

A Review of Hydrogel Application in Wastewater Purification

Lilyan Alsaka ¹ , Lina Alsaka ¹, Ali Altaee ^{1,*} , Syed Javaid Zaidi ², John Zhou ¹  and Tayma Kazwini ¹

¹ Centre for Green Technology, School of Civil and Environmental Engineering, University of Technology Sydney, 15 Broadway, Ultimo, NSW 2007, Australia; lilyan.y.alsaka@student.uts.edu.au (L.A.)

² UNESCO Chair for Desalination and Water Treatment, Center for Advanced Materials (CAM), Qatar University, Doha P.O. Box 2713, Qatar

* Correspondence: ali.altaee@uts.edu.au

Abstract: Hydrogels have garnered considerable interest in water purification owing to their distinctive physicochemical characteristics, including high porosity, modifiable surface chemistry, and superior water retention capacity. This paper provides a thorough examination of the use of hydrogels in wastewater treatment. It encompasses their categorization and separation procedures, including size exclusion, adsorption, electrostatic interactions, and non-sieving processes. Furthermore, it examines how functional groups improve the efficiency of pollutant removal. The review examines hydrogel composites and their filtering processes, measuring their efficacy in adsorption and evaluating the benefits and limits of hydrogels, especially regarding regeneration capacities. It explores hydration processes in hydrogels, emphasizing the fundamental mechanisms and measuring methods involved. Future research must prioritize optimizing hydrogel design to enhance mechanical strength and reusability, investigate innovative functionalization techniques to address emergent contaminants, and establish scalable manufacturing methods for extensive industrial use.

Keywords: hydrogels; wastewater; filtration; adsorption; swelling; hydration



Academic Editor: Hao Wu

Received: 20 January 2025

Revised: 5 February 2025

Accepted: 8 February 2025

Published: 13 February 2025

Citation: Alsaka, L.; Alsaka, L.; Altaee, A.; Zaidi, S.J.; Zhou, J.; Kazwini, T. A Review of Hydrogel Application in Wastewater Purification. *Separations* **2025**, *12*, 51. <https://doi.org/10.3390/separations12020051>

Copyright: © 2025 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

Two million metric tons of pollutants from sewage, industry, and agriculture are released into the water, threatening public health and resulting in 1.4 million fatalities [1]. Efficient methodologies are necessary for converting seawater and wastewater into potable water via water purification. The specific procedures used depend on the source of water and its intended application [2]. Many technologies have been used to desalinate seawater and wastewater to provide fresh water. Conventional biological processes are an example of wastewater treatment and reuse in farming activities instead of rivers and groundwaters. Several countries use tertiary treated wastewater for irrigation, compensating for the shortage of freshwater resources. Biological and membrane bioreactors (MBR) technologies for wastewater treatment are widely used for domestic wastewater treatment to supply a source of clean water for irrigation [3,4]. Hyperfiltration technology desalinates seawater for various purposes, such as irrigation, industrial use, and drinking water. Membranes with different molecular weight cut-offs are developed for various applications. Nanofiltration (NF) membranes, derived from ultrafiltration (UF), are designed to separate divalent ions and partially remove monovalent ions, showing less than 20% rejection rate for the latter [5,6]. NF membranes are used for water softening, industrial wastewater treatment, and desalinating low-salinity water. Reverse osmosis (RO) membranes can selectively eliminate solutes, making them energy-efficient and adaptable [7]. Developed in the 1970s, RO membranes for seawater desalination achieved a 99% rejection rate for monovalent

ions like NaCl, replacing thermal processes as the dominant technology [8]. Despite their advantages, NF and RO membranes face challenges such as intensive pretreatment, fouling, and high energy and cost requirements.

Seawater desalination often needs over 60 bars for a TDS above 40 g/L [9,10]. Chemical purification techniques are slow and generate large quantities of wastewater, while adsorption methods face challenges in finding ideal adsorbents and high regeneration costs. New, cost-effective water purification methods are needed, especially in emergencies [11,12]. In recent years, the research focus has shifted to designing and developing new polymeric adsorption matrixes that provide economical and efficient treatment technology. In this view, hydrogel materials provide a novel technology that enables cost-effective, reusable, and highly effective methods for purifying and decontaminating water. Hydrogel materials consist of an interconnected three-dimensional polymer network. These materials possess a high surface area, which enables them to adsorb a wide range of pollutants from water, such as heavy metals, organic contaminants, and colors, with significant selectivity. In addition, hydrogel materials have a rapid regenerative capacity, which reduces the amount of waste produced during the purifying process, making them a possible substitute for conventional purification methods and significantly reducing waste production [13]. Hydrogels have received considerable interest because they can be used in many different sectors, such as water treatment, agriculture, and biomedicine. Chitosan-based hydrogels successfully remove pollutants like heavy metals and organic contaminants from water because they have a high capacity for adsorption [14]. In agriculture, hydrogels serve as soil conditioners, augmenting water retention and nutrient accessibility and promoting crop productivity and resilience in dry areas [15]. In the biomedical domain, hydrogels are used in drug delivery systems, wound dressings, and tissue engineering owing to their biocompatibility and capacity to replicate natural tissue characteristics [16]. Hydrogel technology remains challenging to use industrially. Scalability, cost-effectiveness, and environmental impact must be considered. Some hydrogels need sophisticated, non-scalable production techniques [17]. Furthermore, a comprehensive assessment of the environmental consequences of hydrogel degrading products is necessary to guarantee sustainability. The limited mechanical strength and possible toxicity of some hydrogel components may also prevent them from being widely used in industry. Despite the improvements in this field, several gaps still exist; firstly, understanding how hydrogel matrices interact with various contaminants is essential for optimizing their design and functionality. Second, although hydrogels have demonstrated potential in laboratory settings, their production and application for large-scale water treatment remain challenging. Key issues that need to be addressed include mechanical stability, regeneration efficiency, and long-term performance for effective practical implementation. Third, it is crucial to thoroughly evaluate the environmental impact of hydrogel synthesis, usage, and disposal to ensure sustainability. Filling these research gaps is vital for advancing hydrogel-based technologies in water purification. This review provides insight into the hydrogel materials used in wastewater treatment, emphasizing the mechanisms involved in removing organic and inorganic pollutants from contaminated solutions and the advantages of hydrogels as an alternative technology for wastewater treatment.

2. Hydrogel Classification

Hydrogels may be categorized in several different ways depending on the processes used in their production. Homopolymeric hydrogels are characterized by a crosslinked skeletal structure produced from a specific monomer or an essential structural component. Table 1 categorizes hydrogels based on various criteria, including polymer composition, charge, crosslinking method, crystallinity, source, and application, highlighting their di-

verse structures and functionalities. The polymer and the manner of crosslinking impact this structure. Copolymeric hydrogels are made up of two or more monomer species, with at least one of them being hydrophilic, that are organized in a variety of configurations inside the framework of the hydrogel [18]. Networks that are formed of two distinct natural or synthetic polymers that have been crosslinked together are known as multipolymeric hydrogels. In addition, hydrogels may be categorized according to their charges (non-ionic, ionic, amphoteric electrolyte, and zwitterionic), the procedures used to crosslink them (chemical or physical), and their crystalline state (amorphous, semicrystalline, or crystalline) [19]. The polymer scaffold may be made from natural, synthetic, or hybrid hydrogels. Hydrogels are formed using natural biopolymers such as gelatine and collagen and polysaccharides like starch, cellulose, chitosan, etc. The use of pHEMA, polyvinyl alcohol, poly(N-isopropyl acrylamide), polyethylene glycol, and polyacrylamide in the production of synthetic hydrogels results in products that exhibit exceptional water absorption and retention capabilities, as well as extended shelf life and enhanced gel strength. Hydrogels derived from biological sources are more durable, porous, biocompatible, and biodegradable [20].

Technology advancements in materials science have made it possible to manufacture composite hydrogel materials, creating novel hybrid structures with distinctive physico-mechanical properties. Physical and chemical methods synthesize and modify these systems, including crosslinking, impregnation, grafting, interpenetration, incorporation, mixing, and imprinting. Depending on their stability, water absorption, and strength, bio-based hydrogels and their modified forms are used in environmental, biomedical, and agricultural applications.

The hydrogel's properties are influenced by the kind of polymer used and the presence of hydrophilic groups ($-\text{OH}$, $-\text{COOH}$, $-\text{CONH}_2$, and $-\text{SO}_3\text{H}$) attached to the polymer chains. The presence of hydrophilic moieties, including alcohol, amides, and carboxylic acids, among other types of molecules, enhances the hydrogel's stiffness and capacity to absorb water [21,22]. Furthermore, the presence of functional groups affects the specific crosslinking approach required to create hydrogel from scratch.

Hydrogels, which are elastic and soft, have limited applications due to their mechanical weakness. Numerous functional groups and the ease with which they may be crosslinked contribute to the mechanical solid strength of these materials. Hydrogel water absorption depends on polymer and network density. Most polymers absorb more water than their bulk. Hydrogels are available in various sizes and forms, such as slabs, membranes, nanogels, beads, microgels, cryogels, and aerogels [23].

Three variables are crucial for successfully manufacturing crosslinked hydrogels: (I) The liquid absorption capacity of the hydrogel network relies on the volume at which the polymer swells. (II) The median molecular weight of the crosslinks determines the crosslinking of the hydrogel. (III) The network mesh's size influences the hydrogel's mechanical strength, degradability, and diffusivity.

Table 1. Hydrogel classification.

| Classification Criteria | Types of Hydrogels | Description |
|------------------------------|--------------------------|--|
| Based on polymer composition | Homopolymeric hydrogels | Crosslinked skeletal networks based on a monomer or key structural component. [24,25] |
| | Copolymeric hydrogels | The hydrogel framework consists of many monomer species, at least one of which is hydrophilic and organized in different configurations. |
| | Multipolymeric hydrogels | Crosslinked networks consisting of two distinct natural or synthetic polymers that are interconnected. |

Table 1. *Cont.*

| Classification Criteria | Types of Hydrogels | Description | |
|------------------------------|----------------------------------|---|---------|
| Based on charge | Nonionic | Hydrogels without any ionic charges. | [26–28] |
| | Ionic | Hydrogels with ionic charges. | |
| | Amphoteric electrolyte | Hydrogels containing both positive and negative charges. | |
| | Zwitterionic | Hydrogels with both positive and negative charges but that are neutral overall. | |
| Based on crosslinking method | Chemically crosslinked hydrogels | Hydrogels crosslinked using chemical methods. | [29] |
| | Physically crosslinked hydrogels | Hydrogels crosslinked using physical methods. | |
| Based on crystallinity | Amorphous hydrogels | Hydrogels without a crystalline structure. | [24,30] |
| | Semicrystalline hydrogels | Hydrogels with partial crystalline structure. | |
| | Crystalline hydrogels | Hydrogels with a crystalline structure. | |
| Based on source | Natural hydrogels | Made from natural biopolymers such as proteins (gelatin, collagen) and polysaccharides (starch, cellulose, chitosan). | [24,31] |
| | Synthetic hydrogels | Made from synthetic polymer compounds such as pHEMA, polyvinyl alcohol, poly(N-isopropyl acrylamide), polyethylene glycol, polyacrylamide, and their derivatives. | |
| | Hybrid hydrogels | Made from a combination of natural and synthetic polymers. | |
| Based on application | Environmental hydrogels | Sorbents and flocculants that purify wastewater and sludge. | [32,33] |
| | Biomedical hydrogels | Biomedical applications including medication delivery systems, tissue engineering, and contact lenses. | |
| | Agricultural hydrogels | Used to release water reservoirs and agrochemicals. | |

3. Hydrogel Separation Mechanisms

The main methods of hydrogel separation rely on their distinct three-dimensional, crosslinked structures, which enable selective absorption and filtering. Size exclusion is a fundamental process in which the pore size of a hydrogel controls the molecules that can flow through it, thereby capturing bigger molecules [34]. Charge interactions are also significant since hydrogels that are ionic or zwitterionic may either attract or repel charged particles, hence increasing separation efficiency [35]. Moreover, due to their hydrophilic properties, the water-absorbing capability of hydrogels facilitates the separation of water from impurities. Hydrogels are essential in water purification and medicine administration [36].

