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Proceeding Paper

Characterization and Removal Efficiency Analysis of MWCNT/Clay Nanocomposites for MB Dye Adsorption †

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Abstract: Multi-walled carbon nanotubes (MWCNTs) combined with clay have shown potential as effective adsorbents for dye removal. This study aims to characterize MWCNT/clay nanocomposites and analyze their removal efficiency for methylene blue (MB) dye under various conditions. The nanocomposites were characterized using techniques such as FESEM, TEM, EDX, TGA, and XRD. The removal efficiency was studied concerning different weights, concentrations, temperatures, pH levels, and comparative amounts of CNT in the composites. The findings revealed distinct properties and behaviors of the nanocomposites, with removal efficiency significantly influenced by weight, MB dye concentration, temperature, and pH. A higher CNT content in the composite corresponded to better removal results. The study demonstrates the potential of MWCNT/clay nanocomposites in wastewater treatment, with insights into optimal conditions for dye removal. The investigation adds valuable knowledge to the field and indicates promising directions for future research.

Keywords: MWCNT/clay nanocomposites; MB dye removal; adsorption efficiency; characterization techniques; wastewater treatment

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1. Introduction

Industrial dyes, often utilized in various manufacturing processes, have been identified as environmental hazards. Their toxicity can directly or indirectly affect living organisms, primarily through their interaction with sunlight entering water bodies. This interaction can lead to interference with the growth of aquatic animals and a decrease in oxygen solubility, thereby inhibiting the proliferation of aquatic biota [1–6]. Dyes are not only detrimental to aquatic life but can also have harmful effects on human health, causing allergic reactions, dermatitis, and skin irritation. This has made dye removal an area of significant research interest, with a focus on reducing environmental contamination and preserving ecosystems. Various water treatment methods have been developed to remove dye molecules from water, such as ion exchange, bio-degradation, coagulation, and precipitation. It is worth noting that each method has its unique advantages and drawbacks in the context of dye removal [7–10]. Among these methods, the adsorption technique has gained popularity for its cost-effectiveness, simple operation, environmental friendliness, and ability to work at ambient temperature and pressure. Organic dyes, in particular, must be removed from effluents, and to achieve this, various well-known adsorbents such as hydrogel, activated carbon, natural fibers, carbon nanotubes, zeolites, and polymer materials have been developed [6,11–15]. The efficacy of adsorption as a vital decolonization method in dyeing wastewater treatment is recognized, with its reliance on kinetic and equilibrium measurements offering simplicity and cost savings [16].

Methylene blue (3,7-bis(dimethylamino) phenothiazine chloride tetra methylthionine chloride), a synthetic dye, is extensively used across industries, including paper, textiles, food, cosmetics, and pharmaceuticals. Its presence, however, poses significant challenges to plants, leading to growth inhibition, reduction in pigment, and protein content of microalgae. Given the harmful effects of wastewater containing methylene blue (MB) dye, comprehensive treatment is mandated prior to industrial discharge [17–19]. This work seeks to explore the advancements, challenges, and future prospects of dye removal techniques, with a particular emphasis on the application and optimization of the adsorption method.

2. Materials and Methods

2.1. Preparation of MB Dye Standard Solution

A standard solution was prepared by dissolving 0.1 g of MB dye in distilled water to create a 100 mg/L concentration. This solution served as the basis for creating a calibration curve, utilizing a series of concentrations ranging from 2 to 20 mg/L. The absorbance of these concentrations was measured at a wavelength of 630 λ_{max}, with the resulting data plotted in Figure 1.

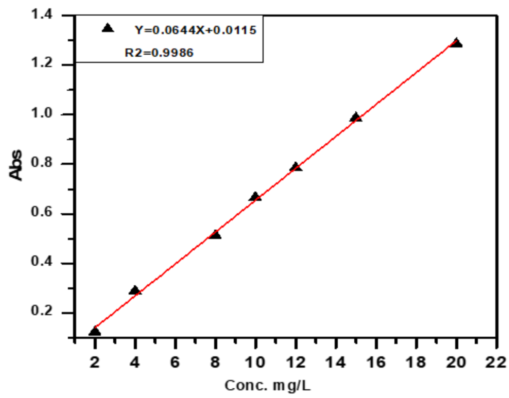


Figure 1. Calibration curve of methylene blue dye.

