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## Article

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# Synthesis of a superabsorbent with low residual monomer content by gamma irradiation and improving gel strength through surface treatment

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## Abstract

In this research, an acrylic acid superabsorbent with low residual monomer content was synthesized using gamma irradiation, followed by a surface treatment method to enhance the characteristics of the hydrogel. A range of superabsorbent hydrogels was produced from partially neutralized acrylic acid through gamma irradiation at doses between 5 and 25 kGy at ambient temperature. The functional groups and morphology of the gel structure were analyzed using Fourier-transform infrared (FTIR) spectroscopy and scanning electron microscopy (SEM), respectively. Furthermore, the influence of radiation dose on several critical properties, including free swelling capacity (FSC), absorption under load (AUL), and thermal characteristics, was investigated. To improve the gel strength and AUL, a surface cross-linking treatment was performed using ethylene glycol di-glycidyl ether (DEGDGE). Following surface cross-linking, the AUL of the sample exhibited a significant increase of 118%. The chromatogram obtained from high-performance liquid chromatography (HPLC) indicated that the residual monomer in the optimal sample was below 1 ppm. These findings illustrate that gamma irradiation serves as an environmentally friendly method for producing high-performance superabsorbents, making them suitable for hygiene and biomedical applications.

**Keywords:** Gamma radiation, Superabsorbent, Acrylic acid, Surface treatment

## 1. Introduction

Superabsorbent polymers (SAP) are polymers that can absorb large amounts of water, often further the absorption capacity of traditional absorbent materials. Generally, they consist of a network of polymer chains that are cross-linked to avoid dissolution [1]. Since their inception, these polymers have been utilized in a variety of hygienic and non-hygienic fields. The distinctive properties and structural elements of these hydrogels are designed by their wide range of applications [2].

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Polyacrylates serve as the primary materials in the superabsorbent polymer (SAP) industry. Superabsorbent polyacrylates are synthesized through the polymerization of acrylic acid in conjunction with a cross-linking agent. Their remarkable moisture-absorption properties render gel-forming network materials derived from acrylic acid (AA) widely applicable in numerous fields. To maintain the presence of anionic groups in acrylic acid, it is desirable to neutralize the acid groups in the liquid phase, typically employing sodium or potassium hydroxide solution, prior to the polymerization process [3].

The polymerization of acrylic acid can be achieved through a variety of methods, including both chemical and radiation-induced cross-linking. Among these techniques, the radiation approach is often considered more beneficial than chemical cross-linking. Ionizing radiation, especially gamma irradiation from Co-60, enables initiator-free polymerization and cross-linking in aqueous conditions by producing radicals in situ. This technique also permits simultaneous sterilization and precise control of network density by varying the radiation dose and formulation parameters. The absence of additional initiators and the solvent-free processing environment can significantly reduce residual monomer levels compared to chemically initiated processes, thus improving biocompatibility and meeting regulatory standards for hygiene and biomedical superabsorbent polymers [4, 5].

One of the primary features of superabsorbent hydrogels is the swollen gel strength (modulus), which influences the liquid absorbency under load (AUL). Instead of using comonomer with acrylic acid to improve the AUL, suitable surface modification is applied to enhance the superabsorbent swollen gel modulus. Furthermore, surface treatment improves the flow characteristics of dried particles and reduces agglomeration by minimizing the deformation of surface cross-linked particles during AUL [6, 7]. The literature has identified several surface cross-linking agents for the treatment of PAA surfaces including: epoxides and alcohols, vinyl ethers, oxazolines, and polyquaternary amine [8-11].

To the best of our knowledge, there is currently no existing literature on the surface treatment of acrylic superabsorbents synthesized through gamma radiation. This study focused on the synthesis of acrylic acid superabsorbent with minimal residual monomer content via gamma irradiation. In this research, ethylene glycol di-glycidyl ether (DEGDGE) was applied as a cross-linking agent for the surface treatment of polyacrylic acid (PAA) to improve its absorbent properties.

## **2. Experimental**

### **2.1 Materials**

AA (99.5% purity), sodium hydroxide (NaOH, 84%), and sodium chloride (NaCl) were provided from Merck. Distilled water was used as the medium for conducting the polymerization and swelling experiments. All reagents employed were of analytical grade and were utilized without any additional purification.

### **2.2 Instrumentation**

The chemical changes of hydrogels were measured using a Fourier Transform Infrared (FTIR) spectrometer, TENSOR 27, Germany. The surface morphology of hydrogels was studied by field emission scanning electron microscope (SEM), using JSM 6510 equipment (JEOL, Japan), with operating voltage of 30 kV. Samples of hydrogels were immersed in distilled water at room temperature until maximum swelling, then was frozen at -80 °C and lyophilized. The freeze-dried hydrogel was fixed on brass holder and coated by sputtering with a thin Au layer in order to avoid charging effects. The thermal stability of the SAP samples was studied on an STA-1500 thermal analyzer (Germany) in a nitrogen atmosphere. Samples were heated from 30 to 600 °C at a heating rate of 10 °C/min. The residual monomer in SAP samples was determined based on the high-performance liquid chromatography (HPLC) technique using an HPLC system (Waters 1525 binary HPLC pump, USA) with a reverse phase Symmetry-C18 column (150 mm × 4.6 mm inner diameter, 5 µm particle size, Waters, USA) at 25±2 °C. The mobile phase was an aqueous 0.01% ortho-phosphoric acid at a flow rate of 1 ml/min. Detection was performed with a UV–Vis detector (2487 dual λ, Waters, USA), and the wavelength for quantification was 210 nm [12]. Rheological measurements were performed by a Paar-Physical oscillatory rheometer (MCR 300, Germany) at 25 °C with parallel plate geometry (plate diameter of 25 mm, gap of 3 mm). The storage modulus ( $G'$ ) was recorded as a function of angular frequency at constant shear strain 0.2 % (to be in the linear viscoelastic zone). All samples were measured at 0.1 g and subsequently allowed to swell in 5 mL of deionized water.

