, 90 and 120 minutes for the contact time experiment in room temperature. After shaking, the absorptive removal of dyes were separated by Whatman® Grade 1 (Merck, Thailand). The each of supernatant was measured by pH meter (Oakton PH 700 Benchtop, USA), and was measured the optical density at the maximum wavelength. The optical density was converted to concentration from the standard curve. The PS power before and after adsorption experiments were dried at 65°C in hot air oven (WiseCube® Fuzzy Control System, Germany) overnight and were collected for the morphological examination by FE-SEM. The amount of equilibrium adsorption, $q_e$ (mg $g^{-1}$), was calculated using the following Equation (1):

$$q_e = \frac{C_i - C_e}{W} V \tag{1}$$

Where $C_i$ and $C_e$ (mg $L^{-1}$) were the liquid-phase concentrations of EO initially and at equilibrium, V (L) was the volume of EO solution, and W (g) was the mass of the dried PS adsorbent.

The percentage removal of EO was calculated using the Equation (2):

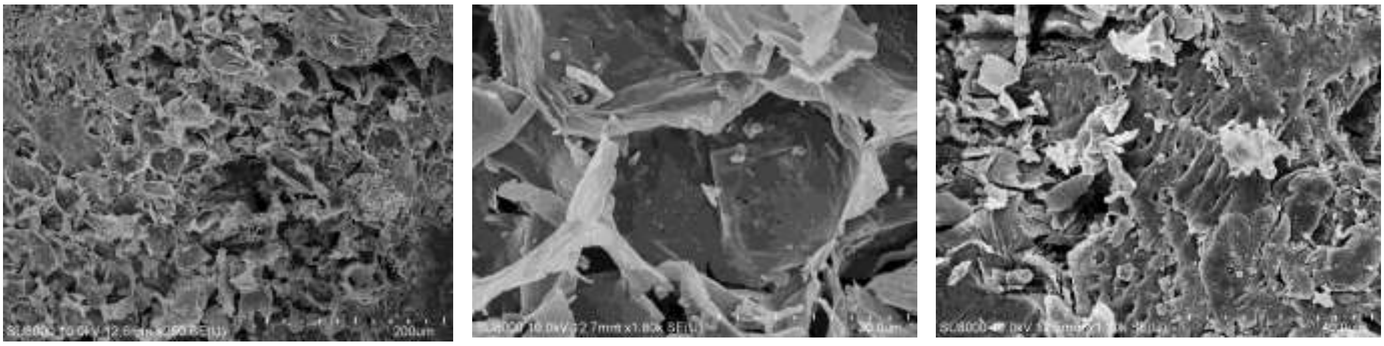
$$\%\ Dry\ removal = \frac{C_i - C_e}{C_i} 100 \tag{2}$$

Where $C_i$ and Ce (mg $L^{-1}$) were the initial and equilibrium concentrations of the EO.