2.2. Preparation of MWCNT/Clay Nanocomposite

The MWCNT/clay nanocomposite was synthesized through a hydrothermal process, as depicted in Figure 2. Approximately 10 g of bentonite clay (sourced from Basra, Iraq) and 0.5 g of MWCNT (supplied from China) were mixed in 50 mL of distilled water each, then combined and stirred for 1 h. The resulting mixture was autoclaved at 160 °C for 24 h. The obtained dark brown precipitate was subsequently filtered, washed with distilled water and ethanol, subjected to ultrasonic treatment in 15 min intervals, and finally dried at 90 °C for 24 h to yield a fine powder.

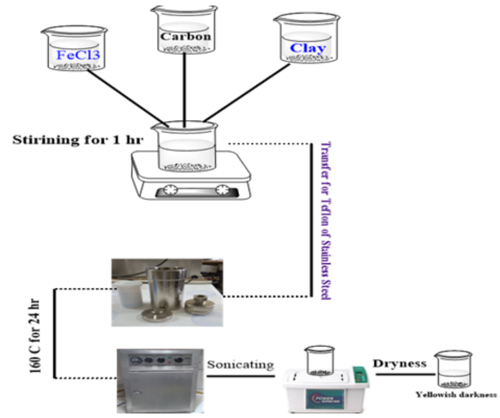


Figure 2. Preparation of multi-walled carbon nanotube (MWCNT)/clay (bentonite) nanocomposite.

2.3. *Adsorption Isotherm*

Adsorption experiments were conducted in a shaker water bath, using 100 mL elementary flasks. Various factors influencing the adsorption process were explored, including pH (3–10), initial dye concentration (10–100 mg/L), and temperature (15–45 °C), under optimal conditions of 25 °C and an agitation rate of 120 rpm. The residual concentration of MB dye in each aliquot was measured using a UV-VIS spectrophotometer. The adsorption efficiency (Qe) and percentage of adsorption ($E\%$) were computed using Equations (1) and (2):

$$Qe = \frac{(C_0 - C_e) \cdot V_{\mathrm{mL}}}{W_{\mathrm{t\ gm}}} \quad (1)$$

$$E\% = \frac{(C_0 - C_e)}{C_0} \times 100 \quad (2)$$

where C_0 and C_e represent the initial and equilibrium concentrations of MB dye, respectively, V_{mL} is the volume in milliliters, and $W_{\mathrm{t\ gm}}$ is the weight in grams. These equations guided the assessment of the adsorption characteristics of the synthesized MWCNT/clay nanocomposite.

2.4. *Characterization of MWCNT/Clay Nanocomposites*

Characterization of the nanocomposites was carried out using various techniques to understand the morphology, composition, and structural properties of the material. Field emission scanning electron microscopy (FESEM) was used to observe the surface morphology and detect porous and rough structures. Transmission electron microscopy (TEM) was employed to study the surface morphology at a more detailed level, including the identification of clusters and carbon nanotube loading on the clay surface. Energy-dispersive X-ray (EDX) spectroscopy was used to analyze the elemental composition and the presence of specific elements in the nanocomposites. Thermogravimetric analysis (TGA) was applied to assess the thermal stability of the nanocomposites. Finally, X-ray diffraction (XRD) was conducted to study the crystalline phases and purity components and to identify any diffracted signals from the carbon nanotubes.

3. Result and Discussion

3.1. *Characterization of MWCNT/Clay Nanocomposites*

Figure 3 provides various insights into the characterization of MWCNT/clay bentonite nanocomposites using different techniques.

3.1.1. FESEM Analysis

The FESEM image, presented in Figure 3A, reveals the lamellar structures of MWCNT/clay bentonite, exhibiting a clearly porous and rough fracture surface. This structure enhances the absorbents, facilitating water diffusion into the absorbent. A noticeable change in morphology is evident after adsorption, as Figure 3B displays new, unequal, large particles on the surface. Such changes result in an increased protuberance and coarseness of the surface structure that was absent prior to adsorption [19,20].

3.1.2. TEM Analysis

The application of TEM as an appropriate technique for studying surface morphology is supported in Figure 3C. The clusters appearing in the form of dark spots are indicative of carbon nanotube loading on the surface of the clay [21].

