ISSN: 0067-2904

Preparation and evaluation of superabsorbent hydrogels for Removing Water from Crude fuel and Gasoline

Mohammed Haitham Ahmed*, Nadia A. Hussein Al-Assady
Department of Chemistry, College of Education for Pure Sciences, University of Basrah, Basrah, Iraq

---

Received: 16/6/2024 Accepted: 30/10/2024 Published: 30/11/2025

Abstract

This research aimed to create eco-friendly superabsorbent gels that excel in swelling and absorption capacity, rapid swelling rate, affordability, and durability, with the ultimate goal of effectively removing water from crude oil and gasoline practically and sustainably. Superabsorbent hydrogels are composed of two main components. The first component is linear polyacrylamide, which acts as a polyelectrolyte. The second component includes varying amounts of acrylamide as a monomer with N, Ǹ-Methylene bis-acrylamide as a cross-linking agent and sodium persulfate as an initiator. This composition is developed using the semi-interpenetrating polymers network technique (semi-IPNs). The hydrogel was characterized using FT-IR, SEM, and XRD techniques. The swelling characteristics was examined of each semi-IPN hydrogel by quantifying the swelling ratio (SR). It was found that it varied from 589 to 5432 ٪depending on component quantities and environments. The results showed that the semi-IPNs containing 2 g acrylamide (sample C) have a high SR as compared to semi-IPNs containing 1 g (sample A) in distilled water and alkaline media but less SR in the acidic medium. The effects of various salt solutions on swelling behavior and the efficacy of semi-IPNs in removing water from crude fuel and gasoline were discussed. The resulting hydrogel after revision of the synthesis temperature indicated an increased swelling rate within a value of SRs ranging from 2700% to 4524% in the water-crude oil mixture and 2700% to 5644% in the water-gasoline mixture when subsequent tests were carried out.

Keywords: Water content; Crude Oil; APAM polyelectrolyte; Gasoline ; superabsorbent hydrogels.

تحضير وتقييم هلاميات فائقة الامتصاص لإزالة الماء من الوقود الخام والبنزين

محمد هيثم احمد* , نادية عاشور حسين
قسم الكيمياء , كلية التربية للعلوم الصرفة , جامعة البصرة , العراق

الخلاصة:

تهدف الدراسة الحالية الى تحضير هلاميات فائقة الامتصاص صديقة للبيئة تعمل على إزالة الماء من النفط الخام والجازولين بشكل عملي ومستدام . تم تحضير الهلاميات من البولي الكتروليت وكميات مختلفة من الاكريلاميد و N,N-مثيلين بس اكريلاميد كعامل شابك وبيرسلفات الصوديوم كعامل بادئ. درجة انتفاخ الهلاميات المحضرة تم قياسها اعتمادا على مجاميعها الوظيفية في أوساط حامضية مختلفة (pH1.2

---

* Email: pgs.mohammed.haithem@uobasrah.edu.iq

pH7.2, , pH12) إضافة الى المحلول الملحي الالكتروليتي NaCl بتراكيز (15,10, 5,1) %. تم تشخيص الهلاميات باستخدام مطيافية الاشعة تحت الحمراء وتم دراسة تحليل XRD لتقدير التراكيب البلورية وجهاز المسح الالكترونيSEM لدراسة بنية السطح . أظهرت النتائج ان نسبة الانتفاخ تعتمد على تركيز الاكريلاميد ودرجة التشابك وان جميع الهلاميات تمتلك نسب انتفاخ تتراوح بين 589٪ إلى 5432٪ وكانت نسبة الانتفاخ في المحيط المتعادل والقاعدي اعلى مما هو عليه في المحيط الحامضي وان نسبة الانتفاخ تنخفض بزيادة تركيز المحلول الملحي . كذلك تم تقييم كفاءة الهلاميات المحضرة في إزالة الماء من مستحلبات النفط الخام والجازولين واستعادة الهلاميات بعد عدة دورات من الاستخدام والتجفيف .

1-Introduction

At a crude fuel production site, a significant volume of aqueous liquid is generated alongside the crude fuel, coming from sources such as fuel reserve, sea, or ground is produced together with the crude fuel, of which amount can be as high as 15–80 % of the total fluid produced [1]. The presence of water in the crude oil is undesirable for several reasons: it reduces the heating value and the quality of the crude fuel; it causes corrosion in the processing facilities; it increases the load on the wastewater treatments and the associated cost; and, more importantly, it increases the extra energy and cost for the removal of the water from the crude fuel [2]. To satisfy global energy demands and enhance production efficiency, it is essential to reduce the cost and energy involved in the removal of water from crude fuel [3]. Currently, the removal of water from crude fuel is primarily achieved through methods such as heating, chemical treatment, gas flotation, and filtration. Heating is the simplest method; however, it is not very effective, especially for heavy fuel, viscous fuel, and fuel with high pour points and cloud points. While the other methods are generally more effective, they often require large amounts of energy and chemical agents, and they are sometimes very complex with unsteady performance [4]. An alternative solution involves the use of water-absorbing polymers. Although compared to the methods mentioned above, this method is more suitable for the offsite application of dewatering the storage tank or the transportation of crude fuel. It involves adding hydrogels to the vessel or the portable mixing unit with the crude oil and leaving it for some time before the hydrogel is removed from the imbibed water. After the hydrogels are available on the market, the most effective way is to develop a custom hydrogel optimized for the specific type of crude fuel and the operating conditions. This customization can be achieved by thoroughly understanding the interaction between the hydrogel, the crude oil, and the environment in which the hydrogel is required to perform [5]. Intelligent materials, commonly known as superabsorbent gel [6], are highly valued for their remarkable characteristics and diverse applications [7-9]. These networks are three-dimensional structures made of polymers that can dissolve very small amounts of water and biological fluids [10-12]. The creation of the network structure may entail the copolymerization of one or more components by ionizing radiation or chemical starting methods [13,14], in contrast to interpolymer. The ability of polymer chains to absorb water from the presence of hydrophilic groups such as -OH, -CONH, $-CONH_2$, -COOH, and $-SO_3H$ [15-17]. These groups enable the polymers to assimilate extra biological fluids and maintain a significant amount of fluid when expanded [18,19]. The establishment of a crosslinking architecture is crucial for the production of hydrogels, as it guarantees their stability in water-based surroundings [20]. Superabsorbent hydrogels are three-dimensional hydrophilic polymer networks through the formation of chemical bonds [21] as well as ionic [22] and supplementary forces [23]. Through hydrophobic or hydrogen bonding interactions, these hydrogels can retain and absorb a significant amount of water while maintaining their physical size [24, 25]. The absorption and retention of water are influenced by the specific type of hydrophilic monomer [26-28] Additionally, the type of polymer used in their production process [29,30], the characteristics of the expanding medium, and the extent of crosslinking [31]. This study aims to develop IPN hydrogels for dewatering crude oil and gasoline. The hydrogel is a cross-linked polymer that swells to a high degree in water and retains a considerable amount of it. The hydrogel being developed must be capable to withstand the high temperatures of the reservoir conditions, it must be elastic and not brittle and it must be resistant to degradation.

