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Radiation-Crosslinked Hydrogels: Material Systems, Functional Applications and Industrialization Challenges

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Abstract. Radiation crosslinking is a green and efficient hydrogel fabrication technique that relies on high-energy rays to induce polymer crosslinking without chemical initiators or crosslinkers. It features sterilization capability, uniform reaction, controllable network structure and no reagent residue, effectively overcoming the limitations of traditional chemical and thermal preparation methods. This review systematically summarizes the basic principles of radiation crosslinking, the characteristics of different radiation sources, and the research progress of synthetic polymer, natural polymer and composite radiation hydrogels. The typical applications of radiation hydrogels in biomedical repair, drug delivery, environmental water treatment, agricultural water retention and flexible intelligent devices are introduced in detail. The current challenges, including uncontrollable crosslinking–degradation competition, radical quenching effects, limited industrial scalability and insufficient standardized evaluation systems, are analyzed. Finally, future development directions focusing on mechanical enhancement, intelligent functionalization, process optimization and mechanistic exploration are proposed. This review aims to provide a systematic reference for the design and application of high-performance radiation-crosslinked hydrogels.

Keywords: radiation crosslinking, hydrogel, polymer, functional materials, biomedical application

1. Introduction

Hydrogels are three-dimensional porous networks of hydrophilic polymer chains. They retain water and exhibit flexible properties similar to biological soft tissues while maintaining a solid shape. High water content, biocompatibility, tunable porosity and reversible swelling–deswelling behavior support applications in biomedicine[1-3], energy sensing [4, 5], catalysis and environmental remediation [6, 7].

Hydrogels are mainly formed by chemical or physical crosslinking. Physical crosslinking uses hydrogen bonds, hydrophobic interactions, ionic coordination and chain entanglement under mild conditions without toxic crosslinkers. These crosslinking points are often weak and reversible, which limits gel strength and stability. Chemical crosslinking connects chains through covalent bonds for stable, strong networks, but often requires initiators, crosslinkers or organic solvents with

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potential residue and biotoxicity risks. Radiation crosslinking employs γ-rays or electron beams to irradiate aqueous polymer solutions or solid matrices. Radicals generated, transferred and recombined on molecular chains initiate polymerization and crosslinking in situ.

Radiation preparation can proceed without additional chemical initiators or crosslinkers, avoiding reagent residues and related safety and environmental concerns. Irradiation can sterilize products simultaneously with gel formation and simplify processing. Dose, dose rate, irradiation time and atmosphere regulate crosslinking density, pore size and mechanical properties, making radiation crosslinking a green, efficient and scalable preparation route [6, 8].

Research has expanded from the crosslinking of single polymers to multicomponent gels and functional applications. This review outlines reaction mechanisms, influencing factors, polymer systems and gelation characteristics, followed by biomedical, energy-sensing, catalytic and environmental applications. It concludes with bottlenecks in performance regulation, irradiation uniformity, continuous production and long-term service stability, and future development trends.

2. Fundamental mechanism and radiation sources for radiation-fabricated hydrogels

2.1. Fundamental reaction mechanism

In figure 1, high-energy irradiation of aqueous polymer solutions induces water radiolysis, generating hydroxyl radicals, hydrated electrons and hydrogen radicals. These reactive species produce macromolecular radicals through hydrogen abstraction. Coupling between different polymer chains induces intermolecular crosslinking, forming three-dimensional hydrogel networks. Intramolecular crosslinking alters chain conformation without forming a continuous macroscopic network. Irradiation may also cleave main chains and induce chain scission, damaging networks and mechanical properties.

Crosslinking–degradation competition governs gel fraction, crosslinking density, swelling ratio and mechanical properties. Dose, dose rate and polymer concentration regulate this process. Crosslinking dominates at low doses, increasing gel fraction; above a critical dose, degradation becomes predominant and weakens networks. High dose rates rapidly generate radicals and facilitate intermolecular crosslinking. Low polymer concentrations increase chain separation, reducing intermolecular collisions and favoring intramolecular crosslinking or radical quenching over stable hydrogel formation.

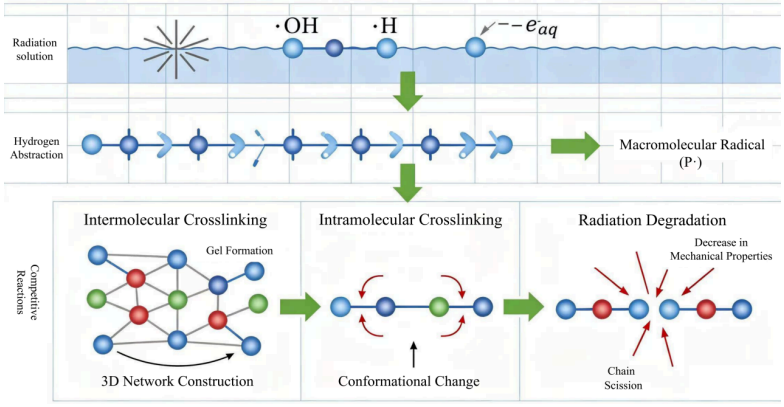


Figure 1. Reaction mechanism of hydrogel preparation by high-energy radiation

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2.2. Comparison of different radiation sources

Various radiation sources differ significantly in energy, penetration capacity and dose rate, which directly govern hydrogel processing conditions, allowable sample thicknesses and final material performance.Table 1 compares gamma rays and electron beam (EB, Electron Beam).

