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RESEARCH PAPER

Role of Carrageenan and Health Approach for Adsorption of Safranin-T Dye from Aqueous Solution by Using Polymer/CNT Surface

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ABSTRACT

A new hydrogel composite (κ-carrageenan-grafted poly(acrylic acid-co-itaconic acid)/carbon nanotube (κC-g-poly(AAc-co-IA)/CNT)) was prepared through free radical graft polymerization in this work. The hydrogel network was prepared by copolymerizing acrylic acid (Ac) and itaconic acid (IA) onto the biopolymer matrix κ-carrageenan and by introducing nanofillers (like carbon nanotubes (CNTs)) for improved structural and functional characteristics. Free radical initiation was employed carefully to support effective grafting and homogeneous dispersion CNTs in the hydrogel network. To optimize the hydrogel formulation, various concentrations of CNTs, κ-carrageenan, AC, and IA were systematically investigated. The effect of these variations on the hydrogel's network structure, porosity, and swelling behavior was studied to identify the composition that provides the best performance for dye adsorption applications. The prepared composite was extensively characterized using Field Emission Scanning Electron Microscopy (FESEM), Transmission electron microscopy I, X-ray Diffraction (XRD), and Fourier Transform Infrared Spectroscopy, (FTIR). These analyses revealed a porous structure with diverse functional groups, thermal stability up to 600 °C, and a high specific surface area of 366.857 m^2/g. The reactivation efficiency of the prepared composite surface initially reached 93% after the first regeneration cycle, but decreased to 69% by the fifth cycle. This gradual decline indicates partial loss of active sites over successive uses. However, the presence of carbon nanotubes (CNTs) played a critical and effective role in maintaining regeneration performance by providing a stable, high-surface-area network that supports dye desorption and the recovery of functional groups.

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INTRODUCTION

Water is essential for human life, and water resources are relied on by human beings for drinking, agriculture, industry and daily life. Nevertheless, the fast population growth, urbanisation, incomplete wastewater sanitation and different types of human activities have resulted in heavy pollution of the water resource. Synthetic dyes are one of the major pollutants that have seriously threatened human health and the environment [1, 2]. Dyes can be categorised has per chemistry — azo, anthraquinone, indigoid, xanthene, phthalocyanine, nitrate, nitroso or ionic as anionic, cationic or neutral. There is a high probability of them entering the aquatic environment due to their widespread application in commercial products from textiles to cosmetics and food production [3, 4]. These dyes are potentially hazardous and can cause irritation, skin and eye damage, as well as respiratory system damage. In addition, the majority of dyes are recalcitrant to biodegradation and are so frequently persistent in the environment. This persistence, coupled with their unsafe nature, highlights the need for their successful separation from the water before discharging into the natural water environments [5-7].

Hydrogels are three-dimensional network structures formed in water-rich environments, typically created from hydrophilic polymers. They are widely utilized due to their excellent biocompatibility, bioadhesion, softness, and extensibility. Hydrogels have applications in various fields, including biomedicine—where they are used for 3D cell culture, tissue engineering, wound healing, and smart wearable device sensors—and in the food industry, for purposes such as the delivery of active substances, fat replacement, and food packaging [8, 9]. The mechanisms for hydrogel formation mainly include physical and chemical cross-linking. Chemically cross-linked hydrogels are made by forming dynamic or permanent covalent bonds through methods such as free radical polymerization, enzymatic induction, Schiff base formation, oxime formation, the Diels-Alder “Click” reaction, and Michael addition. These processes can be quite complex and costly. Furthermore, some cross-linking agents used in these reactions may be toxic. Therefore, it is crucial to consider the food safety of any residual cross-linking agents or newly formed compounds. On the other hand, hydrogels that are physically cross-linked through hydrophobic interactions, hydrogen bonds, ionic bonds, and electrostatic interactions can avoid these drawbacks and thus play a more significant role in the food industry. The physical and chemical characteristics of hydrogels, such as gel

