

Research Article

Modification of Superabsorbent Hydrogels for Industrial Wastewater Treatment

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A breakthrough in superabsorbent hydrogel (SAH) preparation was studied in the current issue by blending potato starch and acrylic acid for wastewater treatment. Gamma irradiation source (⁶⁰Co irradiation) was used to irradiate SAH from 1 to 10 kGy dose at room temperature (~27°C). The swelling ratio, water absorption, equilibrium water content, and gel fraction properties of the hydrogel were investigated. The as-prepared hydrogel treated with KOH (THG) showed excellent absorption capacity but less mechanical stability compared to untreated hydrogel (UHG). The gel fraction of treated SAH was slightly lower in methanol, but the utmost in water at 5 kGy infers the proper grafting of SAH at this point. The prepared SAH was characterized using scanning electron microscopy (SEM) and Fourier transform infrared (FTIR) to investigate the surface morphology and molecular interaction, respectively. Moreover, this study's focal point is to propose an alternative method to remove chromium and methylene blue by SAH from industrial wastewater. The Cr adsorption capacity of UHG was higher than that of THG because the proton's replacement is easier than that of K by Cr. On the other hand, THG was found to be more efficient in removing methylene blue from industrial wastewater due to the presence of an easily ionized group (-COOK) in SAH. Therefore, the hydrogel can be proposed as a potential superabsorbent to remove heavy metals and organic dyes from industrial wastewater.

1. Introduction

Hydrogels are three-dimensional cross-linked polymeric substances that can absorb water in an aqueous environment. Among various types of hydrogels, there is a particular category that can absorb water several hundred times its dry weight named superabsorbent hydrogels (SAHs). They can absorb and retain vast amounts of water even under high pressure or temperature because of the existence of several hydrophilic groups, e.g., -OH, -COOH, -CONH₂, -CONH, and -SO₃H. SAH does not allow its substance to dissolve in water due to its grafted polymeric network

[1–6]. SAH has found extensive use mostly in the manufacturing of baby diapers, sanitary napkins, candles, composites and laminates, controlled drug delivery, filtration, fire-retardant gels, flood control, fragrance carriers, medical waste solidification, surgical pads, water-absorbent pads, wound dressings, in agricultural land as a soil conditioner and fertilizer, etc. [7].

Many types of preparation processes are available to prepare hydrogels of various categories. In general, hydrogels can be prepared by cross-linking of monofunctional and multifunctional monomers or by cross-linking of the copolymer, homopolymer in solution, and simultaneous

copolymerization. The cross-linking process has two steps to complete. The first step is synthesizing the linear polymers in the absence of agents, and the second step is cross-linking of the synthesized polymers using either chemical reagents or irradiation [8–10]. The radiation-induced polymerization technique is a modern, worldwide, eco-friendly concept. It can promote cross-linking in a wide range of polymers and copolymers without additives during the polymerization process. The chemical, physical, and mechanical properties can be improved by cross-linking of polymers [11], and the intensity of cross-linking is controlled by the radiation dose [12]. The cross-linked structure of the polymer is imperative for the packed column application. Additionally, the cross-linked polymer might form a more stable complex with metal ions due to its rigid network structure [13]. The use of the radiation polymerization technique has the following advantages: (i) ionizing radiation initiates radical polymerization at ambient temperature in the absence of chemical initiators [14, 15]; (ii) the initiation step of radiation polymerization is temperature-independent, and the overall activation energies are much smaller than in the chemically initiated process [14, 16]; (iii) the resulting polymer is homogeneous and free from any impurities, and the molecular weight can be controlled by varying doses and dose rate [14, 17].