### **2.3 Preparation of polyacrylic superabsorbent**

A series of acrylic acid solutions were prepared from different concentrations of acrylic acid (5, 10 and 15 wt.%) dissolved in certain amount of distilled water, and then partially

neutralized with NaOH solution at 50% neutralization degree. The mixtures were stirred with 200 rpm at room temperature. Then, they were poured into the vials, sealed and irradiated in a Gamma-cell 220 (dose rate 2.52 kGy/h calibrated by Fricke dosimeters) at doses of 10, 15, 20 and 25 kGy at room temperature. The hydrogels were dried at 40 °C for 24 h, and milled up to 30-70 mesh.

## 2.4 Gel Fraction

The SAP samples, after achieving a constant weight ( $W_i$ ), were submerged in distilled water for 24 hours. After this period of immersion, the samples were removed from the distilled water and dried in a vacuum oven until a constant weight ( $W_1$ ) was achieved. The gel fraction was then determined using the following formula; where  $W_1$  is the weight of the dry sample after extraction in water, and  $W_i$  is the initial weight of the dry sample.

$$\text{Gel fraction (\%)} = W_1 / W_i \times 100 \quad (1)$$

## 2.5 Free swelling capacity (FSC)

The tea bag method was applied to measure the swelling capacity both in deionized water and a physiological saline solution (0.9% sodium chloride). A sample of 0.1 g of the superabsorbent ( $W_1$ ) was immersed in 500 g of either deionized water or saline solution for thirty minutes, allowing the sample to reach equilibrium swelling. Next, the bag was removed from the liquid, hanged for a determined period to eliminate excess fluid before weighing it ( $W_2$ ). A control bag, empty of the sample, was weighed as well ( $W_0$ ). The absorption capacity was determined using the following formula.

$$\text{FSC} = (W_2 - W_0) / W_1 \quad (2)$$

## 2.6 Absorption under load (AUL)

To measure the absorbency under load (AUL) of SAPs, dried SAP sample (0.5 g) was weighed, uniformly dispersed on the surface of polyester gauze. Polyester gauze was on a microporous sintered glass filter plate placed in a Petri dish. A cylindrical solid load was put on the dry SAP particles in a glass cylinder. The load with pressure of 0.3 psi was placed on the sample. Then, 0.9% saline solution was added to the Petri dish. After 1 h, the swollen particles

were removed and reweighed. The AUL value was calculated via formula (3), where  $W_1$  and  $W_2$  are the weights of dried hydrogel and swollen hydrogel [3]:

$$AUL=(W_2-W_1)/W_1 \quad (3)$$

## **2.7 Surface treatment**

Certain amounts of a solvent containing acetone and DW were used to dissolve given amounts of the cross-linking agent (DEGDGE). The resultant solution was then used for soaking the dried SAP particles. They were allowed to remain at room temperature for 30 min. The soaked particles were filtered and cured at a temperature of 140 °C for different times. The amount of SAP particles in the solution was fixed at 1 g SAP per 20 g of the treating solution.

## **3. Results and discussion**

The mechanism of the cross-linking of hydrogels in solution by gamma irradiation was studied by Rosiak et al. [4]. Here, the mechanism is discussed briefly for  $\gamma$ -irradiation of aqueous AA solution. Polymerization reaction will be occurred in these solutions with the following the radiation mechanism stages such as initiation, propagation and termination. In the initiation stage, radiolysis of water occurs, resulting in H and OH radicals. The radicals are recombined to produce  $H_2$  and  $O_2$  gases. In addition, the H or OH radicals attack the double bond of acrylic acid and sodium acrylic acid to produce their radicals, respectively. The reactions of these radicals proceed to the propagation stage, ultimately resulting in the formation of a stable product [3]. In this study, methods of improving the performance and main factors that affect the radiation synthesis of SAPs, such as radiation dose, monomer concentration and neutralization degree were discussed.

