University of Aden Journal of Natural and Applied Sciences., vol.29(1) (2025) 29 – 41

University of Aden Journal of Natural and Applied Sciences

Journal homepage: https://uajnas.adenuniv.com

Research Article

Removal of Crystal violet and Methyl orange dyes from aqueous solutions using Green-Synthesized Activated Carbon prepared from plastic (PET) wastes

Omeima A. Abdul Ghani[1,*], Shaif M.K.Saleh[2], Zeid A.Alothman[3], Mohamed A.Habila[4]

1 Department of Chemistry, Faculty of Education, Aden University, Aden, Yemen.

2 Department of Chemistry, Faculty of Science, Aden University, Aden, Yemen

3,4 Department of Chemistry, Faculty of Science, King Saud University, Riyadh, Saudi Arabia

https://doi.org/10.47372/uajnas.2025.n1.a04

ARTICLE INFO

Received: 01/09/2025

Accepted: 20/10/2025

Keywords:

Removal of (CV) and (MO) dyes,
Green Synthesis,
Activated carbon
(PET) wastes

Abstract

One easy way to remove synthetic colors from aqueous solutions is to adsorb them on inexpensive solid sorbents. Utilizing activated carbon from polymer waste has been the focus of recent sorption process initiatives, therefore it offers a more appealing and affordable option than commercial activated carbon, which is often more costly. This study seeks to investigate the equilibrium and sorption kinetics of two artificial cationic and Ionic dyes from aqueous media which are respectively crystal violet and methylene orange utilizing Polyethylene Terephthalate PET waste to produce activated carbon. Based on FTIR analysis, the adsorbent possesses an OH functional group in $C_6H_8O_7$ modified carbon-2 and KOH modified carbon-2 of a ratio preparation of 1:2, C-H , C=O and C-O. SEM analysis indicated the presence of pores of different sizes and shapes in micrometer diameter. Additionally, XRD images detected $C_6H_8O_7$ modified carbon-2/C-500 had small sharp peaks revealed in $2\theta = 29°$, $2\theta = 41°$and $2\theta = 51°$. The results suggest that these peaks reflect the condensed aromatic carbonized planes. Batch adsorption studies were performed to investigate the a effect of initial PH, contact time, initial CV and MO concentration, adsorbent dosage and temperature on adsorption. The Langmuir and Freundlich isotherm models provided the explanation for experimental data with the highest correlation coefficient with CV dye, whereas the Freundlich isotherm model is suitable to describe the adsorption reaction of the MO dye. Sorption thermodynamic data show that the adsorption process is spontaneous in all samples except ($C_6H_8O_7$modified carbon-1) was nonspontaneous. Moreover endothermic in all the samples except ($C_6H_8O_7$modified carbon-2) was exothermic sample and the clearing PET waste obtained after surface modification by $C_6H_8O_7$ treatment showed a higher adsorption capacity from KOH.

1. Introduction

Artificial coloring is extremely harmful to the environment [1]. Furthermore , some dyes have toxic, carcinogenic and mutagenic properties [1]. It has already been shown that colors alter the way that sunlight takes and harms the aquatic habitat[2] (stops light from passing through water)[3][4]. This significantly reduces the dissolved oxygen [2] and decreases in the rate of photosynthesis which has an impact on the dissolved oxygen levels throughout the aquatic biota[3] and carcinogens which persist as environmental pollutants. These pollutants and move up the food chain through biomagnification which causes higher trophic-level organisms more contaminated than their prey[3]. However, Crystal violet (CV) is frequently utilized in biological staining, textile dyeing painting[4][5]. CV is as well as utilized in veterinary medicine, as external disinfectant, pH indicator and as a dermatological agent[5]. It can enter cells, concentrate in the cytoplasm and interact with negatively charged cell membrane surfaces with ease despite being a carcinogenic dye.[5]. Additionally Methyl orange (MO) is extensively utilized in research labs as well as in printing paper, pharmaceutical and food processing industries.[6]. Death

---

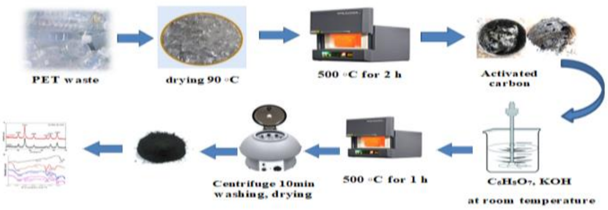
*Department of Chemistry, Faculty of Education, Aden University, Aden, Yemen
E-mail address: kqasem1981@gmail.com

may ensue from high MO exposure levels. Intestinal microbes also metabolize it into aromatic amines[6]. Since methyl orange is soluble in water stable and has low biodegradability, it is challenging to remove from aqueous solutions using standard techniques for water filtration or treatment. [7][6]. Thus, removing colors from wastewater is a difficult operation[8]. Additionally, the synthetic origins and mostly aromatic structure of the dye non-degradable make it harder to handle biologically. [9]. They are very soluble in water which makes it challenging to eliminate them from conventional methods [3]. The fight against environmental pollution has grown in importance [10]. Various approaches are frequently employed for this objective among these therapeutic alternatives are biological and physio-chemical[11] [8], membrane filtration, ozonation, advanced oxidation and integrated treatment processes. However, major drawbacks of these technologies are long operation time, high sludge production, high cost not being eco-friendly, etc[2]. Adsorption technology is one of the most efficient ways to remove dye among a variety of chemical, physical and biological treatment techniques as the result of its low cost, high efficiency, simplicity and insensitivity to harmful compounds[8][12][2] [13][14], in recent years. Potassium hydroxide KOH is one of the most often used activators as under the same circumstances, it may create activated carbon with a large surface area and a fine pore size distribution [15], reduced cost, decreased corrosiveness and minimal environmental contamination. The efficiency of KOH-activated carbon in the adsorption of many organic compounds including (phenol, dyes, heavy metals and pesticides) has been the subject of numerous investigations. [16]. Citric acid (CA) is an organic acid which is widely used in the food and daily chemical industries, it is non-toxic with good solubility, anti-oxidation and mild reaction. Moreover [17]. It has been reported in the literature that citric acid (CA) is an effective modifier for AC to improve the removal of contaminants such as copper, aniline, lead, uranium and methylene blue. [18].

2. Materials and Methods

2.1 Preparation of Activated Carbon

Polyethylene Terephthalate (PET) waste bottles were harvested from the local restaurant in Riyadh (Saudi Arabia) disposal areas. All the collected bottles are colorless and labels were removed , caps were detachment and dirt were remove by water, and finally were dried in the oven at 90 ◦C, for 24 h. The dried bottles were then cut into small pieces of 1 cm by 1 cm dimensions and shredded into flakes for further processing. Activated carbon (AC) was prepared by carbonizing 10 g of cleaned PET in a furnace at 500 ◦C for 2 h. The modification of AC from PET was undertaken by acid and base of Chemical activation for preparation adsorbent. activation mainly was carried out using given amounts of AC and KOH, citric acid in aqueous solution at the AC/KOH, weight ratios (g) 1:1 and 1:2[19], AC/C6H8O7 weight ratios 1:1 and 1:2[20]. Two beakers with 100 ml distilled water for every chemical with continuous stirring by a mechanical stirrer for 24 h at room temperature after drying in an oven at 105◦C to be activated, the 4 samples were put in a furnace at 500 ◦C for 1 h, the samples were grinding to get powder and were sifted by a sieve of 0.011 µm(size of particles). Subsequently, they were wash for half an hour with 10 ml ethanol by sonication to remove inorganic impurities and dissolve inorganic compounds like salts then they were twice washed with deionized water by centrifuge at 10.000 rmp for 10 min, and then were dried in an oven at 105 ◦C for 24 h.