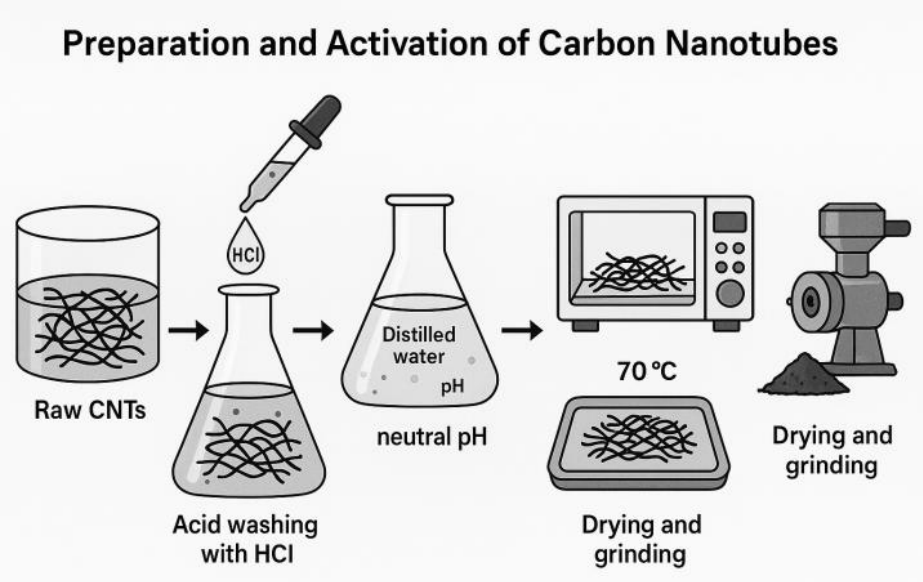

Fig. 1. Preparation and Activation of Carbon Nanotubes (CNTs).

strength, swelling degree, rheological properties, thermal stability, crystal structure, and secondary structure, are usually investigated to assess their performance [10-13].

MATERIALS AND METHODS

Preparation and Activation of Carbon Nanotubes (CNTs)

Carbon nanotubes (CNTs) used in this study were sourced from industrial manufacturing facilities. Due to the presence of residual oily substances and surface contaminants commonly associated with as-produced CNTs, a purification step was necessary before their application. Initially, the raw CNTs were washed thoroughly with hydrochloric acid (HCl) to remove surface-bound impurities and eliminate any residual metal catalysts or oily substances. This acid treatment also enhanced the surface reactivity of the nanotubes. Following acid washing, the CNTs were repeatedly rinsed with distilled water until a neutral pH was achieved, ensuring the complete removal of excess acid. The washed CNTs were then dried in an oven at 70°C to remove moisture, and subsequently ground into a fine powder using a mechanical grinder to enhance dispersion during composite preparation. The activation of CNTs through acid treatment is a crucial step, as it increases their surface area, introduces functional groups (such as –OH and –COOH), and improves their hydrophilicity. These modifications enhance their compatibility with hydrophilic polymer matrices and improve interfacial bonding, which is essential for the efficient incorporation of CNTs into hydrogel networks and for optimizing their adsorption and mechanical performance as shown in Fig. 1.

Preparation and Characterization of κC-g-poly (AAc-co-IA)/CNT Composites

The hydrogel composite κC-g-poly(acrylic acid-co-itaconic acid)/carbon nanotubes (CNT) was

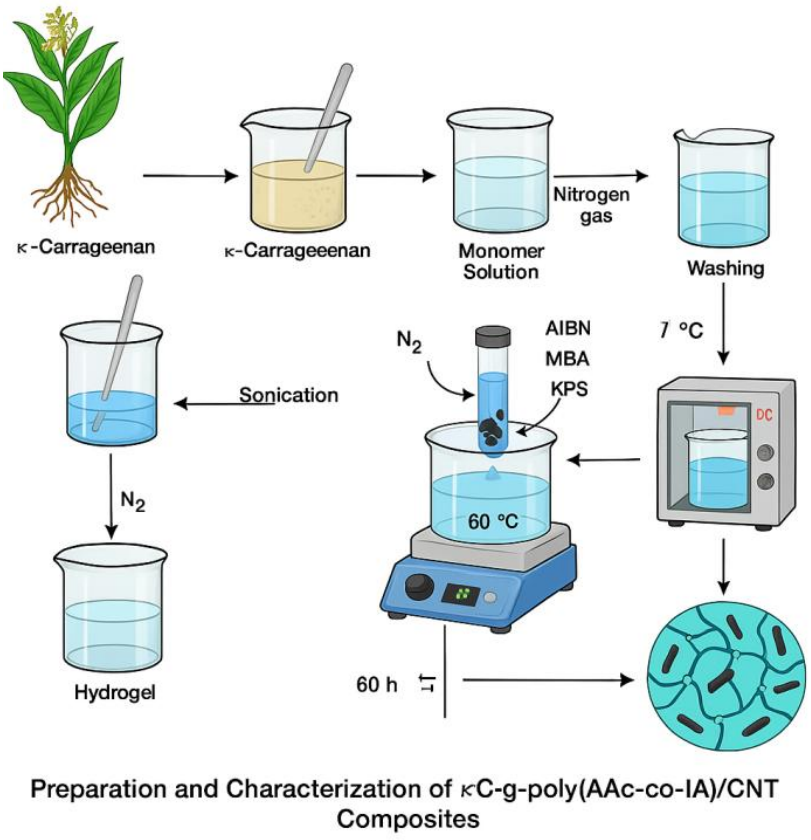

Fig. 2. Preparation of κC-g-poly(AAc-co-IA)/CNT Composites.

