



Gamma Irradiation-Induced Synthesis of Dextrin/N-Vinylpyrrolidone Biohydrogel for High-Efficiency Adsorptive Removal of Nitrofurantoin and Methylene Blue from Aqueous Media

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Abstract

Biomedical pollutants, including pharmaceutical residues such as nitrofurantoin (NF) and medical dyes such as methylene blue (MB), are increasingly detected in surface water and wastewater, posing environmental and health concerns due to their durability and biological activity. In this study, dextrin/N-vinylpyrrolidone (Dex/NVP) biohydrogel was successfully synthesized by Co-60 gamma irradiation and evaluated as a green adsorbent for the removal of NF and MB from aqueous media. The optimized hydrogel, prepared with 5 wt% dextrin at an irradiation dose of 30 kGy, achieved a gel fraction of $\approx 90\%$. Structural (by FTIR and XRD), morphological (by SEM and AFM), and thermal (by TGA) characterization confirmed the successful formation of a porous, cross-linked hydrogel with a high swelling capacity (780–800%) and good thermal stability. Under the optimum adsorption conditions (pH 3.0, adsorbent dosage 0.05 g, and contact time 420 min), the hydrogel removed approximately 78% of NF and 98% of MB, with maximum Langmuir adsorption capacities of 40.93 and 42.51 mg g⁻¹, respectively. Adsorption kinetics followed the pseudo-second-order model, while the Langmuir isotherm provided the best fit, indicating monolayer adsorption on homogeneous active sites. Frontier molecular orbital analysis further supported the contribution of hydroxyl, carbonyl, and amide functional groups to the adsorption process. These findings demonstrate that the radiation-fabricated Dex/NVP biohydrogel is a sustainable and environmentally friendly adsorbent with promising potential for wastewater treatment applications.

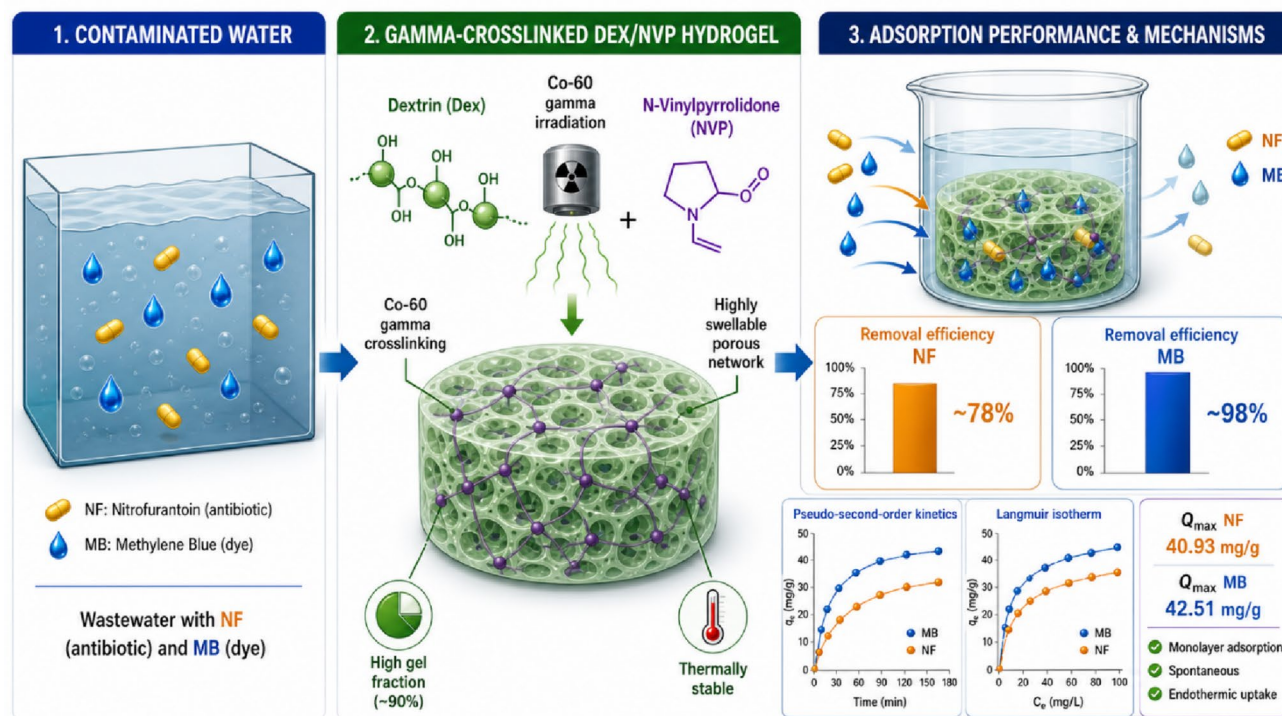
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Graphical Abstract



Keywords Nitrofurantoin · N-vinylpyrrolidone · Methylene blue · Biohydrogel · Adsorptive removal · Gamma irradiation · Wastewater treatment

Introduction

Freshwater is essential for human health, food security, industrial development, and ecological sustainability. Even though water covers 71% of the Earth's surface, less than 1% is freshwater for human and biological use (UN, [55]).

However, rapid industrialization, urbanization, agricultural activities, and increasing pharmaceutical consumption have resulted in the continuous discharge of a wide range of pollutants into aquatic environments, leading to a decline in water quality and threatening both ecosystem and public health [18, 41].

Water pollutants include heavy metals, pharmaceuticals, dyes, pesticides, nutrients, pathogens, and microplastics. Emerging pollutants like antibiotics and synthetic dyes are continuously released into aquatic environments and even remain at low but biologically active concentrations. These substances can enhance antimicrobial resistance, interrupt biological processes, and damage water quality [32, 43].

Among emerging contaminants, antibiotics have attracted considerable attention. Recent studies indicate that around 8500 tons of antibiotics are discharged annually into river systems [12, 20]. Antibiotics promote the creation

of antibiotic-resistant bacteria and facilitate the transfer of resistance genes, causing serious ecological and public health risks. In addition, antibiotic residues can interfere with biogeochemical processes and exert toxic effects on aquatic organisms [4, 19, 22].

Various physical, chemical, and biological technologies have been developed to remove pollutants from aqueous media, including coagulation–flocculation [53], membrane filtration, ion exchange [23], biological treatment [30], advanced oxidation processes [5], photocatalysis [44], electrochemical treatment, and adsorption [49]. Each method offers specific advantages but also suffers from inherent limitations. For example, membrane processes are highly effective but are often limited by membrane fouling and high operational costs, while biological treatments are generally ineffective for non-biodegradable contaminants. Advanced oxidation processes and photocatalysis can efficiently degrade organic pollutants through the generation of reactive oxygen species; however, they typically require high energy input, specialized catalysts, or UV irradiation. García-Ávila et al. [25], Matesun [39], Zhang [56]. In contrast, adsorption is considered one of the most promising and sustainable wastewater treatment technologies owing to its operational simplicity, high removal efficiency, low

energy consumption, cost-effectiveness, and applicability to a wide range of contaminants [17, 40].

Hydrogels are an effective adsorbent substrate owing to their three-dimensional porous networks, which absorb large amounts of water, and functional groups that facilitate strong interactions with polar organic contaminants [36]. These properties often translate to high adsorption capacities and rapid uptake kinetics, especially when hydrogels are derived from biodegradable polymers [8, 29, 57].

Dextrin is a biodegradable, renewable, and inexpensive polysaccharide possessing abundant hydroxyl groups that provide active adsorption sites and excellent hydrophilicity [52]. However, dextrin alone exhibits limited mechanical strength and poor water stability. N-vinylpyrrolidone (NVP) is a hydrophilic vinyl monomer widely used in hydrogel fabrication due to its excellent water solubility and biocompatibility [24]. Upon irradiation, NVP readily undergoes polymerization to form poly(N-vinylpyrrolidone) (PVP), introducing polar lactam groups into the hydrogel network that enhance water uptake and provide additional active sites for interactions with pollutant molecules through hydrogen bonding and dipole–dipole interactions.

Radiation-induced polymerization and crosslinking using gamma irradiation provides an efficient and environmentally friendly approach [9]. This technique eliminates the need for chemical initiators or crosslinkers [33], producing structurally homogeneous, mechanically stable, and sterile polymer networks with tunable physicochemical properties.

Nitrofurantoin, a widely prescribed nitrofurantoin antibiotic, represents a clinically important yet underexplored emerging contaminant. Due to its extensive therapeutic use and high urinary excretion rate, nitrofurantoin is continuously introduced into municipal wastewater streams [34]. Importantly, it exhibits documented genetic toxin and potentially carcinogenic properties, which substantially elevate its environmental and toxicological relevance [3, 11]. Nevertheless, nitrofurantoin has received limited attention in adsorption-based remediation studies, which have largely focused on other antibiotic classes [46].

Methylene blue (MB), a cationic dye widely used in textile fabrication, medical diagnostics and laboratories, represents a prevalent problem in industrial effluent [10, 13]. MB is resistant to biodegradation, inhibits light penetration and photosynthetic activity, and can cause serious health consequences like oxidative stress and methemoglobinemia. While its existence in aquatic environments reduces light penetration, inhibits photosynthesis in aquatic plants and algae, and exerts toxic effects on aquatic organisms, thereby disrupting ecosystem balance [35].

Therefore, its removal before discharge is environmental priority worldwide [42].

In the present study, a biodegradable Dex/NVP biohydrogel was fabricated by Co-60 gamma irradiation using a green, initiator-free, and chemical-crosslinker-free strategy. Unlike previously reported polysaccharide-based adsorbents, the developed hydrogel was designed for the simultaneous removal of nitrofurantoin and methylene blue. Moreover, this study systematically correlates the structural, morphological, swelling, adsorption, kinetic, isotherm, and frontier molecular orbital analyses to provide a comprehensive understanding of the adsorption mechanism. These findings contribute to the development of sustainable radiation-engineered biohydrogels for advanced wastewater treatment.

Materials and Methods

Materials

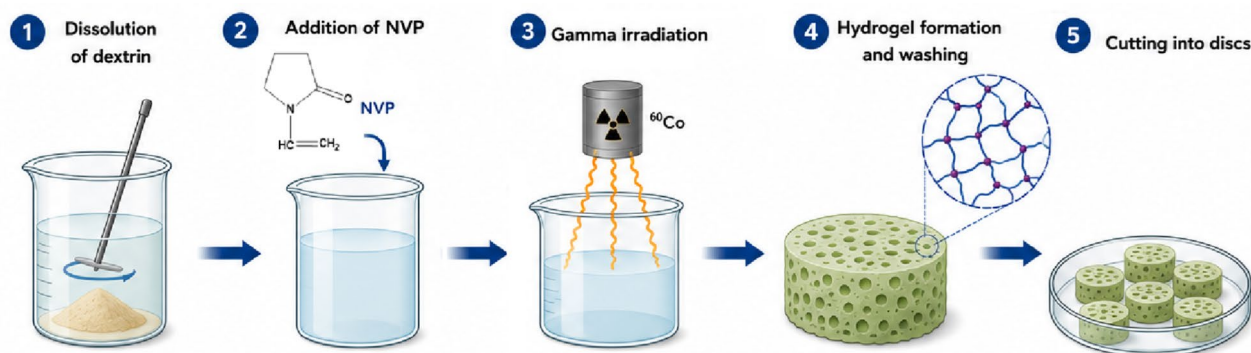
Dextrin and N-Vinyl-2-Pyrrolidone monomer (NVP, 99%) were supplied by Sigma-Aldrich Co. and used as received. Citric acid ($\geq 99.5\%$) and disodium hydrogen phosphate ($\geq 99.0\%$), hydrochloric acid (37%, analytical grade), and Sodium hydroxide ($\geq 98\%$, analytical grade) were purchased from the El-Gomhorea Company for Chemicals, Cairo, Egypt. Nitrofurantoin (NF, pharmaceutical grade) as a drug was obtained from Kahira Pharmaceuticals and Chemical Industries Company, Egypt. Methylene blue (MB) was purchased from El-Nasr Co. for Chemical Industries, Nasr, Egypt, and used as received. Deionized water was used throughout all experiments.

Preparation of Dex/NVP Biohydrogel

The Dextrin/N-vinylpyrrolidone (Dex/NVP) Biohydrogel was set by dissolution of different weights of Dextrin (1, 2, 3, 4, 5, and 6 wt%) in 10.5 mL basic water for 30 min. After mixing and stirring at room temperature, 4.5 mL of NVP was added and mixed for an hour to create a homogeneous mixture. The solutions were placed in glass tubes and exposed to Co-60 γ -ray source at different irradiation doses, and then. The obtained hydrogel rods were sliced into nearly equal smaller disks and drying in air to constant weight, Scheme 1. All samples were submerged in distilled water for 24 h at 70 °C to remove the unreacted component.

Gamma Irradiation

To achieve the desired radiation doses, the materials were exposed to gamma radiation using a cobalt-60 (^{60}Co) gamma cell (Canada) at a dose rate of 0.470 kGy/h, corresponding to approximately 2.13 h per 1 kGy of absorbed



Scheme 1 Schematic illustration of the synthesis of Dex/NVP biohydrogel by gamma irradiation [47]

dose. This source was operated at National Center for Radiation Research and Technology Egyptian Atomic Energy Authority.

Instrumentations and Characterization

FTIR Analysis

FTIR analysis of the DEX/NVP hydrogel was performed using a Vertex 70 spectrometer (Bruker Optik GmbH, Ettlingen, Germany).

Scanning Electron Microscopy (SEM)

The surface morphology of the prepared hydrogel was analyzed using a high-resolution scanning electron microscope (SEM) equipped with a ZEISS LEO 1530 Gemini Optics Lens. The microscope operated at an accelerated voltage of 30 kV, allowing for detailed imaging of the nanoparticle surfaces.

Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) was performed at a heating rate of 10 °C/min in a nitrogen atmosphere using a Shimadzu TGA-30 (Japan).

X-ray Diffraction (XRD)

The crystallographic features were monitored using an XRD model (Rigaku Ultima-IV). The sample was crushed into fine powder, placed onto the sample holder, and scanned in the reflection mode at a 2θ angle over a range from 5° to 90° at a speed of 10° min⁻¹.