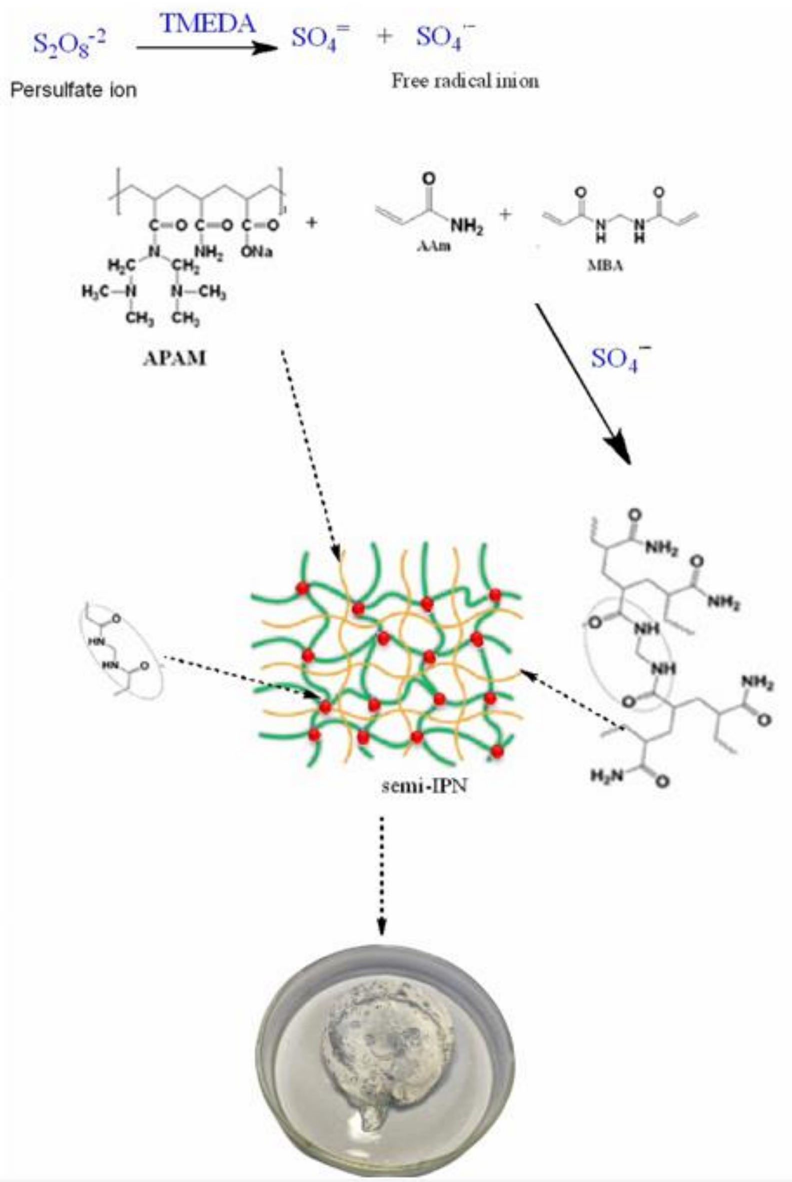
2- Materials and methods

2-1 Materials

The chemicals used in this study were supplied from several sources. Acrylamide (AAm) was supplied by Aldrich Co. (Germany). Ethanol, N, N, Ǹ, Ǹ-Tetramethylethylene diamine (TMEDA), N, Ǹ-Methylene bis-acrylamide (MBA), and sodium persulfate (SPS) were supplied by Fluka Co. (Switzerland). The polyacrylamide APAM, (polyelectrolyte), was supplied by the State Company for Petrochemical Industry in Basra, Iraq. The investigation was carried out utilizing purified water acquired from the Department of Chemistry at the University of Basra, also located in Iraq.

2-2 Preparation of the New IPNs Gel

Dissolve 1 g of the polyelectrolyte APAM in 25 mL of distilled water while continuously heating and stirring at 50°C until a homogeneous solution is obtained. In a separate bowl, dissolve varying amounts of acrylamide (AAm), MBA, and SPS (see Table 1) in 10 mL of distilled water. Gradually add the acrylamide solution to the polyelectrolyte solution with constant stirring for 30 minutes, then heat the mixture in a water bath at 50 °C under nitrogen protection. Nest, sodium persulfate was dissolved in 5 mL and added as an initiator for the acrylamide polymerization process. Following this, 7.0 mL of tetramethylethylenediamine (TMEDA) was added as an accelerating agent with good stirring for 30 minutes until the polymerization and crosslinking were completed. Cut the resulting hydrogel into small pieces. Wash it once with water and leave it to dry at a temperature of 40°C for one day [32].



Scheme 1: The synthesis of superabsorbent semi-IPN hydrogel

Table 1: Amounts of acrylamide, crosslinking agent and initiator

Hydrogel code	APAM (g) polyelectrolyte	AAm (g) monomer	MBA (g) crosslinking agent	SPS (g) initiator
A	1	1	0.2	0.07
B	1	1.5	0.2	0.07
C	1	2	0.2	0.07
D	1	2	0.4	0.07
E	1	2	0.6	0.07
F	1	2	0.2	0.03
G	1	2	0.2	0.1

2-3 **Characterization**

Fourier transform infrared (FT-IR) was used to characterise the synthesized polymer. The experiment used KBr particles coated with Perkin-Elmer model 4100 type A (USA) and covering a wavelength range from 400 to 4,000 cm^{-1}. The produced samples were examined using the X-ray diffraction (XRD) method on a Philips X'Pert-MPD system. The analysis was conducted at a Cu Kκ wavelength of 1.5418.

2-4 **Swelling Measurement**

Dried pieces of APAM IPN hydrogel were used to assess the degree of swelling. To determine the swelling ratio (SR), the hydrogels (0.25± 0.01 g) of each sample were immersed in 100 mL of distilled water for 30 minutes at room temperature. Samples were collected from the water every 30 minutes, dried to remove surface water using filter paper (110 mm, CAT 62010110), weighed, and the swelling ratio (SR) was calculated using the equation (1) [33]:

$$\%SR = \frac{Ws - Wd}{Wd} X100 \qquad \dots\dots\dots\dots 1$$

Where SR= swelling ratio, Ws =weight of swollen gel, and Wd= weight of dry gel, respectively.

2-5 **Saturated crude Fuel - Gasoline Samples**

To achieve a saturated state, 50 mL of fuel samples were mixed with 50 mL of distilled water to create a mixture of petrol and crude fuel. After confirming proper phase contact by rapidly agitating the mixture for 30 minutes using a magnetic mixer and leaving it to settle for 2 hours, 0.25 g of dehydrated APAM IPN hydrogel was introduced into the saturated solution and left to soak at room temperature. After a three-hour interval, the hydrogel fragments retrieved from the saturated solution were weighed. The swelling ratio was calculated after using filter paper filter paper (110 mm, CAT 62010110) to separate the surface water [33].

3- **Results and discussion**

3.1 **The FT- IR spectrum**

From the FT-IR spectra of AAm monomer, APAM polyelectrolyte, and semi-IPN hydrogel revealed that all characteristic peaks of Aam and APAM were present in the semi-IPN hydrogel. This indicates a compatibility of the APAM and semi-IPN hydrogel. IR spectra are shown in **Figures** (1-3), the data shown in **Table** 2.

