

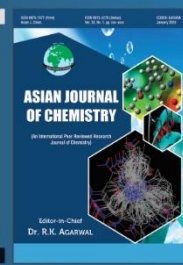


Asian Journal of Chemistry;

Vol. 38, No. 10 (2026), 2586-2600

ASIAN JOURNAL OF CHEMISTRY

<https://doi.org/10.14233/ajchem.2026.36444>



## Experimental and Computational Studies of Buffalo Horn-derived Activated Carbon for the Adsorption of Crystal Violet Dye

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Received: 16 May 2026

Accepted: 4 September 2026

Published online: 9 October 2026

AJC-22489

This study reports the utilization of abundantly discarded animal biomass specifically buffalo horn as a sustainable, low-cost precursor for producing functionalized activated carbon (BHAC) for the removal of crystal violet (CV) dye from textile wastewater. Comprehensive structural and morphological characterization (including SEM, TEM, XRD, XPS, BET) revealed that the KOH-mediated chemical activation successfully introduced heterogeneous pore structures and an iodine number of  $213.73 \text{ m}^2 \text{ g}^{-1}$ . Batch adsorption optimization studies involving adsorbent dose, pH, initial concentration, contact time and temperature achieved an impressive 95.72% removal efficiency of CV at pH 8 with equilibrium data fitting well to the Freundlich multilayer isotherm model ( $R^2 = 0.992$ ) and pseudo-second-order kinetics ( $R^2 = 0.999$ ). Thermodynamic parameters including Gibbs free energy change ( $\Delta G^\circ$ ), change in enthalpy ( $\Delta H^\circ$ ) and entropy ( $\Delta S^\circ$ ) confirmed that the adsorption process is spontaneous and exothermic, while regeneration study demonstrated stable reusability over six successive cycles. Furthermore, density functional theory (DFT) simulations fundamentally validated the adsorption energetics confirming that surface carboxyl groups ( $-\text{COOH}$ ) serve as the primary chemical binding sites driving favourable electrostatic interactions with the cationic adsorbate.

**Keywords:** Buffalo horn, Activated carbon, Cationic dye, Adsorption, Thermodynamics, DFT.

### INTRODUCTION

Industrial effluent pollution has become a major concern owing to its high toxicity and the severe risks it poses to both the environment and human existence. Likewise, the potential health risks associated with hazardous textile waste substances entering the human and animal food chains have garnered global attention. Annually, an incessant 40,000 to 50,000 tonnes of dyes are released into waterbodies as a result of inadequate treatment methods and outdated dyeing techniques within the textile industry [1,2]. These industrial effluents contain thousands of different chemicals and hazardous substances. They encompass both organic and inorganic contaminants that can deoxygenate the receiving waters thereby encouraging harmful microbial growth while introducing chemicals, which are detrimental to the aquatic ecosystem. Consequently, treating these wastewaters is pivotal. Among the most significant categories of adulterants found in wastewater discharged by the textile sector are organic dyes. Even at the low concentrations, the release of coloured effluents leads to severe environ-

mental degradation, causing aesthetic pollution and disrupting critical aquatic habitats [3,4].

Activated carbon is widely used for wastewater treatment due to its high porosity, cost-effectiveness and strong adsorption capacity. It effectively removes dyes and various toxic pollutants including pesticides, herbicides, heavy metals, phenols and chlorinated hydrocarbons [5]. Plant-based biowastes have become a prevalent choice for deriving biowastes, encompassing diverse forms such as flowers, roots, leaves, bark, etc. These plant-based biowaste has been the prime source for production of activated carbon besides the readily found carbon in the form of coal underneath the earth's surface. Agricultural wastes have also become a major source for activated carbon such as sugar beet pulp [6], rice husk [7], *Lawsonia inermis* seeds powder [8], Brazil nutshells [9], tea and plane tree leaves [10], pecan shell [11], etc. On the other hand, recent studies have made a good progress on the utilization of animal biowastes in producing activated carbon. Animal biowastes including chicken feathers, cattle bones, crab shells, etc. are a plentiful source of raw materials so also eco-

onomically alternative for producing activated carbon. Animal waste disposal causes environmental pollution but the production of activated carbon offers a vital eco-friendly method of treating wastewater. Biowastes such as waste human hair [12,13], chicken feather and eggshell [14], cow bones [15], cow manure [16], etc. have shown good absorbance towards different environmental pollutants.

Physical and chemical activation are the two principal techniques for producing activated carbon. Physical activation usually involves the use of high temperatures to pore-form the carbon material. On the contrary, chemical activation occurs at lower temperatures providing higher yield and is accomplished by two common methods (acid and alkali activation). Different activating agents are being used during chemical activation such as  $\text{ZnCl}_2$  [9,17],  $\text{HNO}_3$  [14],  $\text{NaOH}$  and  $\text{H}_3\text{PO}_4$  [18],  $\text{KOH}$  [15,16,19],  $\text{HCl}$  [20], etc. Potassium hydroxide ( $\text{KOH}$ ) is one of the prominent chemical activators employed in the synthesis of activated carbon. Its remarkable properties lie in its ability to yield exceptionally microporous activated carbons while concurrently promoting the creation of  $-\text{OH}$  groups of function within the activated carbon matrix. And this dual effect sets  $\text{KOH}$  apart as a superior option for generating activated carbon with enhanced porosity and surface functionality [19].

Synthetic organic dyes are widely used colouring compounds, with more than 100,000 varieties commercially available and global production exceeding 1 million tons annually. Some synthetic dyes have also been used as pharmacologically active substances in human and veterinary medicine [21]. Among which, crystal violet (CV), a triphenylmethane dye, is extensively utilized for colouring in the textile, paper and pharmaceutical sectors. Crystal violet (CV) in industrial effluents can cause serious environmental and health concerns due to its toxic, carcinogenic and mutagenic effects. It is widely used in textile, paint, printing, medical staining, Gram staining and veterinary applications [17,22].

Upcycling discarded buffalo horn biomass into activated carbon (BHAC) offers a sustainable dual benefit by reducing slaughterhouse/tannery biowaste and removing hazardous synthetic dyes from industrial effluents. Converting this abundant waste into an adsorbent reduces raw-material costs and adds value to the biomass. The synthesized BHAC was characterized using different analytical techniques, and the effects of CV concentration, adsorbent dose, pH and contact time were investigated, followed by equilibrium, kinetic and thermodynamic studies. DFT calculations at the B3LYP/6-31G level provided molecular-level insights, with  $-\text{COOH}$  showing the strongest binding affinity ( $-19.99 \text{ kJ mol}^{-1}$ ; bond length  $1.70 \text{ \AA}$ ) compared with  $-\text{OH}$  and  $-\text{CHO}$  groups. BHAC retained substantial adsorption capacity over multiple regeneration cycles using dilute acid or solvent eluents, highlighting its potential for cost-effective and sustainable wastewater treatment.

## EXPERIMENTAL

The antecedent substance, *i.e.* buffalo horn was collected from the butchery of Mokokchung district, India ( $26^\circ 19' 38''$  N latitude and  $94^\circ 31' 26''$  E longitude) and from the campus

of Nagaland University, Lumami, Zunheboto, Nagaland, India ( $26^\circ 13' 29.60''$  N latitude and  $94^\circ 28' 35.0''$  E longitude). Firstly, the raw materials were efficiently cleansed with tap water and distilled water after which the materials were sun dried for three days. The horns were then removed from the horn core and grounded into smaller pieces (3-5 cm) which was again rinsed with de-ionized water and kept for complete dry in an oven at  $110^\circ \text{C}$ . The horns being dried were then labelled as buffalo horn biomass (BHB) and subjected for further use. Potassium hydroxide (Merck, India) was used as the activating agent, while crystal violet (CV) was purchased from Rankem, Avantor. All chemicals used were of analytical grade and were used without any further pretreatment.

**Activated carbon through pyrolysis and activation:** The dried buffalo horn biomass (BHB) was cut into  $\sim 1 \text{ cm}$  pieces and carbonized at  $600^\circ \text{C}$  for 1 h in a muffle furnace (NSW 101 MF-3). The carbonized material was pulverized, sieved to  $106 \mu\text{m}$ , designated buffalo horn unactivated carbon (BHUC) and stored in a desiccator. For activation, BHUC was impregnated with  $\text{KOH}$  at  $\text{KOH}:\text{BHUC}$  ratios of 0.5:1, 1:1, and 3:1, stirred for 3 h, and dried at  $110^\circ \text{C}$ . The impregnated samples were then pyrolyzed at  $700^\circ \text{C}$  for 2 h under an autogenous atmosphere. After cooling, the carbon was washed thoroughly with  $0.1 \text{ M HCl}$ ,  $0.1 \text{ M NaOH}$  and deionized water until neutral pH, followed by drying at  $110^\circ \text{C}$ . The resulting buffalo horn activated carbon (BHAC) was stored in sealed containers for further analysis.

**Characterization:** The proximate analysis of raw and activated biocarbons was carried out according to ASTM D2866-94 [23]. The physico-chemical properties were characterized using several analytical techniques. Surface morphology was examined by scanning electron microscopy (SEM; JEOL JSM-6360) at different magnifications, while transmission electron microscopy (TEM; JEOL JEM-2100Plus) was used to investigate the nanoscale structure and surface features of BHC and BHAC using well-dispersed samples in ethanol. Fourier transform infrared spectroscopy (FTIR; Spectrum Two, Perkin-Elmer) was used to identify the functional groups in the carbon over  $4000\text{--}500 \text{ cm}^{-1}$  at a resolution of  $4 \text{ cm}^{-1}$ . X-ray diffraction (XRD) analysis was performed using an X-ray diffractometer (ULTIMA IV, Rigaku, Japan), and X-ray photoelectron spectroscopy (XPS; Thermo Fisher Scientific, ESCALAB 250Xi, USA) was used to examine the surface chemistry and electronic states. The surface area, pore-size distribution and pore volume were determined using a Brunauer–Emmett–Teller (BET) surface area analyzer (Quantachrome Autosorb iQ Station 1). CHN contents were determined using a CHN elemental analyzer (varioMACROcube, S/No. 2010BB 1028).