synthesized by free radical polymerization. First, 2 g of κ-carrageenan were dissolved in 30 mL of distilled water at 65°C under constant stirring for 30 min to allow the complete dissolution and homogeneity of the polysaccharide base. Using a different beaker, a monomer solution was prepared by dissolving 10 mL of acrylic acid (AAc) and 1.0 g of itaconic acid (IA) in 5 mL of distilled water. The carrageenan solution was subsequently added to the monomer mixture. After that, nitrogen gas was flowed through the reaction mixture to exclude moisture and prevent the pre-oxidation on subsequent polymerisation. A suspension of CNTs was prepared by suspending 0.1 g of CNTs in 50 mL of distilled water. Following sonication, 5mL of CNT suspension was added to the leading reaction solution. Subsequently, 0.5g of stabiliser AIBN was added to the AC/IA monomer solution under stirring (400 rpm) for another 3 min. After that, crosslinker N,N'-methylenebisacrylamide (MBA) and initiator potassium persulfate (KPS) were dropped with the concentration of 0.5g dissolved in 2mL water one by one. Nitrogen gas was purged after each addition to ensure an inert atmosphere. The entire solution was then poured into sealed tubes, which were placed in a 60 °C water bath for 2 hours to allow the polymerisation to finish. The resulting hydrogel was cleaned using distilled water to wash away unreacted monomers and impurities and dried in an oven at 70°C. Fig. 2 shows the process of the synthesis.

RESULTS AND DISCUSSION

SEM Image Interpretation of Hydrogel Loaded with Carbon Nanotubes (CNTs)

The SEM image in Fig. 3 a, aligned network of multi-walled carbon nanotubes (CNTs) with a uniform tubular structure. The nanotubes appear

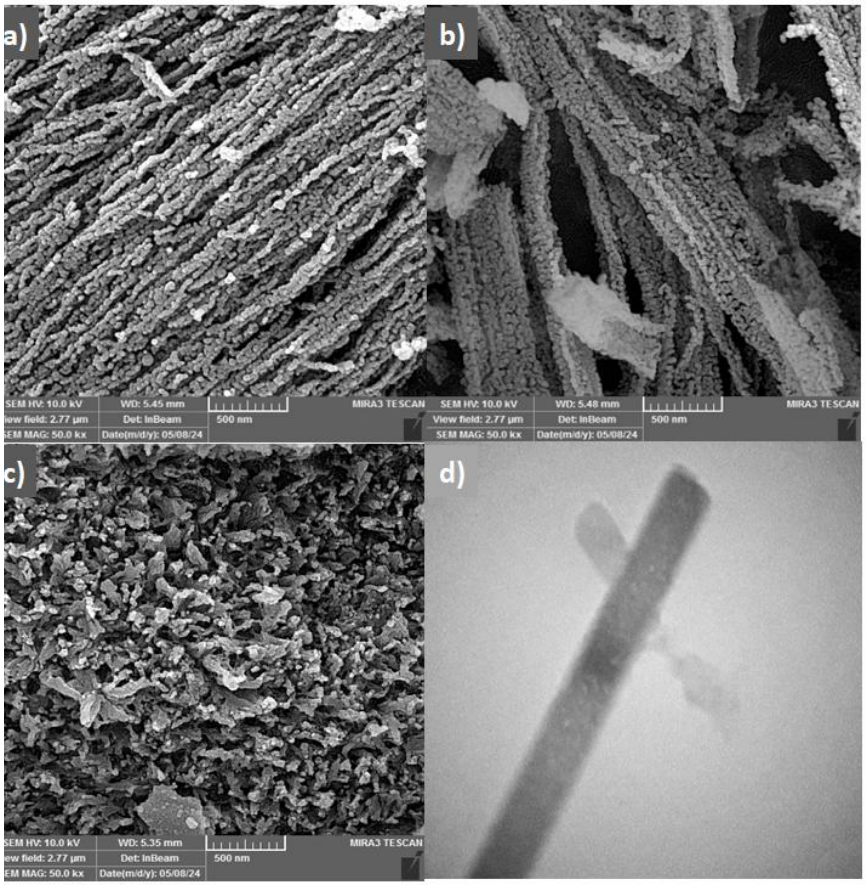

Fig. 3. ESM image of a) CNTs , b) hydrogel before adsorption , c) hydrogel after adsorption safranin-T dye d) TEM image of hydrogel composite.

entangled and closely packed, forming a fibrous, layered surface. The rough texture and attached particles suggest possible surface functionalization or the presence of residual catalysts. The well-defined structure indicates successful synthesis of CNTs with high aspect ratios [14]

The SEM image in Fig. 3b, a fibrous hydrogel matrix embedded with multi-walled carbon nanotubes (CNTs). The CNTs appear as tubular, thread-like structures coated with or surrounded by the hydrogel material. The rough surfaces and attached particles indicate strong interaction and physical integration between the CNTs and the hydrogel. This structure suggests effective dispersion of CNTs within the hydrogel, which enhances its mechanical strength, porosity, and potential for adsorption or conductivity [15].