Atomic Force Microscopy (AFM)

The surface topographies, height and roughness measurements of the hydrogel before and after metal loading were

investigated through Flex-Axiom AFM (Nanosurf- Switzerland) at room temperature by using tapping mode with a silicon cantilever of resonance 110 kHz.

UV/Vis Spectrophotometer

The quantitative analysis was performed using a Unicam 8625 UV/Visible spectrophotometer over a wavelength range of 195–1100 nm. The instrument has a spectral bandwidth of 2.0 nm, a wavelength accuracy of ± 1.0 nm, and a photometric accuracy of ± 0.005 Abs. NF and MB concentrations were determined at their respective maximum absorption wavelengths of 365 and 664 nm. The calibration equations were $A = 0.00892C + 0.00469$ for NF ($R^2 = 0.9928$) and $A = 0.11976C + 0.08671$ for MB ($R^2 = 0.9953$). Based on the calibration sensitivities and the instrumental absorbance uncertainty, the estimated instrumental detection limits were 1.85 mg/L for NF and 0.138 mg/L for MB, while the corresponding quantification limits were 5.61 and 0.418 mg/L, respectively.

Swelling Properties

A known-weight of dextrin/N-vinylpyrrolidone (Dex/NVP) biohydrogel was immersed in 50 mL distilled water until equilibrium was proven. After wiping the surface water with filter paper, the swollen samples were reweighed.

The swelling properties were tested at different pH levels. Calculating swelling % utilized this equation:

$$\text{Equilibrium swelling (\%)} = \frac{(W_s - W_d)}{W_d} \times 100 \quad (1)$$

where W_s is the weight of the sample after swelling for 24 h, and W_d is the initial weight of the dry sample.

Removal Study

An accurate weight of 0.05 g of dry composite was introduced to 100 mL of a known starting concentration of NF and MB dye solution at pH 3.0, and stirred for a defined time. To calculate the removal percentage the following Eq.

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

where (C_o) and (C_e) denote the initial and equilibrium concentrations of NF or MB dye, respectively, both expressed in mg/L. The residual concentrations of NF and MB dye in the solution were measured using a Unicam 8625 UV/Visible spectrophotometer at their corresponding maximum absorption wavelengths, namely 365 nm for NF and 664 nm for MB dye.

Reusability Study

To investigate the reusability of the (Dex/NVP) for removal of both NF and MB pollutants, an accurate weight of 0.05 g of dry composite was introduced to 100 mL of a known starting concentration of NF and MB dye solution at pH 3, and stirred for a defined time. After this, the composites were submerged in a 0.01 M HNO₃ solution for 30 min and washed over and over until a neutral solution was obtained. This sorption–desorption cycle was repeated five times to assess the reusability of the materials.

Results and Discussions

The aim of the present study is to prepare a green biohydrogel (Dex/NVP), using gamma irradiation as a green method, for the removal of Nitrofurantoin (NF) antibiotic and MB dye. The (Dex/NVP) biohydrogel will be characterized using Fourier Transform Infrared Spectroscopy (FTIR), Thermogravimetric Analysis (TGA), and Scanning Electron Microscopy (SEM) Analysis. The swelling properties were evaluated. Finally, the adsorption study was studied as a function of the solution pH, bioadsorbent weight (g), contact time, and initial concentration.

Characterization and Properties of (Dex/NVP) Biohydrogel

To emphasize the interactions between the components of (Dex/NVP) biohydrogels, and their potential applications; a characterization was carried out investigating their structural, thermal, and functional properties. These analysis employed several advanced techniques, including FTIR,

SEM, TGA, and swelling studies, which provided valuable insights into.

Gelation Behavior of DeX/NVP Biohydrogel as a Function of Dextrin Content and Irradiation Dose

The percent of gel is an important measure for assessing crosslinking efficiency in radiation-induced hydrogels. It indicates a harmony among polymer crosslinking and chain scission. Hydrogels with a high gel fraction are ideal for wastewater treatment applications like removing nitrofurantoin as they maintain resilience, structural integrity and reusability during multiple adsorption-desorption cycles [50].

Figure 1 highlights how the irradiation dose (10–40 kGy) impacts the gel formation in hydrogels at different dextrin levels (1–6%). For each formulation, the gel fraction increased up to 30 kGy before decreasing at 40 kGy. The first increase is due to the higher concentration of free radicals produced at moderate doses, causing crosslinking along polymer chains. The maximum gel fraction observed at 30 kGy indicates that this irradiation dose provides an optimal balance between crosslinking and chain scission (degradation). At 40 kGy, the decline in gel% shows that radiation-induced chain scission resulting in partial decomposition of the polymer structure and less insoluble gel. This pattern appears throughout all dextrin levels, demonstrating that extreme irradiation degrades hydrogel integrity. Figure 2 shows gel percentage as an indicator of dextrin concentration at a specific dosage of 30 kGy. The gel fraction gradually increased as the dextrin content increased from 1% to 5%, reaching a maximum value of approximately 90–92% at 5% dextrin. This behavior suggests that elevated dextrin levels improve crosslinking efficiency through enhancing radical production and intermolecular bonding during irradiation [1]. However, boosting the dextrin content to 6% produced a significant decrease in gel% due to higher solution viscosity, which inhibits radical mobility, favoring chain termination and network defects over effective crosslinking. Therefore, a dextrin content of 5% was identified as the optimum composition for achieving the maximum gel fraction at an irradiation dose of 30 kGy.

Swelling Behavior of Dex/NVP Hydrogel

Swelling tests are crucial to evaluating hydrogel performance since they govern diffusion, functional group exposition, adsorption efficiency, and discharge behavior. Hydrogels developed for environmental applications should have high and stable swelling to effectively interact with aquatic media. As we can see in Fig. 3, during the first 60 min, the swelling percentage rises from about 120% to almost 300%,

Fig. 1 Effect of irradiation dose and Dex content on the gelation %

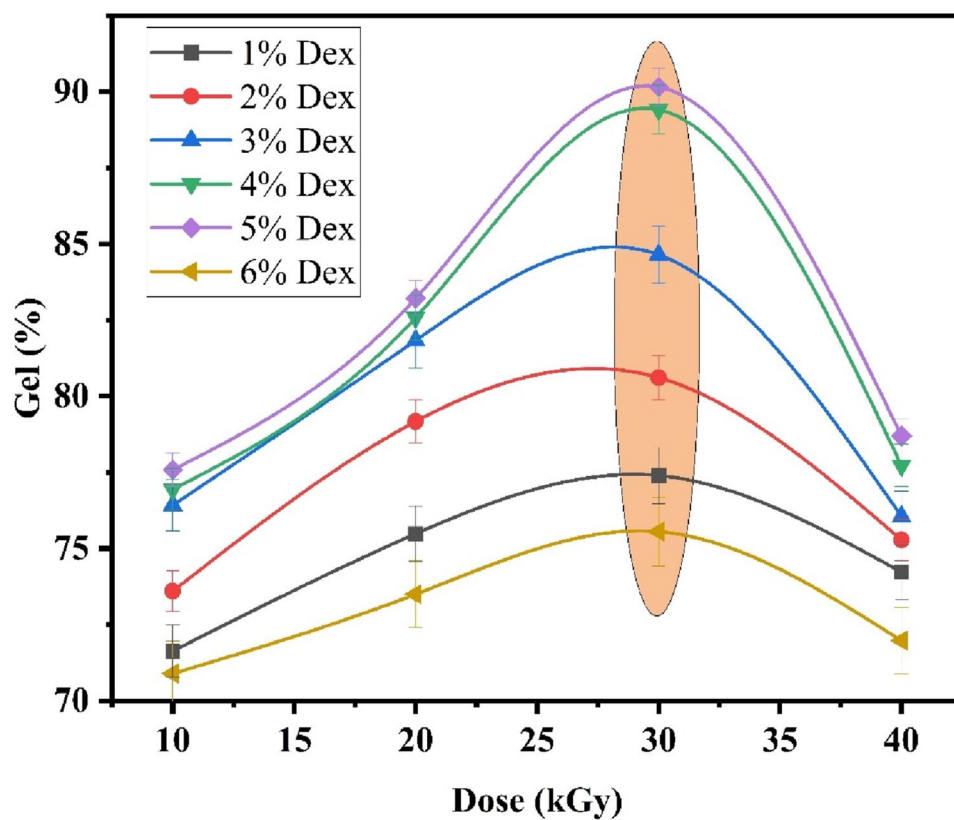


Fig. 2 The impact of Dextrin content (%) on gelation at 30 kGy irradiation dose

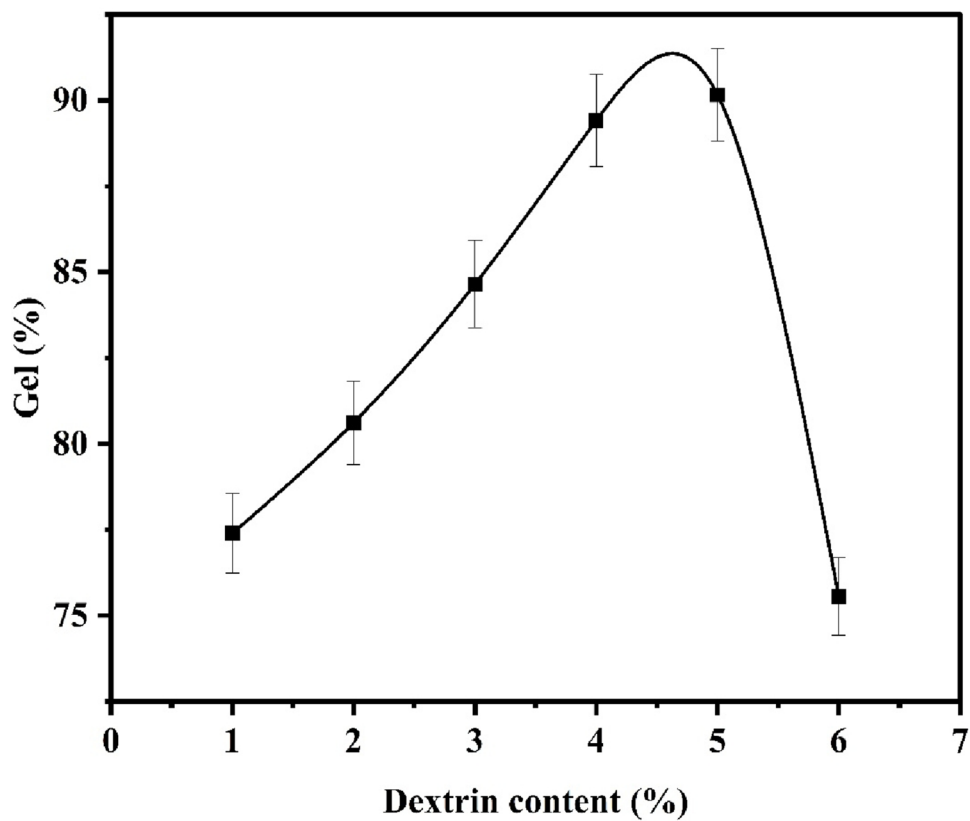
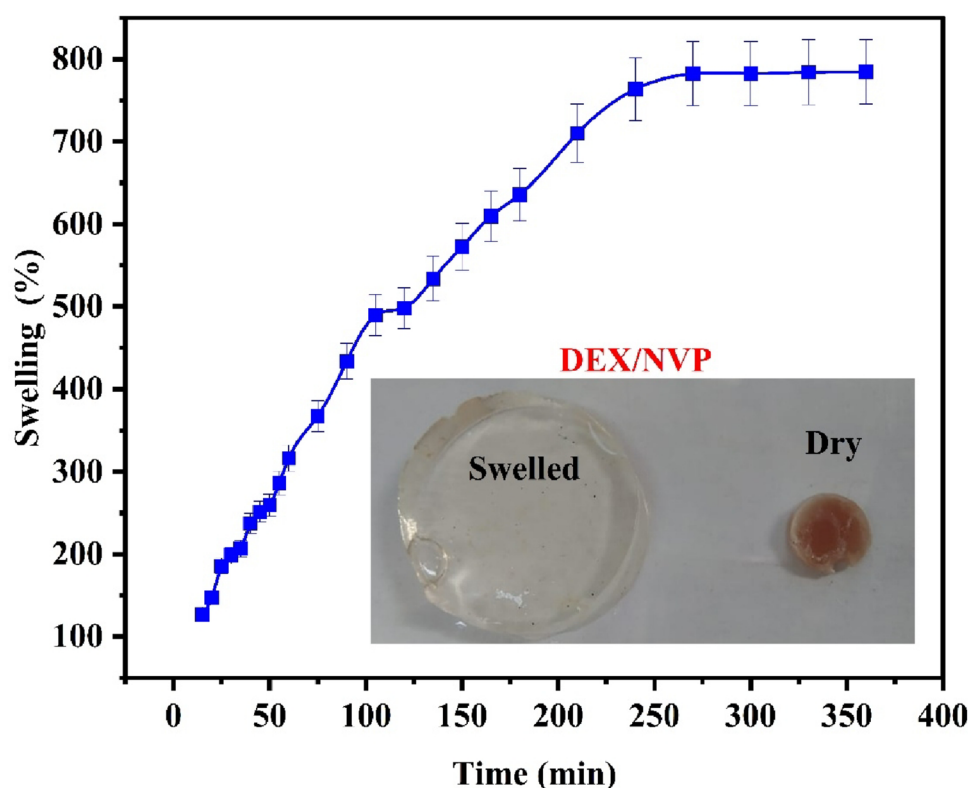


