Effect of Physical Form on the Structural Characteristics and Congo Red Adsorption of CTAB-Modified Clinoptilolite

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The persistence of Congo Red (CR) dye in industrial effluents poses serious environmental and health risks, as it is a known carcinogen that can degrade into benzidine, a compound highly toxic to aquatic organisms. Clinoptilolite, a natural zeolite, is a low-cost and eco-friendly adsorbent with a strong inherent adsorption capacity. Surface modification with cetyltrimethylammonium bromide (CTAB) at concentrations of 0.05–5.0 mM has proven effective in enhancing its adsorption performance. This study comparatively evaluated the CR removal efficiency of CTAB-modified clinoptilolite in granular, powdered, and pelletized forms. The modified samples were characterized using FTIR, XRD, SEM and BET surface area analyses. FTIR and XRD confirmed successful surface modification without altering the zeolitic framework, while SEM revealed distinct morphological and porosity differences among the forms. BET analysis showed that pelletized CTAB-clinoptilolite had a surface area of 8.92 m^2/g and an average pore diameter of 10.51 nm. The optimal CTAB concentration was 1.0 mM for the granule and powder forms, which achieved maximum adsorption capacities of 6.79 and 7.60 mg/g, respectively, whereas pellets performed optimally at 5.0 mM with a capacity of 5.35 mg/g. This study highlights the potential of tailored clinoptilolite for practical wastewater treatment by balancing adsorption performance and structural stability.

Keywords: Adsorption, clinoptilolite, Congo Red, CTAB, modification

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Water-related issues such as scarcity, pollution, and quality degradation are among the most critical environmental concerns in the contemporary global context. While rapid technological advancement has brought many benefits, it has also introduced new challenges, including increased contamination and environmental disruption [1]. Currently, the continuous discharge of industrial effluents from sectors like textiles, rubber, cosmetics, paper, and leather, has introduced toxic and potentially carcinogenic compounds in aquatic environments and hospitals [2]. Among various pollutants, synthetic dye pollution is particularly concerning due to the widespread use of dyes in modern industries and their harmful effects on aquatic environments. It is estimated that up to 30 % of dyes may be wasted during processing and released into the environment [1]. These pollutants not only degrade water quality but also pose significant risks to ecosystems and human health [3]. Among these, Congo Red (CR) is an anionic azo dye with a molecular weight of 696.66 g/mol, characterized by a symmetrical aromatic structure and sulfonate functional groups which contribute to its high chemical stability and resistance to biodegradation. CR is widely used as a colourant in the textile, wood pulp, paper and food industries. In water, it initially forms a red colloidal solution before breaking down into benzidine, a known carcinogen to humans and a mutagen to aquatic organisms [4-5].

Currently, the most widely applied methods for treating dye-contaminated wastewater include membrane separation, biodegradation, photocatalytic degradation, chemical oxidation and adsorption [6-9]. Among these, adsorption technology receives research attention due to its advantages which include simplicity, cost-effectiveness, high efficiency, environmental compatibility, and the wide availability of adsorbent materials [10-12]. Adsorption is a surface-based process in which particles adhere to the surface of an adsorbent through physical interactions or chemical bonding [13]. Therefore, the development of efficient and low-cost adsorbents is essential for advancing the practical application of this technique [14].

Various natural and bio-based adsorbents have been reported in the literature, including cellulose-

based materials, agricultural waste, and natural zeolites. Cellulose-derived adsorbents, for instance, offer advantages such as biodegradability and abundant availability, while modified biomass materials have demonstrated promising adsorption capacities for dye removal [15-16]. Among these, adsorption using green materials such as clinoptilolite, a type of natural zeolite, offers several advantages over activated carbon, including lower cost and wide availability [17]. Clinoptilolite has been extensively studied for applications including water and wastewater treatment due to its excellent adsorption properties [18]. Its general unit-cell formula, $(Na_2K_2CaMg)_3[(AlO_2)_6(SiO_2)_{30}].24H_2O$, classifies it as a porous aluminosilicate with a robust three-dimensional framework that facilitates outstanding adsorption capabilities [19]. Furthermore, clinoptilolite is a naturally mined zeolite that can be obtained from commercial suppliers, typically with a purity ranging from 70 - 90 % [20]. Owing to its negatively charged framework, it effectively attracts cations and adsorbs positively charged substances [21].