### **3.1 Effect of the radiation dose and monomer concentration on the gel formation**

A series of aqueous acrylic acid solutions containing 5, 10 and 15 wt.% monomer was irradiated at the range of 10-25 kGy. The results of sample irradiation are shown in Table 1. Samples in Table 1 were designated as the amount of dose, concentration and neutralization degree respectively. It shows that at AA concentration of 5 wt.%, the monomer concentration was insufficient to generate an interconnected three-dimensional network, resulting in low-viscosity solutions rather than stable hydrogels. Qualitative gel formation was observed at the concentration of 10 % in all absorbed doses from 10 to 25 kGy while the viscosity increases with increasing

absorbed doses. At the concentration of 15% with increasing radiation dose from 10 to 15 kGy the percentage of gel increases and an elastic form of gel is formed. The gel fractions of samples with 15% concentration are shown in Fig. 1. The gel fraction is 70 % at the dose of 10 kGy. The value of gel fraction reaches to 85.7 % at the radiation doses of 15 kGy. After that, it gradually declines to 74% at 25 kGy. These results display that irradiation plays a dual role in the formation of acrylic acid-based hydrogels by simultaneously inducing cross-linking and chain scission reactions. Upon irradiation, water molecules undergo radiolysis to generate highly reactive species, including hydroxyl radicals, hydrogen atoms, and hydrated electrons. These reactive species initiate the formation of free radicals on acrylic acid and sodium acrylate molecules, leading to chain propagation and intermolecular recombination. At relatively low irradiation doses, the concentration of polymer radicals is sufficient to promote intermolecular crosslinking, resulting in the formation of a three-dimensional network and a progressive increase in gel fraction. Consequently, the gel fraction reaches its maximum at 15 kGy, where the balance between radical generation and recombination favors network formation. However, further increasing the irradiation dose produces an excessive number of radicals, which enhances chain scission and oxidative degradation reactions. These degradation processes compete with the crosslinking mechanism, reducing the effective cross-link density and leading to a gradual decrease in gel fraction at higher doses. Therefore, the observed variation in gel fraction reflects the competition between radiation-induced cross-linking and degradation processes.

Table 1: Effect of the radiation dose and monomer concentration on the gel formation

| <b>sample</b> | <b>Monomer (%)</b> | <b>Dose (kGy)</b> | <b>Gel mode</b>     |
|---------------|--------------------|-------------------|---------------------|
| 10K-5-0.5     | <b>5</b>           | <b>10</b>         | <b>dilute</b>       |
| 15K-5-0.5     | <b>5</b>           | <b>15</b>         | <b>dilute</b>       |
| 20K-5-0.5     | <b>5</b>           | <b>20</b>         | <b>dilute</b>       |
| 25K-5-0.5     | <b>5</b>           | <b>25</b>         | <b>dilute</b>       |
| 10K-10-0.5    | <b>10</b>          | <b>10</b>         | <b>low viscose</b>  |
| 15K-10-0.5    | <b>10</b>          | <b>15</b>         | <b>low viscose</b>  |
| 20K-10-0.5    | <b>10</b>          | <b>20</b>         | <b>high viscose</b> |
| 25K-10-0.5    | <b>10</b>          | <b>25</b>         | <b>high viscose</b> |
| 10K-15-0.5    | <b>15</b>          | <b>10</b>         | <b>gel</b>          |
| 15K-15-0.5    | <b>15</b>          | <b>15</b>         | <b>gel</b>          |

|            |    |    |     |
|------------|----|----|-----|
| 20K-15-0.5 | 15 | 20 | gel |
| 25K-15-0.5 | 15 | 25 | gel |

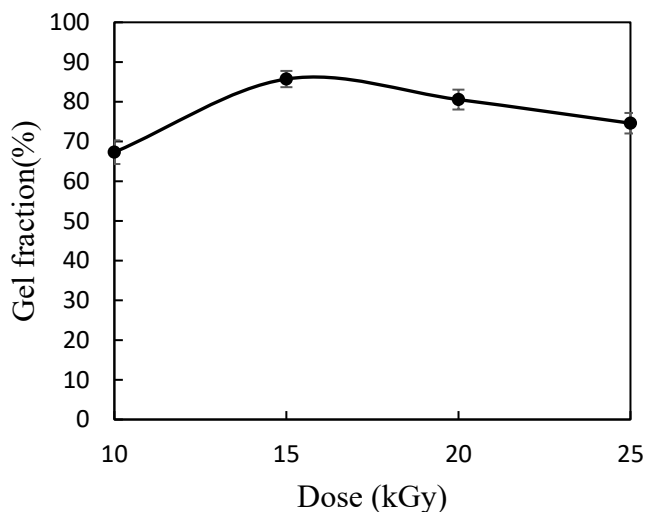


**Fig. 1** Effect of irradiation dose on the gel fraction of SAP samples at 15% concentration of monomer

### 3.2 FTIR Characterization of SAP

FTIR spectra of acrylic acid and SAP formed at 15 kGy with the highest gel fraction (15K-15-0.5) are shown in Fig. 2. Compared to the monomer, some peaks have disappeared in the synthesized polymer. In Fig. 2a related to acrylic acid, the peak assigned to the vibration of C-H at  $985\text{ cm}^{-1}$  related to out-of-plane deformation of the vinyl C-H band is very clear; however, it disappears in the spectrum of the polyacrylic acid gels (Fig. 2b), showing that polymerization has occurred. Carboxylic acid group in AA also has strong C=O peak, which appears at  $1728\text{ cm}^{-1}$ . While the major peak in the SAP spectrum is the C=O stretch at  $1730\text{ cm}^{-1}$ . The disappearance of the vinyl C=C band together with the persistence of the carbonyl absorption confirms successful polymerization and the formation of cross-linked poly(acrylic acid-co-sodium acrylate network). In addition, the characteristic peaks at  $2854$  and  $2925\text{ cm}^{-1}$ , attributed to the aliphatic C-H group in synthesized gels, as well as the blue shift of OH vibration from  $3080\text{ cm}^{-1}$  in AA to  $3438\text{ cm}^{-1}$  in the SAP indicating successful polymerization [13, 14].