Scheme 1. Graphical Abstract

2.2 Batch Adsorption Studies

These examinations were conducted at Saud University's College of Science in the Kingdom of Saudi Arabia. The prepared PET AC (0.01 g in each tube) was transferred to 10 ml polyethylene tube and 8 ml of dye solution (50 ppm) and 2 ml Acid or base buffer were added at pH levels (2,4,6,8,10). The mixture was placed on a shaker at 150 rpm for 24 h at 25°C. The solution was filtered and the C.V and M .O dyes concentrations were determined by spectrometry at the wavelength of maximum absorbance, 590 nm CV , 465 nm MO using a double beam UV–Vis spectrophotometer (Thermo Scientific). Through the calibration curve of the device, the following statistical parameters were obtained: which are correlation coefficient (R^2) 0.9996, intercept -0.01613, LOD 0.309 and LOQ 0.939 factors affecting accuracy. The same procedures were followed to evaluate the effect of pH, time, concentration, dose and temperature on the adsorption capacity of the AC samples. The adsorption capacity of the AC samples was evaluated using the following Eq. (1):

$$\mathrm{Qe} = (C_0 - \mathrm{Ce}) \times \frac{V}{m} \qquad (1)$$

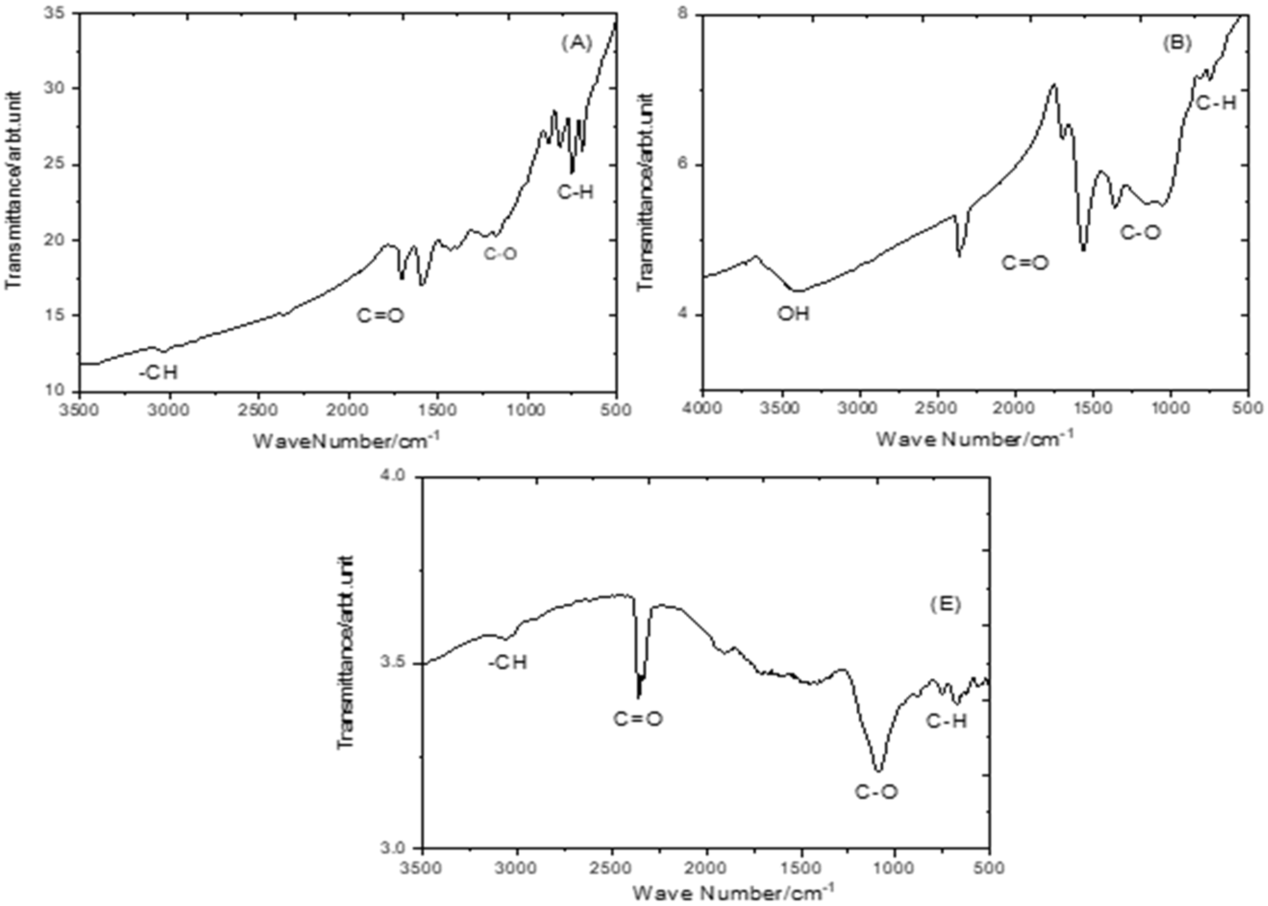


Figure 1: FTIR spectra of Active Carbon(A) $C_6H_8O_7$ modified carbon-1,(B) $C_6H_8O_7$ modified carbon-2 and(E) Unmodified carbon-500.

where Qe is the adsorption capacity (mg /g), C0 is the initial concentration of the dyes, Ce is the equilibrium concentration of the dyes, V is the volume of the solution (L) and M is the mass of the adsorbent (g) [21]. Experiments were conducted and measurements were taken in three replicates. The results are presented as average with standard deviation "Mean ± SD".

3. Results and Discussion

3.1. Structural and Morphological Characteristics

Fig.1 shows the FTIR spectra of carbons were prepared with C6H8O7 modification1-2, 1g-2g. The change in the surface functional groups after chemical modification as shown in Fig1 (A)(B) was compared with the FT-IR spectrum of (E). Fig1 is shows the vibration shifts in E, A and B samples from (1092 to 1226-1320) cm-1, respectively at C-O with medium intensity. Moreover, the C=O group shifts from (2354 to 1730- 1703) cm-1 respectively in samples E, A and B. as result of stretching vibration due to Carboxylic acid modification. While C-H shifts from (3071 to 3036-3411) cm-1 respectively in samples E, A and B as shown in Fig.1 and Effluent appears at a wavenumber of 3411cm1 which indicates the vibration stretch of hydroxyl OH with a wide band [1] [20] [22] [23].

Fig.2 shows the FTIR spectra of carbons which was prepared with KOH modification1-2, 1g-2g. The change of in the surface of functional groups after chemical modification as shown in Fig2 (C)(D) was compared with the FT-IR spectrum of (E). Fig2 is shown, vibration shifts in E, C, and D samples from (1092 to 879-1109) cm-1 respectively in C-O.also the C=O group shifts from (2354 to 1585- 1564) cm-1 respectively in samples E, C and D. Fig2 illustrates the attributed shifting chiefly due to impregnation with KOH. While C-H shifts from (3071 to 3044 -3418) cm^{-1}, respectively in samples E, C, and D as shown in Fig.2. 3418 cm-1band is attached especially for the stretch mode of hydroxyl OH [23]. Shifts and appearance bands indicate that some functional groups of Carboxylic and hydroxyl have been successfully introduced into the surface of the PET Activated carbon after chemical modification. These functional groups are very effective in capturing dyes from aqueous solution [20].

