



RGO-CBPAC versus CBPAC for diluted leachate treatment: adsorption isotherms, kinetics and thermodynamics

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Abstract

This study compares carbon-based activated carbon (CBPAC) and reduced graphene oxide-modified CBPAC (RGO-CBPAC) for removing COD, $\text{NH}_3\text{-N}$, and turbidity from 1:4 diluted leachate from the Bingöl landfill site. Brunauer Emmett Teller (BET) analysis indicated severe micropore blockage in CBPAC after adsorption ($887 \rightarrow 0.76 \text{ m}^2/\text{g}$), whereas RGO-CBPAC retained a higher surface area ($908 \rightarrow 23 \text{ m}^2/\text{g}$) and mesoporous structure, demonstrating enhanced structural stability due to graphene incorporation. Fourier-transform infrared spectroscopy (FTIR) and scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM–EDX) confirmed the presence of surface functional groups and effective pollutant uptake, with RGO-CBPAC showing stronger surface–pollutant interactions. Zeta potential analysis showed pH-dependent surface charge behaviour, with CBPAC exhibiting amphoteric characteristics and RGO-CBPAC maintaining a more negative surface. After adsorption, both became more negatively charged, indicating the accumulation of negatively charged species. Kinetic analysis showed that COD adsorption on CBPAC followed pseudo-first-order and intraparticle diffusion models, whereas $\text{NH}_3\text{-N}$ and turbidity followed pseudo-second-order kinetics. In contrast, RGO-CBPAC exhibited pseudo-second-order and intraparticle diffusion behaviour for all pollutants, suggesting chemisorption with diffusion-controlled contributions. Adsorption equilibrium was best described by the Langmuir isotherm, yielding maximum capacities of 500 and 588 $\text{mg O}_2/\text{g}$ for COD, 17.5 and 36.5 mg/g for $\text{NH}_3\text{-N}$, and 18.1 and 58.5 mg/g ($(\text{NH}_2)_2\text{H}_2\text{SO}_4$)/g for turbidity on CBPAC and RGO-CBPAC, respectively. Thermodynamic analysis demonstrated that COD and turbidity removal using RGO-CBPAC were spontaneous ($\Delta G^\circ < 0$) and endothermic ($\Delta H^\circ > 0$), whereas $\text{NH}_3\text{-N}$ removal remained non-spontaneous for both adsorbents. RGO incorporation significantly enhanced adsorption performance.

Keywords Leachate · COD · $\text{NH}_3\text{-N}$ · Turbidity · CBPAC · RGO-CBPAC · COD

Introduction

Leachate originating from landfills forms when water penetrates the waste strata and participates in multiple biogeochemical and hydrological processes (Mor et al. 2006). Landfill leachate is characterized by elevated levels of total solids, chloride ions, colour, turbidity, total organic carbon (TOC), chemical and biochemical oxygen demands (COD

and BOD_5), $\text{NH}_3\text{-N}$, pH, and various heavy metal ions (KaurKamalpreet Mor et al. 2012; Ravindra et al. 2015). Due to their ability to contaminate surrounding land and aquatic systems, the organic, inorganic, and chlorinated constituents of leachate—together with volatile fatty acids, natural and synthetic ligands, and resistant organic fractions—are recognized as critical environmental pollutants (Wang et al. 2002; Aziz et al. 2011; Nor et al. 2013). The negative consequences of these toxic substances may be seen throughout every food chain level. According to research on the toxicity of 56 landfill leachate samples, 32 of the chemicals have been linked to cancer, 10 to congenital impairments, and 21 to genetic harm (Achankeng 2004). These problems have received much attention, leading to stringent regulations in several nations. Further investigation on landfill leachate treatment is encouraged by stricter pollution control regulations and revised discharge limits (Top

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et al. 2019). Therefore, landfill leachate must be treated to remove contaminants before it is released into water supplies or the environment.

To remove organic contaminants from leachate, several treatment techniques, including physicochemical, biological, leachate recirculation, advanced oxidation, and adsorption, have been reported. The majority of these alternative treatments are beneficial but have one or more drawbacks. For instance, biological processes only work well with young leachate and are ineffective with older leachate (Kurniawan et al. 2006; Gupta et al. 2012). Leachate recirculation is one of the least expensive techniques, although it limits methanogenic activity inhibition and saturation (Şan and Onay 2001). Advanced oxidation techniques are economically expensive and have chlorine oxidation challenges (Murray and Parsons 2004). For the treatment of leachate, alternative treatment approaches such as anaerobic processes, coagulation-flocculation processes (Zhang et al. 2012), and oxidation with ozonation (Oh et al. 2004) have also been researched, but they are hardly cost-effective.

Due to its universal applicability and ease of usage, adsorption is the most preferred technique among these techniques (Mor et al. 2007; Mittal et al. 2009). Commercially accessible carbon-based products, such as powdered activated carbon (PAC), are particularly effective for leachate treatment because of their exceptional adsorption capacities and microporous nature (Kaur et al. 2016). One of the most efficient adsorbents is activated carbon (AC), particularly for refractory organic molecules that resist decay by bacteria and remain in the environment (Zeid et al. 1995). It can additionally act as a highly efficient adsorbent for eliminating pollutants from leachate (Morawe et al. 1995). Activated carbon derived from biomass has been extensively studied as an adsorbent for the treatment of landfill leachate (Azmi et al. 2015; Lee and Hur 2016; Ab Ghani et al. 2017; Deng et al. 2018; Mahdavi et al. 2018). Activated carbon is commonly used in leachate treatment, although it is often combined with other processes to enhance treatment efficiency.

Graphene is a nanomaterial made of carbon with two dimensions and a certain atomic thickness. The oxidised version of graphene is called graphene oxide (GO). A wide range of functional groups-including carbonyl (C=O), epoxy (C–O–C), carboxyl (–COOH), alkoxy (C–O–R), and hydroxyl (–OH)-are present in its structure, influencing its chemical behaviour. These functional groups play an important role in the use of GO in various applications, especially as an adsorbent. GO may therefore swell in water and is highly hydrophilic. RGO, the reduced form of GO, is achieved through chemical, thermal, or photo-thermal reduction processes. Through these processes, most oxygen-containing functional groups are effectively removed, producing an RGO structure that approximates pure graphene, though not completely. Due to the incomplete reducing process, the conversion to graphene cannot be achieved entirely, leaving low amounts of oxygen groups such as epoxy, hydroxyl,

and carboxylic acid still present in its structure. However, its hydrophilic nature is significantly reduced. The reduction process diminishes the availability of active sites on RGO for polar contaminants; however, the restored conjugated π -system and heightened hydrophobicity facilitate the adsorption of non-polar and hydrophobic pollutants via π - π interactions and hydrophobic effects. Additionally, after reduction, RGO typically exhibits a more porous structure, which can further enhance its adsorption capacity for certain contaminants (Li et al. 2013; Perreault et al. 2015; Othman et al. 2018; Mombeshora and Muchuweni 2023). Similar improvements in pollutant removal efficiency associated with surface engineering of nanomaterials have also been reported in previous studies (Abazari et al. 2016; Safaralizadeh et al. 2017).

Current studies on landfill leachate treatment have not examined the application of coal-based powdered activated carbon (CBPAC) for the removal of COD, $\text{NH}_3\text{-N}$, and turbidity from *diluted leachate* at the Bingöl municipal solid waste disposal facility. To enhance the novelty and functional performance of the adsorbent, CBPAC particles were modified with high-surface-area, microporous reduced graphene oxide (RGO) nanoparticles, producing RGO-CBPAC composites. This strategy aims to integrate the inherent adsorption capacity of CBPAC with the hierarchical porosity and additional surface functionalities conferred by RGO, potentially improving pollutant uptake and diffusion kinetics. This study systematically compared the adsorption capacities and removal efficiencies of CBPAC and RGO-CBPAC under real landfill leachate conditions (1:4 dilution), highlighting the effects of RGO incorporation on micropore availability, surface functional groups, and electrostatic interactions. The results show that RGO-CBPAC enhances the removal of COD, $\text{NH}_3\text{-N}$, and turbidity while maintaining adsorption performance via preserved mesoporosity and functional sites, demonstrating the benefits of nanostructural modifications for improving conventional adsorbents and advancing leachate treatment.

Materials and methods

The leachate used in this study was collected from a landfill in Bingöl, Turkey, designed for household solid waste with a daily capacity of approximately 831.950 m³. The landfill base features a multilayer insulation system, including a mineral-based impermeable layer and a synthetic geomembrane, and a drainage layer was added to efficiently collect and transport the leachate.

A 20 L leachate sample was collected from the landfill using grab sampling in a polyethylene container. The sample was stored at 4 °C to maintain uniform physicochemical properties. Initial turbidity, $\text{NH}_3\text{-N}$, and COD were recorded for adsorption experiments (Table 1), with values of 228 NTU, 816 mg/L, and 4175 mg O₂/L. ECS Chemical



(Turkey) supplied the coal-based PAC (CBPAC), marketed as ENCARB CPW900, for the adsorption studies. Its main physical characteristics were presented in Table 2.

Reduced Nano Graphene Oxide (RGO), supplied by Graphene-Biotech Nanotechnology Engineering Ind. and Trad. (in Konya, Turkey), was employed in experimental studies.

A fixed-bed column system (100 mm in length and 16 mm in diameter) was employed to impregnate CBPAC particles with the prepared RGO solution, with upward flow from the bottom inlet to the top outlet (Fig. 1). In the experimental procedure, RGO nanoparticles in powder form were deposited onto CBPAC particles via a basic column arrangement. Based on the jar test results, an RGO suspension with a concentration of 50 mg/L was selected as the optimal operating condition to ensure adequate dispersion within the packed CBPAC bed, facilitate effective transport and attachment onto the surface, and prevent excessive agglomeration and pore blockage. This concentration was found to provide a balanced and controlled loading environment. The procedure was as follows: after the jar tests, the RGO nanoparticles were added to 500 mL beaker to prepare 50 mg/L suspensions. This suspension was then stirred for 3 h at 45 ± 2 °C and 500 rpm until a homogeneous dispersion was achieved in deionised water. The procedure was repeated until a sufficient quantity of RGO-CBPAC was obtained.

A peristaltic pump was employed to deliver the solution into the fixed-bed column system. Initially, 2 g of CBPAC was loaded into the column after placing a supporting layer at its base. The CBPAC bed was subsequently saturated by flushing deionised water through the column. Following this, the RGO suspension was introduced from the bottom inlet using the peristaltic pump, and the process was continued until the appearance of RGO in the effluent, indicating near-saturation of the CBPAC surface with respect to RGO loading. At this point, the flow was stopped, and the modified adsorbent was recovered from the column. The collected RGO-CBPAC particles were dried in an oven at

Table 2 CBPAC's physical characteristics

Physical properties	Value
Iodine number (mg/g)	> 900
Surface Area (m ² /g)	900–950
Particle Size (min. 90%)	200 mesh
Volumetric density (kg/m ³)	~250–550
Humidity (%)	< 5
Ash content (%)	< 18
Apparent density (kg/m ³)	500

80 °C overnight and then cooled to room temperature in a desiccator prior to weighing. This drying procedure was applied to remove physically retained moisture and to minimise its influence on mass measurements. The final dry mass of the adsorbent was determined to be 2.415 g. Based on the difference between the initial and final masses, the impregnation rate of RGO onto CBPAC was calculated to be approximately 20.75%. Under the applied drying conditions, the observed mass increase was attributed primarily to the immobilisation of RGO onto the CBPAC surface rather than residual moisture or impurities. Hereafter, the modified adsorbent is referred to as RGO-CBPAC. The diluted leachate was subsequently used to assess the removal efficiencies of COD, NH₃-N, and turbidity, along with the adsorption performance of CBPAC and RGO-CBPAC.

Batch adsorption investigations

To alleviate excessive organic and particulate content, the raw leachate was diluted at a ratio of 1:4 with deionised water. After dilution, the leachate characteristics were determined as

Table 1 Characterization of the physicochemical nature of leachate

Parameter	Unit	Value
SS	mg/L	221
VSS	"	35
TVS	"	2871
TDS	"	6654
TS	mg/L	7700
Conductivity	mS/cm	47
pH		8.35
Chloride	"	2872
NH ₃ -N	"	816
Turbidity	NTU	228
BOD ₅	mg O ₂ /L	1650
COD	mg O ₂ /L	4175
BOD ₅ / COD	–	0.4

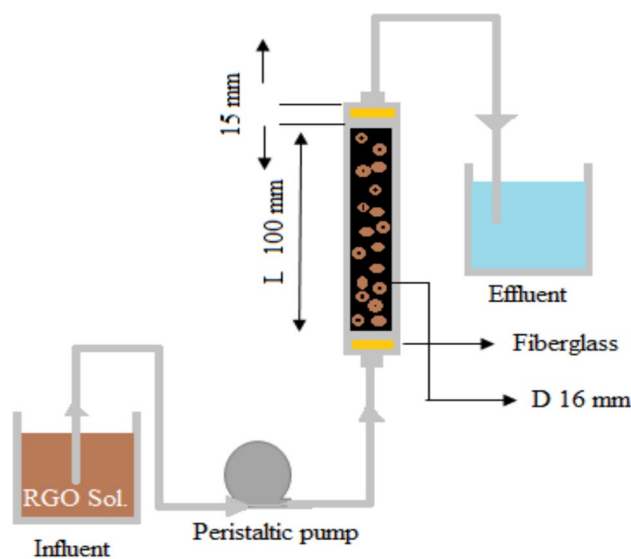


Fig. 1 Schematic representation of the fixed bed column



1044 mg O₂/L COD, 204 mg/L NH₃-N, and 57 NTU turbidity. In the initial stage of the adsorption experiments, 400 mL of the diluted leachate was transferred into a 500 mL cylindrical glass reactor. A predetermined dosage of CBPAC or RGO-CBPAC was then added, and adsorption trials were conducted under the leachate's natural pH of 8.35, at 20 °C, and 300 rpm agitation. The removal efficiencies and adsorption capacities for turbidity, NH₃-N, and COD were subsequently examined under varying conditions, including temperature (20–60 °C), pH (5–9.5), agitation rate (100–300 rpm), and adsorbent dosage (1–5 g/L). A Daihan JT-M6C six-stirrer jar-test apparatus was employed to assess the removal performance. For pH adjustment of the 400 mL samples, 10⁻² M NaOH and H₂SO₄ solutions were used.

Jar-test results indicated that the adsorption of COD, NH₃-N, and turbidity using 5 g of CBPAC and RGO-CBPAC particles reached equilibrium within approximately 40 min; therefore, 40 min was selected as the optimum adsorption time. During this period, samples were withdrawn from the batch reactor at 5, 10, 15, 20, 25, 30, and 40 min. Prior to COD, NH₃-N, and turbidity analyses, the collected samples were filtered through 0.45 µm filter paper (Whatman No. 42) and subsequently diluted fivefold with deionized water. This pre-treatment ensured the suitability of the samples for accurate measurement.