3.1.3. EDX Analysis

Figure 3D shows the EDX analysis of MWCNT/clay nanocomposites. The synthesized nano-composite consists of elements O, C, Si, Ca, Al, Mg, Fe, and K. This composition indicates the presence of bentonite clay in MWCNT. The modified MWCNT exhibits values of the lowest and highest elements at 40.3 wt.% and 0.4 wt.%, respectively [10,22].

3.1.4. TGA Analysis

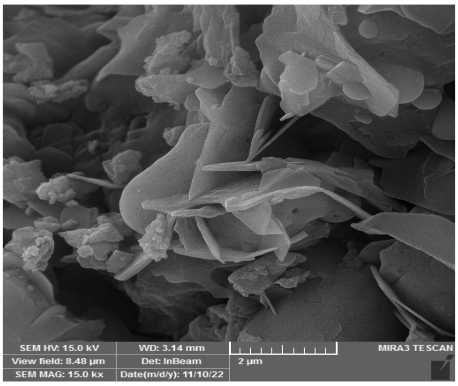
The TGA technique reveals an approving effect of clay incorporation on the thermal stability of the surface, as well as a decrease in the weight loss percentage, as shown in Figure 3E. This result underscores the positive impact of the MWCNT/clay combination on thermal properties [23].

3.1.5. XRD Analysis

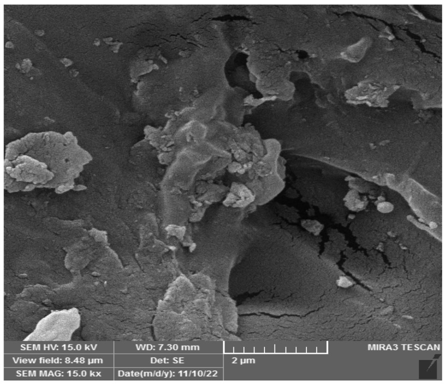
The XRD measurement, used to study the crystal phases and purity components of nanomaterials, is presented in Figure 3F. The X-ray diffraction pattern of the MWCNT/clay nanocomposites shows that all patterns are predominantly semi-similar. Interestingly, there are no diffracted signals of the 0.2, 0.05, and 0.1 g ratios of the carbon nanotube. This phenomenon is attributed to the low weight percentage or non-crystallinity of doping materials on the crystal surface with the 2θ values at 20.1, 21.2, and 27.7 for three types of the 0.2, 0.1, and 0.05 g ratio of the carbon nanotube [19,22,24].

3.2. Effect of Weight of MWCNT/Clay Nanocomposites on Adsorption

In this experiment, the removal percentage and adsorption efficiency were studied based on the utilization of different weights (0.025–0.15 g) of MWCNT/clay nanocomposites. The study was conducted at a concentration of MB (methylene blue) of 40 mg L^{-1} at a pH of 6, and the results are depicted in Figure 4. The removal percentage of MB increased from 70.11% to 95.23% with an increase in the weight of MWCNT/clay from 0.025 to 0.15 g. Correspondingly, the adsorption capacity of MB decreased from 94.0 to 25.4 mg/g. This trend is consistent with previous studies [25], confirming the inverse relationship between the weight of the adsorbent and the adsorption capacity. Consequently, a weight of 0.1 g was determined as the optimal amount for the adsorbent in this particular study. The observed increase in adsorption efficiency (E%) can be attributed to the increase in the number of adsorption sites and the overall increase in surface area [26]. These findings provide valuable insights into optimizing the weight of MWCNT/clay nanocomposites for enhanced adsorption performance.



(**A**)



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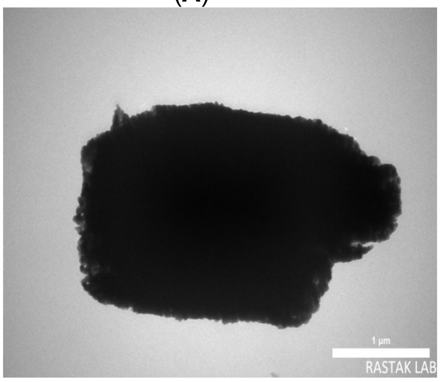