## RESULTS AND DISCUSSION

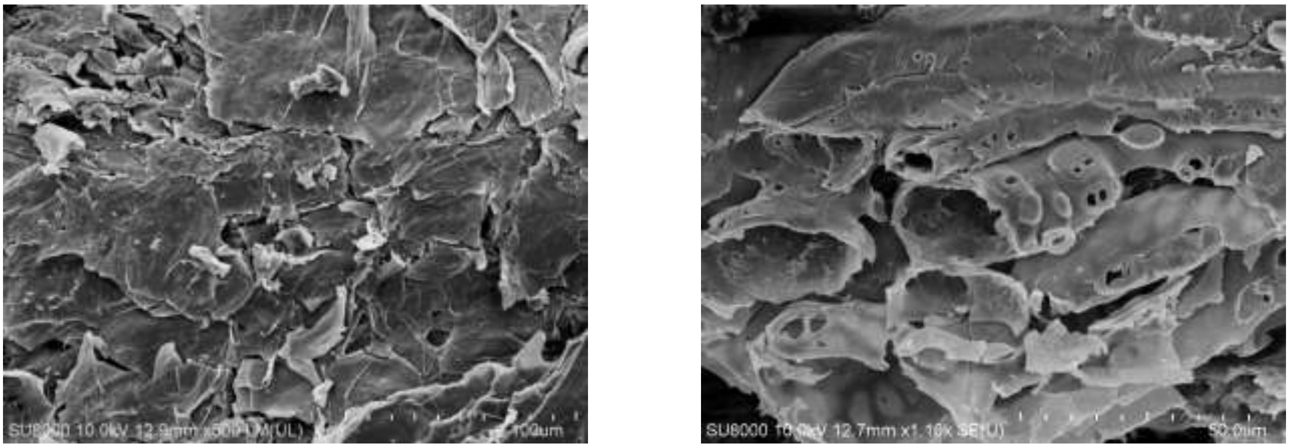
### Characterization of peanut shell

The morphological characterization of PS before adsorption of dye was shown in Figure 1 and after adsorption of dye was shown in Figure 2. PS had the fiber structure shell, multi-layers and many linkages among the fibers which could create to various pores with different sizes. The comparison of PS before and after adsorption for removal EO, both of fiber structures were not changed, they still had many pores. However, the PS after absorptive dye was shown the dye that adhered with the fiber structure and pores. Therefore, the characterization of PS was suitable for bio-adsorbent for EO or any dyes in an anionic group. This result was in agreement with the previous reports. Mojsov and coauthors [31] analysed the morphology of powder peanut hull and used it to adsorb Reactive Black 5, an anionic dye. Xu and coauthors [32] observed the morphology of the activated carbon from PS, and reported that it also had the fiber structure, poly pores and used for removal the organic pollutants from soil eluent. Garga and coauthors [33] also studied the surface morphology of PS and reported that it was showed the porous structure and high amount of cellulose and used for removal Acid Yellow 36, an anionic dye.

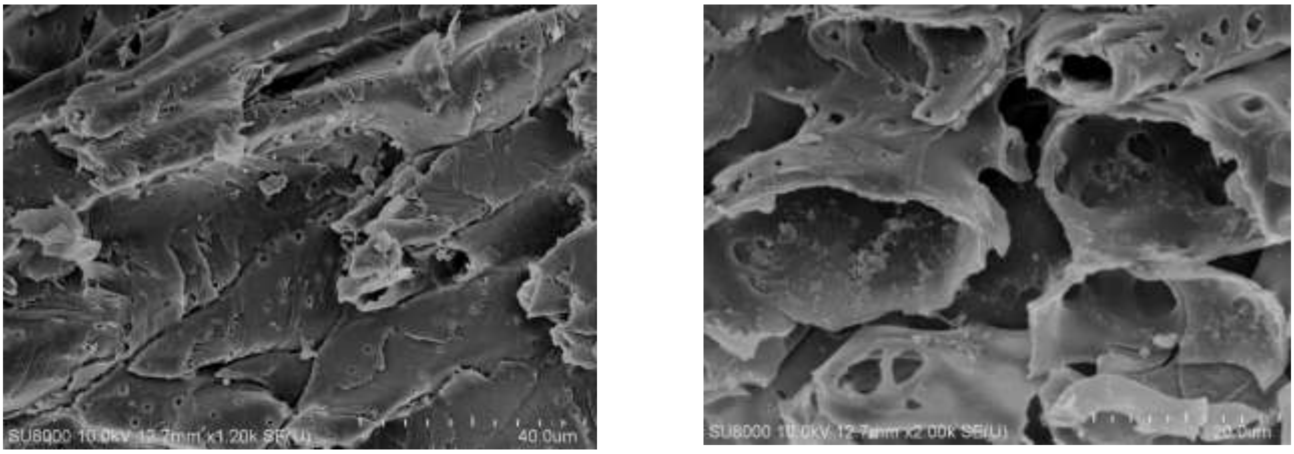


(a) (b) (c)

**Figure 1.** FE-SEM Images of PS before adsorption for EO: (a) PS wasn't grind at High power 12.6 mm x 250 SE(U), size 200 µm; (b) PS wasn't grind at High power 12.7 mm x 1.80 k SE(U), size 30.00 µm; (c) PS was grind at High power 12.5 mm x 1.30k SE(U), size 40.0 µm.



(a) (b)



(c) (d)

**Figure 2.** FE-SEM Images of PS powder after adsorption for EO: (a) Low power 12.9 mm x 500 LM(UL), size 100 µm; (b) High power 12.7 mm x 1.10 k SE(U), size 50.00 µm; (c) High power 12.7 mm x 1.20k SE(U), size 40.0 µm; (d) High power 12.7 mm x 2.00k SE(U), size 20.0 µm.