Adsorption among other water purification technologies is fast, low-cost, easy operation, and high efficiency without producing undesirable by-products [18, 19]. Natural polymer-based hydrogels are revealed as non-toxic, environmentally friendly compounds, which treated as renewable or biologically degradable polymers with lower toxicity [20]. Monir et al. [21] prepared hydrogel from polyethylene oxide (PEO) and acrylic acid (AAc) blends by applying gamma radiation. Afroz et al. [4] prepared hydrogel by aqueous mixture blend of polyethylene oxide (PEO)/N,N-dimethylacrylamide (DMA) using gamma radiation. Sultana et al. [22] synthesized hydrogels from a dilute solution of carboxymethyl cellulose in the presence of mono- and divalent salts by applying gamma radiation. Dafader et al. [23–25] produced three different categories of hydrogels from acrylamide/HEMA, polyvinyl pyrrolidone/AAc, and cassava starch/AAc by graft copolymerization method using gamma radiation of different doses. Bhuyan et al. [26] prepared hydrogels from a blend of potato starch and AAc with gamma radiation and compared the properties with the addition of NaOH in the hydrogel and used only to remove dye. Recently, a few types of hydrogels, i.e., micelle-laden [27], scavenging nanocomposite [28], chitosan [29], and porous nanocomposite [30], are used for water treatment available in the open literature.

However, it has no extensive studies to remove chromium (Cr) and methylene blue (MB) by SAH from lather and textile industrial wastewater (IWW) collected from Savar industrial area, Bangladesh. Therefore, in this present study, potato starch and acrylic acid blends were used to get superabsorbent hydrogels (SAHs) which were irradiated with Co-60 gamma radiation. A number of them were prepared with the addition of alkali (KOH)

solution, and this is also a novelty of this study. The swelling ratio, water absorption, equilibrium water content, and gel fraction were measured and compared with KOH-treated hydrogel (THG) and untreated hydrogel (UHG). The surface morphology and molecular interaction of SAH were investigated by scanning electron microscopy (SEM) and Fourier transform infrared (FTIR), respectively. Additionally, the development of an alternative method was established to remove chromium (Cr) and methylene blue (MB) by SAH from IWW.

2. Experimental

2.1. Chemicals. Potato starch was collected from Sigma-Aldrich, India. Acrylic acid and KOH pellets were purchased from Guangdong Guanghua Chemical Factory Co. Ltd., China, and Merck Private Co. Ltd., India, respectively. All chemicals were reagent grade and used without further purification.

2.2. Apparatus. Atomic Absorption Spectrophotometer (AAS) (model: AA-6800, supplied by SHIMADZU, Japan), Cobalt-60 gamma radiation source (model: GSCIE-11), Fourier transform infrared (FTIR) spectrophotometer (model: AIM-8800, supplied by SHIMADZU, Japan.), and scanning electron microscopy (SEM) (model: JSM-6490, JEOL) were employed for sample characterization.

2.3. Preparation of Superabsorbent Hydrogel (SAH)

2.3.1. Preparation of SAH from Starch and AAc Solution. 150 ml distilled water and 20 g starch were mixed in a 400 ml beaker and stirred with a magnetic stirrer under heating conditions until the dissolution of starch at 80°C for 30 min. Then, the solution was cooled slowly until room temperature, and 30 ml saturated acrylic acid was added to the beaker. The beaker was sealed with aluminum foil. The solution was stirred continuously (without heating) for 30 minutes to make a homogeneous mixture. Then, the solution contained in the glass test tubes was poured into polyethylene bags, and after pouring the samples, the polyethylene bags were sealed. Then, the solution was sent to a gamma radiation source for irradiation.

2.3.2. Preparation of SAH from Starch, AAc, and KOH Solution. To prepare a 3.86 M KOH solution, 50 ml distilled water and 10.8 g KOH were mixed in a 100 ml reagent bottle. It was stirred with a glass rod until KOH dissolved in water. In the prepared starch solution, KOH solution was mixed with stirring for 20 minutes. Then, 30 ml acrylic acid was added into the beaker and sealed with aluminum foil. The solution was stirred continuously for 30 minutes to get a homogeneous mixture. Then, the prepared solution was taken in several test tubes from beaker and poured into polyethylene bags, and after pouring the samples, the polyethylene bags were sealed. Then, the solution was sent to a gamma radiation source for irradiation by ⁶⁰Co source with radiation doses of 1, 3, 5, 7, and 10 kGy at 6.8 kGy/h of radiation dose rate. The obtained SAHs were