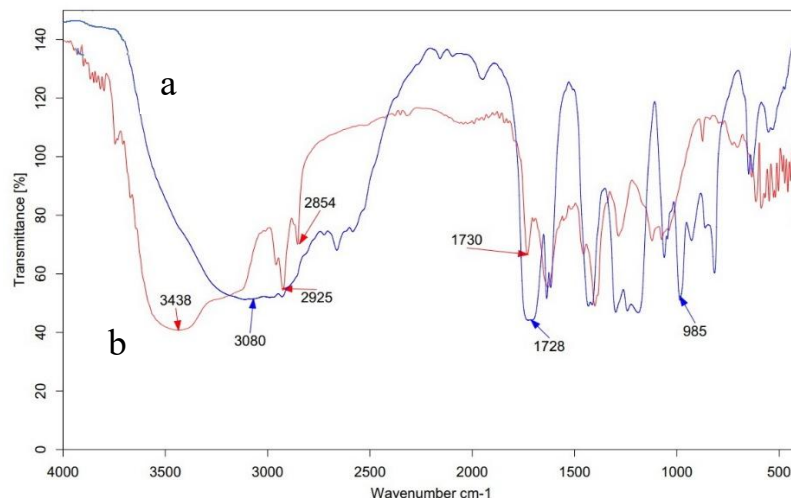
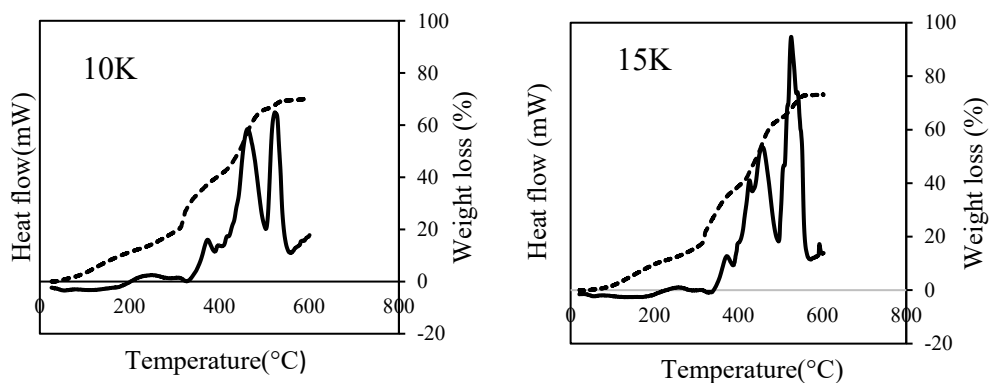


**Fig. 2** FTIR spectrum of a) acrylic acid monomer and b) polymer sample 15K-15-0.5

### 3.3 Thermal analysis

Four superabsorbent hydrogel samples (10K-15-0.5, 15K-15-0.5, 20K-15-0.5, 25K-15-0.5) were structurally characterized through differential scanning calorimetry (DSC) analysis (Fig. 3). As shown in Fig. 3 in all absorbed doses, the two thermal events observed between 300 and 500 °C are attributed to the multistep degradation of the crosslinked poly (acrylic acid-co-sodium acrylate) network. The first peak is associated with decarboxylation and decomposition of the carboxylate groups, whereas the second peak corresponds to degradation of the polymer backbone and decomposition of the more highly crosslinked network [15, 16].



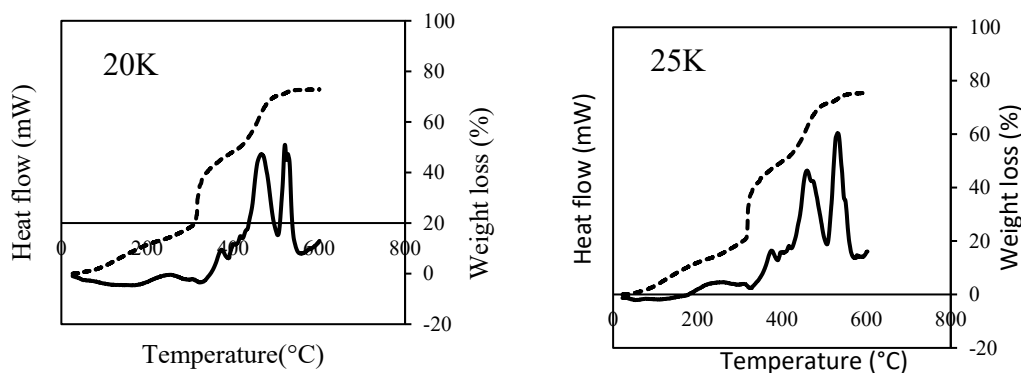


**Fig. 3** DSC analysis of SAPs samples

Fig. 4 also shows the results of thermal degradation in terms of percentage weight loss-temperature. Initial weight loss below 100 °C is attributed to the moisture evaporation. Also, the decrease in initial weight in the temperature range of 100 to 300 °C can be caused by the evaporation of water from the superabsorbent networks and the evaporation of bonded water. The 10K-15-0.5, 15K-15-0.5, 20K-15-0.5, 25K-15-0.5 samples show a significant weight loss at 314 °C, 319 °C, 315 °C, 316 °C respectively at the first stage of degradation. In comparison to other samples, 15K-15-0.5 sample starts to degrade at the highest temperature, suggests a relatively higher cross-link density, confirming gel fractions data. The final weight loss for the samples irradiated at 10, 15, 20 and 25 kGy are 71, 73, 72 and 76% respectively, showing that greater degradation occurs at 25 kGy. This trend is consistent with increased chain scission.

