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Smart and Self-Healing Hydrogels: Design Principles, Mechanisms and Biomedical Applications

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Abstract : *Hydrogels are three-dimensional, water-swollen polymer networks that resemble living tissue and are central materials in drug delivery, wound care, tissue engineering and regenerative medicine. Conventional hydrogels, however, are often fragile and, once damaged, cannot recover. Two developments address these limits. Smart, or stimuli-responsive, hydrogels change volume, structure or permeability in reaction to temperature, pH, light, ions, enzymes or other cues, allowing controlled and targeted release. Self-healing hydrogels restore their structure and function after damage through reversible covalent and non-covalent bonds, so they last longer, can be injected through a needle and adapt to irregular tissue. This paper describes the classification, preparation and properties of hydrogels, the stimuli to which smart hydrogels respond, and the mechanisms that provide self-healing. It reviews applications in drug delivery, cancer therapy, wound dressings and tissue regeneration, discusses the balance between mechanical strength and healing ability, and examines challenges such as reproducibility, long-term safety, scale-up and clinical translation. Smart and self-healing hydrogels combine adaptability, injectability and durability and have strong potential as next-generation biomaterials..*

Keywords: hydrogels; self-healing; stimuli-responsive; smart hydrogels; injectable hydrogels; dynamic covalent bonds; drug delivery; wound dressing; tissue regeneration.

I. INTRODUCTION

A hydrogel is a network of hydrophilic polymer chains, joined by chemical or physical cross-links, that can absorb and retain very large amounts of water without dissolving. The high water content, soft and elastic consistency and porosity of hydrogels resemble the extracellular matrix of living tissue, which explains their good biocompatibility and their use in biology and medicine [1][2][3]. Their porous structure allows nutrients, oxygen and drugs to diffuse in and out, and their softness limits irritation of surrounding tissue [1][2][4].

Despite these advantages, classical hydrogels have weaknesses. Many are mechanically weak and brittle, so they break under stress such as joint movement or the forces of injection. Once cut or cracked, a conventional chemically cross-linked hydrogel cannot repair itself. Their behaviour is also usually fixed: they swell or release drug according to the environment in a passive way and cannot adjust to changing physiological conditions [4][5][6][7]. These limits led to two lines of innovation, smart hydrogels and self-healing hydrogels, which are now often combined [6][8][9][10].

The aim of this paper is to provide a structured account of smart and self-healing hydrogels, from design principles and mechanisms to biomedical applications and translational challenges. The objectives are to describe the structure, classification, preparation and properties of hydrogels; to explain how stimuli-responsive behaviour is achieved; to describe the covalent and non-covalent mechanisms of self-healing; to review applications in drug delivery, cancer therapy, wound healing and tissue engineering; and to discuss limitations and future directions.

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II. CLASSIFICATION, PREPARATION AND PROPERTIES

A. Classification

Hydrogels can be classified in several ways, and a clear understanding of the categories helps to place smart and self-healing systems [1][2][3] (Table I).

TABLE I: CLASSIFICATION OF HYDROGELS

Basis of classification	Categories	Examples or comments
Source of polymer	Natural, synthetic, hybrid	Chitosan, alginate, hyaluronic acid, gelatin; polyethylene glycol, polyvinyl alcohol, poly(N-isopropylacrylamide); hybrids combine both
Cross-linking	Physical (non-covalent), chemical (covalent), dynamic	Physical gels are reversible; chemical gels are strong but static; dynamic gels use reversible bonds
Response to environment	Conventional, stimuli-responsive (smart)	Smart hydrogels change with temperature, pH, light and other triggers
Network structure	Homopolymer, copolymer, interpenetrating, composite	Interpenetrating and composite networks improve strength
Physical form	Bulk, film, microgel, nanogel, injectable	Injectable and nanogel forms suit minimally invasive delivery
Charge	Non-ionic, ionic, amphoteric	Charged gels respond to pH and ionic strength
Degradability	Stable, biodegradable	Degradable gels suit temporary scaffolds and depots