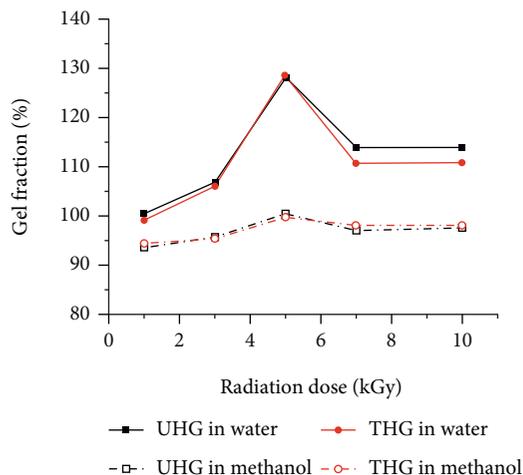


FIGURE 3: Gel fraction of THG and UHG in water and methanol.

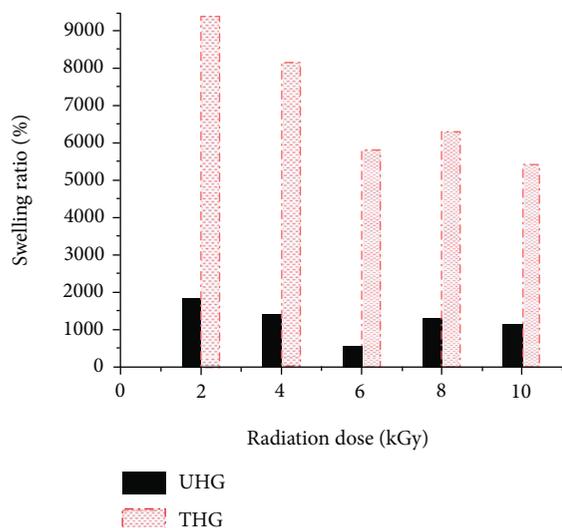


FIGURE 4: The swelling ratio of THG and UHG with radiation dose.

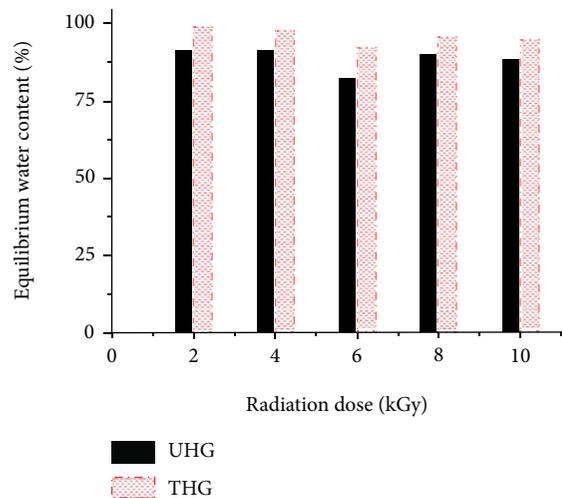


FIGURE 5: The equilibrium water content of THG and UHG with radiation dose.

determine the insoluble content in the samples gravimetrically.

$$\text{Gel content (\%)} = \left[\frac{W_1}{W_0} \right] \times 100, \quad (1)$$

where W_1 is the weight of dry gel after extraction in distilled water and W_0 is the initial weight of dry gel.

2.5. Determination of the Swelling Ratio. SAH samples dried to a constant weight were immersed in distilled water until maximum swelling was obtained at room temperature ($\sim 27^\circ\text{C}$). The prepared hydrogel contains cross-linking polymer network that increases its volume by absorbing water. The temperature of water affects the internal energy and entropy of hydrogel, which leads to the penetration of water into the hydrogel, which means swelling ratio. In the present study, the water temperature was maintained at around 25°C throughout the experiment. The swollen samples were removed with distilled water and weighed after removing the surface water with soft tissue paper. The experiment was repeated three times for each sample, and the average weight of the swollen sample was determined. The swelling ratio was calculated as follows:

$$\text{Swelling ratio} = \frac{[W_2 - W_1]}{W_1}, \quad (2)$$

where W_1 and W_2 are the weight of the dried and swelled gel, respectively.