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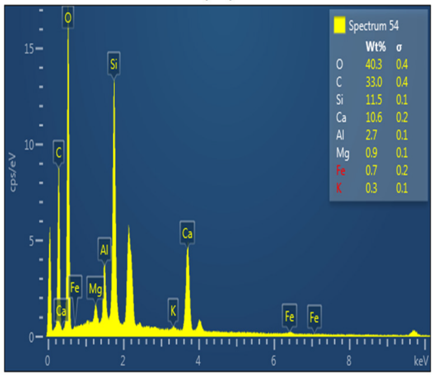

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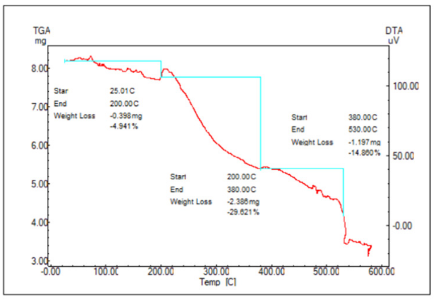
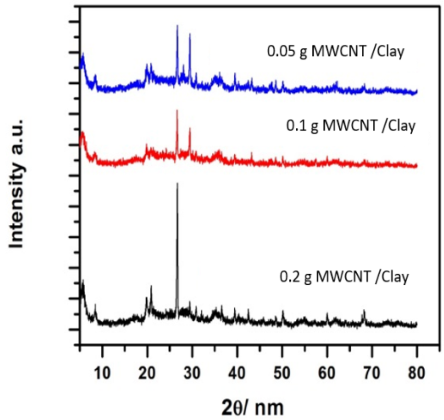


(**E**)



(**F**)

Figure 3. Field emission scanning electron microscopy (FESEM) image of multi-walled carbon nanotube (MWCNT)/clay (bentonite) (**A**) before and (**B**) after adsorption. (**C**) Transmission electron microscopy (TEM) image of MWCNT/clay. (**D**) Energy-dispersive X-ray (EDX) spectroscopy image of MWCNT/clay. (**E**) Thermogravimetric analysis (TGA) image of MWCNT/clay. (**F**) X-ray diffraction (XRD) image of MWCNT/clay.

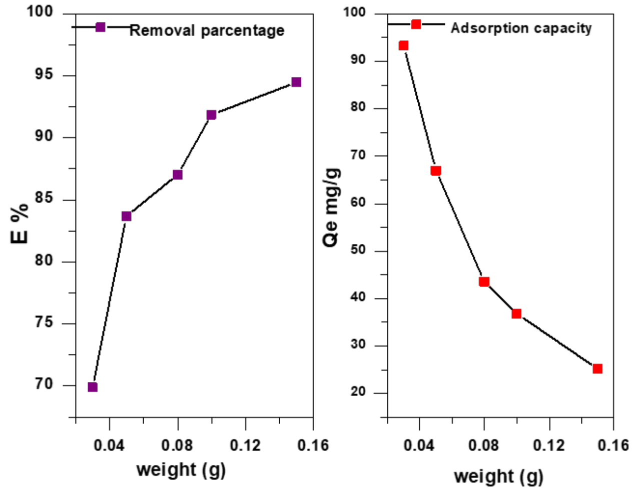


Figure 4. Effect of the weight of multi-walled carbon nanotube (MWCNT)/clay nanocomposites on the surface.

3.3. Effect of Various Concentrations of MB Dye

The influence of different concentrations of MB (methylene blue) dye on the removal percentage of the dye and the quantity of adsorbed MB was investigated. The experiment considered MB concentrations ranging from 10 to 100 mg L^{-1}, and the obtained data revealed distinct trends in the adsorption behavior of MWCNT/clay nanocomposites. For a 100 mL solution with an MB concentration of 40 mg L^{-1}, the percentage removal achieved was 93.65%, with an adsorption capacity of 36.66 mg/g for MWCNT/clay [27]. Interestingly, it was found that the adsorption capacity increased with the concentration of MB dye, while the removal percentage decreased [10]. This inverse relationship between concentration and removal percentage provides key insights into the behavior of the adsorbent and can guide further optimization of the system. Figure 5 illustrates the relationship between the MB dye concentration and both the removal percentage and the adsorption capacity of the MWCNT/clay system.