To enhance the removal of anionic contaminants such as Congo Red (CR) dye from aqueous solutions, surface modification of clinoptilolite with cationic surfactants, particularly cetyltrimethylammonium bromide (CTAB), has been extensively investigated and demonstrated as a promising strategy [22]. This modification forms a CTAB layer on the clinoptilolite surface, imparting improved affinity for both hydrophobic and hydrophilic compounds due to the alkyl chain and the hydrophilic head containing the positively charged nitrogen group [23]. The arrangement of CTAB molecules on the clinoptilolite surface will depend on the initial surfactant concentration [23]. Moreover, this modification process enhances adsorption performance by minimizing pore blockages from impurities and altering surface charge, enabling CTAB-modified clinoptilolite to more effectively capture CR dye molecules with varying characteristics [18,24].

The morphology and pore structure of the adsorbent can affect the accessibility of adsorbate molecules, thereby influencing adsorption kinetics and equilibrium behaviour. Powdered adsorbents generally provide a high surface area and rapid adsorption kinetics; however, their application in large-scale operations and regeneration processes remains impractical. A major limitation of powdered adsorbents at the industrial scale is the requirement of post-filtration to prevent material loss, which can lead to secondary contamination, sediment accumulation in pipelines, and operational issues such as valve blockage and pump failure [25]. Therefore, the development of pelletized adsorbents is essential to address these limitations. Pelletized adsorbents offer several practical advantages, including improved handling, reduced risk of clogging, enhanced flow characteristics within treatment systems, and easier regeneration. The use of surfactant-modified pelletized clinoptilolite represents a promising approach to enhance the practicality of conventional powdered adsorbents in wastewater treatment, potentially eliminating the need for post-filtration, reducing equipment wear and maintenance, and minimizing adsorbent loss during operation.

This study aims to evaluate and compare the adsorption capacity of CTAB-modified clinoptilolite in granular, powdered, and pelletized forms for the removal of Congo Red (CR) dye from aqueous solutions. The modified materials were characterized using X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and Brunauer–Emmett–Teller (BET) surface area analysis. In addition, the effect of the initial CTAB concentration on the adsorption process was examined.

EXPERIMENTAL

Chemicals and Materials

Congo Red ($C_{32}H_{22}N_6Na_2O_6S_2$) and cetyltrimethyl-ammonium bromide ($C_{19}H_{42}NBr$) were obtained from Chemiz and R&M Chemicals, Selangor, Malaysia. Clinoptilolite granules, a type of natural zeolite, were purchased from ACME Chemicals Pte Ltd, in Klang, Selangor.

Preparation of Clinoptilolite

Natural clinoptilolite granules were washed multiple times with tap water to remove impurities, then oven-dried at 60-80 °C for 24 hours. The dried granules were ground into powder using a Fritsch Pulverisette 6 ball mill, and the particle size was measured to be approximately ±0.42 µm using a Zetasizer Nano ZS90. The resulting powdered clinoptilolite was stored in an airtight container to prevent contamination and moisture uptake. To prepare pellets, 0.075 g of the powder was pressed into a single pellet using a hydraulic press at 1 ton of pressure. The pellets were then thermally treated in a furnace at 300-350 °C for 48 hours. The diameter and thickness of the pellets were measured using a digital vernier calliper.

Surface Modification

A cetyltrimethylammonium bromide (CTAB) solution was prepared at concentrations ranging from 0.05 to 5.0 mM. For the granule and powder forms of clinoptilolite, 2 g of each were added to the CTAB solution and stirred continuously using a rotary shaker at 120 rpm for 24 hours at room temperature to allow surface modification via cation exchange. In the case of pelletized clinoptilolite, the pellets were immersed in 200 mL of CTAB solution under the same conditions. After modification, the clinoptilolite samples were separated by filtration, washed several times with distilled water to remove unbound

CTAB, and dried at 60-80 °C for 24 hours. The resulting CTAB-modified clinoptilolite was stored in airtight containers for subsequent characterization and adsorption experiments.

Scanning Electron Microscopy (SEM)

The surface morphology of the clinoptilolite samples was examined using a JEOL scanning electron microscope (SEM, Model JSM-5910). The SEM was operated at an accelerating voltage of 15–20 kV and a current of 10–20 μA, enabling high-resolution imaging of surface features. For analysis, the samples were mounted on aluminium stubs using carbon tape placed on a graphite support unit. To enhance electron conductivity and minimize surface charging, the samples were coated with a thin layer of gold using a sputter coater. SEM imaging was then performed under high vacuum conditions, allowing for the capture of detailed surface images at various magnifications.

Fourier Transform Infrared (FTIR) Analysis

Fourier Transform Infrared (FTIR) spectra were recorded using an Agilent Cary 630 spectrometer equipped with a diamond ATR (Attenuated Total Reflectance) crystal, covering a frequency range of 650–4000 cm^{-1}. This setup enabled the identification of characteristic absorption bands related to chemical bonds and functional groups present in the samples, particularly those introduced through surface modification of the clinoptilolite.

