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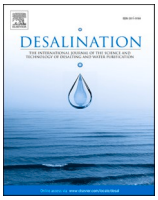

Hydrogels in next-generation energy solutions

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Abbreviations and symbols: AAc, Acrylic acid; AAm, Acrylamide; AAZn, Zinc acrylate; AC, Activated carbon; AEMA, 2-Aminoethylmethacrylate hydrochloride; AG, Alginate; AgNWs, Silver nanowires; AMPSZn, 2-Acrylamido-2-methyl-1-propane sulfonate zinc; BC, Bacterial cellulose; BMAP, Bis [2-(methacryloyoxy) ethyl] phosphate; BNP-HGH, Boron, nitrogen, and phosphorus ternary-doped holey graphene hydrogel; CB, Carbon black; CD, Current density; CMC, Carboxymethyl cellulose; CMCh, Carboxymethyl chitosan; CNF, Cellulose nanofibers; CNT, Carbon nanotube; CPE-K, Self-doped conjugated polyelectrolyte; CTO, Cesium tungsten oxide; DN, Double network; DRGO, Defective reduced graphene oxide hydrogel; EDAB, Ethylene diamine bisborane; EET, Extracellular electron transfer; EIS, Electrochemical impedance spectroscopy; FmoCLL, Fluorenyl methyloxy carbonyl-diphenylalanine; GHG, Greenhouse gases; GO, Graphene oxide; HA, Hyaluronic acid; HBPS, 2-hydroxy-3-butoxypropyl starch; HBSC, Hydrogel-based supercapacitors; HMA, Hexadecyl methacrylate; HPC, Hydroxypropyl cellulose; HPMA, N-(Hydroxypropyl) methacrylamide; HPMC, Hydroxypropylmethyl cellulose; HVAC, Heating, ventilation, and air-conditioning; IPN, Interpenetrating polymer network; ITO, Indium tin oxide; KCA, Carrageenan; KPS, Potassium persulfate; LCST, Lower critical solution temperature; $Li_mCs_nWO_3$, Lithium cerium niobium tungsten oxide; MEG, Moisture electric generator; MFC, Microbial fuel cell; MMA, Methacrylic acid; N, S-GH, Nitrogen and sulfur co-doped graphene Hydrogel; NADES, Natural deep eutectic solvent; NIPAM, N-isopropylacrylamide; N-RGOH, Nitrogen-doped reduced graphene oxide hydrogel; NSG40-γ-MPS, NSG40-3-(Trimethoxysilyl) propyl methacrylate; OCV, Open-circuit voltage; P(SPA), Poly (3-sulfopropyl acrylate); P(VdF-HFP), Poly(vinylidene fluoride-*co*-hexafluoropropene); PAA, Polyacrylic acid; PAAM, Poly(acrylic acid-co-acrylamide); PAAm, Polyacrylamide; PAANa, Polysodium acrylate; PAMPS, Poly(2-acrylamido-2-methyl-1-propanesulfonic acid); PAN, Polyacrylonitrile; PANa, Sodium polyacrylate; PANI, Polyaniline; PAO, Polyamidoxime; PBXHE, Polysaccharide-enhanced hydrogel electrolyte; PD, Power density; PDA, Polydopamine; PDMS, Polydimethylsiloxane; PEA, Phenoxyethyl acrylate; PEDOT, Poly(3,4-ethylenedioxythiophene); PEDOT:PSS, Poly(3,4-ethylene dioxythiophene): Poly(styrene sulfonate); PEG, Polyethylene glycol; PEGDMA, Polyethylene glycol di-methacrylate; PEGMA, Polyethylene glycol methacrylate Hydrogel; PEI, Poly ethyleneimine; PENG, Piezoelectric nanogenerator; PEO, Polyethylene oxide; PET, Polyethylene terephthalate; PHC-Gel, PNIPAM-HPC-CMC-Gelatin; PHF, Polyvinyl alcohol / sulfuric acid / ferric chloride hexahydrate; PHPA, Poly(hydroxypropyl acrylate); PNAH, Poly(N-isopropylacrylamide-*co*-acrylamide) hydrogel; PNIPAm, Poly(di-isopropylacrylamide); PNIPAM, Poly(N-isopropylacrylamide); PPG, Poly(propylene glycol); PPy, Polypyrrole; PS, Potassium tartrate hemihydrate (PTH) - sodium dodecyl sulfate (SDS); PTH, Potassium tartrate hemihydrate; PVA, Polyvinyl alcohol; PVA-g-TMAC, Polyvinyl alcohol grafted with trimethyl ammonium chloride; PVB, Polyvinyl butyral; PVDF, Poly(vinylidene fluoride); PVP, Poly(vinylpyrrolidone); RED, Reverse electrodialysis; rGO, Reduced graphene oxide; SA, Sodium alginate; SCC, Short-circuit current; SCo, Seebeck coefficient; SCs, Supercapacitors; SDS, Sodium dodecyl sulfate; SEM, Scanning electron microscopy; SPAK, Sulfopropyl acrylate potassium; SPMA-Zn, Sodium polymethyl acrylate - zinc sulfate; SSTH, Sandwich structure thermal homeostasiser; TA, Tannic Acid; TEAOH, Tetraethylammonium hydroxide; TEC, Thermoelectro-chemical cells; TEG, Triboelectric generator; TENG, Triboelectric nanogenerator; Ti_3C_2Tx, Titanium Carbide (MXene); TMEDA, N,N,N′*N*′-tetramethylethylenediamine; TR, Thermo-responsive; TRS, Thermo-responsive starch; VOC, Volatile organic compounds; W-VO_2, W-doped vanadium dioxide.

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HIGHLIGHTS

- Critical evaluation of the role of hydrogels in renewable energy revolution.
- Explore advanced hydrogel-based technologies for energy harvesting.
- Investigated the hydrogel's potential in energy storage and conversion systems.
- Hydrogel's perspective to improve the overall energy efficiency and conservation.