3.1. Size Exclusion

Size exclusion is a fundamental separation approach based on the idea that particles cannot flow through holes that are smaller than their diameter (Figure 1a). It is rejected when the particles' hydrated ionic diameters exceed the pore size [37]. Many studies have been performed on the influence of size exclusion on the separation process, particularly emphasizing various molecular features such as size, shape, and molecular weight (MW). Yoon and Lueptow (2005) and Van der Bruggen and Vandecasteele (2002) have confirmed the vital necessity of knowing these molecular features to increase the predictability and effectiveness of size exclusion approaches [38,39]. MW has been used as an essential parameter for assessing rejection, especially for chemicals that are not charged; nonetheless, its capacity to provide comprehensive insights is restricted. In particular, molecular weight does not consider molecule shape, significantly affecting how chemicals interact with membrane pores [40]. Recent research has reached for a more thorough analysis of molecular

size features other than MW to satisfy this problem. For example, Miron et al. emphasized the need to precisely describe the size properties of molecules to have a more accurate knowledge of the separation process [41]. This process enhances the design and use of membrane technologies for targeted solute separation in various industries, from water purification to biochemical processing, and helps explain the dynamics of size exclusion.

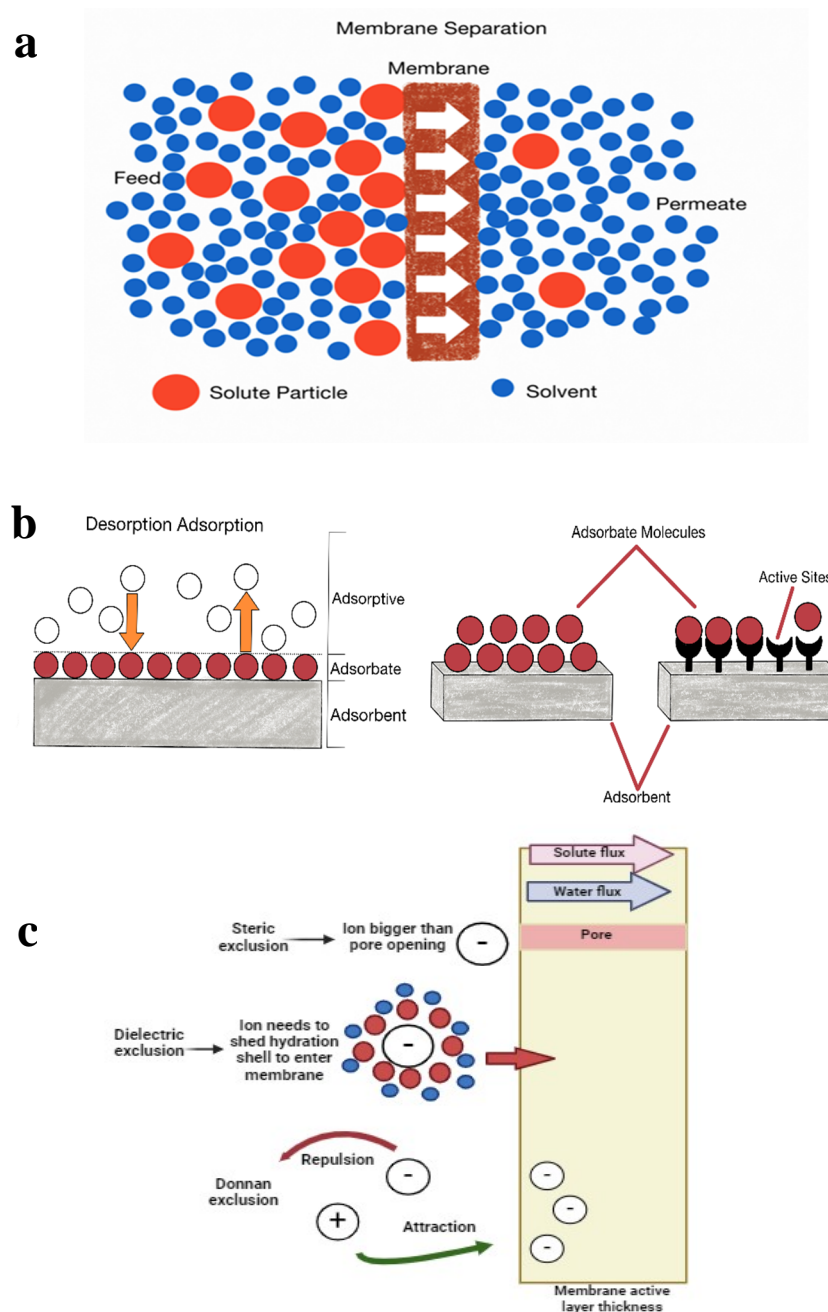


Figure 1. (a) Molecular segregation through a semipermeable membrane, (b) schematic representation of the adsorption process, and (c) schematic representation of solute exclusion mechanisms [42].

3.2. Adsorption

Adsorption is vital in hydrological membrane-based techniques, serving as a critical separation mechanism. Some aspects in the liquid phase, such as organic compounds, heavy metals, or impurities, attach to the membrane's surface through these methods. This method improves filtration and purification efficiency by selectively eliminating undesired substances and enhancing permeate quality (Figure 1b). This process is driven by

attractive forces between the molecules, including chemical affinity, hydrogen bonding, electrostatic interactions, and Van der Waals forces [43]. The effectiveness of adsorption is influenced by the characteristics of the adsorbent, such as hydrophobicity, the presence of functional groups, and density, as well as the properties of the adsorbate, including solubility and molecular size. The competitive dynamics between inorganic and organic substances at active adsorptive sites can enhance or hinder adsorption. Environmental conditions like solution pH, temperature, and other factors critically impact adsorption performance, modifying how substances interact with the membrane [44,45]. The performance of adsorbents is also affected by several operational factors, including the initial ion concentration, temperature, pH, adsorbent dosage, contact time, stirring speed, and ionic strength [46]. Hydrophobic interactions further influence the adsorption of solute molecules onto membranes, playing a pivotal role in the selective rejection of specific micropollutants. Adsorption offers a highly efficient and cost-effective approach to pollutant removal from wastewater. At the same time, membrane filtration, although more costly and requiring a smaller operational area, is particularly effective in removing heavy metal ions [47]. Materials like activated carbon are favored in adsorption processes for their extensive surface area, effectively eliminating micropollutants and adsorbing microbes and organic matter from contaminated waters. Similarly, silica gel, with its vast network of voids and a surface area of about $800 \text{ m}^2/\text{g}$, shows a remarkable capacity for moisture absorption, especially at 40°C , underscoring its utility in various adsorptive applications.

3.3. Electrostatic Interactions

The electrostatic interactions during the use of hydrogel for filtration are crucial and affect the degree to which various materials are separated [48]. A porous hydrogel functions as a physical barrier within a particular filtration system, allowing some molecules or ions to flow through while preventing others from doing so. The charge on the hydrogel's surface and the particles in the feed solution are essential aspects of this process. These charges can either attract or repel particles, substantially impacting the selectivity and efficiency of the separated particles. Electrostatic interactions may be classified as either electrostatic attraction or electrostatic repulsion. When the hydrogel and the particles have the same charge, they repel each other. This repulsion prevents the particles from approaching the hydrogel surface, hindering the particles from getting close to the surface. Alternatively, attraction can facilitate the separation of certain species when the charges are of opposite polarities, promoting adsorption or deposition onto the hydrogel. The efficiency of electrostatic interaction is determined by several factors, including the magnitude and distribution of charges on the hydrogel surface, the charge density of the particles or solutes, the ionic strength of the solution, functional groups in the hydrogel, and pH affecting the hydrogel and contaminants' ionization state [49]. Many theoretical models have been developed to quantify these interactions. Additionally, experimental methods such as zeta potential measurements have been used to obtain information on the surface charge [50,51]

3.4. Non-Sieving Process

The diffusion of ions in two solutions separated by a semipermeable barrier is known as Donnan exclusion. The membrane restricts ionic transport selectively. The electrostatic repulsion between ionic compounds and the fixed charges of the membrane matrix causes Donnan exclusion [52]. Figure 1c shows the Donnan exclusion and dielectric exclusion processes. The membranes reject the negative charges while allowing positive charges to pass through them; thus, some of the dissolved substances are transported, and the ions are trapped in the pores of the membranes. Substances are moved by electromigration, diffusion, and convection depending on the membrane potential gradient [42]. According

to Donnan's exclusion principle, ions with significant charges are rejected more than those with lower charges; hence, it is more effective with divalent ions [53].

Dielectric exclusion occurs because of ion interactions with bound electric charges at interfaces between dielectrically distinct media, such as a membrane structure and a medium. The well-known forces operating on an electric charge near an infinite planar interface are caused by interactions with polarized interfaces. The reaction with a polarized interface is comparable to an imaginary charge on the opposite side of the interface at the same distance from it as the real charge [54–56].

Figure 2 shows techniques for removing pollutants from a water system using hydrogel; these techniques enable separation based on various factors such as the size, chemical composition, hydrophilicity, and charge of the contaminants.

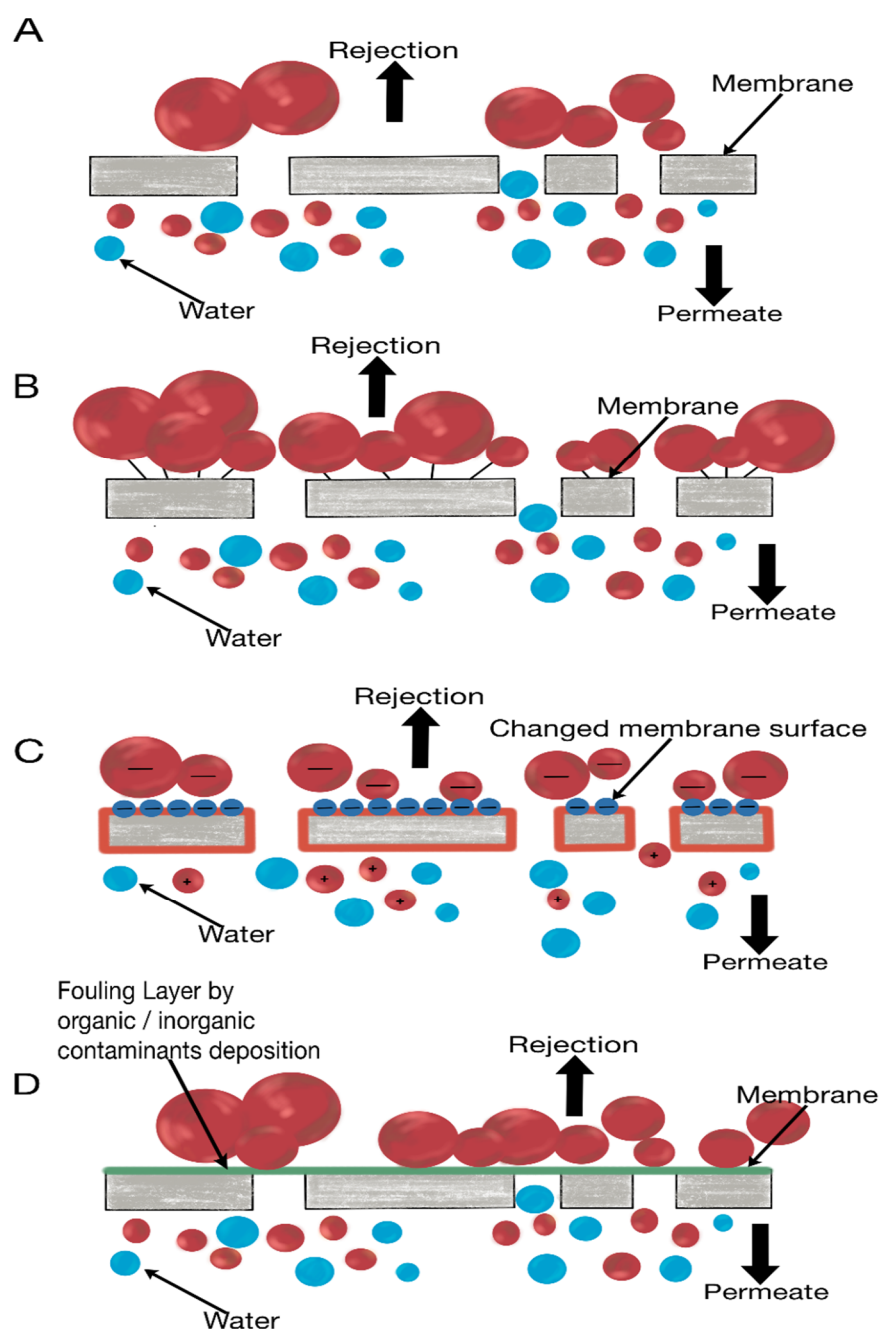


Figure 2. Hydrogel filtration removal mechanisms: (A) size exclusion, (B) hydrophobicity, (C) electrostatic interaction, and (D) adsorption.