X-Ray Diffraction (XRD)

X-ray diffraction (XRD) analysis (Rigaku XRD D/Max 2200V/PC) was used to determine the crystal structure of clinoptilolite before and after surface modification. The sample was scanned over a 2θ range of 5° to 80° using CuKα radiation (λ = 1.5405 Å), with an operating voltage of 40 kV and an emission current of 30 mA.

Brunaeur-Emmett Teller (BET) Method

A surface area analyzer (Micromeritics Tristar II Plus) was employed to measure the total pore volume, specific surface area and average pore size of the clinoptilolite. This instrument operates by measuring the amount of nitrogen (N_2) gas adsorbed onto the sample's surface at roughly -195 to -196 ℃. The adsorption data obtained from this analysis provided critical information regarding the porosity and surface characteristics of the modified clinoptilolite, which are essential for evaluating its effectiveness as an adsorbent.

Adsorption Experiments

A stock solution of Congo Red (CR) dye at a concentration of 100 mg/L was prepared by dissolving 0.1 g dye in 1000 mL of distilled water in an Erlenmeyer flask. Batch adsorption experiments were carried out to investigate the effect of CTAB concentration, ranging from 0.05 to 5.0 mM. For each test, a fixed amount of modified clinoptilolite was added to 100 mL of CR dye solution at a concentration of 30 mg/L. The mixture was agitated at 120 rpm using an orbital shaking incubator for a specified period to ensure uniform mixing and effective contact between the adsorbent and the dye molecules. At one-hour intervals, the residual dye concentration in the solution was measured using a UV-Visible spectrophotometer (Agilent Cary 60) at 495 nm, the maximum absorbance wavelength for Congo Red. For powdered clinoptilolite samples, centrifugation was performed at 3000 rpm for 10 minutes to separate the adsorbent from the solution. The adsorption capacity (q_e, mg/g) and percentage removal of CR dye (%) were calculated using Equations (1) and (2), which provide a quantitative evaluation of the adsorption performance of the CTAB-modified clinoptilolite.

$$q_e = (C_i - C_f)\frac{V}{m} \quad (1)$$

$$\text{Removal of dye (\%)} = \left[\frac{C_i - C_f}{C_i}\right] \times 100 \quad (2)$$

where: C_i is the initial concentration (mg/L) and C_f is the final concentration (mg/L) of Congo Red (CR), V represents the solution volume (L), and m is the mass of the pellets, respectively [26].

RESULTS AND DISCUSSION

Pellet Thickness and Diameter

The clinoptilolite pellets prepared in this study measured 6.0 mm in diameter and 2.3 mm in thickness, as shown in Figure 1. These dimensions result in a compact structure that offers practical advantages for adsorption applications [27]. The relatively small thickness reduces intraparticle diffusion resistance, enhancing dye molecule access to active sites. Similar findings were reported in a study [27] on activated carbon pellets, where smaller pellet size improved both adsorption capacity and uptake kinetics. Moreover, the 6.0 mm diameter provides sufficient mechanical strength for repeated handling and regeneration, essential for large-scale or continuous operations such as packed-bed columns [28]. Figure 2 presents the top and side views of the pellets. Here, thickness refers to the height of the flat, cylindrical portion of the pellet, excluding the convex top and bottom surfaces [29].

Figure 1. Digital calliper measurements of a clinoptilolite pellet showing **(a)** diameter and **(b)** thickness.

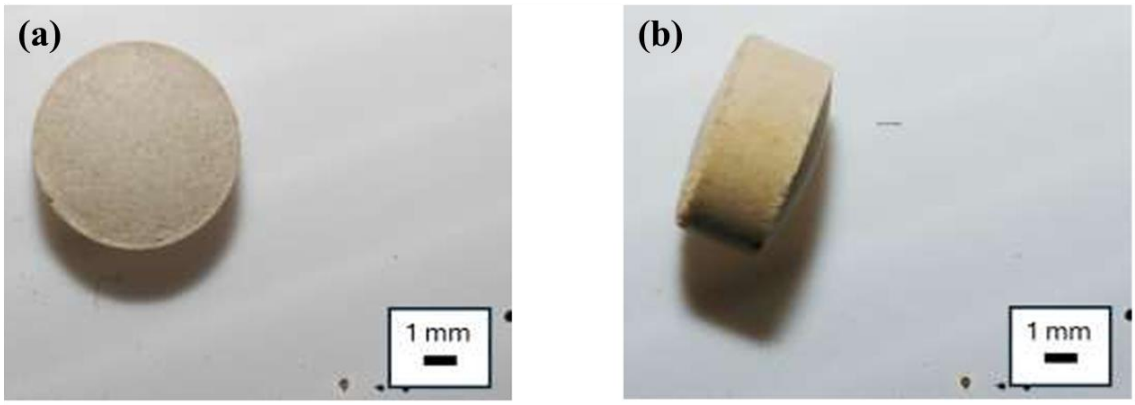

Figure 2. A clinoptilolite pellet: **(a)** top view and **(b)** side view.