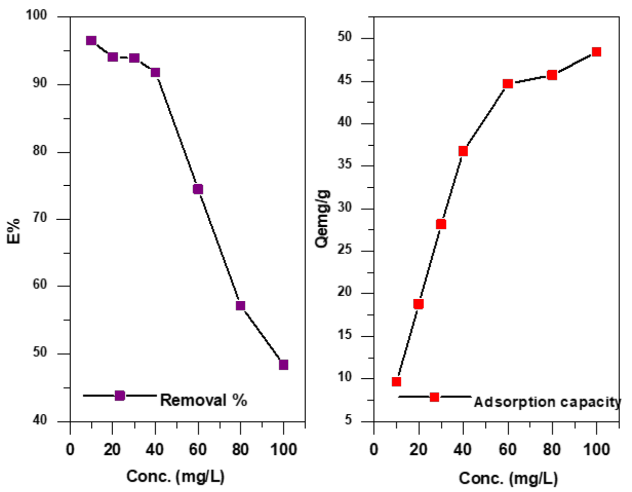


Figure 5. Effect of various concentrations of methylene blue dye on removal percentage and adsorption capacity.

3.4. Effect of Temperature

The impact of solution temperature on the removal efficiency (*E*%) of 0.1 g of MWCNT/clay nanocomposites for the elimination of MB dye was explored.

The experiment was conducted over the course of 1 h at various temperatures, using a dye concentration of 40 mg/L. The removal of MB dye was estimated at different temperatures, and the resulting removal efficiencies were plotted against the corresponding temperatures (Figure 6). Interestingly, the data revealed that the influence of temperature on the removal percentage of MWCNT/clay nanocomposites is relatively minor. Particularly, at temperatures around 45 °C, adverse effects were observed, which could be attributed to the disruption of the physical adsorption of MB dye on the studied surface. In contrast, within the temperature range of 15 to 25 °C, an increase in removal efficiency was noted. Specifically, these temperatures yielded higher removal percentages, ranging from 96.67% to 92.12% [21]. This discovery suggests an optimal temperature window for the adsorption process, offering valuable guidance for potential applications and further investigations.

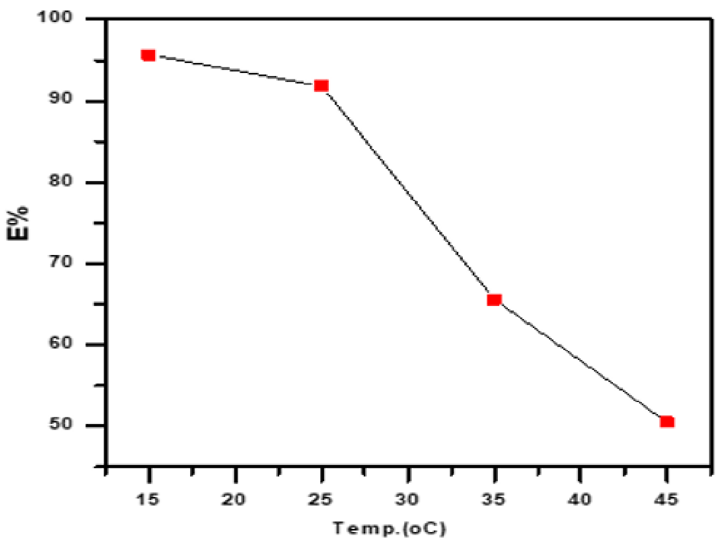


Figure 6. Effect of solution temperature on the removal efficiency of multi-walled carbon nanotube (MWCNT)/clay nanocomposites for methylene blue dye.

3.5. Effect of pH Solution

The pH of the solution is a critical parameter in the adsorption process of MB dye. It affects the adsorption behavior by influencing the surface characteristics of the adsorbent and the dissociation or ionization of the adsorbate.

An experiment was conducted to evaluate the adsorption of MB dye over a pH range of 6 to 10. The results revealed that adsorption decreased appreciably as the pH of the MB dye solution increased to 6. Beyond this point, further increases in pH from 4 to 10 resulted in insignificant changes in adsorption [22]. The higher removal efficiency (*E*%) at acidic pH levels may be attributed to chemical reactions between the encapsulated clay and MB, as well as the electrostatic attraction between the positively charged dye molecules and the negatively charged adsorption sites on the adsorbent. These findings align with previous research and contribute to our understanding of the pH-dependent behavior of the adsorption process, as illustrated in Figure 7 and corroborated by other researchers [28].