2.2.1. Gamma rays (^{60}Co radiation source)

Gamma rays are ionizing radiation with strong penetration. Uniform radiation crosslinking can be achieved for thick bulk samples and hermetically packaged samples. Hydrogel fabrication can be accomplished without chemical initiators or crosslinkers, and sterilization is realized simultaneously during irradiation. Gamma rays are highly suitable for bulk hydrogels for biomedical applications [8, 9]. The main limitations are a low dose rate and long irradiation times. Some polymers are more prone to main-chain degradation under prolonged gamma irradiation, damaging the hydrogel network.

2.2.2. Electron beam (EB)

Electron beams are a form of ionizing radiation generated by accelerators. Electron-beam irradiation has a higher dose rate than gamma rays, leading to fast crosslinking reactions and high processing efficiency for continuous assembly-line production. High-energy electrons can directly act on polymer chains to generate radicals with lower dependence on water. Nevertheless, the penetration depth of electron beams is limited, making them particularly suited to thin-film and thin-layer hydrogel samples. Uneven dose distribution inside thick samples results in insufficient crosslinking in deep layers. The accelerator equipment can be started and stopped, and there is no residual radioactive waste.

2.2.3. Ultraviolet (UV) radiation

Table 1. Comparison of characteristics and application performances of common radiationsources for hydrogel preparation

Radiation type	Dose rate characteristics	Penetration depth	Crosslinking uniformity	Processing efficiency	Equipment and cost	Application scenarios	Main drawbacks
Gamma rays	Low (2-7 kGy/h)	Extremely strong, suitable for thick bulk and large-volume samples	Uniform dose throughout the sample, good consistency of internal and external crosslinking	Low, long irradiation period	Scarce equipment, high protection requirements and high operating and maintenance costs	Large bulk hydrogels, scenarios requiring uniform bulk crosslinking, biomedical applications with in situ sterilization	Long processing cycle; prone to induce chain degradation of some natural polymers; limited batch capacity
Electron beam (EB)	Extremely high (200-300 kGy/h)	Limited, particularly suitable for thin-film and thin-layer samples	Uniform within thin layers; over-crosslinking on the surface of thick samples while insufficient crosslinking in the interior	Extremely high, enabling rapid continuous production	Widely available equipment, high controllability, good adaptability for industrialization	Thin film dressings, thin functional hydrogels, continuous mass fabrication	Uneven crosslinking for thick samples, obvious size effect

UV is non-ionizing radiation and generally cannot directly excite polymers to generate radicals. Most systems require photoinitiators that absorb UV energy and generate radicals for crosslinking. Low equipment costs and selective curing support microstructured and 3D-printable hydrogels. Weak penetration limits curing to thin transparent samples. Photoinitiator residues and

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decomposition products may be cytotoxic, limiting biomedical applications. UV therefore differs from initiator-free ionizing-radiation routes using gamma rays or electron beams.

2.3. Main preparation processes of radiation-fabricated hydrogels

Based on the precursors and the state of the reaction system, radiation-fabricated hydrogels are prepared through five main processes.

(1) Radiation crosslinking of aqueous polymer solutions: The prepared aqueous polymer solution is directly irradiated. Crosslinking occurs between polymer chains to form hydrogels. This simple process is the most common preparation method [10, 11].

(2) Radiation polymerization and crosslinking of monomers: Irradiation triggers polymerization of small-molecule monomers. Crosslinking between chain segments takes place simultaneously with polymerization, completing polymer synthesis and hydrogel formation in one step [6, 12, 13].

(3) Radiation graft copolymerization: Active free radicals are generated on the substrate polymer chains via irradiation to initiate graft polymerization of a second monomer component. This modifies the hydrogel at the molecular level to tune functions such as hydrophilicity/hydrophobicity and biological activity [14, 15].

(4) Solid-state irradiation: Radiation crosslinking is carried out in solid polymer films with no or low water content. Hydrogels are obtained after subsequent water absorption and swelling, which is suitable for preparing high-strength thin-film hydrogels.

(5) Blending and composite irradiation: Two or more polymers and nanofillers are solution-blended followed by simultaneous radiation crosslinking to obtain multicomponent composite hydrogels with improved mechanical properties, electrical conductivity, catalytic performance and other functionalities[7-9, 16-18].

3. Research progress of material systems for radiation-fabricated hydrogels

Synthetic polymer systems exhibit excellent irradiation controllability and process repeatability. By adjusting polymer concentration and irradiation parameters, the gel fraction, swelling behavior and mechanical properties of hydrogels can be regulated. Figure 2 shows the classification of radiation-crosslinked hydrogels on the basis of their matrix composition.