Batch adsorption experiments for the cationic dye were monitored using a UV-Vis spectrophotometer (Lambda 365, Perkin-Elmer, USA) at a maximum absorbance wavelength of  $586 \text{ nm}$ . Zeta potential was measured by laser Doppler anemometry using a Zetasizer Nano ZS (Malvern Instruments). The point of zero charge ( $\text{pH}_{\text{pzc}}$ ), representing the pH at which the net surface charge of the adsorbent is zero, was determined from the intersection of the  $\text{pH}_{\text{(final)}}$  versus  $\text{pH}_{\text{(initial)}}$  curve with the line  $\text{pH}_{\text{(final)}} = \text{pH}_{\text{(initial)}}$  [24]. For this determination,  $0.1 \text{ g}$  of BHAC was added to  $50 \text{ mL}$  of  $0.1 \text{ M KNO}_3$

solutions in conical flasks, with the initial pH adjusted from 2 to 12 using 0.1 N HCl or NaOH. The suspensions were placed on a rotary shaker for 24 h, after which the final pH was measured [25].

**Batch adsorption experiments:** The adsorption efficacy of BHAC for removing crystal violet (CV) from wastewater was evaluated using batch experiments. The effects of initial CV concentration, adsorbent dose, pH, contact time, agitation speed and temperature on the adsorption process were investigated. Briefly, 50 mL of CV solution with concentrations of 10–50 mg L<sup>-1</sup> was placed in 250 mL Erlenmeyer flasks containing BHAC at doses of 0.1–1 g L<sup>-1</sup>. The suspensions were agitated at 100–250 rpm and temperatures of 298–328 K for 10–180 min. After filtration, the residual CV concentration was determined using a double-beam UV-Vis spectrophotometer at  $\lambda_{\text{max}} = 586$  nm. All experiments were performed in triplicate. The adsorption capacity of BHAC and CV removal efficiency were calculated using the equations given below:

$$q_e = \frac{(C_o - C_e)}{m} \times V \quad (1)$$

$$\text{Removal (\%)} = \frac{C_o - C_e}{C_o} \times 100 \quad (2)$$

where  $q_e$  = adsorptive capacity (mg g<sup>-1</sup>),  $C_o$  = initial dye concentration (mg L<sup>-1</sup>),  $C_e$  = concentration at equilibrium (mg L<sup>-1</sup>),  $V$  = volume of CV solution (L) and  $m$  = adsorbent mass (g).

The experimental data were analyzed using the Langmuir, Freundlich, Temkin and Dubinin–Radushkevich (D–R) isotherm models. For the isotherm study, CV solutions with initial concentrations of 10–50 mg L<sup>-1</sup> were equilibrated with BHAC at an adsorbent dose of 0.75 g L<sup>-1</sup>. Adsorption kinetics were evaluated to determine the adsorption rate, equilibrium time and possible adsorption mechanism using the pseudo-first-order (PFO), pseudo-second-order (PSO), Elovich and intraparticle diffusion (IPD) models.

**Error analysis:** The chi-squared test ( $\chi^2$ ) and root mean square error (RMSE) [26] were used to determine the best-fit isotherm and kinetic models for the experimental data. The  $\chi^2$  test was calculated from the sum of the squared differences between the experimental and calculated data points.

The equation for chi-squared test can be obtained from the given equation [27]:

$$\chi^2 = \sum_{i=1}^n \frac{(q_{e(\text{exp})} - q_{e(\text{cal})})^2}{q_{e(\text{cal})}} \quad (4)$$

where  $q_{e(\text{cal})}$  = calculated equilibrium capacity (mg g<sup>-1</sup>),  $q_{e(\text{exp})}$  = equilibrium capacity of the experimental data (mg g<sup>-1</sup>),  $n$  = number of observations.

If the values obtained from the isotherm and kinetic models closely matched the experimental data, the  $\chi^2$  value was expected to be low. A higher  $\chi^2$  value indicated a higher deviation between the calculated and experimental data.

The RMSE was calculated from the given equation:

$$\text{RMSE} = \sqrt{\frac{1}{n-2} \sum_{i=1}^n (q_{e(\text{exp})} - q_{e(\text{cal})})^2} \quad (5)$$

**Theoretical study of crystal violet dye adsorption onto AC:** All molecular structures were modelled using Gauss-

View 6 and calculated with Gaussian 16W. Density functional theory (DFT) calculations were performed using the B3LYP hybrid functional with the 6-31G basis set in a dielectric medium with a permittivity of  $\epsilon = 80$ , corresponding to water, to optimize the ground-state geometries. Perry *et al.* [28] reported that carbonaceous materials primarily consisted of 3–7 fused benzene rings. Accordingly, fused benzene-ring cluster models were used to represent the carbon surface, as commonly applied in theoretical studies of adsorption on carbonaceous materials.

A zig-zag cluster model was used to represent the BHAC surface. The edge atoms at the active site were left unsaturated, while the remaining carbon atoms were terminated with hydrogen atoms. Three oxygen-containing functional groups, hydroxyl (–OH), aldehyde (–CHO) and carboxyl (–COOH), were introduced at the active sites to examine their effects on CV adsorption. The optimized models were designated pristine AC, AC–OH, AC–CHO, and AC–COOH (Fig. 1) [29]. The adsorption of CV on these carbon models was then examined to evaluate the possible interaction pathways. The adsorption energy of CV on the AC surface was calculated using eqn. 6:

$$E_{\text{Ad}} = E_{\text{CV+AC}} - (E_{\text{CV}} + E_{\text{AC}}) \quad (6)$$

where  $E_{\text{CV+AC}}$  = total energy of the CV dye and AC system,  $E_{\text{AC}}$  = total energy of AC and  $E_{\text{CV}}$  = total energy of CV dye. Adsorption energies below –30 kJ mol<sup>-1</sup> were considered indicative of physisorption, whereas values higher than –50 kJ mol<sup>-1</sup> were associated with chemisorption [30,31]. The H...N bond lengths between hydrogen atoms of the oxygen-containing functional groups on AC and nitrogen atoms of CV were evaluated to identify the strongest interaction and assess the adsorption affinity of the different functionalized carbon models.

## RESULTS AND DISCUSSION

**Physico-chemical characterization:** The results showed that activated carbon prepared at 700 °C with a KOH:BHUC impregnation ratio of 2:1 exhibited the highest surface area. Proximate and ultimate analyses were used to evaluate the properties of the activated carbon. As shown in Table-1, BHAC (2:1) had low moisture, volatile matter and ash contents of 4.91%, 19.8% and 0.60%, respectively, indicating favourable properties for adsorption. The results were consistent with previous reports [32–34]. The high fixed-carbon content further supported its potential as an effective adsorbent [34], which may be attributed to the removal of volatile components during thermal treatment and chemical activation [35]. Fixed carbon was calculated using eqn. 7:

$$\text{Fixed carbon (\%)} = 100 - (n_1 + n_2) \quad (7)$$

where  $n_1$  = volatile matter content (%),  $n_2$  = ash content (%).

As shown in Table-1, BHAC exhibited a specific surface area of 330.174 m<sup>2</sup> g<sup>-1</sup> and a pore volume of 0.319 cm<sup>3</sup> g<sup>-1</sup>. The specific surface area of commonly used activated carbons has been reported to range from 215 to 3532 m<sup>2</sup> g<sup>-1</sup> [36,37]. Comparable values have been reported for animal-biomass-derived adsorbents. Liu *et al.* [38] reported a BET surface area of 62.2 m<sup>2</sup> g<sup>-1</sup> for amino-functionalized pig bone-derived

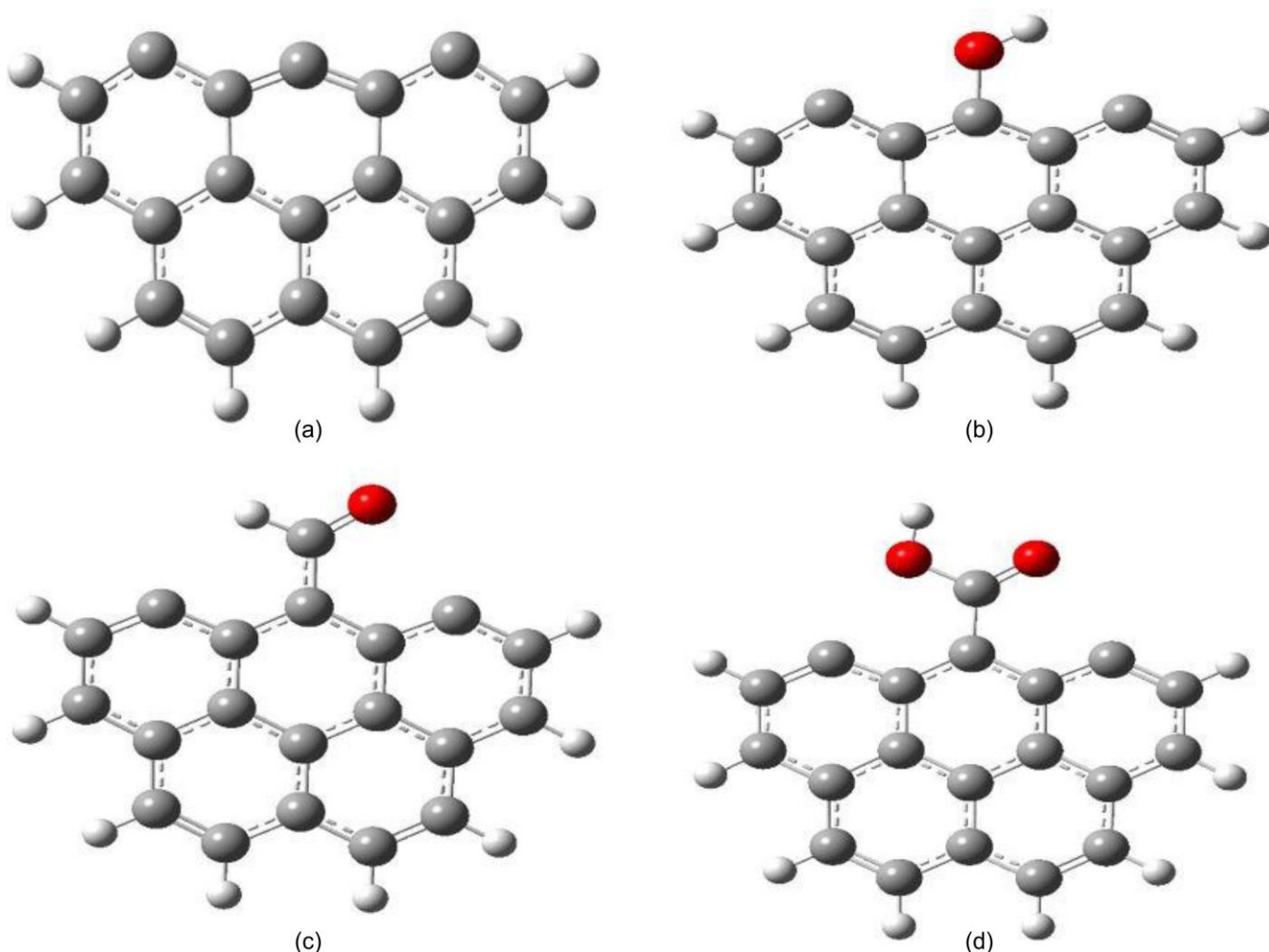


Fig. 1. The optimized models of (a) pristine AC (AC-P); (b) functionalized hydroxyl groups (AC-OH); (c) aldehyde-functionalized armchair (AC-CHO) and (d) functionalized carboxyl groups (AC-COOH)