2.6. Determination of Equilibrium Water Content. The gel sample was dried to a constant weight and was kept in distilled water at room temperature. Swelling of the gel was continued to reach the constant weight and weighed after removing any surface water with tissue paper. The equilibrium water content was calculated as follows:

$$\text{Equilibrium water content (\%)} = \left(\frac{[W_2 - W_1]}{W_2} \right) \times 100, \quad (3)$$

where W_1 and W_2 are the weight of dried gel and gel after water absorption, respectively.

2.7. Determination of Water Absorption. After immersing the hydrogel samples in distilled water at room temperature, the samples were periodically weighed for blotting excess surface water on filter paper. The water absorption of the samples was calculated as follows:

$$\text{Water absorption (\%)} = \left(\frac{[W_t - W_1]}{W_1} \right) \times 100, \quad (4)$$

where W_1 and W_t are the weight of dried and swelled gel at time t , respectively.

2.8. Determination of Chromium Adsorption. Atomic Absorption Spectrometry (AAS) is a prevalent and reliable technique for detecting metals and metalloids in

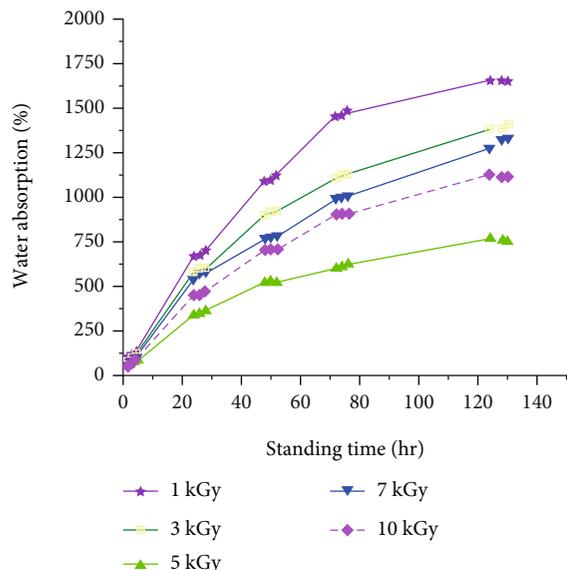


FIGURE 6: The effect of radiation dose and standing time on water absorption of UHG.

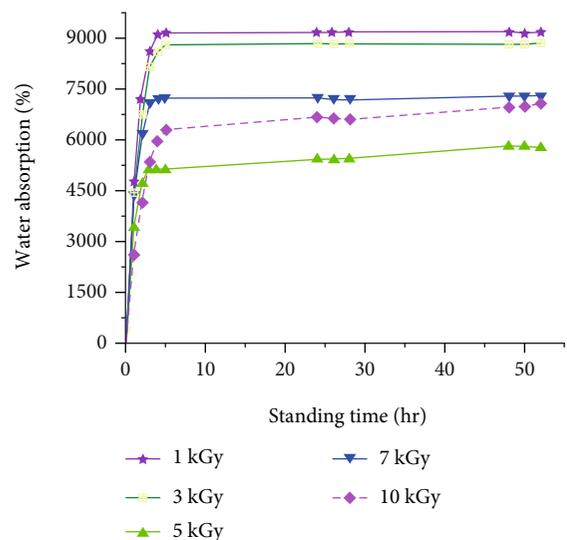


FIGURE 7: Effect of radiation dose and standing time on water absorption of THG.

environmental samples. The total chromium content in water and SAH samples was measured by Flame Atomic Absorption Spectrometry. At first, a typical set of standard calibration curves with good linear regression and better relative standard deviation was attained. The absorption capacity of SAH was determined by chromium adsorption as follows:

$$\text{Metal adsorption capacity} = \frac{V(C_1 - C_2)}{W}, \quad (5)$$

where V is the volume of the chromium solution in liter, W is the weight of SAH in gram, C_1 is the concentration of

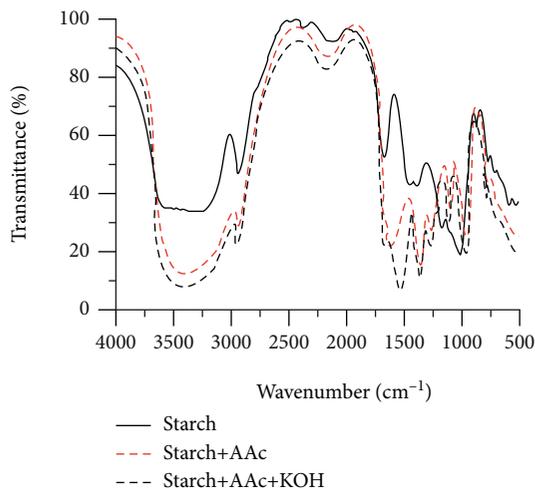


FIGURE 8: FTIR spectra of hydrogels.