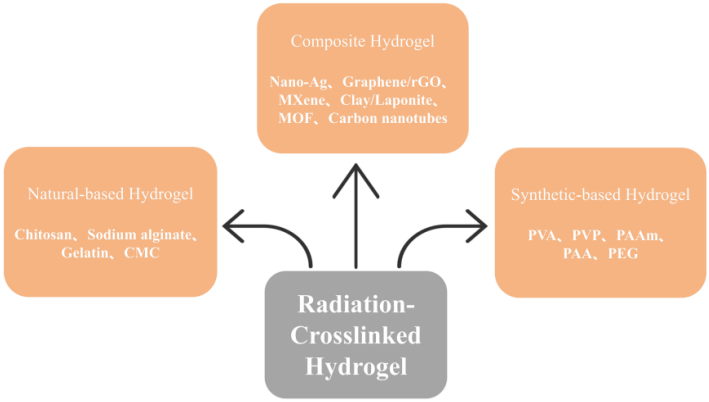


Figure 2. Classification of radiation-crosslinked hydrogels according to matrix composition

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3.1. Synthetic polymer-based radiation hydrogels

Synthetic polymers have stable molecular chain structures, tunable molecular weights and wide irradiation processing windows, making them among the earliest widely used substrates for radiation-crosslinked hydrogel fabrication. Common systems include poly(vinyl alcohol) (PVA), poly(vinylpyrrolidone) (PVP), polyacrylamide (PAAm), poly(acrylic acid) (PAA), and poly(ethylene glycol) (PEG). Coupling between macromolecular radicals forms a three-dimensional crosslinked hydrogel network.

PVA contains abundant hydroxyl groups and produces hydrophilic, biocompatible and wear-resistant hydrogels. Irradiation dose and initial concentration determine gel fraction and crosslinking density. Increasing dose improves crosslinking and tensile strength and decreases equilibrium swelling; excessive doses cause chain scission, defects and mechanical deterioration. γ-irradiated PVA hydrogels support sandy-soil water retention and urea loading, with irradiation conditions regulating swelling, retention, morphology and crystallinity [19]. In PVA/ZrO_2:Eu^{3+} composites, dose alters filler crystal phase structure and PVA crosslinking and optical properties [20].

PVP offers biocompatibility, water solubility and film-forming properties, making it useful for hydrogels used as dressings and drug carriers. Pure PVP has relatively low irradiation crosslinking efficiency, and blending improves gel fraction and stability. γ-copolymerized PVP/sodium alginate/silver nanoparticle hydrogels load and release prednisolone [21]. γ-crosslinked PVP-PEG-$BaTiO_3$ hydrogels have optimized networks and exhibit dielectric and piezoelectric responses [22].

PAAm and PAA contain polar amide and carboxyl groups, respectively, enabling high swelling and adsorption of heavy metals and dyes. PAA has pH-responsive swelling. γ-irradiated sodium allylsulfonate modified starch-acrylic acid hydrogels achieve high water absorption and maintain swelling under alkaline conditions [12]. γ-synthesized HA/CS/PAAc hydrogels are formed through acrylic acid polymerization and crosslinking and exhibit high water content and controlled drug release [13]. γ-crosslinked GO/cellulose nanofiber/PAA patches load chromogenic reagents for non-invasive colorimetric detection of sweat urea [23].

Poly(ethylene glycol) (PEG) resists protein adsorption and is biologically inert. PEG-based radiation hydrogels have low immunogenicity and are highly suitable for biomedical applications and cell culture scaffolds. However, hydrogels obtained by radiation crosslinking of pure PEG have poor mechanical properties, and modifications such as blending and grafting are generally required to improve mechanical performance.

Most pure synthetic polymers are poorly biodegradable under natural conditions, complicating the disposal of single-use medical materials and environmental adsorbents. Conductive hydrogels based on acrylamide-aniline grafted gum ghatti were prepared by γ-ray irradiation to construct semi-interpenetrating/interpenetrating networks. Soil burial experiments demonstrate a certain degree of biodegradation, providing a strategy to address poor degradability [24].

3.2. Natural polymer-based radiation hydrogels

Natural polymers are mainly derived from animals and plants. Representative materials include chitosan, sodium alginate, gelatin and carboxymethyl cellulose. Such polymers offer excellent biocompatibility, biodegradability and low toxicity, together with wide availability, low cost and environmentally friendly characteristics. Most polysaccharide-based natural polymer chains are prone to preferential chain scission and degradation under irradiation, whereas crosslinking is less

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favored. It is difficult to form stable three-dimensional crosslinked hydrogel networks merely by irradiation.

Chitosan has amino and hydroxyl groups and bioactivity for dressings, antibacterial materials and scaffolds, but irradiation alone mainly causes degradation. High-dose irradiation rapidly reduces sodium alginate molecular weight, preventing high-strength gels. Gelatin irradiation induces peptide cleavage; excessive doses cause denaturation and structural damage. Cellulose derivatives such as carboxymethyl cellulose also tend to degrade and are difficult to gel by irradiation alone.

To address the susceptibility of natural polymers to degradation and their low crosslinking efficiency under irradiation, two main modification strategies are used: blending and radiation grafting.

Blending natural polymers with radiation-resistant synthetic polymers produces crosslinked skeletons that improve gel formation while retaining bioactivity. γ-crosslinked chitosan/polyacrylamide hydrogels form precursors for carbon nanotube aerogels for oil-water pollutant removal, with PAM providing the skeleton [25]. γ-irradiated PVA/chitosan and PVA/carboxymethyl cellulose hydrogels retain water and slowly release urea [19]. Acrylic acid/plasticized starch/montmorillonite/chitosan radiation copolymerization combines bioactivity and gel formation for dressings [26]. γ-copolymerized PVP/sodium alginate incorporates alginate into swelling drug-carrier networks [21].