4. Functional Groups in Hydrogels

Based on the crosslinking technique, hydrogen is physically entangled with transitory bonds or chemically linked with excellent stability. Functional groups such as hydroxyl, carboxyl, and amine provide essential qualities such as hydrophilicity and reactive potential, allowing their use in different fields, including medication delivery and water purification [36]. According to, hydrogels respond to motivating such as via pH and temperature. Therefore, they can be used to develop intelligent materials for sophisticated applications [57]. Hydrogels are increasingly used because they selectively remove contaminants via functionalized networks. Preparing hydrogels for this purpose often involves integrating specific functional groups that improve their interaction with pollutants. For instance, hydrogels containing carboxyl groups ($-\text{COOH}$) are effective in binding heavy metal ions such as lead and cadmium, leveraging the ionic interactions between the negative charge of the carboxyl and the positive charge of the metal ions [58]. In addition, using amine groups ($-\text{NH}_2$) in hydrogels can effectively remove anionic dyes and other negatively charged contaminants from water through electrostatic attraction [59,60]. It was discovered that composite hydrogels, including natural and synthetic polymers, can improve their structural and functional properties (Table 2). For example, a chitosan–polyacrylamide composite hydrogel has shown remarkable effectiveness in removing dyes and heavy metals from aqueous solutions. This hydrogel combines the biocompatibility and cationic character of chitosan with the strength of polyacrylamide [61].

Table 2. The functional groups for different hydrogels for contaminant removal from water.

| Hydrogel Type | Functional Group(s) | Contaminants Removed | Example of Application | Reference |
|---|--|---|--|-----------|
| Carboxymethyl cellulose hydrogel Polyacrylic acid hydrogel | Carboxyl ($-\text{COOH}$) | Heavy metals (Cd, Pb, Cu, and phosphates) | Removal of heavy metals from wastewater | [58] |
| Amine-functionalized hydrogel | Amine ($-\text{NH}_2$) | Anionic dyes, heavy metals | Electrostatic removal of negatively charged contaminants | [59,60] |
| Chitosan–polyacrylamide hydrogel | Amine ($-\text{NH}_2$), Amide ($-\text{CONH}_2$) | Dyes, heavy metals | Dual functionality for broad-spectrum contaminant removal | [61] |
| Kappa carrageenan hydrogel | Sulfate ($-\text{OSO}_3$) | Heavy metals and organic pollutants | Effective in binding with divalent metal ions and various organics | [62] |
| Poly(N-isopropyl acrylamide) hydrogel | Amide ($-\text{CONH}_2$) | Temperature-sensitive pollutants | Temperature-responsive contaminant capture | [63] |
| Iron oxide nanocomposite hydrogel | Magnetic groups | Arsenic, chromium | Magnetic separation of heavy metals from drinking water | [64] |
| Cyclodextrin hydrogel | Hydroxyl ($-\text{OH}$) | Organic pollutants, pesticides | Encapsulation and removal of organic chemicals | [65] |

5. Hydrogel Composites

Polymerization is a simple procedure involving the utilization of monomers and initiators carried out at high temperatures, and it produces very dense hydrogels. Upon contact with water, these hydrogels become more pliable and are stabilized by surfactants. Ionic or neutral monomers are used in solution polymerization, and UV or redox initiators are used to commence the process. Following the polymerization process, the solvents are extracted by immersing the hydrogel in water, which results in phase separation and purification. Hydrogels are characterized by significant swelling, hydrophilicity, and coordination interactions with heavy metals. FTIR, XPS, SEM, optical microscopy, and compressive strength tests are some methods used for characterization [66]. Hydrogel composites are becoming more critical in environmental applications and water purification. These composites combine hydrogels with biodegradable, inexpensive materials such as starch, cellulose, lignin, and charcoal, successfully addressing the problems associated

with the cost and environmental impact of artificial hydrogels [67,68]. This integration enhances the composite's adsorptive capacity, augments its surface properties, and introduces essential functional groups such as carboxylic, amino, and hydroxyl groups, increasing hydrophilicity and improving pollutant removal efficiency. Integrating biochar with hydrogels, for instance, substantially elevates the composite's swelling capacity and contaminant adsorption effectiveness. Hydrophilic functional groups in the hydrogel–biochar composite allow for more effective capture and removal of waterborne pollutants than either material alone [67,69]. Research suggests that these composites may significantly enhance the rate at which pollutants like arsenic are absorbed, showing a higher sorption capacity than more straightforward biochar settings. Moreover, these composites can be tailored according to individual water treatment needs by including different essential ingredients and functional groups. For instance, adding biochar high in silica improves the strength of hydrogel composites and optimizes their ability to adsorb substances due to silica's involvement in the composite synthesis [67]. Researchers have developed a dual-network hydrogel composed of PEDOT:PSS and PAAm, which effectively produces solar steam for desalination and sewage purification. This hydrogel demonstrates a high solar absorption rate of approximately 99.8% and a steam production rate of $2.15 \text{ kg m}^{-2} \cdot \text{h}^{-1}$, providing a sustainable solution for water purification [70].

Additionally, a Ti_4O_7 membrane, modified with a hydrophobic surface and a thermal insulation layer, attained a high evaporation rate of $7.51 \text{ kg} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$, producing water that met WHO drinkable water requirements [71]. A Ti_4O_7 -PVA nanocomposite hydrogel has also been designed to generate solar steam efficiently, achieving an evaporation rate of $4.45 \text{ kg} \cdot \text{m}^{-2} \cdot \text{h}^{-1}$ under one-sun irradiation and demonstrating an energy efficiency of 90.69%. This hydrogel effectively desalinated and purified water samples containing high salinity, strong acids or bases, heavy metals, and organic colors [72]. Moreover, researchers blended and crosslinked PEDOT:PSS nanofibrils with poly(vinyl alcohol), resulting in a mechanically resilient nanocomposite hydrogel. In a 60-day seawater evaporation test, this hydrogel maintained an average energy efficiency of approximately 93.3%. Its scalable manufacturing process enables these solar water evaporators to desalinate saltwater and purify wastewater to meet WHO drinking standards [73]. Researchers are making significant strides in water purification technologies. By incorporating TiO_2 nanoparticles and single-walled carbon nanotubes into a polyacrylamide matrix, they developed a cost-effective hydrogel composite with 88.40% porosity, significantly enhancing photothermal and photocatalytic properties for solar-driven purification [74]. Kaolin/CNTs/PAM hydrogel (KCAH), a clay-based hydrogel evaporator, exhibits over 99% desalination efficiency and efficient photothermal evaporation, adsorbing heavy metal ions at different salinities [75]. The co-gelation of reduced graphene oxide (rGO), polyvinyl alcohol (PVA), and poly(acrylamide oxime) has produced an integrated solar-driven hydrogel evaporator (GPPH) with 1289 mg/g uranium adsorption capacity in simulated seawater with 64 ppm uranium [76]. The development of hydrogel composites represents a notable improvement in water treatment technology, providing an efficient, cost-effective, and environmentally friendly solution to address water contaminants.

6. Hydrogel Filtration Mechanisms

Hydrogels play an essential part in the removal of contaminants via a variety of methods that are controlled by the characteristics of the hydrogel as well as the parameters of the environment, such as the pH and the ionic strength [77,78], and ion exchange (Figure 3). These mechanisms are brought about by functional groups within the hydrogel composition. According to Zhou et al. [76], the adsorption capabilities of hydrogels are mainly influenced by the physical and chemical characteristics of the pollutants as well as the

circumstances of the microenvironment that exist in the surrounding area [77]. For example, using FTIR analysis, Zhou et al. [76] demonstrated functional groups' role in adsorption. They found that shifts in the bands of the hydroxyl and acetyl groups indicated changes in chemical interactions that occurred after the binding of contaminants. As validated by this research, complexation promoted by electron donation from nitrogen atoms inside the hydrogel structure was shown to be the predominant mechanism for the adsorption of lead ions into the hydrogel structure.

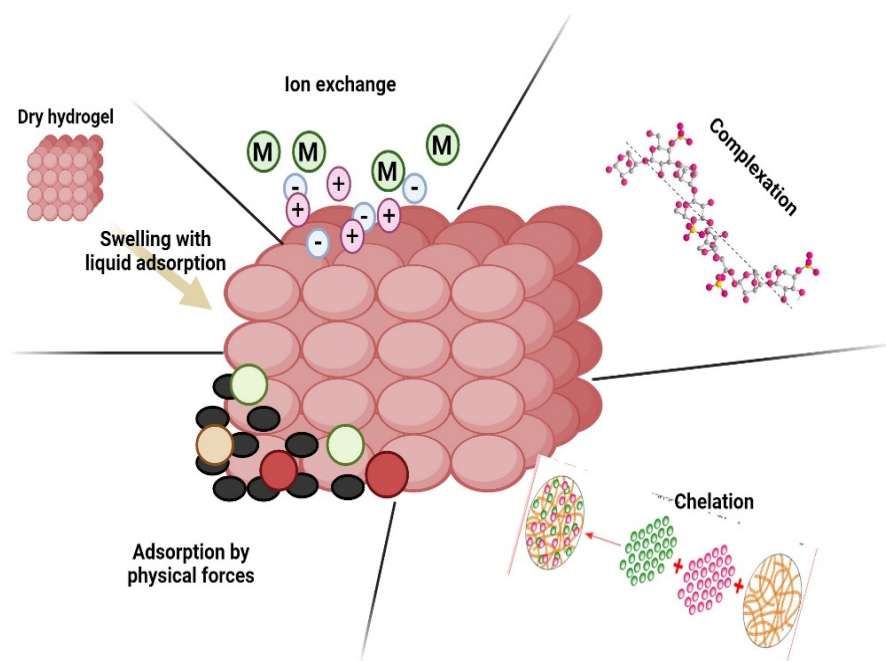


Figure 3. Mechanisms involved in contaminant removal with hydrogels.

As a consequence, the molecular weight of the hydrogel gradually increased [77]. X-ray diffraction (XRD) examination showed changes in the hydrogel's crystalline structure after the contaminants' adsorption, which indicates interactions with contaminants affect the crystalline phases of the hydrogel [34]. The XRD patterns demonstrated a decrease in crystallinity, which indicates that the amorphous regions of the hydrogel did play a substantial part in the adsorption process, which was accomplished by the hydrolysis product precipitation and complexation processes. The pH rise detected throughout the adsorption process supported these results, similar to the results on chitin cadmium absorption [77]. Moreover, cationic hydrogels have a relatively straightforward mechanism in terms of how they incorporate ionic contaminants into their structure. According to Barakat and Sahiner [78], these hydrogels can absorb water in aqueous conditions, expanding the network. Because of this, negatively charged metal ions, such as arsenate, can diffuse into the network and participate in electrostatic interactions, making it feasible for other ions to do the same. The pH significantly influences the solution, which in turn has a significant effect on the electrostatic interactions [79].