Fig. 3 The swelling percentage of (DEX/NVP) biohydrogel as a function of time

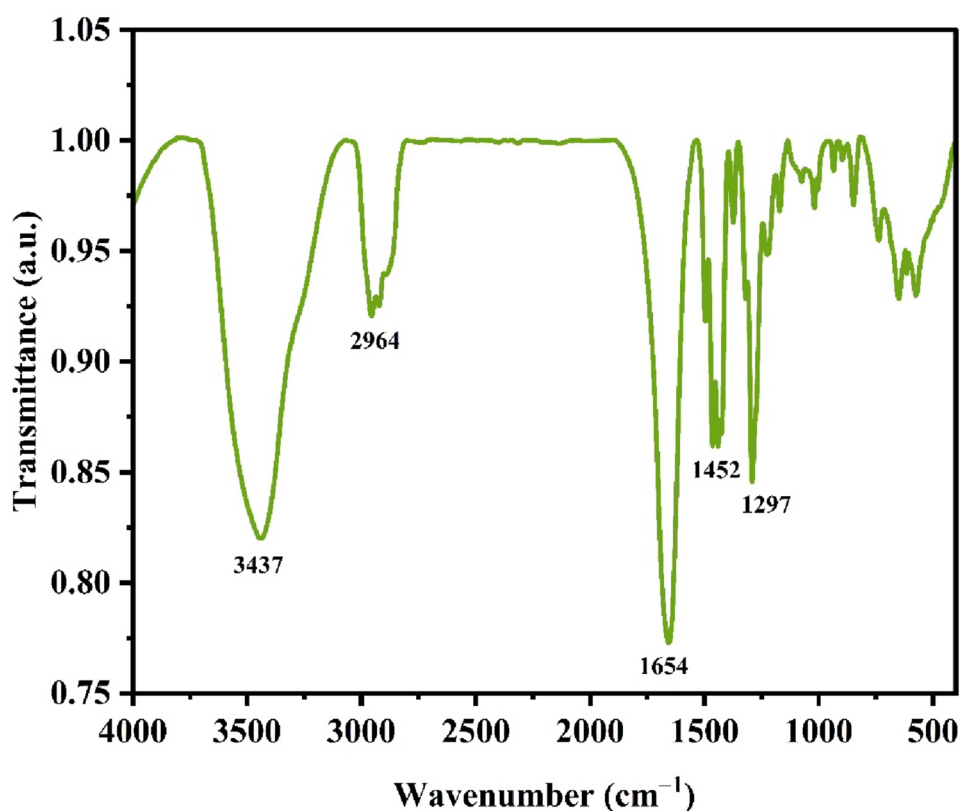


indicating fast water intake. This rapid response is caused by the large number of hydrophilic groups ($-OH$ and $C=O$) in the dextrin and NVP regions, which easily form hydrogen bonds between water molecules. During 60 and 200 min, the swelling rate decreases but the total percentage rises, reaching approximately 700% as water enters deeper into the network and polymer chains loosen. After 240 min, the swelling curve settles at an equilibrium value of 780–800%. This point of stability indicates a well crosslinked and rigid hydrogel that may retain huge quantities of water without splitting apart observed in swelled and dried samples. The swelling behavior of the Dex/NVP hydrogel is comparable to that of previously reported green starch/graphene oxide (GO) hydrogel nanocomposites, which exhibited equilibrium swelling ratios of 400–600%, depending on the GO content and crosslinking density [45]. Dex/NVP exhibits larger equilibrium swelling ($\sim 800\%$) and this is indicated due to dextrin and NVP are extremely hydrophilic, the polymer system is flexible allowing for large chain expansion and there are no hard nanofillers that could limit network swelling. Despite GO based hydrogels have improved mechanical strength and controlled release behaviour, the Dex/NVP hydrogel has a higher water uptake capacity making it ideal for adsorption driven purposes. The swelling time pattern of the Dex/NVP hydrogel demonstrates normal three stage kinetic behaviour. The existence of hydrophilic functional groups causes a quick increase in swelling in the

initial stage, reflecting fast water diffusion into the hydrogel's outer surface. This is followed by a slower, more progressive swelling stage in which polymer chain relaxation and internal network rearrangement regulate water intake. Lastly, the swelling curve approaches a stable plateau. Finally, the swelling curve reached a stable plateau, indicating that the hydrogel had attained equilibrium swelling while maintaining sufficient structural stability.

Fourier Transform Infrared Spectroscopy (FTIR) Analysis

The FTIR spectrum of the Dex/NVP hydrogel illustrated in Fig. 4, and Table 1. It reveals a broad band at $\sim 3437\text{ cm}^{-1}$ which is due to OH stretching vibrations of hydroxyl groups in dextrin, playing a vital part in intra and intermolecular hydrogen bonds where the extent and intensity of this band correspond to a highly hydrated and tightly linked polysaccharide network, a critical feature for the hydrogel's swelling capacity and water uptake activity [21]. The absorption at $\sim 2964\text{ cm}^{-1}$ is from aliphatic C-H stretching vibrations of $-CH$ and $-CH_2$ bonds, showing that the organic framework of dextrin and NVP was intact after gamma irradiation and the ongoing existence of this band illustrates that the irradiation dose prefers crosslinking over severe chain scission, preserving the polymer matrix's integrity in structure [38]. NVP units were effectively inserted into the dextrin

Fig. 4 The FTIR spectrum of Dex/NVP hydrogel**Table 1** FTIR characteristic peaks of Dextrin/NVP hydrogel [21, 38]

Wavenumber (cm ⁻¹)	Assignment
~ 3437	Broad O–H stretching vibration of hydroxyl groups in dextrin (hydrogen bonding)
~ 2964	Aliphatic C–H stretching vibrations (–CH, –CH ₂)
~ 1654	C = O stretching vibration of the amide group (NVP)
~ 1452	C–H bending (scissoring) vibrations
~ 1297	C–N stretching vibration (amide III) / C–O stretching

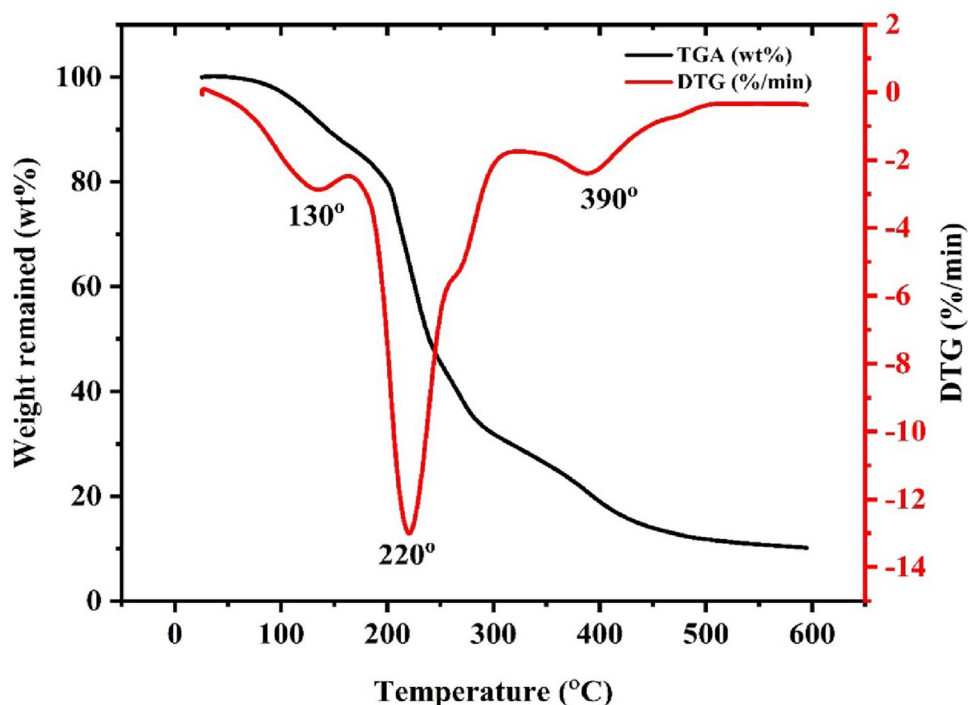
based network, as confirmed by a distinct band at ~ 1654 cm⁻¹ matching to the C = O stretching vibration of the amide group. A carbonyl band inside the large O–H envelope illustrates how NVP carbonyls and dextrin hydroxyls can generate hydrogen bonds and interact dipolar-dipolar where this strengthens the crosslinked structure and increases its rigidity during repeated swelling deswelling cycles. The peak at ~ 1452 cm⁻¹ generated by C–H bending (scissoring) vibrations confirms the existence of saturated aliphatic chains and supports the creation of a compact and flexible polymer structure [21]. The band near ~ 1297 cm⁻¹ is due to C–N stretching (amide III) and C–O stretching showing the existence of amide containing segments. This supports the concept that NVP engages chemically not physically in the system [38]. In general, the FTIR data illustrate that

gamma irradiation efficiently produces a chemically cross-linked Dextrin/NVP hydrogel with dextrin hydroxyl groups and NVP amide/carbonyl activities that are tightly related. The resultant structure is rich in hydrophilic OH, C = O, and C–N groups, that are expected to provide considerable swelling capacity and a diverse set of active sites, providing the material appropriate for aqueous adsorption and ecological remediation purposes.

TGA/DTG of the Prepared Dex/NVP Hydrogel

Thermogravimetric analysis (TGA) must be performed for newly constructed hydrogels since this test indicates thermal stability, free or bound water content, and disintegration pattern, which all these factors have a direct impact on utilization in treatment of water [27]. Understanding the safe temperature range before chemical breakdown is essential. Furthermore, the final residue (char/ash) provides an indirect measure of the integrity of the polymer network and the quantity of inert material left after heating. Figure 5 illustrates the DEX/NVP hydrogel's TGA (black) and DTG (red) curves, which demonstrate a multistep loss of mass behaviour common of hydrated crosslinked polysaccharide/synthetic polymer networks. Phase 1 involves dehydration and loss of physically bonded water (up to ~ 130 °C). Weight loss is low before ~ 130 °C with a weak DTG sign about ~ 130

Fig. 5 Thermogravimetric (TGA, wt%) and derivative thermogravimetric (DTG, %/min) curves of the prepared DEX/NVP hydrogel



°C. This process is primarily responsible for the removal of free and weakly attached water retained within the hydrogel pores and bonds of hydrogen between the hydroxyl groups of dextrin and polar groups in NVP derived sections. This verifies the Dex/NVP structure's extremely hydrated character which promotes pollutant molecules diffusion into the network subsequently adsorption. Phase 2 involves major polymer (about 180–300 °C; DTG maximal $\approx$ 220 °C). The major mass loss takes place in the moderate temperature region, with the highest DTG peak at around 220 °C. This region represents the thermal scission of glycosidic and C-O/C-C bonds in the polysaccharide backbone (dextrin), as well as the dissolution of the organic network produced upon crosslinking [31]. Equivalent main step degradation ranges have been observed for dextrin based systems and crosslinked hydrophilic polymers. Phase 3 shows gradual decomposition and formation of carbon (about 300–450 °C; DTG feature $\approx$ 390 °C). Following the major step, the mass decreases gradually with the DTG curve indicating a larger event about $\approx$ 390 °C degradation. This phase is defined by the decomposition of more stable organic particles as well as the creation of carbon based charcoal. In these parts, NVP/PVP containing materials usually undergo greater temperature decomposition of residual chains and charcoal conversion with mass of residuals (up to 600 °C). At 600 °C, the hydrogel breaks down leaving a low but visible remnant ($\sim$ 10 wt%), signifying a thermally stable carbonaceous section. This demonstrates that Dex/NVP is stable at common regeneration and water treatment temperatures (drying at or below $\sim$ 180 °C), with degradation only occurring at

much greater temperatures than those applied in adsorption desorption cycles.

Morphological Study

The surface morphology of the pristine Dex/NVP hydrogel and the pollutant-loaded hydrogels after adsorption of nitrofurantoin (NF) and methylene blue (MB) was investigated using SEM and AFM, as shown in Figs. 6 and 7.

Scanning Electron Microscopy (SEM) Analysis

The surface morphology and internal microstructure of the gamma irradiated Dex/NVP hydrogel were investigated using scanning electron microscopy (SEM) at Fig. 6 to elucidate the effect of radiation induced crosslinking on the hydrogel architecture [2].

The SEM image of the pristine Dex/NVP hydrogel shows a highly porous and interconnected three-dimensional network. The hydrogel surface contains large cavities with pore diameters ranging approximately from 17 to 81 μm , indicating the successful formation of an open porous structure during gamma irradiation crosslinking. This porous morphology is beneficial for water uptake and pollutant diffusion, as it provides accessible internal channels and active adsorption sites within the hydrogel matrix [45].