Table-1  
PHYSICO-CHEMICAL CHARACTERISTICS OF BHAC AT DIFFERENT KOH: BHAC IMPREGNATION RATIOS

| Impregnation ratio (KOH: BHAC) | Proximate analysis   |                     |                 |                          | Water soluble matter (%) | Acid soluble matter (%) | Iodine number ( $\text{m}^2 \text{g}^{-1}$ ) |
|--------------------------------|----------------------|---------------------|-----------------|--------------------------|--------------------------|-------------------------|----------------------------------------------|
|                                | Moisture content (%) | Volatile matter (%) | Ash content (%) | Fixed carbon content (%) |                          |                         |                                              |
| 0.5:1                          | 11.9                 | 33.3                | 3.2             | 63.5                     | 16.5                     | 2.53                    | 145.03                                       |
| 1:1                            | 5.49                 | 25.2                | 1.58            | 73.22                    | 7                        | 2.50                    | 174.04                                       |
| 2:1                            | 4.91                 | 19.8                | 0.60            | 79.6                     | 1.5                      | 1.08                    | 213.73                                       |
| 3:1                            | 5.86                 | 31.9                | 2.79            | 65.31                    | 18                       | 5.82                    | 116.03                                       |

char used for heavy-metal removal. Isik *et al.* [12] reported a BET surface area of  $3.4107 \text{ m}^2 \text{g}^{-1}$  for hydrochar derived from waste human hair for the adsorption of cationic and anionic dyes, while Wang *et al.* [39] reported  $3.387 \text{ m}^2 \text{g}^{-1}$  for bovine bone waste used for cadmium and fluoride removal.

The CHNS analysis of BHAC prepared at a 2:1 impregnation ratio showed 72.08% carbon, 1.41% hydrogen, 8.94% nitrogen and 0.17% sulfur, with oxygen calculated by difference as 16.96%. With increasing impregnation ratio and activation temperature, the carbon content increased, whereas the hydrogen and oxygen contents decreased [27]. The prepared carbon exhibited low water-soluble matter, indicating its suitability for wastewater treatment. Its low acid-soluble

matter further indicated resistance to acid solubilization, supporting its potential application in acidic water treatment [40]. Pongener *et al.* [24] reported similarly low acid solubility for activated carbon prepared from *Manihot esculenta*. The iodine number served as a relative measure of porosity, with higher values generally associated with a higher degree of activation and enhanced adsorption capacity. The textural properties of the prepared activated carbon were evaluated by  $\text{N}_2$  adsorption-desorption analysis.

**Morphological studies:** The structural and surface characteristics of the activated carbon were examined using SEM. As shown in Fig. 2, BHAC exhibited a more porous surface than BHUC. BHUC showed a relatively smooth surface with

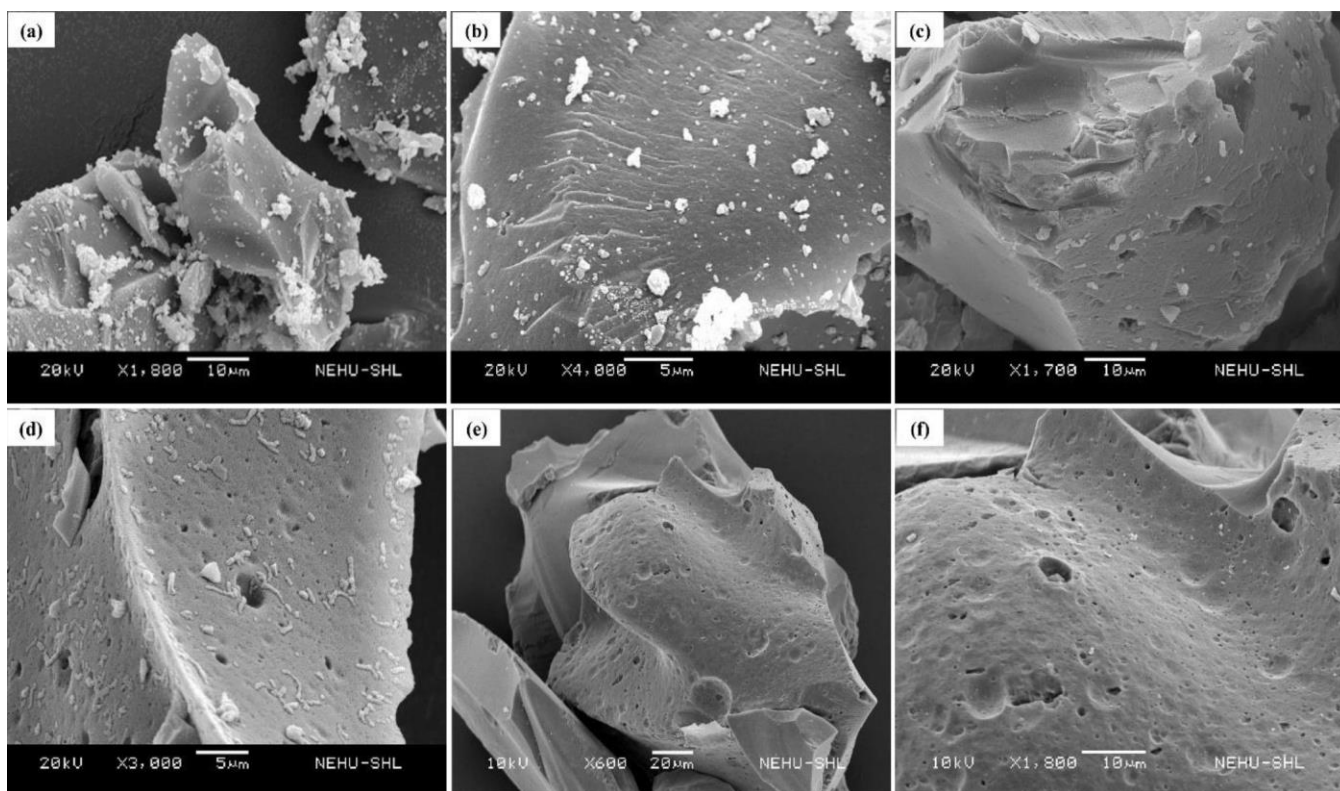


Fig. 2. SEM images under different magnification of (a,b) BHUC, (c,d) BHAC and (e,f) after adsorption

few visible pores and fissures, whereas BHAC displayed an irregular and rough surface with pores of different sizes and shapes, providing more accessible sites for adsorption. The irregular morphology of BHAC could be attributed to the interaction of the precursor with KOH during activation, which promoted pore formation and the release of volatile components during carbonization and activation. Sharma *et al.* [41] reported a similar irregular morphology following KOH activation, while Supong *et al.* [29] observed fractures, voids and pores in KOH-activated carbon. After CV adsorption, the morphology remained similar, although the surface appeared more degraded and partially blocked, supporting the occurrence of pollutant adsorption.

The TEM micrographs of BHUC and BHAC (Fig. 3), along with the corresponding SAED patterns shown in the insets, provided further information on their structural features. BHAC exhibited a distribution of dark and bright regions, where the dark regions were associated with pores and the brighter regions with pore walls. The TEM images suggested that KOH activation increased the accessible porous structure compared with BHUC [42]. During activation, the spaces between neighbouring structural domains may have expanded, forming channels and pores. Similar irregular surface features, including cracks and pores, were reported by de Souza *et al.* [43] for KOH-activated carbon, supporting the role of KOH in pore development.

**FTIR studies:** The FTIR spectrum of BHAC (Fig. 4) showed a broad band at  $3453\text{ cm}^{-1}$ , which was attributed to the O–H stretching vibration of hydroxyl groups [42]. The bands at  $2920$  and  $2847\text{ cm}^{-1}$  were assigned to aliphatic C–H stretching vibrations, while the band at  $1634\text{ cm}^{-1}$  was asso-

ciated with aromatic C=C stretching vibrations [44]. The bands observed in the  $1300\text{--}900\text{ cm}^{-1}$  region were assigned to C–O stretching vibrations of alcohol and phenol groups [45].

**XRD studies:** The XRD analysis was performed over a  $2\theta$  range of  $10\text{--}80^\circ$  to examine the structural characteristics of the carbon. The results showed that the activated carbon possessed a disordered microcrystalline structure, with graphitic microcrystallites arranged in a random orientation. As shown in Fig. 5, both BHUC and BHAC exhibited broad diffraction peaks. BHAC showed two broad peaks at approximately  $2\theta = 25^\circ$  and  $42^\circ$ , corresponding to the (002) and (100) planes, respectively. The broad nature of these peaks suggested the presence of poorly ordered graphitic microstructures in the prepared carbon [46]. The crystallite size of BHAC was estimated using the Debye–Scherrer equation [47]:

$$D = \frac{k\lambda}{\beta \cos \theta} \quad (8)$$

with regard to crystallographic planes,  $D$  denotes the average crystallite size,  $\lambda$  stands for X-ray wavelength ( $1.54\text{ \AA}$ ), while  $\beta$  represents the peak broadening at half maximum intensity (FWHM) in radians,  $k$  as the Scherrer constant (0.94) and the Bragg's diffraction angle as  $\theta$ .

The degree of crystallinity was derived from the following equation:

$$\text{Crystallinity} = \frac{\text{Area of crystalline peak}}{\text{Area of all peaks (crystalline + amorphous)}} \times 100 \quad (9)$$

Hence from the above equation, the BHAC crystallite size on average was determined as  $84.36\text{ nm}$  and the degree of crystallinity was observed to be  $49.19\%$ .