Table 2: Interpretation of FT-IR spectrums of AAm monomer, APAM polyelectrolyte, and semi-IPN hydrogel

S.no	IR Spectrum	Peaks(cm^{-1})	Groups	Stretching / Deformation
1	AAm	(3340.33-3171.99)	N-H	Stretching
		2811.94	C-H (alkyl)	Stretching
		1665.44	C=O(amide)	Stretching
		1465.11	C=C	Stretching
		1608.33	N-H(amide)	Bending
		1276.87	C-O	Stretching
2	APAM	(3479.58-3415.93)	N-H	Stretching
		1662.49	C=O	Stretching
		2920.23-2850.79	C-H (alkyl)	Stretching
		1128.36	C-O	Stretching
		1562.34	N-H(amide)	Bending
3	semi-IPN hydrogel	3400.27	N-H	Stretching
		(2933.50-266.02)	C-H(alkyl)	Stretching
		1689.64	C=O	Stretching
		1168.78	C-O	Stretching
		1409.96	N-H	Stretching

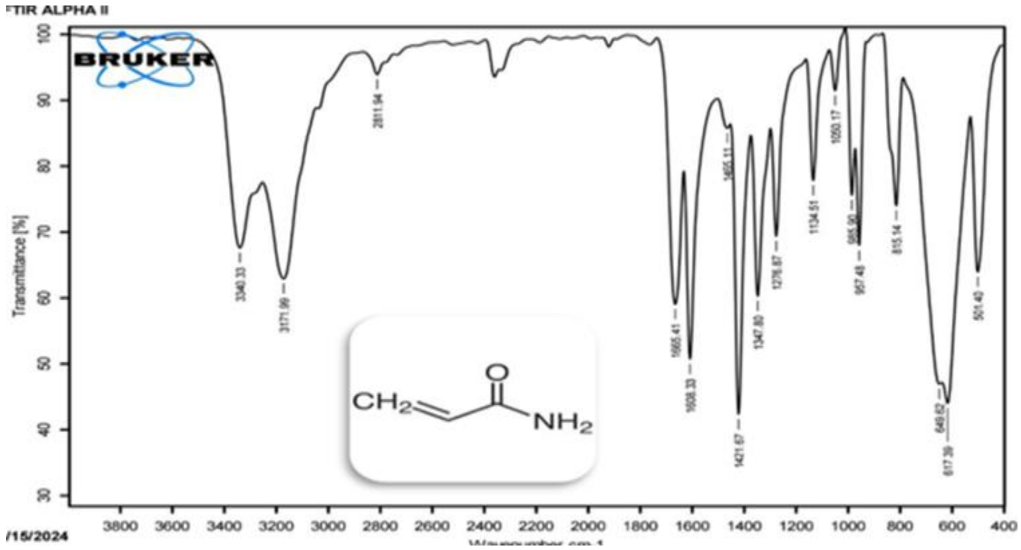


Figure 1: FT-IR of acrylamide monomer

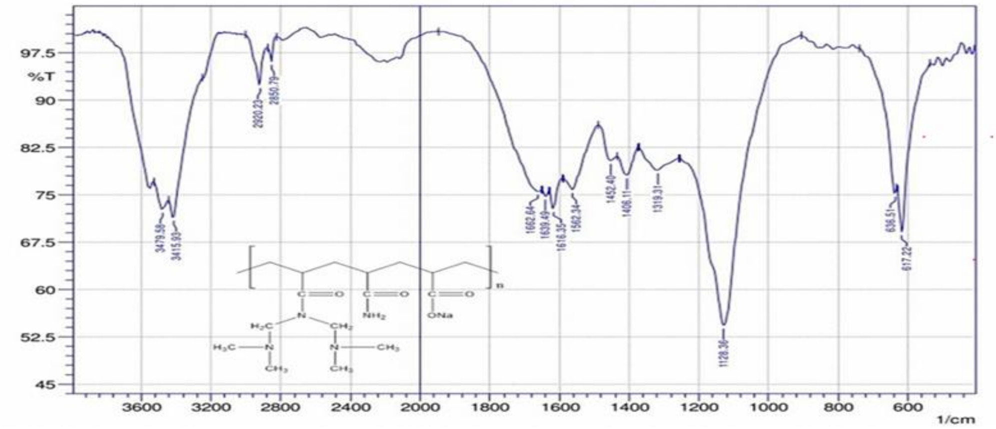
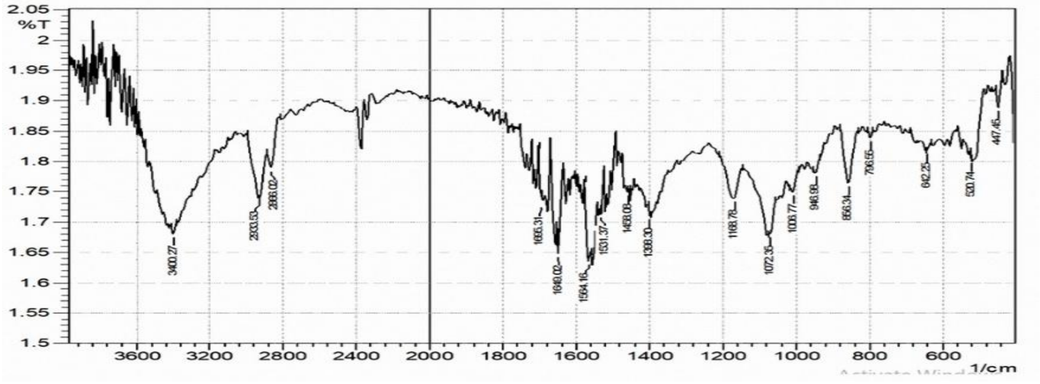


Figure 2: FT-IR spectra of the APAM polyelectrolyte



c

Figure 3: FT-IR of the semi-IPNs hydrogel

3.2 X-ray diffraction (XRD)

Figure 4 depicts the X-ray diffraction pattern of the semi-IPN hydrogel, which consists of a polyelectrolyte and a polyacrylamide superabsorbent. The acrylamide spectra displayed a single band, featuring a prominent peak at 2θ 24.6°. In contrast, the APAM polyelectrolyte pattern exhibited two distinct peaks at 21.07° and 26.1°, suggesting that the polyelectrolyte phase is amorphous. The pattern of the semi-IPN hydrogel revealed two distinct peaks at 2θ 6.458° and 26.76°. The patterns exhibited by the APAM polyelectrolyte and the semi-IPN hydrogels are similar, with slight differences in the peaks, and are higher than acrylamide.

This difference is attributed to an increase in the degree of ordering of chains of the prepared hydrogels that the combination between AAm and MBA individually increases the crosslinking of the hydrogel structure during the polymerization process. Also, due to an increase in H-bonding interactions among the amide group of AAm and carboxylic, groups of APAM chains lead to increased crystallinity of hydrogels than AAm [33].

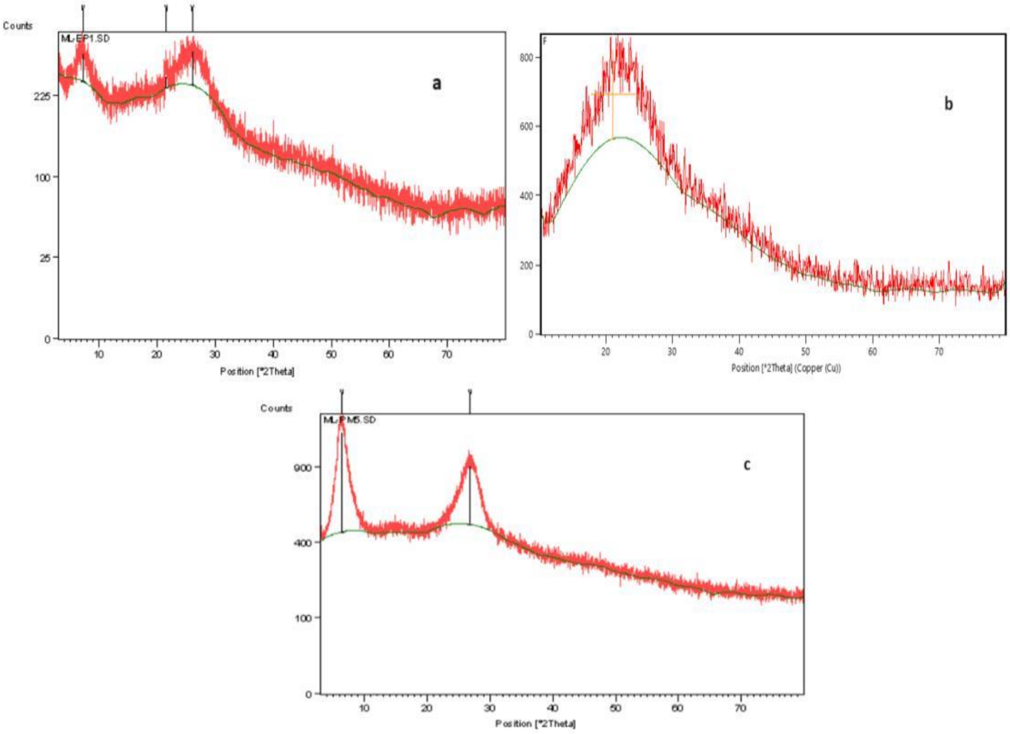


Figure 4: XRD patterns of a) APAM polyelectrolyte, b) AAm, and c) semi-IPN hydrogel