After NF adsorption, the morphology of NF@Dex/NVP became more compact compared with the pristine hydrogel. The pores appeared smaller, with measured diameters mostly in the range of 5–29 μm . Some cavities were partially

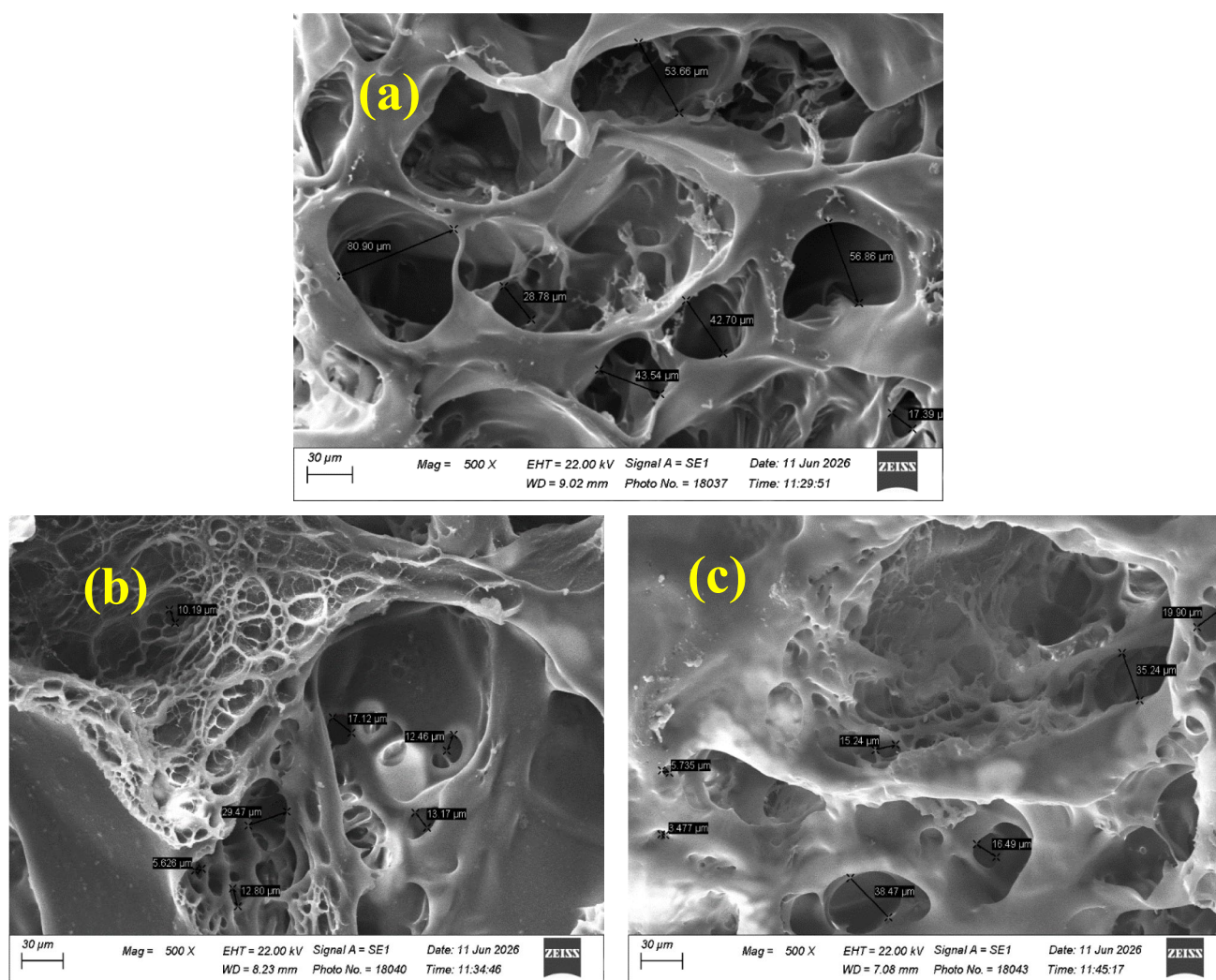


Fig. 6 SEM micrographs of (a) pristine Dex/NVP hydrogel, (b) NF-loaded Dex/NVP hydrogel (NF@Dex/NVP), and (c) MB-loaded Dex/NVP hydrogel (MB@Dex/NVP)

blocked or covered by thin layers, suggesting the successful diffusion and accumulation of NF molecules inside the hydrogel network [28]. This reduction in pore size confirms the interaction between NF and the functional groups of Dex/NVP, such as hydroxyl, carbonyl, and amide groups.

Similarly, after MB adsorption, the MB@Dex/NVP hydrogel retained its porous structure but showed clear morphological changes. The surface became more filled and less open than the pristine sample, with pore sizes approximately ranging from 3.5 to 38 μm. The partial filling and narrowing of pores after MB uptake indicate the successful adsorption of dye molecules inside the hydrogel network. These changes support the high MB removal efficiency, as the porous structure allowed MB molecules to penetrate and interact with the internal adsorption sites [37]. SEM observations confirm that Dex/NVP hydrogel possesses a porous structure favorable for adsorption, while the decrease in

pore size and partial pore filling after NF and MB removal provide strong morphological evidence for pollutant uptake.

AFM Analysis

As we can see in Fig. 7, AFM was used to further evaluate the surface topography of Dex/NVP hydrogel before and after adsorption. The pristine Dex/NVP hydrogel displayed an irregular and rough surface, confirming the heterogeneous nature of the crosslinked polymeric network. The AFM height profile reached approximately 433.7 nm, indicating relatively high surface roughness. Such roughness increases the available surface area and improves contact between the hydrogel and pollutant molecules.

After NF adsorption, the surface topography became less rough, with the height scale decreasing to about 77.9 nm. This decrease suggests that NF molecules covered or filled part of the surface irregularities and pores, producing a

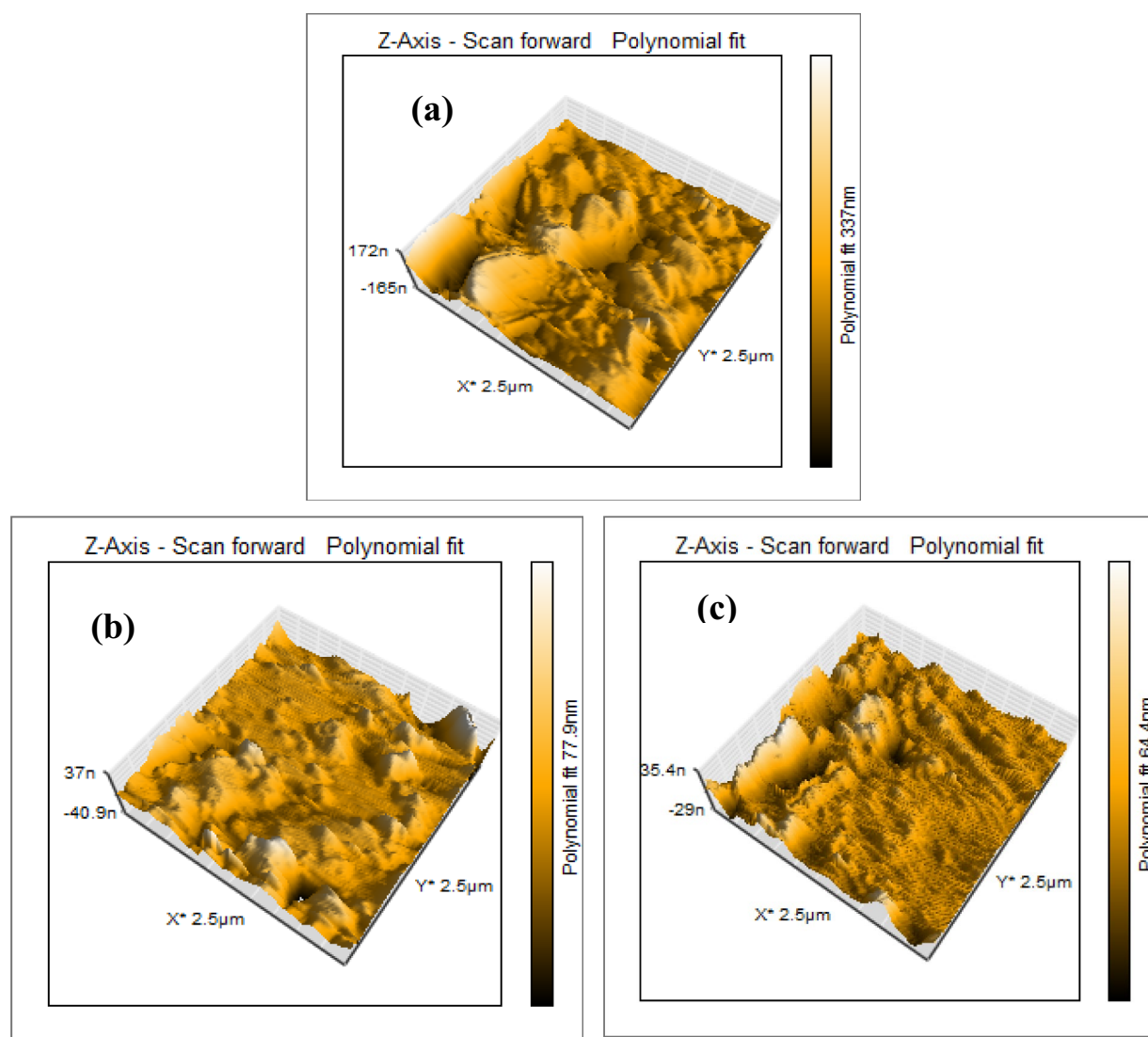


Fig. 7 AFM three-dimensional surface topography of (a) pristine Dex/NVP hydrogel, (b) NF-loaded Dex/NVP hydrogel (NF@Dex/NVP), and (c) MB-loaded Dex/NVP hydrogel (MB@Dex/NVP)

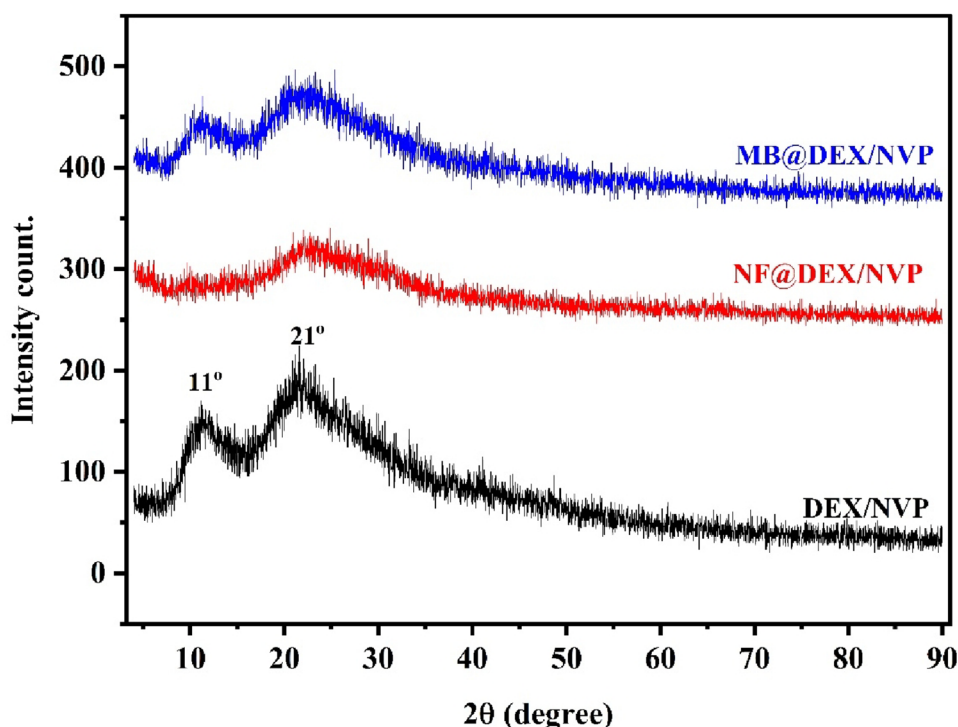
smoother surface. This result agrees with the SEM observation, where partial pore filling was observed after NF uptake.

For MB-loaded Dex/NVP, the AFM image also showed a decrease in surface height compared with the pristine hydrogel, reaching about 66.4 nm. The surface appeared more compact and smoother after MB adsorption, which may be attributed to the deposition and distribution of MB molecules over the hydrogel surface and within the network. The decrease in roughness after MB adsorption confirms the strong interaction between MB molecules and the active sites of Dex/NVP.

X-ray Diffraction (XRD) Analysis

The XRD patterns of Dex/NVP hydrogel before and after adsorption of nitrofurantoin (NF) and methylene blue (MB) are shown in Fig. 8. The pristine Dex/NVP hydrogel exhibited two broad diffraction peaks centered at approximately $2\theta = 11^\circ$ and 21° , indicating the semi-crystalline/amorphous nature of the prepared hydrogel network. The broad peak around 21° is mainly attributed to the polymeric backbone of dextran and N-vinylpyrrolidone, while the weak peak near 11° may be related to ordered regions formed through intermolecular hydrogen bonding within the crosslinked hydrogel structure. The broadness of these peaks confirms that gamma irradiation produced a crosslinked hydrogel with a largely amorphous structure, which is favorable for

Fig. 8 XRD patterns of pristine Dex/NVP hydrogel, NF-loaded Dex/NVP hydrogel (NF@Dex/NVP), and MB-loaded Dex/NVP hydrogel (Dex/NVP–MB) after adsorption



swelling and adsorption because it allows easier diffusion of pollutant molecules into the polymer matrix.

After NF adsorption, the XRD pattern of NF@Dex/NVP showed a noticeable reduction and broadening of the main Dex/NVP diffraction features. No sharp crystalline peaks of nitrofurantoin were clearly observed, suggesting that NF molecules were successfully adsorbed and well dispersed inside the hydrogel network rather than deposited as separate crystalline particles on the surface. The decrease in peak intensity may be attributed to interactions between NF molecules and the active functional groups of Dex/NVP, such as hydroxyl, carbonyl, and amide groups. These interactions can disturb the ordered regions of the hydrogel and increase the amorphous character of the material. Similarly, after MB adsorption, the XRD pattern of Dex/NVP–MB showed broad diffraction bands with reduced crystallinity compared with the pristine hydrogel. The absence of distinct sharp peaks related to MB indicates that methylene blue was incorporated within the hydrogel matrix in a non-crystalline or molecularly dispersed form. The slight change in peak position and intensity after MB adsorption confirms the interaction between MB molecules and the Dex/NVP hydrogel network. These interactions may occur through electrostatic attraction, hydrogen bonding, and possible π – π interactions between the aromatic structure of MB and the polymeric network.