Characterization of Clinoptilolite

FTIR Analysis

The FTIR spectrum of CTAB displayed strong C–H stretching peaks around 2915–2850 cm^{-1} and bending vibrations near 1450 cm^{-1}, as shown in Figure 3, confirming the presence of long alkyl chains at a concentration of 1.0 mM. This is consistent with a previous work [22], which reported peaks at 2918 cm^{-1} and 2850 cm^{-1} corresponding to the asymmetric and symmetric C-H stretching vibrations of methyl ($-CH_3$) and methylene ($-CH_2$) groups in the alkyl chains, respectively. The band at 1630 cm^{-1} corresponds to the bending vibration of the H–O–H bond, indicating water adsorption on the surface of the samples [21-22].

The strong and broad peak around 990 cm^{-1} was attributed to the asymmetric stretching vibration of the Si–O–Al bond. Peaks at 773 cm^{-1} and 800 cm^{-1} were assigned to the symmetric stretching vibrations of internal tetrahedral structures and the vibrations of double-ring external linkages, respectively, both related to stretching vibrations in the alumino- and silico-oxygen bridges. The FTIR spectra of CTAB-modified clinoptilolite showed no significant shifts in the characteristic band frequencies of clinoptilolite, indicating that CTAB adsorption occurred mainly on the zeolite surface without altering the Si–O and Al–O linkages. These results suggest that the incorporation of CTAB did not compromise the integrity of the Si–O and Al–O framework, confirming that the overall structure of natural clinoptilolite remained intact after surfactant modification. However, the modified clinoptilolite displayed a new weak peak at 2915 cm^{-1}, associated with the C–H stretching vibrations of the surfactant's hydrocarbon chains. This finding further confirmed successful CTAB loading onto the clinoptilolite surface [21]. According to a recent work [22], the characteristic C-H peaks at 2918 cm^{-1} and 2850 cm^{-1} in CTAB-modified zeolite FTIR spectra also indicate successful surfactant incorporation.

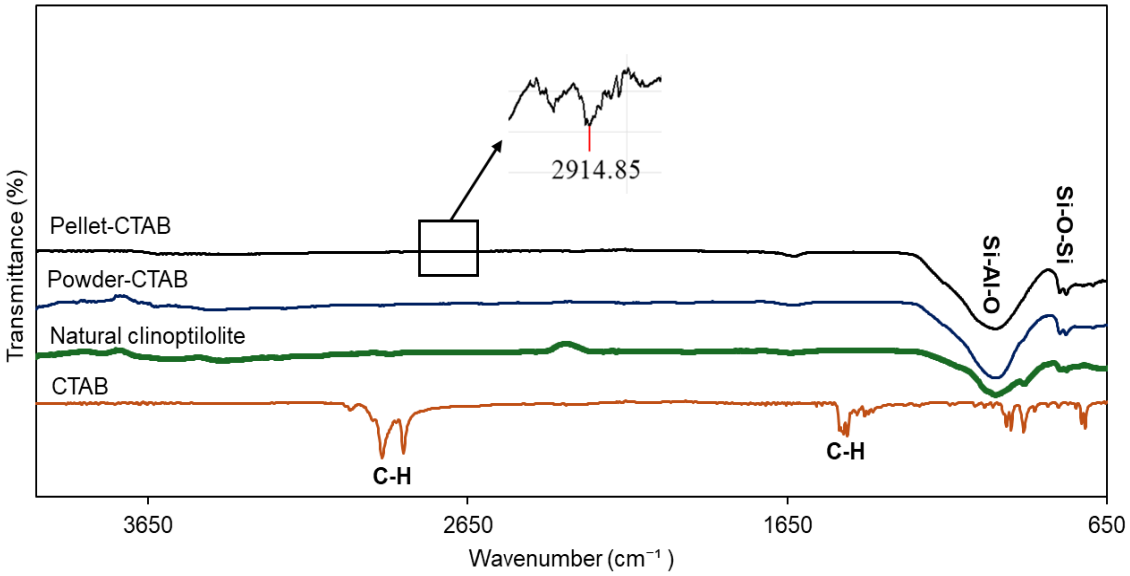

Figure 3. FTIR analysis of CTAB, natural clinoptilolite, modified powder, and pellets.