Radiation grafting attaches chains derived from polymerizable monomers such as acrylamide and acrylic acid to natural polymer backbones. Irradiation-induced crosslinking forms bridges that suppress excessive backbone degradation and increase crosslinking efficiency and gel fraction. One-step γ-grafting of acrylic acid onto potato soluble starch without added initiators or crosslinkers yields hydrogels for desert agriculture, integrating water retention, fertilizer control and metal-ion adsorption [6]. γ-grafted sodium allylsulfonate-starch/acrylic acid networks provide water absorption and salt-resistant swelling [12].

Natural polymer-based radiation hydrogels modified by blending and grafting retain the merits of natural materials including biodegradability and biocompatibility, while overcoming the drawbacks of difficult irradiation molding and poor mechanical performance. These strategies provide a feasible route for developing green biomedical and eco-friendly hydrogels.

3.3. Composite/functionalized radiation hydrogels

Single-component polymer hydrogels rarely meet multiple requirements simultaneously, such as mechanical strength, electrical conductivity, adsorption capacity and stimulus responsiveness. Researchers incorporate inorganic nanofillers, conductive components and functional nanoparticles into polymer matrices to fabricate organic-inorganic composite radiation hydrogels. Common inorganic fillers include silver nanoparticles, graphene, clay and MOFs (metal-organic frameworks).

Incorporating silver nanoparticles can give hydrogels broad-spectrum antibacterial properties, making them useful for wound dressings. Graphene has excellent electrical conductivity and mechanical reinforcing effects; a small amount of doping can markedly improve the tensile strength and conductivity of hydrogels. Clay nanosheets can toughen gel networks via non-covalent interactions, build energy dissipation pathways and enhance the toughness of hydrogels. MOF materials have abundant pore structures and numerous active sites. After doping, they can greatly increase the adsorption capacity of hydrogels toward heavy metal ions and organic pollutants.

γ-irradiated graphene/CuS/PAM hydrogels combine photothermal effects and optimized microchannels for solar evaporation, water purification and heat reuse [16]. Carbon nanotube/acrylamide-acrylic acid hydrogels integrate photothermal desalination, power generation,

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sensing and flame retardancy [17]. In γ-irradiated PAAK/carbon nanotube electrolytes, covalent ether bonds and water-state regulation enable high elongation, ionic conductivity and antifreeze performance at −40 °C for flexible zinc-air batteries [8]. PAM-GM hydrogels containing rGO/MXene, synthesized by one-step γ-irradiation, combine solar utilization, evaporation and photothermal catalytic pollutant degradation [7]. PAM/graphene hydrogels containing radiation-generated silver combine antibacterial and photothermal functions for desalination and purification [18]. Similarly, one-pot γ-synthesized PAM-GO-Ag provides catalytic pollutant reduction, antibacterial activity and convenient recycling [9]. MXene/γ-Al_2O_3 self-healing hydrogels with FeNiMn-MOF networks improve specific capacitance and cycling stability in asymmetric hybrid supercapacitors [27].

However, inorganic nanofillers generally exhibit radical quenching effects during radiation preparation. The filler surface consumes many macromolecular radicals generated by irradiation and inhibits polymer crosslinking reactions, resulting in decreased gel fraction and reduced crosslinking degree. At present, surface modification of fillers, control of filler loading and optimization of pre-dispersion processes are commonly adopted to mitigate the negative effects of quenching.

Dispersing conductive fillers into aqueous polymer solutions before radiation crosslinking yields flexible conductive hydrogels for strain and pressure sensors and energy-storage electrodes. γ-irradiated PAAK/carbon nanotube electrolytes combine high ionic conductivity and large deformation for flexible zinc-air devices [8]. γ-irradiated gum ghatti/acrylamide-aniline hydrogels use polyaniline interpenetrating networks for conductivity and biodegradability [24].

Radiation grafting introduces pH- or temperature-sensitive groups. pH-responsive swelling supports controlled drug release. Thermo-responsive hydrogels with a lower critical solution temperature (LCST) undergo temperature-triggered volume changes suitable for smart actuators and controlled separation. γ-irradiated laponite-PNIPAM hydrogels use clay nanosheets for self-healing thermo-responsive gels without extra crosslinkers or initiators [11]. Two-step γ-grafting of acrylic acid onto PEO forms pH-sensitive oral insulin carriers that protect insulin in gastric acid and release it through intestinal swelling [28].

Apart from bulk macroscopic hydrogels, intramolecularly crosslinked nanogels are also an important research direction of functional radiation hydrogels. Irradiation is performed in highly dilute polymer solutions. Radicals preferentially undergo intramolecular crosslinking within a single polymer chain rather than intermolecular crosslinking, yielding crosslinked microspheres at the nanoscale, namely nanogels. Nanogels have small particle sizes and large specific surface areas, showing unique advantages in tumor-targeted drug delivery, protein delivery and biological diagnosis and treatment. Radiation preparation of nanogels requires no additional chemical crosslinkers and produces high-purity products, which is a green and feasible fabrication route.