## Preparation of standard curve for eosin Y solution

The EO solutions in 0, 2, 4, 6, 8 and 10 mg $L^{-1}$, were measured by optical density at maximum wavelength 523 nm, which had the average optical density at 0, 0.106, 0.200, 0.289, 0.388 and 0.477, respectively. The R-squared ($R^2$) of this standard curve was 0.9988 and y= 0.0483x. The graph of standard curve was shown in Figure 3. According to y value of standard curve can be calculated the concentration of absorptive removal dye [34].

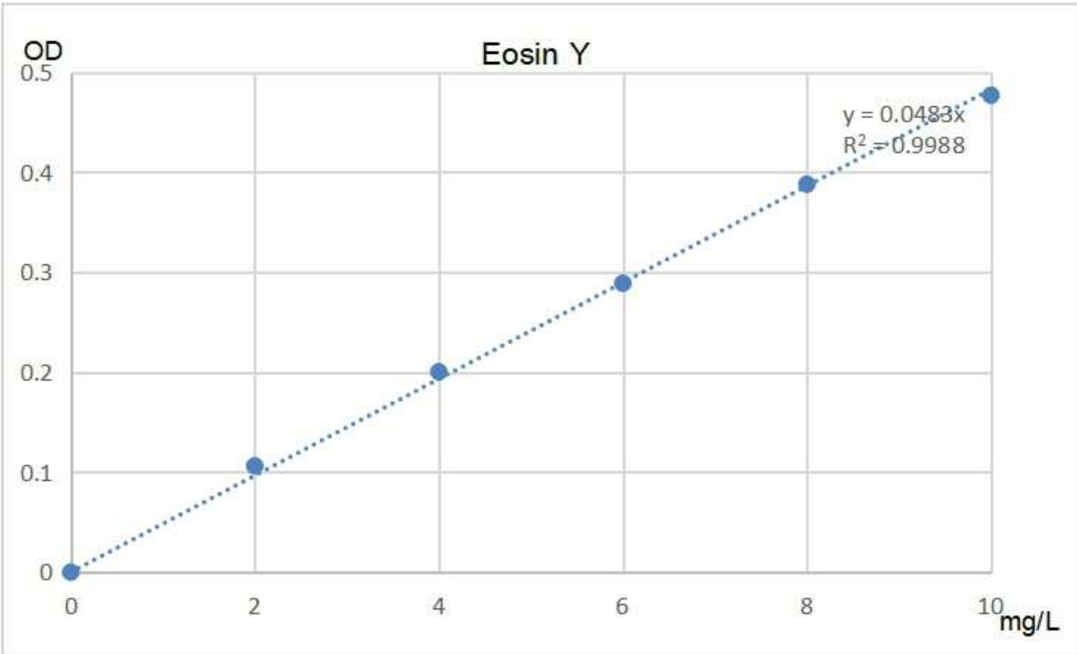



**Figure 3.** Graph of standard curve in various EO solutions at $OD_{523}$.

## Efficiency of peanut shell for removal eosin Y solution

### *Effect of adsorbent dosage*

The effect of PS adsorbent dose for the removal of 100 ml of 10 mg $L^{-1}$ EO was studied by variation in the dose of PS from 5, 25 and 50 g $L^{-1}$. In addition, the contact times for absorbed dye were varied from 30, 60, 90 and 120 minutes. The results were shown in Figure 4. The concentration of absorptive removal dye was calculated by value of y from standard curve on eosin solutions at $OD_{523}$. More addition, the 5 g $L^{-1}$ of

PS can adsorb from the 10 mg $L^{-1}$ EO to 1.82, 1.68, 1.41 and 1.41 mg $L^{-1}$ EO at times 30, 60, 90 and 120 minutes, respectively. The 25 g $L^{-1}$ of PS can adsorb from the 10 mg $L^{-1}$ EO to 1.23, 1.70, 1.70 and 1.73 mg $L^{-1}$ EO at times 30, 60, 90 and 120 minutes, respectively. Finally, the 50 g $L^{-1}$ of PS can adsorb from the 10 mg $L^{-1}$ EO to 1.41, 2.05, 2.51 and 2.39 mg $L^{-1}$ EO at times 30, 60, 90 and 120 minutes, respectively. These results were showed that the efficacy of PS was time and dose dependence.