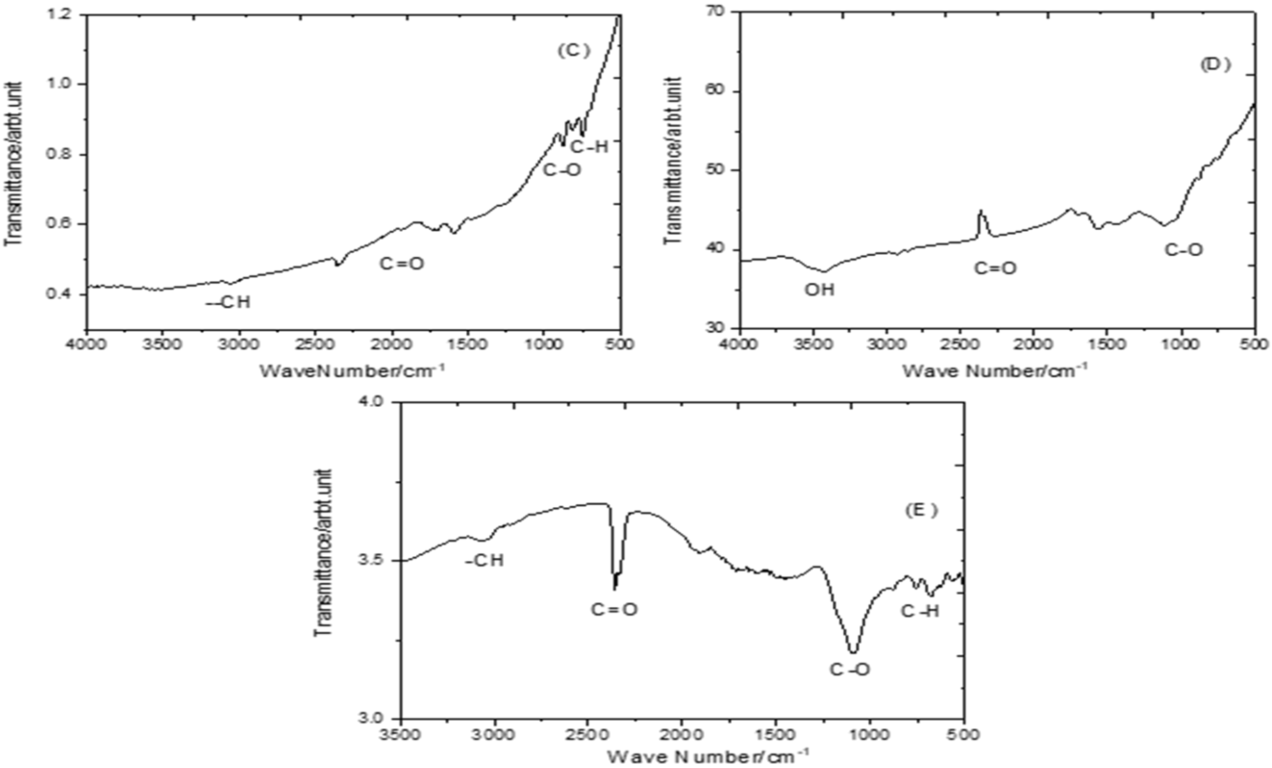


Figure 2: (C) FTIR spectra of Active Carbon KOH modified carbon-1, (D) KOH modified carbon-2, and (E) Unmodified carbon.

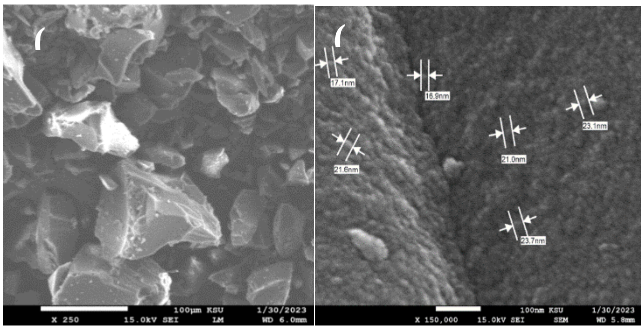


Fig.3: Morphology of the porous Unmodified carbon/C-500 of 250 (a) and 150,000 (b)

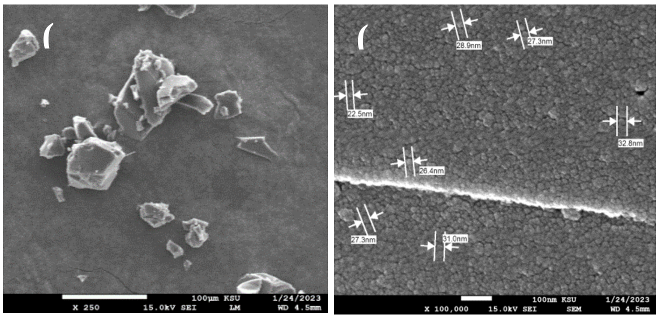


Fig.4: Morphology of the porous C6H8O7 modified carbon-1/C-600 of 250 (a) and 100,000

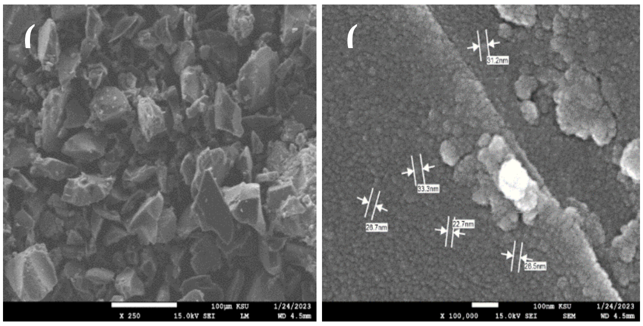


Fig.5: Morphology of the porous $C_6H_8O_7$ modified carbon-2/C-600 of 250 (a) and 100,000

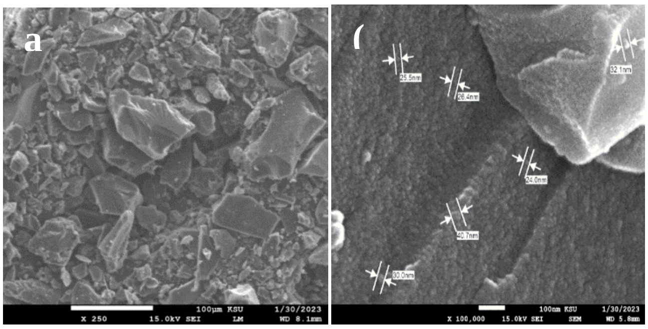


Fig.6: Morphology of the porous KOH modified carbon-1/C-600 of 250 (a) and 100,000

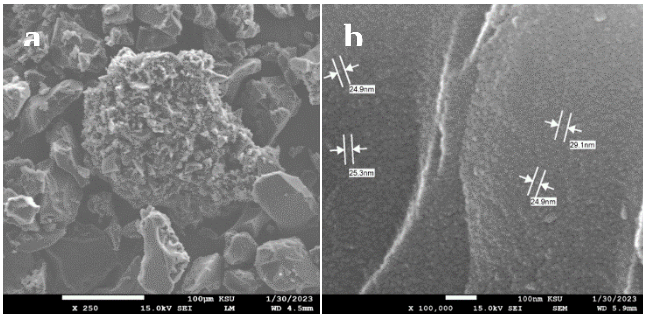


Fig.7: Morphology of the porous KOH modified carbon-2/C-500 of 250 (a) and 100,000 (b)

The prepared activated carbon samples were examined by SEM to analyze the surface of the adsorbents before and after activation. The SEM photographs of the sample are as shown in Fig 3 explains that (a) is Unmodified carbon PET wastes at 500. It can be seen that the micrographs have heterogeneous surfaces with large cracks. These big cracks don't have a specific surface area for adsorption, while in Fig 4,5,6,7 (a) is prepared chemically activated carbons by KOH and citric acid showed a tiff surface full of a variety of random holes forming complicated channel networks due to the modification, which increased the number of cavities on the carbon surface. It demonstrates at higher magnification. in Fig 3 shows (b), Unmodified carbons with large cracks and holes. While Fig. 4,5,6,7 (b) shows the porous of different sizes and shapes in micrometer diameter (µm). These micropores on the external surface attract the dyes to the adsorption sites from solutions.

XRD spectrum in Fig.8 illustrates (a) Appearance a broad diffraction peaks from 2θ between 10° and 35° which is ascribed to the diffraction of amorphous carbon. According to previous studies [24], (b) showed diffraction peaks at 2θ = 20° and 2θ = 10° with the absence of a sharp peak, revealing a predominantly amorphous structure with low crystallinity. According to previous studies [25]. indeed (c) had small sharp peaks revealed in 2θ = 29°, 2θ = 41°and 2θ = 51°. Results suggest that these peaks reflect the condensed aromatic carbonized planes. It is indicate the sharper the peak, the higher the degree of condensation of the aromatic ring according to previous studies[24]. The amorphous structure in(d,e) was identified by the peak at 2θ = 23◦ and peak at 2θ = 43◦ according to previous studies[26][27][28].