7. Hydrogel Adsorption Effectiveness

Hydrogels are three-dimensional networks that retain water. Polar or nonpolar monomers make hydrophilic or hydrophobic hydrogels, although application-specific hydrophobic hydrogels are made. Monomers, initiators, and crosslinkers are typical. Polar monomers are polymerized in situ to make hydrophilic hydrogels. Hydrogels are made by bulk, emulsion, and solution polymerization. In removing pollutants from water, the adsorption effectiveness of hydrogels is affected by several parameters, such as the hydro-

gel's surface morphology, the functional group involved, and ambient circumstances, such as pH and the presence of other ions [80]. Hydrogels have a network structure, making their surface area much more significant than their monomers or other adsorbents. This characteristic is one of the most critical factors in increasing the adsorption capacity of hydrogels. This surface area may be considerably improved using techniques such as freeze-drying during hydrogel manufacturing. These techniques include creating a highly porous structure with a broad pore size distribution, ultimately improving the hydrogel's total adsorption capacity [81]. Adsorption efficiency is also significantly impacted by environmental conditions, which play a vital effect. Hydrogels' efficiency in removing contaminants may be diminished by the presence of inorganic ions or natural organic materials that compete with them. Studies conducted by Tang et al. [79] indicated that when the sulfate concentration in groundwater increased, the Cr(VI) adsorption capacity decreased because of the decrease in the number of competitive ion exchange sites. Furthermore, pH variations may substantially impact the performance of hydrogels [80]. These changes affect the speciation of pollutants such as Cr (VI) and the surface charge of the hydrogel, which in turn modify the properties of adsorption [79,82].

8. Pros and Cons of Hydrogels and Regeneration

Modifying hydrogels' physical and chemical properties also increases their adsorption capacities. Ali et al. [82] and Sanyang et al. [66] discovered that adjusting parameters impacted the hydrogel's swelling capacity and mechanical strength. These factors included the monomer content and the crosslinking density. These characteristics are necessary for efficient adsorption and stability during several usage cycles [67,83]. Compared to other adsorbents, hydrogels have several significant benefits when removing contaminants. Even though some adsorbents may have more excellent adsorption capabilities, hydrogels are especially advantageous since they are simple to separate from water, which contrasts with other materials, such as activated carbon and biochar, that are difficult to recover after treatment. In addition, hydrogels may be used in a simplified purification method that only requires one step. According to Liang et al. [83], alternative adsorbents, on the other hand, often need intricate pH modifications to function well, which ultimately limits their use in natural water circumstances [84]. Because of this pH dependence, the application of many adsorbents is restricted. One example is carbon nanofiber membranes, which only work best within particular pH ranges for certain pollutants, such as Cr(VI) and Pb. Hydrogels, however, are effective throughout a more comprehensive pH range, increasing their applicability in various situations. They are economically more viable, especially in low-income regions, due to the lower production costs than those of synthetic adsorbents. Hydrogels can be regenerated and reused multiple times, offering long-term sustainability and cost-effectiveness that is not achievable with most low-cost adsorbents when used independently.

One of the distinguishing characteristics of hydrogels is their capacity to be regenerated and reused, making them an environmentally friendly choice for removing contaminants. Desorbing the adsorbed pollutants is often a part of the regeneration process, which can be accomplished by either pH modifications or magnetic separations [85]. More specifically, it has been shown that successful desorption may be accomplished using both strong and weak acids, influencing the hydrogel's ability for desorption and its lifetime [83,86]. HNO₃ and HCl, for example, break down the hydrogel after a few cycles, but H₂SO₄ retains its efficacy even after several cycles [86]. Also, magnetic hydrogels have demonstrated various degrees of endurance when subjected to different acids. UV irradiation is another unique method that may be used to regenerate graphene oxide–TiO₂ hydrogels. These hydrogels maintain a high adsorption capacity even after being subjected to several cycles [87].

Tang et al. [79] also showed the regeneration and reusability of hydrogels via a column experiment for removing Cr(VI) by a cationic hydrogel [80].

Interestingly, after six adsorption and desorption cycles, the adsorption capacity remained unchanged (27 mg/g, 90%), and the desorption efficiency was 93% of the average for each cycle [80]. During seven adsorption cycles, Liu et al. [84] demonstrated that 95% of copper, iron, and lead could be extracted from an aqueous solution. Furthermore, they revealed that the hydrogel retained its ability to adsorb metals. It has been shown that hydrogels may be reused for an extended period, increasing their efficiency and reducing the amount of money spent on manufacturing the contaminant removal procedures that take place in aqueous medium [85]. The capacity to reuse hydrogels increases their economic feasibility and demonstrates that they are safe for use in water purification operations from an environmental standpoint. In contrast to some sorbents, which lose their efficacy over time, hydrogels keep their adsorptive capabilities even after regenerating them. This trait makes hydrogels an excellent option for applications that involve the removal of contaminants over short periods.

Table 3 presents a comparison of various hydrogels for contaminant removal. Overall, natural hydrogels, such as lignin, kappa carrageenan, and chitosan, are biodegradable, reusable, and cost-effective media for contaminant removal from aqueous solution. They can be applied as standalone or composite hydrogels for heavy metals and dye treatment. Natural hydrogels' mechanical strength and stability were reported to control their applications in water and wastewater treatment. Combining natural hydrogels with organic and nonorganic nanoparticles was suggested to overcome these problems. Graphene oxide and carbonaceous nanofiber hydrogels were also used for contaminant treatment, although they are more expensive and complicated to fabricate than kappa carrageenan and chitosan ones.

Lignin hydrogels are biodegradable and cost-effective but have limited mechanical strength. They have the ability to remove metals like Pb^{2+} , Cr^{3+} , and Cu^{2+} via various interactions, with adsorption capacities of 308, 65.72, and 541 mg/g, respectively [88–90]. The vast surface area of graphene oxide-based hydrogels makes them particularly effective in removing dyes; nevertheless, manufacturing them is expensive and hazardous. Their adsorption capacities for methylene blue are 324 mg/g, whereas their adsorption capacities for rhodamine B are 114.4 mg/g [91].

Carbonaceous nanofiber membranes eliminate Pb^{2+} , Cr^{3+} , and methylene blue by electrostatic interactions and complexation, with high adsorption capacity but possible performance reduction over cycles [84]. Chitosan, maltodextrin, and gum arabic hydrogels efficiently remove Cd^{2+} while retaining biodegradability but lacking mechanical strength. Chitosan hydrogels are low cost and biodegradable; they have poor mechanical strength [92]. Aluminum hydroxide polyhydrate hydrogels reject multiple ions but perform poorly with monovalent ions [93]. Kappa carrageenan–vanillin hydrogels are biodegradable and efficient but need optimization owing to the limited mechanical strength to remove metal ions [94].

The kappa carrageenan–graft–poly(N hydroxyethyl acrylamide)/magnetite nanocomposite hydrogel removes cationic dyes and heavy metals via adsorption and magnetic separation. However, manufacturing is complex, and stability is uncertain [95]. Kappa carrageenan/tamarind kernel powder hydrogel absorbs brilliant green and Rose Bengal colors. Biodegradable and cost-effective, it may need regeneration and has water stability difficulties [96]. Kappa carrageenan-coated magnetic nanoparticles remove methylene blue swiftly, are eco-friendly, magnetically separate easily, and may be reused four times. While the kappa carrageenan/cellulose hydrogel is cost-effective and eco-friendly, it may lose its adsorption capacity at low pH and in overuse cycles [97].

The kappa carrageenan/graphene oxide hydrogel efficiently removes Na^+ and Mg^+ ions via electrostatic interactions and increased surface area, resulting in higher capacity and mechanical strength. While graphene oxide has a 95% removal efficiency for MgSO_4 and 23% for NaCl , its high cost and production complexity prevent its widespread application [98]. Despite high manufacturing costs and complex synthesis, the titanium dioxide nanoparticle-loaded hydrogel degrades methylene blue via adsorption and photocatalytic degradation under UV light, resulting in excellent adsorption capacity, mechanical strength, and biocompatibility. Adsorption capabilities range from 349.65 mg/g to 465.12 mg/g, depending on the TiO_2 concentration [99].

Chitosan and cellulose composites efficiently remove heavy metals (e.g., Cu, Zn, Cr, Ni, and Pb) by chelation and ion exchange. Biodegradable and eco-friendly, they lack mechanical stability [81]. The Fe_3O_4 -polyvinyl alcohol/chitosan combination adsorbs Cr^{6+} by electrostatic attraction and electron sharing, allowing magnetic separation and recycling, although efficiency drops at neutral pH [100]. Through multiple interactions, the dual-network chitosan-based hydrogel (CMAPP) eliminates methylene blue (MB) and malachite green (MG) dyes at >2000 mg/g and high recyclability, but not anionic dyes [101]. Though swelling is minimized, the CMC-g-poly(AM-co-ITA)/clay nanocomposite removes vivid green dye at 1513.55 mg/g [102]. Finally, the chitosan-ethylene glycol hydrogel (CEGH) eliminates perfluorooctanoic acid (PFOA) via ionic hydrogen bonding at 1275.9 mg/g, but just PFOA [103].

Table 3. Comparison of various hydrogels for contaminant removal.

| Hydrogels Type | Removal Mechanism | Metal Ions | Advantage | Disadvantage | Adsorption Capacity (mg/g) | References |
|---|--|--|--|--|---|------------|
| Lignin hydrogels (lignosulphonate-based graphene hydrogel Lignin grafted polyacrylic hydrogel Lignocellulose-based composite hydrogel | Hydrogen bonding, electrostatic, and covalent interactions Lignin-grafted polyacrylic hydrogel and lignocellulose-based composite hydrogels remove contaminants through adsorption, ion exchange, complexation, hydrogen bonding, and electrostatic interactions | Pb ²⁺ Cr ³⁺ Cu ²⁺ | The ability to biodegrade, a natural polymer, cost-effective, renewable, and versatile, with hydroxyl, methoxy, carboxyl, phenolic, and ether groups enhancing adsorption | Lignin hydrogels suffer from limited mechanical strength, poor hydrolysis, and chemical resistance; require pretreatment; and demonstrate potential leaching and lower swelling capacity | 308 65.72 541 | [88–90] |
| Graphene oxide-based hydrogels | Graphene oxide-based hydrogels can effectively remove dyes by using π – π interactions, van der Waals forces, hydrogen bonding, electrostatic interactions, and the utilization of their large surface area and porosity | Food Blue 2 and Food Red 17 | Superior adsorption for dyes due to the large surface area and functional groups Easily functionalized to target specific contaminants Robust and durable due to GO Maintains performance under varying temperatures Safe for the environment Efficient dye removal within approximately 4 h Reusable 3–5 times, with some loss of effectiveness | High synthesis and processing costs Possible toxicity if not adequately purified Environmental impact Potential long-term stability issues | For both methylene blue (MB) and rhodamine B (RhB), the absorption capacity was 324 mg/g and 114.4 mg/g | [91] |
| Carbonaceous nanofiber membranes | Adsorption through electrostatic interactions, hydrogen bonding, and surface complexation | Pb ²⁺ , Cr ³⁺ , methylene blue | High surface area, fast adsorption rates, high adsorption capacity, regenerable and reusable over multiple cycles, and efficient at high flux | Potential for performance decline over multiple use cycles, complex manufacturing process | Methylene blue: up to 818.6 mg/g Pb(II): up to 423.7 mg/g Cr(VI): up to 221.3 mg/g | [84] |
| Hydrogels based on chitosan, maltodextrin, and gum arabic | Adsorption by physical forces, ion exchange, complexation | Cd ²⁺ | Biodegradable, low cost, abundant, high efficiency | Limited mechanical strength, biodegradability | 80–150 mg/g | [92] |
| Polyvinyl alcohol/chitosan | PVA/chitosan hydrogels remove Mn (II) ions from aqueous solutions by physical adsorption, ion exchange, and complexation | Mn ²⁺ | Biodegradable, high adsorption capacity, low cost | Biodegradability, limited mechanical strength | 10.5 mg/g | [104] |
| Aluminum hydroxide Polyhydrate hydrogel | Adsorption, ion exchange, electrostatic attraction | Pb ²⁺ , Cu ²⁺ , Mg ²⁺ , Ca ²⁺ , Ni ¹⁺ , Zn ²⁺ , As ⁵⁺ | High rejection rates, stable performance, easy to fabricate, reusable 4 times | Limited effectiveness for monovalent ions, potential for clogging | Removal efficiency for Pb, Cu, and Zn was 99.9%, Ni: 100%, Mg: 85%, Ca: 84% | [93] |
| Kappa carrageenan–vanillin hydrogel | Adsorption, ion exchange, chelation mechanism | Pb ²⁺ , Cu ²⁺ , Mg ²⁺ , Ca ²⁺ , Na ¹⁺ | High removal efficiency, low energy consumption, biodegradable | Low mechanical strength requires optimization for stability | Cu: 1.22 mg/g Removal efficiency for Mg: 88%, pb: 79%, Cu: 82%, Ca: 82%, Na: 38% | [94] |
| Kappa carrageenan–graft–poly(N hydroxyethyl acrylamide)/magnetite nanocomposite (κ -g-PHEAA/Fe ₃ O ₄) hydrogel | Adsorption, magnetic separation | Cationic dyes (MB, R6G), heavy metals (Cu ²⁺ , Hg ²⁺) | High adsorption capacity and reusable, magnetic properties facilitate separation | Complex synthesis, potential stability issues | MB: 196.1, R6G: 307.8, Cu(II): 243.4, Hg(II): 229.9 | [95] |
| Kappa carrageenan/tamarind kernel powder hydrogel | Adsorption | Brilliant green (BG), Rose Bengal (RB) | Biodegradable, cost-effective, high adsorption capacity, pH-responsive | May require regeneration, stability issues in water | 840.33 mg/g (BG), 168.06 mg/g (RB) | [96] |