Adsorption Study

The adsorption performance of the Dex/NVP hydrogel toward nitrofurantoin (NF) and methylene blue (MB) was investigated under different experimental conditions, including solution pH, adsorbent dosage, contact time, and initial pollutant concentration, as presented in Fig. 9a–d. These parameters are essential for understanding the adsorption behavior because they influence the surface charge of the hydrogel, the availability of active adsorption sites, the diffusion of pollutant molecules, and the strength of the interactions between the adsorbates and the functional groups within the Dex/NVP hydrogel network. Multi-variable statistical approaches, particularly response surface methodology, can provide additional information concerning interactions among adsorption parameters and facilitate the identification of optimum operating conditions. For example, Saha et al., applied a five-level, three-factor RSM design to optimize adsorbent dosage, contact time, and initial dye concentration during the adsorption of Victoria blue from aqueous solution [48]. In the present study, however, the experimental parameters were investigated individually using a one-factor-at-a-time approach. Therefore, the current dataset does not allow statistically reliable evaluation of interactions among the studied variables through RSM or ANN modeling. Future investigations should employ a structured experimental design, such as a central composite or Box–Behnken design, together with a sufficiently large dataset to enable reliable RSM optimization and

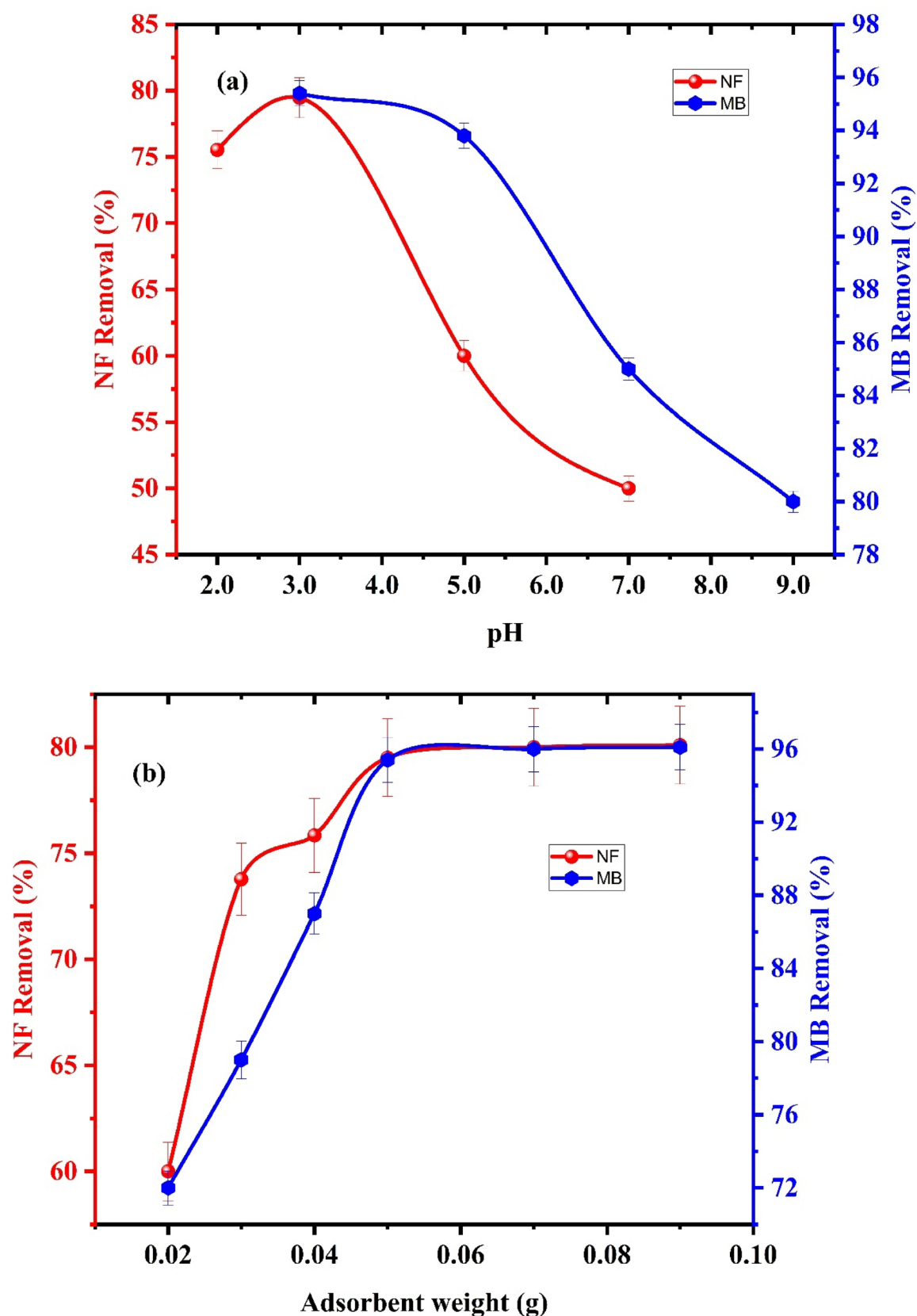


Fig. 9 Effects of solution pH, adsorbent weight, contact time, and initial pollutant concentration on NF and MB removal by the Dex/NVP hydrogel. Optimum conditions were pH 3.0, 0.05 g adsorbent, and 420 min contact time using 100 mL solution. Data are presented as mean $\pm$ SD ($n=3$)

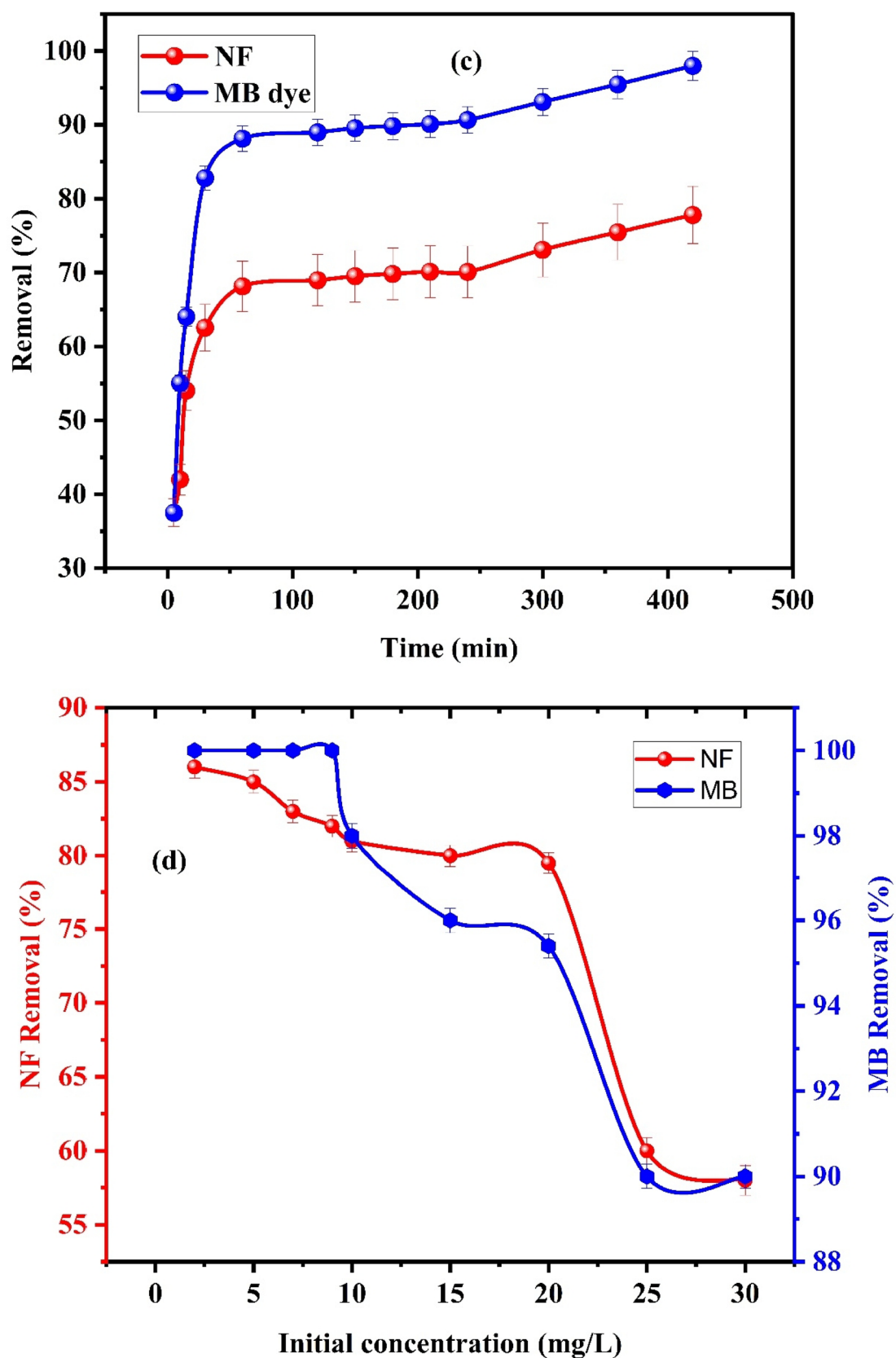


Fig. 9 (continued)

ANN prediction of NF and MB adsorption by the Dex/NVP hydrogel. The solution pH is one of the most effective factors affecting adsorption efficiency, as it controls both the ionization state of the pollutant molecules and the surface charge of the hydrogel. As shown in Fig. 9a, the removal efficiency of NF increased from about 75.7% at pH 2 to a maximum value of approximately 79.6% at pH 3.0. However, further increase in pH caused a significant decrease in NF removal, reaching about 60% at pH 5.0 and nearly 50% at pH 7.0. This decrease may be attributed to the weakening of the interaction between NF molecules and the active functional groups of the Dex/NVP hydrogel under higher pH conditions. In addition, NF showed precipitation behavior at higher pH values, which may interfere with the real adsorption process and affect the reliability of the removal evaluation. Therefore, pH 3.0 was considered the most suitable medium for NF adsorption [54].

For MB dye, the maximum removal efficiency was also observed at pH 3.0, reaching about 95%. The removal efficiency remained relatively high at pH 5.0, around 93–94%, but decreased gradually with further increase in pH, reaching nearly 85% at pH 7.0 and about 80% at pH 9.0. The high adsorption efficiency under acidic conditions may be related to the strong interaction between MB molecules and the functional groups of Dex/NVP hydrogel, such as hydroxyl, carbonyl, and amide groups. The decrease in MB removal under alkaline conditions may be associated with changes in electrostatic interactions and possible competition between hydroxide ions and dye molecules for the available adsorption sites. Based on these findings, pH 3.0 was selected as the optimum pH for the removal of both NF and MB dye.

The effect of adsorbent weight was investigated to determine the suitable hydrogel dosage required for efficient pollutant removal, as shown in Fig. 9b. Increasing the Dex/NVP hydrogel weight from 0.02 to 0.05 g markedly enhanced the removal efficiency of both NF and MB dye. For NF, the removal efficiency increased from about 60% at 0.02 g to nearly 79–80% at 0.05 g. Similarly, MB removal increased from about 72% to approximately 95–96% over the same dosage range. This improvement is mainly attributed to the increased number of available adsorption sites and the larger accessible hydrogel surface area at higher adsorbent weight. Beyond 0.05 g, the removal efficiency showed only a slight change and reached an almost constant value. This plateau behavior indicates that most pollutant molecules present in the solution were already adsorbed under the fixed experimental conditions. Therefore, 0.05 g was selected as the optimum adsorbent weight for the subsequent adsorption experiments.

The influence of contact time on NF and MB dye removal is presented in Fig. 9c. The adsorption process proceeded rapidly during the initial stage, indicating fast diffusion of

pollutant molecules toward the active sites of the hydrogel. For NF, the removal efficiency increased sharply during the first 30–60 min, reaching approximately 68–69%, followed by a slower increase with prolonged contact time. The removal efficiency reached about 77.8% after 420 min. For MB dye, the adsorption was faster and more efficient than NF, increasing rapidly to about 88–90% within 60–120 min and then gradually increasing to approximately 98% after 420 min. The rapid adsorption at the beginning can be explained by the abundance of vacant active sites on the hydrogel surface and within its porous network. At longer contact times, the adsorption rate decreased due to the progressive occupation of these active sites and the reduction in the concentration gradient between the solution and the hydrogel surface. Accordingly, 420 min was selected as the equilibrium contact time for both pollutants.

The effect of initial pollutant concentration on the removal efficiency is shown in Fig. 9d. For NF, the removal efficiency decreased gradually with increasing initial concentration from 2 to 20 mg/L, decreasing from about 86% to nearly 79–80%. A more pronounced decrease was observed at higher concentrations, where the removal efficiency dropped to about 60% at 25 mg/L and approximately 58% at 30 mg/L. This behavior can be explained by the limited number of active adsorption sites on the hydrogel. At low initial concentrations, the available adsorption sites are sufficient to capture most NF molecules. However, as the pollutant concentration increases, the number of NF molecules becomes higher than the available binding sites, resulting in a lower removal percentage [7]. A similar trend was observed for MB dye. The removal efficiency remained very high, about 100%, at low concentrations up to nearly 9 mg/L, then decreased gradually to about 96% at 15 mg/L and 95% at 20 mg/L. At higher concentrations, the removal decreased more clearly, reaching about 90% at 25–30 mg/L. Although the removal percentage decreased with increasing initial MB concentration, the hydrogel still exhibited high adsorption efficiency toward MB compared with NF. This suggests a stronger affinity of MB molecules toward the Dex/NVP hydrogel network, most likely due to more favorable interactions with the hydrogel functional groups and easier diffusion through the porous structure.

Dex/NVP hydrogel exhibited efficient adsorption performance toward both NF and MB dye, with superior removal efficiency for MB dye. The optimum adsorption conditions were pH 3.0, adsorbent weight of 0.05 g, and contact time of 420 min. Under these conditions, the hydrogel achieved approximately 77.8% removal for NF and about 98% removal for MB dye. These results confirm the promising potential of Dex/NVP hydrogel as an effective adsorbent for the treatment of wastewater contaminated with pharmaceutical residues and synthetic dyes.