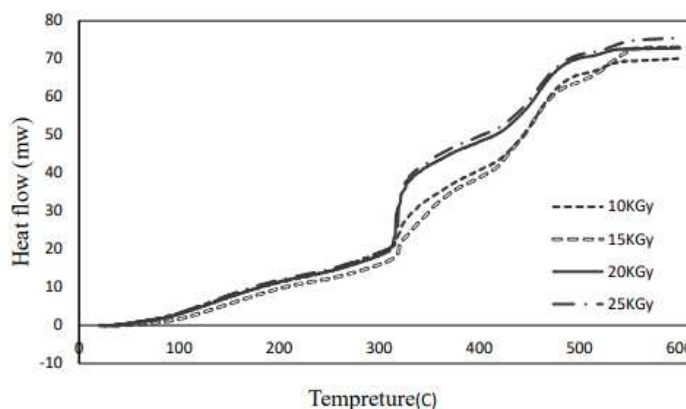


**Fig. 4** The results of thermal degradation of the SAP samples irradiated at 10 to 25 kGy

### 3.4 Swelling behavior

Fig. 5 illustrates the equilibrium swelling of SAPs which are obtained from 15% AA solution as a function of the irradiation dose in distilled water and saline solutions. It is observed that the swelling ratios of hydrogels in deionized water are 333, 373, 423 and 362 g/g at the dose of 5, 10, 15 and 25 kGy, respectively. The maximum swelling ratio of hydrogel was reached to 423 g/g (shown in Fig. 5) at the dose of 15 kGy. The maximum swelling ratio is obtained from hydrogel at the dose of 15 kGy indicating the highest possibility of repealing carboxylate anions in the hydrogel networks. The swelling of SAPs in saline solution at the dose range of 10-25 kGy is in the range of 56-62 g/g, demonstrating no significant changes at different absorbed doses. Also, the amount of AUL is in the range of 13 -13.8 g/g, representing the small effect of radiation cross-linking in the range of 10-25 kGy on AUL amounts.

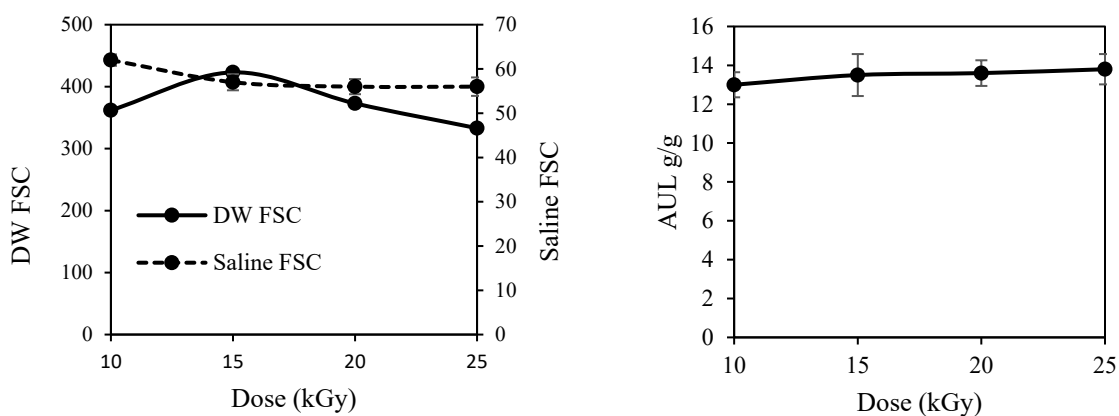


**Fig. 5** Effect of irradiation dose on free (left) and loaded (right) absorption of the SAP samples

### 3.5 Effect of neutralization on swelling behavior

The effect of neutralization degree (Dn) on the FSC and AUL of the 15k-15-0.5 sample is presented in Fig. 6. The swelling ratios of hydrogels with neutralization degree of 0.25, 0.5 and 0.75 is approximately 347, 423, and 412 g/g for DW solution, and 46, 57, and 78 g/g for saline solution. The effect of neutralization degree was also investigated on AUL. The results show that increasing the neutralization degrees from 0.25 to 0.75 results in an increase in AUL from 13.2 to 15.6 g/g. When the ND content is low, the acidity of the aqueous phase is high, causing the

protonation of carboxylate anions under acidic conditions. This phenomenon results in a decrease in electrostatic repulsion. As the ND content rises, there is an increase in both the hydrophilic groups and sodium content within the branch chain, which subsequently enhances the electrostatic repulsion and internal osmotic pressure of the cross-linking network. Such changes are beneficial for improving absorbency [14].