B. Preparation and Cross-Linking

Hydrogels are prepared by polymerising monomers in the presence of a cross-linker, by cross-linking existing polymers, or by letting polymers self-assemble through physical interactions [1][3]. Chemical cross-linking forms covalent bonds, for instance through radical polymerisation, click chemistry or reaction with aldehyde cross-linkers, and gives stable, strong networks, though residual cross-linker or initiator may be toxic [1][3]. Physical cross-linking uses hydrogen bonds, ionic interactions, hydrophobic association or crystallisation, which avoids chemical cross-linkers but usually gives weaker gels [1][2][3]. This distinction matters for self-healing, since reversible non-covalent and dynamic covalent bonds can break and re-form, whereas permanent covalent cross-links cannot [6][7].

C. Properties and Characterisation

The usefulness of a hydrogel depends on measurable properties [1][2][3]: swelling behaviour, which governs water content, pore size and release; mechanical properties, including stiffness, strength, toughness and elasticity; network structure and porosity; degradation and stability; biocompatibility; and drug loading and release. For self-healing gels, additional properties include healing efficiency, healing time, recovery of mechanical strength after damage, and the ability to shear-thin and recover, evaluated by rheology and by cutting and re-joining tests [5][6][7].

III. SMART (STIMULI-RESPONSIVE) HYDROGELS

Smart hydrogels undergo a reversible change in swelling, structure, mechanical properties or permeability when exposed to a stimulus. The change arises from polymer groups that ionise, form or break hydrogen bonds, or alter hydrophilicity in response to the environment [8][11][12]. The main stimuli are summarised in Table II. A well-studied example is a thermoresponsive hydrogel formed by self-assembly of poly(N-isopropylacrylamide) with clay, which shows self-healing together with temperature-dependent behaviour, illustrating how smart and self-healing properties

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can be built into the same network [13]. Dual or multi-responsive gels, which react to more than one stimulus, are of increasing interest because physiological conditions seldom change in only one way [8][11][12]

TABLE II: STIMULI TO WHICH SMART HYDROGELS RESPOND

Stimulus	Mechanism of response	Typical polymers or groups	Biomedical use	Ref.
Temperature	Polymer changes solubility at a critical temperature, giving sol-gel transition or collapse	Poly(N-isopropylacrylamide), poloxamers	In situ gelling injectables; thermally triggered release	[12][13]
pH	Ionisable groups gain or lose charge, changing swelling	Polymers with carboxyl or amine groups, chitosan	Release in tumours, inflamed tissue or gut	[8][11][12]
Light	Photosensitive groups isomerise, cleave or generate heat	Azobenzene, o-nitrobenzyl groups, photothermal agents	Remote, on-demand release	[8][12]
Enzymes and biomolecules	Enzymes cleave cross-links or glucose alters binding	Peptide cross-links, boronate groups	Disease-triggered release	[8][11][12]
Redox	Disulfide or other bonds cleave in a reducing environment	Disulfide-containing polymers	Intracellular and tumour release	[11][14]
Ionic strength and ions	Charge screening or ionic cross-linking	Alginate, polyelectrolytes	Controlled swelling and gelation	[2][12]
Electric or magnetic field	Conductive or magnetic components move or heat	Conductive polymers, magnetic nanoparticles	Remote actuation; electroactive dressings	[12][15][16]

IV. SELF-HEALING HYDROGELS

A. Concept

Self-healing is the ability of a material to repair damage and restore its original properties, partly or fully, without external help or with a mild trigger. A self-healing hydrogel is designed so that, when cut or cracked, the broken surfaces come together and the cross-links re-form across the interface, restoring a continuous network (Fig. 1) [6][7]. Benefits for biomedical use include longer lifetime under mechanical stress, injectability, ability to fill irregular defects and a degree of safety, since damaged material can recover rather than fail suddenly [5][6][9][10].