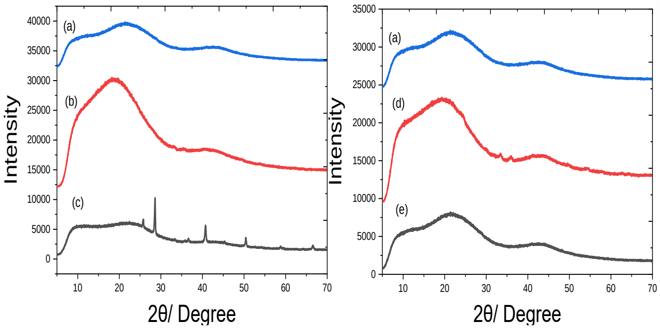
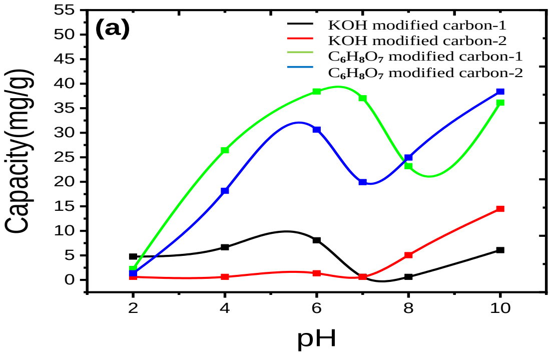


Fig.8: XRD patterns of the carbon from PET(a) unmodified carbon-500 (b) $C_6H_8O_7$ modified carbon-1/C-500 (c) $C_6H_8O_7$ modified carbon-2/C-500 (d) KOH modified carbon-1/C-500 (e) KOH modified carbon-2/C-500

3.2. Adsorption Studies:

3.2.1. Effect of pH on (CV) and (MO) Removal:

As shown in Figure 9, pH of 6 was the best medium for adsorption of dyes CV and MO ,by the samples of C6H7O8 modified carbon-1, C6H7O8 modified carbon-2, KOH modified carbon-1and KOH modified carbon-2. This may denoted to the fact that affects the solubility of the metal ions in water, the concentration of the counter ions on the functional groups of the adsorbent and the degree of ionization of the adsorbate during reaction [29].



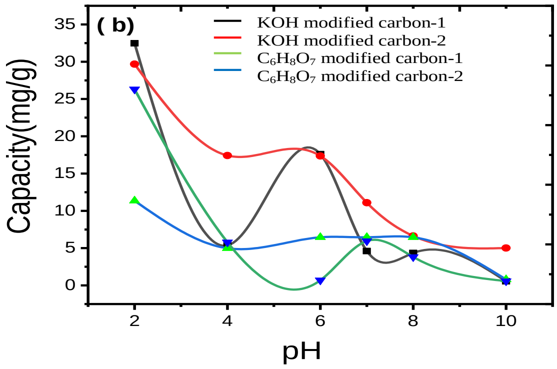


Fig.9: The investigation of the effect of pH on the adsorption of (a)CV and (b) MO at 25 ◦C with a contact time of 24 h. using adsorbent dose of 0.05 g and metal concentration of 50 mg/L, (n=3)

3.2.3. Effect of Contact Time:

studying the time effect was investigated by changing the time of contact from 5min to 4500 min Fig.10 on the adsorptions of CV was investigated by increasing the time of the adsorption process at 1500 min and the capacity until reaching (38.4, 9.8) mg/g for (C6H7O8 modified carbon-1, KOH modified carbon-1) respectively. while in MO at 1600 min and the capacity stopped increasing until reaching (6.458, 5.9) mg/g for (C6H7O8 modified carbon-2, and KOH modified carbon-2) respectively. Moreover, there were no further increases in the adsorption on the adsorbent surfaces. This indicate the rat of adsorption equal to the rat of desorption which were previous to the adsorption reached to the equilibrium state.

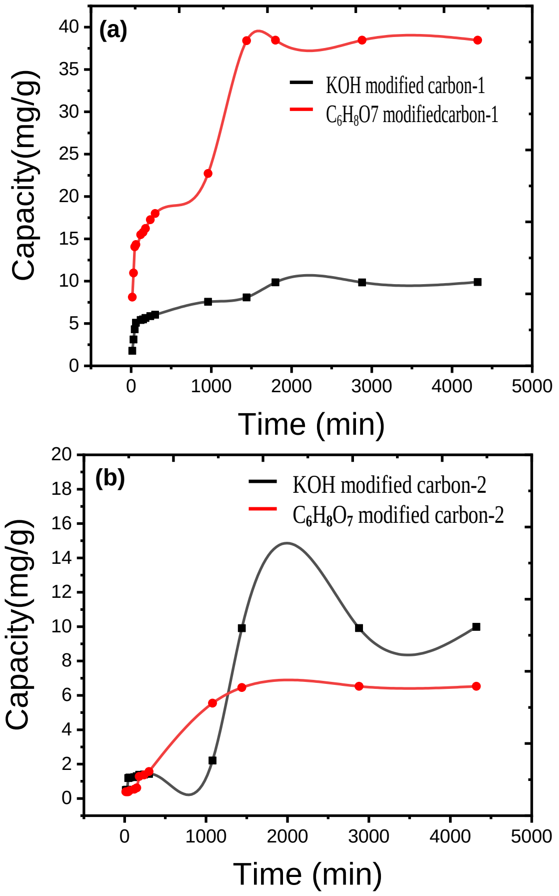


Fig.10: Contact time investigation for adsorption of CV on to(C6H7O8 modified carbon-1, KOH modified carbon-1) and MO on to(C6H7O8 modified carbon-2, and KOH modified carbon-2) adsorbent dose 0.05 g, and metal concentration of 50 mg/L ,pH 6, (n=3).

Effect of Adsorbent Dosage:

It is essential to estimate the optimum amount of the adsorbent dosage[30]. Long removal efficiency is adversely affected by increasing adsorbent doses as excess adsorbent dosage reduces removal efficiency due to particle agglomeration and decrease active sites on the surface area of the adsorbent [31]. As shown in Fig 11 the values of the capacity varied depending on the adsorbent dosages from 0.2 to 1 g L-1. The capacity decreased during adsorption of CV, from 38.4 to 0.3 and from 8 to 0.6 in (C6H7O8 modified carbon-1, KOH modified carbon-1), respectively. While, during adsorption of MO from 5.5 to 3.1 and from 17.6 to 1.8 in (C6H7O8 modified carbon-2, KOH modified carbon-2) respectively decreased for an initial metal concentration of 50 mg/L.

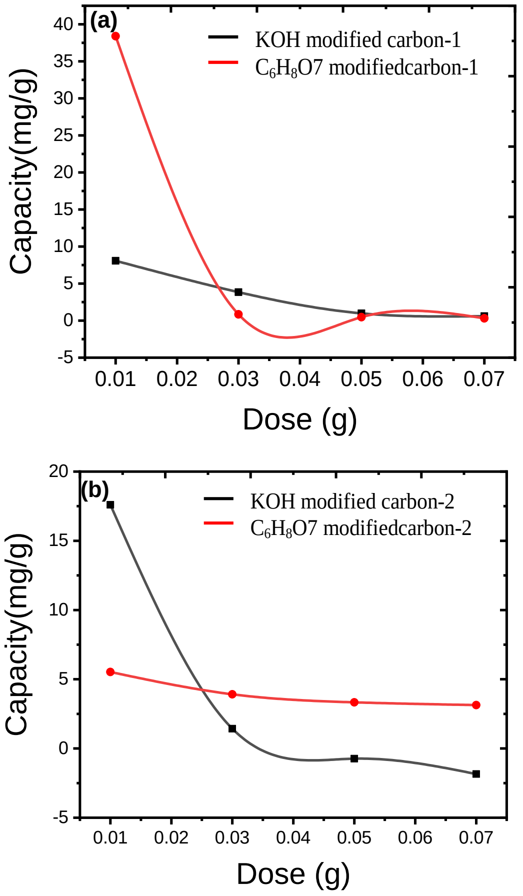


Figure 11: Effect of adsorbent dose on (a)CV and (b) MO adsorption; metal concentration of 50 mg/L ,pH 6, (n=3).