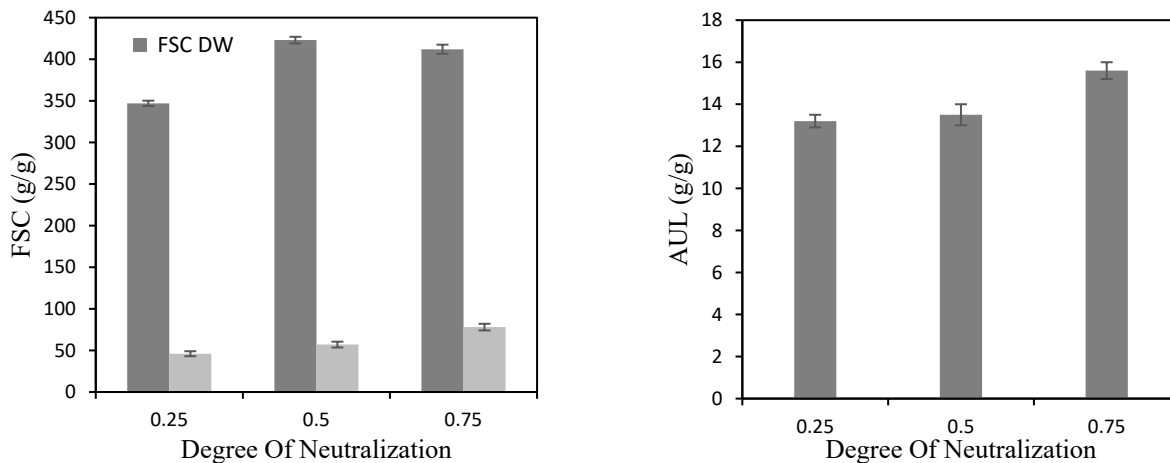


**Fig. 6** Effect of neutralization degree on the FSC and AUL of the 15k-15-0.5 sample

### 3.6 SEM analysis

The SEM image illustrates the geometry and dimensions of the pores in relation to their specific positions within the hydrogels. As a result, it provides relevant insights into both the uniformity and diversity of the hydrogels' network. For this purpose, SEM images of the sample with higher swelling ratio, the 15K-15-0.5 sample, was taken and shown in Fig. 7. The hydrogel exhibits a surface characterized by irregularly shaped pores. This porous configuration enhances the surface area of the superabsorbent polymer (SAP), allowing it to retain a greater volume of water.

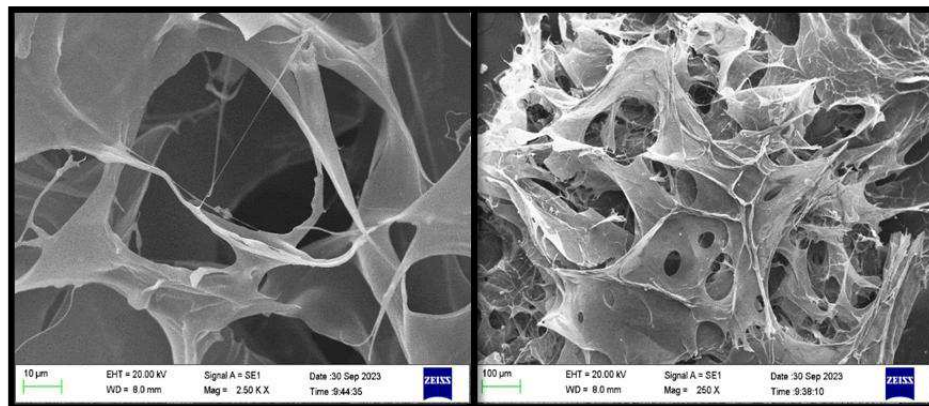


**Fig. 7** SEM images of 15k-15-0.5 sample with two magnifications

### 3.7 Residual monomer

The reduction of residual monomer is a critical necessity for the production of mature industrial products, especially in categories like superabsorbent polymers (SAPs), which are primarily made from sodium polyacrylic acid (PAA-Na). Based on the reports, the amount of residual monomer in thermal initiation methods is higher than 200 ppm [17, 18]. In this study, instead of thermal initiators, radiation was used to initiate the polymerization. Here, the synthesized sample with higher swelling ratio, 15K-15-0.5, was selected and its residual monomer was measured by means of high-performance liquid chromatograph. Fig. 8 indicate the chromatograph of standard solution of 1 ppm (red line) and sample solution black line. The content of AA was calculated from the area under the peak after around 7 min from injection of the sample as shown in the diagrams. It can be seen that the content of residual monomer in the sample is lower than 1 ppm, indicating the irradiation as a suitable method for medical SAP synthesis.