GRAPHICAL ABSTRACT

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ABSTRACT

Hydrogels hold significant potential to revolutionize the renewable energy sector due to their unique properties, tunable functionalities, and diverse applications. This study provides a critical evaluation of hydrogels in renewable energy, focusing on their fundamental properties and mechanisms relevant to energy technologies. Advanced hydrogel-based energy harvesting technologies, including hydrogen production, osmotic energy, bioenergy, uranium extraction, as well as mechanical, thermoelectric, and hygro-gradient energy harvesting, are explored in detail. The role of hydrogels in energy storage systems, such as supercapacitors and electrode materials, is analyzed, emphasizing advancements in energy density, cyclic stability, and performance enhancement techniques. The review also addresses the critical challenges of hydrogels, such as scalability, cost-effectiveness, mechanical stability, and environmental sustainability, which must be overcome for broader adoption in energy applications. Furthermore, the transformative potential of hydrogels in energy efficiency and conservation, in advanced energy-saving technologies was highlighted. This paper underscores hydrogels' role in renewable energy security, emphasizing their ability to achieve sustainable development goals (SDGs) and net-zero emissions by decarbonizing the global energy sectors. The present study not only bridges existing research gaps but also provides valuable insights into the future of hydrogels as a cornerstone of sustainable energy ecosystems.

1. Introduction

Energy, a quantitative property that can be converted into various forms, underpins human advancement. With the expanding population and advancing technology, energy demand has become a primary concern. The quest for sustainable and efficient energy solutions is one of the most pressing challenges of the 21st century. The transition to renewable energy sources can be facilitated by innovative materials like hydrogels, which are promising candidates due to their unique structural and functional properties to achieve sustainable energy security and ensure net-zero emissions and sustainable development goals (SDGs). Hydrogels are a class of materials characterized by a water-swollen, three-dimensional structure with controllable intrinsic properties [1–3]. The tunable properties of hydrogels make them promising candidates for applications in energy storage [3–7], production [8–11] and conversion [12–15]. The semi-liquid state of hydrogels enables the conduction of electrical signals, while their mechanical integrity and flexibility make them ideal for green energy applications, including next-generation wearable electronic devices, flexible energy storage devices, and flexible electrodes [1,3,16].

Recent efforts to develop energy-efficient [17,18], reliable, and high-performing renewable energy technologies have highlighted the significant potential of hydrogel varieties and their composites in providing cost-effective solutions i.e., hydrogen production [19,20], uranium extraction from seawater [21,22], osmotic energy harvesting [23,24] and bioenergy harvesting [25,26]. Hydrogels also hold the potential to revolutionize novel sustainable technologies such as thermoelectric materials [27,28], moisture gradient energy [29,30] and mechanical energy [31,32], thereby enabling efficient energy harnessing from the environment. The groundbreaking and classical applications of hydrogels are extensive in different fields, i.e., biomedical [33,34], medicine and biopharmaceuticals [35–37], drug and RNA delivery and sensing [35,38–40], bone and tissue engineering [41,42], robotics [43,44] and human-machine interfaces [45], bioelectronics and ionotronics interfaces [46,47], desalination and water treatment including solar interfacial evaporation [48–50], atmospheric water harvesting [51] and enzyme catalysis [52].

The utilization of hydrogels within the energy sector is extensive and widespread but sporadic. However, to date, most of the review papers on the energy applications of hydrogels focus primarily on their fundamental properties and classifications. They also cover hydrogel-based energy storage devices, including wearable energy storage systems, supercapacitors, metal-ion and metal-air batteries, water-based energy conversion systems, as well as electrocatalysts together with water purification technologies [53–61]. Despite the burgeoning interests in hydrogels, there is a significant gap in the literature when it comes to comprehensive reviews that encapsulate their multifaceted potential within the energy sector. While several studies have explored the fundamental properties of hydrogels and their applications in isolated contexts, none have comprehensively examined their versatile role across a wide spectrum of energy harvesting technologies, energy storage devices and their role in energy efficiency and conservation. This review seeks to thoroughly investigate the potential of hydrogels in transforming the energy sectors (Fig. 1). We intend to analyze recent progressions in hydrogel-based advanced energy harvesting technologies, energy conversion devices, energy storage systems, and energy-saving technologies. Furthermore, this study critically assesses the methodologies and mechanisms utilized to enhance the properties of

hydrogels for energy harvesting, conservation and efficiency. It aims to elucidate the transformative influence of hydrogels in the renewable energy sectors.

Moreover, it endeavors to identify significant challenges and forthcoming directions within this rapidly expanding field. Through bridging the current research gap, this review not only emphasizes the significance of hydrogels in sustainable energy solutions but also lays the groundwork for future investigations aimed at maximizing their efficacy. Given the evolving energy landscape, comprehending the role of innovative materials such as hydrogels will be paramount in shaping a sustainable and resilient renewable energy future.

2. Fundamentals of hydrogels in renewable energy applications

Hydrogels are hydrophilic molecular arrays that retain water, which are not particularly solid or liquid but hold exclusive characteristics of both solid and liquid [3,62]. Hydrogels are ubiquitous in the energy sector because of hydrogel's polymer network that can hold water molecules within and their superior softness, bioactivity, and biocompatibility [2]. The polymer network causes the hydrogels to be elastic solids, maintaining their shape and size with deformability and softness [2,47], while the water molecules within the polymer network make the hydrogel an ionic conductor [47]. The permeability and transferability of a wide range of chemicals and biological molecules are attributed to the high-water content in the hydrogel matrix that gives it liquid-like attributes [2]. Hydrogels have been employed and are currently the subject of extensive investigation in the domain of batteries, supercapacitors, fuel cells, energy harvesting technologies and devices for energy storage [3,47]. The use of hydrogels can revolutionize the energy sector due to their versatile applications merged with notably better