The SEM image in Fig. 3 c displays the surface morphology of the hydrogel after Safranin-T dye adsorption. The structure appears denser and more compact compared to the unloaded hydrogel, indicating successful dye uptake. The surface shows irregular, aggregated particles likely representing dye molecules adhered to or absorbed within the hydrogel matrix. These morphological changes confirm that the adsorption process altered the surface texture, indicating an effective interaction between the hydrogel and the dye [16, 17].

TEM Image Interpretation of CNT-Loaded Hydrogel

The TEM image in Fig. 3d reveals the structural characteristics of the carbon nanotube/hydrogel composite. The elongated rod-like structures are the CNTs, and the lighter or uneven areas are the hydrogel matrix. The CNTs are generally well-dispersed in the hydrogel, as indicated by this image, although some minor agglomeration is apparent. This distribution facilitates a good interaction between CNTs and hydrogel, which is beneficial for improving the mechanical strength, conductivity, or adsorption performance of the composite. In general, the picture portrays the efficient incorporation of CNTs in the hydrogel microstructure as well as gives an imtroduce into the nanoscale organization of the composite [18-20].

XRD Analysis and Structural Interaction Between Hydrogel and CNTs

The XRD pattern confirmed the presence of graphitic structures in the CNTs. In the composite spectrum (Fig. 4), two broad peaks observed at $2\theta \approx 22^\circ$ and $2\theta \approx 38^\circ$ indicate significant structural

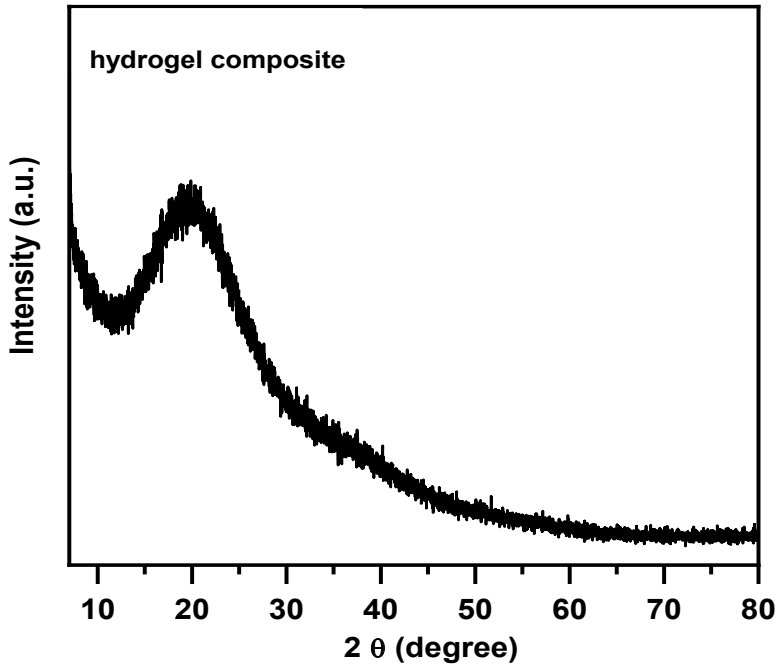

Fig. 4. XRD Analysis and Structural Interaction Between Hydrogel Composite.

interaction between the hydrogel matrix and the CNTs. These wide and irregular peaks reflect a reduction in crystallinity, suggesting that the incorporation of CNTs into the hydrogel disrupted their ordered structure. This loss of crystallinity confirms the successful integration and physical interaction between the hydrogel network and the carbon nanotubes, a key feature in tailoring composite properties for functional applications.

Functional Group Analysis of the Composite Before and After Safranin-T Dye Adsorption

FTIR spectra (Fig. 5) reveal notable changes in the composite before and after Safranin-T adsorption. A broad absorption band in the 3150–3600 cm^{-1} range (black line) shifted after dye interaction (blue line), indicating hydrogen bonding between dye molecules and hydroxyl (O–H) groups on the composite surface. Additionally, a decrease in intensity at 1680 cm^{-1} suggests interaction of carboxyl (C=O) groups with the dye, further confirming chemical bonding. Shifts and intensity reductions in the 1500–1600 cm^{-1} region indicate perturbation of aromatic C=C bonds, supporting π–π interactions between the dye and the composite. Moreover, spectral changes in the 500–1000 cm^{-1} range (blue line) suggest adsorption onto the dye's cyclic framework, highlighting interactions with the modified CNTs. Overall, spectrum C confirms successful adsorption of Safranin-T onto the composite, primarily through hydrogen bonding and π–π stacking mechanisms, as evidenced by distinct spectral shifts [21].