Cr solution before adsorption (ppm), and C_2 is the concentration of Cr solution after adsorption (ppm).

2.9. Determination of Methylene Blue Dye Adsorption.

Water-soluble dye methylene blue solution was prepared by dissolving the dye in deionized water. The SAH was added into the dye solution, and the concentration of dye solution was 20 ppm, 40 ppm, and 60 ppm. The amount of residual dye in an aqueous solution was determined by UV-visible spectroscopy periodically. The absorption capacity of SAH was determined for methylene blue adsorption as follows:

$$\text{Dye adsorption capacity} = \frac{L(C_1 - C_2)}{M}, \quad (6)$$

where L is the volume of the dye solution in liter, M is the weight of SAH in gram, C_1 is the concentration of dye solution before adsorption (ppm), and C_2 is the concentration of dye solution after adsorption (ppm).

3. Results and Discussions

3.1. The Mechanism of the Reaction. Gamma radiation leads to cross-linking in the starch matrix. The impact of gamma radiation on starch was reported before by [31]. They provide insight into the improvement of the physical and cross-linking properties by gamma radiation. Figure 1 shows the schematic view of a cross-linked polymer network. In the radiated solution, three reactive species were formed in water as hydrated electrons, hydroxyl radicals, and hydrogen radicals. The free radicals generated in the starch molecule (amylopectin) were observed after exposure to the solution with radiation treatment. In the aqueous blend of starch and AAc with the use of radiation, the free radicals formed in acid react with radiated starch to get a long-chain graft structure of hydrogels shown in Figure 2. The grafting process of acrylic acid onto potato starch was carried out using a radiation source, and a polymeric network of negatively charged $-\text{COO}^-$ groups was generated. Finally, the alkali- (KOH-) treated SAH was carried

TABLE 1: Characteristic bands of different functional groups in hydrogels.

Functional group	Frequency range (cm ⁻¹)	Reference (cm ⁻¹)
-OH stretching of starch	3450	3423 [26]
C-O-C stretching of starch	1020-1170	1022-1161 [26]
C-H stretching of absorption	2950	294 [26]
C=O stretching of the carboxyl group of AAc	1720	171 [26]
-COOK in SAH	1550	153 [33]

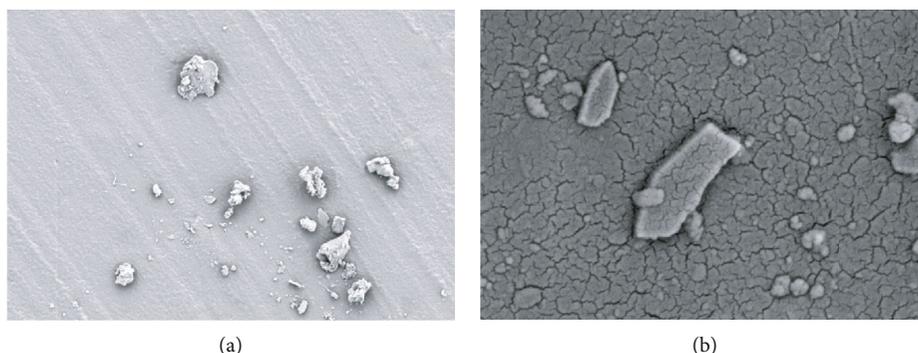


FIGURE 9: SEM images of SAH: (a) UHG and (b) THG.

out by the -COOK group by replacing the proton with potassium ions as shown in Figures 1 and 2.