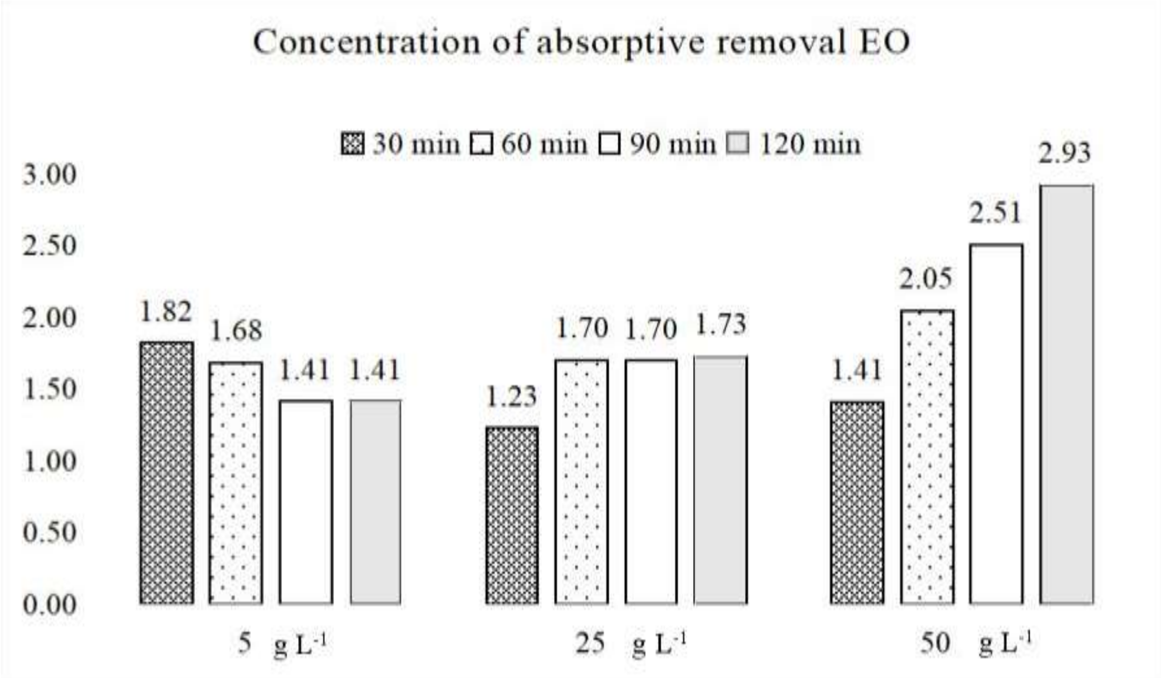



**Figure 4.** Graph of concentration of absorptive removal EO from aqueous solution.

However, at 90 to 120 minutes, the PS were invariable for adsorption. The present results were in agreement with the previous reports. Salleh and coauthors [35] had reviewed the effect of cationic and anionic dye adsorption by agricultural solid wastes for the examples peanut shell, rice husk and activated carbon biochar. The concentration of dyes such as methylene blue, brilliant cresyl blue, neutral red, congo red and reactive black 5 were decreased in low adsorbent dose at the highest time. However, when increased dose of adsorbent, the concentrations of dyes were rapid decreased in lower time [35]. In addition, Sattar and coauthors [36] evaluated the efficiency of peanut shell and peanut shell biochar for removal of arsenic from water, the results of the concentration of arsenic metal were decreased from 5 mg $L^{-1}$ in various adsorbent doses at 2 hour which was associated with equilibrium adsorption capacity that increased adsorbent dose can lead to reduced concentration of dye and reached to equilibrium adsorption at lower time. Moreover, the peanut shell adsorbent was used to removal Acid Yellow 36, at initial dose 100 mg $L^{-1}$ with various adsorbent doses 2 to 6 g $L^{-1}$ at contact times 20 to 160 min [33]. The pH values of EO were measured in before (pH = 4.91) and after adsorption, the results were shown in Figure 5. The pH values of EO after shaking with the 5 g $L^{-1}$ of PS at times 30, 60, 90 and 120 minutes were 4.09, 4.00, 3.93 and 3.90, respectively. Next, the pH values of EO after shaking with the 25 g $L^{-1}$ of PS at times 30, 60, 90 and 120 minutes were 3.57, 3.53, 3.51 and 3.49, respectively. Finally, the pH values of EO after shaking with the 50 g $L^{-1}$ of PS at times 30, 60, 90 and 120 minutes were 3.37, 3.36, 3.36 and 3.33, respectively.