3.3 **SEM studies**

To ensure the structural integrity of the superporous hydrogel, scanning electron microscopy (SEM) was performed. Figure 5 demonstrates that the hydrogel possesses a porous structure, facilitating rapid expansion. The highly porous nature of hydrogels enables increased water absorption within the material. The hydrogel's highly permeable structure allows for increased water entry through the pores [34].

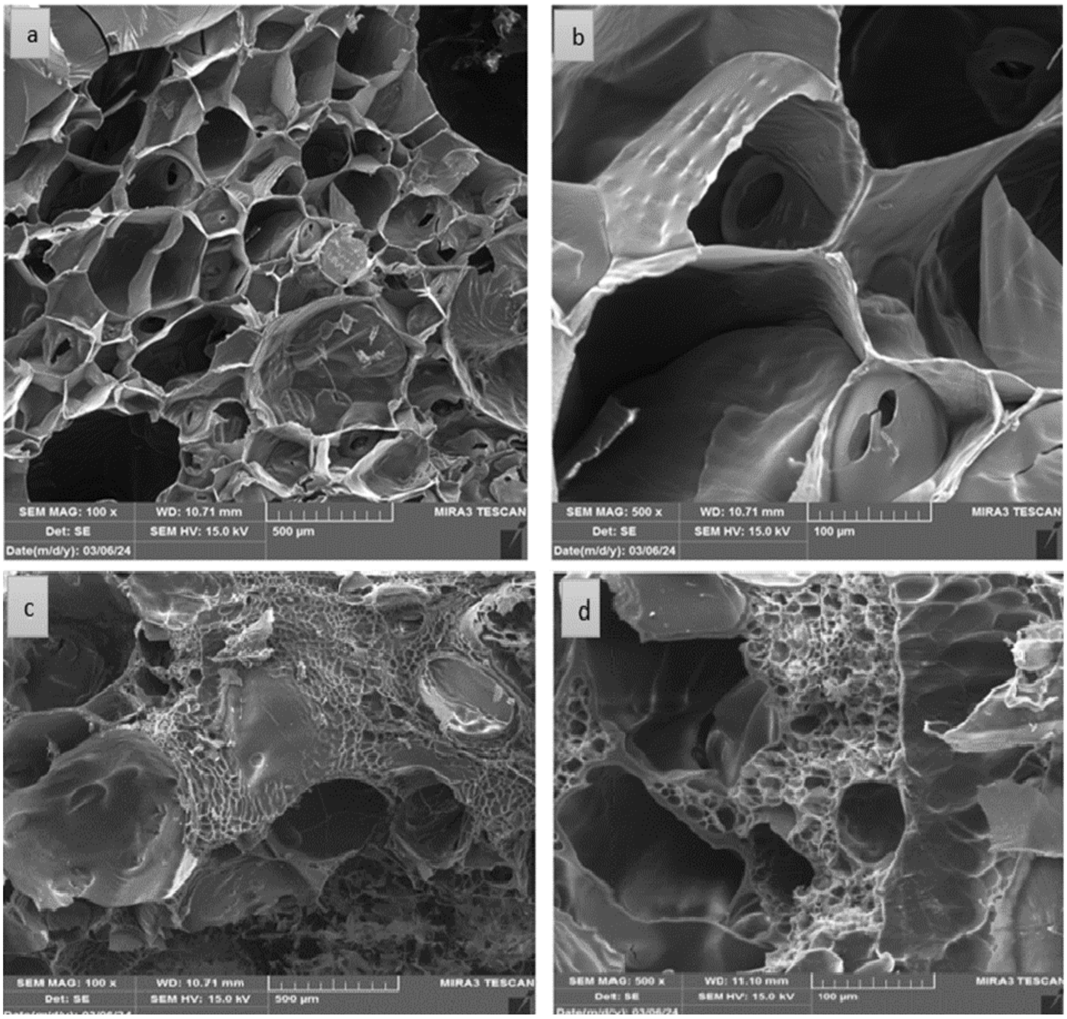


Figure 5: Scanning electron microscopy (SEM) for the selected formula (C) at different magnification powers: a)100kx, b)500kx, c) 100kx, and d) 500kx

3.4 **Swelling Property of superabsorbent semi-IPN hydrogel**

The polyacrylamide contains hydrophilic amide groups, while the APAM polyelectrolyte contains hydrophilic -N+$(CH_3)_3$ and -COOH groups. This study investigated the swelling properties of the hydrogel [35], and Figure 5 demonstrates the impact of AAm on the swelling properties of the semi-IPN hydrogel. (a) Throughout the swelling test, the behavior of the substance in deionized water at ambient temperature is observed as a function of time. Figure 6 clearly shows the exponential growth in the expansion rate of each semi-IPN hydrogel during a period of 0 to 360 minutes. The hydrogel sample containing 2 g of AAm exhibits the highest swelling rate, while sample A exhibits the slowest. The swelling rate of the hydrogel escalated with the elevation of AAm content, which can be attributed to the augmented abundance of hydrophilic groups.