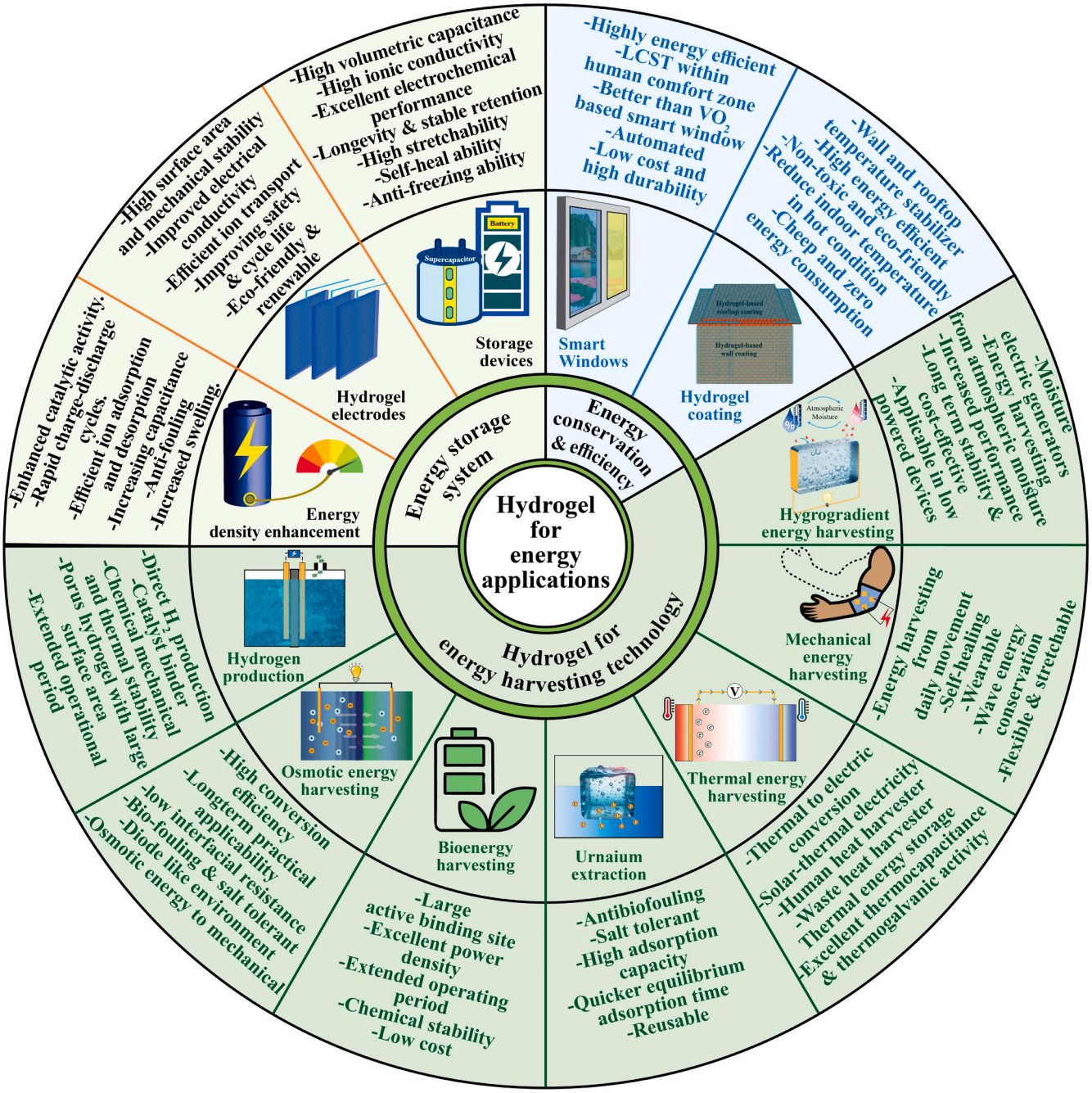


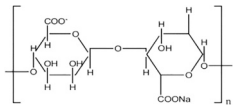
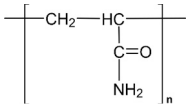
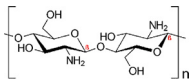
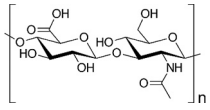
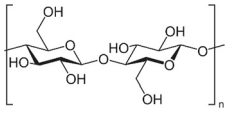
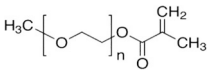
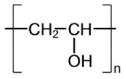
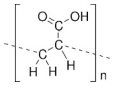
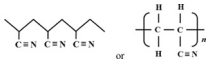
Fig. 1. Schematic illustration of hydrogel's applications in the renewable energy domain.

performance, stability and durability. In this section, we discuss the unique properties of hydrogels, emphasizing their suitability in the energy sector.

2.1. Hydrogels: An emerging horizon in the energy sector

The polymer network that consists of the structure of hydrogel has a mesh size of about 10 nm, which enables the water molecules absorbed by the hydrogels to have the same chemical and physical properties as water in a liquid state [47]. In the liquid state, water can conduct electric charges by conveying ions. However, liquid electrochemical substances tend to have short-term electrochemical stability [63]. That is where hydrogels act as the bridge between solids and liquids, as hydrogels maintain superior mechanical and chemical stability [64]. Hydrogels can have tunable porosity, water content, mechanical properties, thermal properties, and electrochemical properties [1–3,47,62], which makes them promising candidates for numerous applications. The variation of properties of hydrogels is attributed to the interaction of water molecules with the hydrophilic groups within the polymer chains due to the influence of surface tension, hydrogen bonding, van der Waals forces, and electrostatic interactions [5,65]. These interactions can be controlled by internal and external factors like chemical composition, degree of crosslinking, temperature, pH, reaction conditions etc. [4,5,65]. The molecular arrays within hydrogels exhibit a capacity for reversible deformation or reformation in response to specific stimuli to which the hydrogels are susceptible [65]. The capability for reversible deformation and reformation of molecular arrays within hydrogels is harnessed to fine-tune their properties, rendering them highly suitable for many applications within the energy sector, such as hydrogen production [19], batteries [7], supercapacitors [6,66,67], electrochemical devices [68], fuel cells [69], energy storage devices [5], flexible electronics [70], printable conductive polymers [16], and triboelectric and piezoelectric devices [71] (see Fig. 1). Recent efforts in hydrogel research within the energy field offer the potential to revolutionize the

Table 1
Summary of hydrogel varieties based on various polymeric backbones in energy applications.