Kinetic Studies

By applying the pseudo-first and second–order model to investigate the rate of the adsorption process, it was found that the values of R21 in CV dye were (0.98, 0.99, 0.8824, 0.9638) whereas, the values of R21 of adsorptions of MO dye were (0.7529, 0.7294, 0.8875, 0.9518) for these samples C6H8O7modified carbon-1, KOH modified carbon-1, C6H8O7modified carbon-2 and KOH modified carbon-2 respectively. The results in Table 1 indicate that the pseudo-second-order model is a better fit for adsorption kinetics than the pseudo-first-order model due to good agreement between experimental and calculated qe values. It can be concluded that the model of the second order can be applied to explain

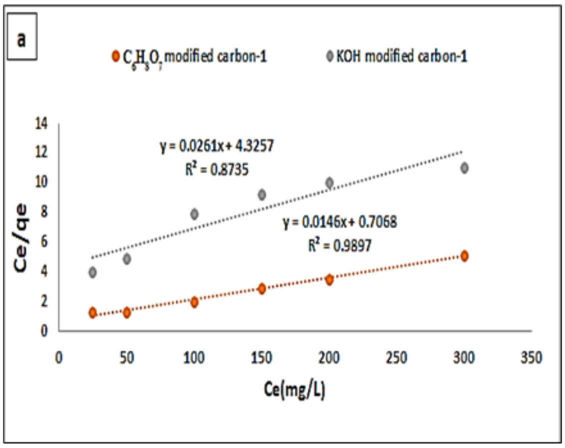
Table 1. Kinetic constants for (a)CV on to (C6H8O7modified carbon-1, KOH modified carbon-1) and (b) MO on to(C6H8O7modified carbon-2, KOH modified carbon-2) modified active carbon from PET waste

Dyes	Samples	Pseudo-first-order			Pseudo-second-order			
		K_1 (g/mg.min)	q_e,cal (mg/g)	$R^2{}_1$	q_e,exp (mg/g)	K_2 (min^{-1})	q_e,cal (mg/g)	$R^2{}_2$
CV	**$C_6H_8O_7$modified carbon-1**	**7.26*10^{-4}**	**2.9*10^{-4}**	**0.9807**	**38.46**	**1.38*10^{-3}**	**27.7**	**0.7529**
	KOH modified carbon-1	**1.14*10^{-4}**	**217220**	**0.9943**	**9.867**	**2.76*10^{-3}**	**6.77**	**0.7294**
MO	**$C_6H_8O_7$modified carbon-2**	**1.1*10^{-4}**	**74**	**0.8824**	**6.458**	**-9.2*10^{-4}**	**6.289**	**0.8875**
	KOH modified carbon-2	**2.5*10^{-4}**	**46**	**0.9638**	**9.909**	**-1.6*10^{-3}**	**5.788**	**0.9518**

the adsorption kinetics. Discussion of these results is consistent with what was stated in the references [32][33]. These observations indicated that the rate of the dye adsorption process was controlled by chemi-sorption process which depended on the chemical character of the dye[34].

Langmuir

The following figures are data for CV and MO dyes adsorption on to the adsorbent which were used to support the assumption. In Fig. 12, the graph shows that vales of correlation coefficient were high for R2 (0.9897, 0.8735) of CV dye on to (C6H8O7modified carbon-1, KOH modified carbon-1) respectively while R2 for adsorption MO dye were low values for R2 (0.5981, 0.4148) in (C6H8O7modified carbon-2, KOH modified carbon-2) respectively. Consequently, the Langmuir assumption is suitable for the adsorption of CV dye more than MO dye and is identical to the monolayer and homogeneous adsorption process[35] [33].



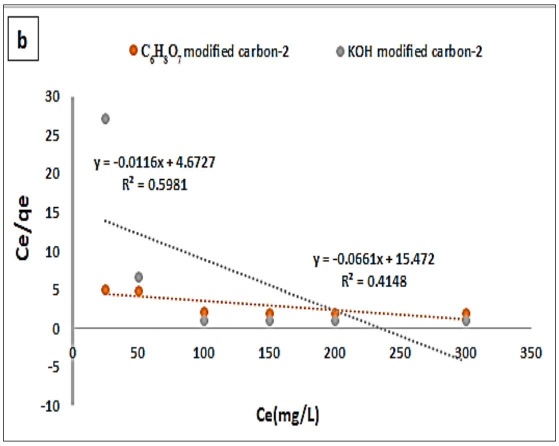


Figure 12: Langmuir adsorption isotherms (a)CV on to (C6H8O7modified carbon-1, KOH modified carbon-1) and (b) MO on to(C6H8O7modified carbon-2, KOH modified carbon-2) (modified active carbon from PET waste)

Freundlich

In Fig. 13, the constants calculated for the Freundlich are shown. The process of CV and MO adsorption was well correlated with the Freundlich equation R2(0.9943, 0.9807) for CV dye on to (C6H8O7modified carbon-1, KOH modified carbon-1) respectively, while the values of R2(0.9638, 0.8824) for M.O dye on to (C6H8O7modified carbon-2, KOH modified carbon-2) respectively, These high correlation coefficient R2 previous to Freundlich isotherm model fitted data and better than the Langmuir isotherm model for both dyes sorption of CV and MO. They exhibited good behavior suggesting and heterogeneous adsorption process on the surfaces of prepared adsorbate or as a multilayer sorption with nonuniform distribution of adsorption heat and affinities over a heterogeneous surface[35] [33].

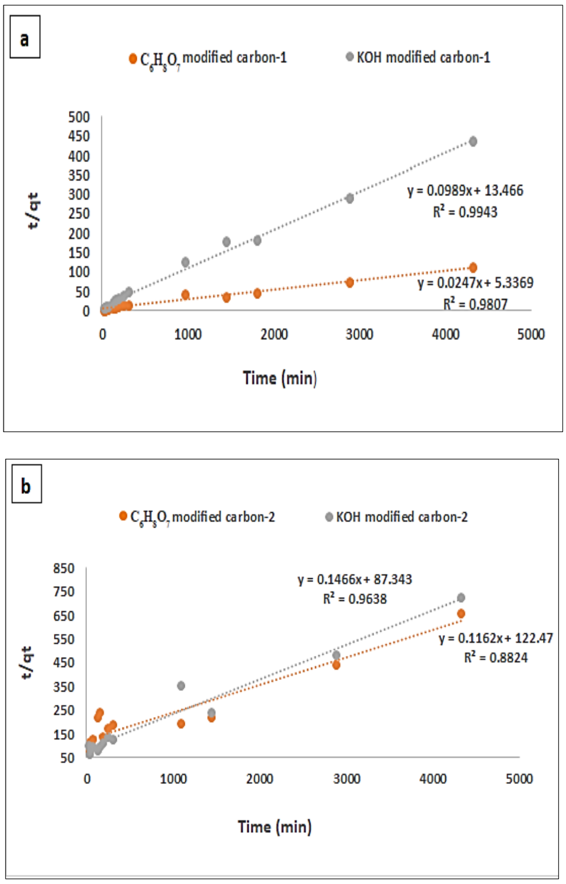


Figure 13: Freundlich Isotherm of (a) CV on to ($C_6H_8O_7$modified carbon-1, KOH modified carbon-1) and (b) MO on to(C6H8O7modified carbon-2, KOH modified carbon-2) (modified active carbon from PET waste.