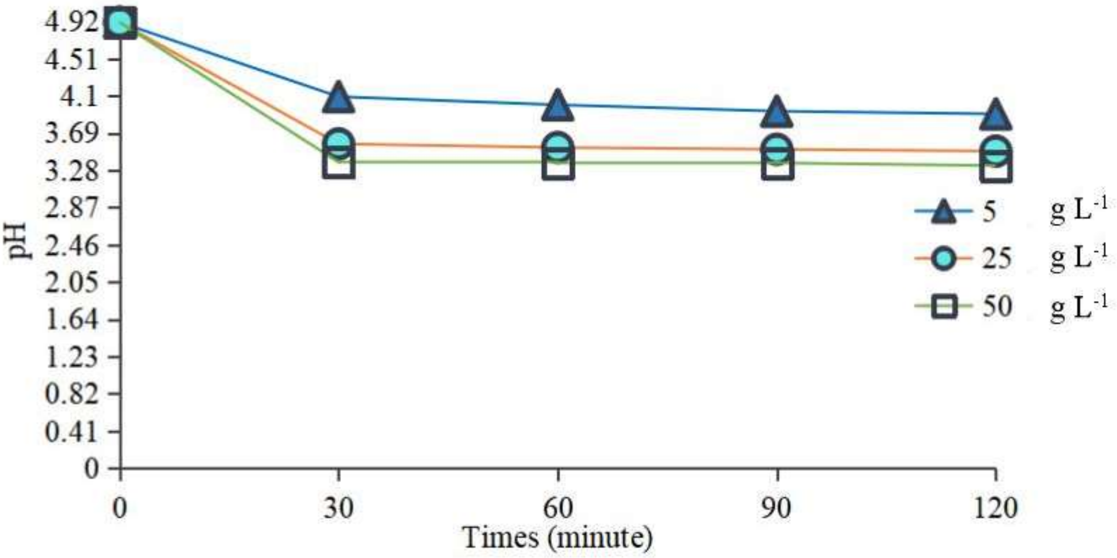



**Figure 5.** pH values after adsorption of EO by PS

The result of pH values were indicated to the adsorptive removal of EO from aqueous solution, there were increased acid in all of adsorbent doses when compared with only dye without adsorption. It might be the ferment of peanut shell that made acid during adsorption for removal dye, and the EO is an acid dye. According to [37, 38], the pH affects the substance surface binding-sites and chemistry of aqueous solutions, and including of the charge on adsorbent surface with the degree of ionization of the dye. The pH of media can change the structure of dye, it gets protonated form in the acidic medium and deprotonated form at higher pH. From the other previous studies, Boumchita and coauthors [23] were adjusted the ranges of pH values of Eriochrome Black T, an anionic dye, they found that the percentage removal of the dye decreased from 99% to 10% as the pH of the solution increased from 2 to 12. Witek-Krowiak and coauthors [39] reported that the pH decreased when the concentrations of protons were increased and competition in binding the active sites on the surface of the biosorbent, by the $H^+$ and metal ions. Protonated active sites were incapable of binding the bind metal ions, leading to free ions remaining in the solution [39]. It might be related to the result of pH value in the present study that shown protonated form, can change the structure of EO. As the pH decreased, the competing effect of H+ ions were increased and the functional groups carrying positive charges were exposed and lead to electrostatic attraction between dye anion and adsorbent [40].

*Effect of contact time*

The effect of contact times on the removal of 100 ml of 10 mg $L^{-1}$ EO by the PS adsorbent were evaluated for various times: 30, 60, 90 and 120 minutes. The amount of equilibrium adsorption, $q_e$ (mg $g^{-1}$), was calculated using the Formula 1. Figure 6 was shown the $q_e$ after shaking with the 5 g $L^{-1}$ of PS at times 30, 60, 90 and 120 minutes were 1.636, 1.664, 1.718 and 1.718 mg $g^{-1}$, respectively. The $q_e$ after shaking with the 25 g $L^{-1}$ of PS at times 30, 60, 90 and 120 minutes were 0.351, 0.332, 0.332 and 0.331 mg $g^{-1}$, respectively. The $q_e$ after shaking with the 50 g $L^{-1}$ of PS at times 30, 60, 90 and 120 minutes were 0.172, 0.159, 0.150 and 0.141 mg $g^{-1}$, respectively. The equilibrium dye uptake capacity ($q_e$) was found to decrease in PS adsorbent ranging from 5, 25 and 50 g $L^{-1}$. The 5 g $L^{-1}$ of PS was slower adsorption capacity in 30 to 60 min, and was approached to equilibrium at 90 min. While, the 25 g $L^{-1}$ of PS was equilibrium adsorption capacity at 30 min and was decreased adsorption of dye from 60 min to 120 min. Moreover, the equilibrium adsorption capacity of 50 g $L^{-1}$ of PS was 30 min and slightly decreased adsorption of EO from 60 min to 120 min. Therefore, the best equilibrium efficiency of adsorption for removal dye by peanut shell adsorbent was 30 min when adding to adsorbent dose.