Chemical agents

Analytical-grade reagents obtained from Sigma-Aldrich and Merck (USA) were used without further purification. The pH of the leachate was adjusted using H₂SO₄ (98%, Merck) and NaOH (≥98%, Sigma-Aldrich). Chloride concentrations were determined with H₂O₂ (30%, Sigma-Aldrich), Al(OH)₃, K₂CrO₄ (≥99%), and AgNO₃ (≥99%), all supplied by Sigma-Aldrich. Turbidity was analysed using hydrazine sulphate and hexamethylenetetramine (≥99%, Sigma-Aldrich). NH₃-N was quantified with reagents from Sigma-Aldrich and Merck, including phenol (99.5%), sodium nitroprusside dihydrate (≥99%), trisodium citrate dihydrate (≥99%), NaOH (≥98%), and NaOCl (~10%). COD analysis was performed using H₂SO₄ (98%), K₂Cr₂O₇ (≥99%), and Ag₂SO₄ (≥99.99%), purchased from Merck and Sigma-Aldrich.

Analyses

Physical analyses

Leachate characterization included measurements of total dissolved solids (TDS), conductivity, and pH using a Thermo Scientific Orion Versa Star instrument, while turbidity, colour, and total suspended solids (TSS) were determined in accordance with standard analytical methods (Rice et al. 2012).

Turbidity was measured using a standard turbidimeter calibrated with formazin-based standard solutions. The turbidity standard was prepared through the reaction of hydrazine sulfate (NH₂)₂H₂SO₄ and hexamethylenetetramine (CH₂)₆N₄, resulting in the formation of stable colloidal formazin particles responsible for light scattering. Turbidity values were initially recorded in NTU. For adsorption isotherm and kinetic evaluations, NTU values were converted into formazin-equivalent concentrations expressed as mg (NH₂)₂H₂SO₄/L using the experimentally determined linear relationship: C_{eq} = 1.25 × NTU. This conversion was applied to ensure consistency between optical turbidity measurements and mass-based adsorption modelling.

Chemical analyses

For COD determination, the SM 5220.D Closed Reflux Colorimetric Method was applied, and absorbance at 600 nm was used to calculate the concentrations (Rice et al. 2012). For NH₃-N determination, the 4500-F phenate method was applied, and sample concentrations were obtained from absorbance values at 640 nm (Rice et al. 2012). For chloride determination, the standard 4500-Cl-B argentometric method was applied, and the blank titration values were subtracted from the sample readings (Rice et al. 2012).

All measurements for COD, NH₃-N, and turbidity were performed in triplicate, with mean values reported and error bars included in all plots.

Zeta potential analysis

A PSS NICOMP 380 ZLS instrument was employed to assess the zeta potentials of CBPAC and RGO-CBPAC particles.

SEM-EDS analysis

CBPAC and RGO-CBPAC particles were characterized using a Zeiss EVA MA10 scanning electron microscope (SEM) equipped with energy-dispersive X-ray spectroscopy (EDS).

FTIR analysis

Fourier-transform infrared (FTIR) spectra of CBPAC and RGO-CBPAC particles were recorded using a Thermo Scientific Nicolet IS 5 instrument.



Adsorption equilibrium and modelling

The condition in which adsorption and desorption rates of molecules at the solid–liquid interface is equal at a certain temperature is referred to as adsorption equilibrium, as illustrated in Eq. (1).

If an equilibrium is reached,

$$q_{eq} = f(C_{eq}) \quad (1)$$

A mass balance between incoming and outgoing substances during adsorption was used to derive Eq. (2).

$$V_L(C_0 - C_{eq}) = m_a(q_{eq} - q_0) \quad (2)$$

Equation (3) is found at $t=0$ and $q_0=0$ when the adsorbent is not loaded.

$$q_{eq} = \frac{V_L}{m_a}(C_0 - C_{eq}) \quad (3)$$

Efficiency in batch adsorption testing was determined using Eq. (4).

$$\text{Eff.}(\%) = \frac{(C_0 - C_e)}{C_0} \times 100 \quad (4)$$

where;

C_0 : the adsorbate's beginning concentration (mg/L),

C_{eq} : the equilibrium concentration of the adsorbate (mg/L),

q_{eq} : the mass of the adsorbent relative to the amount of pollutant absorbed at equilibrium (mg/g),

V_L : volume (L),

m_a : the amount of adsorbent (g).

Assessment of potential leaching from CBPAC and RGO-CBPAC

A preliminary leaching test was performed to evaluate the potential release of soluble species from CBPAC and RGO-CBPAC under different pH conditions. Batch reactors containing 400 mL of deionised water were prepared at initial pH values of 5, 7, 8.35, and 9.5, respectively, while the temperature was maintained at 20 °C. In each reactor, 5 g of adsorbent was introduced, followed by agitation at 300 rpm for 40 min to simulate operational mixing conditions. After equilibration, the suspensions were filtered through a 0.45 µm membrane filter, and the filtrates were analysed for COD, $\text{NH}_3\text{-N}$, and turbidity. The same procedure was repeated for RGO-CBPAC under identical experimental conditions. The results indicated that only very low levels of COD (approximately 0.5–5 mg/L), $\text{NH}_3\text{-N}$ (generally below 0.05 mg/L), and turbidity (very low NTU values) were detected for CBPAC. These values were determined to be

negligible and not significant enough to influence the overall experimental results. For RGO-CBPAC, COD (approximately 0.5–6 mg/L), $\text{NH}_3\text{-N}$ (mostly below the detection limit), and turbidity (typically < 1 NTU) were observed. Similarly, these values were considered negligible and did not contribute meaningfully to the aqueous phase contamination.

In general, the obtained results demonstrate that both CBPAC and RGO-CBPAC exhibit stable behaviour under the tested conditions, with only minimal release of dissolved and particulate species into the solution. This indicates that secondary contamination from the adsorbents is negligible under the experimental conditions applied in this study.

Adsorption isotherms

The amount of pollutant adsorbed per unit mass of adsorbent is directly related to the concentration of the pollutant remaining in solution at equilibrium for a given temperature. Adsorption isotherms are widely employed to evaluate the maximum adsorption capacity of an adsorbent. In this study, the adsorption behaviour of CBPAC and RGO-CBPAC was analysed using Dubinin–Radushkevich (D–R), Langmuir, Freundlich, and Temkin isotherm models. The linearized forms of these models, applied to determine the corresponding isotherm parameters, are briefly summarized below.

Dubinin–Radushkevich isotherm

The D–R theory indicates that the Polanyi curve, illustrating the adsorption potential of micropores, undergoes shifts that are Gaussian in nature. The adsorption potential ($\mathcal{E}$) is defined by an experimental equation, and the corresponding adsorption energy can be calculated using the D–R isotherm. Specifically, the slope of the resulting graph is employed to determine the adsorption energy, the magnitude of which indicates the nature of the adsorption process (Dubinin 1947). Adsorption energy, defined as the energy released during the removal of one mole of adsorbate by the adsorbent, is obtained from the D–R isotherm, with the linearized expression shown in Eq. (5).

$$\ln q_e = \ln X'_m - K' \mathcal{E}^2 \quad (5)$$

Equation (6) yields the adsorption energy (E , kJ/mol), which offers information on the adsorption characteristic.

$$E = \frac{1}{\sqrt{2K'}} \quad (6)$$

Equation (7) is used to get the value ($\mathcal{E}$) of the D–R constant.

$$\mathcal{E} = RT \ln \left[1 + \frac{1}{C_e} \right] \quad (7)$$



The letters E , R , T , C_e , X'_m , and K correspond to the Polanyi potential, gas constant (8.314 J/mol K), temperature in Kelvin, equilibrium adsorbate concentration, adsorption capacity, and adsorption energy constant.

The line slope is denoted by K' , and $\ln X'_m$ is determined from the intercept on the $\ln q_e$ axis in a plot of $\ln q_e$ versus E^2 .

Langmuir isotherm

In the Langmuir model, adsorption occurs at a limited number of equivalent sites, leading to the formation of a saturated monolayer on the adsorbent surface, with pollutants competing for these active sites (Langmuir and Herman 1980).

The linearized Langmuir equation is represented by Eq. (8).

$$\frac{1}{q_e} = \frac{1}{bK} \times \frac{1}{C_e} + \frac{1}{b} \quad (8)$$

Freundlich isotherm

Freundlich's isotherm, developed by H. M. F. Freundlich, models adsorption on heterogeneous solid surfaces (Freundlich 1906). Equation (9) represents the Freundlich isotherm after linearization.

$$\log q_e = \log K_f + \left(\frac{1}{n}\right) \log C_e \quad (9)$$

In the $\log C_e$ – $\log q_e$ plot, the intercept gives $\log K_f$, while the slope is used to compute $(1/n)$. At equilibrium, the amount adsorbed per unit adsorbent is q_e (mg/g), Freundlich constants are K_f and n , and C_e (mg/L) represents the pollutant concentration in the liquid phase.

Temkin isotherm

According to Temkin's model, the adsorption heat decreases linearly with increasing surface coverage because of adsorbent–pollutant interactions. The corresponding linearized equation is given in Eq. (10) (Temkin 1940).

$$q_e = B \times \ln A_T + B \times \ln C_e \quad (10)$$

In Temkin analysis, the slope of $\ln C_e$ versus q_e determines B , while the intercept with the q_e axis provides $\ln A_T$, with B and A_T representing the isotherm constant (L/g) and adsorption heat (J/mol), respectively.

Adsorption kinetics

Adsorption performance is typically evaluated using three parameters: percentage removal (Ads. %), time-dependent adsorption capacity q_t (mg/g), and equilibrium adsorption capacity q_e (mg/g) (Makrygianni et al. 2019). The correlation between them is as follows:

$$\% \text{ Ads.} = \frac{(C_0 - C_e)}{C_0} \times 100 \quad (11)$$

$$q_t = \frac{V \times (C_0 - C_t)}{m} \quad (12)$$

$$q_e = \frac{V \times (C_0 - C_e)}{m} \quad (13)$$

C_0 , C_t , C_e , m , and V denote the initial concentration, concentration at time t , equilibrium concentration, adsorbent mass, and reactor leachate volume, respectively, with units of mg/L, mg/L, mg/L, g, and L.

Adsorption kinetics are employed to identify the factors influencing the adsorption rate and to investigate equilibrium relationships. In aqueous adsorption systems, the Pseudo-first-order (PFO) (Chys et al. 2015), Pseudo-second-order (PSO) (Zhang et al. 2009), and Intraparticle Diffusion (IPD) (Aziz et al. 2011) models are widely used to describe adsorption kinetics.

Pseudo-first-order kinetics

For fast adsorption processes, Lagergren introduced the PFO kinetic model, with its linearized equation shown in Eq. (14) (Lagergren 1898).

$$\ln (q_e - q_t) = \ln q_e - k_1 t \quad (14)$$

In this kinetic model, q_e denotes the equilibrium adsorption capacity, q_t the amount adsorbed at any time, k_1 the rate constant, and t the elapsed time, with units mg/g, mg/g, 1/min, and min, respectively.

Plotting $\ln (q_e - q_t)$ against time yields a negatively sloped line, from which q_e is derived from the intercept and k_1 from the slope.

Pseudo-second-order kinetics

For time-dependent adsorption studies, the PSO kinetic model by Ho and McKay (Ho and McKay 1999) is used, and its linearized form is provided in Eq. (15).



$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (15)$$

For the PSO model, k_2 is the rate constant in g/mg min. A plot of t/q_t against time provides q_e from the slope and k_2 from the ordinate intercept.

Intraparticle diffusion model kinetics

In cases where the PFO and PSO models do not sufficiently represent adsorption kinetics, the Intraparticle Diffusion (IPD) model is used, with its linear equation provided in Eq. (16).

$$q_t = k_{id} t^{1/2} + C \quad (16)$$

In the IPD model, K denotes the diffusion rate constant, $t^{1/2}$ is the root time variable, q_t indicates adsorption at a given time, and C corresponds to a boundary layer-related constant.

In the intraparticle diffusion model, the C value reflects boundary layer thickness. The slope of a $t^{1/2}$ versus q_t plot provides the rate constant, while the ordinate intercept gives C (McKay et al. 1980).

Thermodynamic Parameters

Adsorption thermodynamics for COD, $\text{NH}_3\text{-N}$, and turbidity were investigated at temperatures ranging from 293 to 323 K, and the standard ΔG° , ΔH° , and ΔS° values were derived using Eqs. (17–19).

$$\Delta G^\circ = -RT \ln K_d \quad (17)$$

$$\ln K_d = \left(\frac{\Delta S^\circ}{R} \right) - \left(\frac{\Delta H^\circ}{R} \right) \left(\frac{1}{T} \right) \quad (18)$$

The determination of the K_d value was carried out with the help of Eq. (19).

$$K_d = \frac{F_e}{1 - F_e} \quad (19)$$

In this study, T , R , and F_e correspond to temperature (K), gas constant (kJ/mol K), and the proportion of turbidity, $\text{NH}_3\text{-N}$, and COD adsorbed at 40 min.

Results and discussion

Comparative surface and morphological characterization of CBPAC and RGO-CBPAC particles before and after adsorptive removal of pollutants via BET, FTIR, and SEM–EDS Analyses

The adsorption characteristics of CBPAC and RGO-CBPAC particles were systematically investigated using nitrogen physisorption, Brunauer–Emmett–Teller (BET) surface analysis, Fourier Transform Infrared Spectroscopy (FTIR), scanning electron microscopy (SEM), and energy-dispersive X-ray spectroscopy (EDS). Both materials exhibited a progressive increase in nitrogen adsorption with rising relative pressure, reaching a plateau at $P/P_0=0.99$ (Fig. 2 (a, b)). This behaviour suggests that adsorption mainly takes place at accessible microporous sites, with gas uptake reaching saturation at high relative pressures due to the full occupation of these active sites.

The CBPAC sample revealed a typical type I isotherm prior to adsorption, confirming micropore-dominated chemisorption, while macropores played an insignificant role because of their wide size distribution. This demonstrates that adsorption in CBPAC is essentially controlled by its microporous structure. On the other hand, RGO-CBPAC also presented a type I isotherm; however, the presence of reduced graphene oxide layers introduced a hierarchical pore arrangement encompassing both micro- and mesopores (Koyuncu and Kul 2020; Chen et al. 2021; Gong et al. 2022). The formation of mesopores in RGO-CBPAC contributes to higher adsorption potential while enabling more rapid transport of contaminants through the pore channels. Consequently, this finding confirms that RGO-mediated structural enhancements positively influence both adsorption kinetics and site accessibility. After exposure to pollutants, CBPAC exhibited a shift from a type I isotherm to a sigmoidal type II profile, indicating nearly full saturation of micropores and partial occupancy of mesopores, whereas macropores remained mostly unutilized (Fig. 2c, d) (Liu et al. 2021; Gong et al. 2022). In comparison with CBPAC, RGO-CBPAC preserved portions of its mesoporous structure after adsorption, indicating that its hierarchical architecture prevents total pore blockage and retains additional sites for continued adsorption. This highlights the contribution of RGO to sustaining long-term adsorption capacity.