Effect of Carrageenan (Kc) Content on Swelling Efficiency (PS%)

Fig. 6a shows the influence of increasing carrageenan (Kc) weight on the dye swelling efficiency (PS%) of the hydrogel. As the amount of Kc increases from 0.5 to 2.5 wt%, the adsorption efficiency significantly improves, reaching a maximum at around 2.0–2.5 wt%. This indicates that carrageenan enhances the hydrogel's

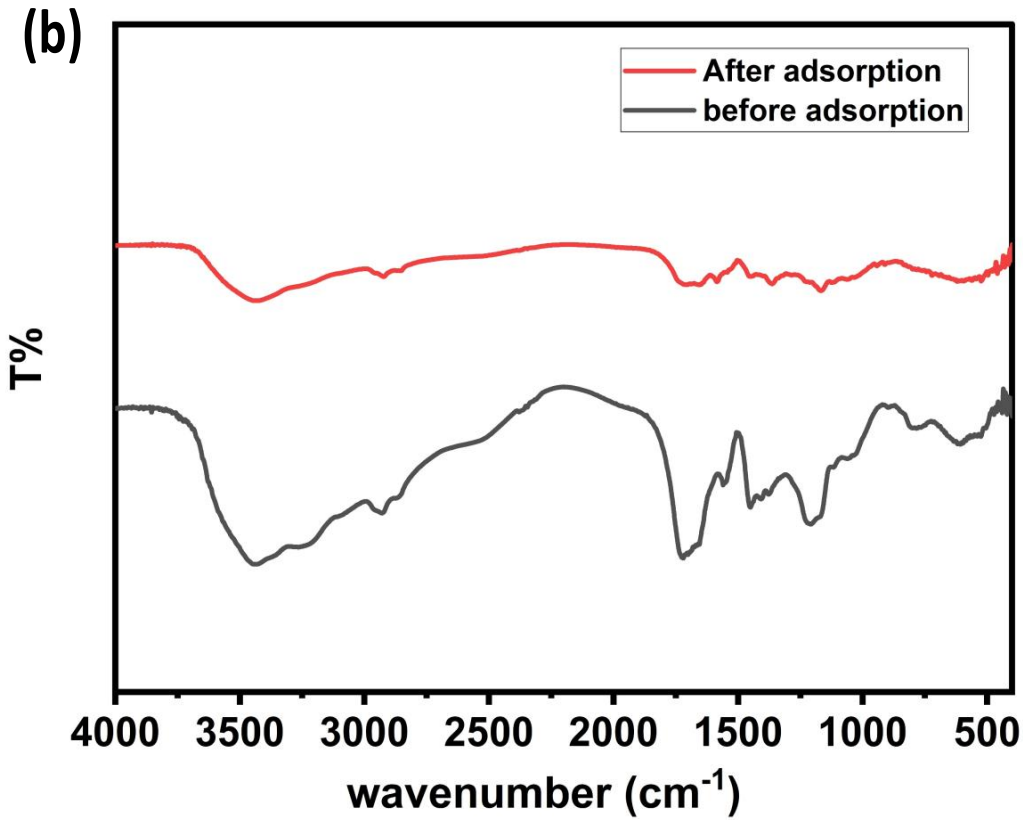

Fig. 5. FTIR of hydrogel before and after adsorption.

adsorption capacity due to its hydrophilic nature and functional groups that interact with dye molecules. However, beyond 2.5 wt%, a decline in PS% is observed, possibly due to excess carrageenan causing structural densification or reduced porosity, which limits dye diffusion and binding [22].

Effect of CNT Loading on Swelling Efficiency (PS%)

Fig. 6b illustrates the impact of increasing CNT content on the dye removal efficiency (PS%) of the hydrogel. As CNT loading rises from 0.01 to 0.04 wt%, the adsorption efficiency significantly improves, reaching an optimal performance at approximately 0.04–0.05 wt%. This enhancement is attributed to the high surface area and active sites provided by CNTs, which facilitate the adsorption of dyes. However, further increases in CNT content beyond 0.05 wt% lead to a sharp decline in adsorption efficiency. This may result from CNT agglomeration at higher concentrations, which reduces their effective surface area and disrupts the hydrogel network, limiting dye accessibility [23, 24].