X-Ray Diffraction (XRD) Analysis

XRD analysis is a valuable, non-destructive technique for identifying crystalline phases in solids and studying their structural properties. Figure 4 presents the X-ray diffraction (XRD) patterns of natural clinoptilolite and CTAB-modified samples in powder, granular, and pellet forms. All samples exhibited the characteristic diffraction peak of clinoptilolite at approximately 26.41° (2θ), corresponding to the (020) plane, confirming the presence of the zeolitic framework [JCPDS 39-1383]. Additional peaks near 22.14° and 29.75° were also consistent with the typical clinoptilolite structure [30].

The XRD patterns of the CTAB-modified samples showed slight differences in the intensity and sharpness of their diffraction peaks compared to natural clinoptilolite, although no changes in their position were observed. Similar findings have been reported previously [21]. These results suggest that the modification primarily occurred on the external surface of the material, in agreement with earlier studies [21,30]. A slight reduction in peak intensity was observed in the modified samples, which may indicate partial pore blockage by CTAB molecules rather than a collapse of the crystal structure.

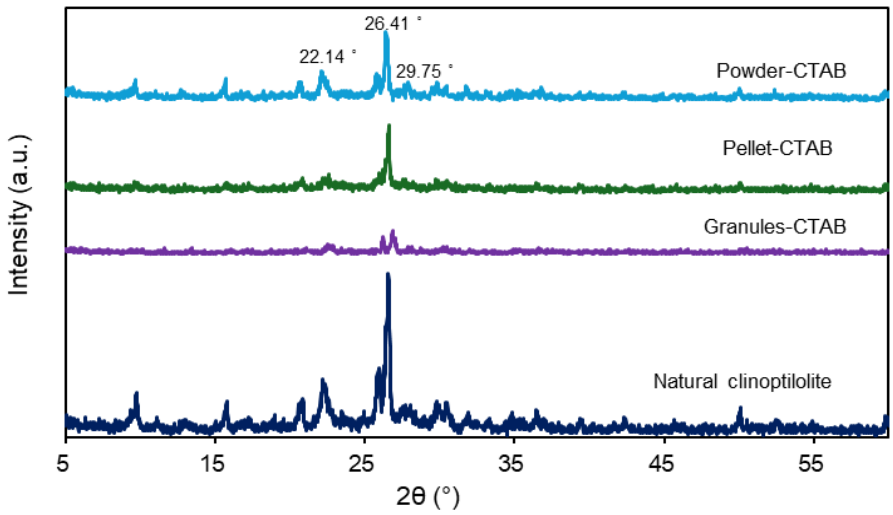

Figure 4. X-ray diffraction (XRD) patterns of natural clinoptilolite and CTAB-modified samples in powder, granular and pellet forms.