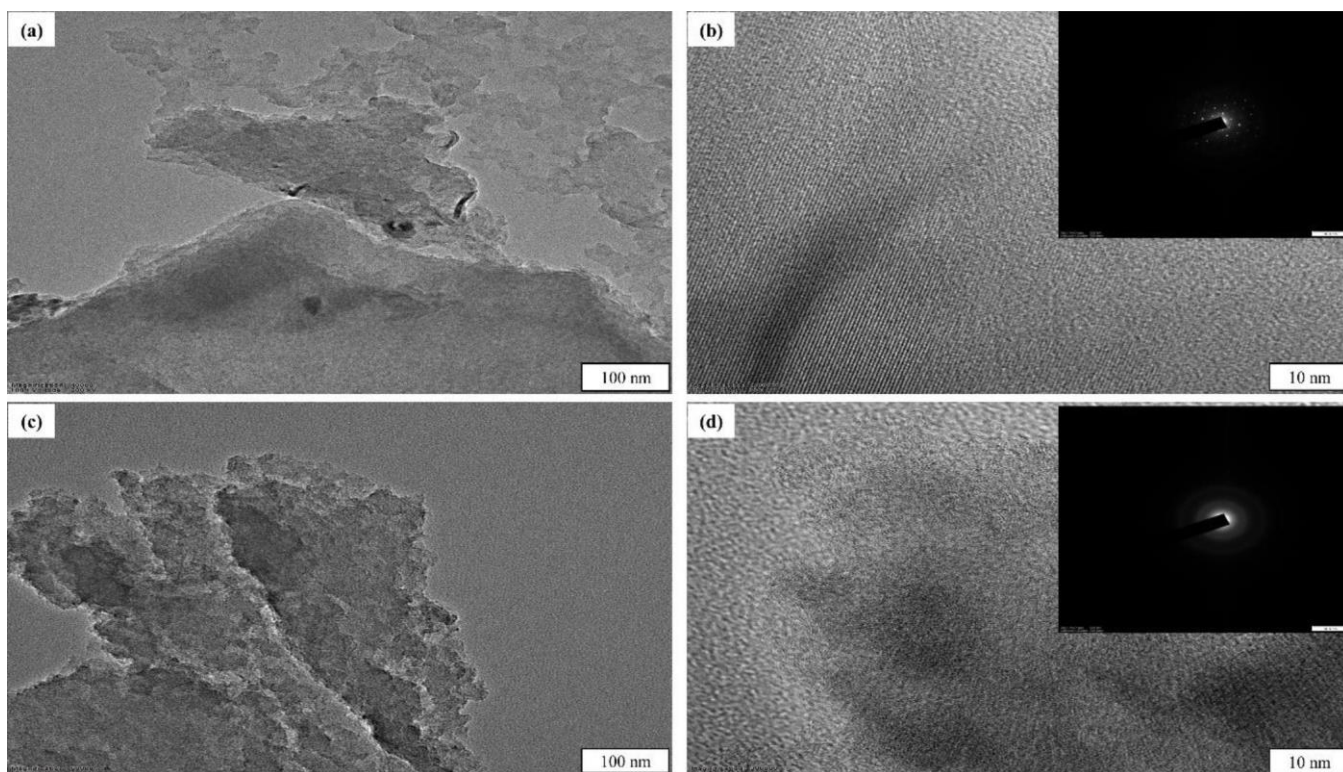


Fig. 3. TEM images under different magnification of (a,b) BHUC, (c,d) BHAC and SAED patterns at the insets of (b) and (d)

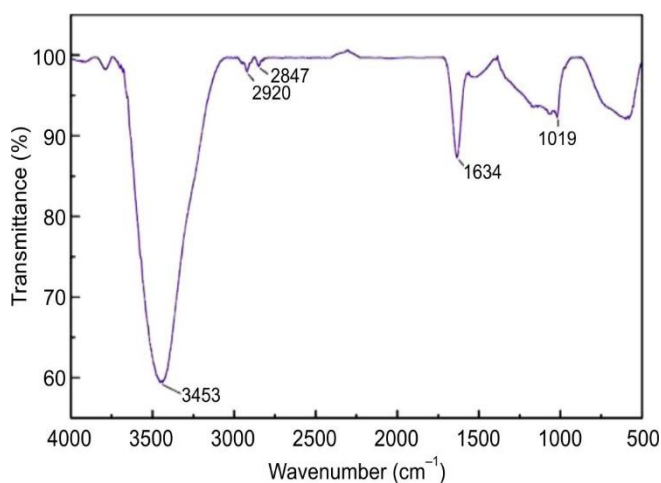


Fig. 4. FTIR spectrum of BHAC

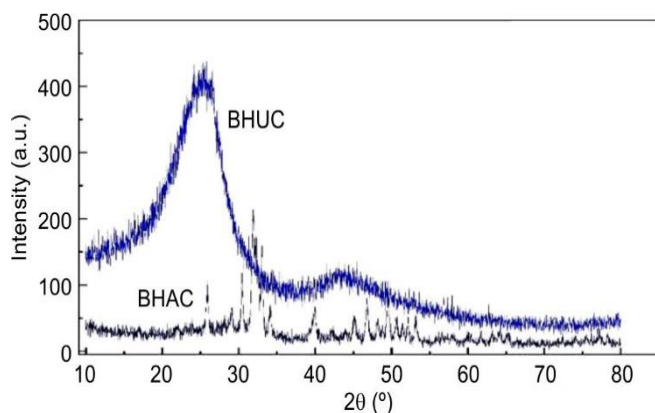


Fig. 5. XRD patterns of BHUC and BHAC

**Zeta potential studies:** Zeta potential measurements provide information on the surface charge characteristics of an adsorbent and can be used to assess its surface acidity or basicity [48]. As shown in Fig. 6, BHAC exhibited a zeta potential of -9.07 mV, indicating a negatively charged surface. A similar observation was reported by Yağmur & Kaya [49]. The point of zero charge ( $\text{pH}_{\text{pzc}}$ ) represents the pH at which the adsorbent surface has no net electrical charge, resulting in the minimal electrostatic attraction or repulsion toward charged species. The  $\text{pH}_{\text{pzc}}$  of BHAC was determined over an initial pH range of 2-12, as shown in Fig. 7.

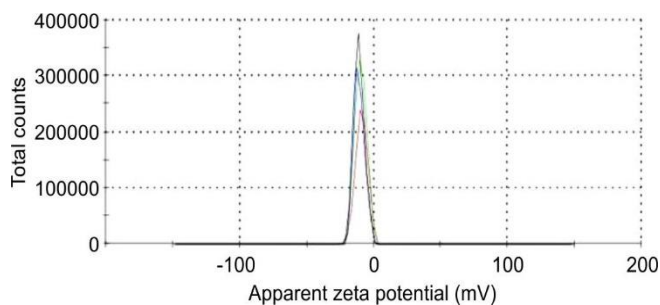


Fig. 6. Zeta potential distribution of BHAC

**XPS studies:** The elemental compositions of BHUC and BHAC shown in Fig. 8a-b revealed clear differences in their carbon and oxygen contents. BHAC exhibited a higher carbon content of 85.83 wt.% and 90.33 at.%, together with a lower oxygen content, compared with BHUC. The higher carbon content of BHAC suggested higher suitability for the removal of metallic species, dyes and other organic contaminants from aqueous solutions, as reported in the literature [50,51].

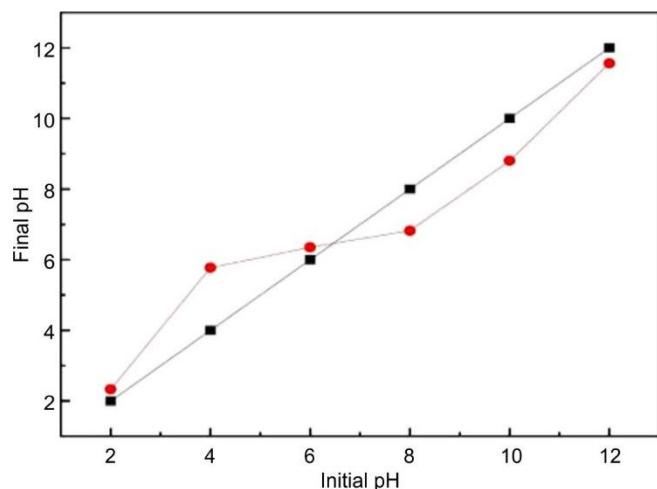


Fig. 7. pH variation plot for the determination of the  $pH_{zpc}$  of BHAC

As shown in Fig. 8c-f, the XPS survey spectra of BHAC and the deconvoluted C1s, O1s and N1s spectra exhibited distinct peaks corresponding to different surface chemical groups. The C1s peak at 284.80 eV was mainly assigned to graphitic C–C bonds [52], while the O1s peak at 530.90 eV was attributed to C=O bonds [19]. The N1s peak near 400 eV was associated with C–NH<sub>2</sub> species. According to Oginni *et al.* [19], KOH activation promotes the formation of oxygen-containing functional groups on the carbon surface. During washing, surface –OK groups can undergo ion exchange with water to form –OH groups, increasing the hydrophilic character and wettability of the activated carbon surface.

**Batch adsorption studies:** The effect of BHAC dosage was investigated over the range of 0.1–1.0 g L<sup>−1</sup> (Fig. 9a). The

CV removal efficiency increased from 45.58% to 96.6% with increasing adsorbent dosage, which was attributed to the availability of a higher number of active adsorption sites [53]. In contrast, the adsorption capacity decreased from 11.39 to 2.37 mg g<sup>−1</sup> at higher dosages, possibly due to the particle aggregation, which reduced the accessibility of active sites. Thus, an inverse relationship was observed between removal efficiency and adsorption capacity. Similar behaviour was reported by Sharma *et al.* [41]. A BHAC dosage of 0.75 g L<sup>−1</sup> was selected for subsequent experiments.

The effect of contact time on CV removal was studied over 10–180 min using a BHAC dosage of 0.75 g L<sup>−1</sup>, an initial CV concentration of 40 mg L<sup>−1</sup>, a temperature of 25 °C and pH 7. As shown in Fig. 9b, CV removal increased rapidly up to 90 min, which was attributed to the availability of abundant active functional groups and vacant surface sites [53,54]. The removal rate then approached equilibrium, and 90 min was selected as the equilibrium contact time for subsequent experiments. The decrease in adsorption rate after this period may be related to the reduced availability of adsorption sites and repulsive interactions between adsorbed and free CV molecules.

The effect of initial CV concentration was investigated over the range of 10–50 mg L<sup>−1</sup> (Fig. 9c). The removal efficiency initially increased with increasing CV concentration, owing to the enhanced concentration gradient and higher driving force for mass transfer between the liquid and solid phases. At higher concentrations, the adsorption efficiency decreased, which could be attributed to the progressive saturation of available active sites on the BHAC surface [55,56].