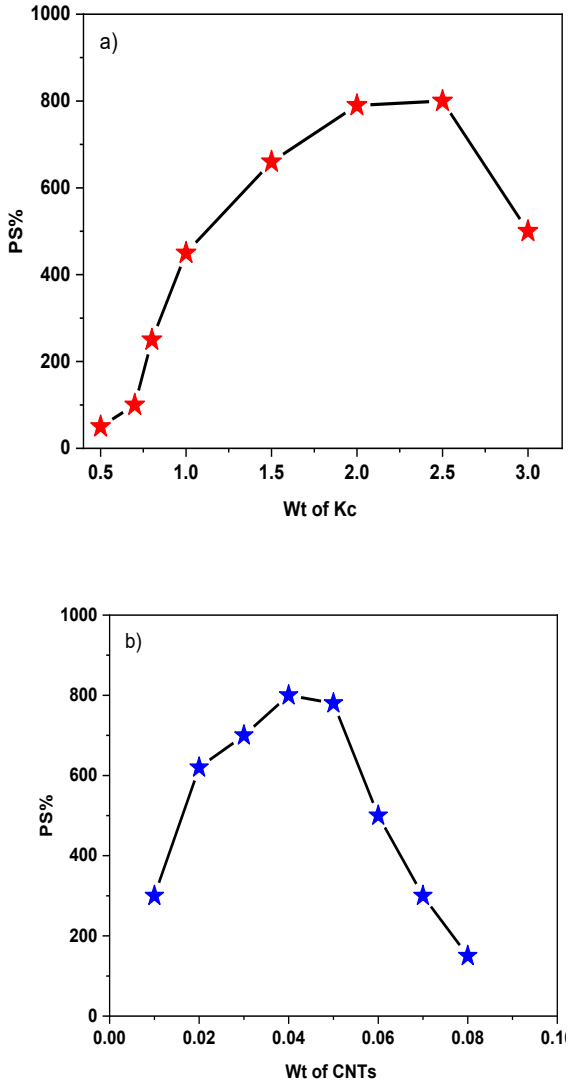

Fig. 6. Effect of a) Carrageenan (Kc) , b) CNTs on the Content of Swelling Efficiency (PS%).

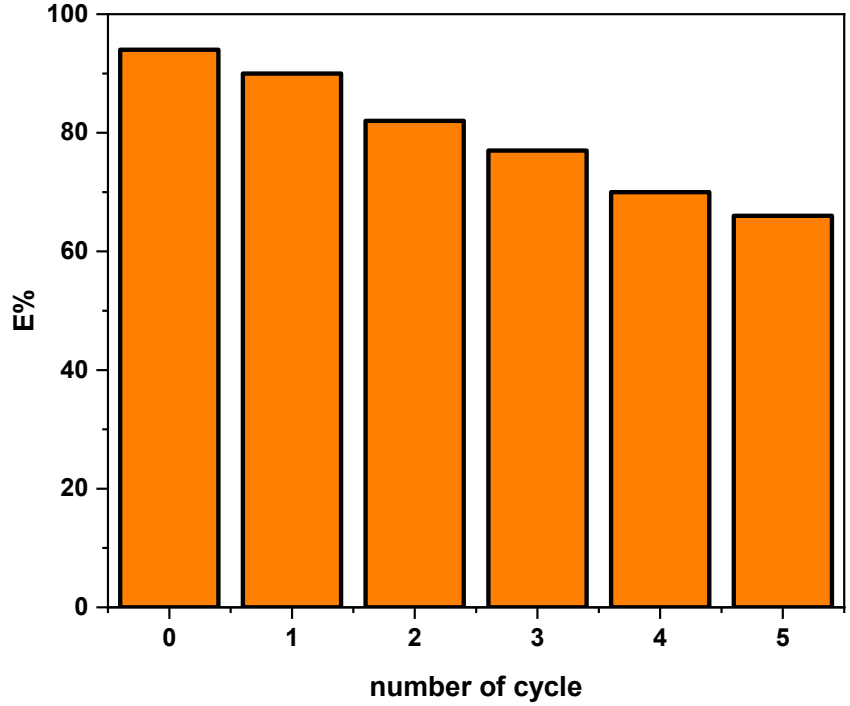

Fig. 7. Reactivation of the Composite Surface After Safranin-T Adsorption. experimental conditions (0.08 gm of the surface with 500 mg/L of the dye under condition 25 °C, pH 7 and equilibrium time 60 mins.).

Reactivation of the Composite Surface After Safranin-T Adsorption

The adsorption process have been done under experimental conditions (0·08 gm of the surface with 500 mg/L of the dye under condition 25 °C, pH 7 and equilibrium time 60 mins.). The surface reactivation of the composite following Safranin-T dye adsorption was evaluated using 0·1 M HCl through five sequential washing cycles. In each cycle, 0·5 g of the composite was mixed with 10 mL of dye solution and stirred at 25°C for 1 hour. The mixture was then centrifuged at 3000 rpm for 10 minutes, and the absorbance of the supernatant was measured using UV-Vis spectrophotometry. The solid residue was dried at 70°C before reactivation [25].