Fig. 10 Nonlinear fitting of the pseudo-first-order (PFO) and pseudo-second-order (PSO) kinetic models for NF and MB adsorption onto the Dex/NVP hydrogel. Experimental conditions: initial pollutant concentration = 20 mg/L, solution volume = 100 mL, adsorbent weight = 0.05 g, and pH = 3.0. Data are presented as mean $\pm$ SD ($n=3$)

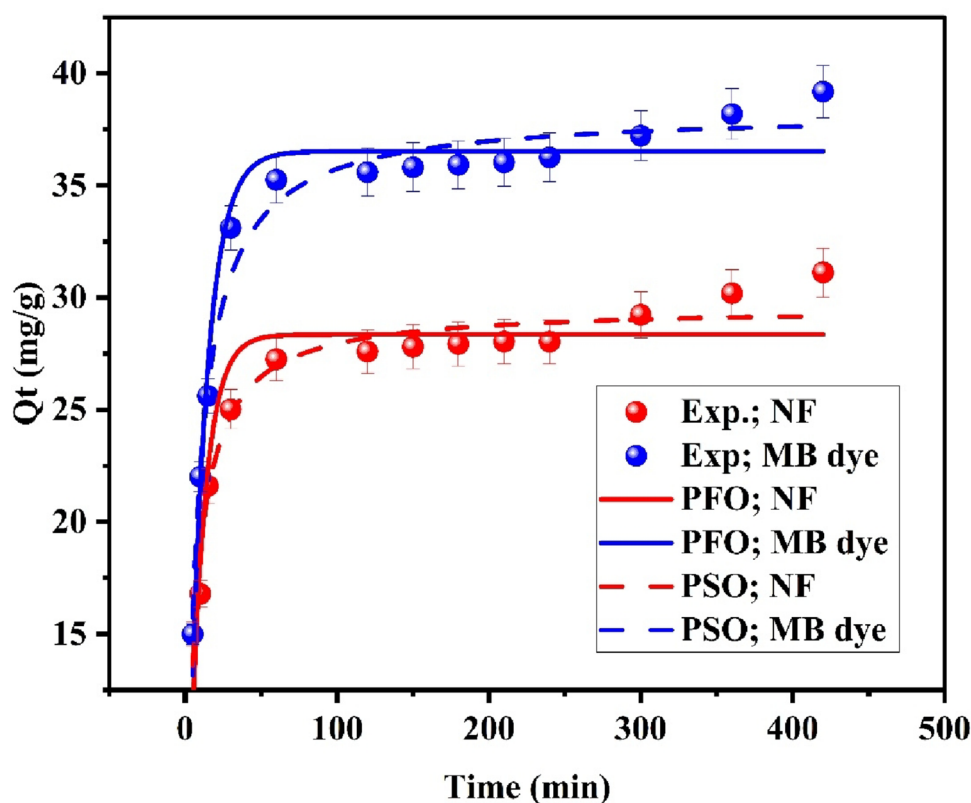


Table 2 Kinetic parameters for NF and MB adsorption onto the Dex/NVP hydrogel under the following conditions: initial pollutant concentration = 20 mg/L, solution volume = 100 mL, adsorbent weight = 0.05 g, and pH = 3.0

Pollutant	Model	q_{exp} mg/g	q (cal.) mg/g	Rate constant	Δq (mg/g)	Reduced Chi-square	(R^2)
NF	PFO	31.12	28.35	$k_1=0.10362 \text{ min}^{-1}$	2.77	3.06526	0.88724
	PSO		29.59	$k_2=0.00574 \text{ g mg}^{-1} \text{ min}^{-1}$	1.53	1.20033	0.95584
MB	PFO	39.19	36.53	$k_1=0.08948 \text{ min}^{-1}$	2.67	1.80202	0.96851
	PSO		38.30	$k_2=0.00365 \text{ g mg}^{-1} \text{ min}^{-1}$	0.89	1.18047	0.97937

Kinetic Study

The adsorption kinetics of NF and MB dye onto Dex/NVP hydrogel are evaluated using pseudo-first-order (PFO; Eq. 3) and pseudo-second-order kinetic (PSO; Eq. 4) models, as shown in Fig. 10 and are summarized in Table 2.

$$q_t = q_e (1 - e^{-k_1 t}) \quad (3)$$

$$q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e t} \quad (4)$$

q_t = Adsorption capacity at time t (mg/g).

q_e = Equilibrium adsorption capacity (mg/g).

k_1 = Pseudo-first-order rate constant (min^{-1}).

k_2 = Pseudo-second-order rate constant ($\text{g mg}^{-1} \text{ min}^{-1}$).

t = Contact time (min).

For NF adsorption, the experimental adsorption capacity was 31.12 mg/g. The PFO model gave a calculated q_e value of 28.35 mg/g, with a difference Δq of 2.77 mg/g and R^2 value of 0.88724. In contrast, the PSO model gave a calculated q_e value of 29.59 mg/g, which is closer to q_{exp} , with a lower Δq of 1.53 mg/g and a higher R^2 value of 0.95584. In addition, the reduced Chi-square value decreased from 3.06526 for PFO to 1.20033 for PSO, confirming the better fitting of the PSO model.

For MB dye adsorption, the experimental adsorption capacity was 39.19 mg/g. The PFO model produced a calculated q_e of 36.53 mg/g, with $\Delta q = 2.67$ mg/g and $R^2 = 0.96851$. The PSO model showed a better agreement with the experimental data, giving $q_{e,cal} = 38.30$ mg/g, a much smaller Δq of 0.89 mg/g, and a higher R^2 value of 0.97937. The reduced Chi-square also decreased from 1.80202 for PFO to 1.18047 for PSO. These results indicate that the PSO model provides a better fit to the experimental

Fig. 11 Nonlinear fitting of the Langmuir and the Freundlich isotherm models for NF and MB adsorption onto the Dex/NVP hydrogel. Experimental conditions: contact time=420 min, solution volume=100 mL, adsorbent weight=0.05 g, and pH=3.0. Data are presented as mean $\pm$ SD (($n=3$))

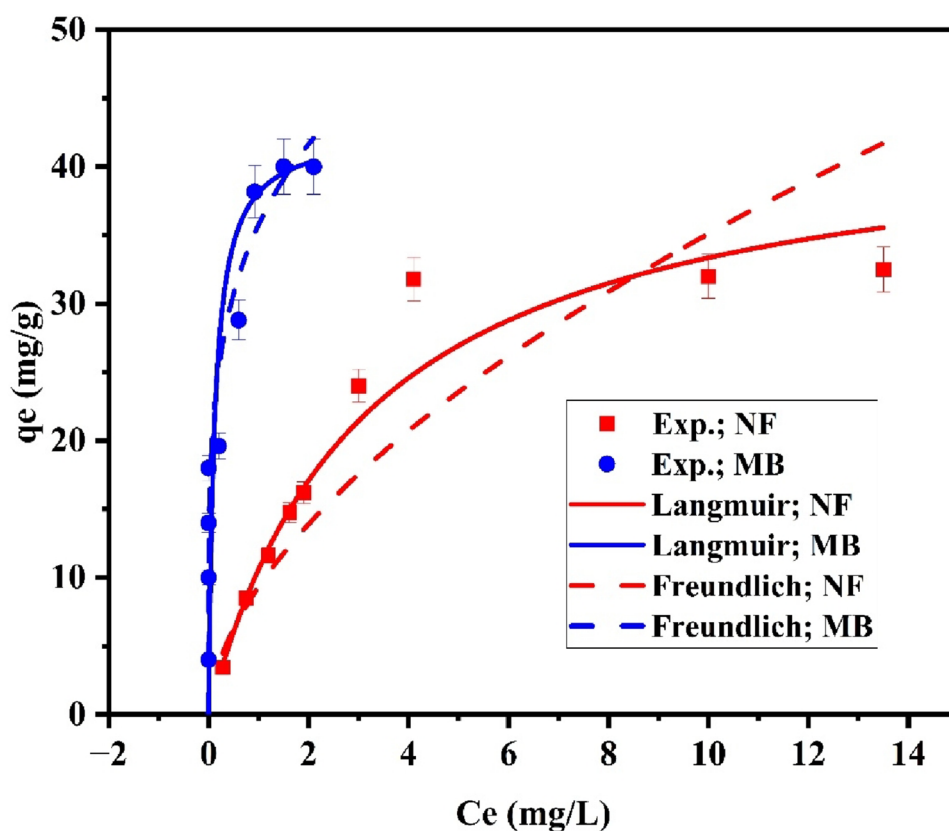


Table 3 Nonlinear isotherm parameters for NF and MB dye adsorption onto Dex/NVP hydrogel

Pollutant	Model	q_{exp} (mg/g)	C_e (mg/L)	q_{cal} (mg/g)	Isotherm constants	Δq (mg/g)	Reduced Chi-square	R^2
NF	Langmuir	32.50	13.50	34.63	$Q_m=40.9279$ mg/g $K_a=0.40715$ L/mg	2.13	7.96185	0.94261
	Freundlich		13.50	36.52	$K_f=12.96023$ $n=2.51201$	4.02	21.26767	0.84670
MB dye	Langmuir	40.00	2.10	40.33	$Q_m=42.51363$ mg/g $K_a=8.81199$ L/mg	0.33	0.03880	0.99982
	Freundlich		2.10	42.85	$K_f=34.53280$ $n=3.43783$	2.85	95.75770	0.54880

data, suggesting that the adsorption process is predominantly governed by chemisorption.

Therefore, the adsorption process is likely controlled mainly by interactions between the pollutant molecules and the active functional groups of the Dex/NVP hydrogel, including hydroxyl, carbonyl, and amide groups. The better fitting of MB dye compared with NF also agrees with its higher removal efficiency and higher adsorption capacity.

Isotherm Study

The adsorption isotherm study was performed to describe the equilibrium interaction between nitrofurantoin (NF) and methylene blue (MB) dye molecules and the active sites of the Dex/NVP hydrogel. The experimental equilibrium data were fitted using the nonlinear Langmuir (Eq. 5) and

Freundlich (Eq. 6) isotherm models, as shown in Fig. 11. The calculated isotherm parameters are summarized in Table 3.

$$q_e = \frac{Q_m K_a C_e}{1 + K_a C_e} \quad (5)$$

$$q_e = K_f C_e^{\frac{1}{n}} \quad (6)$$

q_e = Equilibrium adsorption capacity (mg/g).

Q_m = Maximum monolayer adsorption (mg/g).

K_a = Langmuir adsorption constant (L/mg).

C_e = Equilibrium concentration of adsorbate in solution (mg/L).

K_f = Freundlich constant.

n =Adsorption intensity.

The Langmuir model assumes monolayer adsorption onto a homogeneous adsorbent surface with a finite number of identical adsorption sites. As shown in Table 3; Fig. 11, the Langmuir model provided the best fitting performance for both NF and MB dye adsorption. For NF, the calculated maximum monolayer adsorption capacity (Q_m) was 40.9279 mg/g, while the Langmuir constant (K_a) was 0.40715 L/mg. At the experimental equilibrium point of $C_e = 13.50$ mg/L and $q_{exp} = 32.50$ mg/g, the Langmuir model gave $q_{cal} = 34.63$ mg/g, with a relatively small Δq value of 2.13 mg/g. The corresponding R^2 value of 0.94261 and reduced Chi-square value of 7.96185 indicate a reasonable agreement between the experimental and calculated data.

For MB dye, the Langmuir model showed excellent agreement with the experimental data, giving a very high R^2 value of 0.99982 and a very low reduced Chi-square value of 0.03880. The calculated Q_m value was 42.51363 mg/g and K_a was 8.81199 L/mg. At $C_e = 2.10$ mg/L and $q_{exp} = 40.00$ mg/g, the calculated adsorption capacity was 40.33 mg/g, giving a very small Δq value of 0.33 mg/g. This close agreement confirms the strong suitability of the Langmuir model for describing MB dye adsorption onto the Dex/NVP hydrogel.

The Freundlich model was also applied to evaluate the possibility of adsorption on a heterogeneous surface and multilayer adsorption behavior. For NF, the Freundlich constants were $K_f = 12.96023$ and $n = 2.51201$. Since n is greater than 1, the adsorption of NF can be considered favorable. However, the Freundlich model gave $q_{cal} = 36.52$ mg/g and $\Delta q = 4.02$ mg/g, which is higher than the Δq obtained

from the Langmuir model. In addition, the lower R^2 value of 0.84670 and the higher reduced Chi-square value of 21.27 indicate that the Freundlich model is less suitable for describing NF adsorption.

For MB dye, the Freundlich model gave $K_f = 34.53280$ and $n_f = 3.43783$, suggesting favorable adsorption based on the n_f value. However, the fitting quality was poor compared with the Langmuir model. The calculated q_{cal} value was 42.85 mg/g, giving $\Delta q = 2.85$ mg/g. Moreover, the low R^2 value of 0.54880 and the high reduced Chi-square value of 95.76 demonstrate that the Freundlich model does not adequately represent the equilibrium adsorption behavior of MB dye onto Dex/NVP hydrogel.

Effect of Temperature and Thermodynamic Analysis of NF and MB Adsorption

The influence of temperature on the adsorption of nitrofurantoin (NF) and methylene blue (MB) by the Dex/NVP hydrogel is presented in Fig. 12a. Increasing the temperature from 25 to 45 °C increased the NF removal efficiency from approximately 92.5% to 96.1%, whereas MB removal increased from approximately 98.0% to 99.5%. The positive temperature dependence indicates that adsorption is favored at higher temperature. This behavior may be associated with enhanced molecular mobility, faster diffusion of NF and MB through the swollen porous hydrogel network, and improved accessibility of the hydroxyl, carbonyl, and amide binding sites. The consistently higher removal of MB indicates a greater apparent affinity of the Dex/NVP hydrogel for MB under the investigated conditions.