The effect of pH on CV adsorption was investigated over the range of pH 3–10 while maintaining the other parameters

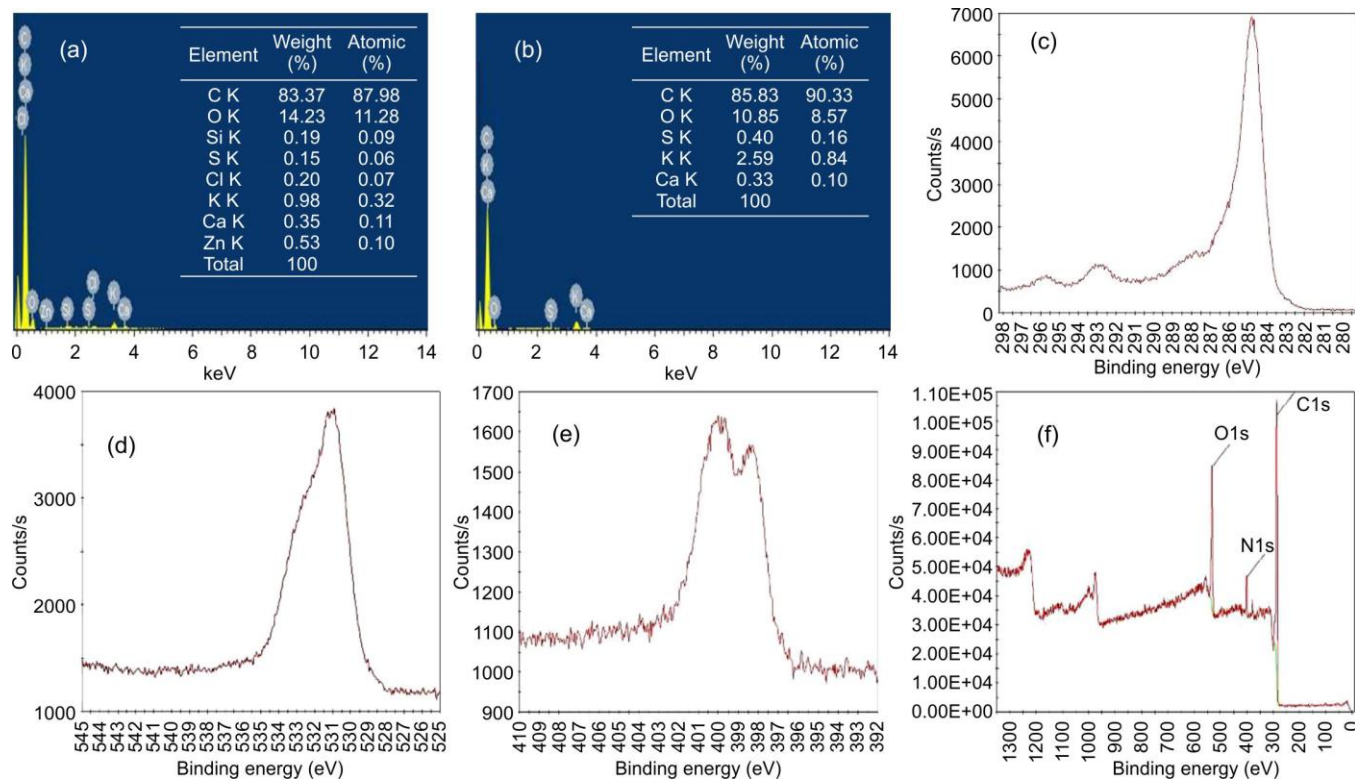


Fig. 8. EDX spectra of (a) BHUC and (b) BHAC, (c-f) XPS spectra of BHAC

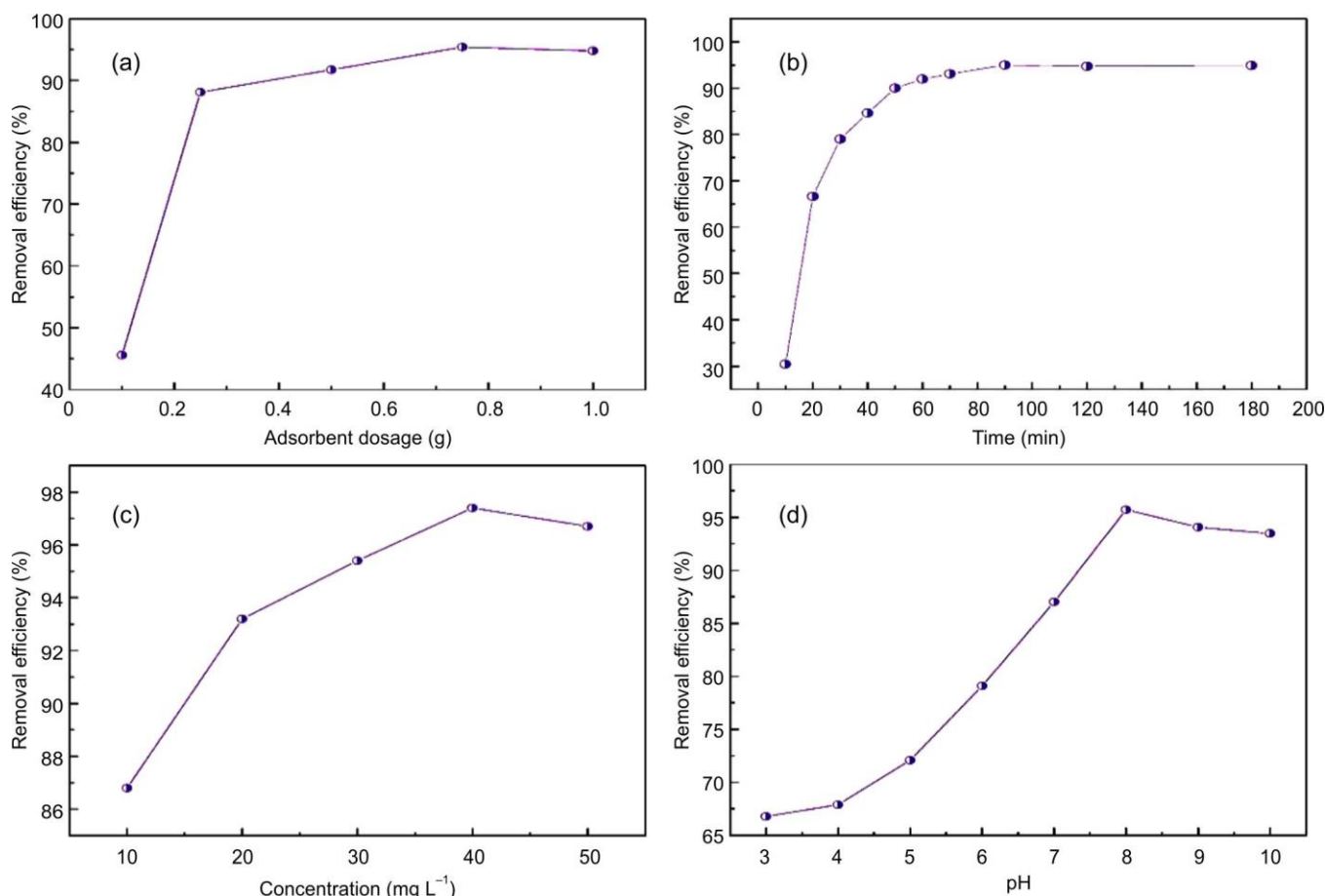


Fig. 9. Effect of (a) adsorbent dose, (b) contact time, (c) initial concentration (d) pH towards CV adsorption

at their equilibrium conditions. As shown in Fig. 9d, the highest removal efficiency of 95.72% was obtained at pH 8. The BHAC  $pH_{pzc}$  was determined to be 6.8, indicating that the surface became increasingly negatively charged above this pH. The higher adsorption of CV in alkaline conditions could be attributed to favourable electrostatic interactions between the cationic CV molecules and the negatively charged BHAC surface. Under acidic conditions,  $H^+$  ions competed with CV molecules for available adsorption sites, resulting in

lower removal efficiency. The observed pH-dependent behaviour was consistent with the findings of Foroutan *et al.* [57]. Patil *et al.* [58] similarly reported efficient CV removal using activated carbon derived from waste sugarcane leaves at pH 9.

**Adsorption isotherm models:** The experimental results concerning the uptake of dye along with the homogeneity and heterogeneity were determined by the four frequently employed isotherm algorithms as shown in Fig. 10. The isotherm data (Table-2) were evaluated using the linear forms of the

TABLE-2  
LIST OF ADSORPTION ISOTHERM EQUATIONS, FITTED PARAMETERS AND THEIR DATA FOR CV ADSORPTION

| Isotherm                   | Equation                                                                                                                                  | Parameters                                                       | $R^2$ | $\chi^2$ | RMSE  |
|----------------------------|-------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------|-------|----------|-------|
| Langmuir                   | $\frac{C_e}{q_e} = \frac{1}{Q_{\max} K_L} + \frac{1}{Q_{\max}} C_e$                                                                       | $q_m = 3.12$<br>$K_L = 0.90$<br>$R_L = 0.02$                     | 0.88  | 0.114    | 0.338 |
| Freundlich                 | $\log q_e = \frac{1}{n} (\log C_e) + \log K_F$                                                                                            | $1/n = 0.318$<br>$n = 3.14$<br>$K_F = 2.21$                      | 0.99  | 0.00062  | 0.025 |
| Temkin                     | $q_e = \frac{RT}{b_T} \ln C_e + \frac{RT}{b_T} \ln A_T$                                                                                   | $b_T = 2.614$<br>$A_T = 18.55$                                   | 0.98  | 0.002    | 0.053 |
| Dubinin-Radushkevich (D-R) | $\ln q_e = \ln q_{DR} - \beta \varepsilon^2$<br>$\varepsilon = RT \ln \left( 1 + \frac{1}{C_e} \right)$<br>$E = \frac{1}{(2\beta)^{1/2}}$ | $q_{DR} = 14.739$<br>$\beta = 7.63 \times 10^{-2}$<br>$E = 2.55$ | 0.96  | 0.014    | 0.121 |