Composite and functionalized hydrogels overcome single-component limitations through synergistic reinforcement, antibacterial activity, conductivity, responsiveness and adsorption. Uneven filler dispersion, phase separation, radical quenching and limited composite-network controllability remain challenges. The characteristics, irradiation behaviors, merits, drawbacks and corresponding modification strategies of radiation-crosslinked hydrogels based on different matrix materials are summarized in Table 2.

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Table 2. Summary of material characteristics and modification strategies for radiationcrosslinked hydrogels

Material category	Typical substrates	Irradiation reaction characteristics	Advantages	Main drawbacks	Common modification methods
Synthetic polymers	PVA, PVP, PAAm, PAA, PEG	Crosslinking dominates; wide crosslinking–degradation window and favorable process stability	High gel fraction, tunable mechanical properties, good repeatability	Most are not naturally biodegradable with limited bioactivity	Blending, freeze-irradiation cycle, grafting of functional groups
Natural polysaccharide polymers	Chitosan, sodium alginate, carboxymethyl cellulose	Molecular chains preferentially degrade under high-energy irradiation; hydrogels are hardly formed by irradiation alone	Excellent biocompatibility, biodegradable, green and low-cost raw materials	Low crosslinking efficiency and poor gel strength for pure systems	Blending with synthetic polymers, radiation grafting of vinyl monomers
Protein-based natural polymers	Gelatin, collagen	High-dose irradiation easily induces peptide chain cleavage and protein denaturation	Good cell affinity, suitable for soft tissue repair	Low radiation tolerance and narrow processing window	Low-dose irradiation, dual-network co-crosslinking
Inorganic–organic composite systems	Polymers + nano-Ag/graphene/clay/MOF	Fillers tend to trigger radical quenching effect and reduce crosslinking efficiency	Mechanical reinforcement, antibacterial property, conductivity, high adsorption capacity, stimulus responsiveness	Filler agglomeration, poor interfacial compatibility, decreased crosslinking degree	Surface modification of fillers, optimization of filler loading, ultrasonic pre-dispersion

4. Research progress on applications of radiation-fabricated hydrogels

Radiation-fabricated hydrogels can be formed in one step without initiators, crosslinkers or high-temperature conditions. The pore structure, swelling behavior, mechanical strength and distribution of functional groups of hydrogels can be precisely regulated. Radiation-fabricated hydrogels exhibit application potential in multiple fields such as biomedicine, environmental water treatment, agricultural water retention and flexible intelligent devices.

4.1. Biomedical field

Biomedicine represents the most mature and highly industrialized application area for radiation-fabricated hydrogels. Hydrogels prepared by traditional chemical methods usually contain residual initiators and crosslinkers, which easily induce cytotoxicity and inflammatory responses. High-energy irradiation can realize material sterilization simultaneously, satisfying the core requirements of biomedical materials for high biocompatibility, safety and sterility.

Radiation-crosslinked dressings have 3D porosity and high water content, which can create a constant-temperature, constant-humidity and hypoxic wound microenvironment. They absorb exudate, isolate bacterial contamination, relieve pain, prevent scab cracking and accelerate epidermal regeneration. Dose-controlled crosslinking provides hemostatic, antibacterial and adhesive properties for superficial injuries, deep burns and chronic wounds, overcoming gauze limitations in permeability, adhesion and healing cycles.

HA/CS/PAAc hydrogels have high water content and cytocompatibility, with in vitro keratinocyte viability above 82%, indicating potential as artificial skin substitutes [13]. γ-crosslinked acrylic acid/plasticized starch-montmorillonite-chitosan dressings with model drug sulfanilamide accelerated rat wound healing [26]. Radiation-fabricated PAM-GO-Ag hydrogels combine broad-spectrum antibacterial activity with convenient wound attachment and removal, showing potential

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for infected wounds [9]. Graphene/silver hydrogels achieve a 24 h antibacterial rate of 99% against Escherichia coli and have potential for medical dressings as well as water purification [18].

The 3D crosslinked networks of radiation-fabricated hydrogels possess tunable pore sizes and swelling-responsive properties, enabling encapsulation and controlled release of small-molecule drugs, protein pharmaceuticals and antibacterial factors. Tuning irradiation parameters can precisely adjust the density of hydrogel networks to realize long-term sustained release, targeted release and pulsatile drug release. They are widely used in topical dermatological medication, local anti-inflammation and targeted anti-tumor therapy.

Insulin is loaded into freeze-dried, two-step γ-grafted PEO-g-PAAc hydrogels, which protect it in gastric acid and release it upon intestinal swelling. Rat intragastric experiments confirmed blood-glucose reduction lasting at least 4 h [28]. γ-copolymerized PVP/sodium alginate/silver nanoparticle hydrogels use prednisolone as a model drug; dose and nanoparticle loading regulate uptake and release kinetics [21]. γ-crosslinked HMA-ATU hydrogels contain thiourea sites for in-situ reduction of Co, Ni and Cu nanoparticles. Beyond catalysis, these coordination-rich hydrogels offer strategies for metal-based drug carriers [29].

Radiation-crosslinked hydrogels with biocompatibility and biomimetic extracellular matrix structure can serve as 3D scaffold materials for bone, skin and cartilage tissue engineering. HA/CS/PAAc hydrogels crosslinked by γ-ray irradiation contain hyaluronic acid and chondroitin sulfate, key components of the extracellular matrix. Their high water content and tunable swelling characteristics match the requirements of skin tissue engineering scaffolds [13].