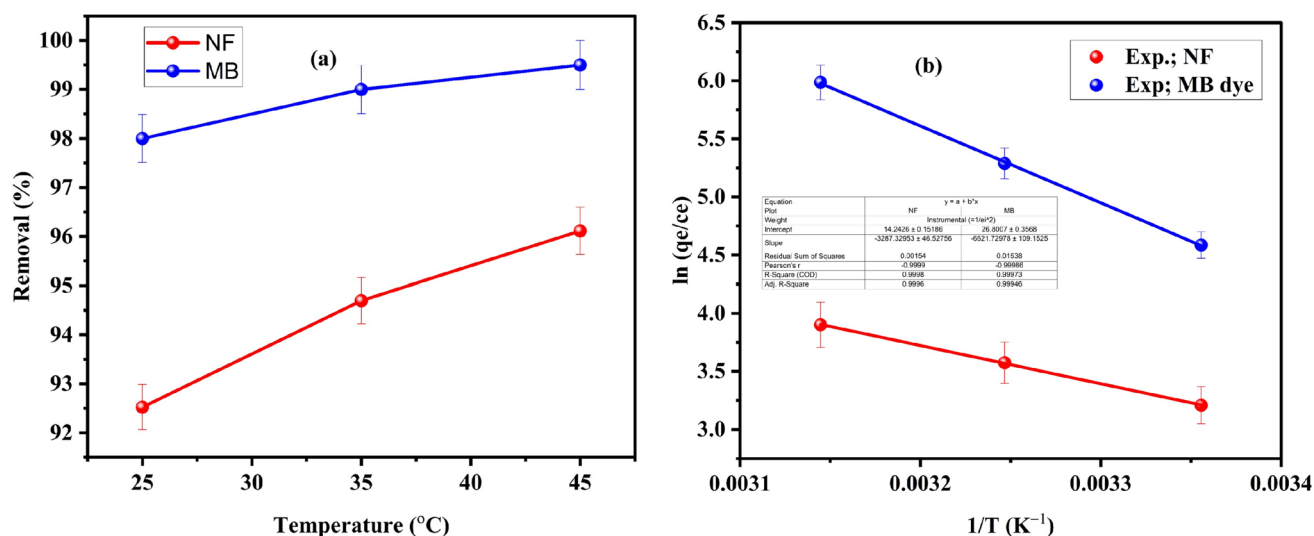


Fig. 12 (a) Effect of temperature on the removal efficiencies of nitrofurantoin (NF) and methylene blue (MB) by the Dex/NVP hydrogel and (b) Van't Hoff plots of $\ln(q_e/C_e)$ versus $1/T$ for NF and MB adsorption.

Experimental conditions: initial pollutant concentration = 20 mg L⁻¹, solution volume = 100 mL, adsorbent weight = 0.05 g, pH = 3.0, and contact time = 24 h. Data are presented as mean ± SD ($n = 3$)

The thermodynamic behavior was evaluated using the Van't Hoff plots of $\ln(q_e/C_e)$ versus $1/T$ shown in Fig. 12b. The linear relationships were described by $\ln(q_e/C_e) = 14.2426 - 3287.3295/T$ for NF and $\ln(q_e/C_e) = 26.8007 - 6621.7298/T$ for MB. The corresponding coefficients of determination were 0.99980 and 0.99973, respectively, indicating excellent linear agreement over the investigated temperature range. Nevertheless, because only three temperatures were examined, these high R^2 values should be interpreted cautiously and additional temperature points would further strengthen the thermodynamic assessment.

Thermodynamic Equations and Assumptions

$$K_e = q_e/C_e$$

$$\ln K_e = -\Delta H^\circ / (RT) + \Delta S^\circ / R$$

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ = -RT \ln K_e$$

where K_e is the apparent equilibrium distribution term, q_e is the equilibrium adsorption capacity, C_e is the equilibrium pollutant concentration, R is the universal gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$), and T is the absolute temperature. The values of ΔH° and ΔS° were determined from the slope ($-\Delta H^\circ/R$) and intercept ($\Delta S^\circ/R$), respectively. The Van't Hoff analysis assumes that equilibrium is attained at each temperature, that the dominant adsorption process does not change over the studied range, and that ΔH° and ΔS° remain approximately constant.

Thermodynamic Parameters

The positive ΔH° values of $27.33 \text{ kJ mol}^{-1}$ for NF and $55.06 \text{ kJ mol}^{-1}$ for MB confirm the endothermic nature of both adsorption processes, in agreement with the increased removal efficiencies observed at higher temperature. The larger ΔH° value for MB indicates that its adsorption is more strongly influenced by temperature. The positive ΔS° values ($118.42 \text{ J mol}^{-1} \text{ K}^{-1}$ for NF and $222.83 \text{ J mol}^{-1} \text{ K}^{-1}$ for MB) indicate increased apparent disorder at the solid–solution interface, which may arise from pollutant uptake accompanied by desolvation and rearrangement of water molecules and hydrogel functional groups [48].

The calculated ΔG° values were negative throughout the studied temperature range and became progressively more negative as the temperature increased. For NF, ΔG° decreased from $-7.97 \text{ kJ mol}^{-1}$ at 298.15 K to $-10.34 \text{ kJ mol}^{-1}$ at 318.15 K , while for MB it decreased from -11.38 to $-15.84 \text{ kJ mol}^{-1}$. These results indicate that adsorption is thermodynamically favorable and that increasing temperature enhances the spontaneity of the process. The more negative ΔG° values for MB are consistent with its higher removal efficiency and stronger apparent affinity for the Dex/NVP hydrogel. Collectively, the thermodynamic findings support an endothermic and favorable adsorption process governed by the combined contribution of diffusion through the porous network and specific interactions with the hydrogel functional groups. Thermodynamic magnitudes alone, however, should not be used as the sole criterion for distinguishing physical from chemical adsorption (Table 4).

Increasing temperature improved the removal of both NF and MB by the Dex/NVP hydrogel. The positive ΔH° and ΔS° values, together with the negative ΔG° values, demonstrate that the adsorption processes are endothermic, thermodynamically favorable, and increasingly spontaneous at higher temperature. MB exhibited more negative ΔG° values and a stronger temperature response than NF, in agreement with its consistently higher removal efficiency. These results further support the suitability of the Dex/NVP hydrogel for the simultaneous removal of pharmaceutical and dye contaminants from aqueous media.

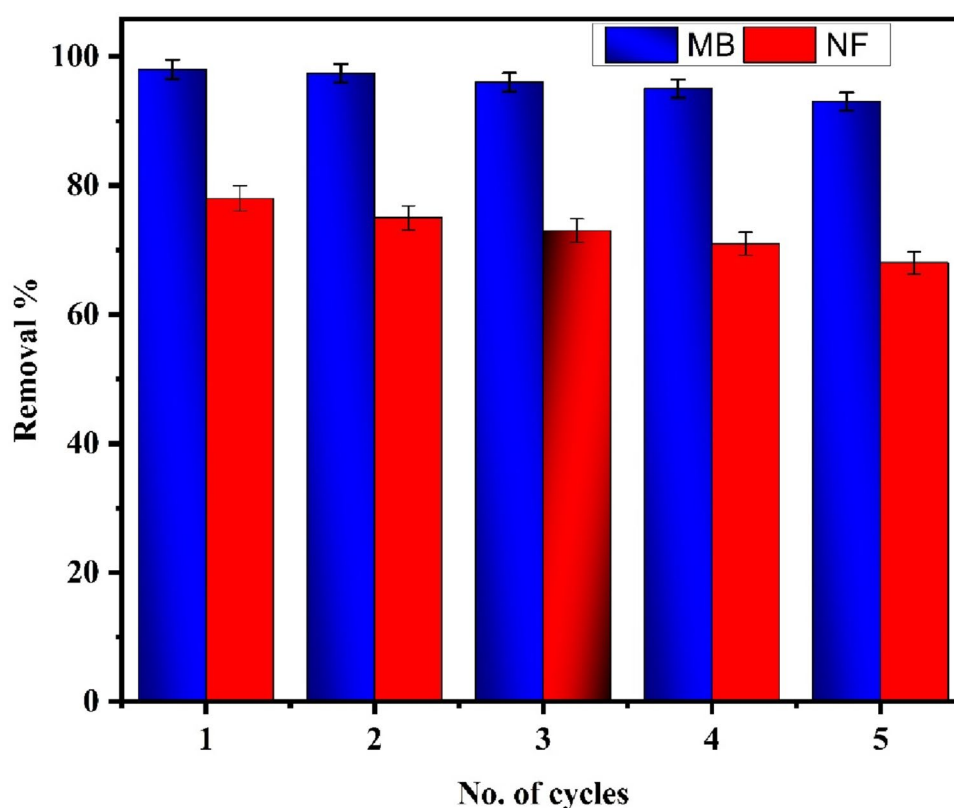
Reusability Study

The reusability of the Dex/NVP hydrogel for the removal of nitrofurantoin (NF) and methylene blue (MB) was evaluated over five consecutive adsorption–desorption cycles. As shown in Fig. 13, the hydrogel maintained a high adsorption efficiency throughout the tested cycles, although a gradual decrease in the removal percentage was observed. For MB, the removal efficiency decreased slightly from approximately 98% in the first cycle to about 93% after the fifth cycle. In comparison, NF removal decreased from approximately 78% to about 68% over the same number of cycles. The relatively small reduction in MB adsorption demonstrates the strong affinity and good regeneration stability of the Dex/NVP hydrogel toward the cationic dye. The greater reduction observed for NF may be associated with incomplete desorption, partial blockage of adsorption

Table 4 Thermodynamic parameters for NF and MB adsorption onto the Dex/NVP hydrogel in the temperature range 298.15 – 318.15 K

Adsorbate	ΔH° (kJ mol^{-1})	ΔS° ($\text{J mol}^{-1} \text{ K}^{-1}$)	R^2	ΔG° at 298.15 K (kJ mol^{-1})	ΔG° at 308.15 K (kJ mol^{-1})	ΔG° at 318.15 K (kJ mol^{-1})
NF	27.33 ± 0.39	118.42 ± 1.26	0.99980	-7.97	-9.16	-10.34
MB	55.06 ± 0.91	222.83 ± 2.97	0.99973	-11.38	-13.61	-15.84

Fig. 13 Reusability of the Dex/NVP hydrogel for NF and MB removal over five consecutive adsorption–desorption cycles. Operational conditions: initial pollutant concentration = 20 mg/L, solution volume = 100 mL, adsorbent weight = 0.05 g, pH = 3.0, and contact time = 420 min. Data are presented as mean $\pm$ SD ($n=3$)



sites, or retention of NF molecules within the porous hydrogel network. Repeated swelling, washing, and regeneration may also cause minor structural changes or loss of accessible functional groups. Nevertheless, the hydrogel retained considerable adsorption performance after five cycles, confirming its potential for repeated wastewater-treatment applications.

The operational stability of the Dex/NVP hydrogel was assessed through five consecutive adsorption–desorption cycles. The retention of substantial NF and MB removal efficiencies after repeated regeneration indicates satisfactory functional stability under the investigated conditions. Nevertheless, further leaching and post-regeneration structural analyses are recommended to fully evaluate its long-term chemical stability and environmental safety. Besides adsorption efficiency, the preparation cost and reusability of an adsorbent are important considerations for its practical application. The principal cost components of the Dex/NVP hydrogel include dextrin, N-vinylpyrrolidone, irradiation processing, washing, drying, and regeneration. The use of a bio-based dextrin backbone and radiation-induced synthesis without an additional chemical initiator or conventional crosslinking agent may simplify the preparation procedure. Furthermore, the retention of adsorption performance over five adsorption–desorption cycles could decrease adsorbent replacement requirements and reduce the generation of spent adsorbent. Similar cost and scale-up considerations

Table 5 Comparison of the maximum adsorption capacity of the developed Dex/NVP biohydrogel with reported adsorbents for nitrofurantoin and a related pharmaceutical contaminant

Adsorbent system	Target contaminant	Qm (mg /g)	Reference
Banana-peel biochar	Nitrofurantoin	3.78	Bernardino et al., [8]
Dex/NVP biohydrogel	Nitrofurantoin	40.93	Present study

have been emphasized for adsorption-based wastewater-treatment materials (Saha, Debnath, & Saha, 2024). However, because the present experiments were conducted at laboratory scale, a detailed pilot-scale techno-economic assessment that includes capital, energy, labour, water, regeneration, and waste-disposal costs is required before industrial implementation.

Comparative Adsorption Performance

To place the adsorption performance of the Dex/NVP biohydrogel in the context of recent literature, its maximum Langmuir adsorption capacities were compared with reported adsorbents for NF, a related pharmaceutical contaminant, and MB. Direct studies on hydrogel-based NF adsorption remain scarce; therefore, Table 5 includes the limited NF literature together with one hydrogel study involving a related antibiotic.

As shown in Tables 5 and 6 the Dex/NVP biohydrogel achieved maximum adsorption capacities of 40.93 mg/g for NF and 42.51 mg/g for MB. Its NF capacity is higher than the values reported for the directly comparable NF adsorbent and the related pharmaceutical hydrogel listed in Table 5. For MB, some nanocomposite hydrogels containing CuO, carbon, orange peel (BOP1) and biosorbent green peel exhibit considerably higher capacities. However, these systems differ substantially in composition, porosity, surface charge, and test conditions, preventing a strict one-to-one comparison. The principal advantage of the present Dex/NVP biohydrogel is therefore not the highest reported MB capacity, but its balanced adsorption of two chemically distinct contaminants within a single dextrin-containing hydrogel platform. This dual-function performance, together with the simple material composition and the absence of metal-containing nanofillers, supports the practical and environmental relevance of the developed adsorbent.

Frontier Molecular Orbital Analysis of Dex/NVP Hydrogel Components

Frontier molecular orbital analysis was performed to investigate the electronic characteristics of the main components forming the Dex/NVP hydrogel network Fig. 14. This analysis provides useful information about the electron-donating and electron-accepting regions of dextrin and N-vinylpyrrolidone (NVP), which may contribute to the adsorption behavior of the hydrogel through polar interactions, hydrogen bonding, and donor–acceptor interactions.

The electronic structure of dextrin showed a HOMO energy of -11.31 eV and a LUMO energy of $+3.99$ eV, giving a wide HOMO–LUMO energy gap of about 15.3 eV. This large energy gap indicates that dextrin is electronically stable and chemically hard, with a low tendency toward charge-transfer or redox reactions. Therefore, dextrin mainly acts as a stable hydrophilic backbone within the hydrogel network. The frontier orbital distribution is mainly localized around the oxygen-rich carbohydrate units, particularly the hydroxyl and ether groups. These functional groups are

expected to provide abundant polar adsorption sites capable of interacting with pollutant molecules through hydrogen bonding and dipole–dipole interactions.

In contrast, NVP exhibited a smaller HOMO–LUMO gap of about 5.81 eV, indicating higher polarizability and greater electronic responsiveness compared with dextrin. The HOMO and LUMO distributions of NVP were mainly localized around the vinyl-amide region, especially near the carbonyl and amide groups. These sites may act as more active centers within the hydrogel network, enhancing its ability to interact with adsorbate molecules through hydrogen bonding, dipole interactions, and possible donor–acceptor interactions. Thus, the incorporation of NVP into the dextrin matrix improves the electronic and functional diversity of the hydrogel.