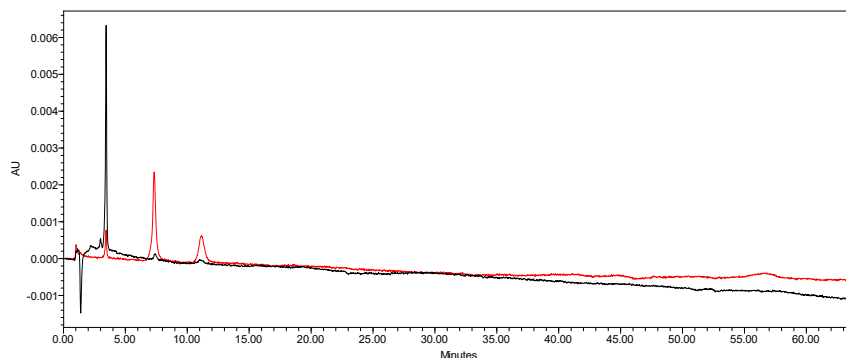
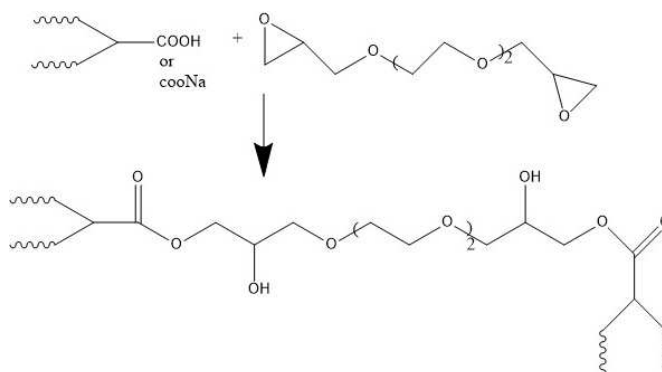


**Fig. 8** HPLC diagram of standard solution (red line) and 15K-15-0.5 sample solution (black line)

### 3.8 Post treatment of SAP

Post surface treatment was performed to enhance the sample swollen gel modulus. The duration and temperature of post-treatment significantly influence the properties of SAP, as they not only improve the efficiency of the reaction but also contribute to degradation. Here, the 15K-15-0.5 sample was post-treated via the surface cross-linking processes by DEGDGE at the temperature of 140 °C for different times. Free swelling behavior in saline and DW solutions as well as AUL was shown in Fig. 9. As shown in Fig. 9a, there are no significant changes on free swelling behavior in saline and DW solutions at all the times before and after treatment. A slight downward trend in swelling capacities is probably due to moisture evaporation with the passing of time, leading to an increase in the  $T_g$  [1]. This increase in  $T_g$  can result in a reduction in diffusion into a deeper length for the reagent. The results in Fig. 9b show that the maximum amount of the AUL, 29.4, is achieved after 30 min surface treatment. The AUL enhancement from 13.5 to 29.4 by DEGDGE showed that surface treatment can increase cross-link density in the SAP surface through DEGDGE reaction with SAP's carboxylic acid groups (scheme 1). As shown in Scheme 1, the carboxylic acid groups in SAP may interact with the oxirane rings in the external cross-linker DEGDGE, resulting in the opening of the epoxy rings and their incorporation into the polymer chains.



**Scheme 1** The interaction between the carboxylic acid group of SAP and the oxirane ring present in the external cross-linker DEGDGE.

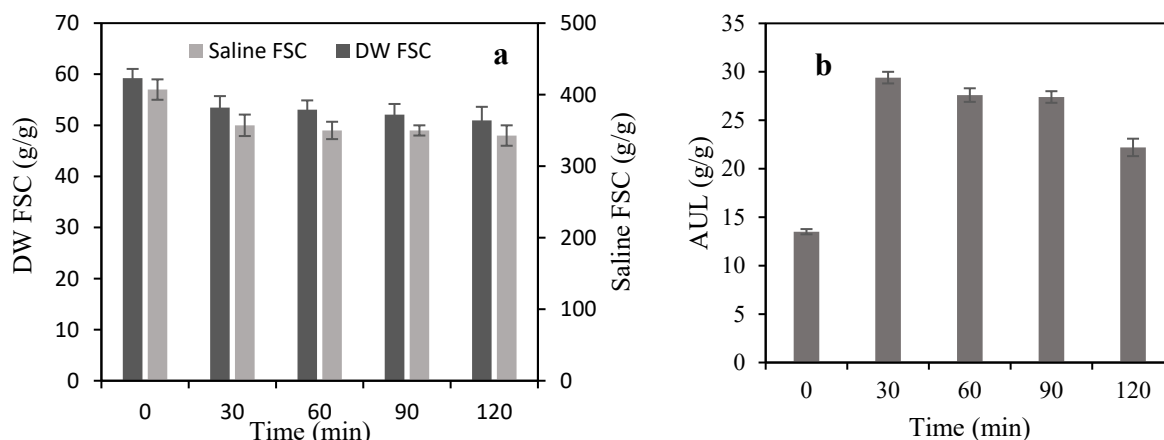


**Fig. 9** Effect of surface treatment time on a) swelling capacity in DW and saline solution and b) AUL

### 3.9 FTIR spectroscopy

The ATR-IR spectroscopy technique was utilized to identify the formation of ester groups resulting from the reaction between  $\text{-COOH}$  groups and DEGDGE. Fig. 10 displays the ATR spectra of 15K-15-0.5 sample and cross-linked surface modified one by DEGDGE. As illustrated in Fig. 10, the absorption peak at  $3358\text{ cm}^{-1}$  corresponds to the stretching vibrations of OH bonds. Following modification, this peak shifts to  $3448\text{ cm}^{-1}$ , indicating the development of a weaker hydrogen bond in comparison to that found in carboxylic acid. The broad absorption peak at  $1167\text{ cm}^{-1}$  attributed to C-O-C which is additional component within the modified SAP network structure. The observed shift in the OH peak and the detection of the C-O-C peak in the spectrum of the modified SAP suggest that the carboxylic acid groups present in SAP may interact with the oxirane rings found in the epoxy cross-linkers. This interaction likely results in the ring opening of the epoxy groups, facilitating the linking of polymer chains. Furthermore, as previously stated, the glycidyl groups present in epoxy resin exhibit a tendency to react with the carboxylic pendant groups of the superabsorbent. Following the modification reaction, the C=O peak of the carboxylic acid is observed to shift to a higher wavenumber (from  $1639$  to  $1717\text{ cm}^{-1}$ ). It is important to highlight that, owing to its high polarity, the carboxyl group can establish stronger hydrogen bonds in comparison to alcohols.