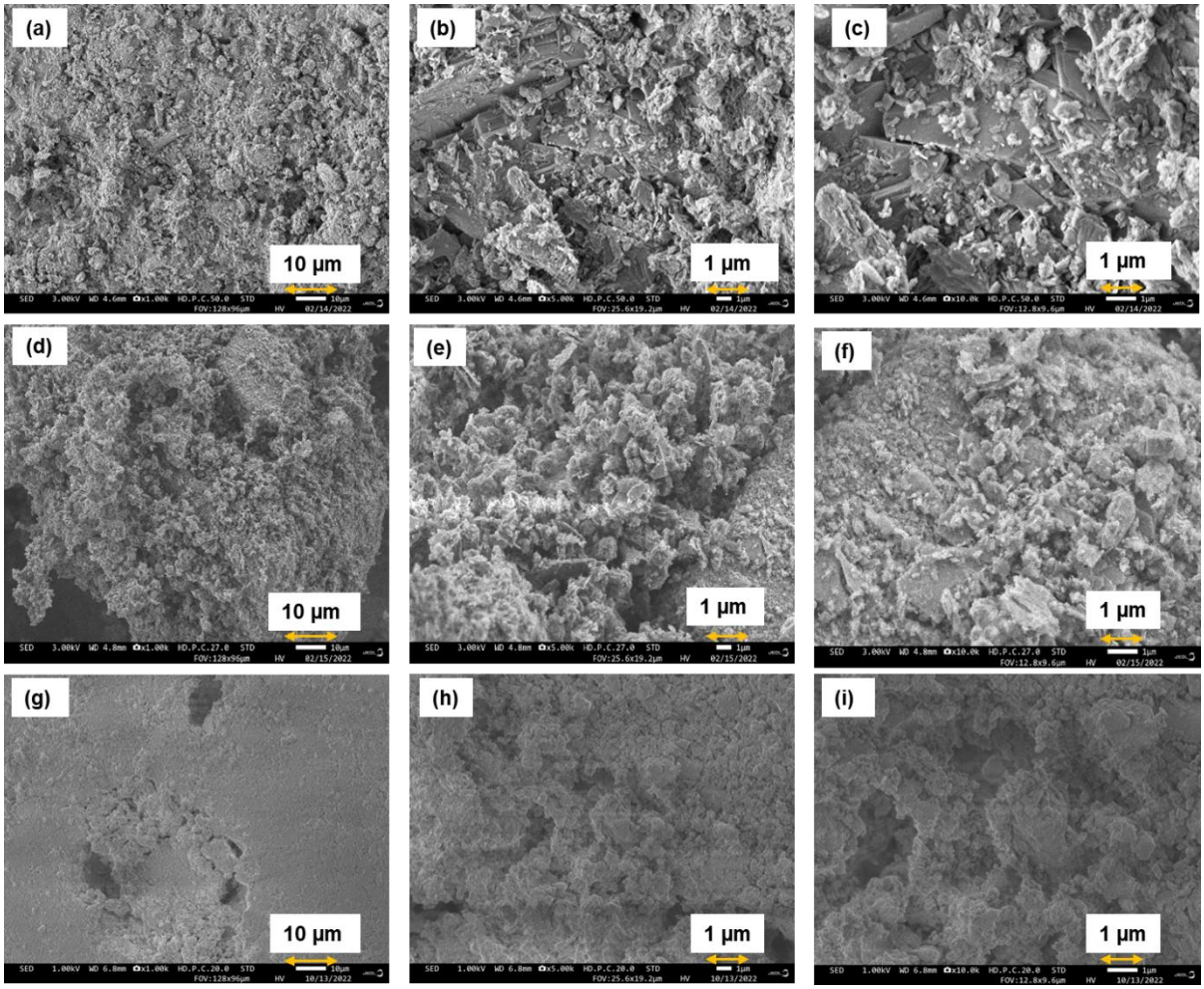

Figure 5. SEM images of clinoptilolite in **(a–c)** granular form, **(d–f)** powder form and **(g-i)** pellet form.

Scanning Electron Microscopy (SEM) Analysis

The surface morphology of clinoptilolite in granular, powdered and pelletized forms was examined using SEM. Figure 5(a)–(c) revealed needle-like structures with geometric, sharp edges. Figure 5(d)–(f) showed that the powdered form exhibited a rough, highly porous, and irregular surface structure with numerous cracks and crevices. This rough and open texture resulted from the grinding of granules into fine powder, which increased surface defects and exposed more internal pores. Consequently, the powdered form possessed a significantly larger specific surface area and more accessible active sites, which greatly enhanced its adsorption capacity by providing additional locations for dye molecules to attach. In contrast, Figure 5(g)–(i) illustrate the pelletized form, which appeared more compact, smoother, and denser, with fewer visible pores and cracks. Pelletization typically compresses the structure and reduces pore accessibility, leading to a decrease in accessible surface area and active adsorption sites, which may result in lower adsorption capacity compared to the powdered form.

Brunauer Emmet Teller (BET) Analysis

The Brunauer–Emmett–Teller (BET) method was used to determine the specific surface area of clinoptilolite based on N_2 adsorption–desorption isotherms. A summary of the textural properties of Pellet-CTAB and powdered forms reported in the literature, including surface area, pore volume and pore diameter, is presented in Table 1. In this study, Pellet-CTAB clinoptilolite exhibited a specific surface area of 8.92 m^2/g, a pore volume of 0.023 cm^3/g, and an average pore diameter of 10.51 nm.

Table 1. Summary of textural properties of zeolitic materials in pelletized and powdered forms.

Sample	Specific surface area (m^2/g)	Total pore volume (cm^3/g)	Average pore diameter (nm)	Reference
Pellet-CTAB (5.0 mM)	8.92	0.023	10.51	This study
Powder (Surfactant Modified Zeolite)	28.90	0.081		[21]
Powder (Natural Clinoptilolite)	11.93	0.094	31.54	[34]
Powder (Surfactant Modified Clinoptilolite)	7.64	0.039	20.52	[34]
Powder (Surfactant Modified Clinoptilolite)	5.41	0.051	27.57	[30]
Powder (Natural Clinoptilolite)	14.68	0.116		[31]