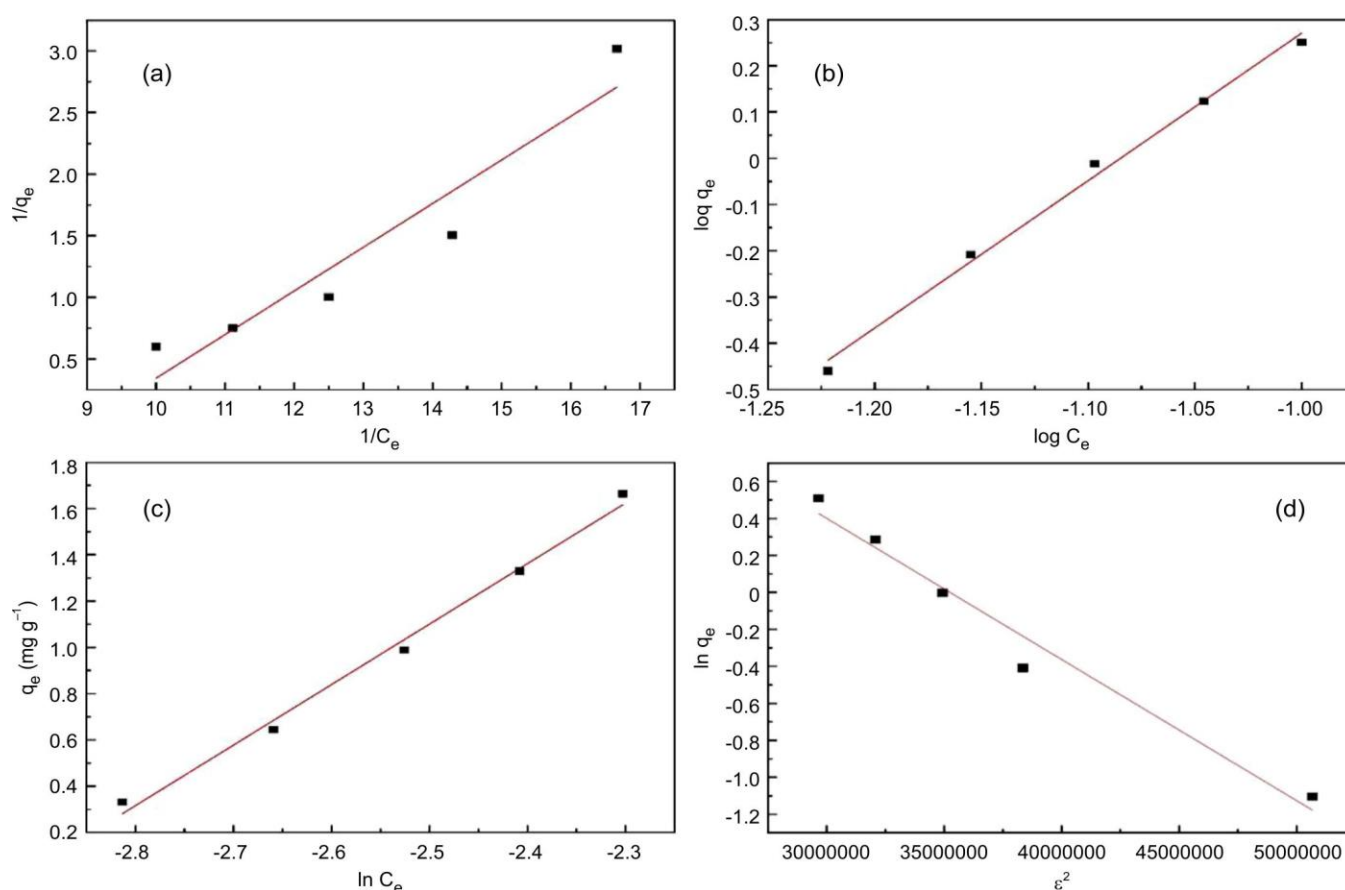


Fig. 10. Adsorption isotherm plots for CV: (a) Langmuir, (b) Freundlich, (c) Temkin and (d) Dubinin-Radushkevich (D-R) models

Langmuir, Freundlich, Temkin and Dubinin–Radushkevich (D–R) models. Among the models studied, the Freundlich isotherm provided the best fit to the experimental data, with a high correlation coefficient ( $R^2 = 0.99$ ). The Freundlich model was evaluated by plotting  $\log q_e$  against  $\log C_e$ . The parameter  $n$ , commonly referred to as the heterogeneity factor, describes the nature of the adsorption process. An  $n$  value of 1 represents linear adsorption, while  $n > 1$  is generally associated with favourable physical adsorption on heterogeneous surfaces. The Freundlich model is applicable to the multilayer adsorption on heterogeneous surfaces. Similar observations have been reported in previous studies [17,59,60].

The Langmuir model assumes monolayer adsorption on a homogeneous surface containing a finite number of equivalent

adsorption sites. Its lower  $R^2$  value indicated a poorer fit to the experimental data compared with the Freundlich model. The Temkin model yielded an adsorption heat constant ( $b_T$ ) of  $2.614 \text{ kJ mol}^{-1}$  and an  $R^2$  value of 0.98. The D–R model gave a theoretical monolayer adsorption capacity ( $q_{DR}$ ) of  $14.739 \text{ mg g}^{-1}$  and a mean free energy ( $E$ ) of  $2.55 \text{ kJ mol}^{-1}$ . Its  $R^2$  value of 0.96 indicated a comparatively weaker fit to the experimental data.

**Adsorption kinetic studies:** Four kinetic models were evaluated to study the adsorption behaviour of CV onto BHAC viz. the pseudo-first-order (PFO), pseudo-second-order (PSO), Elovich and intraparticle diffusion (IPD) models (Fig. 11). The linear equations and corresponding parameters are shown in Table-3. The PFO model described the adsorption of solute

TABLE-3  
KINETIC PARAMETERS OBTAINED FROM DIFFERENT MODELS FOR THE ADSORPTION OF CV

| Model                   | Equation                                              | Parameters                                                                | $R^2$ | $\chi^2$ | RMSE  |
|-------------------------|-------------------------------------------------------|---------------------------------------------------------------------------|-------|----------|-------|
| Pseudo-first order      | $\log(q_e - q_t) = \log q_e - \frac{K_1}{2.303} t$    | $q_{e, \text{exp}} = 1.33$<br>$q_{e, \text{cal}} = 4.91$<br>$k_1 = 1.327$ | 0.617 | 0.008    | 0.093 |
| Pseudo-second order     | $\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e}$ | $q_{e, \text{exp}} = 1.33$<br>$q_{e, \text{cal}} = 1.42$<br>$k_2 = 0.631$ | 0.999 | 0.00016  | 0.012 |
| Elovich model           | $q_t = \frac{1}{b} \ln(ab) + \frac{1}{b} \ln t$       | $a = 0.027$<br>$b = 1.045$                                                | 0.770 | 0.276    | 0.525 |
| Intraparticle diffusion | $q_t = K_p t^{1/2} + C$                               | $k_p = 0.0076$<br>$C = 1.096$                                             | 0.725 | 0.00019  | 0.014 |

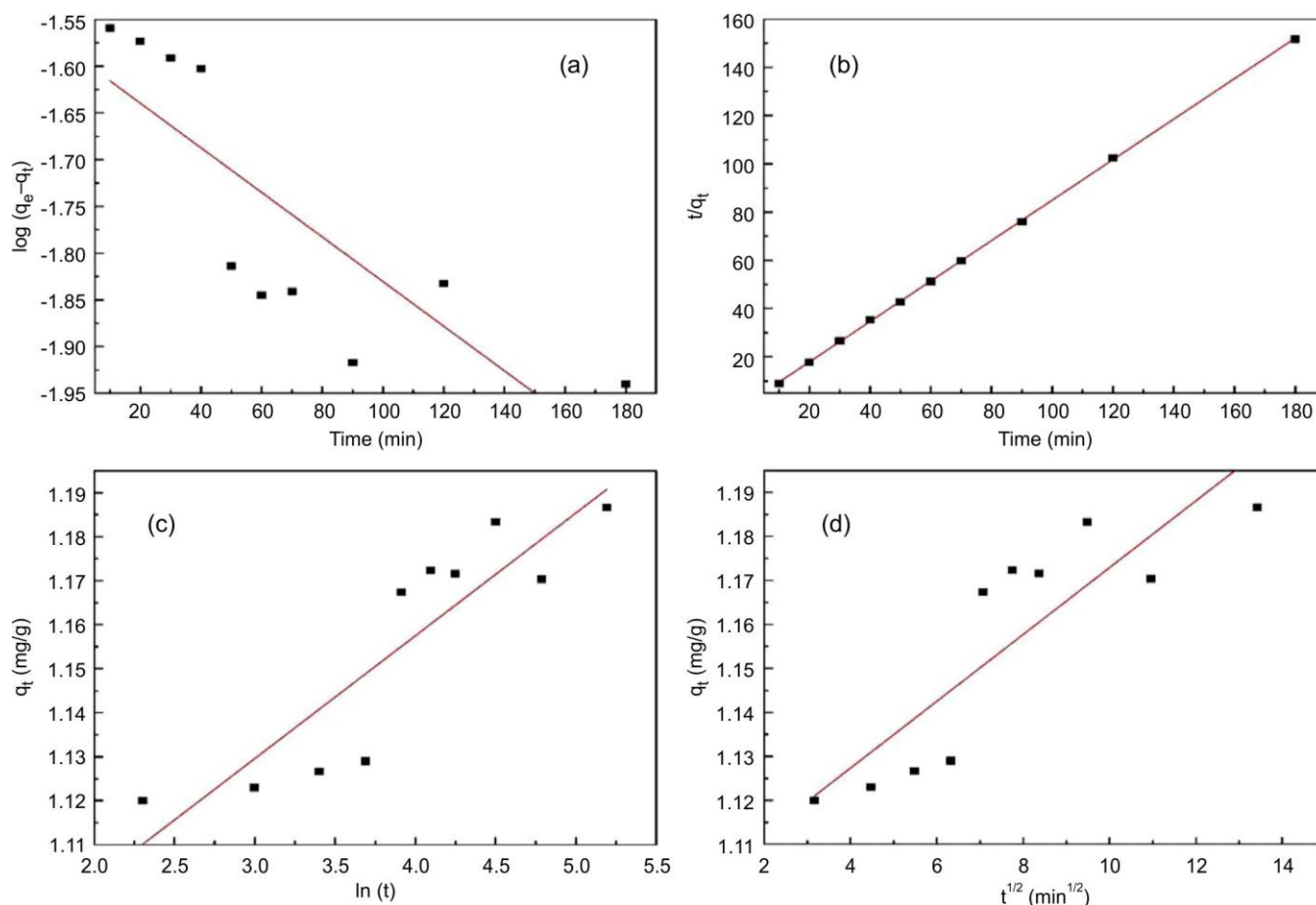


Fig. 11. Kinetic linear plots of (a) pseudo-first order, (b) pseudo-second order, (c) Elovich model, (d) intraparticle diffusion model for the adsorption of CV

molecules onto the adsorbent as a function of contact time [61,62]. It showed a correlation coefficient ( $R^2$ ) of 0.617 and a rate constant ( $K_1$ ) of 1.327. The low  $R^2$  value and poor agreement between the calculated and experimental adsorption capacities indicated that the PFO model did not adequately describe the adsorption kinetics. In contrast, the PSO model showed close agreement between the calculated and experimental adsorption capacities, with a high correlation coefficient ( $R^2 = 0.999$ ), indicating a better fit to the experimental adsorption data [62].

