Comparative between different Natural Sources of Activated Carbon for the Removal Reactive Green Dye from Aqueous Solution

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Abstract

Water pollution is one of the biggest problems facing us, so it requires the need to develop a very high-efficiency and inexpensive absorbent material from natural surfaces and compare it with expensive commercial sorbents. Where in this study an absorbent substance was prepared from the natural surfaces of Syzgium Aromaticum Flower Bud (SAFB), Nigella Sativa (NS) Converted to activated carbon to activated carbon Activated (SAFB), and Activated (NS), as these surfaces are characterized by high porosity, cheap price and very high efficiency in removing dye. The composite was illustrated via different analysis apparatus included FTIR, and FE-SEM, The adsorption result were fitted with Freundlich isotherms. The experimental kinetic result at different primary reactive green dye concentrations, were also analyzed through first model, second model and Elovich model. The obtained data appear that the model pseudo-first model fits the adsorption kinetic result through R^2 0.96028, R^2 0.9486, R^2 0.9016 and R^2 0.9620 of SAFB and ACTIVE-SAFB and Nigella Sativa (NS) and Activated (NS) in the same order.

Key Words: Adsorption, Activated Carbons, Dye, Kinetic, Equilibrium.

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Introduction

Water pollution becomes a serious global concern threatening the entire biosphere and affecting the lives of many millions of people around the world. Water contamination causes a variety of diseases and millions of people die annually because of illness related to the dirty water. The dye contaminant is the common water pollution which should be treated before their drop (Aljeboree 2019). The small amounts of dyes (below 1 ppm) change the color and quality of water and considerably influence the water environment. The current traditional methods change one kind of contamination in to another. like, adsorbing polluting from water in to the adsorbent solid cleared water, but relocate the contamination onto the solid form. Now the adsorbent is a novel contamination that should be treated. In increment, imitative treatments water method need a convoluted design.

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Thus researchers are having for ecofriendly treatment way for degradation of the contamination instead of converting them from one form to another (Y.F. Mestre 2001). Adsorption can be an advantageous method over other approaches (filtration, coagulation, ion-exchange, precipitation, osmosis reverse, and oxidative methods), mostly as it is design simple and minimum cost system, in addition to effective towards a wide series of contaminants (Liu, Wang et al.). The option of adsorbent reuse is another desirable aspect (Pasamontes and Callao 2004). Presently, more than a few studies have been devoted to preparing adsorbent materials for treatment waste water (Pasamontes and Callao 2003; M.H. Chiu, et al. 2009). Natural materials, agricultural, and industrial wastes have been utilized as adsorbents very minimum cost (K. Shah 2017; Y. Peng 2017). Too, advanced engineered materials like as AC, GO, CNT, hydrogels and silica based materials, have found broad applicability in this research field (Sultan 2003; De Baere and De Backer 2007; Ayad F. Alkaim 2016). In this article, it is significant to remind that the utilize of developed and ultra-treated materials can suppress the minimum cost label often related with adsorption.

Experimental Part

Activated Carbons

Natural surfaces like Syzgium Aromaticum Flower Bud (SAFB), Nigella Sativa (NS) was washed, depicted, crushed and eventually ground to a size of 0.5- 3.0 mm in a laboratory mill and then dried in an air oven. A typical activation scheme was accompanied: H2SO4 impregnation followed by 300^{o}C carbonization at 1 hr. Past experience has shown that this temperature is ideal for many agricultural raw materials. The sample was washed different times with D.W until a neutral filtrate was obtained, then dried at 110 $_{0}$C to a constant weight and placed in a clean, dry glass container.

Adsorption Studies

Obtain it maximum removal efficiency, all tests are carried out in many sets. Each set contains 0.05 gm of (SAFB), Activated (SAFB), (NS) and Activated (NS) in different stoppered conical flasks, to each flasks 100ml from freshly prepared BG dye solution of primary conc. (50 mg.L^{-1}). The shaking Incubator used the shaking samples at 160 rmp at 60 min. then separated by centrifugation at 6000 rpm for 10 min. and the solutions you obtained reading by, UV-Visible spectrophotometer PC 1800 at λmax = 625 nm and used the following equation to determined adsorption capacity at equilibrium:

$$qe = \frac{Vsol.(Co - Ce)}{m} \qquad (1)$$

The adsorption efficiency and percentage removal of the dye on the adsorbents were calculated according to the following equations:

$$\% E = \frac{(Co - Ce)}{Co} x100 \qquad (2)$$

Where qe (mg.g-1) is the amount of dye adsorbed on to the adsorbent at equilibrium, Co (mg.L^{-1}) and Ce (mg.L^{-1}) are the primary and equilibrated dye concentration, at the same order, V(L) is the volume of dye solution , m (g) is the weight of the surface.