Table 3. *Cont.*

| Hydrogels Type | Removal Mechanism | Metal Ions | Advantage | Disadvantage | Adsorption Capacity (mg/g) | References |
|---|---|--|---|---|---|------------|
| Kappa carrageenan-coated magnetic nanoparticles | Kappa carrageenan sulfonate groups adsorb and electrostatically interact with methylene blue (MB) | Methylene blue (MB) | High adsorption capacity, quick removal (5 min), eco-friendly, facile magnetic separation, and neutral and alkaline chemical stability Reusable 4 times | This product features rapid removal (5 min) and high adsorption capacity, is eco-friendly, has quick magnetic separation, and shows excellent chemical stability in neutral and alkaline environments | 185.3 mg/g at pH 9 | [97] |
| Kappa carrageenan/cellulose hydrogel | Adsorption, electrostatic interactions, and ion exchange between Pb ²⁺ ions and functional groups (such as –OH and –COOH) | Pb ²⁺ ions | High adsorption capacity, eco-friendly, reusable (with an effectiveness of over 79% after eight cycles), and cost-effective | At low pH, adsorption capability may decrease during reuse cycles. | 486 ± 28.5 mg/g | [62] |
| Kappa carrageenan/graphene oxide hydrogel | Electrostatic interactions, increased surface area, and hydrophilicity caused by graphene oxide and kappa carrageenan contribute to adsorption | Na ¹⁺ , Mg ²⁺ | High adsorption capacity, improved mechanical strength, enhanced selectivity, and antifouling properties | Graphene oxide is expensive, and manufacturing complex and performance deteriorate over numerous cycles | Removal efficiency MgSO ₄ : 95%, NaCl: 23% | [98] |
| Titanium dioxide nanoparticle-loaded κ-carrageenan hydrogel | Adsorption and photocatalytic degradation TiO ₂ nanoparticles, when exposed to ultraviolet light, produce reactive oxygen species, which contribute to the degradation of dyes | Methylene blue | High adsorption capacity, improved mechanical strength, UV-responsive degradation, and biocompatibility | High production costs, complex synthesis process | Methylene blue: 349.65 mg/g to 465.12 mg/g depending on TiO ₂ concentration and crosslinking | [99] |
| Chitosan (chitin)/cellulose composite hydrogel | Adsorption via chelation and ion exchange through hydroxyl and amine groups | Cu ²⁺ , Zn ²⁺ , Cr ⁶⁺ , Ni ²⁺ , Pb ²⁺ | High adsorption efficiency, eco-friendly, biodegradable, improved stability with freeze-drying | Limited mechanical stability requires regeneration | Cu ²⁺ (0.417 mmol/g), Zn ²⁺ (0.303 mmol/g), Cr ⁶⁺ (0.251 mmol/g), Ni ²⁺ (0.225 mmol/g), Pb ²⁺ (0.127 mmol/g) | [81] |
| Fe ₃ O ₄ -polyvinyl alcohol/chitosan composite hydrogel | Adsorption via electrostatic attraction and electron-sharing | Cr ⁶⁺ | High adsorption efficiency, magnetic separability, good repeatability | Decreased swelling ratio, limited adsorption under neutral conditions | Cr ⁶⁺ (0.267 g/g) | [100] |
| Dual-network chitosan-based hydrogel (CMAPP) | Adsorption via electrostatic attraction, hydrogen bonding, and n-π interactions | Methylene blue (MB), malachite green (MG), crystalline violet (CV), basic fuchsin (BF) | High adsorption efficiency, strong anti-interference, excellent recyclability | Limited adsorption for anionic dyes | MB: 1800 mg/g, MG: 1797 mg/g, CV: 2495 mg/g, BF: 2938 mg/g | [101] |
| Carboxymethyl cellulose(CMC)-g-poly(AM-co-ITA)/clay nanocomposite hydrogel | Adsorption via electrostatic interactions and multilayer adsorption | Brilliant green (BG) dye | High adsorption capacity, enhanced stability with clay, easy separation, strong reusability | Reduced swelling ratio with higher clay content | BG: 1513.55 mg/g | [102] |
| Novel chitosan–ethylene glycol hydrogel (CEGH) | Ionic hydrogen bond interaction between carbonyl groups of PFOA and protonated amine groups of CEGH | Perfluorooctanoic acid (PFOA) | High adsorption capacity, simple synthesis via freeze-thawing, eco-friendly | Limited to specific contaminants like PFOA | PFOA: 1275.9 mg/g | [103] |

9. Hydration Phenomena in Hydrogel: Mechanisms, Measurements

Hydrogel hydration phenomena include water absorption and swelling, which affect their physical and chemical characteristics. Hydrogels made from kappa carrageenan, chitosan, and cellulose absorb water well owing to their hydrophilic and porous properties. When these gels come into contact with water, they undergo swelling driven by osmosis, where water molecules penetrate the gel matrix, causing it to expand. This hydration enhances the gel's mechanical strength and flexibility, making it appropriate for medication administration, wound healing, and environmental cleanup. The gel's crosslinking density affects swelling, water absorption, ionic groups, pH, and temperature. Hydration mechanisms are crucial to the polymer hydrogel sol–gel transition, as responsive hydrogel systems show. A polymer hydrogel's structural makeup is 90% water; hence, understanding hydration processes is vital. Hydrogels may expand their dry weight 1000-fold in water or salt water [34], with Gibas and Janik [105] expressing sorbed water as equilibrium water content (EWC) in Equation (1).

$$EWC\% = \frac{W_w}{W_t} \times 100 \quad (1)$$

W_w is the dry gel weight, and W_t is the hydrated gel weight.

Figure 4 shows a multi-step technique for hydrating and expanding swellable polymer systems [105,106]. Water molecules first hydrate the hydrogel matrix's most polar, hydrophilic active spots to generate primary bound water. This water is essential to the hydrogel structure and cannot be separated. Hydrophobic sites interact with water molecules to create secondary bonded water in the next phase. Total bound water has intermediate qualities from primary and secondary bound fluids [106]. In the next stage, the covalent or physical crosslinks resist the polymer network's osmotic driving force toward infinite dilution. Thus, they absorb water equal to the polymer's equilibrium swelling capacity. Most free water is between network chains and bigger holes, micropores, or voids [107]. Due to electrostatic repulsion, polymer chains stretch, generating vacuum regions in the polymer network where much loosely bound water may be absorbed. Thus, the polymer crosslinking strategy during hydrogel fabrication is crucial to network formation. It increases the number of surface-active sites and interstitial domains, makes the copolymer insoluble in water, and increases the polymer network's mechanical strength and swelling.

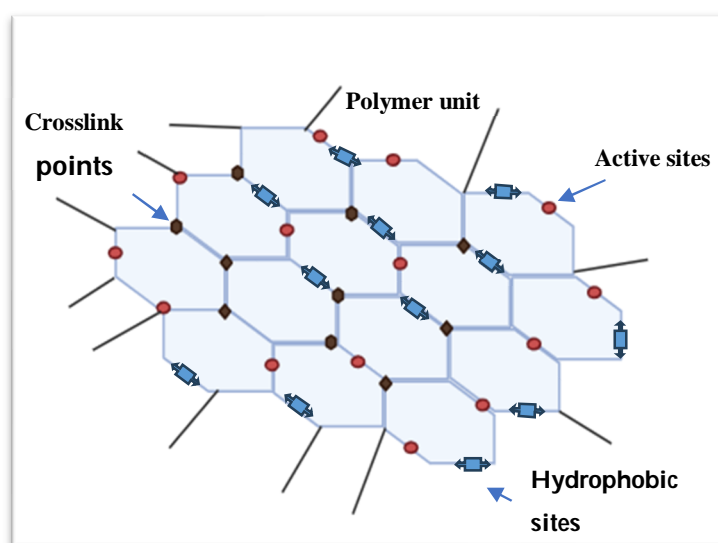


Figure 4. Illustration depicting the structure of a polymer network.

To quantify polymer networks' water absorption (uptake) features, water adsorption/desorption, water retention time (WRt), swelling rates, and repeatability performance were estimated. Equations (2) and (3) are used to calculate the water adsorption/swelling rate and the reproducibility performance of a hydrogel substance. $WR(t)$ is defined as the ratio of the weight of the water retained by the hydrogel at a specific time to the weight of the dry hydrogel by Equation (4).

$$\text{Swelling ratio}(SR\%) = \frac{(W2 - W1)}{W2} \times 100 \quad (2)$$

$$\text{Water uptake} \left(WU \frac{g}{g} \right) = \frac{(W2 - W1)}{W1} \quad (3)$$

$$\text{Water retention time} \left(WRt \frac{g}{g} \right) = \frac{(Wt - Wd)}{Wd} \quad (4)$$

$$\text{Deswelling}(DS\%) = \frac{Ws}{W0} \times 100 \quad (5)$$

Equations (2) and (3) measure the swelling ratio and water uptake. After 24 h of equilibrium swelling, the hydrogel weighs $W2$ in g, and $W1$ is the dry material's weight after filtering or oven drying to constant weight. In Equation (4), Wt is the weight of the hydrogel at time t after it has absorbed water, and Wd is the hydrogel's initial dry weight. Deswelling refers to the process by which a hydrogel releases absorbed water and contracts, returning closer to its original dry state. This characteristic is essential for applications that require hydrogels to undergo multiple swelling–deswelling cycles while maintaining their structural integrity and functionality. Deswelling (DS) was calculated using Equation (5), with Ws representing the weight of the swollen hydrogel and $W0$ the weight after vacuum drying for 30 min.

The hydrogel's swelling and deswelling phenomenon demonstrates its ability to retain and release water. Equation (6) calculates the moisture desorption rate by allowing a sample with a known weight W (g) to absorb and release water at a particular temperature, relative humidity, and time. $W1$ and $W2$ are sample weights after sorption and desorption, while WD is the water desorption rate.

$$\text{Water desorption}(WD\%) = \frac{(W1 - W2)}{W} \times 100 \quad (6)$$

The elastic response of polymer chains and chemical and physical factors affect hydrogel composite water absorption and swelling. Polymer networks swell when water enters the hydrophilic matrix via diffusion and capillary action. The flexibility of crosslink sites allows hydrogel structures to expand and contract in response to physical or chemical stimuli. Hydrogel swelling is affected by crosslinking density, particular surface area, hydrophile–lipophile character, and medium ionic strength.