Furthermore, with the development of flexible bioelectronics, radiation-fabricated conductive hydrogels and stimuli-responsive hydrogels have become active research areas. By combining conductive polymers, MXene, graphene and other functional fillers through radiation processing, sensing hydrogels integrating flexibility, conductivity and biocompatibility can be fabricated for real-time monitoring of human physiological signals. Meanwhile, radiation-synthesized nanogels feature small size, strong permeability and high drug loading efficiency, demonstrating broad prospects in tumor-targeted delivery and precision biomedical diagnosis and treatment. GO/cellulose nanofiber/PAA composite hydrogel sensing patches were prepared by γ-ray crosslinking. The hydrogel network can immobilize enzymes and chromogenic reagents for non-invasive colorimetric detection of urea in sweat. The hydrogel can also act as an LDI-MS matrix for urea quantification, providing medical sensing materials for non-invasive home diagnosis of chronic kidney disease [23]. Laponite-PNIPAM self-healing thermo-responsive hydrogels were fabricated by γ-ray irradiation without additional chemical crosslinkers. The uniform network combines thermally triggered volume phase transition and self-healing capacity, making these hydrogels promising for wearable biosensors and intelligent drug release devices [11].

4.2. Environmental water treatment field

Water pollution remediation and water-saving agriculture constitute important application scenarios of radiation-fabricated functional hydrogels. Radiation grafting modification can precisely introduce carboxyl, amino, hydroxyl, sulfonic and other active adsorption groups onto polymer chains, producing functional hydrogel adsorbents with high adsorption performance and recyclability.

Radiation hydrogels have 3D porosity and active sites for adsorption of Pb, Cd, Cu and Cr ions and azo and aromatic dyes. Compared with chemical modification, radiation grafting proceeds more uniformly and efficiently and can enhance capacity and selectivity without secondary preparation pollution. Desorption allows repeated reuse, lowering costs for industrial wastewater, domestic sewage and heavy-metal-contaminated water treatment.

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PAM-GO-Ag catalyzes methylene blue and rhodamine B reduction, provides antibacterial activity and is recoverable for reuse [9]. rGO/MXene hydrogels integrate solar evaporation and photothermal catalysis, achieving an evaporation rate of 4.671 $kg \cdot m^{-2} \cdot h^{-1}$ under one-sun illumination. Methylene blue and rhodamine B degradation rates are 91.5% and 86.3%, respectively, with ion removal up to 99% [7]. Hydrogels with graphene@CuS microchannels achieve an evaporation rate of 8.1 $kg \cdot m^{-2} \cdot h^{-1}$ and provide photocatalytic methylene blue degradation, salt resistance and anti-crystallization performance [16]. Graphene/silver evaporators desalinate water; silver inhibits microbial growth and improves long-term stability [18]. Carbon nanotube aerogels derived from γ-crosslinked chitosan-polyacrylamide have hydrophobic pores for pump-oil and organic-solvent adsorption [25]. Radiation-grafted dodecyl methacrylate/melamine sponges reach a 154° water contact angle, absorb 66–168 times their mass in oils and continuously separate floating oil [14]. Radiation grafting and in-situ reduction produce silver nanocluster composite membranes. PAA chains on PE membranes immobilize silver nanoclusters whose fluorescence is specifically quenched by Cr^{3+}, enabling their use in visual water-screening strips [15].

In agricultural applications, radiation-crosslinked superabsorbent water-retention hydrogels can absorb, store and retain hundreds of times their own mass of water. When incorporated into soil, they can improve soil pore structure, reduce soil water evaporation and deep percolation, and enhance soil water and fertilizer retention capacity. These hydrogels can slowly release water and nutrients to support crop growth.

One-step γ-synthesized Urea/PSS-g-PAA from potato soluble starch-acrylic acid without added crosslinkers or initiators combines water retention, salt resistance, controlled urea release and Cu^{2+} adsorption, promoting maize growth in drought pot experiments [6]. SAS5wt%-SBSH reaches a pure-water absorption capacity of 951.17 g/g and maintains swelling in saline and alkaline solutions [12]. γ-irradiated PVA/chitosan and PVA/carboxymethyl cellulose load and slowly release urea; comparative swelling and retention tests provide strategies for sandy-soil water and fertilizer regulation [19].

4.3. Other intelligent functional application fields

Apart from biomedicine and environmental remediation, radiation-synthesized smart hydrogels have undergone rapid development in flexible electronics, smart responsive materials and energy storage devices.

Radiation-crosslinked conductive composites combine high flexibility, sensitivity and fatigue resistance. Flexible pressure, strain and human-machine interaction sensors support wearable electronics and motion monitoring.

Radiation-polymerized acrylamide-acrylic acid hydrogels with multi-walled carbon nanotubes and ionic liquids integrate photothermal desalination, power generation, sensing and flame retardancy [17]. γ-irradiated PAAK/CNT hydrogels achieve 328.54 $mS \cdot cm^{-1}$ ionic conductivity, tensile elongation at break over 3022.7% and stable performance at −40 °C. Assembled flexible zinc-air batteries provide high specific capacity and stable output under deformation [8]. γ-crosslinked PVP-PEG-$BaTiO_3$ hydrogels exhibit piezoelectric and dielectric responses for biomimetic flexible sensors [22]. γ-irradiated gum ghatti/acrylamide-aniline forms semi- and full-interpenetrating conductive networks, with biodegradability verified by soil burial [24]. MXene/γ-Al_2O_3 self-healing hydrogels with FeNiMn-MOF electrodes form asymmetric hybrid supercapacitors, delivering 435.71 $F \cdot g^{-1}$ at 1 $A \cdot g^{-1}$ and retaining 90.40% capacitance after 10000 charge-discharge cycles [27].