BET surface area and pore volume measurements further clarified the effects of adsorption. For CBPAC, the surface area sharply declined from 887 m^2/g to 0.76 m^2/g , while the pore volume dropped from 0.494 cm^3/g to 6.5×10^{-4} cm^3/g , accompanied by an increase in mean pore diameter from 2.23 nm to 6.4 nm. These results suggest extensive micropore filling along with slight expansion due to adsorbate presence. In contrast, RGO-CBPAC did not undergo a complete reduction in surface area (from 908 m^2/g to 23 m^2/g) or pore volume (from



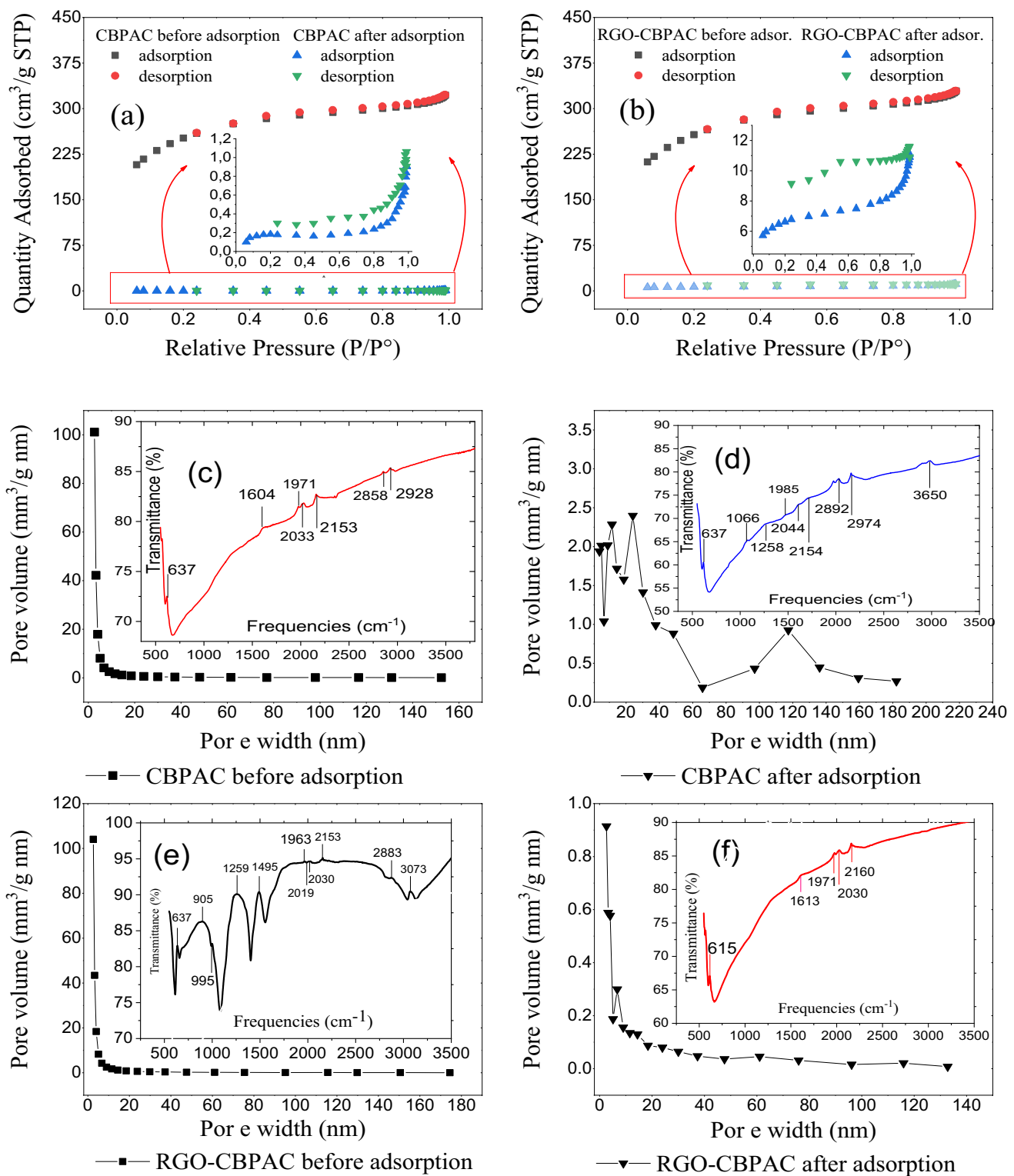


Fig. 2 Adsorption-desorption N_2 isotherms of CBPAC and RGO-CBPAC at 373 K (panels a, b), relationships between pore dimensions and volumes (c-f), and FTIR spectra of the adsorbents before and after adsorption (c-f)

0.504 cm^3/g to 0.018 cm^3/g); meanwhile, the pore width showed only a minor increase from 2.22 nm to 2.87 nm (Fig. 2c-f). This retention of mesoporous channels indicates that RGO-CBPAC

maintains accessible adsorption sites even after substantial pollutant uptake, reflecting its enhanced structural stability.



Pore size distribution analysis supported these findings. CBPAC initially exhibited pores ranging from 2.6 to 152.4 nm, with volume peaks at 2.6, 3.26, and 4.0 nm (101.1, 42.2, and 17.9 mm³/g, respectively). After adsorption, the pores expanded to 4–182.3 nm, with volumes sharply reduced to 0.0193–0.0027 mm³/g nm, reflecting nearly complete micropore occupation. RGO-CBPAC displayed initial pore sizes of 2.62–174.4 nm, with peaks at 104.1, 43.44, and 18.4 mm³/g, which post-adsorption narrowed to 2.60–132.8 nm with volumes of 0.9–0.0072 mm³/g nm. These observations indicate that while CBPAC experiences near-total pore blockage, RGO-CBPAC retains mesoporous channels, allowing sustained adsorption capacity (Fig. 2c–f).

FTIR spectroscopy revealed significant modifications in surface functional groups following adsorption, as shown in Fig. 2c–d). Before adsorption, CBPAC exhibited characteristic vibrations at 637 cm⁻¹ (CH₂ in vinyl groups), 1604 cm⁻¹ (C=N), 1971 cm⁻¹ (>C=C=C), 2033 cm⁻¹ (R₁R₂C=N=N), 2153 cm⁻¹ (>C=C=O), and 2858–2928 cm⁻¹ (–CH₃/–CH₂) (Colthup 2012). Post-adsorption, notable peak shifts (e.g., 2033 → 2044 cm⁻¹) and the appearance of a new OH peak at 3650 cm⁻¹ confirmed the adsorption of COD, NH₃-N, and turbidity-inducing compounds. As shown in Fig. 2e, the FTIR spectrum of RGO-CBPAC before adsorption exhibits key vibrations: vinyl (CH₂) at 637 cm⁻¹, azoxy C=N at 1495 cm⁻¹, nitro-amines (>N=NO₂) at 1259 cm⁻¹, allenes (C=C=C) at 1963 cm⁻¹, diazo (R₁R₂C=N=N) at 2019 cm⁻¹, ketenes (>C=C=O) at 2153 cm⁻¹, 1,1-dialkyl alkenes (R₂C=CH₂) at 3073 cm⁻¹, and –CH₃ at 2883 cm⁻¹ (Colthup 2012). RGO-CBPAC exhibited similar spectral shifts and the emergence of new peaks at 1613 and 1971 cm⁻¹, indicating active participation of RGO surface functional groups in adsorption (Fig. 2f). These results suggest that adsorption proceeds through a combination of pore filling and chemical interactions.

SEM analysis provides important insight into the morphological evolution of CBPAC before and after adsorption. Initially, CBPAC particles exhibit an aggregated and heterogeneous structure containing macro-, meso-, and micropores, indicating a well-developed porous network with high adsorption potential (Fig. 3a). Following adsorption, a significant structural change is observed: micropores become completely filled, while mesopores remain only partially occupied (Fig. 3b). This behaviour suggests that adsorption preferentially occurs in micropores due to their higher adsorption energy and stronger affinity for pollutant molecules. The complete saturation of micropores indicates that CBPAC reaches its adsorption capacity rapidly, which explains its strong initial removal performance. However, the partial occupancy of mesopores implies that these larger pores continue to act as diffusion pathways and secondary adsorption sites. This reflects an ongoing mass transfer process, where pollutants gradually migrate from the bulk solution into the internal pore structure. Consequently, while micropores

dominate the early stages of adsorption, mesopores contribute to sustained adsorption over time.

In general, these findings highlight a key limitation of CBPAC: the rapid saturation of micropores may restrict long-term adsorption efficiency. The presence of partially filled mesopores suggests that improving mesoporous structure could enhance pollutant transport and extend adsorption capacity. This also supports the rationale for material modification strategies, such as RGO incorporation, which aim to preserve pore accessibility and improve overall adsorption performance.

In this study, RGO nanoparticles were not employed as a catalyst but rather as a modifying component to enhance the adsorbent properties of CBPAC. The incorporation of RGO into the CBPAC structure induced significant changes in surface texture, pore distribution, and the availability of active adsorption sites, which directly influenced adsorption performance. Accordingly, RGO-CBPAC exhibited a layered and aggregated morphology, where RGO sheets bridged fissures and cracks. Post-adsorption observations revealed that micropores reached full saturation while mesopores remained partially open, emphasizing the advantage of hierarchical structuring for improved pollutant transport and adsorption efficiency (Fig. 4a, b).

These morphology-related modifications facilitated stronger interactions between the adsorbent surface and pollutants (COD, NH₃-N, and turbidity-related constituents), thereby enhancing removal efficiency. Furthermore, SEM and BET analyses support these findings by demonstrating clear differences in surface morphology, pore structure, and specific surface area between CBPAC and RGO-CBPAC, highlighting the direct relationship between adsorbent morphology and adsorption efficiency.

The results from EDS elemental characterization corroborated the morphological and textural findings. The elemental composition of CBPAC shifted from C 29.6 wt.%, O 42 wt.%, and Na 21.96 wt.% to O 50.68 wt.%, Cl 18.22 wt.%, Na 22.75 wt.%, Mg 6.71 wt.%, and K 1.64 wt.% after adsorption, indicating effective uptake of leachate pollutants. Similarly, RGO-CBPAC composition changed from C 93.57 wt.% and O 4.85 wt.% to O 37.67 wt.%, C 14.62 wt.%, Al 30.68 wt.%, K 9.42 wt.%, Na 5.39 wt.%, and Mg 2.22 wt.% post-adsorption, further confirming successful pollutant adsorption (Fig. 3c, d; Fig. 4c, d).

In conclusion, while CBPAC and RGO-CBPAC both achieve substantial removal of COD, NH₃-N, and turbidity, RGO-CBPAC exhibits greater resilience, maintains mesoporous channels, and sustains adsorption potential over time. CBPAC's reliance on micropores leads to rapid saturation, limiting its prolonged use. The hierarchical pore configuration and RGO incorporation in RGO-CBPAC, however, ensure continuous accessibility of active sites, improved



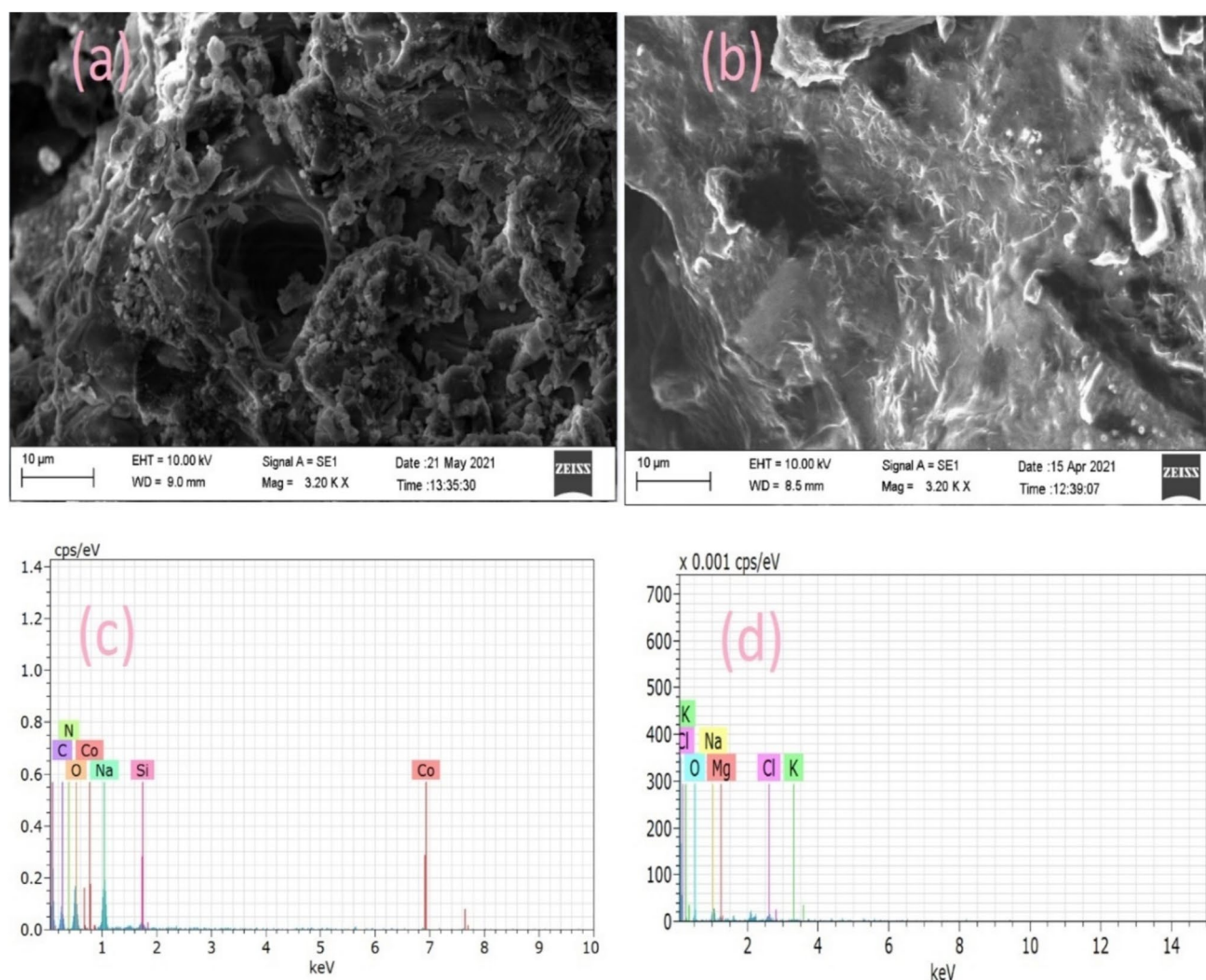


Fig.3 SEM–EDX micrographs of CBPAC particles obtained prior to adsorption (**a, c**) and following adsorption (**b, d**)

diffusion rates, and higher reusability, positioning it as a superior candidate for real-world wastewater remediation.

Comparative analysis of the effect of adsorbent dosage on COD, $\text{NH}_3\text{-N}$, and turbidity removal performance and adsorption capacity using CBPAC and RGO-CBPAC particles

To assess the impact of adsorbent dosage on leachate treatment, batch adsorption experiments were conducted using CBPAC and RGO-CBPAC at dosages ranging from 1 to 5 g/L, 20 °C, natural pH 8.35, and 300 rpm. Removal efficiencies and adsorption capacities were recorded after 40 min and were illustrated in Fig. 5. For CBPAC, increasing the adsorbent dosage resulted in a marked improvement in pollutant removal: COD removal increased from 30% at 1 g/L to 42% at 5 g/L, $\text{NH}_3\text{-N}$ from 13.5 to 22%, and

turbidity from 33 to 46% (Fig. 5). Conversely, the adsorption capacity per unit mass decreased with increasing dosage, reaching maximum values at the lowest dosage of 1 g/L, with 125.3 mg $\text{O}_2\text{/g}$ for COD, 11 mg $\text{NH}_3\text{-N/g}$, and 9.4 mg $(\text{NH}_2)_2\text{H}_2\text{SO}_4\text{/g}$. This decline is attributed to aggregation of CBPAC particles at higher dosages, which blocks active sites and reduces accessibility of adsorption surfaces (Hamdy et al. 2018).