Thermodynamic Studies

The values of ΔH°, ΔS°, and ΔG° for the CV and MO adsorption are presented in Table 2. The effect of temperature was investigated at 298, 308, 318 and 328 K. As can be noticed that the ΔG° tends to decrease or increase. If ΔGº is a negative value at a given temperature it indicates that the particular reaction is spontaneous. On the other hand if ΔGº is positive value it indicates that adsorption process is a non-spontaneous reaction. The adsorption process indicates that the particular reaction is spontaneous when the bonds formed between the metal ions and the binding sites rupture at high temperature. This decreases the percentage removal and the spontaneity of an adsorption process can be explained by the free energy change (ΔGº). In addition, the interactions with free energy indicated that the processes were favorable for the formation of electrostatic interactions between metal ions and the active site of the adsorbent, where the results in Table 2 showed that the reaction in the samples (KOH modified carbon-1, C6H8O7modified carbon-2, KOH modified carbon-2) was non-spontaneous owing to ΔGº positive. While in the sample (C6H8O7modified carbon-1) was the reaction spontaneous due to ΔH°< TΔS°, the energy of enthalpy which had less effective from effect of adsorption entropy (freedom-degree) [33], [36]. The negative ΔH° value denotes the exothermic process of obtaining adsorption. As shown in Table 2, this happened in sample (C6H8O7modified carbon-2) and

Table 2. The Thermodynamic Parameters of (a)CV on to (C6H8O7modified carbon-1, KOH modified carbon-1) and (b) MO on to(C6H8O7modified carbon-2, KOH modified carbon-2)

Dyes	**Smples**		**Thermodynamic Parameters**		
		Temperature T(K)	**ΔG° (KJ/mole)**	**ΔS°(J/mole/K)**	**ΔH° (KJ/mole)**
CV	**$C_6H_8O_7$modified carbon-1**	**298 308 318 328**	**-7878.1 -8222.4 -12180.5 -12700.4**	**8.8**	**2.1**
	KOH modified carbon-1	**298 308 318 328**	**3402.8 3441.5 3372.6 2816.5**	**-5.5**	**0.39**
MO	**$C_6H_8O_7$modified carbon-2**	**298 308 318 328**	**4081.6 4616.8 5140.5 8029.3**	**-3.9**	**-1.4**
	KOH modified carbon-2	**298 308 318 328**	**655.9 4624.1 12130.3 13210.0**	**-10.5**	**1.7**

Table 3. Comparison between literature and the most signification results obtained in the current work

Dye	**Adsorbent**	**Maximum adsorption capacity (mg/g) based on the references optimization condition**	**References**
CV	Activated carbon	87.20	[41]
	Activated carbon Lemon ACL/Fe_2o_3 magnetic nanocomposite	35.31	[42]
	Clay (PFB-Mzvi adsorbent)	89.9	[43]
	Activated carbon (commercial)	116	[44]
	KOH modified carbon-1	9.86	In this study
	$C_6H_8O_7$ modified carbon-1	38	
MO	Activated carbon from date pits	434	[45]
	Activated carbon from Lemon peels	33	[46]
	biological activated carbons	3.18-3.66	[47]
	activated carbons	8.78-16.90	[48]
	KOH modified carbon-2	5.9	In this study
	$C_6H_8O_7$ modified carbon-2	6.4	

the positive ΔH° value involves an endothermic process ($C_6H_8O_7$modified carbon-1, KOH modified carbon-1, KOH modified carbon-2). while the negative value was shown in ($C_6H_8O_7$ modified carbon-2) exothermic reaction [37]. As reported in Table 2, the positive values of entropy change (ΔS) for the sorption process, which indicates an entropy value above zero and confirms the irreversibility of the adsorption process [38]. On the other hand, the negative value of entropy change ΔS suggests the desorption of the adsorbate molecules from the solid to the liquid during the adsorption process due to the decreased disorder at the solid/ liquid interface [39]. Moreover, the degree of freedom of the adsorbed species is increased as a result of the positive value of ΔS [40].

To demonstrate the efficacy of AC from PET as an effective adsorbent for CV and MO, it is necessary to compare its adsorption capacity to that of other known adsorbents. Table 3compares the q_{max} values for CV.
and MO adsorption on different adsorbents to those of our adsorbents.

4. Conclusions

In this study, the optimum conditions for the maximum removal of CV and MO dyes by the adsorbent were found to be: 0.05 g in 10 ml of the adsorbate solution at pH 6, a contact time of 1500-1600 min respectively at a temperature of 25 ◦C. The percentage removal of CV and MO dyes reached a maximum of around 50 ppm initial metal ion concentration with an adsorption capacity of KOH modified carbon-1, C6H8O7 modified carbon-1, (9.86 - 38) mg/g for cv respectively, and KOH modified carbon-2, C6H8O7 modified carbon-2 (5.9 - 6.4) mg/g for MO respectively. The adsorption equilibrium data fitted well to the Langmuir and Freundlich isotherm models suggesting the presence of monlayer and multilayer adsorption mechanisms for the CV adsorption by the adsorbent while in MO dye multilayer adsorption mechanisms. The thermodynamic studies indicated that the adsorption process is spontaneous in all samples except (C6H8O7modified carbon-1), which was a nonspontaneous, as well as endothermic process was excited in all the samples except (C6H8O7modified carbon-2) which was an exothermic sample. The FTIR analysis indicated the presence of various functional groups in modified PET waste such as the OH functional group in C6H8O7 modified carbon-2 and KOH modified carbon-2, with a ratio preparation of 1:2, C-H, C=O, and C-O functional groups. The SEM analysis suggested that the adsorbent possesses porous structures of different sizes and shapes in micrometer diameter XRD images detected that C6H8O7 modified carbon2/C-500 had small sharp peaks revealed in 2θ = 29°, 2θ = 41°and 2θ = 51°. Results suggest that these peaks reflect the condensed aromatic carbonized planes which indicates that the activated carbon prepared from PET was predominantly amorphous and this can be which could be related to the dyes removing by the adsorbent. The surface modified PET waste showed higher potential for the removal of CV and MO when compared with surface unmodified PET waste. However, further investigations should be carried out to identify the optimum conditions for the removal of CV and MO from aqueous solutions using surface modified PET waste as an adsorbent.