Polymeric backbones	Chemical formulas	Sources	Applications	Advantages	Disadvantages
Alginate		brown algae (Phaeophyceae)	– Osmotic energy harvesting [8]	– Biocompatible – Non-toxic – Cost-effective	– Low retention capacity – Limited chemical stability
Polyacrylamide (PAAm)		Synthetic	– Solar thermoelectricity energy harvesting [74]	– High hydrophilicity – Host-effective – Super absorbent	– Unreacted monomers leach out – Low thermal stability
Chitosan		Crustacean shell	– Electrochemical energy storage [75] – Uranium recovery [72]	– Biocompatible – Antimicrobial activity	– Brittle – Limited electroconductivity
Hyaluronic Acid (HA)		Animal tissues and streptococci bacteria	– Triboelectric nano generator [76]	– Biocompatible – Lubricating properties	– Expensive – Limited mechanical strength
Cellulose		Plants and bacteria	– Flexible batteries [77] – Thermocells for low-grade heat harvesting [9]	– Abundant and low-cost – Non-toxic – Strong and flexible	– Low water solubility – Requires chemical modification for electroconductivity
Collagen	$C_4H_6N_2O_3R_2{\cdot}(C_7H_9N_2O_2R)n$	Animal tissues and bones	– Flexible electronic device – Electrochemical energy storage [78]	– Natural biomaterial – Tunable mechanical properties	– Expensive – Difficult processing
Polyethylene oxide (PEO)		Synthetic	– Thermoelectric material [10]	– Highly hydrophilic – Non-toxic – Biocompatible	– Weak mechanical properties – Limited ionic conductivity – Low water retention capacity
Polyvinyl alcohol (PVA)		Synthetic	– Solar thermoelectric conversion [79] – Triboelectric nanogenerator [73] – Hydrogen production from direct seawater [19]	– ost-effective – good chemical stability	– Poor mechanical properties – Low electroconductivity
Polyacrylic acid (PAA)		Synthetic	– Thermochromic energy saving smart windows [17]	– High ionic conductivity – Can make a complex with metal ions – Tunable water content	– Low water retention capacity – Unreacted monomers leach out
Polyacrylonitrile (PAN)		Synthetic	– Supercapacitor [80]	– Good mechanical strength – Thermally stable – Chemically resistant – Can be doped with conductive materials	– Difficult processing

sector through numerous advancements. Over the years, hydrogels have undergone thorough investigation and tailored modifications for their application in the energy sector. Table 1 provides an overview of some common hydrogels based on the different polymeric backbones with their sources, applications in the energy field, and their advantages and disadvantages. Viewing from their chemical formula, hydrogels are mostly polymeric structures, and their sources vary from natural sources like algae, bacteria, animal tissues, and plants to synthetic sources.

Recent studies suggest hydrogels have indeed opened a new horizon in the energy sector and unveiled monumental advancements in ionotronic devices, wearable electronics, triboelectric devices, fuel cells, supercapacitors, and energy storage devices. In a recent experiment, Xie et al. developed a membrane-based seawater electrolyzer device in which they used a self-dampening electrolyte based on polyvinyl alcohol (PVA) soaked in KOH-based (PVA-KOH) hydrogel [19]. This hydrogel's self-dampening property enabled them to develop a cost-effective water electrolysis system for hydrogen production. Hydrogels are also used in batteries for improved performance. Production of uranium is primarily from naturally occurring ores until recently, recovery of uranium from seawater has seen promising avenues by using hydrogels. Zhang et al. developed a semi-interpenetrating hydrogel that proved to be an impressive absorbent with efficient uranium harvesting from real seawater [72]. Recently, a salinity-tolerant metal-free ionic hydrogel was used to extract uranium from seawater [11]. The newly developed hydrogel had a peak adsorption capacity that exceeded most available adsorbents. Wang et al. modified polymeric zwitterion polymer by molecular scale lubrication mechanism to develop lean water hydrogel electrolyte for zinc ion batteries [3]. Their hydrogel-based batteries had better coulombic efficiency, outstanding cycling performance and a wider stable voltage window. Hydrogels are also used for energy-efficient applications; for instance, low vaporization enthalpy hydrogels for deriving pure water using solar energy only [48]. The most promising development brought about by hydrogel is in wearable and flexible electronic devices. Zhou et al. developed a wearable electronic device that can generate electricity by triboelectric nanogenerator based on PVA hydrogel [73]. Another study reported a flexible supercapacitor based on bacterial cellulose (BC) aerogel with sodium polyacrylate (PANa) soaked in KOH (PANa/KOH) hydrogel reinforcement [4].

Hydrogels emerge as pivotal entities for pioneering a novel frontier within the energy sector, showcasing remarkable prevalence in diverse applications. Cutting-edge renewable energy harvesting technologies, including energy storage devices and energy-efficient materials for diverse applications, are substantially refined and optimized through the integration of various hydrogel types.