Reactivation involved stirring the dried residue with 20 mL of 0·1 M HCl for 2 hours. This acid washing step was repeated five times with fresh HCl portions. Afterwards, the composite was rinsed with deionized water until neutral pH and dried for reuse in a new adsorption cycle, following the same procedure. Results (Fig.7) indicated that acidic treatment effectively regenerated the composite surface by removing adsorbed dye, restoring functional groups, and converting COO^- groups to COOH. This disruption of electrostatic interactions facilitated the desorption of the dye. The CNTs-COOH composite exhibited excellent regeneration performance due to its multilayer structure and high density of active sites, supporting its potential for repeated adsorption applications [26-28].

CONCLUSION

In this study, a novel κ-carrageenan-g-poly(acrylic acid-co-itaconic acid)/carbon nanotube (κC-g-poly(AAc-co-IA)/CNT) hydrogel composite was successfully synthesized via free radical polymerization. The incorporation of κ-carrageenan significantly enhanced the hydrogel's dye-swelling efficiency due to its hydrophilic nature and functional groups. Optimal adsorption performance was achieved at 2·0–2·5 wt% carrageenan, while excess loading reduced efficiency due to structural densification. Similarly,

the introduction of CNTs improved adsorption capacity up to an optimal concentration of 0.04–0.05 wt%, beyond which CNT agglomeration negatively impacted the hydrogel network. The composite exhibited excellent regeneration capability through repeated acid washing cycles, maintaining high adsorption performance and restoring functional groups. These findings highlight the composite's potential as a cost-effective, recyclable, and high-performance adsorbent for removing cationic dyes such as Safranin-T from aqueous environments.