RGO-CBPAC consistently outperformed CBPAC across all tested dosages. At 5 g/L, removal efficiencies were 52% for COD, 31% for $\text{NH}_3\text{-N}$, and 55% for turbidity, with adsorption capacities of 43.4 mg $\text{O}_2\text{/g}$, 3.6 mg $\text{NH}_3\text{-N/g}$, and 3.2 mg $(\text{NH}_2)_2\text{H}_2\text{SO}_4\text{/g}$ (Fig. 5). Maximum adsorption per unit mass was observed at 1 g/L, reaching 146.2 mg $\text{O}_2\text{/g}$ for COD, 13.9 mg $\text{NH}_3\text{-N/g}$, and 11.4 mg $(\text{NH}_2)_2\text{H}_2\text{SO}_4\text{/g}$. This improvement is due to the presence of RGO, which enhances surface area, pore volume, and functional groups, thereby



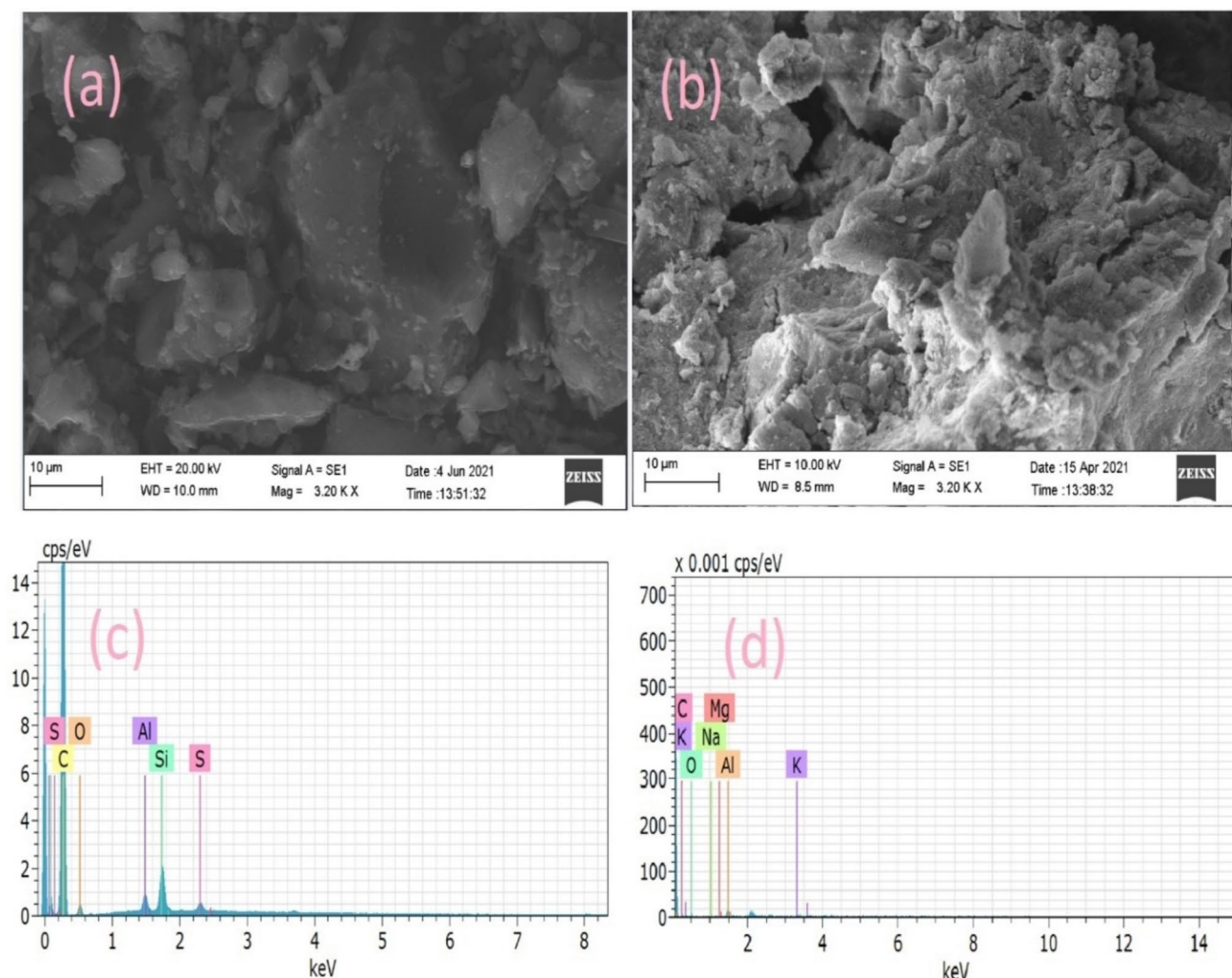


Fig.4 SEM-EDS images of RGO-CBPAC particles before adsorption (a, c) and after adsorption (b, d)

maintaining adsorption site accessibility even at higher dosages (Hamdy et al. 2018).

The comparative assessment showed that RGO-CBPAC outperformed CBPAC by approximately 10%, 9%, and 9% in COD, $\text{NH}_3\text{-N}$, and turbidity removal efficiencies, respectively, with corresponding increases in adsorption capacities of 20.9 mg O_2/g , 2.9 mg $\text{NH}_3\text{-N}/\text{g}$, and 2 mg $(\text{NH}_2)_2\text{H}_2\text{SO}_4/\text{g}$ (Fig. 5) (Gotvajn et al. 2009; Mahdavi et al. 2018; Kinidi and Salleh 2020). RGO incorporation enhances the adsorption efficiency per unit mass and ensures consistent performance over a range of dosages. While both CBPAC and RGO-CBPAC exhibit dosage-dependent adsorption behaviour, RGO-CBPAC consistently achieves superior pollutant removal, higher structural integrity, and higher adsorption capacity per unit. Maximum removal efficiencies were obtained at 5 g/L, whereas peak adsorption capacities were recorded at 1 g/L, emphasizing the significance of optimizing adsorbent dosage to achieve a balance between total removal and unit adsorption performance.

Comparative analysis of the effect of adsorption time on COD, $\text{NH}_3\text{-N}$, and turbidity removal efficiency and adsorption capacity using CBPAC and RGO-CBPAC particles

Figure 6 depicts the time-dependent behaviour of COD, $\text{NH}_3\text{-N}$, and turbidity concentrations, along with their respective removal efficiencies and adsorption capacities, for CBPAC and RGO-CBPAC. Both adsorbents showed a rapid uptake during the initial 20 min, indicating fast occupation of available active sites. Beyond this period, adsorption slowed and approached equilibrium at around 40 min. After 40 min, CBPAC achieved COD, $\text{NH}_3\text{-N}$, and turbidity concentrations of 607 mg O_2/L , 159 mg $\text{NH}_3\text{-N}/\text{L}$, and 38.5 mg $(\text{NH}_2)_2\text{H}_2\text{SO}_4/\text{L}$, corresponding to removal efficiencies of 41.9%, 22%, and 45.9%, and adsorption capacities of 34.9 mg O_2/g , 3.58 mg $\text{NH}_3\text{-N}/\text{g}$, and 2.62 mg $(\text{NH}_2)_2\text{H}_2\text{SO}_4/\text{g}$. In contrast, RGO-CBPAC demonstrated enhanced performance, with final concentrations of



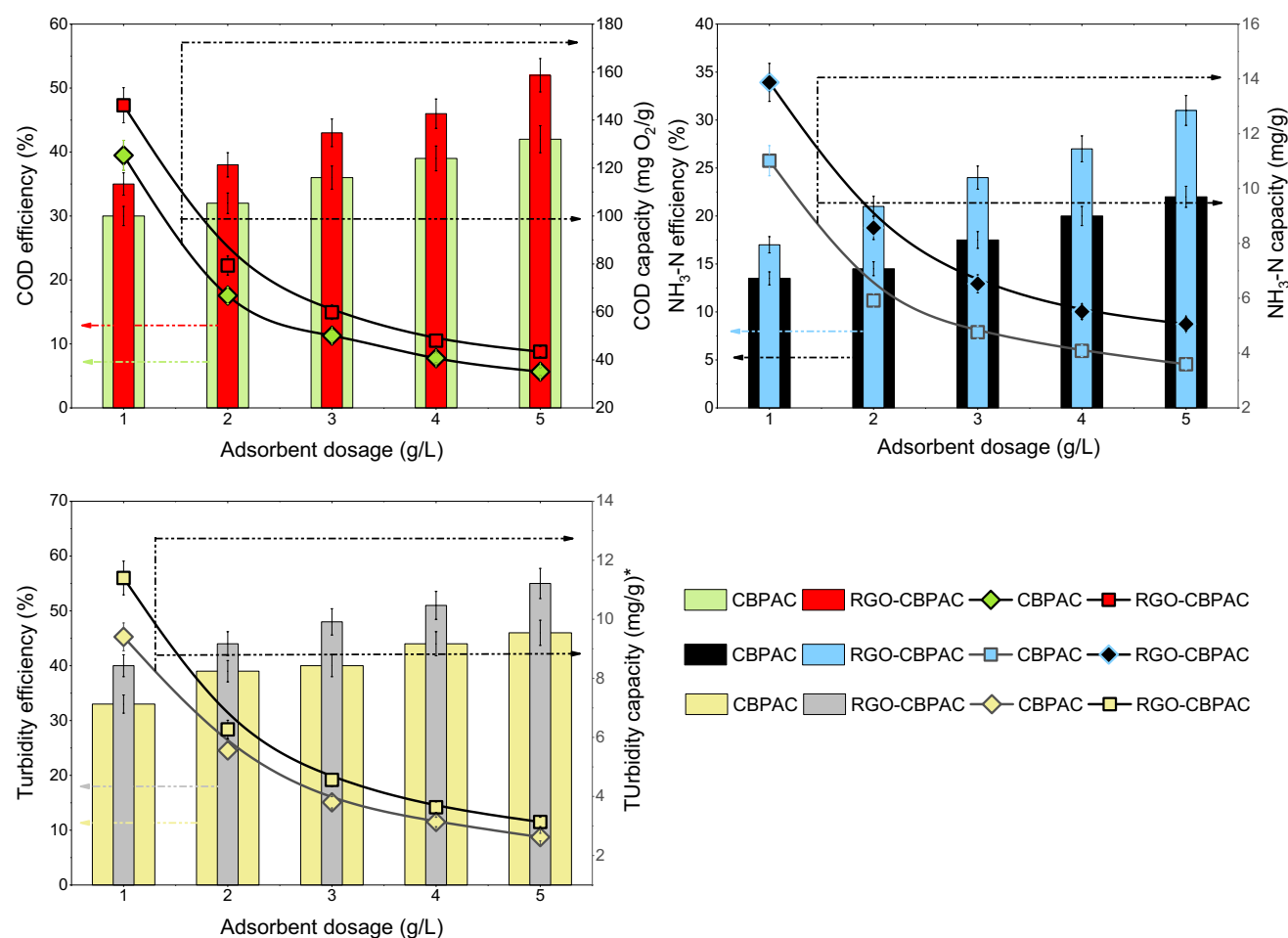


Fig. 5 Comparison of COD, NH₃-N, and turbidity removal efficiencies and adsorption capacities under varying CBPAC and RGO-CBPAC loadings

521 mg O₂/L, 142 mg NH₃-N/L, and 32.2 mg (NH₂)₂H₂SO₄/L, removal efficiencies of 50.1%, 30.3%, and 54.8%, and adsorption capacities of 41.8 mg O₂/g, 4.9 mg NH₃-N/g, and 3.2 mg (NH₂)₂H₂SO₄/g (Fig. 6). These findings were consistent with literature reports of rapid adsorption within the first 15–25 min due to abundant active sites (Bhattacharya et al. 2008; Vieira et al. 2010).

Although the contact time was increased, adsorption beyond 25 min showed only slight enhancement. This behaviour indicates that initial adsorption proceeds rapidly via available surface sites, but subsequent adsorption is hindered by pore saturation and repulsive interactions between remaining pollutants and already adsorbed molecules. Consequently, a 40-min contact duration was selected as optimal for balancing high removal efficiencies with efficient adsorbent utilization.

A comparative evaluation demonstrates that the addition of RGO significantly improves both the removal efficiencies and adsorption capacities. Compared with CBPAC, RGO-CBPAC achieved increases of ~8.2% for COD (6.9 mg O₂/g), 8.3% for NH₃-N (1.1 mg NH₃-N/g),

and 8.9% for turbidity (0.58 mg (NH₂)₂H₂SO₄/g) (Fig. 6). These improvements are associated with RGO's contribution to enhanced surface area, hierarchical pore architecture, and additional functional groups, which promote faster pollutant transport and more efficient utilization of adsorption sites (Bhattacharya et al. 2008; Vieira et al. 2010). The findings highlight adsorption time as a key parameter influencing leachate treatment performance, with RGO-CBPAC consistently exceeding CBPAC in all measured outcomes, notably during the fast-initial adsorption stage and in achieving greater equilibrium capacities.