3.2. Effect of Radiation Dose and KOH on Gel Fraction. The KOH-treated hydrogel (THG) and untreated hydrogel (UHG) were prepared by gamma radiation at different doses, and the as-prepared hydrogels were dissolved in two different solutions of pure methanol and water. The results obtained from the experiment are presented in Figure 3. The gel fraction of SAH increased with grafting with radiation dose in a specific value up to 5.0 kGy. Above the radiation 5.0 kGy, the gel fraction shows a decreasing trend due to the degradation of molecules. The gel fraction of SAH is almost 15% better in water than that of methanol. In literature, the gel fraction was measured after removing the dissolved parts from solvent-immersed hydrogel as similar studies reported elsewhere [21, 22].

3.3. Effect of Radiation Dose and KOH on Swelling Ratio. The effect of radiation dose on the swelling ratio of treated (THG) and untreated hydrogels (UHGs) was observed in Figure 4. The swelling ratio decreases with increasing radiation dose, due to an increase in cross-linked density and lower most value at 5.0 kGy. This might be due to the proper grafting of SAH at 5.0 kGy implying the optimum radiation dose. The swelling ratio of THG was found to be about 5.0 times better than that of UHG due to the higher water absorption capacity of KOH-treated SAH. The hydrophilicity of the potassium carboxylate group (-COOK) is higher than that of the carboxyl group (-COOH). Bhuyan et al. and Lou et al. [26, 32] measured the swelling ratio of NaOH-treated hydrogels and found a better swelling ratio of THG than that of UHG.

3.4. Effect of Radiation Dose and KOH on Equilibrium Water Content. Figure 5 shows the effect of radiation dose and KOH on the equilibrium water content. It can be found that the equilibrium water content decreases with increased radiation dose because more grafting ability implies less amount of water absorbed. A similar phenomenon was also reported in [22]. The minimum equilibrium water content was found at 5.0 kGy for excellent grafting. In addition, the equilibrium water content was found 8% better in THG than UHG due to higher water absorption capacity. Nevertheless, the THG was easily broken after water absorption because of its less mechanical stability of it.

3.5. Effect of Radiation Dosage on Water Absorption of UHG. The impact of the radiation dose on water absorption of UHG is presented in Figure 6. The patterns of all studied data obtained from water absorption showed uphill trends withstanding time. The water absorption capacity of SAH was maximum at 1.0 kGy radiation dose and gradually decreased until 10.0 kGy except at 5.0 kGy. The lowest water absorption occurred at 5.0 kGy due to better cross-linking at this point, and above it is the initiation of degradation of molecules of SAH. Considering other doses, the water absorption capacity of SAH reduces in the periodic order. The established view of the point is that the cross-linking increases with the increase of radiation dosage as well as void spaces in the polymer network reduced for free water entrance and imply low water absorption capacity [23].

3.6. Effect of Radiation Dosage on Water Absorption of THG. The effect of radiation dose on water absorption of THG is shown in Figure 7. Water absorption capacity of the alkali-treated sample (THG) increases sharply withstanding time from 1.0 to 5.0 h and becomes constant after a pick point

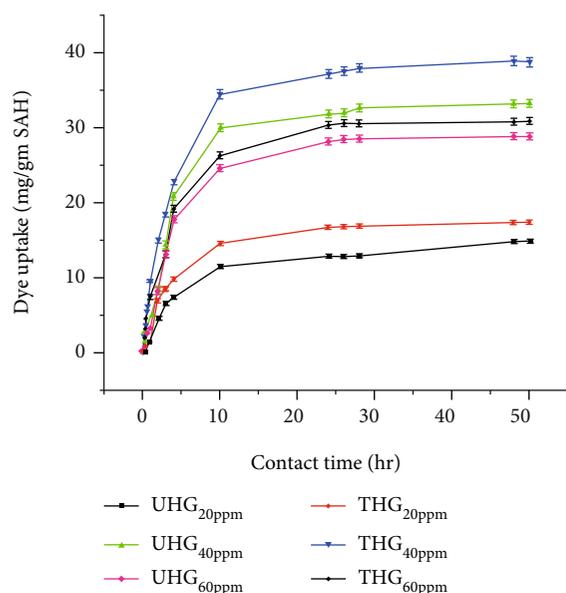


FIGURE 10: The effect of contact time on the removal of methylene blue dye.