The Elovich model, which is commonly used to describe adsorption on heterogeneous surfaces, gave an  $R^2$  value of 0.770, indicating a weaker fit than the PSO model. The Weber–Morris intraparticle diffusion model was used to assess the possible diffusion mechanism and rate-limiting step. The resulting plots did not pass through the origin, suggesting that intraparticle diffusion was not the sole rate-controlling step. The applicability of the kinetic models was further assessed using  $\chi^2$  and RMSE values. As shown in Table-3, the PSO model exhibited the highest  $R^2$  and the lowest  $\chi^2$  and RMSE values, which confirm its closest fit to the experimental kinetic data.

**Thermodynamic studies:** The thermodynamic parameters of CV adsorption were evaluated at a temperature variation between 298–328 K. Furthermore, models of thermodynamic parameters such as the Gibbs free energy ( $\Delta G^\circ$ ),

enthalpy ( $\Delta H^\circ$ ) and entropy ( $\Delta S^\circ$ ) were computed to assess the feasibility and nature of the sorption process with the help of the following equations [54,63]:

$$\ln K_d = \frac{\Delta S^\circ}{R} - \frac{\Delta H^\circ}{RT} \quad (11)$$

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad (12)$$

$$K_d = \frac{q_e}{C_e} \quad (13)$$

where  $K_d$  = coefficient of adsorption distribution,  $q_e$  = adsorbed concentration of CV on BHAC ( $\text{mg g}^{-1}$ ),  $C_e$  = equilibrium concentration of CV ( $\text{mg L}^{-1}$ ),  $R$  = universal gas constant ( $8.314 \text{ J mol}^{-1} \text{ K}$ ),  $T$  = reaction/absolute temperature (K),  $\Delta G^\circ$  = Gibbs free energy change ( $\text{kJ mol}^{-1}$ ),  $\Delta H^\circ$  = enthalpy change ( $\text{kJ mol}^{-1}$ ),  $\Delta S^\circ$  = entropy change ( $\text{kJ mol}^{-1} \text{ K}^{-1}$ ).

Although the van't Hoff plot showed limited linearity ( $R^2 = 0.72$ ), the calculated thermodynamic parameters provided complementary information on the energetic characteristics of CV adsorption onto BHAC. Due to the deviation from ideal linearity, these parameters were interpreted together with the kinetic, equilibrium and other experimental results rather than as unrelated evidence of the adsorption mechanism. As shown in Table-4, the negative  $\Delta G^\circ$  values indicated that CV adsorption onto BHAC was spontaneous. The  $\Delta G^\circ$  values within the range of  $-20$  to  $0 \text{ kJ mol}^{-1}$  are generally

TABLE-4  
THERMODYNAMIC PARAMETERS FOR CV  
ADSORPTION AT DIFFERENT TEMPERATURES

| Temp. (K) | $\Delta G^\circ$<br>(kJ mol <sup>-1</sup> ) | $\Delta H^\circ$<br>(kJ mol <sup>-1</sup> ) | $\Delta S^\circ$<br>(kJ mol <sup>-1</sup> K <sup>-1</sup> ) |
|-----------|---------------------------------------------|---------------------------------------------|-------------------------------------------------------------|
| 298       | -3.87                                       |                                             |                                                             |
| 308       | -2.64                                       |                                             |                                                             |
| 318       | -1.41                                       | -40.66                                      | -0.124                                                      |
| 328       | -0.17                                       |                                             |                                                             |

associated with physisorption [57]. The negative  $\Delta H^\circ$  value indicated that the adsorption process was exothermic, while the negative  $\Delta S^\circ$  value suggested decreased randomness at the solid–liquid interface during adsorption. El Bourachdi *et al.* [64] reported that although the magnitude of  $\Delta H^\circ$  was relatively high for purely physical adsorption, it remained close to the range associated with weak adsorbate–adsorbent interactions, supporting physisorption as the predominant mechanism [57].

**Adsorbent regeneration:** In the present study, BHAC (1 g L<sup>-1</sup>) was stirred continuously with 50 mL of CV solution for 24 h to achieve saturation. The saturated adsorbent was then treated with 0.1 M NaOH under continuous stirring for 4 h for desorption. The regenerated carbon was filtered, thoroughly rinsed, dried at 110 °C and reused for subsequent CV adsorption cycles. As shown in Fig. 12, the CV removal efficiency decreased from 92.45% in the first cycle to 77.75% in the sixth cycle. This decrease could be attributed to progressive pore blockage and reduced accessibility of adsorption sites during repeated cycles. The incomplete desorption of CV by NaOH may have further contributed to the decline in adsorption performance. Residual NaOH on the carbon surface may likewise affect the availability of active sites and adsorption capacity [56]. SEM images (Fig. 2e-f) showed

noticeable changes in the BHAC surface after adsorption, including particle agglomeration and a rougher surface caused by CV deposition. The observed morphological changes, together with the regeneration results, indicated that a substantial fraction of the adsorbed CV could be removed during regeneration and that BHAC retained its adsorption performance over multiple cycles.

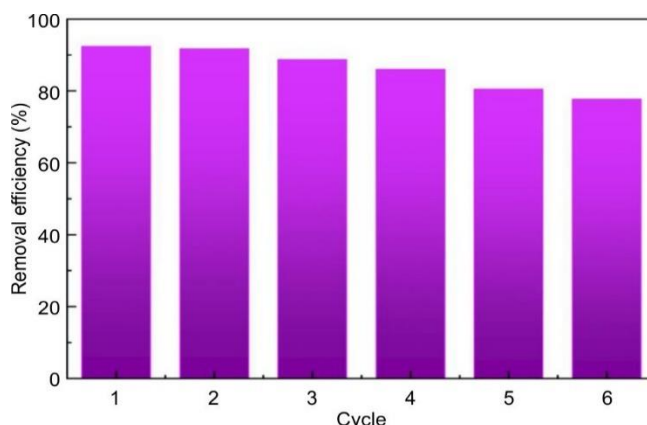


Fig. 12. Reusability and regeneration performance of BHAC over successive cycles

**Theoretical calculations:** To understand the interactions occurring during the adsorption process of CV onto BHAC, density functional calculations were applied. The adsorption phenomena of AC with CV were investigated using a zig-zag model since such research study provides valuable input to improving the preparation methods for BHAC. Fig. 13 displays the optimum configurations of the interactions between CV and BHAC. A summary of the computed adsorption energy ( $E_{ad}$ ) and bond lengths for different mode of interactions is shown in Table-5.

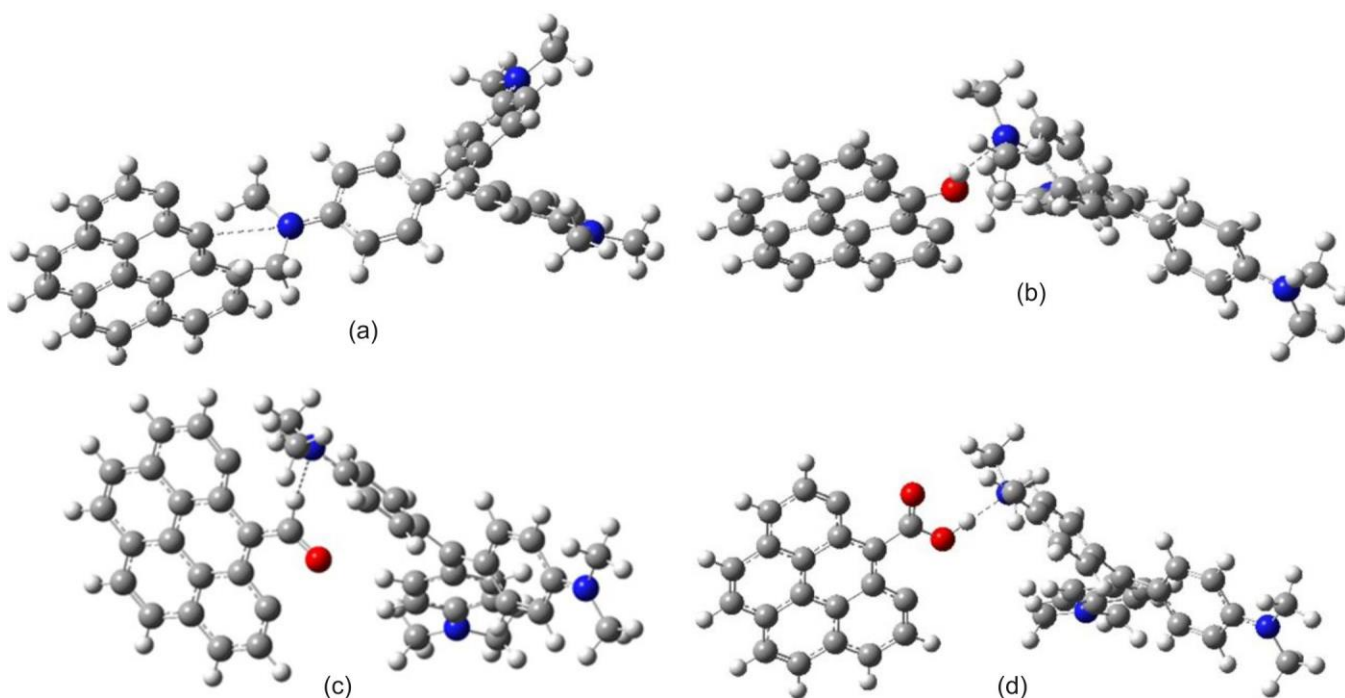


Fig. 13. Optimized structures of CV adsorption onto BHAC (a) (AC)C---N(CV) (b) (AC-OH)H---N(CV) (c) (AC-CHO)H---N(CV) (d) (AC-COOH)H---N(CV)

TABLE-5  
ADSORPTION ENERGIES AND  
BOND LENGTHS OF CV DYE AND BHAC SYSTEM

| System      | Mode of interaction | Adsorption energy (kJ/mol) | Bond length (Å) |
|-------------|---------------------|----------------------------|-----------------|
| CV + AC     | (CV)N---C(AC)       | -12.50                     | 2.57            |
| CV + AC-OH  | (CV)N---OH(AC)      | -16.43                     | 2.58            |
| CV + AC-CHO | (CV)N---HOC(AC)     | -15.29                     | 2.51            |
| CV+AC-COOH  | (CV)N---HOOC(AC)    | -19.99                     | 1.70            |

#### Adsorption of CV dye on pristine activated carbon:

For the CV and AC system, (C<sub>25</sub>H<sub>30</sub>ClN<sub>3</sub>)–N···C(AC) interactions mode was considered, in which one of the N groups of CV was aligned with the carbon atom of zig-zag edge site. Fig. 13 illustrates the bond formation and the optimized structures which clearly shows that the N group of CV molecule get adsorbed onto the zig-zag model of AC. The adsorption energy at -12.50 kJ mol<sup>-1</sup> with bond length of 2.57 Å, indicates that the CV adsorption on pristine activated carbon is favourable and physisorptive.