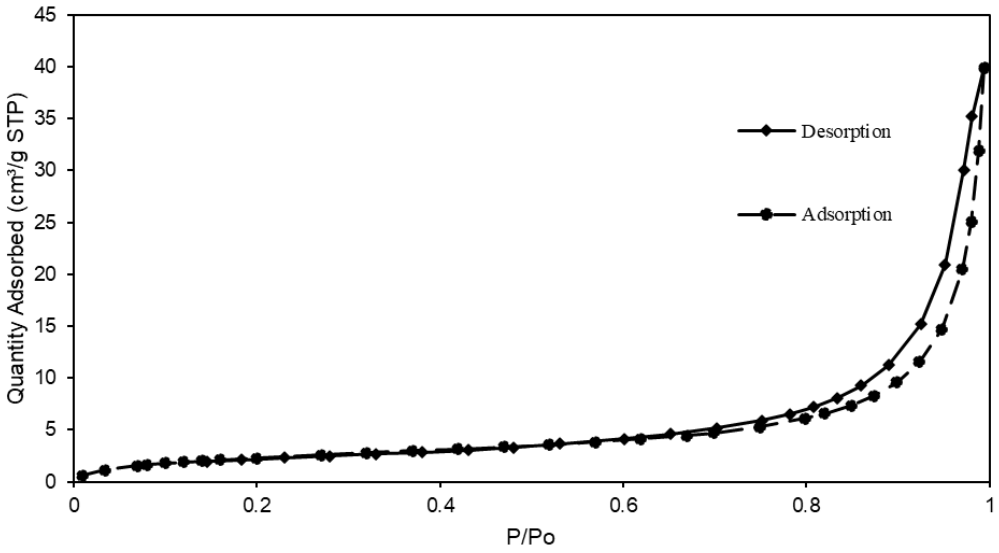

Figure 6. N_2 adsorption-desorption isotherms of Pellet-CTAB clinoptilolite.

The isotherm in Figure 6 displays a typical type IV curve with an H3 hysteresis loop, indicating the presence of mesopores (2–50 nm) [31-32], which is consistent with recent findings [33] and supports the measured average pore diameter. The decrease in the adsorbed volume of the modified clinoptilolite suggests that the presence of cationic surfactant on the surface reduced the available surface area. As shown in Table 1, the surface area decreased by 26 % and 14 % compared to previously reported values [31,34]. The reduction in surface area is likely due to pore blocking by the surfactant, as reported by other researchers [31].

Adsorption Study for CR Removal

Effect of CTAB Concentration

Figure 7 shows the effect of CTAB concentration on (a) the adsorption capacity, and (b) the percentage removal of Congo Red (CR) dye using modified clinoptilolite in granular, powder, and pellet forms. The experiments were conducted using a 40 mg/L CR solution, and 0.375 g of modified adsorbent, under equilibrium contact time conditions. The adsorption capacity of the granular form steadily increased with CTAB concentration, rising from 2.52 mg/g at

0.05 mM to 6.79 mg/g at 1.0 mM. Beyond this point, at 4.0 and 5.0 mM, no significant improvement in adsorption capacity was observed. Similarly, CR removal efficiency rose from 31.47 % at 0.05 mM to 84.61 % at 1.0 mM but declined slightly to 76 % at higher CTAB concentrations. The adsorption capacity of the powder form increased from 6.10 mg/g at 0.05 mM to 7.60 mg/g at 1.0 mM, then plateaued slightly around 7.5 mg/g at 4.0 and 5.0 mM. The highest CR removal efficiency, 96.09 %, was achieved at 1.0 mM, with only a slight decrease to 95 % at higher concentrations. Overall, the powder form demonstrated the highest uptake performance among the three forms. The adsorption capacity of the pellet form increased from 3.15 mg/g at 0.05 mM to 5.35 mg/g at 5.0 mM, while the maximum CR removal efficiency of 99.28 % was observed at 5.0 mM. The lower adsorption capacity of the pellet form compared to the powder is likely due to the short contact time during the adsorption experiment, which may have limited the diffusion, interaction, and proper arrangement of CTAB molecules on the pellet surface.

Although the critical micelle concentration (CMC) of CTAB is 1.0 mM [22], the pellet form achieved optimal adsorption capacity at 5.0 mM, deviating from the expected trend which is likely a result of insufficient time for the CTAB molecules to fully bind at lower concentrations. The amount of CTAB molecules attached to the clinoptilolite surface increased with concentration up to about 1.0 mM, but declined beyond that point. This finding suggests that the dye adsorption performance was strongly influenced by how much CTAB was bound to the surface. The enhanced adsorption can be attributed to stronger hydrophobic interactions between the aromatic rings of the CR dye and the alkyl tails of the CTAB molecules. This behaviour aligns with the findings reported by another study [14], where increased dye uptake was observed up to the CMC, and attributed to enhanced hydrophobic interactions and improved surface functionalization. As the CTAB concentration increased from 0.05 to 1.0 mM, both adsorption capacity and CR removal efficiency also increased, especially for the granular and powder forms. Therefore, 1.0 mM was identified as the optimal concentration for modifying the adsorbent.