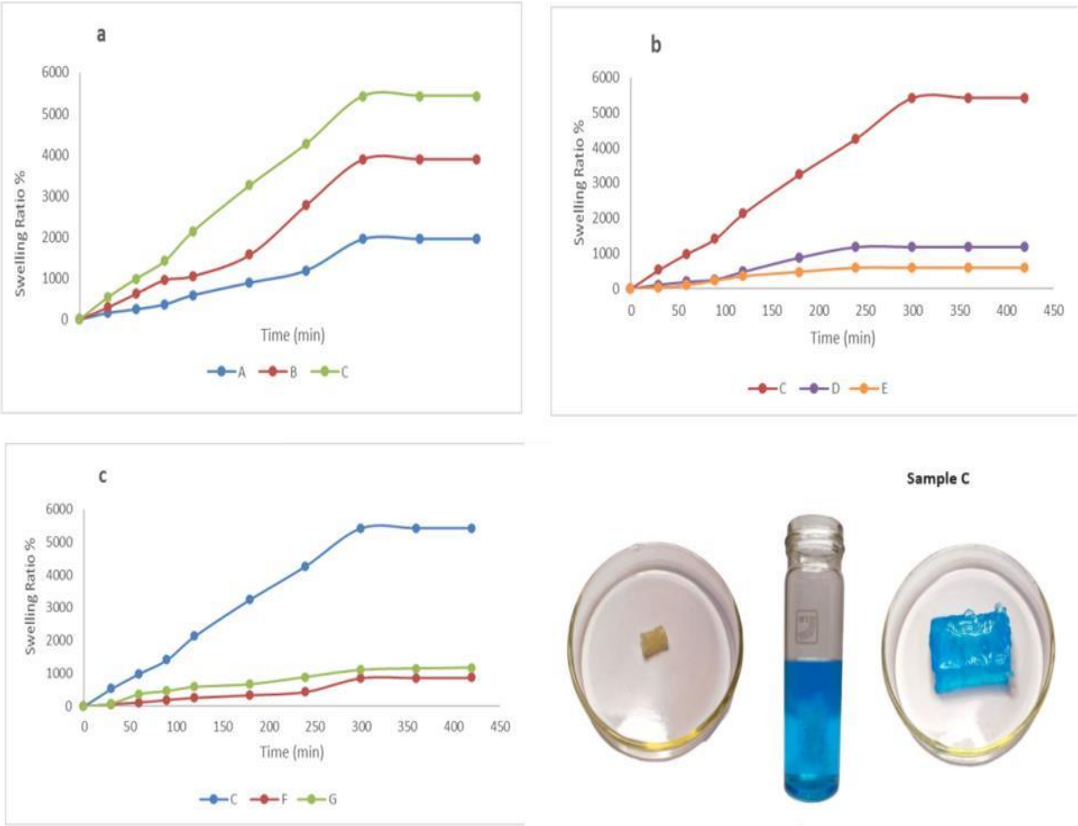


Figure 6: Swelling ratio concerning time: a) effect AAm weights b) effect with different MBA weights c) effect with different initiator weights. d) Images of hydrogels (sample C) swelling in water.

3.5 **Effect of Cross-Linker and Initiator weights on Swelling** (b).

To investigate the effect of MBA weights as a cross-linker on the properties of the semi-IPN hydrogel formulas C, D, and E, these formulations were prepared and subsequently evaluated. The results are shown in Figure 6 (b). As illustrated in Figure 5, an increase in the weights of the cross-linker led to a rise in the density of the IPN hydrogel. This increase occurred because the network space was reduced and porosity decreased due to the formation of stronger bonds between polymer chains and the cross-linker, which reduced the size of the interconnecting pores (Sample E) and reduced the swelling ratio of the polymer [36]. To examine the effect of different weights of initiators on the swelling ratio of the hydrogel, the weights of SPS were altered within the range of 0.03-0.1 g (Figure 6), (c) The maximum swelling ratio (5432)% was attained at an initiator weight of 0.07 g. When the weight is less than 0.07 g, the number of active free radicals in the polymer backbone decreases, resulting in a decrease in the degree of polymerization and, thus, a reduction in the final water absorbency. The fall in the swelling value of PAAm hydrogel can be attributed to self-cross-linking, which increases the termination step reaction by bimolecular collision and reduces the molecular weight of the hydrogel. The negative correlation between initiator weight and the molecular weight explains the reason behind the latter.

3.6 **Effect of different pH on swelling ratio**

In Figure 7 (A), The APAM polyelectrolyte demonstrates a substantial increase in size when immersed in both pure water and basal medium. The presence of carboxylic group inions (-COO^-) along polymer chains results in greater electrical repulsion between the negatively charged atoms. This, in turn, drives a rapid swelling process in the cross-linked polyacrylamide network [37-40]. Furthermore, the presence of hydrophilic groups (such as -

$CONH_2$ and -N+R4) in the semi-interpenetrating networks (IPNs) of hydrogels has a substantial effect on swelling behavior. It accelerates the rate of swelling by facilitating enhanced hydrogen bonding between the polymer networks and water. In contrast, the semi-IPNs showed a reduced swelling ratio when exposed to an acidic environment due to the protonation of the carboxylate groups. The protonation caused the APAM chains to wrap around each other, and the lack of expansion in the polyacrylamide network structure helped this process [36]. The presence of amide and ammonium functional groups is responsible for the resulting oedema. Improvement and ammonium groups are commonly found. In general, the acrylamide network consists of many interactions, such as hydrogen bonding interference and ionic interference caused by the polyelectrolyte. Internal hydrogen bonds are created through the collaboration of amide groups inside the polyacrylamide network. The ionic groups on both polyelectrolytes are surrounded by a stable carapace created by these linkages. The acrylamide network is responsible for absorbing water (water uptake WU) from the surroundings [40,41]. Observations indicated variations in swelling ratios among polyelectrolytes due to the presence of quaternary ammonium and carboxyl groups at pH levels of 1.4, 7.2, and 12.

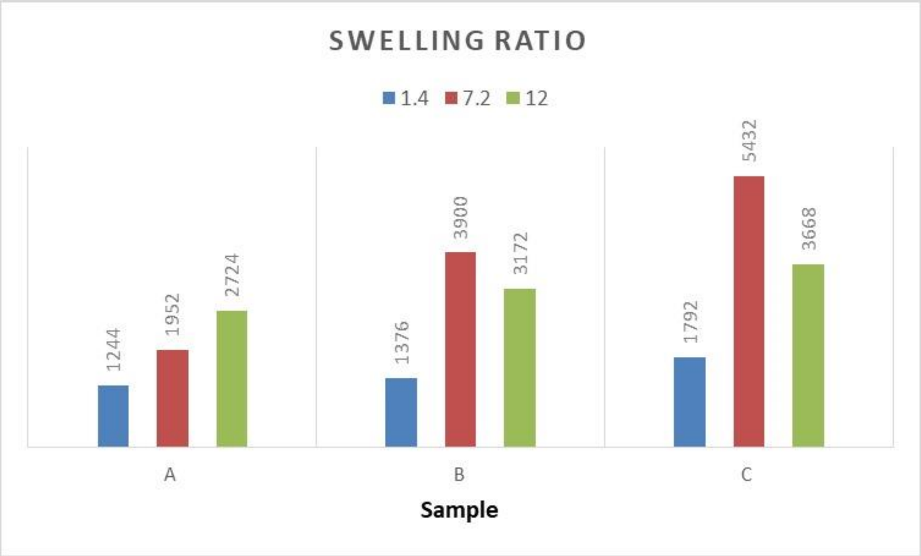


Figure 7: (A) : Effect of different pH 1.4, 7.2, and 12 on swelling ratio

3.7 **Study the effect of salt solutions on absorption ratio**

This test is valuable for examining the impact of salt concentration on the absorption ratio of the semi-IPNs of hydrogels. Four concentrations of NaCl salt (1%, 5%, 10%, and 15%) were prepared, and 0.25 g of the semi-IPNs of hydrogels was immersed for 4 hours. The absorption ratio was measured, and these were the results: Table 2 and Figure 7 (B) show

Table 3. The effect of salt solutions on absorption ratio

C	B	A	NaCl concentration %
2404	**1948**	**1724**	**1**
2108	**1884**	**1600**	**5**
1852	**1580**	**1220**	**10**
1764	**1444**	**1120**	**15**

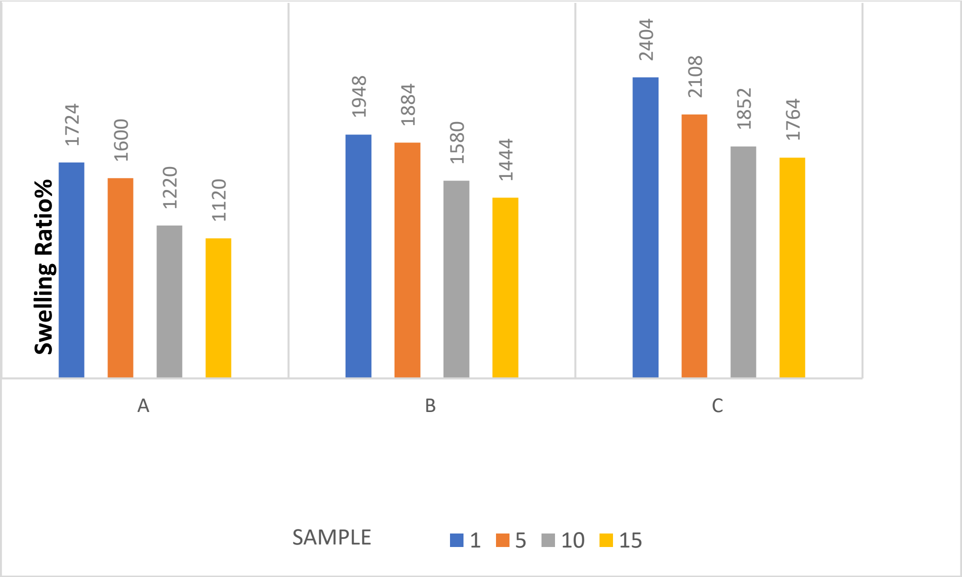


Figure 7(B): Effect of different salt concentrations (1%,5%,10%,15%) on swelling ratio%.