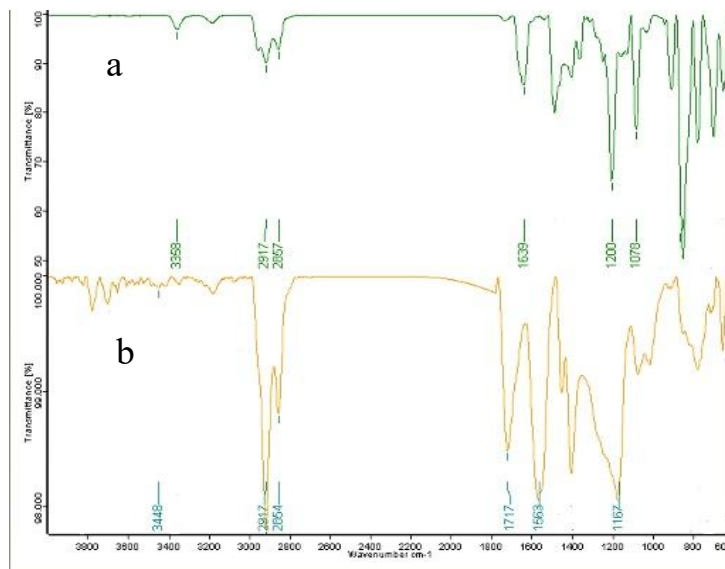


**Fig. 10** ATIR spectra of (a) base SAP and (b) surface modified SAP in the presence of DEGDGE

### 3.10 Rheological analysis

Figure 11 shows storage modulus versus angular frequency for 15K-15-0.5 SAP and surface treated SAP using DEGDGE. In all frequency the SAP has the low storage modulus and surface treated SAP show enhanced storage modulus. The higher storage modulus observed in the treated SAP samples indicates a network with a greater cross-linking density near the hydrogel surface. Surface cross-linking effectively increases the overall cross-linking density within the hydrogel, thereby improving its mechanical strength. The AUL and rheological measurements both show that DEGDGE is an effective surface cross-linking agent [19].

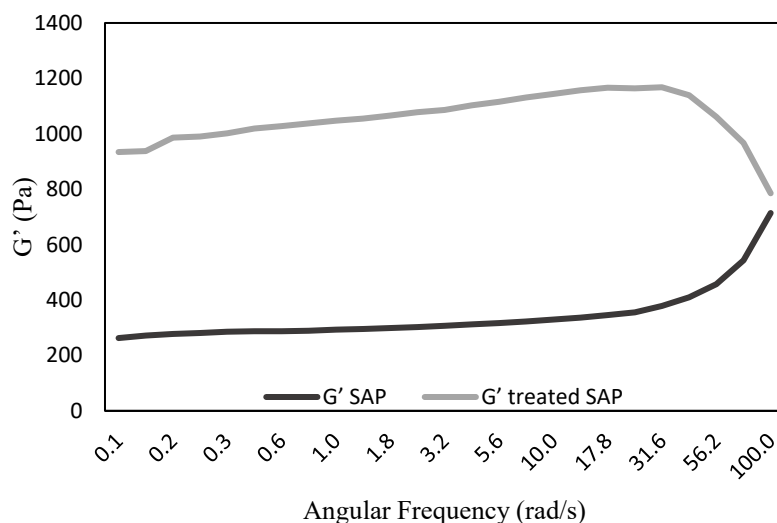


**Fig. 11** Storage modulus vs. angular frequency for SAP and surface-treated SAP with DEGDGE

#### **4. Conclusion**

Polyacrylic acid superabsorbent hydrogel with low residual monomer content was successfully synthesized via gamma irradiation. The results show that monomer concentration, dose amount, neutralization degree are the main factors that affect the swelling behavior. Water uptake potential of the optimum sample, 15K-15-0.5, goes as high as 42,300 % of its own weight in a short period of time. SEM micrographs illustrated interconnected porous networks, DSC and Surface modification using DEGDGE effectively increased gel strength while preserving swelling capacity. Optimal surface curing time identified as 30 min for maximized mechanical and swelling performance balance. Based on the results, this green, efficient method offers a compelling strategy for producing SAPs for commercial applications requiring high purity and mechanical resilience, including hygiene products and medical devices.

#### **Data availability**

The datasets used and/or analyzed during the current study available from the corresponding author on reasonable request.

#### **Author contributions**

**Azam Akhavan:** Conceptualization, Supervision, Writing – review & editing, Writing – original draft, Data curation, **Farahnaz Nasiri:** Writing – review & editing, Investigation, Data curation. **Vagihe Nikfarjam:** Writing – review & editing, Formal analysis. **Peiman rezaeian:** Writing – review & editing, Formal analysis. **Mohamad reza Dadfar:** Writing – review & editing, Formal analysis.

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