Results and Discussion

FT-IR Characterization for Adsorbent/Adsorbate

The A(ASFB) and A(NS) was detected using F.T-IR spectroscopy. F.T-IR spectra in the mid-IR range of 4000 to 400 cm-1 were obtained with a resolution of 1 cm^{-1}. Fig. 1 a and b shows the FT-IR A(NS) and A(ASFB) range before and after RG dye adsorption. Near the adsorption, the band intensity of the F.T-IR pattern appears to decrease, As a consequence, there is a substantial difference between (AC) before and after RG interaction, leading to the hypothesis that a phy-sorption mechanism occurs as information of the attractive forces under examination between the (AC) surface and RG dye. (Ayad F. Alkaim 2013; Enas M Alrobayi 2017; Ibrahim Jooda Sahib 2017).

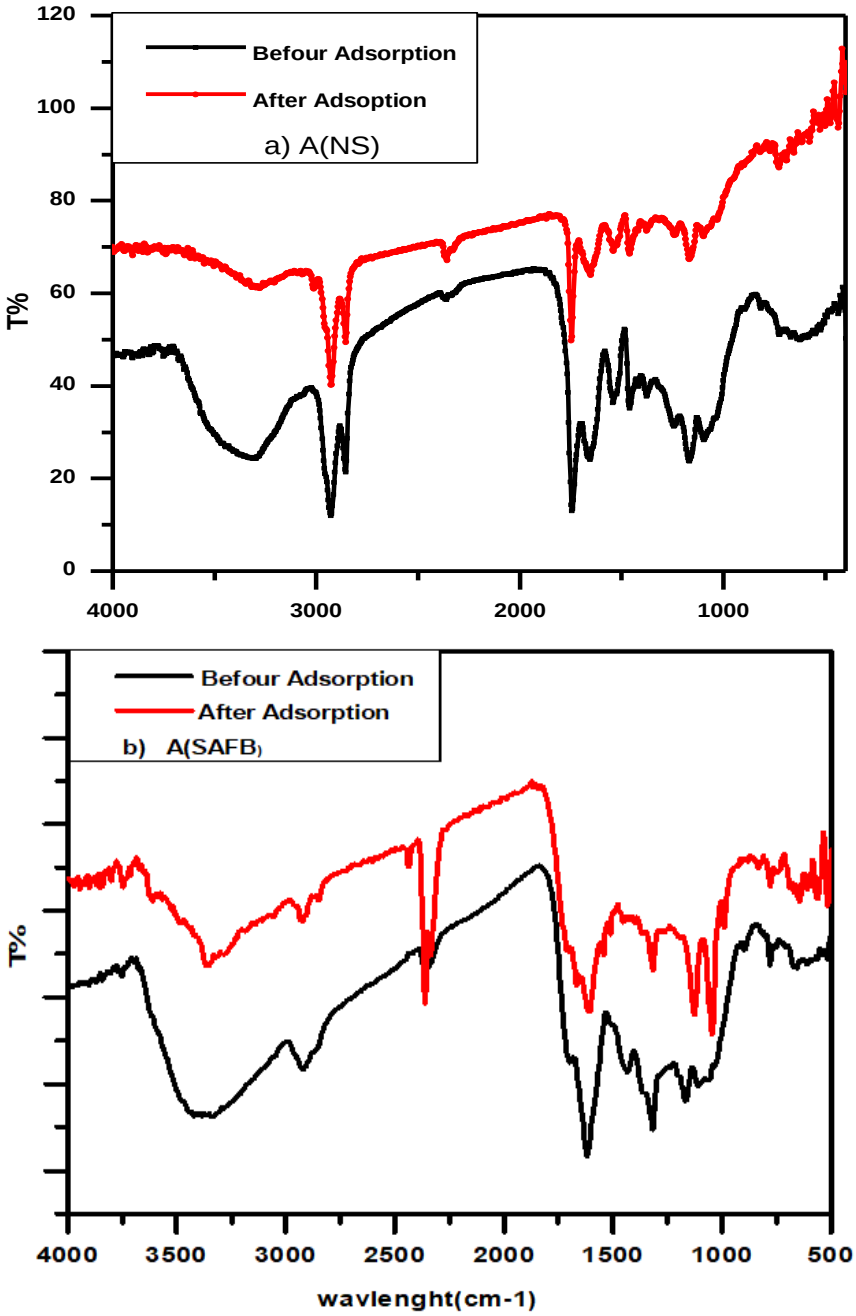

Figure 1. FTIR-spectra of a) A(NS) and b) A(ASFB) before and after adsorption of RG dye.

Field Emission Scanning Electron Microscopy (FE-SEM)

The scanning electron micrograph of RPAC at 200 magnification is shown in Fig. 3. The image shows that a) and c) NS before Adsorption b) and d) Activated NS after Adsorption We notice that the surfaces before adsorption from figure a) and c) are in the form of small clusters with each other as the activated surface A(NS) these clusters are more clustered with each other compared to the non-activated surface (NS). However, after adsorption from figure b) and d) , it is noticed that the surface is more swollen, which is an indication of the dye loading inside the surface. The surfaces can be compared after adsorption as the activated surface A(NS) is more swollen and this is evidence of the increase in the surface area and the increase in the effective sites (Layth S. Jasim 2021).