Figure 7(B) illustrates that the absorption ratio decreases as the concentration of salt increases. This phenomenon can be attributed to the fact that increasing the concentration of the salt solution reduces the solution pressure difference between the solution immersed outside and inside the hydrogel network. Consequently, the primary driving force decreases because water enters the hydrogel network, so absorption decreases as well due to the deposition of salt inside the pores of the hydrogel, as shown in Figure 8 [42].

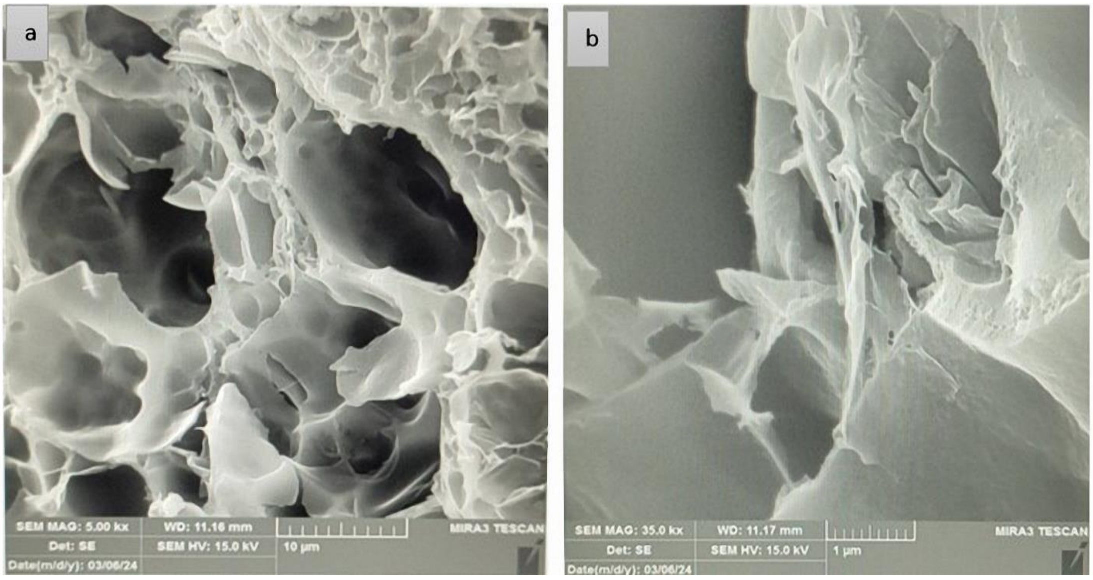


Figure 8: Scanning electron microscopy for the selected formula (C) after being immersed in salt solution at different magnification powers: a) 5.00kx, b) 35.0kx.

3.8 Application and recovery of dry semi-IPN hydrogels to remove the free and soluble water contents

The saturated petrol samples were combined with dry APAM IPN hydrogels to ensure adequate interaction between the hydrogel particles and the liquid phase. The hydrogels were then allowed to dissolve in the saturated solution at room temperature. As shown in Figure 9, the volume of the water began to decrease, while the volume of the fuel samples remained unchanged. The hydrogel pieces turned blue due to it absorbing the colored water. After 3 hours, the hydrogel pieces were removed from the water, blotted with filter paper to remove surface water, and weighted, and the swelling ratio was calculated. Following the removal of water content, the hydrogel pieces were recovered, washed with water and dehydrated using ethanol, then dried in an oven at 70 ºC and retested in another mixture of fuel and water.

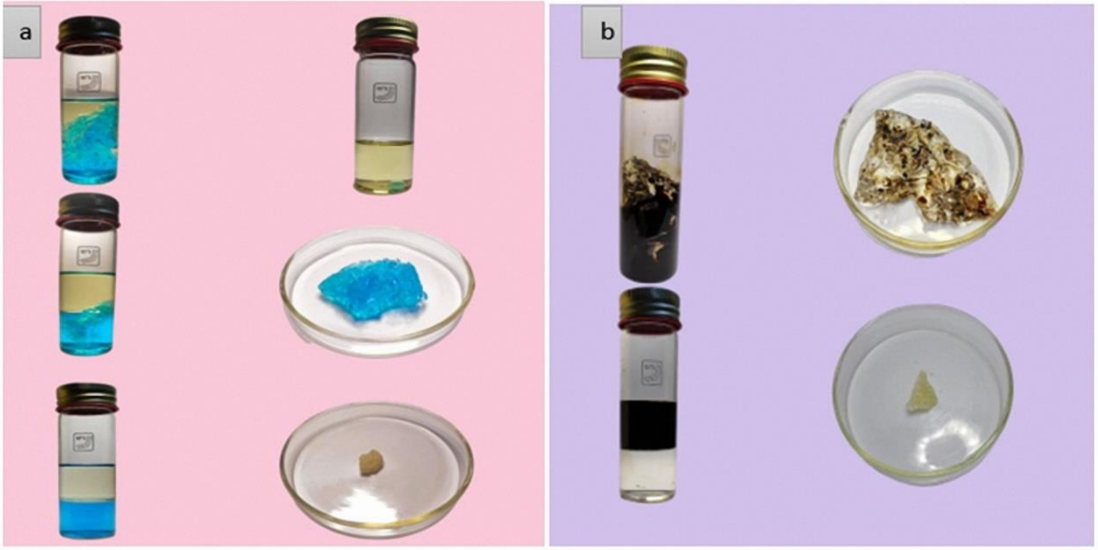


Figure 9: Images of hydrogels remove water saturated with a) Gasoline, b) Crude Fuel

Water retention persists after repeated cycles of fuel water removal, dehydration, and purification. The swelling ratio of hydrogels containing APAM IPNs remained unchanged. As the study progressed, it improved and showed a higher swelling rate than the first swelling after preparation. As can be seen in Figure 10, this could be due to the expansion of the polymer chains in the APAM IPN hydrogel network, which leads to a higher degree of swelling after each cycle. The best formulation is sample C, where the swelling ratio in the gasoline-water mixture is 5644%, while the swelling ratio in the crude oil-water mixture is 4524% after four cycles. The hydrogels with the same properties were reused and the extent of swelling was checked regularly. Comparable results were found when the severity of edema was analyzed eight months after the hydrogel was prepared. During synthesis, the petroleum fuel exhibited a swelling ratio between 1588 and 2700 percent. This swelling ratio was then maintained in a range of 1365 to 3364 percent over eight months. The swelling ratio of the fuel was initially between 1424 and 2700 percent, and after eight months it remained between 1298 and 3144 percent.