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Radiation grafting produces thermo-, pH-, ion- and photo-responsive hydrogels with reversible swelling, shrinkage and deformation. γ-irradiated laponite-PNIPAM forms self-healing thermo-responsive networks for actuators and controlled release [11]. Methacrylated POSS-crosslinked PNIPAM hydrogels synthesized by one-step γ-irradiation use POSS as an inorganic crosslinker to regulate pore size and thermal response, with controlled water absorption-release for potential smart separation [30].

Radiation hydrogels offer green fabrication, controllable structure, tunable performance and biosafety. Continued development of processing and functional modification will expand applications and strengthen their industrial potential in biomedicine, environmental remediation and flexible electronics. Figure 3 schematically depicts the material systems, key properties and multi-field applications of radiation-crosslinked hydrogels with typical application examples.

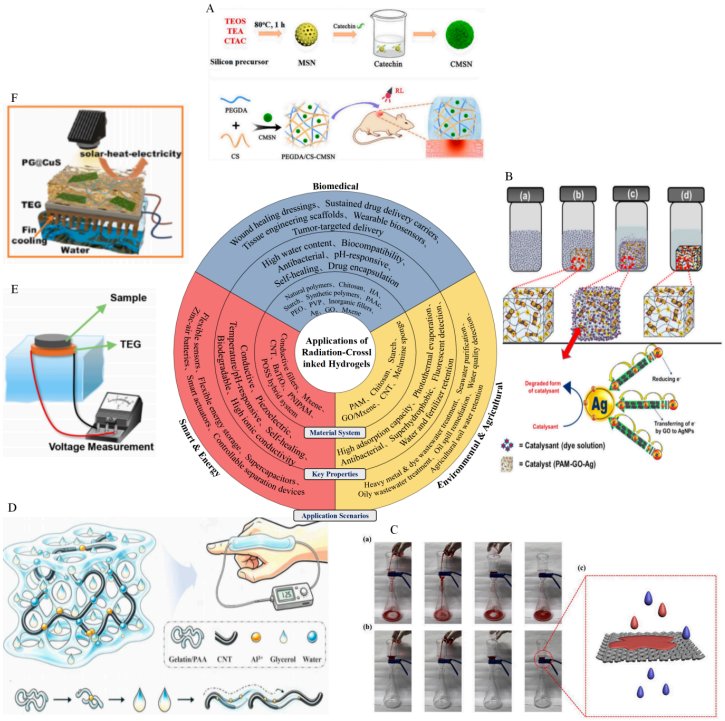


Figure 3. Schematic illustration of the material systems, key properties and multi-field applicationsof radiation-crosslinked hydrogels, together with representative application examples. Panel Aadapted from[31]; Panel B adapted from[9]; Panel C adapted from[32]; Panel D adapted from[33]; Panel E adapted from[17]; Panel F adapted from[16]

5. Challenges and limitations

Bottlenecks remain in performance regulation, large-scale fabrication, composite construction, as well as clinical and industrial translation. Key challenges concern intrinsic material properties, preparation technologies, composite systems and application translation.

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5.1. Poor radiation stability of polymer substrates: dilemma between cross-linking and degradation

Natural polysaccharides and proteins are biocompatible but radiation-sensitive. For most natural polymers, irradiation tends to induce chain scission, lowering molecular weight and loosening networks. Insufficient doses produce inadequate radicals, limited cross-linking and low gel fractions. Excessive doses favor degradation, uncontrolled swelling and instability, making cross-linking–degradation balance a core challenge.

Blending or grafting with radiation-resistant synthetic polymers suppresses natural-polymer degradation [6, 19, 25, 26]. One-step acrylic-acid grafting builds starch networks while retaining biodegradability and water retention [6]. γ-irradiated PAM forms a skeleton encapsulating chitosan, balancing moldability and bioactivity [25]. Modified systems still have narrow dose windows: insufficient doses reduce grafting and gel fraction; excessive irradiation causes backbone scission and impairs swelling behavior and mechanical properties [26].

Furthermore, hydrogels with a single network formed solely by radiation cross-linking generally show unsatisfactory mechanical performance. Lacking effective energy-dissipation pathways and suffering from network heterogeneity, such hydrogels tend to fracture, collapse and fatigue under tension, compression or bending. Their inferior strength, toughness and fatigue resistance cannot meet requirements of engineering scenarios such as biomedical load-bearing devices and complex-environment sensors, limiting practical deployment.

Nanofillers such as carbon nanotubes, graphene, MXene and inorganic oxides are incorporated for mechanical reinforcement [7, 8, 17]. PAAK/CNT hydrogels achieve tensile elongation over 3022.7 %, yet performance depends on specific formulations and irradiation parameters. Poor filler dispersion or network defects cause mechanical deterioration under cyclic loading [8]. Long-term fatigue studies under complex service conditions are insufficient.