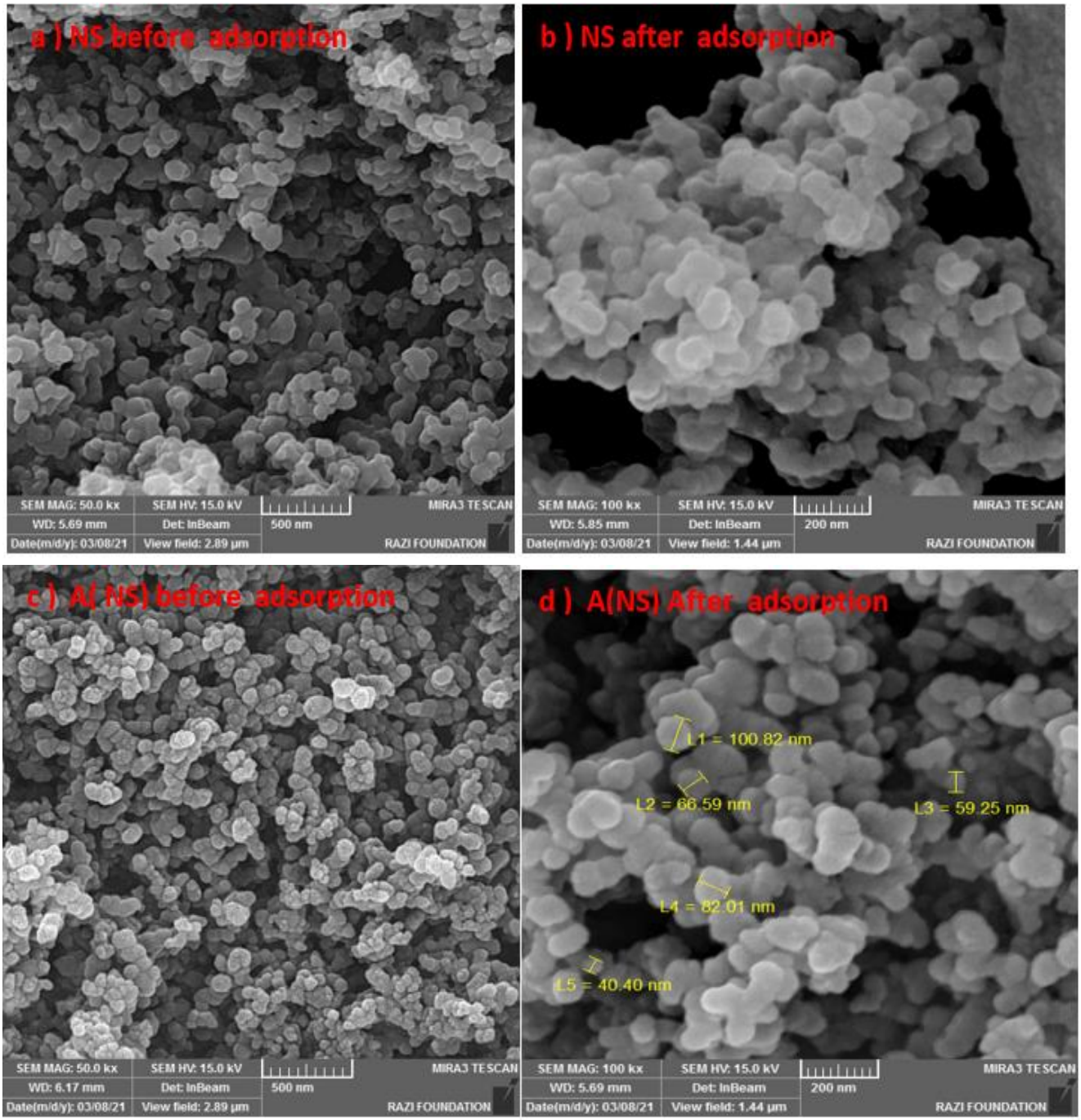

Figure 2. FESEM image of (NS) (a) and A(NS) (b) after and before adsorption of RG

It is noticed through the figure 3 a) and c) that the activated A(ASFB) and non-activated (ASFB) surface before adsorption contains many cavities non regular, but after the adsorption process we notice that all these cavities are full and the shape became more regular and resembles a cloud, and this is evidence of the fullness the surfaces cavities into dye (Aseel M Aljeboree 2021).