The molecular orbital results support the adsorption performance observed experimentally. Dextrin provides structural stability, hydrophilicity, and abundant hydroxyl groups, while NVP introduces more polarizable amide-containing active sites. The cooperative contribution of these two components can explain the efficient adsorption behavior of the Dex/NVP hydrogel under the optimized experimental conditions. The presence of hydroxyl, ether, carbonyl, and amide groups within the hydrogel network facilitates interaction with pollutant molecules and improves their uptake from aqueous solution.

These results also agree with the adsorption isotherm study, where the Langmuir model provided a better description of the adsorption process than the Freundlich model. This suggests that adsorption mainly occurs through monolayer coverage on available active sites distributed within the Dex/NVP hydrogel network. The electronic stability of dextrin and the higher polarizability of NVP together provide a balanced structure: dextrin maintains the stability of the hydrogel matrix, while NVP enhances the availability and activity of adsorption sites.

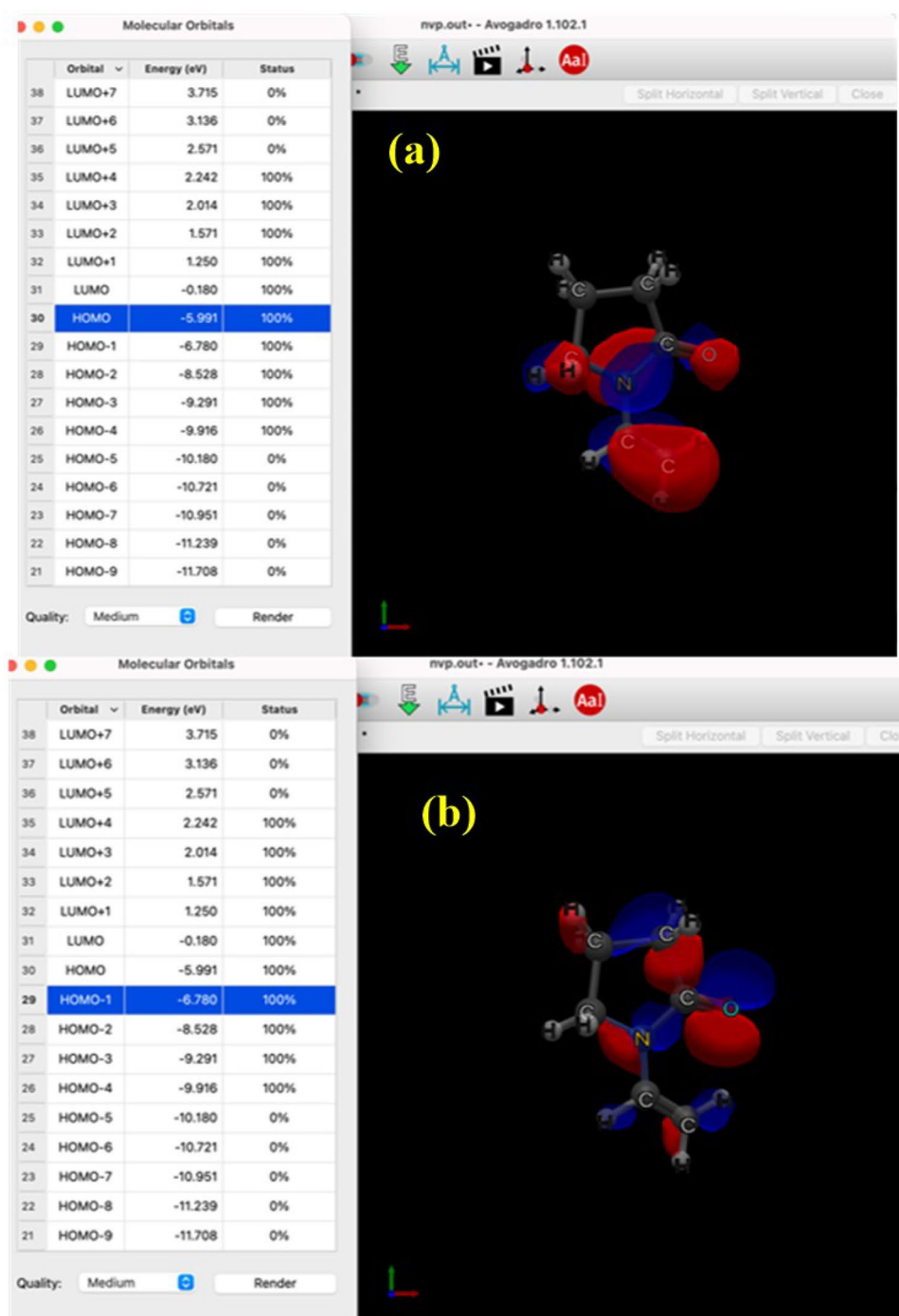
Proposed Adsorption Mechanism

The proposed adsorption mechanism of NF and MB onto the Dex/NVP hydrogel is illustrated in Fig. 15. Initially, the pollutant molecules diffuse from the aqueous phase into the interconnected pores of the swollen hydrogel network. The hydroxyl, carbonyl, and amide groups present in the Dex/NVP matrix provide active adsorption sites for both contaminants. NF adsorption is mainly promoted by hydrogen bonding between its oxygen- and nitrogen-containing groups and the hydroxyl or amide groups of the hydrogel, together with dipole–dipole interactions involving carbonyl groups [14, 15]. MB adsorption is facilitated by diffusion into the porous network and electrostatic and dipole-related interactions between the cationic dye and the

Table 6 Comparison of the maximum adsorption capacity of the developed Dex/NVP biohydrogel with recently reported hydrogel-based and bio-based adsorbents for methylene blue

Adsorbent system		
Pectin/Arabic gum-based CuO nanocomposite hydrogel	219.82	Sayed et al., [49]
Green Peel	207.90	Benkouachi et al., [6]
H ₃ PO ₄ -modified orange-peel/sodium-alginate beads, BOP1	764.92	Guediri et al., [26]
Superabsorbent polymer/activated-charcoal composite hydrogel	213.2	Shah et al., [51]
Dex/NVP biohydrogel	42.51	Present study

Fig. 14 Frontier molecular orbital distributions of N-vinylpyrrolidone (NVP) (a) and dextrin (b)



polar functional groups of the hydrogel [16]. Therefore, the overall adsorption process is proposed to arise from the synergistic contribution of physical diffusion and specific intermolecular interactions. This interpretation is consistent with the FTIR, SEM, AFM, adsorption-isotherm, kinetic, and frontier molecular orbital results. Under the optimum conditions of pH 3.0, an adsorbent dose of 0.05 g per 100 mL, and a contact time of 420 min, removal efficiencies of approximately 77.8% for NF and 98% for MB were achieved.

Limitations and Future Work

The present study demonstrates the successful synthesis of a gamma radiation-induced Dex/NVP biohydrogel and its efficient adsorption of nitrofurantoin (NF) and methylene blue (MB) under controlled laboratory conditions. However, several aspects remain to be investigated to facilitate its practical implementation.

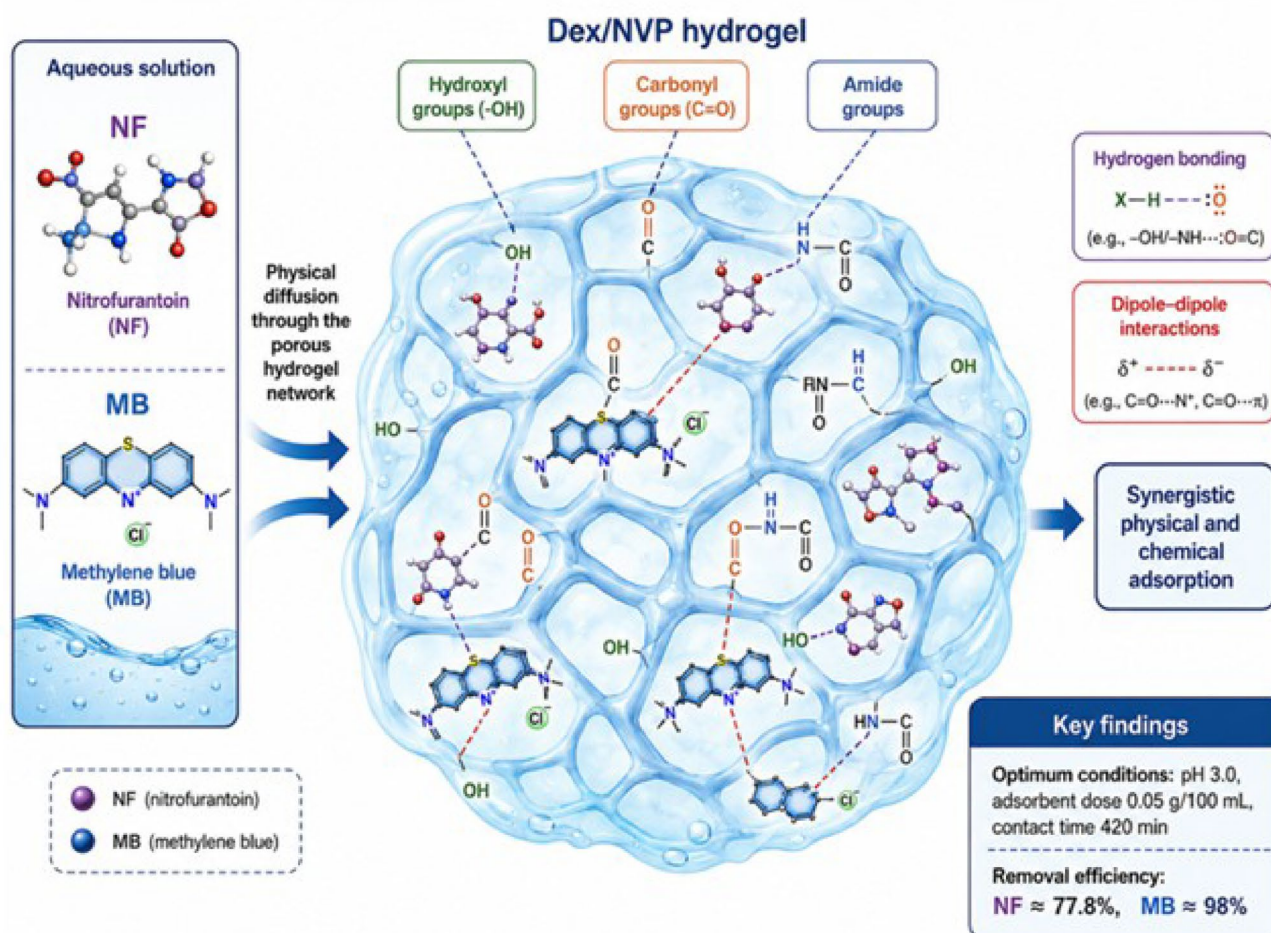


Fig. 15 Schematic representation of the proposed adsorption mechanism of nitrofurantoin (NF) and methylene blue (MB) onto the Dex/NVP hydrogel through diffusion within the porous network, hydrogen

bonding, and dipole-dipole interactions involving hydroxyl, carbonyl, and amide functional groups. Optimum operational conditions: pH 3.0, adsorbent dose = 0.05 g per 100 mL, and contact time = 420 min

The adsorption experiments were performed using synthetic aqueous solutions; therefore, future studies should evaluate the adsorption performance in real wastewater containing complex matrices and competing coexisting ions. In addition, long-term storage stability and operational stability under different environmental conditions should be considered to verify the durability of the developed adsorbent. Furthermore, pilot-scale investigations and continuous-flow column studies are necessary to validate its applicability under realistic treatment conditions.

Future work should also include a comprehensive techno-economic assessment, considering raw material consumption, irradiation cost, processing, and regeneration expenses, to evaluate the economic feasibility of large-scale production and practical wastewater treatment applications. Finally, additional investigations into the adsorption mechanism using complementary characterization techniques and advanced kinetic and isotherm analyses may provide further

insight into the interaction between the adsorbent surface and different classes of contaminants.

Conclusion

A sustainable dextrin/N-vinylpyrrolidone (Dex/NVP) biohydrogel was successfully synthesized via Co-60 gamma irradiation using a green, initiator-free, and chemical-crosslinker-free approach. The optimized hydrogel, prepared with 5% dextrin at an irradiation dose of 30 kGy, exhibited a high gel fraction ($\sim 90\%$) and excellent swelling capacity ($\sim 800\%$). Structural, morphological, and thermal characterization confirmed the successful formation of a crosslinked hydrogel network with a porous architecture suitable for adsorption applications.

The prepared Dex/NVP biohydrogel demonstrated efficient simultaneous removal of nitrofurantoin (NF) and

methylene blue (MB), achieving removal efficiencies of approximately 78% and 98%, respectively, under the optimized adsorption conditions. Equilibrium data were best described by the Langmuir isotherm, with maximum adsorption capacities of 40.93 mg g⁻¹ for NF and 42.51 mg g⁻¹ for MB, indicating favorable monolayer adsorption. Combined experimental characterization and frontier molecular orbital analysis suggested that the adsorption process is governed by the synergistic contribution of the hydrogel functional groups through hydrogen bonding, electrostatic, dipole–dipole, and donor–acceptor interactions.

Overall, the radiation-synthesized Dex/NVP biohydrogel offers a simple, environmentally friendly, and effective adsorbent for the simultaneous removal of pharmaceutical and dye contaminants from water. The promising adsorption performance, together with the green synthesis strategy, highlights its potential for sustainable wastewater treatment applications. Further studies should focus on evaluating its performance in real wastewater, long-term reusability, continuous-flow systems, and techno-economic feasibility to support practical implementation.

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Declarations

Competing interests The authors declare no competing interests.

Consent to Participate The authors confirm that all information they provide for this study is realistic experimental data and findings.

Consent to Publish The authors give their consent for the publication of identifiable details, which can include photograph(s) and/or videos and/or case history and/or details within the text (“Material”) to be published in the above Journal and Article.

Ethical Approval Not applicable. This study does not involve any human participants or animals.

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