5.2. Limitations of radiation equipment and processes: barriers against industrial scale-up

Limited access to γ-ray facilities, high operating costs and strict radiation-protection requirements can constrain routine large-scale material processing. Electron-beam irradiation has limited penetration and is particularly suited to thin-film samples. For bulky hydrogels, surface over-cross-linking and insufficient internal cross-linking cause structural inhomogeneity.

Most reported studies use small, thin samples, for which uniform dose fields help maintain stable gel performance[6-8, 16, 18]. Equipment constraints limit reproduction of laboratory conditions for thick dressings and bulk adsorptive hydrogels. The limited number of studies on solid-state irradiation of carbon-based composite hydrogels indicates that this approach remains underdeveloped.

Large-scale industrial systems face uneven dose fields, shielding caused by sample stacking and fluctuations in ambient temperature. These factors alter gel fraction, pore size, mechanical and swelling properties, making it difficult to replicate optimal lab-scale conditions.

Laboratory process optimization studies investigate irradiation dose, monomer concentration and filler content [12, 20, 22], whereas pilot-scale studies are limited. PVA-based composite hydrogels and starch-acrylic acid superabsorbent hydrogels exhibit swelling and water retention at the laboratory scale [12, 19]. After scale-up, uneven internal irradiation dose produces cross-linking-density gradients and batch-to-batch discrepancies. The dose-performance correlation for scale-up remains unclear without mature scaling models.

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5.3. Poor compatibility of multi-component systems: insufficient controllability over cross-linking efficiency and networks

Most inorganic and metallic nanoparticles exert free-radical-scavenging effects. They consume radiation-generated free radicals, inhibit polymer cross-linking and reduce gel yield, producing loose, defective networks and degraded performance.

Carbon nanotubes, graphene, MXene, silver nanoparticles and metal oxides are functional fillers[1-3, 7, 9, 13]. Filler loadings are mostly determined through trial-and-error experiments to mitigate scavenging [2, 9]. Quantitative research on interactions between fillers and free radicals is scarce. Higher filler content improves conductivity, photothermal performance and adsorption capacity, yet it usually decreases gel fraction and cross-linking density.

Agglomeration and phase separation hinder control over pores, cross-linking and functional sites. Fabrication of double networks and interpenetrating polymer networks (IPNs) remains insufficiently controllable, with unclear filler–matrix synergy. rGO/MXene, PAM-GO-Ag and CNT performance relies on dispersion [7, 11, 13]. Irradiation dynamics linking radical evolution, dispersion and network formation are seldom explored, leaving structure–activity relationships empirical. Although gum ghatti-based conductive IPNs have been realized [24], formation conditions and network proportions remain difficult to control.

5.4. Lack of standardization system: difficulties in clinical and industrial transformation

Unified standards for preparation procedures, performance evaluation and quality inspection are absent. Diverse formulations, irradiation parameters and modification approaches lead to inconsistent stability and repeatability, impeding standardized product development.

Irradiation dose, dose rate, atmosphere, polymer concentration and post-treatment protocols vary significantly across studies. Certain chitosan composite dressings are prepared at 15 kGy [26], and HA/CS/PAAc biomedical composite hydrogels achieve high gel fractions at 15 kGy [13]. By contrast, PVP/sodium alginate/silver nanoparticle hydrogels for drug delivery were prepared at 30 kGy [21]. Test conditions for gel fraction, swelling ratio and cytocompatibility are not unified, hindering comparisons and the development of common evaluation criteria.

Clinical translation of biomedical hydrogels requires long cycles and strict approval criteria. Clinical data are required to verify biosafety, long-term histocompatibility and in-vivo degradation toxicity of radiation-processed materials. The costs of irradiation facilities and complex production processes raise prices compared with conventional chemically cross-linked hydrogels and restrict commercialization of dressings and tissue-engineering scaffolds.

Most biomedical studies use cell assays or small-animal tests [13, 26, 28]. Data on large animals, long-term in vivo compatibility, degradation metabolism and toxicology are lacking. HA/CS/PAAc has been evaluated using in vitro viability tests [13], PEO-g-PAAc was evaluated in rat intragastric experiments [28], and chitosan dressings were evaluated in short-term rat wound-healing tests [26]. Environmental studies mainly use simulated wastewater or seawater [1, 7, 13], with insufficient pilot tests using real water and soil or assessing long-term cyclic stability.

Future efforts should optimize substrate modification, upgrade irradiation processes, improve the precision of composite-network design and advance industrial processing technologies for high performance, multi-functionality, standardization and industrialization.

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6. Conclusion and future outlook

Radiation cross-linking provides initiator- and cross-linker-free hydrogels for biomedical and environmental applications, with sterilization possible under appropriate conditions. Cross-linking–scission competition remains difficult to control, especially in natural polymers. Double networks and nanofillers can improve mechanical properties and functionality but may alter radical reactions and network formation. Electron penetration and nonuniform doses constrain scale-up. Future studies could combine controlled irradiation, reinforcement and functional modification with application-specific stability and performance evaluation. Electron paramagnetic resonance spectroscopy, gel-fraction analysis and kinetic modeling may clarify radical evolution and cross-linking–degradation competition. Practical implementation will depend on improved material performance, reproducible processing, quality control and application-specific safety validation.

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