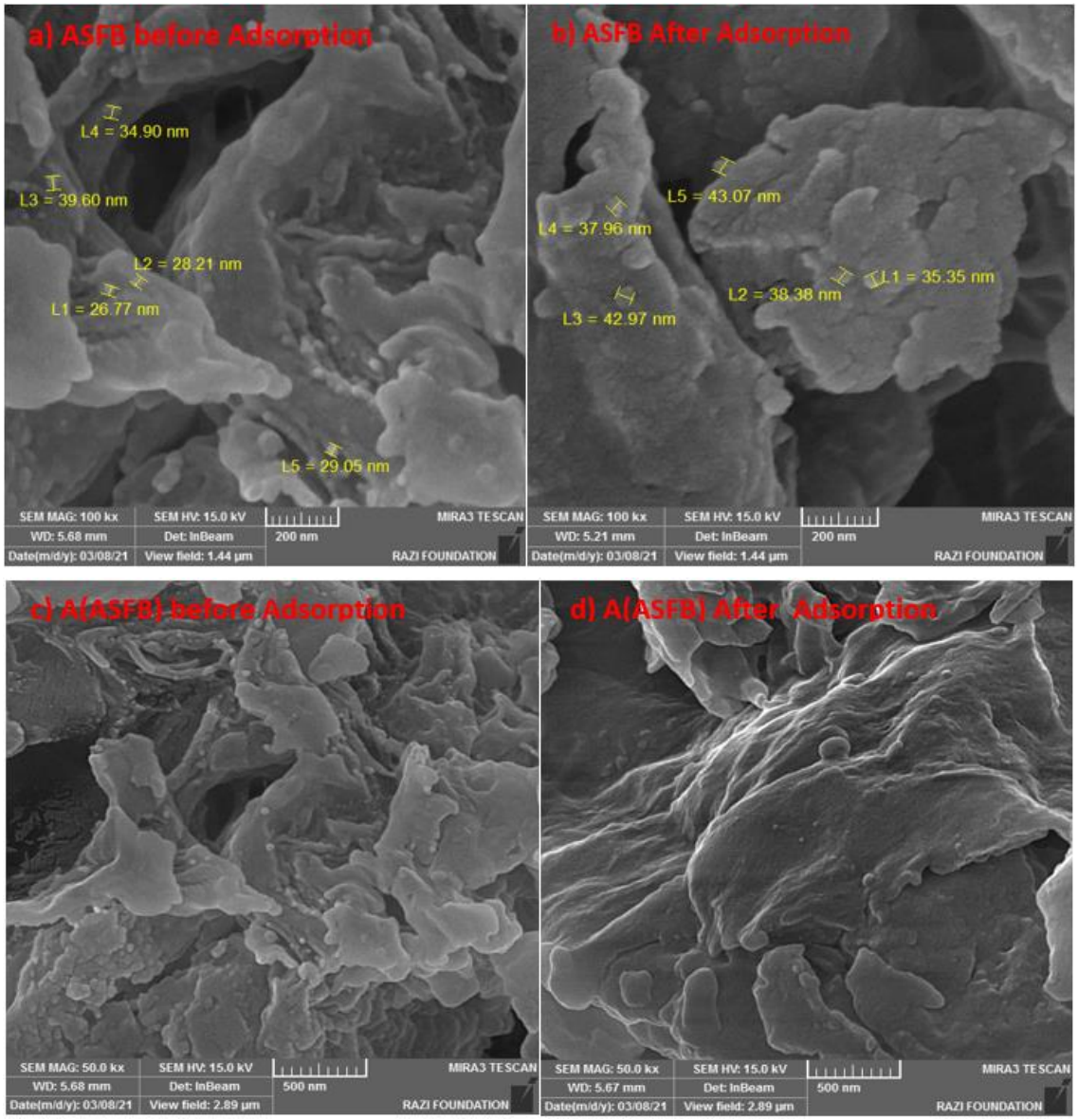

Figure 3. FESEM image of (SAFB) (a) and A(SAFB)(b) after and before adsorption of RG.

Compare Study of different Surfaces

A comparison between the prepared natural and activated surfaces was studied, where it was noticed that the activated surfaces were of high efficiency compared to non-activated surfaces, due to the fact that the activated surfaces are characterized by high porosity and increase the effective sites and increase the surface area so that it has the ability to retain the dye inside. The top data of removal (E%) for dye the order increasing: A(SAFB) > (SAFB) > A(NS) >NS (Aseel M. ALjeboree 2019). shows in Figure 4.

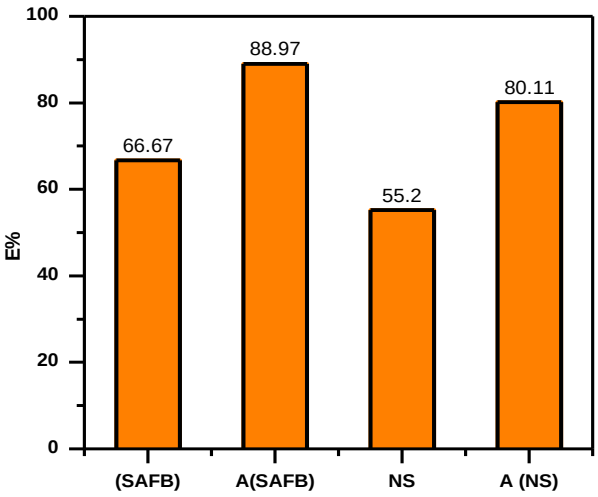

Figure 4. Comparative study of different surfaces at 20°C, 1hr and 0.05g in (50 mg/L) 100 mL water.