B. Dynamic Covalent Bonds

Self-healing in hydrogels is generally intrinsic: reversible bonds that form part of the network allow repeated healing, whereas capsule-based extrinsic healing allows only limited repair [6][7]. Dynamic covalent bonds are chemical bonds that can break and reform under mild conditions. Reviews describe imine (Schiff base), acylhydrazone, boronate ester, disulfide and Diels-Alder bonds [6][7][14]. They give stronger and more stable networks than purely physical interactions while still allowing repair, and many respond to pH or redox conditions, linking self-healing to stimuli-responsive behaviour [6][14]. Schiff-base chemistry is particularly popular for injectable biomedical hydrogels because it proceeds in water without toxic catalysts [14][15][16].

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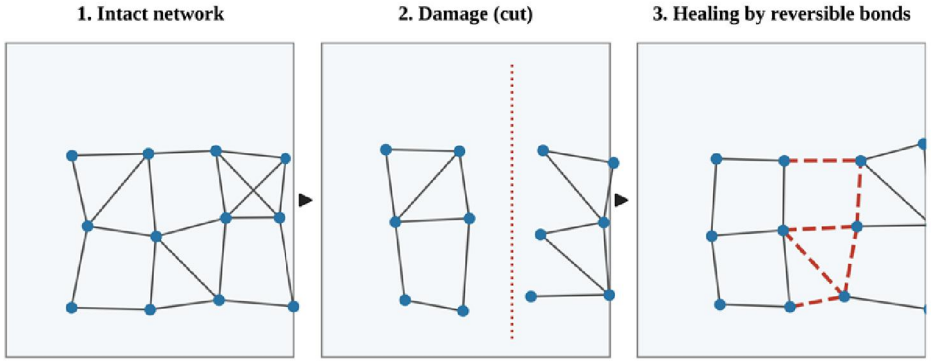

Fig. 1 Principle of self-healing in a hydrogel: an intact network is damaged and then restores its structure through reversible bonds re-forming across the interface.

C. Dynamic Non-Covalent Interactions

Non-covalent interactions are weaker but easily reversible and include hydrogen bonding, hydrophobic association, electrostatic or ionic interactions, host-guest interactions, metal-ligand coordination and pi-pi stacking [5][6][7]. Gels based on them heal rapidly, often without a trigger. Nanocomposite designs in which polymer chains interact with clay have also been used; a poly(N-isopropylacrylamide)-clay hydrogel illustrates a thermoresponsive network that heals through reversible attachment of polymer chains to clay platelets [13]. Their drawback is lower strength and stability, so such gels may be soft and may erode in the body [5][6][7]. The interaction types are compared in Table III.

TABLE III: DYNAMIC INTERACTIONS USED TO IMPART SELF-HEALING TO HYDROGELS

Type of interaction	Examples	Healing behaviour	Strength	Ref.
Dynamic covalent	Imine, acylhydrazone, boronate ester, disulfide, Diels-Alder	Self-healing, sometimes needs mild pH, heat or time	Moderate to high	[6][7][14]
Hydrogen bonding	Polymers with amide, hydroxyl or carboxyl groups	Fast, spontaneous	Low to moderate	[5][6][7]
Hydrophobic association	Amphiphilic polymers, micelle cross-links	Fast, spontaneous	Moderate	[5][7]
Ionic and electrostatic	Alginate with divalent ions; polyelectrolyte complexes	Spontaneous	Moderate	[6][7]
Host-guest	Cyclodextrin with guest groups	Reversible, spontaneous	Moderate	[6][7]
Metal coordination	Catechol-metal, polymer-metal ion complexes	Spontaneous	Moderate to high	[5][6][7]
Polymer-nanoparticle interaction	Clay and polymer chains	Spontaneous	Moderate	[5][13]

D. Strength Versus Healing

A fundamental design problem is that strong networks and fast healing pull in opposite directions: strong cross-links give a tough network but heal slowly or not at all, whereas labile cross-links heal rapidly but give a soft, weak gel. Strategies for resolving this conflict include combining dynamic covalent and non-covalent bonds in one network,

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double-network and interpenetrating structures in which one network provides toughness and another reversible energy dissipation, nanocomposite fillers that reinforce the matrix and add reversible junctions, and multiple hydrogen-bonding or metal-coordination domains that dissipate energy under stress [5][6][7]. Healing at physiological conditions without a trigger and with full recovery of strength nevertheless remains difficult [5][6][7].