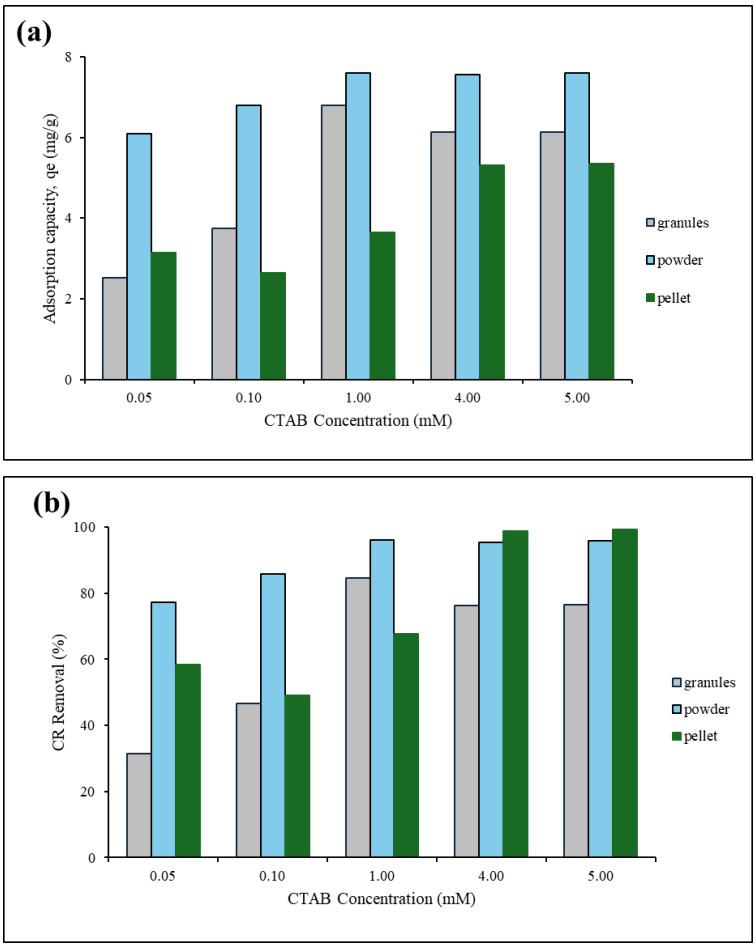

Figure 7. Effect of CTAB concentration on **(a)** adsorption capacity, and **(b)** percentage removal of CR.

CONCLUSION

This study highlights the comparative performance of CTAB-modified clinoptilolite in three physical forms, powder, granules and pellets, for the removal of Congo Red (CR) dye from aqueous solutions. The surface of the clinoptilolite was successfully modified using a range of CTAB concentrations, from 0.05 mM to 5.0 mM. Characterization techniques including Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), Brunauer–Emmett–Teller (BET) surface area analysis, and scanning electron microscopy (SEM) were used to study the effect of CTAB concentration on the structural properties and the nature of the surfactant layer formed on the modified clinoptilolite surfaces. The FTIR and XRD results confirmed that the clinoptilolite was successfully coated with CTAB. Among the three forms, BET analysis showed that the Pellet-CTAB clinoptilolite had a surface area of 8.92 m^2/g and an average pore diameter of 10.51 nm. The primary aim of this study was to determine the optimal CTAB concentration for maximizing both the adsorption capacity (q_e) and the removal efficiency of CR dye (%). The optimal concentration for both the powdered and granular forms was found to be 1.0 mM. At this concentration, the powdered form achieved a maximum adsorption capacity of 7.60 mg/g with a CR removal efficiency of 96.09 %, while the granular form reached 6.79 mg/g and 84.61 % removal efficiency. Increasing the CTAB concentration beyond the CMC did not result in any further enhancement of adsorption performance. In contrast, for the pelletized form, the optimal CTAB concentration was found to be 5.0 mM, which resulted in a maximum adsorption capacity of 5.35 mg/g and a CR removal efficiency of 99.28 %. By comparing the adsorption behaviour and structural properties of each physical form, this study offers a detailed understanding of the adsorption mechanisms of CR dye on CTAB-modified clinoptilolite and supports the use of this natural material in environmental remediation. Powdered clinoptilolite demonstrated the highest surface area and the most irregular porous structure, which significantly contributed to its superior adsorption performance. Additionally, the CTAB-modified clinoptilolite, especially in powder and pellet forms, showed great promise as a versatile and effective adsorbent for treating dye-polluted wastewater. Scaling up the pellet form for integration into continuous flow systems will be essential for practical application in industrial wastewater treatment processes.

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