Kinetic Models

Kinetics Adsorption provides input about the mechanism of adsorption and possibility of steps controlling. That fundamental for the practical design of adsorption methods. The experimental result were analyzed utilizing the first model, second model and Elkovich kinetic models. The linear expression of the first model is given by:

$$q_t=q_e[1-\exp(k_f t)] \qquad (3)$$

where qe is the quantity of RG adsorbed (mg/g) at equilibrium, qt is the quantity of RG adsorbed at time t (min) and k_1 is the rate constant of adsorption of first model. The values of the rate constant, k_1, were determined from the linear plots of the graph and their values are given in Table 3.

The kinetics of the adsorption method may also be described in pseudo-second order rate equation (Ho 1998). The linearized form of t equation is expressed as:

$$qt = \frac{K2qe2t}{1+K2qet} \qquad (4)$$

The kinetic result from first model, second model and Elkovich model adsorption kinetic, are given in Table 1. The nonlinear plots of qt versus t indicated a best arrangement among the experimental and calculated qe values for several primary A(SAFB), (SAFB), A(NS), NS concentrations. Also, the correlation coefficients of the pseudo- First kinetic model (R^2 ⩾0.96028), (R^2 ⩾0.916), (R^2 ⩾0.9486), (R^2 ⩾0.9620) at the same order). N adher D. Radhy 2019; Waleed K. Abdulsahib 2020 (Through practical experiments, the kinetic models for absorption of dye were accurately determined on the union surface A(SAFB), (SAFB), A(NS), NS by analyzing the three false models of the first and second-order as in Figure (5). (Adriano 2005; A. Gulkowska 2008).