2.2. Ion conduction in hydrogels

Metals have long been recognized for their ion conductivity, yet beyond metals, diverse materials exhibit the capacity to conduct electricity. Since the advent of electricity, there has been a continuous development and enhancement of conductive materials for various applications. Ion-conducting materials typically fall into the categories of metallic solids or electrochemical liquids. Hydrogels, on the other hand, represent a unique class of materials that possess characteristics of both semi-solids and liquids, enabling them to conduct ions. The tunable conductivity of hydrogels has prompted extensive research to enhance

Table 2
Summary of techniques used for ion conductivity enhancement in hydrogels.

Hydrogel's polymers	Techniques used	Increased ion conductivity (S cm^{-1})	Applications	Remarks
Alginate	– Addition of $CaSO_4$, then dried and rehydrated with $CaCl_2$	0.0022	– Energy storage device and flexible conductive material [70]	– Exceptional mechanical stability
	– Combination of chitosan by anti-polyelectrolyte effect and semi-dissolution acidification sol-gel transition method	0.296 0.049	– Flexible supercapacitor [82]	– Excellent capacitance retention – High specific capacitance
PAAm	– Cross-linked by methacrylated graphene oxide	0.127	– Compressible supercapacitors [83]	– Good thermal stability – Wide temperature tolerance
Chitosan	– In-situ crosslinking with dimethyl sulfoxide and saltwater	0.0137	– All-solid-state supercapacitor [84]	– Can operate at a wide range of temperatures
Poly(N-isopropylacrylamide) (PNIPAM)	– Gelation and in situ polymerization	0.002 and 0.008	– Motion sensing [85]	– Change in electroconductivity under mechanical stimuli
Cellulose	– Cellulose nanofibers are used in copolymerized acrylamide monomers and sodium acrylate monomers	0.103	– High voltage flexible supercapacitor	– Excellent rate performance – High cycling stability
PEO	– Incorporation with polyvinylidene fluoride and $LiClO_4$	0.006	– Rechargeable lithium-ion batteries [86]	– Ideal choice for next-generation rechargeable high-energy lithium batteries
PVA	– Soaked in KOH and swelled	0.340	– Zinc-air batteries [81]	– Different amount of KOH used to prepare and check electroconductive performances
	– Soaked in KOH and composite created with tetraethylammonium hydroxide	0.064 0.034	– Flexible zinc–air battery [7]	– Improved water retention and delayed dehydration and crystallization
	– Blending with sodium alginate and polyvinyl glycol	0.0314	– Flexible ionic thermoelectric supercapacitor [87]	– Cutting edge thermally chargeable supercapacitor
PAA	– *Co*-polymerization of acrylic acid and octadecyl methacrylate	0.015	– Flexible supercapacitor [88]	– Remarkable physical resistance
PANa	– Soaked in KOH and filled in with bacterial cellulose aerogel	0.019	– Flexible supercapacitor [4]	– Very well adapted to extreme conditions – Strong, anti-freezing property
PAN	– Ionic liquid impregnation (EMIM-BF4)	0.0424	– Wearable strain sensor [89]	– Works in extremely low temperature.
Poly(vinylpyrrolidone) (PVP)	– Hydrothermal synthesis technique used for direct fabrication of silver within PVP nanowire hydrogel	0.125	– Energy storage [90]	– Very stable in aqueous and organic solvents

ion conductivity, aiming to optimize the performance of electronic devices. Ion-conducting electrolytes are prepared by entrapment of liquid electrolytes into different hydrogel hosts [81]. The morphology and electroconductive properties of the hydrogels depend on the type and content of the hydrogel polymer host, electrolyte, and solvent present in the polymer. In the case of hydrogels, water acts as the solvent.

The ion conductivity of hydrogel is increased for several compelling reasons, such as enhancing the performance of electronic devices, efficient energy harvesting, efficient energy storage, optimized sensitivity of sensors, and enhanced electrochemical activity [3–5,7,47,62,70]. Table 2 summarizes the ion conductivity enhancement techniques of hydrogels used in recent energy applications.

The enhancement of ion conductivity in hydrogels is accomplished through techniques like copolymerization, which involves incorporating various functional groups onto the same main chain. This strategy serves to mitigate the limitations associated with certain polymers [5,88]. An alternative approach for enhancing ion conductivity in hydrogels involves crosslinking modification (Fig. 2). This method entails the incorporation of suitable crosslinking agents into polymer monomers, which bind the polymeric monomers and form a network that inherently holds water in the matrices. This mechanism not only enhances the mechanical strength of the hydrogel, but also aids in the augmentation of ionic conductivity [5,83]. The grafting of branched monomers, which facilitates ion transport, represents another avenue for enhancing ion conductivity in hydrogels. It is imperative to recognize the existing challenges associated with the improvement of ionic conductivity in hydrogels. A systematic exploration into the long-term stability and potential side effects resulting from enhanced ion conductivity in various applications is pivotal for the practical implementation of these strategies in energy applications.

2.3. Stability of hydrogels

While hydrogels exhibit promising attributes for applications in energy storage devices, fuel cells, supercapacitors, etc., it is undeniable that they often lack stability in terms of thermal, mechanical, and electrochemical aspects. Extensive research efforts have been devoted to addressing these stability issues over the years, and numerous studies have yielded impressive results in enhancing the stability of hydrogels for various applications. Long-term operation of devices reliant on hydrogels has been observed to detrimentally impact the electrochemical performance of these devices. To mitigate this issue, researchers have devised various mechanisms aimed at enhancing not only electrochemical performance but also mechanical and thermal stability (Fig. 2). Furthermore, recent advancements in research have led to the development of self-healing hydrogels, introducing an innovative dimension to address longevity and operational efficiency concerns in hydrogel-based devices [10,88]. A multi-responsive and healable supercapacitor, employing a Fe_3O_4@Au/PAAm hydrogel, had been developed. This unique device demonstrates responsiveness to light, capable of self-healing upon exposure to light. Additionally, it exhibits automatic healing in response to a magnetic field and elevated temperature, showcasing a versatile and self-repairing capability [6]. A recent study has successfully engineered a hydrogel utilizing poly(3,4-ethylene dioxythiophene): poly(styrene sulfonate) (PEDOT: PSS). This hydrogel demonstrates adjustable mechanical properties and presents an innovative application in the realm of 3D printing for the fabrication of conductive hydrogels [91].