E. Injectability and Shear-Thinning

A gel with reversible cross-links can be pushed through a needle or catheter, during which the network breaks and flows like a liquid (shear-thinning), and then reassembles at the target as stress is removed, filling the defect and forming a stable gel in situ [9][10]. Injectable self-healing hydrogels allow minimally invasive delivery of drugs, cells and biomaterials without surgery, and injectability, self-healing and responsiveness together make them especially suited to local, targeted therapy [9][10]. Successful injection requires that the gel recovers quickly after shear and that cross-linking is fast enough to hold at the site but slow enough to allow injection [9][10].

V. BIOMEDICAL APPLICATIONS

A. Controlled Drug Delivery

Hydrogels have long been explored as drug carriers because they hold hydrophilic drugs in their water-filled network, protect fragile molecules such as proteins and peptides, and release drug by diffusion, swelling or degradation [2][4]. Limitations include limited loading of hydrophobic drugs, burst release and difficulty controlling release over long periods [4]. Smart and self-healing hydrogels address several of these weaknesses: release can be switched on or modulated by pH, enzymes, glucose or reactive oxygen species, and injectable self-healing depots can stay at the injection site and release drug locally over a prolonged period, reducing systemic exposure and dosing frequency [8][9][10][11]. Reviews of smart self-healing hydrogels for controlled delivery note that drugs delivered range from small molecules to proteins, growth factors and nucleic acids, and that predictable, reproducible release remains the main goal [8][9].

B. Cancer Therapy

Tumour tissue has a more acidic, more reducing and enzyme-rich environment than healthy tissue, and these differences can serve as triggers. A review of stimuli-responsive and self-healing hydrogels for cancer therapy describes injectable gels placed in or near tumours that release chemotherapeutic, immunotherapeutic or photothermal and photodynamic agents in a controlled way, and that can adhere to the tumour site or surgical cavity after resection, reducing systemic toxicity and the risk of recurrence [11]. Self-healing allows the gel to withstand tissue movement and conform to an irregular cavity [9][11]. Most of this work is in cell culture and animal models [11].

C. Wound Dressings

An ideal dressing maintains a moist environment, adheres to tissue, conforms to the wound, resists infection, reduces oxidative stress and supports repair, and a self-healing hydrogel can remain intact when the patient moves [15][16]. An injectable, conductive self-healing hydrogel dressing with dual-antioxidant and antibacterial properties accelerated wound healing, with conductivity thought to support the electrical signals involved in repair while antioxidant and antibacterial functions limited oxidative damage and infection [15]. A related antibacterial, antioxidant, electroactive injectable hydrogel was designed as a self-healing dressing with high adhesiveness and conductivity [16]. These examples show a trend towards multifunctional dressings in which self-healing provides durability and injectability while other functions address the biology of the wound [15][16].

D. Tissue Engineering and Regenerative Medicine

Hydrogels are widely used as scaffolds because they mimic the extracellular matrix and can carry cells and growth factors; reviews discuss their use for cartilage, bone, nerve, cardiac and skin repair and stress the importance of

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mechanical properties, degradation and bioactivity for each tissue [17][18]. Rational design, in which chemistry, architecture and biological cues are matched to the target tissue, is regarded as the way forward [17]. Injectable self-healing gels can fill irregular defects and deliver cells with limited damage during injection, while stimuli-responsive gels can release growth factors on demand or change stiffness in response to cell or tissue signals [10][12]. Tissues under continuous load, such as cartilage and heart muscle, particularly benefit from materials able to repair microdamage [5][10][12]. Applications are summarised in Table IV.