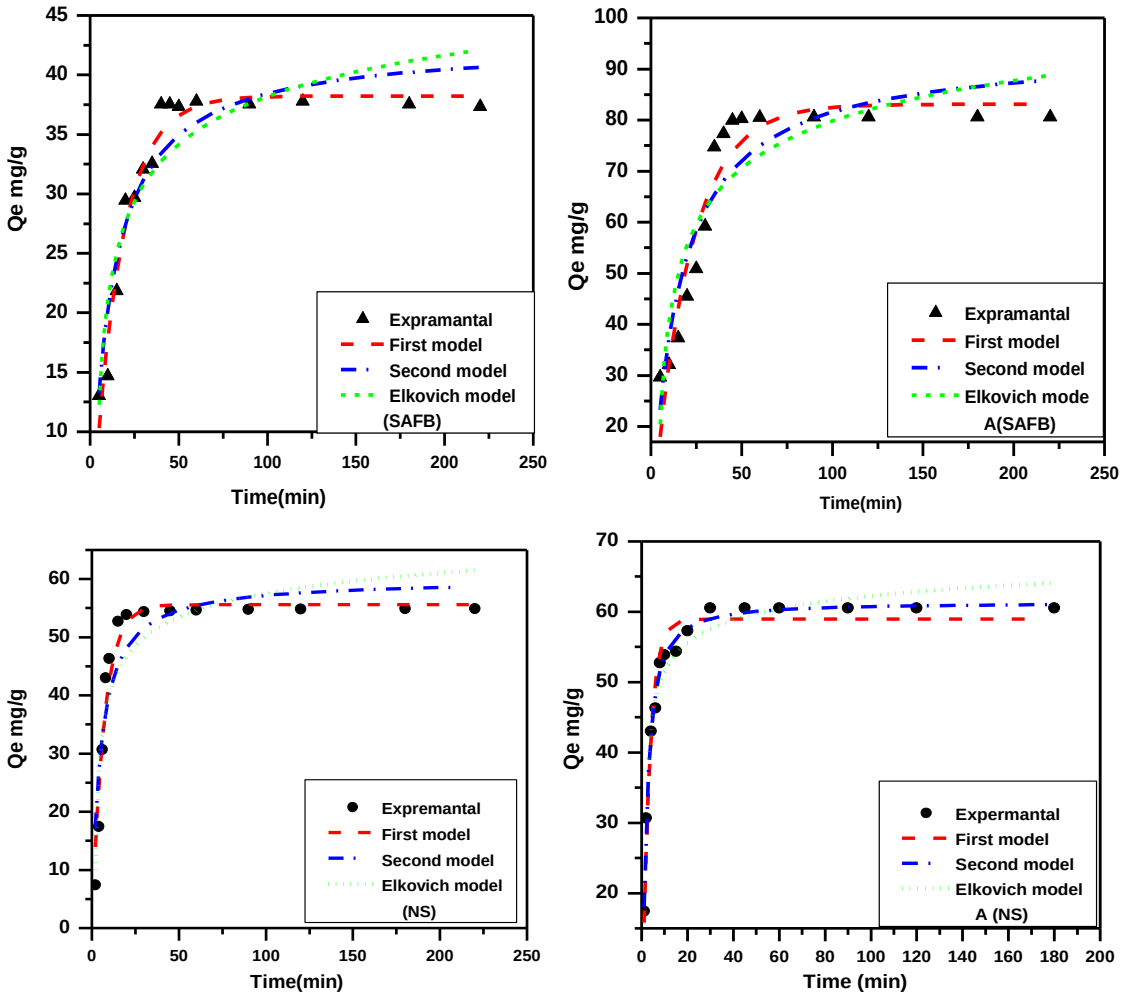

Figure 5. Kinetics model of dye adsorption on A(SAFB), (SAFB), A(NS), NS. Conditions: adsorbent dosage 0.05 gm and temperature 20°C

Table 1. Adsorption kinetics factors the adsorption of RG dye

Model	Equation	Parameters	Value (NS)	Value A(NS)	Value (SAFB)	Value A(SAFB)
First-order	$q_t=q_e[1-\exp(k_f t)]$	K1(min-1)	0.14426	0.120	0.0628	0.916
		qe(calc)(mgg-1)	55.5901	58.962	38.251	83.0989
		R2	0.9486	0.9620	0.96028	0.9136
Second-order	$q_t=\frac{K2qe2t}{1+K2qet}$	K2(g/mg/min)	0.8629	0.4141	0.08945	0.0666
		qe(calc)(mgg-1)	59.9644	61.276	42.6422	93.883
		R2	0.86295	0.9121	0.8945	0.8576
Elkovich model	$q_t=\frac{1}{\beta}1\beta\ln(\alpha\beta)+\frac{1}{\beta}\ln t$	α (mg g−1 min−1)	25.0981	15.454	24.499	56.486
		β (g min−1)	2.1487	1.245	1.0222	0.8918
		R2	0.8556	0.821	0.8499	0.7958

Adsorption Model

Numerous isotherm model equations are existing for investigating sorption equilibrium factor, and the utmost public being is the isotherms Freundlich, Langmuir, The model of Langmuir isotherm (Figure 6) is built on the theory that there is a fixed quantity of active sites, which regularly dispersed over the surface of the adsorbent; these sites have identical desirability for adsorption of a monomolecular layer and no interaction among adsorbed molecules. The Freundlich isotherm (Figure 6) applicable for heterogeneous surface adsorption. This model supposes a positive relationship among adsorbate concentration adsorbent quantities on the surface. Similarly, the energy sorption proportionally declines at the end of the sorption centers of the adsorbent. Calculation of correlation coefficients done via fitting the experimental equilibrium result for the dye- A(ASFB) and dye- A(NS) system utilizing Langmuir, Freundlich isotherms. Figures 5 and Table (2) appear the highest correlation coefficients (R^2 = 0.9769) and (R^2 = 0.9777) related to the model Langmuir (Langmuir 1918; Adeyemo, Adeoye et al. 2017; Hayder Obaid Jamel 2017; Aseel M Aljeboree 2019; Alkaim and Aljobree 2020).