The salt sensitivity factor, calculated from equilibrium swelling in saline solution to deionized water, assesses how ionic strength affects polymer swelling. Smaller f -values suggest a lesser electrolyte swelling reaction. Higher cation charges promote crosslinking, diminishing swelling dynamics. Zhang and Francisco et al. examined the effects of monovalent (K^+ , Na^+ , NH_4^+), divalent (Ca^{2+}), and trivalent (Al^{3+}) ions on chitosan- and starch-based hydrogel swelling. Monovalent, divalent, and trivalent cations limit water absorption. Cationic charge enhances crosslinking density, minimizing swelling [108,109]. The salt sensitivity factor (f) shows this link, with higher f -values affecting swelling more. The swelling capacity (g/g) of chitosan-based polymers declines with larger f -values: NH_4^+ (0.69; 43.3 g/g) < Na^+ (0.76; 33.6 g/g) < Ca^{2+} (0.93; 10.2 g/g) < Al^{3+} (0.97; 4.7 g/g). Chemical complexation and electrostatic interaction with polymer matrix let multivalent

cations produce more crosslinks [110]. Higher crosslink density reduces swelling by limiting access to interstitial polymer domains. Increased ionic strength also lowers osmotic swelling pressure, causing the gel to contract. Alam et al. found that cellulose-based hydrogels' swelling and WR significantly decreased from 725 g/g in deionized water to 118 g/g in saline water due to the charge screening effect [111].

$$\text{Salt sensitivity factor} = 1 - \frac{W_{\text{saline}}}{W_{\text{water}}} \quad (7)$$

Water absorption and swelling are stronger in hydrogels of hydrophilic polymers like hydroxyl or carboxyl groups. Due to charge screening, cellulose-based hydrogels expand greatly in deionized water but less in saline [111]. Higher environmental ionic strength minimizes the osmotic pressure differential, contracting the hydrogel. The polymer's molecular weight and chemical structure impact the hydrogel's environmental response. Due to differences in polymeric chains and functional groups, chitosan-based hydrogels swell differently than cellulose-based ones [108]. Higher-molecular-weight polymers make stronger, more durable hydrogels but may expand less.

10. Conclusions

Water purification methods have undergone tremendous development to meet the rising concerns over water pollution produced by contaminants from industrial, agricultural, and sewage sources. The efficiency of membrane technologies such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) in removing a wide variety of pollutants depends on size exclusion, adsorption, and electrostatic interactions. These technologies have become a vital part of the different approaches. Fouling, high energy consumption, and high operating expenses are obstacles these technologies confront, even though they have beneficial applications. In light of this, novel materials such as hydrogels provide potentially promising alternatives. Because of their one-of-a-kind three-dimensional polymer networks, large surface area, and quick regeneration capabilities, hydrogels provide a solution for water purification that is both cost-effective and efficient. They can absorb various pollutants, including organic toxins and heavy metals, making them appropriate for emergencies and environmentally responsible water treatment procedures. Hydrogel composites have been developed to further improve their functioning by merging the characteristics of natural and synthetic materials, increasing these composites' adsorptive capacity and compatibility with the environment. The adsorption processes of hydrogels, which functional groups and ambient circumstances regulate, make it possible for hydrogels to effectively remove pollutants via complexation, ion exchange, and physical adsorption. Hydrogels are a better option than traditional adsorbents because they can undergo regeneration via various mechanisms. This guarantee of long-term sustainability and cost-effectiveness positions hydrogels as a superior choice. Despite these advantages, several challenges remain that require further investigation. These include improving the structural integrity of hydrogels to withstand prolonged operational use, tailoring hydrogel compositions to maximize adsorption capacity for specific pollutants and developing cost-effective synthesis methods for large-scale production and real-world implementation. Additionally, there is a need to explore the long-term environmental impact and degradation mechanisms of hydrogel-based materials and investigate multifunctional hydrogels that incorporate nanomaterials for selective adsorption and self-regeneration properties. Addressing these research gaps will pave the way for more effective, sustainable, and commercially viable hydrogel-based water purification technologies.

Author Contributions: Conceptualization, L.A. (Lilyan Alsaka), L.A. (Lina Alsaka), A.A. and T.K.; methodology, L.A. (Lilyan Alsaka), S.J.Z., T.K. and L.A. (Lina Alsaka); validation, L.A. (Lilyan Alsaka), L.A. (Lina Alsaka), A.A. and S.J.Z.; formal analysis, L.A. (Lilyan Alsaka) and L.A. (Lina Alsaka); investigation, L.A. (Lilyan Alsaka), T.K., A.A., J.Z. and S.J.Z.; data curation, J.Z., A.A. and L.A. (Lilyan Alsaka); writing—original draft preparation, L.A. (Lilyan Alsaka), L.A. (Lina Alsaka) and A.A.; writing—review and editing, L.A. (Lilyan Alsaka), A.A. and J.Z.; visualization, L.A. (Lilyan Alsaka) and A.A.; supervision, A.A. and J.Z.; project administration, A.A.; funding acquisition, A.A. and S.J.Z. All authors have read and agreed to the published version of the manuscript.

Funding: This research is made possible by a food security research award (MME04-0607-230061) from the Qatar National Research Fund (QNRF) in partnership with the Ministry of Municipality. The statements made herein are solely the responsibility of the authors.

Conflicts of Interest: The authors declare no conflict of interest.

References

- Bolisetty, S.; Peydayesh, M.; Mezzenga, R. Sustainable technologies for water purification from heavy metals: Review and analysis. *Chem. Soc. Rev.* **2019**, *48*, 463–487. [\[CrossRef\]](#) [\[PubMed\]](#)
- Ray, S.S.; Iroegbu, A.O.C. Nanocellulosics: Benign, Sustainable, and Ubiquitous Biomaterials for Water Remediation. *ACS Omega* **2021**, *6*, 4511–4526. [\[CrossRef\]](#) [\[PubMed\]](#)
- Acikgoz, C.; Gül, Ü.D.; Özkan, K.; Borazan, A.A. Degradation of Reactive Blue by the mixed culture of *Aspergillus versicolor* and *Rhizopus arrhizus* in membrane bioreactor (MBR) system. *Desalination Water Treat.* **2016**, *57*, 3750–3756. [\[CrossRef\]](#)
- Prajapati, M.; Shah, M.; Soni, B. A review of geothermal integrated desalination: A sustainable solution to overcome potential freshwater shortages. *J. Clean. Prod.* **2021**, *326*, 129412. [\[CrossRef\]](#)
- Mulyanti, R.; Susanto, H. Wastewater treatment by nanofiltration membranes. In *IOP Conference Series: Earth and Environmental Science*; IOP Publishing: Bristol, UK, 2018; Volume 142.
- Shon, H.K.; Phuntsho, S.; Chaudhary, D.S.; Vigneswaran, S.; Cho, J. Nanofiltration for water and wastewater treatment—A mini review. *Drink. Water Eng. Sci.* **2013**, *6*, 47–53. [\[CrossRef\]](#)
- Anis, S.F.; Hashaikeh, R.; Hilal, N. Reverse osmosis pretreatment technologies and future trends: A comprehensive review. *Desalination* **2019**, *452*, 159–195. [\[CrossRef\]](#)
- Macedonio, F.; Drioli, E.; Gusev, A.; Bardow, A.; Semiat, R.; Kurihara, M. Efficient technologies for worldwide clean water supply. *Chem. Eng. Process.* **2012**, *51*, 2–17. [\[CrossRef\]](#)
- Jiang, S.-X.; Li, Y.-N.; Ladewig, B.P. A review of reverse osmosis membrane fouling and control strategies. *Sci. Total Environ.* **2017**, *595*, 567–583. [\[CrossRef\]](#)
- Qasim, M.; Badrelzaman, M.; Darwish, N.N.; Hilal, N. Reverse osmosis desalination: A state-of-the-art review. *Desalination* **2019**, *459*, 59–104. [\[CrossRef\]](#)
- Mishra, N.S.; Reddy, R.; Kuila, A.; Rani, A.; Nawaz, A.; Pichiah, S.; Mukherjee, P. A Review on Advanced Oxidation Processes for Effective Water Treatment. *Curr. World Environ.* **2017**, *12*, 469–489. [\[CrossRef\]](#)
- Parsons, S. *Advanced Oxidation Processes for Water and Wastewater Treatment*; IWA Publishing: London, England, 2004.
- Lee, A.; Elam, W.; Darling, S.B. Membrane materials for water purification: Design, development, and application. *Environ. Sci. Water Res. Technol.* **2016**, *2*, 17–42.
- Chelu, M.; Musuc, A.M.; Popa, M.; Moreno, J.M.C. Chitosan hydrogels for water purification applications. *Gels* **2023**, *9*, 664. [\[CrossRef\]](#) [\[PubMed\]](#)
- Patra, S.K.; Poddar, R.; Brestic, M.; Acharjee, P.U.; Bhattacharya, P.; Sengupta, S.; Pal, P.; Bam, N.; Biswas, B.; Barek, V.; et al. Prospects of hydrogels in agriculture for enhancing crop and water productivity under water deficit condition. *Int. J. Polym. Sci.* **2022**, *2022*, 4914836. [\[CrossRef\]](#)
- Ho, T.-C.; Chang, C.-C.; Chan, H.-P.; Chung, T.-W.; Shu, C.-W.; Chuang, K.-P.; Duh, T.-H.; Yang, M.-H.; Tyan, Y.-C. Hydrogels: Properties and applications in biomedicine. *Molecules* **2022**, *27*, 2902. [\[CrossRef\]](#)
- Ansar, R.; Saqib, S.; Mukhtar, A.; Niazi, M.B.K.; Shahid, M.; Jahan, Z.; Kakar, S.J.; Uzair, B.; Mubashir, M.; Ullah, S.; et al. Challenges and recent trends with the development of hydrogel fiber for biomedical applications. *Chemosphere* **2022**, *287*, 131956. [\[CrossRef\]](#)
- Cai, J.; Guo, J.; Wang, S. Application of Polymer Hydrogels in the Prevention of Postoperative Adhesion: A Review. *Gels* **2023**, *9*, 98. [\[CrossRef\]](#)
- Zhao, L.; Zhou, Y.; Zhang, J.; Liang, H.; Chen, X.; Tan, H. Natural polymer-based hydrogels: From polymer to biomedical applications. *Pharmaceutics* **2023**, *15*, 2514. [\[CrossRef\]](#)