TABLE 2: Characteristics/efficiency value of SAH at 5.0 kGy.

Property/performance	Unit	Value	
		THG	UHG
Gel fraction	%	128.43	128
Swelling ratio	%	5871.5	570.65
Equilibrium water content	%	92.5	82.38
Water absorption	%	5798.61 _{min}	755.06 _{min}
MB removal at 40 ppm	mg/g	38.77 _{max}	33.31 _{max}
Cr removal at 40 ppm	mg/g	5.93 _{max}	10.13 _{max}

min: minimum; max: maximum.

beyond 5.0 h. However, the water absorption of the gels showed utmost at 1.0 kGy radiation dose due to lack of proper cross-linking and lowest at 5.0 kGy because of desired cross-linking. Moreover, the water absorption capacity of the treated gels was observed to be lowest at 5.0 kGy due to the deficiency of void space in the system.

3.7. FTIR Analysis of SAH. FTIR analysis can present the molecular interaction of potato starch/AAC blend to form a hydrogel. Figure 8 presents the infrared spectra of potato starch, starch/AAC, and starch/AAC/KOH blended hydrogel. The peaks observed around 3450 cm^{-1} and $1020\text{--}1170\text{ cm}^{-1}$ can be attributed to the O–H stretching and C–O–C skeletal vibration of starch. The absorption peak at 2950 cm^{-1} corresponds to C–H stretching of the acrylate unit and starch, while the bands at 1395 and 1265 cm^{-1} correspond to the angular deformation of C–H. The peak at 1720 cm^{-1} can be ascribed to the C=O stretching of the carboxyl group of AAC. In addition, peak at 1550 cm^{-1} corresponds to --COOK , which is absent in the spectra of starch/AAC and pure starch. This result indicates the interaction of AAC with starch. The FTIR spectra are listed and compared with previous work in Table 1.

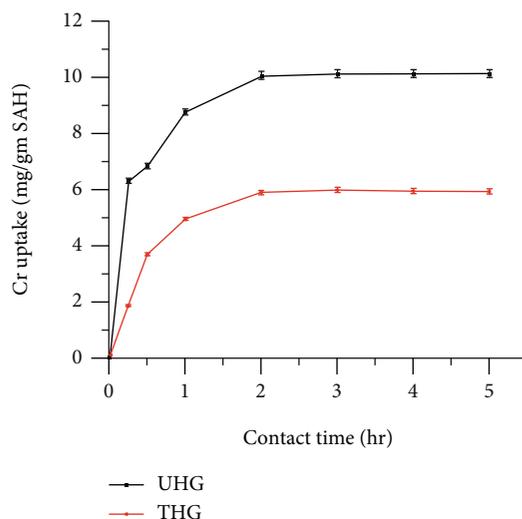


FIGURE 11: The effect of contact time on the removal of chromium.

3.8. SEM Analysis of SAH. Figure 9 presents the SEM micrographs of starch, starch/AAC blend, and starch/AAC/KOH blend hydrogels. The surface morphologies of the hydrogels show different characteristics. The starch/AAC hydrogel exhibits a relatively rough and compact surface pattern in SEM images (Figure 9(a)), which might be due to the hydrogen bond attraction between the grafted --COOH chains. On the other hand, Figure 9(b) shows a relatively expanded structure in SEM images, which may be due to the repulsion between grafted --COO^- chains with large electrostatic force. This expansion can facilitate the treated SAH to adsorb methylene blue. Bhuyan et al. [23] reported almost the same pattern of surface morphology for hydrogels.

3.9. Application of As-Prepared Hydrogels. Adsorption is one of the most important industrial mass transfer processes through which the dissolved components from an aqueous solution can remove by solid adsorbent to attract the dissolved solute towards its surface. The accumulation of concentrated matter at surface or the interphase is involved in this process. The liquid, solid, gas, or dissolved solute phase can exist in the adsorbent. The chemical adsorption occurred due to the exchange of electrons, where the adsorbate is chemically bonded to the surface. On the other hand, the physical adsorption occurred due to the attachment of adsorbate with the adsorbent surface by hydrogen bonding, Vander Waals forces, polarity, dipole-dipole interactions, etc. The above description notices extensive use in removing dyes or heavy metals from aqueous medium. The pH 6.5 of prepared hydrogel was maintained for dye and chromium removal.