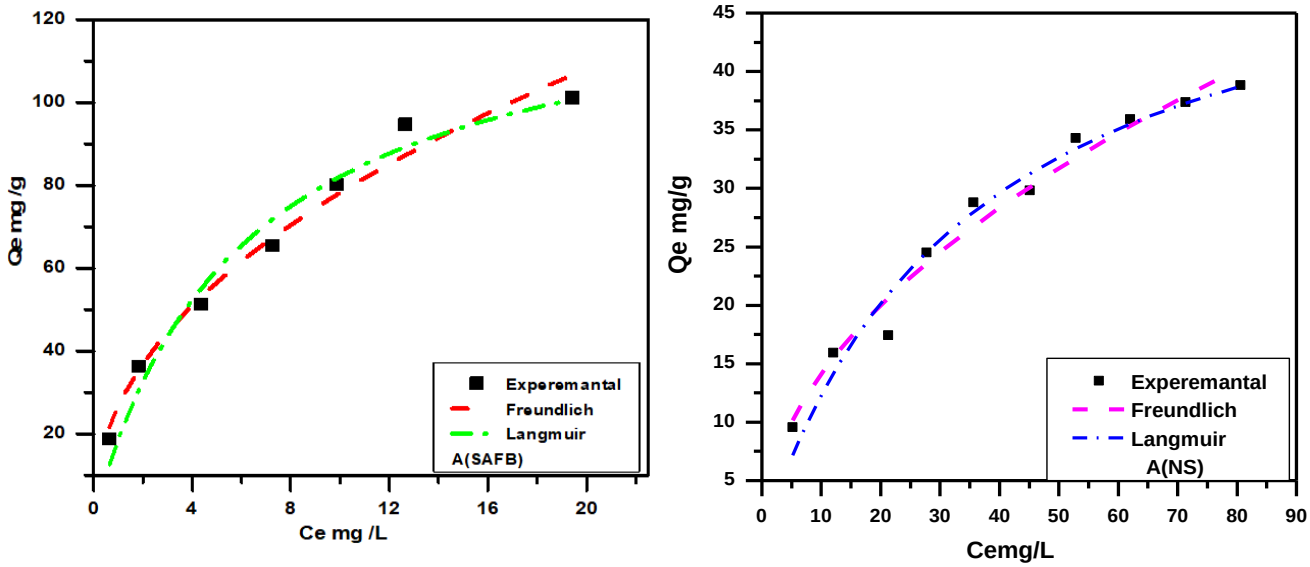

Figure 6. Several adsorption models nonlinear fit of adsorption RG dye on A(SAFB) and A(NS) primary conc. = 50 mg. L^{-1}, Temp. = 20°C, and weight of surface 0.05 g/L).

Table 1. Freundlich and Langmuir isotherms model factors RG dye adsorbed on the surface of A(SAFB) and A(NS) at 20°C.

Isotherm models	Parameters	A(NS)	ACTIVE-SAFB
Freundlich	K_F	4.5111 ± 0.541	26.706 ± 2.481
	1/n	0.5111 ± 0.022	0.466 ± 0.037
	R^2	0.9777	0.97694
Langmuir	q_m (mg/g)	65.771 ± 2.0121	132.410 ± 12.209
	K_L(L/mg)	0.0311 ± 0.002	0.163 ± 0.038
	R^2	0.8133	0.96622

Conclusion

1. Good removal of Reactive green dye from aqueous solution via utilizing A(SAFB).
2. comparative between the Maximum adsorption of removal percentage 66.67% of (SAFB) and 88.97% of ACTIVE-SAFB .and 55.2% of (NS) and 80.12% of ACTIVE-NS.
3. The best removal percentage of ACTIVE-SAFB. Comparative between of ACTIVE-NS. Thus Due to the greater activity of ACTIVE-SAFB in the dye's adsorption, it might be utilized as adsorbent wastewater treatment for dye removal.
4. The obtained data appear that the model pseudo-first-order.
5. The data appear isotherm Freundlich gave the better than langmuir for the both surfaces.

References

Gulkowska A, Leung HW, So MK, Taniyasu S, Yamashita N, Yeung LW, Lam PK. Removal of antibiotics from wastewater by sewage treatment facilities in Hong Kong and Shenzhen, China. *Water research* 2008; 42(1-2): 395-403.

Adeyemo AA, Adeoye IO, Bello OS. Adsorption of dyes using different types of clay: a review. *Applied Water Science* 2017; 7(2): 543-568.

Adriano WS, Veredas V, Santana CC, Gonçalves LB. Adsorption of amoxicillin on chitosan beads: Kinetics, equilibrium and validation of finite bath models. *Biochemical engineering journal* 2005; 27(2): 132-137.

Aljeboree AM. Removal of Vitamin B6 (Pyridoxine) Antibiotics Pharmaceuticals From Aqueous Systems By ZnO. *International Journal of Drug Delivery Technology* 2019; 9(02): 125-129.

Alkaim AF, Aljobree AM. White marble as an alternative surface for removal of toxic dyes (Methylene Blue) from aqueous solutions. *International Journal of Advanced Science and Technology* 2020; 29(5): 5470-5479.

Aljeboree AM, Al-Baitai AY, Abdalhadi SM, Alkaim AF. Investigation Study of Removing Methyl Violet Dye From Aqueous Solutions Using Corn-Cob as A Source of Activated Carbon. *Egyptian Journal of Chemistry* 2021; 64(6): 2873-2878. https://doi.org/10.21608/EJCHEM.2021.55274.3159

Aljeboree AM, Al-Gubury HY, Said MH, Alkaim AF. (2019). The effect of different parameters on the removal of vitamin B12 drug (as a model biochemical pollutants) by AC/clay. *Biochemical and Cellular Archives* 2019; 19(1): 755-759.

ALjeboree AM, ALshirfi AN, ALkaim AF. Removal of Pharmaceutical Amoxicillin drug by using (CNT) decorated Clay/Fe2O3 Micro/Nanocomposite as effective adsorbent: Process optimization for ultrasound-assisted adsorption. *International Journal of Pharmaceutical Research* 2019; 11(4): 80-86.