3.6. Comparative Study

A comparative study was conducted to evaluate the efficiency of different weights of CNT/clay (bentonite) surfaces, specifically 0.2 g, 0.1 g, and 0.05 g, as adsorbents for MB dye removal. The results revealed that the removal percentage (*E*%) increased in the order of 0.2 g CNT/Clay > 0.1 g CNT/Clay > 0.05 g CNT/Clay. This trend is attributed to the increase in surface area that comes with the increase in the amount of CNT, leading to enhanced removal efficiency [7,8]. The comparative assessment is visually represented in Figure 8.

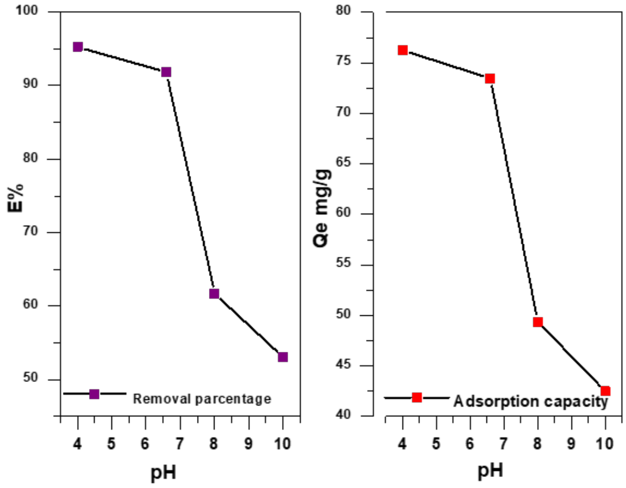


Figure 7. Effect of solution pH on the adsorption of methylene blue dye using multi-walled carbon nanotube (MWCNT)/clay nanocomposites.

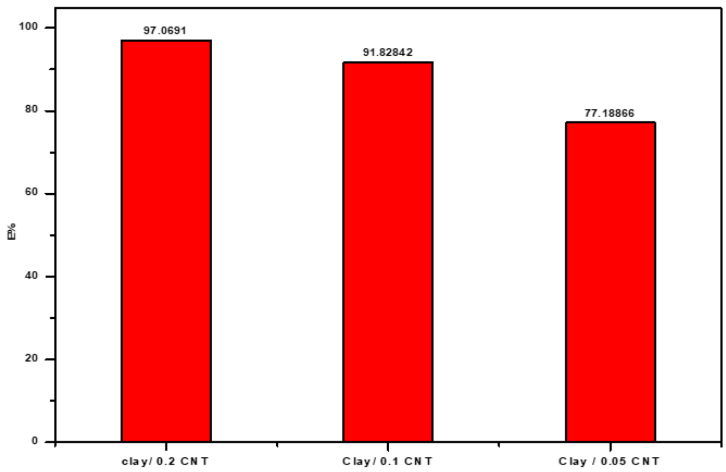


Figure 8. Comparative study among 0.2 g carbon nanotube (CNT)/clay (bentonite), 0.1 g CNT/clay, and 0.05 g CNT/clay surfaces as adsorbents for methylene blue dye removal.

4. Conclusions

The comprehensive study on the characterization and effects of MWCNT/clay nanocomposites on MB dye removal offers significant insights into the applications of these materials in wastewater treatment. Through various techniques, such as FESEM, TEM, EDX, TGA, and XRD, the distinct properties and behaviors of the nanocomposites were effectively characterized. The removal efficiency of the MWCNT/clay nanocomposites was found to be influenced by several factors, including the weight of the composite, the concentration of MB dye, solution temperature, and pH. An increase in weight and concentration of MB dye was shown to enhance removal efficiency, while the temperature effect was found to be optimal within the range of 15–25 °C. Acidic pH was also identified as more favorable for adsorption. Furthermore, the comparative study revealed the correlation between the amount of CNT in the nanocomposites and the removal efficiency, with a higher CNT content leading to better results. This investigation contributes valuable knowledge to the ongoing research in the field and demonstrates the promising potential of MWCNT/clay nanocomposites for efficient and sustainable dye removal. Future work may focus on optimizing the fabrication process and exploring the potential of these nanocomposites in various industrial applications, considering economic and ecological aspects.

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