Comparative analysis of the effect of agitation rate on COD, NH₃-N, and turbidity removal performance and adsorption capacity using CBPAC and RGO-CBPAC particles

The role of agitation rate in influencing the adsorption performance of CBPAC and RGO-CBPAC particles was systematically examined, with COD, NH₃-N, and turbidity removal efficiencies and adsorption capacities evaluated



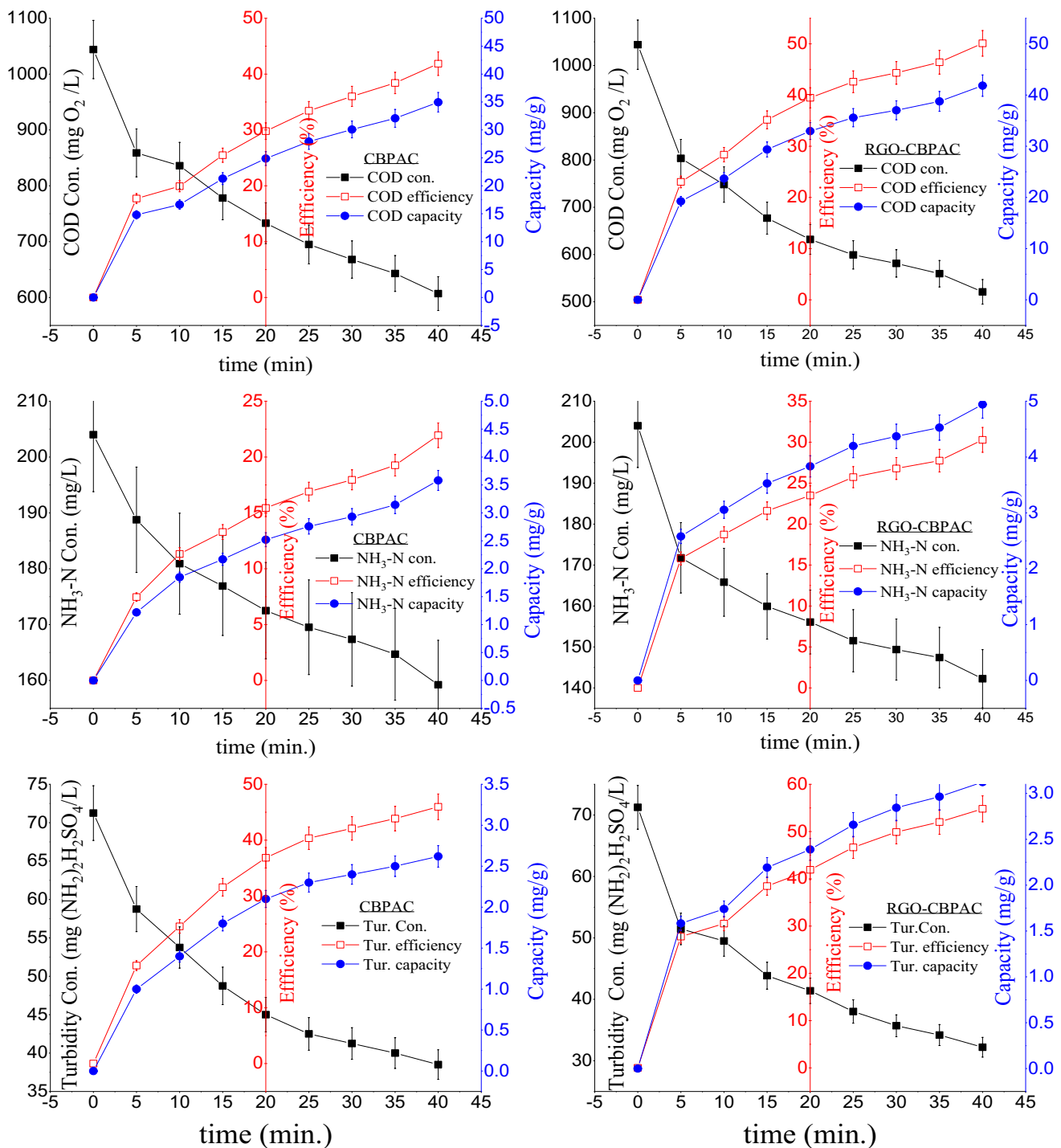


Fig. 6 Time-dependent changes in pollutant concentrations, removal efficiencies of COD, NH₃-N, and turbidity, and adsorption capacities of CBPAC and RGO-CBPAC particles

(Fig. 7). All experiments were conducted at a natural pH of 8.35, temperature of 20 °C, and adsorbent dosage of 5 g/L, while agitation rates were set at 100, 200, and 300 rpm. After 40 min, samples were filtered through a 45 µm membrane, and the corresponding adsorption metrics were recorded. For CBPAC, adsorption performance increased with rising agitation rates.

COD removal efficiency improved from 36% at 100 rpm to 42% at 300 rpm, with corresponding adsorption capacities increasing from 30.1 to 35.1 mg O₂/g. NH₃-N removal increased from 17 to 22%, with adsorption capacities rising from 2.8 to 3.6 mg NH₃-N/g, while turbidity removal enhanced from 25 to 30%, with capacities increasing from 1.43 to 1.7 mg (NH₂)₂H₂SO₄/g. In contrast, RGO-CBPAC consistently



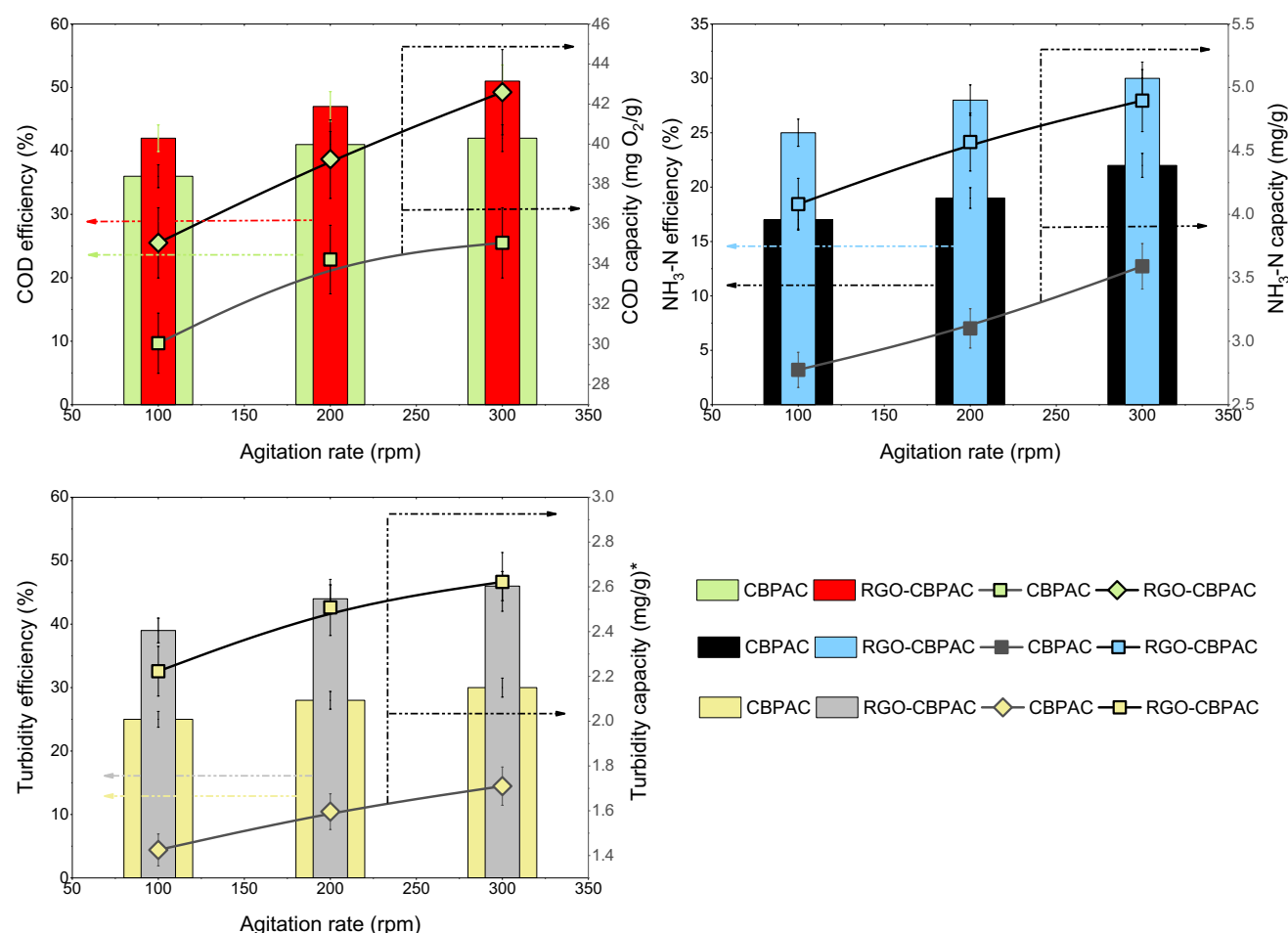


Fig. 7 Comparison of COD, NH₃-N, and turbidity removal efficiencies and adsorption capacities under varying agitation rates

outperformed CBPAC across all agitation rates, achieving COD removal efficiencies of 42%, 47%, and 51% with adsorption capacities of 35.1, 39.3, and 42.3 mg O₂/g, NH₃-N removal of 25%, 28%, and 30% with capacities of 4.1, 4.7, and 4.9 mg NH₃-N/g, and turbidity removal of 39%, 44%, and 46% with capacities of 2.2, 2.5, and 2.6 mg (NH₂)₂H₂SO₄/g at 100, 200, and 300 rpm, respectively.

The enhancement in adsorption performance with increasing agitation rate can be attributed to improved mixing and dispersion of the adsorbent particles and pollutants. Elevated stirring reduces the boundary layer thickness around the adsorbents and enhances the external mass transfer coefficient, thereby increasing the frequency of interactions between CBPAC or RGO-CBPAC and the contaminants (Bellebia et al. 2009). After 40 min at 300 rpm, RGO-CBPAC outperformed CBPAC, achieving improvements of ~9% and 7.5 mg O₂/g for COD, 8% and 1.3 mg NH₃-N/g, and 16% and 0.92 mg (NH₂)₂H₂SO₄/g for turbidity (Fig. 7). These results highlight the structural benefits of RGO-CBPAC, such as larger surface area, additional functional groups, and preserved micropore accessibility, which together enable more efficient pollutant uptake under higher agitation. In conclusion, both adsorbents display

agitation-dependent improvements, with RGO-CBPAC showing better performance due to its hierarchical pore architecture and functionalized surface.

Comparative analysis of the effect of initial solution temperature on COD, NH₃-N, and turbidity removal performance and adsorption capacity using CBPAC and RGO-CBPAC particles

The role of initial solution temperature on the adsorption performance of COD, NH₃-N, and turbidity and their corresponding adsorption capacities was systematically examined using CBPAC and RGO-CBPAC in batch experiments. All experiments were performed at natural pH 8.35, 5 g/L adsorbent dosage, and 300 rpm stirring. Five initial temperatures (20–60 °C) were tested. Following a 40-min contact period, the samples were filtered through a 45 µm membrane, and the removal efficiencies and adsorption capacities were determined (Fig. 8).

The results indicated that both removal efficiencies and adsorption capacities for COD, NH₃-N, and turbidity increased progressively with rising temperature. For CBPAC, COD removal improved from 42% at 20 °C to 51% at 60 °C,



corresponding to an increase in adsorption capacity from 35.5 to 42.6 mg O₂/g. Similarly, NH₃-N removal rose from 22 to 30%, with adsorption capacities increasing from 3.6 to 4.9 mg NH₃-N/g, while turbidity removal increased from 46 to 57%, with adsorption capacities increasing from 2.6 to 3.3 mg (NH₂)₂H₂SO₄/g. RGO-CBPAC exhibited even greater enhancements under the same conditions, with COD removal increasing from 52 to 60% (43.4–50.1 mg O₂/g), NH₃-N removal from 31 to 39% (5.1–6.4 mg NH₃-N/g), and turbidity removal from 55 to 66% (3.1–3.8 mg (NH₂)₂H₂SO₄/g).

The increase in adsorption performance observed at higher temperatures suggests that the adsorption process is endothermic in nature (Zhang et al. 2016). This effect is explained by multiple mechanisms: higher temperatures increase the kinetic energy of COD and turbidity molecules, enhancing their diffusion into CBPAC and RGO-CBPAC pores (Kaur et al. 2016); they also reduce the thickness of the boundary layer around the adsorbents, improving mass transfer rates. In addition, the solubility of NH₃ decreases at elevated temperatures, favouring its release from the aqueous phase and potentially enhancing its interaction with the adsorbent surface. Consequently, both removal efficiencies and adsorption capacities improved as

temperature increased. After 40 min, a comparison between RGO-CBPAC and CBPAC indicated significant performance gains: COD removal increased by 9% with adsorption capacities rising by 7.52 mg O₂/g, NH₃-N by 9% and 1.46 mg NH₃-N/g, and turbidity by 11% and 0.52 mg (NH₂)₂H₂SO₄/g, as presented in Fig. 8. These results demonstrate the better adsorption efficiency of RGO-CBPAC, highlighting the synergistic effect of RGO incorporation in improving both surface area and active site availability under elevated temperatures.

Comparative analysis of the effect of initial pH value on COD, NH₃-N, and turbidity removal performance and adsorption capacity using CBPAC and RGO-CBPAC particles

Initial leachate pH was identified as a key factor influencing the adsorption efficiency of CBPAC and RGO-CBPAC particles for COD, NH₃-N, and turbidity, as well as the related adsorption capacities. Both adsorbents exhibit surface charge properties that are highly sensitive to pH changes, directly affecting electrostatic interactions with pollutants (Renou et al. 2008; Wu et al. 2012). Batch adsorption

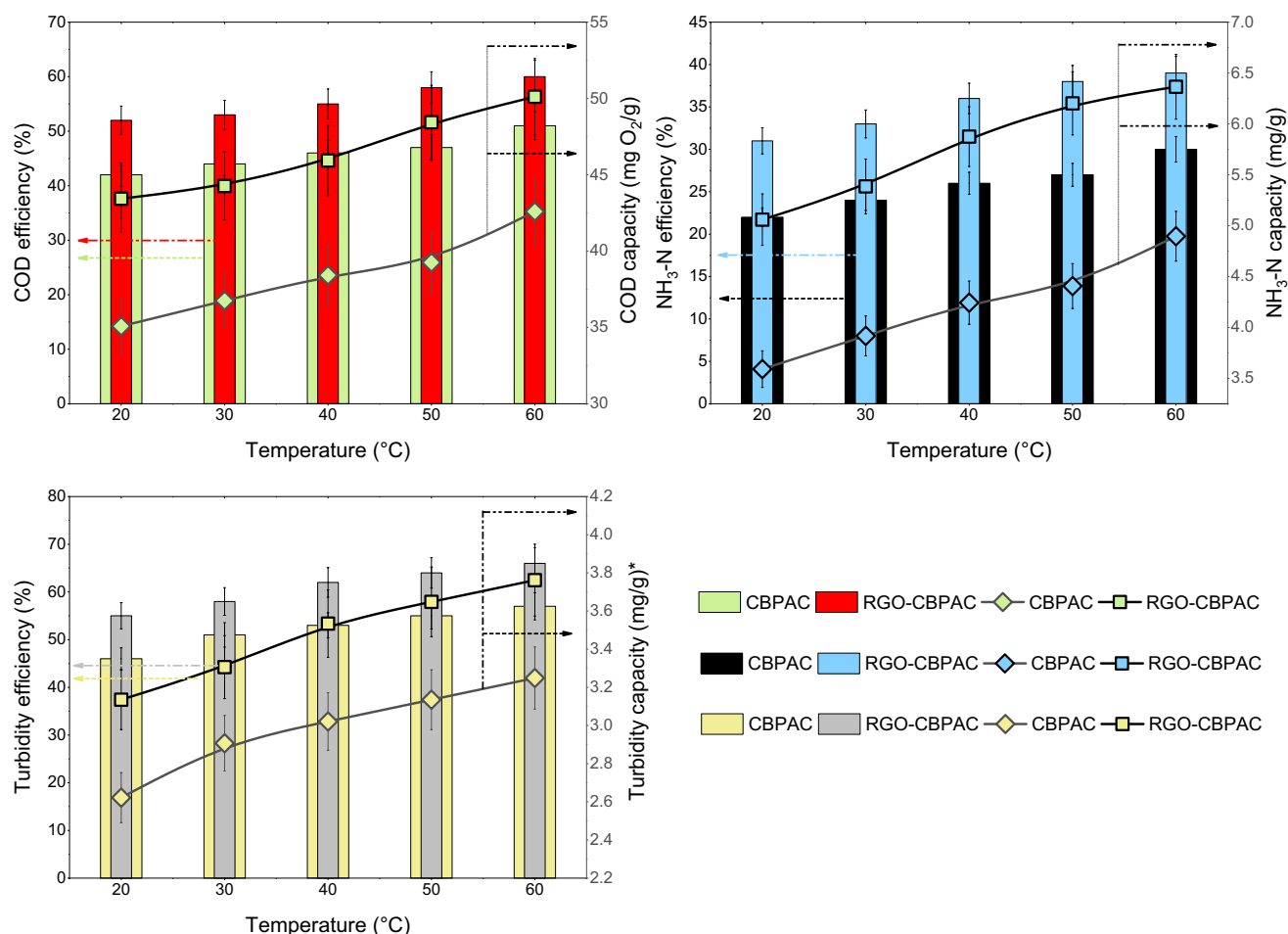


Fig. 8 Comparison of COD, NH₃-N, and turbidity removal efficiencies and adsorption capacities under varying initial temperatures



experiments were conducted at 20 °C, 300 rpm stirring, and an adsorbent dosage of 5 g/L, with initial pH value of 5, 7, 8.35 (natural), and 9.5. Removal efficiencies and adsorption capacities under these conditions were shown in Fig. 9. At pH 5 and after 40 min, CBPAC exhibited COD, $\text{NH}_3\text{-N}$, and turbidity removal efficiencies of 48%, 27%, and 52%, with corresponding adsorption capacities of 40.1 mg O_2/g , 4.4 mg $\text{NH}_3\text{-N}/\text{g}$, and 2.9 mg $(\text{NH}_2)_2\text{H}_2\text{SO}_4/\text{g}$. In contrast, RGO-CBPAC showed better removal efficiencies and capacities under the same conditions: 57%, 42%, and 63%, and 47.6 mg O_2/g , 6.9 mg $\text{NH}_3\text{-N}/\text{g}$, and 3.6 mg $(\text{NH}_2)_2\text{H}_2\text{SO}_4/\text{g}$. Performance decreased progressively with increasing pH, reaching minimum values at pH 9.5, underscoring the pH sensitivity of both adsorbents.