Alkaim AF, Alrobayi EM, Algubili AM, Aljeboree AM. Synthesis, characterization, and photocatalytic activity of sonochemical/hydration–dehydration prepared ZnO rod-like architecture nano/microstructures assisted by a biotemplate. *Environmental technology* 2017; 38(17): 2119-2129. https://doi.org/10.1080/09593330.2016.1246615

Alkaim AF, Alqaraguly MB. Adsorption of basic yellow dye from aqueous solutions by activated carbon derived from waste apricot stones (ASAC): equilibrium, and thermodynamic aspects. *International journal of chemical sciences* 2013; 11(2): 797-814.

Abd Alrazzak N, Aowda SA, Atiyah AJ. Removal Bismarck Brown G dye from aqueous solution over a composite of triazole-polyvinyl chloride polymer and zinc oxide. *Oriental Journal of Chemistry* 2017; 33(5): 2476-2483.

De Baere S, De Backer P. Quantitative determination of amoxicillin in animal feed using liquid chromatography with tandem mass spectrometric detection. *Analytica chimica acta* 2007; 586(1-2): 319-325.

Alrobayi EM, Algubili AM, Aljeboree AM, Alkaim AF, Hussein FH. Investigation of photocatalytic removal and photonic efficiency of maxilon blue dye GRL in the presence of TiO2 nanoparticles. *Particulate Science and Technology* 2017; 35(1): 14-20.

Jamel HO, Adnan S, Mugheer TH, Jasim LS. Synthesis and Characterization of the Ligand (S, S'-Bis (Benzo [d] Oxazol-2-yl) 2, 2'-((Methylenebis (4, 1 Phenylene)) Bis (Azanediyl)) Diethanethioate (BMPAE) and it's Complexes with Some Transition Metals Ions. *Journal of Global Technology* 2017; 8(9): 76-85.

Ho YS, McKay G. Sorption of dye from aqueous solution by peat. *Chemical Engineering Journal* 1998; 70(2): 115-124.

Sahib IJ, Jasim LS, Alkaim AF. Synthesis and characterizations of reduced graphene oxide/iron oxide: as a model of water treatment. *International Journal of Applied Engineering Research* 2017; 12(24): 14874-14877.

Shah K, Hassan E, Ahmed F, Anis I, Rabnawaz M, Shah MR. Novel fluorene-based supramolecular sensor for selective detection of amoxicillin in water and blood. *Ecotoxicology and environmental safety* 2017; 141: 25-29.

Langmuir I. The adsorption of gases on plane surfaces of glass, mica and platinum. *Journal of the American Chemical society* 1918; 40(9): 1361-1403.

Layth S, Jasim AMA. Removal of Heavy Metals by Using Chitosan/Poly (Acryl Amide-Acrylic Acid) Hydrogels: Characterization and Kinetic Study. *NeuroQuantology* 2021; 19(2): 31-37.

Liu Y, Wang F, Chen X, Zhang J, Gao B. Cellular responses and biodegradation of amoxicillin in Microcystis aeruginosa at different nitrogen levels. *Ecotoxicology and environmental safety* 2015; 111: 138-145.

Chiu MH, Chang JL, Zen JM. An Analyte Derivatization Approach for Improved Electrochemical Detection of Amoxicillin. *Electroanalysis: An International Journal Devoted to Fundamental and Practical Aspects of Electroanalysis* 2009; 21(14): 1562-1567.

Radhy ND, Jasim LS. Synthesis of graphene oxide/hydrogel composites and their ability for efficient adsorption of crystal violet. *Journal of Pharmaceutical Sciences and Research* 2019; 11(2): 456-463.

Pasamontes A, Callao MP. Determination of amoxicillin in pharmaceuticals using sequential injection analysis (SIA): Evaluation of the presence of interferents using multivariate curve resolution. *Analytica chimica acta* 2003; 485(2): 195-204.

Pasamontes A, Callao MP. Determination of amoxicillin in pharmaceuticals using sequential injection analysis and multivariate curve resolution. *Analytica chimica acta* 2004; 515(1): 159-165.

Sultan M. Spectrophotometric determination of acyclovir in some pharmaceutical formulations. *Il Farmaco* 2002; 57(11): 865-870.

Abdulsahib WK, Ganduh SH, Radia ND, Jasim LS. New Approach for Sulfadiazine Toxicity Management using Carboxymethyl Cellulose Grafted Acrylamide Hydrogel. *International Journal of Drug Delivery Technology* 2020; 10(2): 259-264.

Peng Y, Liu M, Chen X, Yuan H, Zhao J. Surface-enhanced Raman spectroscopy coupled with gold nanoparticles for rapid detection of amoxicillin residues in duck meat. *Spectroscopy Letters* 2017; 50(10): 579-584.

Mestre YF, Zamora LL, Calatayud JM. Determination of phenylephrine hydrochloride by flow injection analysis with chemiluminescence detection. *Journal of AoAc international* 2001; 84(1): 13-18.

Zheng J, Zhou Y. Brain-computer interface data classification based on support vector machine. *NeuroQuantology* 2018; 16(6): 638-642.