**Adsorption of CV on the functionalized activated carbon surface:** In this study, the interactions of CV with three oxygen-containing functional groups on BHAC (–OH, –CHO and –COOH) were examined using DFT calculations. The H···N bond lengths between the hydrogen atoms of the functional groups and the nitrogen atom of CV were evaluated alongside the adsorption energies to assess the strength of the interactions (Table-5). The AC–COOH model showed the strongest interaction, with an adsorption energy of -19.99 kJ mol<sup>-1</sup> and an H···N bond length of 1.70 Å. This was followed by AC–OH (-16.43 kJ mol<sup>-1</sup>; 2.51 Å) and AC–CHO (-15.29 kJ mol<sup>-1</sup>; 2.58 Å). The results indicated that the –COOH functional group contributed more strongly to CV adsorption than the –OH and –CHO groups. The adsorption energy profiles for the optimized CV–BHAC configurations were further analyzed to determine the most favourable interaction mode (Fig. 14) [37].

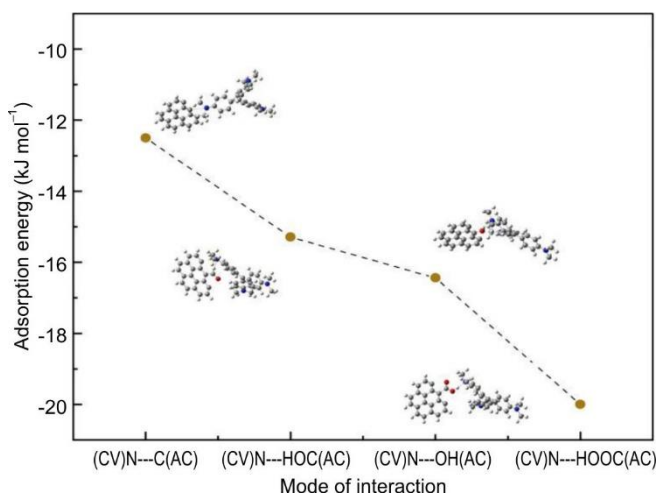


Fig. 14. Energy diagram for the adsorption of CV dye onto BHAC

**Cost analysis and comparison of BHAC with other adsorbents:** The total estimated cost of producing 223 g of BHAC was calculated by considering the expenses associated

with electricity, KOH, HCl, water and the raw precursor. The cost breakdown is shown in Table-6. Approximately 1 kg of raw buffalo horn biomass yielded 223 g of activated bio-carbon, with an estimated production cost of INR 502.89. This estimate excluded the initial capital investment, labour costs, capital expenditure, operational profit margins, recovery and reuse of KOH. The use of readily available and relatively inexpensive raw materials further supported the economic feasibility of the process. The production cost could decrease substantially upon scale-up from laboratory to industrial production.

TABLE-6  
ESTIMATED PRODUCTION COST OF 250 g OF BHAC

| Materials used                     | Consumption (approx.) | Estimated amount (approx.) in INR |
|------------------------------------|-----------------------|-----------------------------------|
| KOH                                | 250 g                 | 336                               |
| HCl                                | 250 mL of 0.1 M HCl   | 1.89 (INR 455/500 mL)             |
| Precursor                          | 1 kg                  | 0                                 |
| Electricity                        | 15 kWh                | 45                                |
| De-ionized water                   | 60 L                  | 120                               |
| Overall expenditure/250 g (in INR) |                       | 502.89                            |

Table-7 presents a comparison of BHAC with previously reported adsorbents for CV removal, considering different adsorption conditions and adsorbent characteristics that influence removal efficiency. Table-8 further compares the adsorption capacity of BHAC with those of other reported adsorbents for CV removal, providing a basis for evaluating its adsorption performance.

#### Conclusion

The successful upcycling of discarded buffalo horn biomass provided a dual environmental benefit by addressing animal-waste disposal while generating a cost-effective adsorbent for wastewater treatment. The study established buffalo horn-derived activated carbon (BHAC) as a sustainable and low-cost adsorbent for CV removal, with its porous structure supporting effective dye uptake. Although the enthalpy value ( $\Delta H^\circ = -40.66$  kJ mol<sup>-1</sup>) suggested a contribution from stronger adsorbate–adsorbent interactions, the low mean free energy ( $E = 2.55$  kJ mol<sup>-1</sup>) and other thermodynamic results supported physisorption as the predominant process. The maximum monolayer adsorption capacity ( $q_m = 3.12$  mg g<sup>-1</sup>) was relatively low compared with several other biomass-derived adsorbents, indicating limited adsorption capacity despite the high removal efficiency achieved under the optimized dilute conditions. BHAC retained a CV removal efficiency of 77.75% after six regeneration cycles, supporting its potential for repeated use. The combination of kinetic analysis and DFT calculations provided molecular-level insight into the adsorption process, with surface –COOH groups showing the strongest binding affinity toward CV among the functional groups investigated.

#### ACKNOWLEDGEMENTS

The author, Ketiyala Ao, gratefully acknowledge the financial support provided through the UGC-NET JRF fellow-

TABLE-7  
COMPARATIVE ANALYSIS OF CV ADSORPTION PERFORMANCE USING DIFFERENT ADSORBENTS

| Adsorbent                                                        | Surface area (m <sup>2</sup> g <sup>-1</sup> ) | Adsorbent dose | Concentration (mg L <sup>-1</sup> ) | Contact time (min) | pH   | Temp. (°C) | Ref.          |
|------------------------------------------------------------------|------------------------------------------------|----------------|-------------------------------------|--------------------|------|------------|---------------|
| Poultry litter                                                   | 148.05                                         | 2.5 g          | 50                                  | 45                 | 10.0 | 25         | [17]          |
| Orange peel                                                      | —                                              | 0.05 g         | 50                                  | 160                | —    | 30         | [56]          |
| Zeolite-montmorillonite                                          | 19.271                                         | 2.0 g          | 40                                  | —                  | 9.0  | 25         | [60]          |
| Peanut shells                                                    | —                                              | 10 mg          | 100                                 | —                  | —    | 30         | [65]          |
| Natural clay                                                     | 76.971                                         | 0.06 g         | 700                                 | 60                 | 8.0  | 22         | [66]          |
| Pumice stone                                                     | 7.06                                           | 0.5 g          | 100                                 | 150                | 6.5  | 25         | [67]          |
| <i>Date palm</i> (L.) dead leaflets                              | 161                                            | 50 mg          | 30                                  | 30                 | 6.5  | 27         | [68]          |
| Palm kernel shell                                                | —                                              | 0.5 g          | 400                                 | 24 h               | 7.0  | 25         | [69]          |
| Dredging sediment from dam                                       | 39.1                                           | 1.0 g          | 30                                  | 240                | 7.0  | 20 ± 1     | [70]          |
| <i>Platanus orientalis</i> (Chinar tree)                         | —                                              | 0.05 g         | 20                                  | 40                 | 6.52 | 25         | [71]          |
| Polymer composite (rubber granulate and scrap polyurethane foam) | 0.9473                                         | 0.5 g          | 50                                  | 200                | 7.0  | 50         | [72]          |
| Buffalo horn                                                     | 330.174                                        | 0.75 g         | 40                                  | 90                 | 7.0  | 25         | Current study |

TABLE-8  
COMPARATIVE ADSORPTION CAPACITIES OF DIFFERENT ADSORBENTS FOR REMOVAL OF CV

| Adsorbent                                                         | q <sub>m</sub> (mg g <sup>-1</sup> ) | Ref.          |
|-------------------------------------------------------------------|--------------------------------------|---------------|
| <i>Typha latifolia</i> activated carbon (TLAC)/chitosan composite | 0.269-2.375                          | [73]          |
| Raw cassava peels powder                                          | -5.15                                | [74]          |
| <i>Cedrus deodara</i> sawdust                                     | 0.673                                | [75]          |
| Sewage sludge                                                     | 3.87                                 | [76]          |
| Fish scales                                                       | 0.40                                 | [77]          |
| Crude chitin (RC)                                                 | -10.80                               | [78]          |
| Buffalo horn                                                      | 3.12                                 | Present study |

ship (NTA Ref. No. 190520065608). Tsenbeni N. Lotha (Award No. 202021-NFST NAG-03968) is thankful to the Ministry of Tribal Affairs, Government of India, under the National Fellowship for Scheduled Tribe (NFST) scheme. Lemzila Rudithongru (NU/RDC-200(1)-11 EvS/NNF/2022-3364) gratefully acknowledges the UGC Non-NET Fellowship for financial assistance. The authors also sincerely acknowledge the Department of Chemistry, Nagaland University, Lumami; SAIF, NEHU, Shillong; Tezpur University and NEIST, Jorhat, for providing the necessary research facilities. The authors further express their sincere gratitude to Dr. Francis A.S. Chipem, Department of Chemistry, Manipur University, for providing access to the computational facilities.

#### CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

#### DECLARATION OF AI-ASSISTED TECHNOLOGIES

During the preparation of this manuscript, the authors used an AI-assisted tool(s) to improve the language and no data, results or scientific conclusions were generated by AI. All analyses, interpretations and conclusions are the authors' own. The authors reviewed and edited the content and take full responsibility for the published work.

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