At acidic pH, adsorption is enhanced due to neutralization or positive charging of CBPAC and RGO-CBPAC surfaces in response to elevated H^+ levels, which strengthens electrostatic interactions with negatively charged COD and turbidity components. Under alkaline conditions, increased OH^- concentrations intensify electrostatic repulsion, reducing removal efficiencies and capacities. NH_3 adsorption was promoted by low pH via NH_4^+ formation, which promoted

electrostatic and ion-exchange interactions, whereas at higher pH, non-ionic NH_3 dominated, and adsorption occurred mainly through weaker hydrogen bonding or Lewis acid–base interactions, resulting in decreased adsorption.

After 40 min of adsorption at pH 5, RGO-CBPAC consistently outperformed CBPAC, with improvements in COD, $\text{NH}_3\text{-N}$, and turbidity removal efficiencies of 9%, 15%, and 11%, and corresponding increases in adsorption capacities of 7.5 mg O_2/g , 2.5 mg $\text{NH}_3\text{-N}/\text{g}$, and 0.63 mg $(\text{NH}_2)_2\text{H}_2\text{SO}_4/\text{g}$, respectively, as shown in Fig. 9. This performance was attributed to the higher surface area, enhanced porosity, and additional functional groups provided by RGO incorporation, which facilitated more effective pollutant interactions and faster adsorption kinetics (Seredych et al. 2009).

In general, the initial pH was a critical factor influencing adsorption performance, with acidic conditions (pH 5) promoting maximum removal of COD, $\text{NH}_3\text{-N}$, and turbidity. Alkaline conditions progressively reduced adsorption efficiency due to electrostatic repulsion and limited pollutant binding. Across all tested pH values, RGO-CBPAC consistently exhibited better performance than CBPAC,

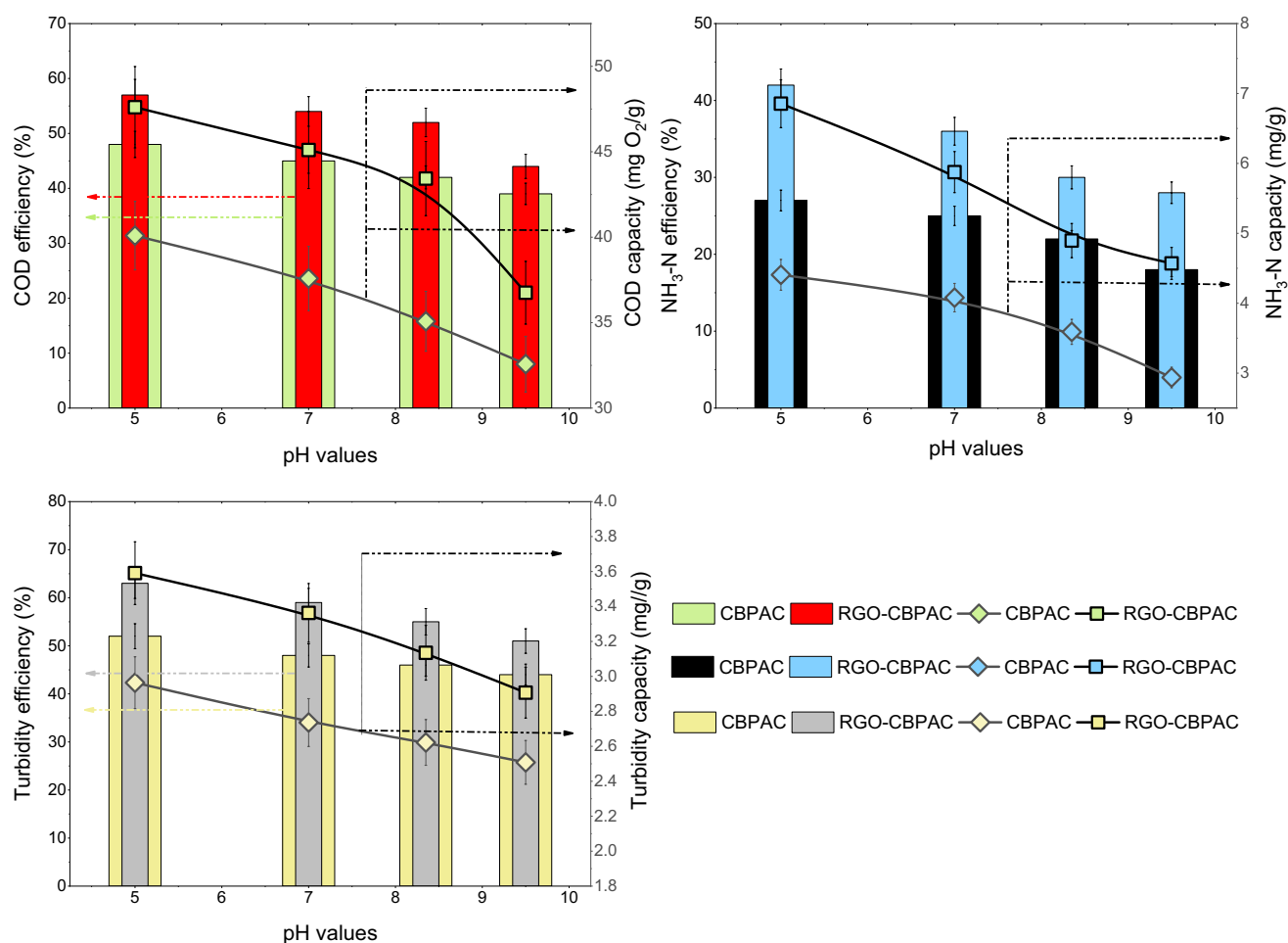


Fig. 9 Comparison of COD, $\text{NH}_3\text{-N}$, and turbidity removal efficiencies and adsorption capacities under varying initial pH values



highlighting the beneficial role of RGO incorporation in advanced leachate treatment.

To investigate the pH-dependent behaviour of CBPAC and RGO-CBPAC adsorbents, as well as their removal performance for COD, $\text{NH}_3\text{-N}$, and turbidity from the diluted leachate at different pH values, zeta potential measurements were conducted at pH 5, 7, 8.35, and 9.5 before and after adsorption. The zeta potentials (ζ) of CBPAC and CBPAC-RGO exhibited strong pH dependence and were significantly altered after adsorption of COD, $\text{NH}_3\text{-N}$, and turbidity-related constituents. Before adsorption, CBPAC showed amphoteric surface behaviour with ζ values of +10 mV at pH 5, 0 mV at pH 7, -18 mV at pH 8.35, and -32 mV at pH 9.5, indicating a transition from a positively charged surface under acidic conditions to a negatively charged surface under alkaline conditions. In contrast, RGO-CBPAC exhibited predominantly negative surface charge across the entire pH range even before adsorption, with ζ values of -2 mV, -16 mV, -30 mV, and -45 mV at pH 5, 7, 8.35, and 9.5, respectively, reflecting the influence of oxygen-containing functional groups introduced by reduced graphene oxide. After adsorption, both materials showed a pronounced shift toward more negative zeta potential values at all pH levels due to the accumulation of negatively charged organic species and colloidal matter on the adsorbent surface. Specifically, CBPAC changed to -5 mV at pH 5, -15 mV at pH 7, -28 mV at pH 8.35, and -45 mV at pH 9.5, while RGO-CBPAC further decreased to -12 mV, -27 mV, -40 mV, and -58 mV at the same pH values. This overall trend indicates that adsorption of COD, $\text{NH}_3\text{-N}$, and turbidity constituents enhances surface electronegativity by introducing additional oxygenated functional groups and forming an organic coating layer, with the RGO-modified composite consistently exhibiting stronger negative surface charge due to synergistic interactions between graphene-based domains and adsorbed species. These results highlight that acidic conditions promote adsorption of dissolved

organic matter and negatively charged species, thereby improving removal efficiencies and adsorption capacities.

In addition to the parametric analysis, visual observations were carried out to assess the changes in adsorbent surface characteristics after the adsorption processes. These observations were presented in Fig. 10. In Fig. 10, when panels (a–d) are evaluated in terms of colour, variations in the tonal characteristics of the adsorbents associated with the adsorption process can be discerned upon close examination. In panel (a), CBPAC prior to adsorption exhibits a uniform dark grey to nearly black coloration, consistent with the typical appearance of conventional activated carbon. Following adsorption, as shown in panel (b), the material becomes slightly darker and less uniform, with localized regions of deeper black tones, indicating the accumulation of contaminants on the surface. In panel (c), RGO-CBPAC before adsorption presents an anthracite-like dark grey colour, appearing somewhat deeper and cooler in tone compared to CBPAC, with a more refined visual character despite reduced surface gloss. After adsorption, panel (d) reveals a further shift toward very dark grey, approaching black, accompanied by increased tonal heterogeneity, including occasional extremely dark regions and subtle brownish undertones, which suggest a greater extent of pollutant deposition.

In general, both adsorbents exhibit a degree of darkening after adsorption; however, the change is more pronounced for RGO-CBPAC, reaching deeper grey–black intensities, which is consistent with its relatively higher adsorption performance, although it does not indicate an extreme contrast between the two materials.

Proposed mechanisms for the removal of COD, $\text{NH}_3\text{-N}$, and Turbidity

Figure 11a, b illustrates the removal mechanisms of COD, $\text{NH}_3\text{-N}$, and turbidity from the diluted leachate using CBPAC and RGO-CBPAC adsorbents. For CBPAC (Fig. 11a), the

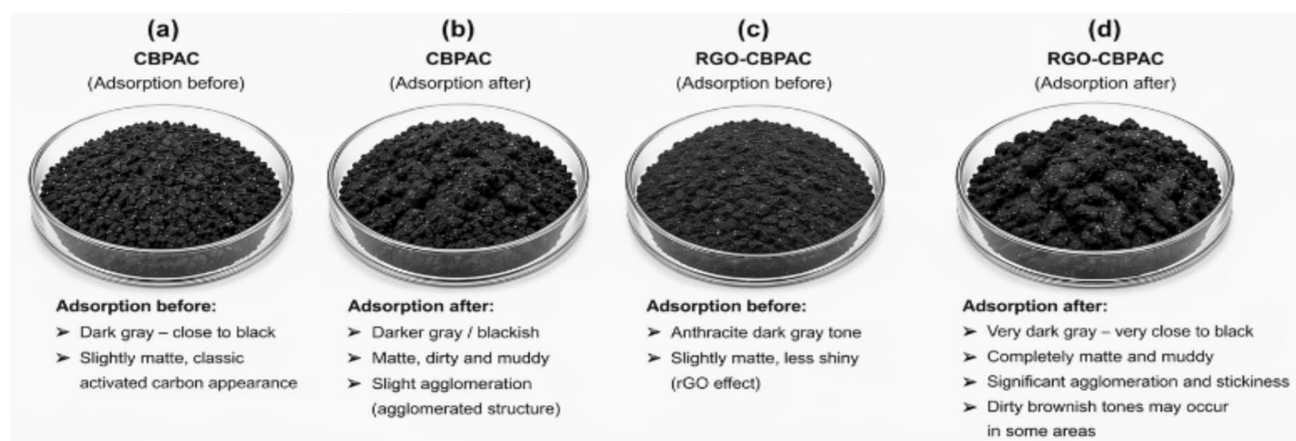


Fig. 10 Visual representation of the adsorption performance of CBPAC and RGO-CBPAC: Petri dish images of the adsorbents before and after adsorption (a–d)



removal process is governed by multiple adsorption mechanisms. Initially, contaminants were transported from the bulk solution to the adsorbent surface, followed by pore diffusion into the micro- and mesoporous structure of CBPAC. Organic compounds contributing to COD were primarily adsorbed through π - π interactions between aromatic structures and the graphitic domains of the carbon surface. In addition, electrostatic attraction played a significant role in the removal of NH_3 -N species (mainly NH_4^+), depending on the surface charge of the adsorbent. Turbidity removal by CBPAC occurred predominantly via physical adsorption and pore entrapment, where suspended and colloidal particles were captured within the porous matrix. Additionally, weak van der Waals forces and surface deposition contributed to the attachment of fine particles onto the carbon surface. However, due to the absence of strong surface functional groups, aggregation of colloids was limited, and turbidity removal mainly depends on adsorption and filtration-like mechanisms.

For RGO-CBPAC, the adsorption performance was enhanced due to the synergistic combination of CBPAC porosity and the functionalized surface of reduced graphene oxide (RGO). The removal mechanism involved dual active surfaces, where adsorption occurred both on the RGO sheets and within the CBPAC pores. The presence of RGO strengthens π - π interactions due to its extended sp^2 carbon network, leading to improved adsorption of aromatic organic compounds. Furthermore, electrostatic interactions were enhanced as a result of improved surface charge distribution. The oxygen-containing functional groups ($-\text{OH}$, $-\text{COOH}$) on RGO contribute to surface complexation, facilitating the binding of NH_4^+ and polar organic molecules. In addition, pore diffusion continued to play a role within the CBPAC matrix. Turbidity removal in RGO-CBPAC is markedly improved due to a combined mechanism of adsorption, charge neutralization, and bridging/flocculation. The RGO sheets acted as a platform that promoted the aggregation of colloidal particles through inter-particle bridging, forming larger flocs that were more easily retained within the porous structure. Moreover, surface functional groups enhance particle destabilization, resulting in more effective turbidity removal compared to CBPAC alone.