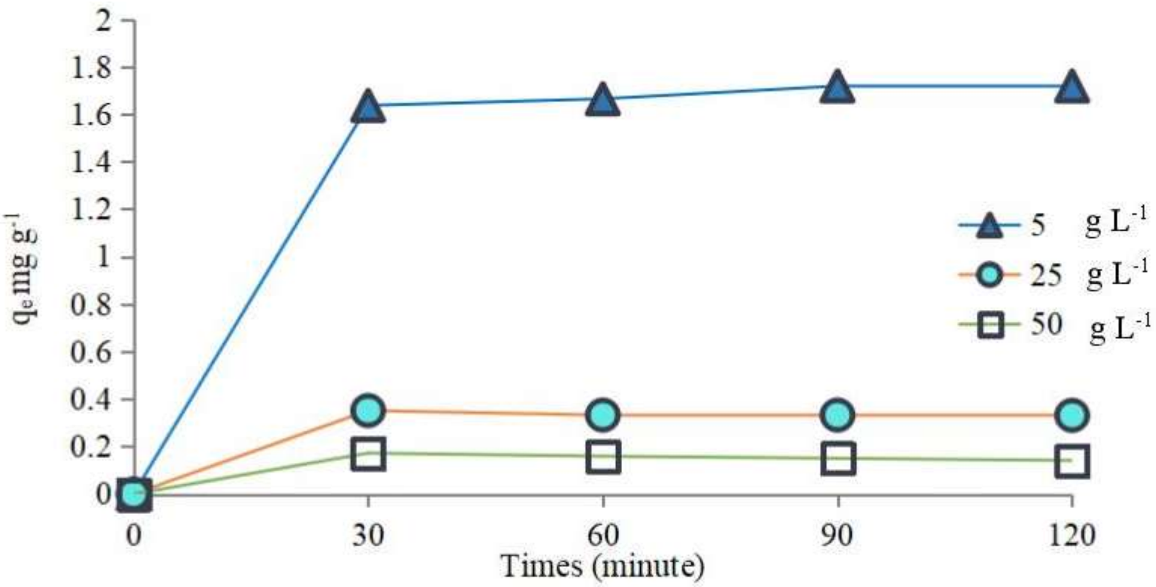



**Figure 6.** The amount of equilibrium adsorption, $q_e$ (mg $g^{-1}$) at various times and different adsorbent doses.

The present results were in agreement with the previous reports. Villarreal and coauthors [41] studied the adsorption capacity of anionic dyes (AB25, AB74 and RB4 dyes) with natural adsorbents (peanut shell and pecan shell). The adsorption capacities ($q_e$) were shown 3.2-8.5 mg $g^{-1}$ with 20 mg $L^{-1}$ initial dye concentrations and 10 g $L^{-1}$ of natural adsorbent dose at various times 20-60 min. According to Gama and coauthors [42] had shown the adsorption capacities of PS with metals that were reduced with the increase in concentration levels of the adsorbent which were shown lower adsorbate/adsorbent ratio. Moreover, the effect equilibrium of adsorption was shown rate slowly increase to reach equilibrium in about 60 min of different concentration of Eriochrome Black T dye (50, 70, 90 and 120 mg $L^{-1}$) with adding to adsorbent dose of PS (0.05 to 2.00 g $L^{-1}$) [10]. In addition, Tahir and coauthors [43] reported the crystal violet (cation) dye was removed with peanut shell by these conditions: 0.1 g adsorbent doses, 50 mg $L^{-1}$ dye initial concentration and 20-120 min contact times, which the equilibrium adsorption capacity was balanced for removal dye at the highest time.