Table 3 presents a comprehensive overview of the mechanisms employed to fine-tune and enhance the mechanical, thermal, and electrochemical performances of hydrogels utilized in energy applications. Recent techniques, such as reinforcement through clay nanoparticles and carbon nanotubes, have emerged as particularly noteworthy, showcasing exceptional outcomes in bolstering the stability and overall performance of these hydrogels [92,93].

Hydrogels, while promising for energy applications, grapple with instability in thermal, mechanical, and electrochemical domains. Despite substantial research efforts, their long-term use negatively impacts electrochemical performance. Innovations like self-healable hydrogels and materials offer remarkable improvement, yet challenges persist. The detailed mechanisms outlined in Table 3 elucidate strategies, including double network hydrogels, clay nanoparticle reinforcement, and CNT incorporation, showcasing intricate trade-offs in mechanical, thermal, and electrochemical enhancements. While these approaches demonstrate progress, the multifaceted nature of hydrogel stability remains apparent.

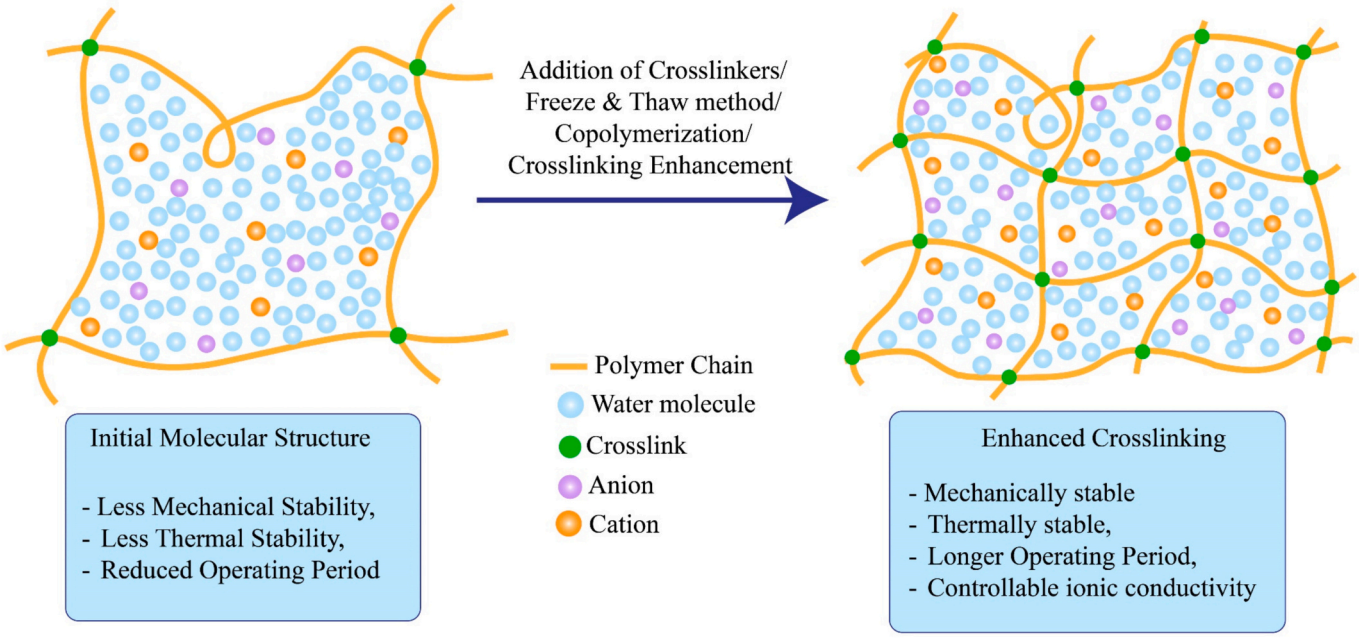


Fig. 2. Molecular array of hydrogels enhancing ionic, mechanical, and thermal properties. Hydrogels' tunable ionic, mechanical, and thermal properties make them ideal for precise modifications in various energy applications.

Table 3
Techniques and mechanisms of enhancing mechanical, thermal and electrochemical stability of hydrogels in energy applications.