CONFLICT OF INTEREST

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

REFERENCE

1. Li Q, Yue Q-Y, Su Y, Gao B-Y, Sun H-J. Equilibrium, thermodynamics and process design to minimize adsorbent amount for the adsorption of acid dyes onto cationic polymer-loaded bentonite. Chem Eng J. 2010;158(3):489-497.
2. Albo Hay Allah MA, Alshamsi HA. Green synthesis of AC/ZnO nanocomposites for adsorptive removal of organic dyes from aqueous solution. Inorg Chem Commun. 2023;157:111415.
3. Adsorption and Removal of pharmaceutical Riboflavin (RF) by Rice husks Activated Carbon. International Journal of Pharmaceutical Research. 2019;11(2).
4. Malek NNA, Jawad AH, Abdulhameed AS, Ismail K, Hameed BH. New magnetic Schiff's base-chitosan-glyoxal/fly ash/Fe_3O_4 biocomposite for the removal of anionic azo dye: An optimized process. Int J Biol Macromol. 2020;146:530-539.
5. Maryudi M, Amelia S, Salamah S. Removal of Methylene Blue of Textile Industry Waste with Activated Carbon using Adsorption Method. Reaktor. 2019;19(4):168-171.
6. Aljeboree AM, Alkaim AF, Al-Dujaili AH. Adsorption isotherm, kinetic modeling and thermodynamics of crystal violet dye on coconut husk-based activated carbon. Desalination and Water Treatment. 2015;53(13):3656-3667.
7. Radoor S, Kandel DR, Park K, Jayakumar A, Karayil J, Lee J. Low-cost and eco-friendly PVA/carrageenan membrane to efficiently remove cationic dyes from water: Isotherms, kinetics, thermodynamics, and regeneration study. Chemosphere. 2024;350:140990.
8. Al-Aidy H, Amdeha E. Green adsorbents based on polyacrylic acid-acrylamide grafted starch hydrogels: the new approach for enhanced adsorption of malachite green dye from aqueous solution. Int J Environ Anal Chem. 2020;101(15):2796-2816.
9. An JK, Wang WB, Wang AQ. Preparation and swelling properties of a pH-sensitive superabsorbent hydrogel based on psyllium gum. Starch - Stärke. 2010;62(10):501-507.
10. Boztepe C, Tosun E, Bilenler T, Sislioglu K. Synthesis and characterization of acrylamide-based copolymeric hydrogel–silver composites: Antimicrobial activities and inhibition kinetics against E. coli. International Journal of Polymeric Materials and Polymeric Biomaterials. 2017;66(18):934-942.
11. Cai Z, Zhou W, Chen W, Huang R, Zhang R, Sheng L, et al. Preparation and properties of cationic starch-carrageenan-sodium alginate hydrogels with pH and temperature sensitivity. Food Chem. 2024;459:140272.
12. Cao L, Li N. Activated-carbon-filled agarose hydrogel as a natural medium for seed germination and seedling growth. Int J Biol Macromol. 2021;177:383-391.
13. Chopra L, Sharma A, Chohan JS, Upadhyay VV, Singh R, Sharma S, et al. Synthesis and characterizations of super adsorbent hydrogel based on biopolymer, Guar Gum-grafted-Poly (hydroxyethyl methacrylate) (Gg-g-Poly (HEMA)) for the removal of Bismarck brown Y dye from aqueous solution. Int J Biol Macromol. 2024;256:128518.
14. Aljeboree AM, Al-Baitai AY, Abdalhadi SM, Alkaim AF. Investigation Study of Removing Methyl Violet Dye From Aqueous Solutions Using Corn-Cob as A Source of Activated Carbon. Egyptian Journal of Chemistry. 2021;0(0):0-0.
15. Irfan J, Ali A, Hussain MA, Haseeb MT, Alsahli TG, Naeem-ul-Hassan M, et al. A superabsorbent and pH-responsive copolymer-hydrogel based on acemannan from Aloe vera (Aloe barbadensis M.): A smart material for drug delivery. Int J Biol Macromol. 2024;270:132306.
16. Abdul-Reda Hussein U, Abd S, Al-Mashhadani ZI, M. Abid F, M. Aljeboree A, F. Alkaim A. Sustainable removal of amoxicillin and tetracycline from aqueous media using Pine-Leaf derived activated carbon: Adsorption performance and regeneration potential. Applied Chemical Engineering. 2025;8(2).
17. Kaith BS, Jindal R, Mittal H, Kumar K. Synthesis, characterization, and swelling behavior evaluation of hydrogels based on Gum ghatti and acrylamide for selective absorption of saline from different petroleum fraction–saline emulsions. J Appl Polym Sci. 2011;124(3):2037-2047.
18. Khan S, Rahman NU, Alam S, Zahoor M, Shah LA, Umar MN, et al. Synthesis of Poly(GG-co-AAm-co-MAA), a Terpolymer Hydrogel for the Removal of Methyl Violet and Fuchsin Basic Dyes from Aqueous Solution. ACS Omega. 2024.
19. Kumari P, Kaushal D, Chauhan V, Shandilya P, Kumar M. Synthesis of gum acacia-cl-acrylic acid-co-itaconic acid hydrogels for efficient removal of toxic dye rhodamine-B: A step for sustainable environment. Int J Biol Macromol. 2025;292:139296.
20. Aljeboree AM, Hasan IT, Al-Warthan A, Alkaim AF. Preperation of sodium alginate-based SA-g-poly(ITA-co-VBS)/RC hydrogel nanocomposites: And their application towards dye adsorption. Arabian Journal of Chemistry. 2024;17(3):105589.
21. Aljeboree AM, Alkaim AF. Removal of Antibiotic Tetracycline (TCs) from aqueous solutions by using Titanium dioxide (TiO_2) nanoparticles as an alternative material. Journal of Physics: Conference Series. 2019;1294(5):052059.
22. Yang X, Zhang X, Feng X, Xu B, Du C, Zhang E, et al. Novel porous hydrogel beads based on amidoxime modified polymer of intrinsic microporosity for efficient cationic dye removal. Microporous Mesoporous Mater. 2024;377:113218.
23. Mao X, Wang L, Gu S, Duan Y, Zhu Y, Wang C, et al. Synthesis of a three-dimensional network sodium alginate–poly(acrylic acid)/attapulgite hydrogel with good mechanic property and reusability for efficient adsorption of Cu^{2+} and

Pb^{2+}. Environ Chem Lett. 2018;16(2):653-658.
24. Thamer BM, Al-aizari FA, Abdo HS. Activated Carbon-Incorporated Tragacanth Gum Hydrogel Biocomposite: A Promising Adsorbent for Crystal Violet Dye Removal from Aqueous Solutions. Gels. 2023;9(12):959.
25. Wei J, Yan L, Zhang Z, Hu B, Gui W, Cui Y. Carbon nanotube/Chitosan hydrogel for adsorption of acid red 73 in aqueous and soil environments. BMC Chemistry. 2023;17(1).
26. Bakhshi V, Poursadegh H, Amini-Fazl MS, Salari D, Javanbakht S. Synthesis and characterization of bio-nanocomposite hydrogel beads based on magnetic hydroxyapatite and chitosan: a pH-sensitive drug delivery system for potential implantable anticancer platform. Polym Bull. 2023;81(8):7499-7518.
27. Ullah N, Ali Z, Khan AS, Adalat B, Nasrullah A, Khan SB. Preparation and dye adsorption properties of activated carbon/clay/sodium alginate composite hydrogel membranes. RSC Advances. 2024;14(1):211-221.
28. Thombare N, Mishra S, Siddiqui MZ, Jha U, Singh D, Mahajan GR. Design and development of guar gum based novel, superabsorbent and moisture retaining hydrogels for agricultural applications. Carbohydr Polym. 2018;185:169-178.