*Percentage of removal eosin dye*

The results of percentage removal (%R) for EO were shown in Figure 7. The %R after shaking with the 5 g $L^{-1}$ of PS at times 30, 60, 90 and 120 minutes were 81.8%, 83.2%, 85.9% and 85.9%, respectively. The %R after shaking with the 25 g $L^{-1}$ of PS at times 30, 60, 90 and 120 minutes were 87.7%, 83.0%, 83.0% and 82.7%, respectively. The %R after shaking with the 50 g $L^{-1}$ of PS at times 30, 60, 90 and 120 minutes were 85.9%, 79.5%, 74.9% and 70.7%, respectively. The result was indicated the percentage of removal EO that shown the best adsorption in 25 g $L^{-1}$ of PS.

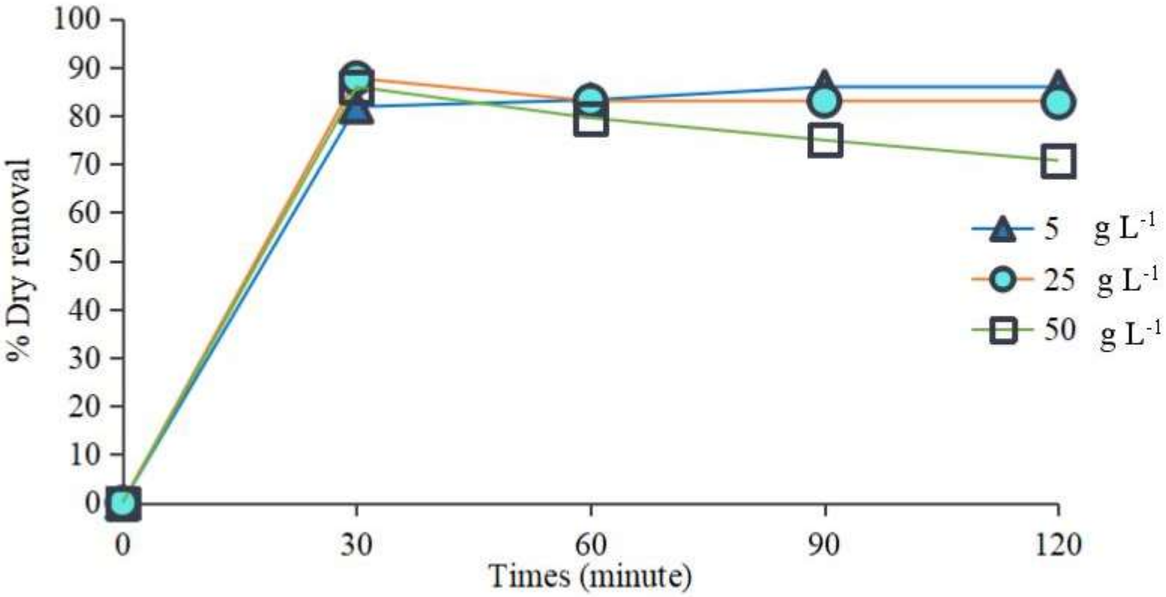



**Figure 7.** Percentage removal (% R) for eosin dye.

The present results were in agreement with the previous reports. Boumchita and coauthors [23] evaluated the efficiency of PS adsorbent with Acid Yellow 36 dye. They reported that the removal of Acid Yellow 36 dye was shown the maximum efficiency at 30 min contact time with the percentage of removal was 50% with an adsorbent dose of 2 g $L^{-1}$. Moreover, Garga and coauthors [33] analysed the effect of 50-120 mg $L^{-1}$ the initial concentration of Eriochrome Black T and the fixed concentration at 2 g $L^{-1}$ of peanut shell. The percentage of removal was decreased with the increase in the initial dye concentration.

## CONCLUSION

The present study was concluded the efficiency of anionic dye (Eosin Y) adsorption onto dried peanut shell as an adsorbent. Due to the peanut shell is an ago-waste, it might be suitable for recyclable waste. The main finding such as the morphological of peanut shell which was found multi-layers of fiber structure. This structure has be created many porous and different sizes. So it is the great characteristic for the adsorptive process. Moreover, the parameters that were used for the efficiency evaluation such as different adsorbent doses, various contact times and initial dye concentration have also been importance. In addition, the Eosin Y is an acid dye, during the adsorption, the pH is more acid in the high dose and in the maximum contact time. Therefore, the PS might be a good alternative bio-sorbent, low cost and has the suitable structure for adhered with Eosin Y dye which it might be depended on electron charge of anionic (negative) dye that bind with positive charge in peanut shell.

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**Conflicts of Interest:** The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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