References

1. A. D. Rosanti, "Adsorption of Methylene Blue and Methyl Orange from Aqueous Solution using Orange Peel and CTAB-Modified Orange Peel," vol. 9, no. 1, pp. 237–246, 2022, doi:org/10.18596/jotcsa.1003132.
2. S. Lairini et al., "The adsorption of Crystal violet from aqueous solution by using potato peels (Solanum tuberosum): Equilibrium and kinetic studies," J. Mater. Environ. Sci., vol. 8, no. 9, pp. 3252–3261, 2017. www.jmaterenvironsci.com.
3. B. Lellis, C. Z. Fávaro-Polonio, J. A. Pamphile, and J. C. Polonio, "Effects of textile dyes on health and the environment and bioremediation potential of living organisms," Biotechnol. Res. Innov., vol. 3, no. 2, pp. 275–290, 2019, doi: 10.1016/j.biori.2019.09.001.
4. S. Chakraborty, A. Mukherjee, S. Das, N. R. Maddela, S. Iram, and P. Das, "Study on isotherm, kinetics, and thermodynamics of adsorption of crystal violet dye by calcium oxide modified fly ash," Environ. Eng. Res., vol. 26, no. 1, pp. 1–9, 2021, doi: 10.4491/eer.2019.372.
5. A. Wathukarage, I. Herath, M. C. M. Iqbal, and M. Vithanage, "Mechanistic understanding of crystal violet dye sorption by woody biochar: implications for wastewater treatment," Environ. Geochem. Health, vol. 41, no. 4, pp. 1647–1661, 2019, doi: 10.1007/s10653-017-0013-8.
6. M. Farhan Hanafi and N. Sapawe, "A review on the water problem associate with organic pollutants derived from phenol, methyl orange, and remazol brilliant blue dyes," Mater. Today Proc., vol. 31, no. 2020, pp. A141–A150, 2020, doi: 10.1016/j.matpr.2021.01.258.
7. V. Yönten, N. K. Sanyürek, and M. R. Kivanç, "A thermodynamic and kinetic approach to adsorption of methyl orange from aqueous solution using a low cost activated carbon prepared from Vitis vinifera L.," Surfaces and Interfaces, vol. 20, no. May, pp. 1–8, 2020, doi: 10.1016/j.surfin.2020.100529.
8. R. Paradelo, X. Vecino, A. B. Moldes, and M. T. Barral, "Potential use of composts and vermicomposts as low-cost adsorbents for dye removal: an overlooked application," Environ. Sci. Pollut. Res., vol. 26, no. 21, pp. 21085–21097, 2019, doi: 10.1007/s11356-019-05462-x.
9. Y. Y. Ling, F. Bukhari, and M. Suah, "Journal of Environmental Chemical Engineering Extraction of malachite green from wastewater by using polymer inclusion membrane," Biochem. Pharmacol., vol. 5, no. 1, pp. 785–794, 2017, doi: 10.1016/j.jece.2017.01.001.
10. C. X. H. Su, L. W. Low, T. T. Teng, and Y. S. Wong, "Combination and hybridisation of treatments in dye wastewater treatment: A review," J. Environ. Chem. Eng., vol. 4, no. 3, pp. 3618–3631, 2016, doi: 10.1016/j.jece.2016.07.026.
11. R. Farma et al., "Corn cob based activated carbon preparation using microwave assisted potassium hydroxide activation for sea water purification," J. Phys. Conf. Ser., vol. 1120, no. 1, 2018, doi: 10.1088/1742-6596/1120/1/012017.
12. A. V. Filho, R. X. Kulman, L. V. Tholozan, A. R. F. de Almeida, and G. S. da Rosa, "Preparation and characterization of activated carbon obtained from water treatment plant sludge for removal of cationic dye from wastewater," Processes, vol. 8, no. 12, pp. 1–13, 2020, doi: 10.3390/pr8121549.
13. J. Ma et al., "Enhanced adsorptive removal of methyl orange and methylene blue from aqueous solution by alkali-activated multiwalled carbon nanotubes," ACS Appl. Mater. Interfaces, vol. 4, no. 11, pp. 5749–5760, 2012, doi: 10.1021/am301053m.
14. A. Demirbas, "Agricultural based activated carbons for the removal of dyes from aqueous solutions: A review," J. Hazard. Mater., vol. 167, no. 1–3, pp. 1–9, 2009, doi: 10.1016/j.jhazmat.2008.12.114.
15. A. A. Ahmad and A. Idris, "Preparation and characterization of activated carbons derived from bio-solid: A review," Desalin. Water Treat., vol. 52, no. 25–27, pp. 4848–4862, 2014, doi: 10.1080/19443994.2013.808797.
16. Z. Heidarinejad, M. H. Dehghani, M. Heidari, G. Javedan, I. Ali, and M. Sillanpää, "Methods for preparation and activation of activated carbon: a review," Environ. Chem. Lett., vol. 18, no. 2, pp. 393–415, 2020, doi: 10.1007/s10311-019-00955-0.
17. M. Zhang et al., "A Prospective Review on the Research Progress of Citric Acid Modified Starch," Foods, vol. 12, no. 3, pp. 1–13, 2023, doi: 10.3390/foods12030458.
18. U. S. Rashid and A. N. Bezbaruah, "Citric acid modified granular activated carbon for enhanced defluoridation," Chemosphere, vol. 252, 2020, doi: 10.1016/j.chemosphere.2020.126639.
19. Z. Guo, J. Zhang, and H. Liu, "Ultra-high Rhodamine B adsorption capacities from an aqueous solution by activated carbon derived from: Phragmites australis doped with organic acid by phosphoric acid activation," RSC Adv., vol. 6, no. 47, pp. 40818–40827, 2016, doi: 10.1039/c5ra25200h.
20. L. Chen, A. Ramadan, L. Lü, W. Shao, F. Luo, and J. Chen, "Biosorption of methylene blue from aqueous solution using lawny grass modified with citric acid," J. Chem. Eng. Data, vol. 56, no. 8, pp. 3392–3399, 2011, doi: 10.1021/je200366n.
21. M. A. Habila, Z. A. Alothman, R. Ali, A. A. Ghafar, and M. S. E. D. Hassouna, "Removal of Tartrazine Dye onto Mixed-Waste Activated Carbon: Kinetic and Thermodynamic Studies," Clean - Soil, Air, Water, vol. 42, no. 12, pp. 1824–1831, 2014, doi: 10.1002/clen.201300191.
22. C. Bhattacharjee, S. Dutta, and V. K. Saxena, "A review on biosorptive removal of dyes and heavy metals from wastewater using watermelon rind as

biosorbent," Environ. Adv., vol. 2, no. July, p. 100007, 2020, doi: 10.1016/j.envadv.2020.100007.

23. K. D. and A. Y. Abdoul Ntieche Rahman, Abdoul Wahabou, Patrick Mountapmbeme Kouotou, Abdelaziz Bacaoui, "Removal of Micro Pollutants From Aqueous Solution Using Activated Carbons From Pet Waste," Can. J. Pure Appl. Sci., vol. 11, no. 1, pp. 4131–4140, 2017. www.cjpas.net.

24. R. Nandi, M. K. Jha, S. K. Guchhait, and D. Sutradhar, "Impact of KOH Activation on Rice Husk Derived Porous Activated Carbon for Carbon Capture at Flue Gas alike Temperatures with High CO 2 /N 2 Selectivity," 2023, doi: 10.1021/acsomega.2c06955.

25. M. S. Shamsuddin, N. R. N. Yusoff, and M. A. Sulaiman, "Synthesis and characterization of activated carbon produced from kenaf core fiber using H 3 PO 4 activation," Procedia Chem., vol. 19, pp. 558–565, 2016, doi: 10.1016/j.proche.2016.03.053.

26. A. Kami, P. Mi, A. Wr, J. Sre, R. Jan, and J. Serafin, "Activated Carbon Modification towards Efficient Catalyst for High Value-Added Products Synthesis from Alpha-Pinene," 2021, doi.org/10.3390/ma14247811.

27. S. Ayyalusamy, S. Mishra, and V. Suryanarayanan, "Promising post-consumer PET-derived activated carbon electrode material for non-enzymatic electrochemical determination of carbofuran hydrolysate," Sci. Rep., no. August, pp. 1–9, 2018, doi: 10.1038/s41598-018-31627-8.

28. M. Pakuła, S. Biniak, M. Bystrzejewski, and R. Szmigielski, "Journal of Analytical and Applied Pyrolysis Characteristics of activated carbon prepared from waste PET by carbon dioxide activation," vol. 100, pp. 192–198, 2013, doi: 10.1016/j.jaap.2012.12.021.

29. M. Zhang et al., "Novel cationic polymer modified magnetic chitosan beads for efficient adsorption of heavy metals and dyes over a wide pH range," Int. J. Biol. Macromol., vol. 156, pp. 289–301, 2020, doi: 10.1016/j.ijbiomac.2020.04.020.

30. S. Madadgar, F. Doulati Ardejani, Z. Boroumand, H. Sadeghpour, R. Taherdangkoo, and C. Butscher, "Biosorption of Aqueous Pb(II), Co(II), Cd(II) and Ni(II) Ions from Sungun Copper Mine Wastewater by Chrysopogon zizanioides Root Powder," Minerals, vol. 13, no. 1, 2023, doi: 10.3390/min13010106.

31. H. Gupta and P. R. Gogate, "Intensified removal of copper from waste water using activated watermelon based biosorbent in the presence of ultrasound," Ultrason. Sonochem., vol. 30, pp. 113–122, 2016, doi: 10.1016/j.ultsonch.2015.11.016.

32. M. A. Habila et al., "Phosphonium-based ionic liquid modified activated carbon from mixed recyclable waste for mercury(II) uptake," Molecules, vol. 24, no. 3, pp. 1–11, 2019, doi: 10.3390/molecules24030570.

33. Z. A. Alothman, M. A. Habila, M. Sheikh, and N. S. Al-kadhi, "Fabrication of renewable palm-pruning leaves based nano-composite for remediation of heavy metals pollution," Arab. J. Chem., vol. 13, no. 4, pp. 4936–4944, 2020, doi: 10.1016/j.arabjc.2020.01.015.

34. D. Sun, Z. Zhang, M. Wang, and Y. Wu, "Adsorption of Reactive Dyes on Activated Carbon Developed from <i>Enteromorpha prolifera</i>," Am. J. Anal. Chem., vol. 04, no. 07, pp. 17–26, 2013, doi: 10.4236/ajac.2013.47a003.

35. E. E. Jasper, V. O. Ajibola, and J. C. Onwuka, "Nonlinear regression analysis of the sorption of crystal violet and methylene blue from aqueous solutions onto an agro-waste derived activated carbon," Appl. Water Sci., vol. 10, no. 6, pp. 1–11, 2020, doi: 10.1007/s13201-020-01218-y.

36. M. T. Amin, A. A. Alazba, and M. Shafiq, "Effective adsorption of methylene blue dye using activated carbon developed from the rosemary plant: Isotherms and kinetic studies," Desalin. Water Treat., vol. 74, pp. 336–345, 2017, doi: 10.5004/dwt.2017.20572.