TABLE IV: BIOMEDICAL APPLICATIONS OF SMART AND SELF-HEALING HYDROGELS

Application	Key design features	Reported benefit	Ref.
Controlled drug delivery	Stimuli-responsive, injectable depots	Triggered, local, prolonged release	[4][8][9]
Cancer therapy	pH-, redox- or enzyme-responsive gels; adhesion to tumour or cavity	Local release, lower systemic toxicity	[9][11]
Wound dressings	Conductive, antibacterial, antioxidant, adhesive, self-healing	Durable dressing; faster healing; infection control	[15][16]
Tissue engineering	Injectable, cell-friendly, responsive scaffolds	Defect filling; on-demand growth factor release	[10][12][17][18]
Load-bearing tissues	Tough, self-healing networks	Resistance to fatigue and microdamage	[5][10]
Smart actuation and sensing	Thermo-responsive nanocomposite gels	Shape or property change on demand	[12][13]

VI. CHALLENGES AND LIMITATIONS

The technology is far from clinical routine, and the reviewed literature identifies a number of challenges [4][6][7][8][14] (Table V). A recurring point is that the complexity of many reported systems is itself a barrier: a multi-component hydrogel with several functional groups, nanoparticles and drugs may perform admirably in the laboratory but be difficult to manufacture, sterilise and register as a medicine or device [4][7][14]. Simpler designs that combine only the functions needed for a particular clinical use are more likely to reach patients [6][7][8]

TABLE V: PRINCIPAL CHALLENGES IN THE DEVELOPMENT OF SMART AND SELF-HEALING HYDROGELS

Challenge	Explanation	Possible response
Strength versus healing	Tough gels heal poorly; fast-healing gels are weak	Dual networks; combined dynamic bonds; nanocomposites [5]
Healing under physiological conditions	Many systems need non-physiological pH, temperature or time to heal	Bonds that are dynamic at pH 7.4 and 37 °C
Biocompatibility and toxicity	Residual cross-linkers, reactive groups and degradation products	Biocompatible components; thorough toxicology [4][14]
Reproducibility and characterisation	Healing efficiency and release measured by non-standard tests	Standard methods for healing and rheology [6][7]
Control of drug release	Burst release; difficulty for hydrophobic drugs	Nanocarrier-loaded gels; stimuli control [4][8]
Degradation control	Mismatch between gel life and tissue repair	Tunable degradable bonds [17][18]

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Challenge	Explanation	Possible response
Sterilisation and storage	Sterilisation may damage dynamic networks	Compatible sterilisation methods; stable formulations
Scale-up and regulation	Complex syntheses; classification as drug, device or both	Simple chemistries; early regulatory dialogue [4][7]
Clinical evidence	Mostly cell and animal studies	Well-designed clinical trials

VII. FUTURE PERSPECTIVES

Several directions appear important. The first is hydrogels that combine toughness and healing under physiological conditions, using hierarchical design and several types of dynamic interaction [5][6][7]. The second is multifunctional materials in which self-healing is combined with antibacterial, antioxidant, adhesive, conductive and bioactive properties, as already shown in advanced wound dressings [15][16]. The third is precision delivery, with gels that respond to specific disease signals and deliver combinations of drugs, cells or biologics [8][9][11]. The fourth is integration of hydrogels with sensors and electronics, a direction suggested by conductive and electroactive systems [15][16]. The fifth is a stronger link between material design and clinical need, with standardised testing, long-term safety assessment and early clinical studies [4][7][17].

VIII. CONCLUSION

Hydrogels are versatile materials whose resemblance to living tissue has made them important in drug delivery, wound care and regenerative medicine, but conventional gels are weak, static and unable to repair themselves. Smart hydrogels, which respond to stimuli such as temperature, pH, light, enzymes and redox conditions, provide control over when and where drugs are released. Self-healing hydrogels, based on reversible covalent and non-covalent bonds, recover from damage, can be injected and reform at the target site, and last longer in mechanically demanding environments. Together these features give injectable, adaptive materials that have shown promise in controlled drug delivery, cancer therapy, multifunctional wound dressings and tissue engineering. Important problems remain: the trade-off between mechanical strength and healing, the need for healing under physiological conditions, safety, reproducibility, sterilisation, scale-up and the scarcity of clinical data. Progress will depend on simpler, well-characterised designs matched to specific clinical needs, standard test methods and rigorous in vivo and clinical evaluation.

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