20. Kalendova, P.; Svoboda, L.; Hroch, J.; Honcova, P.; Drobna, H.; Slang, S. Hydrogels based on starch from various natural sources: Synthesis and characterization. *Starch-Stärke* **2021**, *73*, 2100051. [[CrossRef](#)]
21. Bhat, M.P.; Lee, J.-H.; Kurkuri, M.; Chen, T.; Kim, C.S.; Lee, K.-H. Diatom contained alginate-chitosan hydrogel beads with enhanced hydrogen bonds and ionic interactions for extended release of gibberellic acid. *Int. J. Biol. Macromol.* **2025**, *291*, 138906. [[CrossRef](#)]
22. Bashir, S.; Hina, M.; Iqbal, J.; Rajpar, A.H.; Mujtaba, M.A.; Alghamdi, N.A.; Wageh, S.; Ramesh, K.; Ramesh, S. Fundamental concepts of hydrogels: Synthesis, properties, and their applications. *Polymers* **2020**, *12*, 2702. [[CrossRef](#)]
23. Kuddushi, M.; Rajput, S.; Shah, A.; Mata, J.P.; Aswal, V.K.; El Seoud, O.A.; Kumar, A.; Malek, N.I. Stimuli responsive, self-sustainable, and self-healable functionalized hydrogel with dual gelation, load-bearing, and dye-absorbing properties. *ACS Appl. Mater. Interfaces* **2019**, *11*, 19572–19583. [[CrossRef](#)]
24. Ullah, F.; Othman, M.B.H.; Javed, F.; Ahmad, Z.; Akil, H.M. Classification, processing and application of hydrogels: A review. *Mater. Sci. Eng. C* **2015**, *57*, 414–433. [[CrossRef](#)] [[PubMed](#)]
25. Singhal, R.; Gupta, K. A review: Tailor-made hydrogel structures (classifications and synthesis parameters). *Polym. Plast. Technol. Eng.* **2016**, *55*, 54–70. [[CrossRef](#)]
26. Sargeant, T.D.; Desai, A.P.; Banerjee, S.; Agawu, A.; Stopek, J.B. An in situ forming collagen-PEG hydrogel for tissue regeneration. *Acta Biomater.* **2012**, *8*, 124–132. [[CrossRef](#)]
27. Mehta, P.; Sharma, M.; Devi, M. Hydrogels: An overview of its classifications, properties, and applications. *J. Mech. Behav. Biomed. Mater.* **2023**, *147*, 106145. [[CrossRef](#)]
28. Erathodiyil, N.; Chan, H.-M.; Wu, H.; Ying, J.Y. Zwitterionic polymers and hydrogels for antibiofouling applications in implantable devices. *Mater. Today* **2020**, *38*, 84–98. [[CrossRef](#)]
29. Maitra, J.; Shukla, V.K. Cross-linking in hydrogels—A review. *Am. J. Polym. Sci* **2014**, *4*, 25–31.
30. Bustamante-Torres, M.; Romero-Fierro, D.; Arcentales-Vera, B.; Palomino, K.; Magaña, H.; Bucio, E. Hydrogels classification according to the physical or chemical interactions and as stimuli-sensitive materials. *Gels* **2021**, *7*, 182. [[CrossRef](#)]
31. Khansari, M.M.; Sorokina, L.V.; Mukherjee, P.; Mukhtar, F.; Shirdar, M.R.; Shahidi, M.; Shokuhfar, T. Classification of hydrogels based on their source: A review and application in stem cell regulation. *JOM* **2017**, *69*, 1340–1347. [[CrossRef](#)]
32. Rabajczyk, A.; Gniazdowska, J.; Stojek, P.; Czyżewska, K.; Trusek, A.; Labus, K. Hydrogels and Their Functionalization—Analysis of the Possibility of Their Application in Post-Fire Water Treatment Processes. *Materials* **2024**, *17*, 5820. [[CrossRef](#)]
33. Liu, Y.; Wang, J.; Chen, H.; Cheng, D. Environmentally friendly hydrogel: A review of classification, preparation and application in agriculture. *Sci. Total Environ.* **2022**, *846*, 157303. [[CrossRef](#)]
34. Ahmed, E.M. Hydrogel: Preparation, characterization, and applications: A review. *J. Adv. Res.* **2015**, *6*, 105–121. [[CrossRef](#)] [[PubMed](#)]
35. Pooremaeil, M.; Namazi, H. Application of polysaccharide-based hydrogels for water treatments. In *Hydrogels Based on Natural Polymers*; Elsevier: Amsterdam, The Netherlands, 2020; pp. 411–455.
36. Caló, E.; Khutoryanskiy, V.V. Biomedical applications of hydrogels: A review of patents and commercial products. *Eur. Polym. J.* **2015**, *65*, 252–267. [[CrossRef](#)]
37. Zhang, R.; Tian, J.; Gao, S.; Van der Bruggen, B. How to coordinate the trade-off between water permeability and salt rejection in nanofiltration? *J. Mater. Chem. A Mater. Energy Sustain.* **2020**, *8*, 8831–8847. [[CrossRef](#)]
38. Yoon, Y.; Lueptow, R.M. Removal of organic contaminants by RO and NF membranes. *J. Membr. Sci.* **2005**, *261*, 76–86. [[CrossRef](#)]
39. Van der Bruggen, B.; Vandecasteele, C. Modelling of the retention of uncharged molecules with nanofiltration. *Water Res.* **2002**, *36*, 1360–1368. [[CrossRef](#)]
40. Chang, E.-E.; Liang, C.-H.; Huang, C.-P.; Chiang, P.-C. A simplified method for elucidating the effect of size exclusion on nanofiltration membranes. *Sep. Purif. Technol.* **2012**, *85*, 1–7. [[CrossRef](#)]
41. Miron, S.M.; Dutournié, P.; Ponche, A. Filtration of Uncharged Solutes: An Assessment of Steric Effect by Transport and Adsorption Modelling. *Water* **2019**, *11*, 2173. [[CrossRef](#)]
42. Huber, F.; Berwanger, J.; Polesya, S.; Mankovsky, S.; Ebert, H.; Giessibl, F.J. Chemical bond formation showing a transition from physisorption to chemisorption. *Sci. Am. Assoc. Adv. Sci.* **2019**, *366*, 235–238. [[CrossRef](#)]
43. de Andrade, J.R.; Oliveira, M.F.; da Silva, M.G.; Vieira, M.G. Adsorption of Pharmaceuticals from Water and Wastewater Using Nonconventional Low-Cost Materials: A Review. *Ind. Eng. Chem. Res.* **2018**, *57*, 3103–3127. [[CrossRef](#)]
44. Grassi, M.; Kaykioglu, G.; Belgiorno, V.; Lofrano, G. Emerging compounds removal from wastewater. *Green Chem. Sustain* **2012**, *2*, 15–38.
45. Ambashta, R.D.; Sillanpää, M. Water purification using magnetic assistance: A review. *J. Hazard. Mater.* **2010**, *180*, 38–49. [[CrossRef](#)] [[PubMed](#)]
46. Ahmed, M.J.K.; Ahmaruzzaman, M. Investigation on the effective remediation of quinoline at solid/solution interface using modified agricultural waste: An inclusive study. *Int. J. Environ. Sci. Technol.* **2016**, *13*, 1177–1188. [[CrossRef](#)]
47. Mulder, M. *Basic Principles of Membrane Technology*; Springer Science & Business Media: Berlin/Heidelberg, Germany, 2012.

48. Pendergast, M.M.; Hoek, E.M. A review of water treatment membrane nanotechnologies. *Energy Environ. Sci.* **2011**, *4*, 1946–1971. [[CrossRef](#)]
49. Hilal, N.; Al-Zoubi, H.; Darwish, N.A.; Mohammad, A.W.; Arabi, M.A. A comprehensive review of nanofiltration membranes: Treatment, pretreatment, modelling, and atomic force microscopy. *Desalination* **2004**, *170*, 281–308. [[CrossRef](#)]
50. Verliefde, A.; Cornelissen, E.; Amy, G.; Van der Bruggen, B.; van Dijk, H. Priority organic micropollutants in water sources in Flanders and the Netherlands and assessment of removal possibilities with nanofiltration. *Environ. Pollut.* **2007**, *146*, 281–289. [[CrossRef](#)]
51. Yaroshchuk, A.E. Rejection mechanisms of NF membranes. *Membr. Technol.* **1998**, *1998*, 9–12.
52. Roy, Y.; Warsinger, D.M.; Lienhard, J.H. Effect of temperature on ion transport in nanofiltration membranes: Diffusion, convection and electromigration. *Desalination* **2017**, *420*, 241–257. [[CrossRef](#)]
53. Liu, Y.; Lin, B.; Liu, W.; Li, J.; Gao, C.; Pan, Q. Preparation and characterization of a novel nanofiltration membrane with chlorine-tolerant property and good separation performance. *RSC Adv.* **2018**, *8*, 3643–3644. [[CrossRef](#)]
54. Bowen, W.R.; Welfoot, J.S. Modelling the performance of membrane nanofiltration—Critical assessment and model development. *Chem. Eng. Sci.* **2002**, *57*, 1121–1137. [[CrossRef](#)]
55. Geng, X.; Wang, J.; Ding, Y.; Zhang, W.; Wang, Y.; Liu, F. Poly(vinyl alcohol)/polydopamine hybrid nanofiltration membrane fabricated through aqueous electrospraying with excellent antifouling and chlorine resistance. *J. Membr. Sci.* **2021**, *632*, 119385. [[CrossRef](#)]
56. Ritt, C.L.; Werber, J.R.; Wang, M.; Yang, Z.; Zhao, Y.; Kulik, H.J.; Elimelech, M. Ionization behavior of nanoporous polyamide membranes. *Proc. Natl. Acad. Sci. USA* **2020**, *117*, 30191–30200. [[CrossRef](#)] [[PubMed](#)]
57. Hoare, T.R.; Kohane, D.S. Hydrogels in drug delivery: Progress and challenges. *Polymers* **2008**, *49*, 1993–2007. [[CrossRef](#)]
58. Perumal, S.; Atchudan, R.; Edison, T.N.J.I.; Babu, R.S.; Karpagavinayagam, P.; Vedhi, C. A short review on recent advances of hydrogel-based adsorbents for heavy metal ions. *Metals* **2021**, *11*, 864. [[CrossRef](#)]
59. Ferfera-Harrar, H.; Benhalima, T.; Sadi, A. Development of functional chitosan-based superabsorbent hydrogel nanocomposites for adsorptive removal of Basic Red 46 textile dye. *Polym. Bull.* **2022**, *79*, 6141–6172. [[CrossRef](#)]
60. Dalalibera, A.; Vilela, P.B.; Vieira, T.; Becegato, V.A.; Paulino, A.T. Removal and selective separation of synthetic dyes from water using a polyacrylic acid-based hydrogel: Characterization, isotherm, kinetic, and thermodynamic data. *J. Environ. Chem. Eng.* **2020**, *8*, 104465. [[CrossRef](#)]
61. Wang, Z.; Zhang, G.; Li, Y. Preparation of chitosan/polyacrylamide/graphene oxide composite membranes and study of their methylene blue adsorption properties. *Materials* **2020**, *13*, 4407. [[CrossRef](#)]
62. Kalaiselvi, K.; Mohandoss, S.; Ahmad, N.; Khan, M.R.; Manoharan, R.K. Adsorption of Pb²⁺ Ions from Aqueous Solution onto Porous Kappa-Carrageenan/Cellulose Hydrogels: Isotherm and Kinetics Study. *Sustainability* **2023**, *15*, 9534. [[CrossRef](#)]
63. Saito, T.; Nishiyama, Y.; Putaux, J.-L.; Vignon, M.; Isogai, A. Homogeneous suspensions of individualized microfibrils from TEMPO-catalyzed oxidation of native cellulose. *Biomacromolecules* **2006**, *7*, 1687–1691. [[CrossRef](#)]
64. Zhang, Y.; Li, Z. Heavy metals removal using hydrogel-supported nanosized hydrous ferric oxide: Synthesis, characterization, and mechanism. *Sci. Total Environ.* **2017**, *580*, 776–786. [[CrossRef](#)]
65. Cova, T.F.G.G.; Murtinho, D.; Pais, A.A.C.C.; Valente, A.J.M. Cyclodextrin-based materials for removing micropollutants from wastewater. *Curr. Org. Chem.* **2018**, *22*, 2150–2181. [[CrossRef](#)]
66. Chen, J.; Jiang, X.; Yin, D.; Zhang, W. Preparation of a hydrogel-based adsorbent for metal ions through high internal phase emulsion polymerization. *ACS Omega* **2020**, *5*, 19920–19927. [[CrossRef](#)] [[PubMed](#)]
67. Sanyang, M.; Ghani, W.A.W.A.K.; Idris, A.; Bin Ahmad, M. Hydrogel biochar composite for arsenic removal from wastewater. *Desalination Water Treat.* **2016**, *57*, 3674–3688. [[CrossRef](#)]
68. Sun, Y.; Ma, Y.; Fang, G.; Li, S.; Fu, Y. Synthesis of acid hydrolysis lignin-g-poly-(acrylic acid) hydrogel superabsorbent composites and adsorption of lead ions. *BioResources* **2016**, *11*, 5731–5742. [[CrossRef](#)]
69. Karakoyun, N.; Kubilay, S.; Aktas, N.; Turhan, O.; Kasimoglu, M.; Yilmaz, S.; Sahiner, N. Hydrogel–Biochar composites for effective organic contaminant removal from aqueous media. *Desalination* **2011**, *280*, 319–325. [[CrossRef](#)]
70. Zhao, Q.; Wu, Z.; Xu, X.; Yang, R.; Ma, H.; Xu, Q.; Zhang, K.; Zhang, M.; Xu, J.; Lu, B. Design of poly (3, 4-ethylenedioxythiophene): Polystyrene sulfonate-polyacrylamide dual network hydrogel for long-term stable, highly efficient solar steam generation. *Sep. Purif. Technol.* **2022**, *300*, 121889. [[CrossRef](#)]
71. Qiu, X.; Kong, H.; Li, Y.; Wang, Q.; Wang, Y. Interface engineering of a Ti₄O₇ nanofibrous membrane for efficient solar-driven evaporation. *ACS Appl. Mater. Interfaces* **2022**, *14*, 54855–54866. [[CrossRef](#)]
72. Xu, X.; Zhao, Q.; Liu, Q.; Qiu, J.; Li, J.; Zheng, W.; Cao, J.; Wang, L.; Wang, W.; Yuan, S.; et al. Full-spectrum-responsive Ti₄O₇-PVA nanocomposite hydrogel with ultrahigh evaporation rate for efficient solar steam generation. *Desalination* **2024**, *577*, 117400. [[CrossRef](#)]
73. Zhao, Q.; Liu, J.; Wu, Z.; Xu, X.; Ma, H.; Hou, J.; Xu, Q.; Yang, R.; Zhang, K.; Zhang, M.; et al. Robust PEDOT: PSS-based hydrogel for highly efficient interfacial solar water purification. *Chem. Eng. J.* **2022**, *442*, 136284. [[CrossRef](#)]