Strategies	Modification techniques	Mechanisms	Mechanical improvement	Thermal improvement	Electrochemical improvement
Double network hydrogels (DN hydrogels) [94]	Introduction of a secondary polymer network	Interpenetrating networks for stress distribution and energy dissipation	Significantly improve mechanical strength and toughness	Enhance thermal stability due to multiple crosslinking networks offering redundant pathways for energy dissipation.	It can improve ionic conductivity due to increased connectivity and water retention.
Clay nanoparticle reinforcement [92]	Dispersion of clay nanoparticles (montmorillonite, halloysite)	Increased crosslinking density and intermolecular interactions	Significantly enhance mechanical strength and stiffness due to strong interfacial interactions between clay and polymer chains.	Improve thermal stability through enhanced heat transfer and restricted polymer chain mobility.	It may slightly decrease ionic conductivity due to the physical blockage of ion pathways.
Carbon nanotube (CNT) incorporation [93]	CNT networks within the hydrogel matrix	Strong interfacial interactions and stress transfer	Highly enhanced mechanical strength, stiffness, and toughness due to CNT's exceptional stiffness and load-bearing capabilities.	Significantly enhance thermal conductivity due to CNT's intrinsic thermal properties.	It can significantly improve ionic conductivity depending on CNT type and dispersion, providing conductive pathways.
Bioinspired mineralization [95]	Mimicking natural calcium phosphate formation	In situ formation of nanocomposite structures	It can significantly improve mechanical strength and elasticity due to the incorporation of mineral nanoparticles within the hydrogel matrix.	Mimics natural tissues with high strength and thermal stability, offering enhanced thermal tolerance.	It could influence ionic conductivity depending on the type and amount of mineralization, potentially providing additional ion channels.
Dynamic covalent bonds [96]	Introduction of crosslinks with reversible bonds (disulfide, Diels-alder)	Energy dissipation through bond breaking and reforming	Enhance mechanical strength and toughness due to the ability of dynamic bonds to redistribute stress and repair damage through bond reformation.	Enable self-healing and stress dissipation, potentially improving thermal stability by repairing network fractures.	It can provide dynamic ion pathways through bond breaking and reforming, potentially enhancing ionic conductivity.
Cryogelation [97]	The freezing and thawing process generates interconnected pores	Tailored pore structure for enhanced compression and shear resistance	Mechanical strength increases due to the repetition of thaw and freezing, enhancing the crosslinking	Creates interconnected porous structures with high surface area, potentially improving thermal insulation.	It can influence ionic conductivity depending on pore size and distribution, potentially promoting or hindering ion transport.
Hydrophobic modifications [98]	Grafting hydrophobic groups (alkyl chains)	Improvement in interchain packing and resistance to water-mediated failure	Improves mechanical characteristics by limiting the hydrophilic properties	Introduce hydrophobic domains that can repel water and enhance air entrapment, improving thermal insulation.	It may decrease ionic conductivity by reducing water content and ion mobility.
Ionic crosslinking [87,99]	Introduction of ionic interactions between oppositely charged polymers	Strong electrostatic interactions and enhanced cohesive forces	It can improve mechanical strength and toughness due to the strong electrostatic interactions between ionic crosslinks.	Utilizes ionic interactions for crosslinking, offering the potential for self-healing and improved toughness.	It can enhance ionic conductivity due to the presence of mobile ions within the network.
Double crosslinking [100]	Combining covalent and ionic crosslinking	Synergistic effect on network stability and energy dissipation	It can significantly enhance mechanical strength and toughness due to the synergistic effect of multiple crosslinking mechanisms.	Introduces two different crosslinking mechanisms for enhanced stability and redundancy.	It can influence ionic conductivity depending on the types of crosslinking used, potentially affecting water content and ion mobility.

2.4. Biodegradability and durability of hydrogels in energy applications

Hydrogels are mostly polymers prepared from petrochemicals and other chemical entities [101].The persistent challenges in the final disposal of hydrogels, often resulting from their non-biodegradable nature and potential environmental persistence, highlight an imperative concern. While ongoing research endeavors aim to enhance the mechanical, thermal, and electrochemical durability of hydrogels utilized in energy applications, the issue of their end-of-life fate remains a formidable challenge. Consequently, concerted efforts have been directed towards the development of eco-friendly hydrogels, aligning with a broader commitment to sustainability and mitigating environmental impacts [102]. A groundbreaking achievement in the development of eco-friendly hydrogels was realized through the creation of an all-natural hydrogel comprising cellulose and bentonite clay, showcasing remarkable biodegradability [101]. This innovative hydrogel not only combines superior mechanical performance with high ionic conductivity but also exhibits exceptional freezing tolerance. This pioneering development represents a significant stride towards the creation of environmentally sustainable hydrogels.

The strategic blending of degradable polymers emerges as a promising avenue for the fabrication of biodegradable hydrogels. A notable example involves the development of a solid-state zinc battery featuring a flexible hydrogel electrolyte based on PVA and BC [103]. The cost-effective nature of cellulose contributes to the affordability of the hydrogels, and its inherent biodegradability is complemented by its high thermal and chemical stability when blended with the PVA hydrogels. In a recent study, a BC-based hydrogel was employed to successfully create thermocells, showcasing its potential for harvesting low-grade heat in a cost-effective and environmentally friendly manner [9]. Kim et al. [76] pioneered the development of a triboelectric nanogenerator (TENG) utilizing a hydrogel film derived from HA, showcasing remarkable biodegradability. A recent study developed a hydrogel composite of alginate, sericin, and cellulose to create an interpenetrating network that has biodegrading properties [104]. Chemical and enzymatic pretreatment of hydrogels can prove to be fruitful in terms of making biodegradable hydrogels. These enzymes increase the hydrophilicity of the hydrogels which facilitates the degradation of hydrogels.

The burgeoning energy sector, particularly in renewable and sustainable avenues, presents exciting opportunities for utilizing hydrogels. However, the environmental burden of persistent hydrogel materials necessitates a critical lens on their biodegradability and durability. Analyzing these aspects will reveal both limitations and opportunities for optimizing hydrogels in energy applications.

Hydrogels exhibit significant promise in the energy sector due to their versatile polymer backbones and unique physicochemical properties. Recent advancements in optimizing ionic conductivity have demonstrated their potential for efficient ion transport, making them highly relevant for energy applications. Efforts to enhance hydrogel stability have further validated their feasibility for long-term use under diverse conditions. Meanwhile, balancing biodegradability and durability remains a critical challenge, underscoring the importance of designing hydrogels that are both high-performing and environmentally sustainable.

3. Advanced hydrogel-based energy harvesting technologies

With the ever-increasing demand for energy, various technologies have been developed to harvest energy from diverse sources. The versatile application of hydrogels in energy harvesting has significantly enhanced effectiveness and advancement in this field. Many challenges encountered during energy harvesting from different sources have been overcome through the utilization of hydrogels. The use of hydrogels in energy harvesting is extensive, encompassing hydrogen generation [13,19,20], uranium extraction [21,105,106], bioenergy harvesting [25,26,107], mechanical energy harvesting [31,108,109], osmotic energy harvesting [8,18,110], hygro-gradient energy harvesting [29,30], thermal energy harvesting [111] and more. There are also reports of hydrogels being employed to mimic biological bodies for energy harvesting [95]. Additionally, combining hydrogels has shown promise in integrating their characteristics for simultaneous energy generation from multiple sources. This section will explore advanced technologies for harnessing energy using hydrogels in detail.