37. K. H. Ranaweera, P. I. Godakumbura, and B. A. Perera, "Adsorptive removal of Co(II) in aqueous solutions using clearing nut seed powder," Heliyon, vol. 6, no. 4, p. e03684, 2020, doi: 10.1016/j.heliyon.2020.e03684.

38. L. N. Rao, S. Feroz, D. Kamalakar, M. V. Rao, J. C. C. E. A, and A. Pradesh, "Equilibrium, Kinetic and Thermodynamics Studies of Methylene Blue Dye from Aqueous Solution by Using Natural Adsorbent," Int. J. Pharm. Chem. Biol. Sci., vol. 4, no. 4, pp. 960–967, 2014. www.ijpcbs.com.

39. A. M. A. El Naggar, M. M. Ali, S. A. Abdel Maksoud, M. H. Taha, A. S. Morshedy, and A. A. Elzoghby, "Waste generated bio-char supported co-nanoparticles of nickel and cobalt oxides for efficient adsorption of uranium and organic pollutants from industrial phosphoric acid," J. Radioanal. Nucl. Chem., vol. 320, no. 3, pp. 741–755, 2019, doi: 10.1007/s10967-019-06529-2.

40. M. F. Cheira, M. N. Rashed, A. E. Mohamed, I. H. Zidan, and M. A. Awadallah, "The performance of Alizarin impregnated bentonite for the displacement of some heavy metals ions from the wet phosphoric acid," Sep. Sci. Technol., vol. 55, no. 17, pp. 3072–3088, 2020, doi: 10.1080/01496395.2019.1675701.

41. A. H. AbdEl-Salam, H. A. Ewais, and A. S. Basaleh, "Silver nanoparticles immobilised on the activated carbon as efficient adsorbent for removal of crystal violet dye from aqueous solutions. A kinetic study," J. Mol. Liq., vol. 248, pp. 833–841, 2017, doi: 10.1016/j.molliq.2017.10.109.

42. R. Foroutan, S. J. Peighambardoust, S. H. Peighambardoust, M. Pateiro, and J. M. Lorenzo, "Adsorption of crystal violet dye using activated carbon of lemon wood and activated carbon/fe3 o4 magnetic nanocomposite from aqueous solutions: A kinetic, equilibrium and thermodynamic study," Molecules, vol. 26, no. 8, pp. 1–19, 2021, doi: 10.3390/molecules26082241.

43. Y. Wang et al., "Preparation of microscale zero-valent iron-fly ash-bentonite composite and evaluation of its adsorption performance of crystal violet and methylene blue dyes," Environ. Sci. Pollut. Res., vol. 24, no. 24, pp. 20050–20062, 2017, doi: 10.1007/s11356-017-9426-2.

44. Z. Hasan, D. W. Cho, I. H. Nam, C. M. Chon, and H. Song, "Preparation of calcined zirconia-carbon composite from metal organic frameworks and its application to adsorption of crystal violet and salicylic acid," Materials (Basel)., vol. 9, no. 4, 2016, doi: 10.3390/ma9040261.

45. K. Mahmoudi, K. Hosni, N. Hamdi, and E. Srasra, "Kinetics and equilibrium studies on removal of methylene blue and methyl orange by adsorption onto activated carbon prepared from date pits-A comparative study," Korean J. Chem. Eng., vol. 32, no. 2, pp. 274–283, 2015, doi: 10.1007/s11814-014-0216-y.

46. Y. Raji et al., "Efficient Adsorption of Methyl Orange on Nanoporous Carbon from Agricultural Wastes : Characterization , Kinetics , Thermodynamics , Regeneration and Adsorption Mechanism," 2022.

47. M. Ennabely, "Removal of methyl orange dye from aqueous solutions using three types of biological activated carbons : Comparative study," pp. 0–12, 2022.

48. I. K. Taquieteu, H. T. Dzoujo, V. O. Shikuku, C. Banenzoué, and D. D. J. Dina, "Fixed-Bed Adsorption of an Azo Dye (Methyl Orange) onto Chemically and Thermally Regenerated Activated Carbons," J. Chem., vol. 2023, 2023, doi: 10.1155/2023/6677710.

University of Aden Journal of Natural and Applied Sciences., vol.29(1) (2025) 29 – 41

مجلة جامعة عدن للعلوم الطبيعية والتطبيقية

Journal homepage: https://uajnas.adenuniv.com

بحث علمي

إزالة الصبغ البنفسجي البلوري (CV) وصبغ الميثيل البرتقالي (MO) من المحاليل المائية باستخدام الكربون المنشّط المُحضَّر بطريقة خضراء من نفايات البلاستيك (PET)

أميمة أمين عبدالغني[1]* ، شائف محمد قاسم صالح [2]، زيد عبدالله العثمان[3] ، محمد أحمد حبيلة[3]

1. قسم الكيمياء، كلية التربية، جامعة عدن، اليمن.
2. قسم الكيمياء، كلية العلوم، جامعة عدن، اليمن
3. قسم الكيمياء، كلية العلوم ، جامعة الملك سعود، المملكة العربية السعودية

https://doi.org/10.47372/uajnas.2025.n1.a04

الملخص

إحدى الطرق السهلة لإزالة الألوان الاصطناعية من المحاليل المائية هي امتصاصها على مواد ماصة صلبة غير مكلفة. كان استخدام الكربون النشط من نفايات البوليمر محور مبادرات عملية الامتصاص الحديثة، وبالتالي فإنه يوفر خيارًا أكثر جاذبية وبأسعار معقولة من الكربون النشط التجاري، والذي غالبًا ما يكون أكثر تكلفة. تسعى هذه الدراسة إلى التحقيق في توازن وحركية الامتصاص لصبغتين كاتيونية وأيونية اصطناعيتين من الوسائط المائية وهما على التوالي البنفسجي البلوري والبرتقالي الميثيلين باستخدام نفايات البولي إيثيلين تيريفثالات PET لإنتاج الكربون النشط. بناءً على تحليل FTIR، تمتلك المواد الماصة مجموعة وظيفية OH في $C_6H_8O_7$ modified carbon-2 و KOH modified carbon-2 بنسبة تحضير 1:2، C-H، C = O و C-O. أشار تحليل المجهر الإلكتروني الماسح إلى وجود مسام بأحجام وأشكال مختلفة في قطر الميكرومتر. بالإضافة إلى ذلك، كشفت صور XRD عن وجود قمم حادة صغيرة في $C_6H_8O_7$ modified carbon-2/C-500 $2\theta = 29°$, $2\theta = 41°$ و $2\theta = 51°$ ؛ تشير النتائج إلى أن هذه القمم تعكس المستويات المتفحمة العطرية المكثفة. أجريت دراسات امتزاز الدفعات للتحقيق في تأثير الرقم الهيدروجيني الأولي ووقت التلامس وتركيز CV وMO الأولي وجرعة المادة الماصة ودرجة الحرارة على الامتزاز. قدمت نماذج تساوي الحرارة Langmuir وFreundlich تفسيرًا للبيانات التجريبية ذات أعلى معامل ارتباط مع صبغة CV، في حين أن نموذج تساوي الحرارة Freundlich مناسب لوصف تفاعل الامتزاز لصبغة MO. تُظهر بيانات الديناميكا الحرارية للامتزاز أن عملية الامتزاز تلقائية في جميع العينات باستثناء ($C_6H_8O_7$ modified carbon-1) التي كانت غير تلقائية. علاوة على ذلك، كانت العينة ماصة للحرارة في جميع العينات باستثناء ($C_6H_8O_7$ modified carbon-2) وكانت العينة طاردة للحرارة وأظهرت نفايات PET المنظفة التي تم الحصول عليها بعد تعديل السطح بواسطة معالجة $C_6H_8O_7$ قدرة امتصاص أعلى من KOH.

مفاتيح البحث

التسليم: 01/ 09 / 2025

القبول : 20 / 10 / 2025

كلمات مفتاحية:

إزالة الأصباغ (CV) و (MO)؛
التخليق الأخضر؛
الكاربون المنشط؛
نفايات البلاستيك (PET)؛