3.10. Application for Dye Removal. An additional important issue was to investigate the ability of SAH to remove hazardous methylene blue (MB) from IWW. Figure 10 illustrates the adsorption of methylene blue dye with different initial dye concentrations (20 to 60 ppm) as a function of standing time. It was found that dye adsorption increases with an increased initial dye concentration in an aqueous solution due to an increase of dye species in the bulk solution,

TABLE 3: Adsorption capacity of hydrogels for Cr and MB dye removal.

Adsorbent	$q_{\max, Cr}$ (mg/g)	$q_{\max, MB}$ (mg/g)	Reference
AGMA-AAm-AAc hydrogel	48.49		[39]
Chitosan cross-linked hydrogel	100		[40]
N-Maleyl chitosan P(AA-Co-VPA) hydrogel	66.89		[41]
Tio ₂ -graphene hydrogel graphite oxide		4.73	[42]
Cellulose/chitosan hydrogel beads		40	[43]
Chitosan-based hydrogel		93.03	[44]
Potato starch/AAc/gamma radiation/KOH	38.77	5.93	This study
Potato starch/AAc/gamma radiation	33.31	10.13	This study

increasing the possibility of dye contact with the gel. To capture MB, the SAH was prepared by 5.0 kGy gamma radiation dose that was optimized earlier. Furthermore, the dye adsorption capacity of THG was higher than that of UHG in every case. This is because the ionizing ability of the $-COOK$ group is more than $-COOH$ for the positively charged dye of MB. The maximum value is 38.95 mg/g of dye was removed by THG at an initial dye concentration of 40 ppm, indicating the possibilities for application in agriculture, pharmacy, and environmental technology.

3.11. Application for Chromium Removal. Hydrogels are excellent adsorbents to remove various types of metal ions from IWW [27]. This vital issue of the present study was to investigate the ability of SAH to remove hazardous chromium metal ions from industrial effluent water. The 40 ppm concentrated chromium was removed by 5.0 kGy of IWW as shown in Figure 11. Chromium adsorption was found to be higher for untreated SAH compared to the alkali-treated hydrogel. The reason is the low activity of Cr to exchange K instead of H. The Cr adsorption attained maximum values around 10.13 mg/g at 3 h by UHG. Therefore, SAH can be considered as an alternative to low-cost adsorbent [34–38] for IWW treatment. The characteristics or efficiency values of SAH at 5.0 kGy are listed in Table 2. Adsorption capacity of different hydrogels for Cr and MB dye removal is shown in Table 3. All experimental data of dye and Cr removal are reproducible and below 1.5% deviated.

4. Conclusion

Hydrogels were prepared from the aqueous solution of potato starch and AAc blend using gamma radiation, and a number of the hydrogels were treated with KOH in this study. The water absorption capacity of treated SAH is better than that of untreated SAH. It was also found that water absorption, swelling ratio, and equilibrium water content decrease with increased radiation dose. The optimum radiation dose is considered at 5.0 kGy concerning gel fraction and swelling ratio. FTIR spectroscopic analysis spectra indicate the interaction of potato starch molecules with acrylic acid to form a hydrogel. Scanning electron microscopy (SEM) discloses that KOH-treated hydrogels have expanded structures, which can facilitate the property of SAH to adsorb methylene blue from an aqueous solution. The pres-

ent scenario is the maximum removal of MB is 38.95 mg/g by treated SAH at an initial dye concentration of 40 ppm. On the other hand, the chromium adsorption was higher by untreated SAH because of the low activity of Cr to exchange K instead of H. The maximum adsorption attained for Cr was around 10.13 mg/g for 3.0 h by untreated SAH. Therefore, the SAH prepared in the present study was satisfactorily performed and proposed to be a practical application for IWW treatment enriched in organic dyes and heavy metals towards a safe environment.

Data Availability

All data generated through experiments and analyzed during this study are included within this article.

Conflicts of Interest

The authors declare no conflict of interest.

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