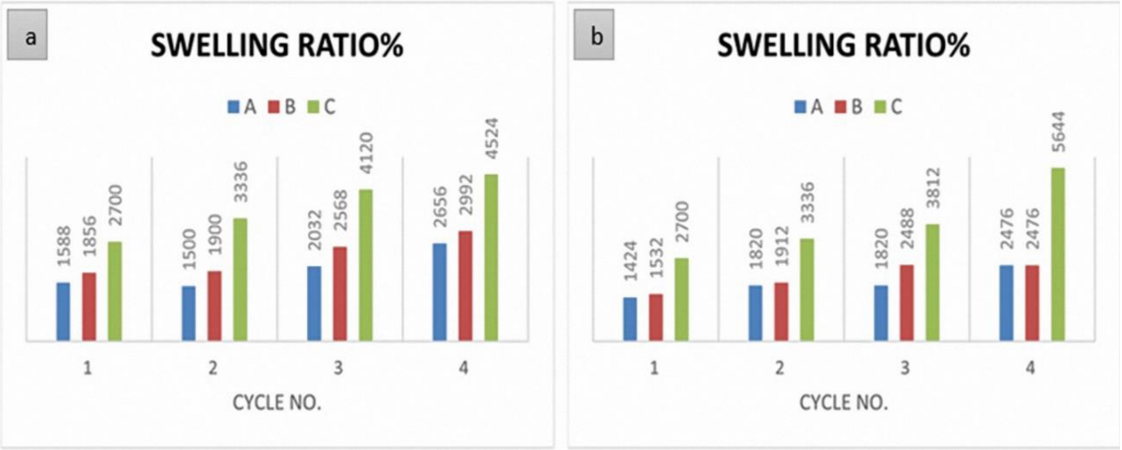


Figure 10: The amount of swelling ratio of the APAM IPNs hydrogels (A, B, C) in several cycles: a) in Crude fuel. b) in Gasoline

Conclusion

In the present study, the APAM IPN hydrogels with superporous structures were synthesized through free radical polymerization. These hydrogels exhibited a rapid swelling rate and a high swelling ratio due to the high porosity and cross-linking of the pores within the polymer chain, which was confirmed by the SEM images. The concentrations of the monomer, crosslinking agent, and initiator affect the swelling and viscoelastic properties of the hydrogel as well as the volume of the solvent. Additionally, the swelling of the hydrogel was also significantly influenced by the external stimuli pH and ionic strength. The equilibrium swelling ratios showed high values at pH 12 and 7.4, which can be attributed to the ionization of the -NH2 and -COOH groups in the hydrogels and the superporous structure. The polymers were also able to rapidly achieve equilibrium between swelling and deswelling when the pH changed between acidic and neutral conditions. Sample C showed a higher swelling ratio than sample A at pH 7.4. Increasing the ionic strength of the swelling medium from 1 % to 15 % NaCl led to a decrease in the degree of swelling. The APAM IPN hydrogels proved to be an effective alternative method for removing the water content of crude oil and gasoline. The experiments showed that the hydrogels can be used to treat saturated crude oil and gasoline for many cycles. Based on the results obtained in this work, new hydrogels will be synthesized from different hydrophilic monomers and tested for their ability to remove water from fuel. The tests can also be extended to other liquid fuels such as jet fuel and other liquid fuels.

Declarations

- Ethical approval Not applicable

-Informed consent Not applicable

-Availability of data and material. The datasets used and analyzed during the current study are available from the corresponding author on reasonable request.

-Competing interests The authors declare that they have no competing interests

-Funding No funds were received to fulfil this work.

-Acknowledgements We'd like to express our gratitude to the Chemistry Department, College of Education for Pure Sciences, University of Basrah, Basrah, Iraq, for supporting us and use their facilities to conduct our research analyses. This research is a part of the Master degree graduate research project.

References

[1] W. Jiang, L. Lin, X. Xu, H. Wang, and P. Xu, “Analysis of Regulatory Framework for Produced Water Management and Reuse in Major Oil- and Gas-Producing Regions in the United States,” *Water*, vol. 14, no. 14, p. 2162, 2022.

[2] L. Henrique, B.Patricia, M. Gabriela , M. R. W. Maciel and L. V. Fregolente.” Development of a Hydrogel Column for Water Removal from Fuels*”, Chemical Engineering Transactions*, vol. 86, pp. 1117-1122, 2021.

[3] J. Pichtel, “Oil and Gas Production Wastewater: Soil Contamination and Pollution Prevention,” *Applied and Environmental Soil Science*, vol. 2016, pp. 1–24, 2016.

[4] N. Hazrati, A. A. Miran Beigi, and M. Abdouss, “Demulsification of water in crude oil emulsion using long chain imidazolium ionic liquids and optimization of parameters,” *The Science and Technology of Fuel and Energy*, vol. 229, pp. 126–134, 2018.

[5] A. Telin L. Lenchenkova, R. Yakubov, K. Poteshkina, P. Krisanova, A. Filatov, and A. Stefantsev, “Application of Hydrogels and Hydrocarbon-Based Gels in Oil Production Processes and Well Drilling,” *Gels*, vol. 9, no. 8, p. 609, 2023.

[6] J. Chen, J. Wu, P. Raffa, F. Picchioni, and C. E. Koning, “Superabsorbent Polymers: From long-established, microplastics generating systems, to sustainable, biodegradable and future proof alternatives,” *Progress in Polymer Science*, vol. 125, p. 101475, 2022.

[7] L. Miao, J. Hu, M. Lu, Y. Tu, X. Chen, Y. Li, and S. Hu "Alkynyl-functionalization of hydroxypropyl cellulose and thermoresponsive hydrogel thereof prepared with P(NIPAAm- co -HEMAPCL)," *Carbohydrate Polymers*, vol. 137, pp. 433–440, 2016.

[8] C. Tuncer and M. Işık, "Boron adsorption studies of poly(styrene-glycidyl methacrylate) latex: effect of modification agents," *Iranian Polymer Journal*, vol. 5, pp. 1-12, 2024.

[9] N. A. Fadil, S. Irawan, N. A. M. Isa, and S. R. Shafian, "Gelation Behavior of Polyacrylamide Reinforced with Nano-Silica for Water Shutoff Treatment in Oil Field," *Solid State Phenomena*, vol. 307, pp. 252–257, 2020.

[10] N. Y. Steinman, N. Y. Bentolila, and A. J. Domb, "Effect of Molecular Weight on Gelling and Viscoelastic Properties of Poly(caprolactone)–b-Poly(ethylene glycol)–b-Poly(caprolactone) (PCL–PEG–PCL) Hydrogels," *Polymers*, vol. 12, no. 10, p. 23722020.

[11] J. M. Rosiak, P. Ulański, and A. Rzeźnicki, "Hydrogels for biomedical purposes," *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, vol. 105, no. 1, pp. 335–339, 1995.

[12] L. A. Shah, M. Khan, R. Javed, M. Sayed, M. S. Khan, A. Khan, and M. Ullah., "Superabsorbent polymer hydrogels with good thermal and mechanical properties for removal of selected heavy metal ions," *Journal of Cleaner Production*, vol. 201, pp. 78–87, 2018.

[13] H. Shoukat, K. Buksh, S. Noreen, F. Pervaiz, and I. Maqbool, "Hydrogels as Potential Drug-Delivery Systems: Network Design and Applications," *Therapeutic Delivery*, vol. 12, no. 5, pp. 375–396, 2021.

[14] A. Adair, A. Kaesaman, and P. Klinpituksa, "Superabsorbent materials derived from hydroxyethyl cellulose and bentonite: Preparation, characterization and swelling capacities," *Polymer Testing*, vol. 64, pp. 321–329, 2017.

[15] O. P. Gbenebor, S. O. Adeosun, G. I. Lawal, S. Jun, and S. A. Olaleye, "Acetylation, crystalline and morphological properties of structural polysaccharide from shrimp exoskeleton," *Engineering Science and Technology, an International Journal*, vol. 20, no. 3, pp. 1155–1165, 2017.