74. Xu, X.; Liu, Q.; Qiu, J.; Zhao, Q.; Yuan, S.; Li, H.; Li, Z.; Fu, A.; Xu, J.; Lu, B. Photothermal-photocatalytic bifunctional highly porous hydrogel for efficient coherent sewage purification-clean water generation. *Desalination* **2025**, *597*, 118364. [\[CrossRef\]](#)
75. Li, Z.; Qiu, J.; Xu, X.; Wan, R.; Yao, M.; Wang, H.; Zhou, Z.; Xu, J. Solar driven kaolin-based hydrogels for efficient interfacial evaporation and heavy metal ion adsorption from wastewater. *Sep. Purif. Technol.* **2025**, *354*, 129243. [\[CrossRef\]](#)
76. Wang, Z.; Li, Y.; Liu, F.; Shao, H.; Yang, Y.; Wang, L.; Jin, Z.; Li, D.; He, X.; Chen, N. Embedded solar-powered hydrogel evaporator for enhancing uranium extraction from seawater. *Chem. Eng. J.* **2025**, *505*, 159063. [\[CrossRef\]](#)
77. Zhou, D.; Zhang, L.; Guo, S. Mechanisms of lead biosorption on cellulose/chitin beads. *Water Res.* **2005**, *39*, 3755–3762. [\[CrossRef\]](#) [\[PubMed\]](#)
78. Benguella, B.; Benaissa, H. Cadmium removal from aqueous solutions by chitin: Kinetic and equilibrium studies. *Water Res.* **2002**, *36*, 2463–2474. [\[CrossRef\]](#) [\[PubMed\]](#)
79. Barakat, M.; Sahiner, N. Cationic hydrogels for toxic arsenate removal from aqueous environment. *J. Environ. Manag.* **2008**, *88*, 955–961. [\[CrossRef\]](#)
80. Tang, S.C.; Yin, K.; Lo, I.M. Column study of Cr (VI) removal by cationic hydrogel for in-situ remediation of contaminated groundwater and soil. *J. Contam. Hydrol.* **2011**, *125*, 39–46. [\[CrossRef\]](#)
81. Sun, X.; Peng, B.; Ji, Y.; Chen, J.; Li, D. Chitosan (chitin)/cellulose composite biosorbents prepared using ionic liquid for heavy metal ions adsorption. *AIChE J.* **2009**, *55*, 2062–2069. [\[CrossRef\]](#)
82. Gao, H.; Sun, Y.; Zhou, J.; Xu, R.; Duan, H. Mussel-inspired synthesis of polydopamine-functionalized graphene hydrogel as reusable adsorbents for water purification. *ACS Appl. Mater. Interfaces* **2013**, *5*, 425–432. [\[CrossRef\]](#)
83. Ali, A.E.-H.; Shawky, H.; El Rehim, H.A.; Hegazy, E. Synthesis and characterization of PVP/AAC copolymer hydrogel and its applications in the removal of heavy metals from aqueous solution. *Eur. Polym. J.* **2003**, *39*, 2337–2344.
84. Liang, H.; Cao, X.; Zhang, W.; Lin, H.; Zhou, F.; Chen, L.; Yu, S. Robust and highly efficient free-standing carbonaceous nanofiber membranes for water purification. *Adv. Funct. Mater.* **2011**, *21*, 3851–3858. [\[CrossRef\]](#)
85. Liu, Z.; Wang, H.; Liu, C.; Jiang, Y.; Yu, G.; Mu, X.; Wang, X. Magnetic cellulose–chitosan hydrogels prepared from ionic liquids as reusable adsorbent for removal of heavy metal ions. *Chem. Commun.* **2012**, *48*, 7350–7352. [\[CrossRef\]](#)
86. Ozay, O.; Ekici, S.; Baran, Y.; Kubilay, S.; Aktas, N.; Sahiner, N. Utilization of magnetic hydrogels in the separation of toxic metal ions from aqueous environments. *Desalination* **2010**, *260*, 57–64. [\[CrossRef\]](#)
87. Zhang, Z.; Xiao, F.; Guo, Y.; Wang, S.; Liu, Y. One-pot self-assembled three-dimensional TiO₂-graphene hydrogel with improved adsorption capacities and photocatalytic and electrochemical activities. *ACS Appl. Mater. Interfaces* **2013**, *5*, 2227–2233. [\[CrossRef\]](#) [\[PubMed\]](#)
88. Thakur, S.; Govender, P.P.; Mamo, M.A.; Tamulevicius, S.; Mishra, Y.K.; Thakur, V.K. Progress in lignin hydrogels and nanocomposites for water purification: Future perspectives. *Vacuum* **2017**, *146*, 342–355. [\[CrossRef\]](#)
89. Kwak, H.W.; Shin, M.; Yun, H.; Lee, K.H. Preparation of silk sericin/lignin blend beads for the removal of hexavalent chromium ions. *Int. J. Mol. Sci.* **2016**, *17*, 1466. [\[CrossRef\]](#)
90. Zhang, L.; Lu, H.; Yu, J.; Fan, Y.; Yang, Y.; Ma, J.; Wang, Z. Synthesis of lignocellulose-based composite hydrogel as a novel biosorbent for Cu²⁺ removal. *Cellulose* **2018**, *25*, 7315–7328. [\[CrossRef\]](#)
91. Guo, H.; Jiao, T.; Zhang, Q.; Guo, W.; Peng, Q.; Yan, X. Preparation of graphene oxide-based hydrogels as efficient dye adsorbents for wastewater treatment. *Nanoscale Res. Lett.* **2015**, *10*, 1–10. [\[CrossRef\]](#) [\[PubMed\]](#)
92. Paulino, A.T.; Belfiore, L.A.; Kubota, L.T.; Muniz, E.C.; Tambourgi, E.B. Efficiency of hydrogels based on natural polysaccharides in the removal of Cd²⁺ ions from aqueous solutions. *Chem. Eng. J.* **2011**, *168*, 68–76. [\[CrossRef\]](#)
93. Karbassiyazdi, E.; Altaee, A.; Ibrar, I.; Razmjou, A.; Alsaka, L.; Ganbat, N.; Malekizadeh, A.; Ghobadi, R.; Khabbaz, H. Fabrication of carbon-based hydrogel membrane for landfill leachate wastewater treatment. *Desalination* **2023**, *564*, 116783. [\[CrossRef\]](#)
94. Ibrar, I.; Alsaka, L.; Yadav, S.; Altaee, A.; Zhou, J.L.; Shon, H.K. Kappa carrageenan-vanillin composite hydrogel for landfill leachate wastewater treatment. *Desalination* **2023**, *565*, 116826. [\[CrossRef\]](#)
95. Kulal, P.; Badalamoole, V. Hybrid nanocomposite of kappa-carrageenan and magnetite as adsorbent material for water purification. *Int. J. Biol. Macromol.* **2020**, *165*, 542–553. [\[CrossRef\]](#)
96. Vaid, V.; Jindal, R. An efficient pH-responsive kappa-carrageenan/tamarind kernel powder hydrogel for the removal of brilliant green and rose bengal from aqueous solution. *J. Appl. Polym. Sci.* **2022**, *139*, 52218. [\[CrossRef\]](#)
97. Salgueiro, A.M.; Daniel-Da-Silva, A.L.; Girão, A.V.; Pinheiro, P.C.; Trindade, T. Unusual dye adsorption behavior of κ-carrageenan coated superparamagnetic nanoparticles. *Chem. Eng. J.* **2013**, *229*, 276–284. [\[CrossRef\]](#)
98. Yadav, S.; Ibrar, I.; Altaee, A.; Samal, A.K.; Zhou, J. Surface modification of nanofiltration membrane with kappa-carrageenan/graphene oxide for leachate wastewater treatment. *J. Membr. Sci.* **2022**, *659*, 120776. [\[CrossRef\]](#)
99. Patel, P.; Gangwar, P.; Thareja, P. Preparation and rheology of titanium dioxide nanoparticles loaded κ-carrageenan hydrogel beads strengthened by mixed salts for multipollutant water remediation. *J. Vinyl Addit. Technol.* **2023**, *29*, 773–794. [\[CrossRef\]](#)

100. Yan, E.; Cao, M.; Ren, X.; Jiang, J.; An, Q.; Zhang, Z.; Gao, J.; Yang, X.; Zhang, D. Synthesis of Fe₃O₄ nanoparticles functionalized polyvinyl alcohol/chitosan magnetic composite hydrogel as an efficient adsorbent for chromium (VI) removal. *J. Phys. Chem. Solids* **2018**, *121*, 102–109. [[CrossRef](#)]
101. Miao, C.; Huang, W.; Li, K.; Yang, Y. Highly efficient removal of adsorbed cationic dyes by dual-network chitosan-based hydrogel. *Environ. Res.* **2024**, *263*, 120195. [[CrossRef](#)]
102. Aljeboree, A.M.; Alkaim, A.F.; Alsultany, F.H.; Issa, S.K. Highly reusable nano adsorbent based on clay-incorporated hydrogel nanocomposite for cationic dye adsorption. *J. Inorg. Organomet. Polym. Mater.* **2024**, 1–22. [[CrossRef](#)]
103. Long, L.; Hu, X.; Yan, J.; Zeng, Y.; Zhang, J.; Xue, Y. Novel chitosan–ethylene glycol hydrogel for the removal of aqueous perfluorooctanoic acid. *J. Environ. Sci.* **2019**, *84*, 21–28. [[CrossRef](#)]
104. Abdeen, Z.; Mohammad, S.; Mahmoud, M. Adsorption of Mn (II) ion on polyvinyl alcohol/chitosan dry blending from aqueous solution. *Environ. Nanotechnol. Monit. Manag.* **2015**, *3*, 1–9. [[CrossRef](#)]
105. Gibas, I.; Janik, H. Synthetic polymer hydrogels for biomedical applications. *Chem. Chem. Technol.* **2010**, *4*, 297–304. [[CrossRef](#)]
106. Pasqui, D.; De Cagna, M.; Barbucci, R. Polysaccharide-based hydrogels: The key role of water in affecting mechanical properties. *Polymers* **2012**, *4*, 1517–1534. [[CrossRef](#)]
107. Hoffman, A.S. Hydrogels for biomedical applications. *Adv. Drug Deliv. Rev.* **2012**, *64*, 18–23. [[CrossRef](#)]
108. Zhang, Q.; Wang, Z.; Zhang, C.; Aluko, R.E.; Yuan, J.; Ju, X.; He, R. Structural and functional characterization of rice starch-based superabsorbent polymer materials. *Int. J. Biol. Macromol.* **2020**, *153*, 1291–1298. [[CrossRef](#)] [[PubMed](#)]
109. Rodrigues, F.H.; Fajardo, A.R.; Pereira, A.G.; Ricardo, N.M.; Feitosa, J.P.; Muniz, E.C. Chitosan-graft-poly (acrylic acid)/rice husk ash based superabsorbent hydrogel composite: Preparation and characterization. *J. Polym. Res.* **2012**, *19*, 1–10. [[CrossRef](#)]
110. Alam, M.N.; Christopher, L. Natural cellulose-chitosan cross-linked superabsorbent hydrogels with superior swelling properties. *ACS Sustain. Chem. Eng.* **2018**, *6*, 8736–8742. [[CrossRef](#)]
111. Alam, M.N.; Islam, M.S.; Christopher, L. Sustainable production of cellulose-based hydrogels with superb absorbing potential in physiological saline. *ACS Omega* **2019**, *4*, 9419–9426. [[CrossRef](#)]

Disclaimer/Publisher’s Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.