In general, the mechanisms presented in Fig. 11a, b) demonstrated that while CBPAC relied primarily on pore diffusion and physical adsorption for turbidity removal,

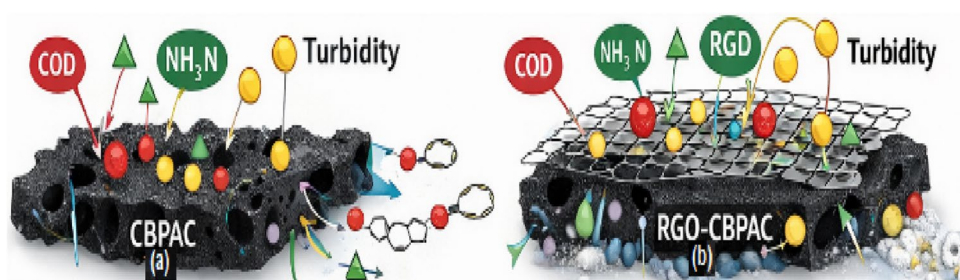
RGO-CBPAC achieved better performance through additional aggregation and bridging effects, leading to enhanced removal of suspended and colloidal matter.

Performance limitations and process integration of CBPAC and RGO-CBPAC for leachate treatment

The moderate removal efficiencies observed for COD (48–60%), NH_3 -N (22–42%), and turbidity (46–66%) are consistent with the highly heterogeneous and refractory nature of landfill leachate as well as the intrinsic limitations of adsorption-based treatment processes. Although CBPAC and RGO-CBPAC exhibit improved surface properties, including enhanced surface area, increased porosity, and additional oxygen-containing functional groups (particularly in the RGO-modified structure), the complex composition of leachate significantly constrains overall treatment performance.

In the case of COD removal, landfill leachate contains a large fraction of high-molecular-weight and structurally complex organic compounds, such as humic- and fulvic-like substances, which exhibit strong steric hindrance and low diffusivity within microporous adsorbent structures. These macromolecular species are not easily accessible to internal adsorption sites, resulting in partial rather than complete removal. In addition, pore blockage and progressive surface saturation during the adsorption process may further reduce the availability of active sites, especially at higher organic loading conditions. For NH_3 -N, the relatively low removal efficiency can be attributed to the dominant presence of NH_4^+ in hydrated ionic form, which exhibits limited affinity toward carbon-based surfaces under near-neutral to alkaline conditions. Even though RGO incorporation introduces additional functional groups and improves electron density distribution on the adsorbent surface, the interaction between NH_4^+ ions and the predominantly π -electron-rich carbon matrix remains weak. Turbidity removal is primarily governed by the entrapment of suspended and colloidal particles. While CBPAC and RGO-CBPAC effectively remove a significant fraction of larger colloids, a portion of fine particles and dissolved organic-inorganic complexes may remain in the effluent due to size exclusion effects and electrostatic repulsion between negatively charged surfaces and colloidal species. In addition, competitive adsorption between dissolved organic

Fig. 11 Mechanisms of COD, NH_3 -N, and turbidity removal using CBPAC (a) and RGO-CBPAC (b)



matter and particulate matter may reduce the overall efficiency of turbidity reduction.

Despite these limitations, the obtained results indicate that both CBPAC and RGO-CBPAC are effective adsorbent materials for partial pollutant removal and, more importantly, for reducing pollutant loads in landfill leachate. Therefore, these materials should not be considered as standalone treatment technologies but rather as key components of an integrated treatment train. In practical applications, the adsorption step can be effectively implemented as a polishing stage following biological treatment to remove residual refractory organics and suspended solids, or as a pre-treatment step to reduce organic loading prior to advanced oxidation processes (AOPs) or membrane-based filtration systems.

Such hybrid integration is particularly important for landfill leachate treatment, where single-unit processes rarely achieve compliance with strict discharge standards. The synergistic coupling of biological degradation, adsorption (CBPAC/RGO-CBPAC), and advanced oxidation processes is expected to significantly enhance overall treatment efficiency by targeting different pollutant fractions through complementary mechanisms. Consequently, the present study demonstrates that while adsorption alone provides moderate removal performance, its strategic integration within a multi-stage treatment system can ensure more robust and environmentally compliant leachate management.

Kinetics of adsorption, isotherm models, and thermodynamic components of CBPAC and RGO-CBPAC particles

The kinetic data for the removal of COD, $\text{NH}_3\text{-N}$, and turbidity using CBPAC and RGO-CBPAC were comprehensively analysed using pseudo-first-order (PFO), pseudo-second-order (PSO), and intraparticle diffusion (IPD) models to elucidate the controlling adsorption mechanisms and rate-limiting steps, and the corresponding model plots were illustrated in Fig. 12. The PFO model generally describes physical adsorption processes governed by weak van der Waals interactions, whereas the PSO model assumes chemisorption involving electron sharing or exchange between adsorbent and adsorbate. In contrast, the IPD model is used to investigate whether pore diffusion within the adsorbent structure controls the overall adsorption rate. A key parameter in the IPD model is the intercept constant (C), which reflects the thickness of the boundary layer; a value of C close to zero indicates that intraparticle diffusion is the sole rate-controlling step, while increasing values of C suggest a greater contribution of external film diffusion and surface adsorption resistance.

For CBPAC, COD removal was best described by the IPD model ($R^2=0.99$, $C=3.97$), indicating that pore diffusion is a dominant mechanism; however, the relatively high C value

clearly reveals that boundary layer effects and external mass transfer limitations also play a significant role, meaning that adsorption does not occur exclusively within the internal pores but is strongly influenced by surface phenomena at the solid–liquid interface. In comparison, $\text{NH}_3\text{-N}$ ($R^2=0.98$, $C=0.1$) and turbidity removal ($R^2=0.98$, $C=0.016$) exhibited very low C values, demonstrating that intraparticle diffusion is highly dominant in these systems, with minimal resistance from the boundary layer. Nevertheless, the similarly high R^2 values obtained from both PFO and PSO models suggest that the overall process cannot be attributed to a single mechanism and instead involves a combination of physical adsorption and chemisorption occurring simultaneously during different stages of the process.

For the RGO-CBPAC system, COD removal showed excellent fitting with both PFO and IPD models ($R^2=0.99$), while the PSO model also provided a relatively good fit ($R^2=0.98$), indicating a complex multi-mechanistic behaviour. Notably, the IPD model yielded a very high C value (14.41), which strongly indicates the presence of significant boundary layer resistance and enhanced surface adsorption effects. This can be attributed to the incorporation of RGO, which increases the number of active adsorption sites, improves surface heterogeneity, and enhances external mass transfer interactions, thereby shifting the mechanism toward stronger surface-controlled adsorption rather than purely pore-controlled diffusion. For $\text{NH}_3\text{-N}$ removal using RGO-CBPAC ($R^2=0.98$, $C=2.94$), the adsorption process appears to be governed by a coupled mechanism involving both chemisorption and intraparticle diffusion, reflecting moderate boundary layer influence. In the case of turbidity removal ($R^2=0.98$, $C=0.078$), the low C value indicates that intraparticle diffusion remains the primary rate-controlling step, although surface adsorption still contributes to the overall removal process. In general, the consistent observation of $C \neq 0$ across all systems confirms that intraparticle diffusion is not the sole rate-limiting step. Instead, adsorption proceeds through a multi-stage mechanism involving (i) external film diffusion from the bulk solution to the adsorbent surface, (ii) surface adsorption onto active sites, and (iii) diffusion into internal pores. Furthermore, the comparison between CBPAC and RGO-CBPAC clearly demonstrates that RGO modification enhances adsorption performance by increasing adsorption capacity (higher q_e values) and intensifying surface interactions, particularly for COD removal, while also altering the balance between diffusion-controlled and surface-controlled mechanisms depending on the target pollutant. The corresponding adsorption rate constants for CBPAC and RGO-CBPAC are summarized in Table 3.

Adsorption isotherms depict the distribution of solutes between the liquid phase and the adsorbent at equilibrium, giving insights into both the adsorption capacity and the heterogeneity of the adsorbent's surface. The equilibrium adsorption behaviour was further investigated using well-established isotherm models, including Freundlich,



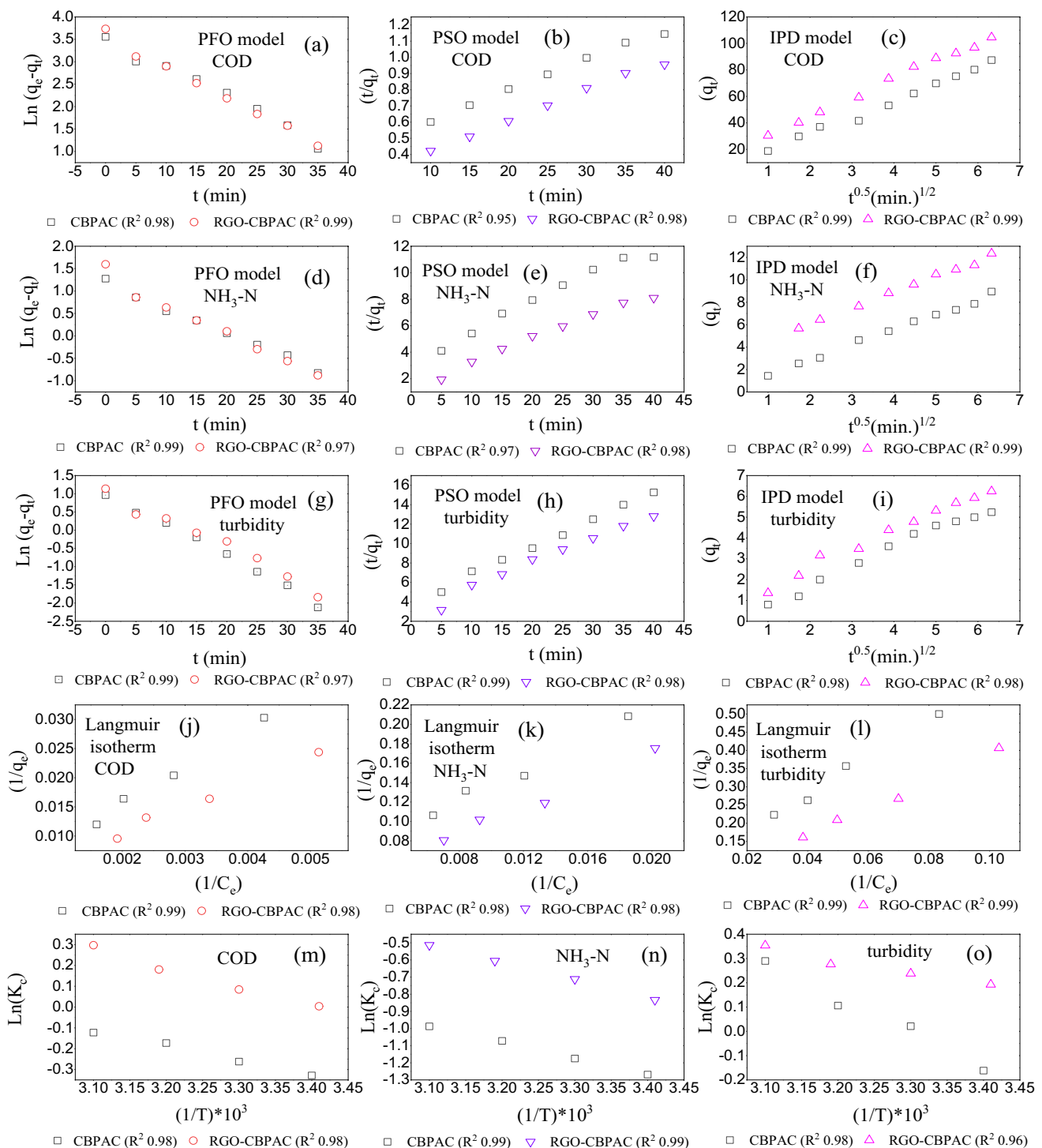


Fig. 12 Adsorption kinetics, Langmuir isotherms, and $\ln(K_c)$ plots versus $(1/T) \times 10^3$ values of turbidity, $\text{NH}_3\text{-N}$, and COD removal from the diluted leachate using CBPAC and RGO-CBPAC

Langmuir, Dubinin–Radushkevich, and Temkin models, with their corresponding R^2 values and isotherm constants summarized in Table 4.

Both CBPAC and RGO-CBPAC showed the highest correlation with the Langmuir model for COD, $\text{NH}_3\text{-N}$, and turbidity removal, with determination coefficients of $R^2 = 0.99$

and 0.99 for COD, $R^2 = 0.98$ and 0.98 for $\text{NH}_3\text{-N}$, and $R^2 = 0.99$ and 0.98 for turbidity, respectively (Fig. 12j–l). The excellent fit of the Langmuir model suggests monolayer adsorption onto homogeneous active sites, indicating that both adsorbents possess a finite number of uniform binding sites with minimal lateral interactions. In contrast,



Freundlich, Temkin, and D–R models exhibited lower R^2 values, implying that heterogeneous adsorption and adsorbate–adsorbate interactions were less significant under the tested experimental conditions. Comparative evaluation of adsorption capacities across studies is inherently limited by differing operational conditions. Despite this, literature data were compiled to contextualize the COD and $\text{NH}_3\text{--N}$ removal performance of CBPAC and RGO-CBPAC. As presented in Table 5, both adsorbents exhibit higher Langmuir adsorption capacities for COD and $\text{NH}_3\text{--N}$ than most activated carbons previously reported in the literature. The maximum monolayer capacities further indicate that RGO-CBPAC has a relatively higher adsorption potential than CBPAC and other activated carbons for all target pollutants. This improvement underscores the effectiveness of RGO incorporation in enhancing surface area, increasing the density of functional groups, and improving overall adsorption performance, highlighting the advantageous physicochemical properties of RGO-CBPAC for leachate treatment. Only a limited number of studies have explored the use of activated carbon for turbidity removal from landfill leachate, as summarised in Table 5. For example, a study utilising a COLUFNAS/AC composite adsorbent derived from *Luffa cylindrica* (sponge gourd), coconut shell, pineapple leaf, and coconut

shell-based activated carbon reported a turbidity removal efficiency of 45.90% at laboratory scale (AbdulKadir et al. 2023). Similarly, Almeida et al. (2017) achieved approximately 36% turbidity removal from intermediate landfill leachate using 1% activated carbon (AC1%) (de Almeida et al. 2017). In contrast, as presented in Table 5, the present study achieved higher turbidity removal efficiencies of 52% and 63% using CBPAC and RGO-CBPAC, respectively.

Thermodynamic parameters of adsorption, namely standard enthalpy (ΔH°), entropy (ΔS°), and Gibbs free energy (ΔG°), were determined by conducting batch adsorption experiments at initial temperatures of 20, 30, 40, 50, and 60 °C. Equilibrium constants (K_c) were calculated for each temperature, and linearized Van't Hoff plots ($\ln K_c$ versus $1/T$) were used to derive the thermodynamic quantities (Fig. 12m–o).

The values of thermodynamic parameters associated with turbidity, $\text{NH}_3\text{--N}$, and COD removal were provided in Table 6. The thermodynamic parameters (ΔH° , ΔS° , and ΔG°) for COD, $\text{NH}_3\text{--N}$, and turbidity removal using CBPAC and RGO-CBPAC were evaluated over the temperature range of 293–323 K to elucidate the feasibility and nature of the adsorption processes.