[16] G. Zain, A. A. Nada, M. A. El-Sheikh, F. A. Attaby, and A. I. Waly, "Superabsorbent hydrogel based on sulfonated-starch for improving water and saline absorbency," *International Journal of Biological Macromolecules*, vol. 115, pp. 61–68, 2018.

[17] K. Rop, D. Mbui, N. Njomo, G. N. Karuku, I. Michira, and R. F. Ajayi, "Biodegradable water hyacinth cellulose-graft-poly(ammonium acrylate-co-acrylic acid) polymer hydrogel for potential agricultural application," *Heliyon*, vol. 5, no. 3, p. e01416, 2019.

[18] W. A. Laftah, S. Hashim, and A. N. Ibrahim, "Polymer Hydrogels: A Review," *Polymer-Plastics Technology and Engineering*, vol. 50, no. 14, pp. 1475–1486, 2011.

[19] H. Huang, Z. Dong, X. Ren, B. Jia, G. Li, S. Zhou, and W. Wang, "High-strength hydrogels: Fabrication, reinforcement mechanisms, and applications," *Nano Research*, vol. 16, no. 2, pp. 3475–3515, 2023.

[20] A. Vashist, A. Kaushik, A. Vashist, V. Sagar, A. Ghosal, Y. K. Gupta, and M. Nair, "Advances in Carbon Nanotubes–Hydrogel Hybrids in Nanomedicine for Therapeutics," *Advanced healthcare materials*, vol. 7, no. 9, p. 1701213, 2018.

[21] S. Kim and H. Chung, "Convenient Cross-Linking Control of Lignin-Based Polymers Influencing Structure–Property Relationships," *ACS Sustainable Chemistry & Engineering*, vol. 11, no. 5, pp. 1709–1719, 2023.

[22] W. Hu, Z. Wang, Y. Xiao, S. Zhang, and J. Wang, "Advances in crosslinking strategies of biomedical hydrogels," *Biomaterials Science*, vol. 7, no. 3, pp. 843–855, 2019.

[23] A. GhavamiNejad, N. Ashammakhi, X. Y. Wu, and A. Khademhosseini, "Crosslinking Strategies for 3D Bioprinting of Polymeric Hydrogels," *Small* ,vol. 16, no. 35, p. 2002931, 2020.

[24] Q. Wang, Y. Zhang, Y. Ma, M. Wang, and G. Pan, "Nano-crosslinked dynamic hydrogels for biomedical applications," *Materials Today Bio*, vol. 20, p. 100640, 2023.

[25] J. Long, A. V. Nand, C. Bunt, and A. Seyfoddin, "Controlled release of dexamethasone from poly(vinyl alcohol) hydrogel," *Pharmaceutical Development and Technology*, vol. 24, no. 7, pp. 839–848, 2019.

[26] J. Li, Q. Shi, X. Wu, C. Li, and X. Chen, "In vitro and in vivo evaluation of 3D biodegradable thermo/pH sensitive sol-gel reversible hydroxybutyl chitosan hydrogel," *Materials Science and Engineering*, vol. 108, p. 110419, 2020.

[27] J. M. Li, C. S. Hu, J. M. Shao, H. J. Li, P. Y. Li, X. C. Li, and W. D. He, "Fabricating ternary hydrogels of P(AM-co-DMAEMA)/PVA/β-CD based on multiple physical crosslinkage," *The International Journal for the Science and Technology of Polymers*, vol. 119, pp. 152–159, 2017.

[28] F. Ullah, M. B. H. Othman, F. Javed, Z. Ahmad, and H. Md. Akil, "Classification, processing and application of hydrogels: A review," *Materials Science and Engineering*: C, vol. 57, pp. 414–433, 2015.

[29] A. M. Hillery, A. W. Lloyd, and J. Swarbrick, *Drug Delivery and Targeting*: For Pharmacists and Pharmaceutical Scientists. CRC Press, 2003.

[30] M. C. Hacker and H. A. Nawaz, "Multi-Functional Macromers for Hydrogel Design in Biomedical Engineering and Regenerative Medicine," *International Journal of Molecular Sciences*, vol. 16, no. 11, pp. 27677–27706, 2015.

[31] N. Zoratto and P. Matricardi, "4 - Semi-IPNs and IPN-based hydrogels in *Polymeric Gels*", *K. Pal and I. Banerjee, Eds., in Woodhead Publishing Series in Biomaterials*, pp. 91–124, 2018.

[32] A. Zaharia, A. L. Radu, S. Iancu, A. M. Florea, T. Sandu, I. Minca, and T. V. Iordache," Bacterial cellulose-poly(acrylic acid-co-N, N - methylene-bis-acrylamide) interpenetrated networks for the controlled release of fertilizers", *The Royal Society of Chemistry*, 8, 17635–17644 .2018.

[33] A. M. Ghaffar, M. B. El-Arnaouty, A. A. Abdel Baky, and S. A. Shama, "Radiation-induced grafting of acrylamide and methacrylic acid individually onto carboxymethyl cellulose for removal of hazardous water pollutants' *Designed Monomers and Polymers*, vol. 19, no. 8, 706–718. 2016.

[34] Y. Chujo, K. Sada, and T. Saegusa, "Iron(II) bipyridyl-branched polyoxazoline complex as a thermally reversible hydrogel," *Macromolecules*, vol. 26, no. 24, pp. 6315–6319, 1993.

[35] N. A. H. Al-Asady, D. A. Hassan, and D. T. Abdulqader, "In vitro tooth whitening efficacy of carbamide peroxide polyelectrolyte gel and colorimetric evaluation, stability, and hydrogen peroxide release," *Chemical Papers*, vol. 76, no. 6, pp. 3841–38512022.

[36] A. V. Dobrynin and M. Rubinstein, "Theory of polyelectrolytes in solutions and at surfaces," *Progress in Polymer Science*, vol. 30, no. 11, pp. 1049–1118, 2005.

[37] B. Zhu, D. Ma, J. Wang, and S. Zhang, "Structure and properties of semi-interpenetrating network hydrogel based on starch," *Carbohydrate Polymers*, vol. 133, pp. 448–455, 2015.

[38] R. Feksa, E. A. Troian, C. D. Muller, F. Viegas, A. B. Machado, and V. C. Rech, "Chapter 11 - Hydrogels for biomedical applications," in *Nanostructures for the Engineering of Cells, Tissues and Organs, A. M. Grumezescu, Ed., William Andrew Publishing*, 2018, pp. 403–438.

[39] S. Xu, H. Li, H. Ding, Z. Fan, P. Pi, J. Cheng, and X. Wen, "Allylated chitosan-poly(N-isopropylacrylamide) hydrogel based on a functionalized double network for controlled drug release," *Carbohydrate Polymers*, vol. 214, pp. 8–14, 2019.

[40] R. V. Kulkarni, R. Boppana, G. Krishna Mohan, S. Mutalik, and N. V. Kalyane, "pH-responsive interpenetrating network hydrogel beads of poly(acrylamide)-g-carrageenan and sodium alginate for intestinal targeted drug delivery: Synthesis, in vitro and in vivo evaluation," *Journal of Colloid and Interface Science*, vol. 367, no. 1, pp. 509–517, 2012.

[41] S. Lankalapalli and V. R. M. Kolapalli, "Polyelectrolyte Complexes: A Review of their Applicability in Drug Delivery Technology," *Indian journal of pharmaceutical sciences*, vol. 71, no. 5, pp. 481–487, 2009.

[42] H. Yang, J. Liu, X. Jia, Y. Zhou, and H. Ji. "Influence of NaCl concentrations on the crack-sealing behavior of superabsorbent polymers in cementitious materials." *Construction and Building Materials*, vol. 243, pp. 118228, 2020.

Graphical Abstract