Table 3 Reaction rate constants and determination coefficients for the turbidity, $\text{NH}_3\text{--N}$, and COD adsorption kinetics using CBPAC and RGO-CBPAC

COD (CBPAC)								
PFO			PSO			IPD		
k_1 (1/min)	q_e (mg/g)	R^2	k_2 (g/mg min)	q_e (mg/g)	R^2	k_{id} (mg/g min ^{1/2})	C	R^2
0.063	78.9	0.98	1.1×10^{-3}	99	0.95	13.1	3.97	0.99
$\text{NH}_3\text{--N}$ (CBPAC)								
k_1 (1/min)	q_e (mg/g)	R^2	k_2 (g/mg min)	q_e (mg/g)	R^2	k_{id} (mg/g min ^{1/2})	C	R^2
0.055	8.02	0.99	9×10^{-3}	10.32	0.99	1.36	0.1	0.98
Turbidity (CBPAC)								
k_1 (1/min)	q_e (mg/g)	R^2	k_2 (g/mg min)	q_e (mg/g)	R^2	k_{id} (mg/g min ^{1/2})	C	R^2
0.084	5.25	0.99	0.013	6.65	0.99	0.878	0.016	0.98
COD (RGO-CBPAC)								
PFO			PSO			IPD		
k_1 (1/min)	q_e (mg/g)	R^2	k_2 (g/mg min)	q_e (mg/g)	R^2	k_{id} (mg/g min ^{1/2})	C	R^2
0.073	89.2	0.99	7.9×10^{-4}	123.5	0.98	13.9	14.41	0.99
$\text{NH}_3\text{--N}$ (RGO-CBPAC)								
k_1 (1/min)	q_e (mg/g)	R^2	k_2 (g/mg min)	q_e (mg/g)	R^2	k_{id} (mg/g min ^{1/2})	C	R^2
0.064	9.32	0.97	6.8×10^{-3}	14.74	0.98	1.45	2.94	0.98
Turbidity (RGO-CBPAC)								
k_1 (1/min)	q_e (mg/g)	R^2	k_2 (g/mg min)	q_e (mg/g)	R^2	k_{id} (mg/g min ^{1/2})	C	R^2
0.0815	5.54	0.97	0.015	7.43	0.98	0.9	0.078	0.98



Table 4 Isotherm model constants for evaluating COD, NH₃-N, and turbidity adsorption on CBPAC and RGO-CBPAC

Isotherm models	CBPAC			RGO-CBPAC		
	COD	NH ₃ -N	Turbidity	COD	NH ₃ -N	Turbidity*
<i>Langmuir</i>						
Q _m (mg/g)	500	17.5	18.1*	588	36.5	58.47
K _L (L/mg)	3 × 10 ⁻⁴	6.9 × 10 ⁻³	1.6 × 10 ⁻²	3.8 × 10 ⁻⁴	3.7 × 10 ⁻³	4.5 × 10 ⁻³
R ²	0.99	0.98	0.99	0.99	0.98	0.99
<i>Freundlich</i>						
K _f (L/mg)	0.23	0.45	0.28	0.344	0.34	0.312
n	1.1	1.66	1.27	1.104	1.36	1.09
R ²	0.97	0.97	0.98	0.97	0.97	0.98
<i>D-R</i>						
Q _m (mg/g)	82.68	9.4	4.6	102.5	12.91	6.48
β	9 × 10 ⁻³	3 × 10 ⁻⁴	2 × 10 ⁻⁵	6 × 10 ⁻³	4 × 10 ⁻⁴	2 × 10 ⁻⁵
E (kJ/mol)	7.45	40.82	158.2	9.12	35.35	158.2
R ²	0.91	0.95	0.93	0.9	0.95	0.93
<i>Temkin</i>						
K _T (L/mg)	48.56	4.07	2.412	60.3	6.32	3.67
B ₁ (kJ/kmol)	8 × 10 ⁻⁴	6 × 10 ⁻²	0.184	9.6 × 10 ⁻³	5 × 10 ⁻²	0.19
R ²	0.95	0.97	0.98	0.94	0.97	0.97

* (NH₂)₂H₂SO₄

For COD removal, both adsorbents exhibited positive enthalpy changes ($\Delta H^\circ = +5.56$ kJ/mol for CBPAC and $+7.65$ kJ/mol for RGO-CBPAC), indicating that the process was endothermic in nature. The positive entropy values ($\Delta S^\circ > 0$) suggest an increase in randomness at the solid–liquid interface, which can be attributed to the displacement of water molecules and weakly bound species during adsorption. However, a clear distinction was observed in the Gibbs free energy values. While COD removal by CBPAC yielded positive ΔG° values throughout the studied temperature range, indicating a non-spontaneous process under the investigated conditions, the ΔG° values for RGO-CBPAC were negative at all temperatures and became increasingly negative with rising temperature. This result demonstrates that the incorporation of reduced graphene oxide significantly enhanced the thermodynamic feasibility of COD removal, rendering the process spontaneous and temperature-favoured.

In the case of NH₃-N removal, positive ΔH° and ΔS° values were obtained for both CBPAC and RGO-CBPAC, confirming that the adsorption process was endothermic and accompanied by an increase in system disorder. Nevertheless, the ΔG° values remained positive for both adsorbents across the entire temperature range, although lower ΔG° values were observed for RGO-CBPAC. These findings indicate that NH₃-N removal was not thermodynamically spontaneous under the examined conditions, but the RGO modification improved the energetic favourability of adsorption. The absence of spontaneous behaviour may be associated with the highly hydrated nature of ammonium species and competitive interactions with co-existing ions in the leachate matrix.

For turbidity removal, positive enthalpy and entropy changes were again observed for both adsorbents, confirming an endothermic process driven by increased interfacial disorder. In contrast to COD and NH₃-N, the ΔG° values for turbidity removal were negative for both CBPAC and RGO-CBPAC at all investigated temperatures, indicating a spontaneous adsorption process. Moreover, the increasingly negative ΔG° values with rising temperature suggest that higher temperatures further promote the removal of colloidal and suspended particles. Notably, RGO-CBPAC exhibited more negative ΔG° values at elevated temperatures despite a lower ΔH° compared to CBPAC, implying that turbidity removal on RGO-CBPAC required less energetic input while maintaining higher thermodynamic favourability.

On the whole, the thermodynamic assessment demonstrates that RGO-CBPAC consistently outperformed CBPAC, particularly for COD and turbidity removal, by providing lower (more negative) Gibbs free energy values and enhanced entropy contributions. The results confirm that RGO modification improves the surface–adsorbate interactions and promotes more favourable adsorption pathways, especially at elevated temperatures.

In conclusion, the integration of kinetic, isotherm, and thermodynamic analyses offers a holistic perspective on the adsorption mechanisms of CBPAC and RGO-CBPAC. The incorporation of RGO markedly enhances adsorption kinetics, equilibrium capacity, and thermodynamic favourability, thereby enabling more efficient removal of COD, NH₃-N, and turbidity. These findings highlight the superior physicochemical properties of RGO-CBPAC over conventional CBPAC, reinforcing its potential as a scalable and



Table 5 A comparative analysis of reported adsorbent capacities and those of CBPAC and RGO-CBPAC for the removal of COD and NH₃-N, and turbidity from the leachate

AC. type/precursor	Adsorbate	Leachate type	Capacity (mg/g)	Rem. (%)	References
Darco	COD	stabilized	–	38	Gotvajin et al. (2009)
Oil Palm shell	COD	stabilized	1460	50	Lim et al. (2009)
Com. PAC	COD	intermediate	–	24.6	Liyan et al. (2009)
Com. PAC	COD	synthetic	–	87	Karginodot (2004)
	ammonia		–	16	Karginodot (2004)
Rice husk	COD	young	–	70	Kalderis et al. (2008)
Com. PAC	COD	stabilized	6.5	–	Hur et al. (2001)
Desotec	COD	stabilized	268	–	Nooten et al. (2008)
SCG A. Carbon	COD	stabilized	40	> 90	Ferraz and Yuan (2020)
Norit 0.8	COD	intermediate	0.253	68	Chan et al. (2002)
Com. PAC	COD	stabilized	4300	38	Hur and Kim (2000)
Cal. Filt. 400	COD	stabilized	564	70	Morawe et al. (1995)
Oat hulls		–	40	90	Foo et al. (2013)
Com. PAC	COD	young	–	49	Aghamohammadi et al. (2007)
	Ammonia		–	78	Aghamohammadi et al. (2007)
Norit 0.8	COD	stabilized	88.80	90	Rivas (2003)
Com. PAC	COD	intermediate	–	75	Uygur and Kargı (2004)
	Ammonia		–	44	Uygur and Kargı (2004)
Act. Carbon	COD	stabilized	37.88	77	Halim et al. (2010)
	Ammonia		6.0790	29	Halim et al. (2010)
ACA	COD	–	47.37	–	Kaur et al. (2016)
TSAC	COD	–	64.93	80	Foo et al. (2013)
COLUFNAS/AC	Turbidity	young	–	45.9	AbdulKadir et al. (2023)
AC1%	Turbidity	intermediate	–	36	de Almeida et al. (2017)
RGO-CBPAC	COD	intermediate	588	57.7	In this study
RGO-CBPAC	Ammonia	intermediate	36.5	42	In this study
RGO-CBPAC	Turbidity	intermediate	58.5	63	In this study
CBPAC	COD	intermediate	500	48	In this study
CBPAC	Ammonia	intermediate	17.5	27	In this study
CBPAC	Turbidity	intermediate	18.1	52	In this study

high-performance adsorbent for advanced wastewater treatment applications.

Conclusion

Extensive characterization and adsorption investigations indicate that CBPAC and RGO-CBPAC particles possess distinct structural and mechanistic advantages for the removal of COD, NH₃-N, and turbidity from the leachate. BET measurements revealed that adsorption in CBPAC is largely controlled by its microporous network, whereas the hierarchical combination of micro- and mesopores in RGO-CBPAC improves overall adsorption capacity and enhances the diffusion of pollutants. FTIR and SEM-EDS analyses further confirmed that RGO integration not only increases surface area but also introduces additional functional groups, strengthening chemical interactions with contaminants. In summary, CBPAC micropores saturate

rapidly, but RGO-CBPAC maintains mesopore accessibility, sustaining adsorption over longer timescales.

Batch adsorption experiments investigating the effects of adsorbent dosage, solution temperature, initial pH, and stirring rate demonstrated consistent trends for both CBPAC and RGO-CBPAC. Adsorption onto CBPAC is primarily limited by rapid micropore saturation, whereas RGO-CBPAC displays higher adsorption capacities and accelerated kinetics. Elevated solution temperatures increase the kinetic energy of COD and turbidity molecules, reduce the thickness of the boundary layer surrounding the adsorbent, and decrease NH₃ solubility, collectively enhancing the adsorption efficiency and capacities of both materials.

Adsorption performance was strongly governed by solution pH, with acidic conditions (pH 5) yielding the highest removal efficiencies for COD, NH₃-N, and turbidity due to favourable electrostatic interactions and NH₄⁺ formation. In contrast, alkaline conditions reduced adsorption efficiency because of



Table 6 Thermodynamic parameters (ΔG° , ΔH° , ΔS°) for the adsorption of COD, $\text{NH}_3\text{-N}$, and turbidity

CBPAC						RGO-CBPAC					
COD						COD					
ΔH° (kJ/mol)	ΔS° (kJ/mol K)	ΔG° (kJ/mol)				ΔH° (kJ/mol)	ΔS° (kJ/mol K)	ΔG° (kJ/mol)			
		Temperature (K)						Temperature (K)			
		293	308	313	323			293	308	313	323
5.56	0.016	0.81	0.71	0.55	0.37	7.65	0.026	−0.03	−0.23	−0.49	−0.75
$\text{NH}_3\text{--N}$						$\text{NH}_3\text{--N}$					
ΔH° (kJ/mol)	ΔS° (kJ/mol K)	ΔG° (kJ/mol)				ΔH° (kJ/mol)	ΔS° (kJ/mol K)	ΔG° (kJ/mol)			
		Temperature (K)						Temperature (K)			
		293	308	313	323			293	308	313	323
7.44	0.015	3.30	2.95	2.81	2.66	8.4	0.022	1.96	1.74	1.16	1.30
Turbidity						Turbidity					
ΔH° (kJ/mol)	ΔS° (kJ/mol K)	ΔG° (kJ/mol)				ΔH° (kJ/mol)	ΔS° (kJ/mol K)	ΔG° (kJ/mol)			
		Temperature (K)						Temperature (K)			
		293	308	313	323			293	308	313	323
11.32	0.037	−0.48	−0.11	−0.26	−0.63	4.1	0.0154	−0.44	−0.59	−0.74	−0.89

increased electrostatic repulsion and weaker interaction mechanisms. RGO-CBPAC consistently exhibited better performance than CBPAC across all pH values, which can be attributed to its enhanced surface area, porosity, and functional groups introduced by RGO incorporation.

Kinetic and isotherm investigations revealed that RGO-CBPAC consistently achieves faster and more efficient adsorption than CBPAC, exhibiting strong correlations with both PFO and PSO models and better maximum monolayer capacities according to the Langmuir model. Thermodynamic analysis revealed that COD and turbidity removal using RGO-CBPAC were spontaneous ($\Delta G^\circ < 0$) and endothermic ($\Delta H^\circ > 0$), with increasing feasibility at elevated temperatures, whereas $\text{NH}_3\text{-N}$ removal remained non-spontaneous for both adsorbents despite improved energetic favourability following RGO modification.

The improved removal of COD, $\text{NH}_3\text{-N}$, and turbidity from diluted leachate by RGO-CBPAC was attributed to surface property modifications arising from the incorporation of RGO nanoparticles into the CBPAC structure. The introduction of reduced graphene oxide enhanced the specific surface area, optimised pore structure, and preserved mesoporous characteristics, thereby improving the accessibility of active sites. These structural modifications promoted electrostatic and $\pi\text{-}\pi$ interactions between the adsorbent surface and pollutants, while kinetic analysis indicated that adsorption on RGO-CBPAC more closely follows the pseudo-second-order model, suggesting a greater contribution of chemisorption, whereas CBPAC exhibited more pronounced diffusion limitations. Collectively, these findings not only explain the superior performance of the RGO-modified adsorbent but also highlight the critical role of

surface engineering in governing adsorption mechanisms. From a broader perspective, this study demonstrates that surface engineering of carbon-based adsorbents represents a highly effective strategy for improving wastewater treatment performance. Future studies should therefore focus on optimising graphene loading and investigating regeneration behaviour to further enhance the sustainability and practical applicability of such composite adsorbents.

Taken together, these results demonstrate that RGO-CBPAC surpasses CBPAC in the diluted leachate treatment due to its hierarchical pore structure, larger surface area, higher functional group density, and improved adsorption kinetics. The retention of mesoporous channels in RGO-CBPAC ensures sustained adsorption capacity and structural stability, making it a more effective adsorbent for long-term wastewater treatment applications.

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Data availability Data used in this work will be made available upon reasonable request to the corresponding author.

Declarations

Conflict of interest The authors declare no competing interests.



Ethical approval Not applicable.

Consent to participate All authors agreed to contribute to this study.

Consent for publication All authors agreed.

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