3.1. Hydrogel-mediated hydrogen production

Hydrogen fuel has the potential to be a gateway to a sustainable future. This clean and sustainable energy source has spurred intensive research and innovative technologies due to its high energy density, recyclability, non-toxicity, and eco-friendly production methods [13]. Numerous researches have been dedicated to enhancing the production of hydrogen with the aid of hydrogels. The efforts encompass performance enhancement of existing strategies, drawbacks minimization, and innovation of newer technologies. Table 4 highlights recent remarkable uses of hydrogels for the production of hydrogen.

Hydrogels have been proven to be an excellent medium to produce hydrogen by electrolysis, photolysis and hydrolysis of a wide variety of substances like seawater, sodium borohydride, human urine, brine, etc. [12,13,19,112,113]. A recent study conducted by Im et al. produced hydrogen while simultaneously producing liquid fertilizer from human urine [12]. The study employed the use of PVA-KOH hydrogel incorporated within melamine foam that achieved as high as 32.48 mL of H_2 production per minute for a current density of 212.60 mA/cm^2. Their demonstration was a successful proof of hydrogen production from human urine. The presence of ions restricts long-term water electrolysis using seawater. Due to this, filtration is required prior to the electrolysis of seawater. However, Xie et al. were successful in producing H_2 directly from seawater without any prior purification [19]. Their novel PVA-KOH hydrogel acted as a self-dampening electrolyte sustaining the motive force for water migration across a hydrophobic membrane without the need for an external energy source. This research pioneered a cost-effective alternative to industrial alkaline water electrolysis systems. A relevant study was recently conducted by Kabir et al. (2025) in which PVA-KOH hydrogel-based electrolyte was modified with tetraethylammonium hydroxide (TEAOH) to achieve self-wetting property that drove the transfer of water molecules thorough a hydrophobic membrane in a water electrolysis cell [113]. In another recent study, Poly ethyleneimine (PEI) hydrogel was coated on a platinum plate and nickel foam electrode that repelled the gas bubbles, enhancing the hydrogen evolution reaction [114]. This inexpensive method demonstrated a stable electrochemical hydrogen production system suitable for practical scalability.

Catalytic hydrolysis is another technique to produce hydrogen, especially from sodium borohydride ($NaBH_4$). This is a preferable technique because, unlike electrolysis, this technique does not require current. As hydrogels can adsorb and trap catalytic ions within their matrices due to electrostatic interaction with the functional groups, they serve as excellent templates for holding catalysts. Ding et al. used cellulose derived from wheat straw to make a composite hydrogel mixed with PVA, which provided the template for Ni and Cu nanoparticles [13]. This eco-friendly hydrogel not only prevented the aggregation of metal nanoparticles but also demonstrated better catalytic efficiency. A similar hydrogen production mechanism can be observed in another study, where poly(3-sulfopropyl acrylate) hydrogel supported the template for Pd nanoparticles that served as catalysts during the hydrolysis of ethylene diamine bisborane (EDAB) [115]. This study demonstrated the suitability of EDAB as a long-term storage of hydrogen. Modified hydrogels containing metallic catalysts can serve as a super porous catalyst system for increasing the catalytic activity due to increased surface area [116]. This porous hydrogel system containing metal ions enables controllable H_2 production by manipulation with a magnetic field.

The utilization of photon energy is yet another significant technique to produce hydrogen. Multifunctional integrated systems composed of hydrogels are deployed for the production of hydrogen utilizing photon energy. For instance, a hydrogel derived from PVA and chitosan filled with wood-based cellulose loaded with photocatalysts simultaneously produced hydrogen and evaporated freshwater from seawater and wastewater [14]. This hydrogel system efficiently intercepted volatile organic compounds (VOC) from wastewater while producing up to 9.7 mmol/g.h of hydrogen, proving the practical applicability. In another study, photocatalyst-added agarose hydrogel generated H_2 from metal-containing industrial wastewater [117]. This mechanism eliminated the catalyst poisoning caused by metal contamination. A recent study on graphene-based hydrogels developed a dual-role composite hydrogel that demonstrated excellent photocatalytic activity during the production of H_2 from seawater and freshwater [112]. This hydrogel composite is a promising corrosion-resistant material for the production of H_2 using visible light absorption. Lee et al. recently developed a hybrid microbial hydrogel of polypropylene glycol (PPG) and hydrophilic polyurethane incorporated by N_2-fixing bacteria [20]. This microbial device can float above the water surface and produce hydrogen from water via the N_2 fixation mechanism. This scalable, environmentally friendly solar H_2 production system is the first demonstration of hydrogen generation using nitrogen-fixing microbes. In a recent study conducted by Grinberg et al., hydrogenase enzymes were immobilized by a fluorenyl methyloxy carbonyl-diphenylalanine (FmocLL) hydrogel matrix that demonstrated highly efficient enzyme-mediated electrochemical H_2 production [118].

Hydrogen fuel has garnered significant global attention; nonetheless, challenges persist in hydrogen generation from diverse sources. In recent years, hydrogels have emerged as a promising solution, effectively mitigating these challenges. The incorporation of hydrogels in hydrogen generation processes has facilitated the development of more efficient, stable, reliable, and environmentally friendly techniques (Fig. 3). Novel hydrogels have addressed previous limitations in hydrogen production, paving the way for innovative technologies that extend beyond laboratory-scale applications. Engineered hydrogel materials act as smart entities that respond to external physical stimuli, allowing for the control of hydrogen production rates. However, challenges remain in the domain of hydrogen production and utilization. Notably, membrane fouling and electrode corrosion during long-term water electrolysis operations reduce hydrogen production efficiency. Additionally, the most commonly applied method for hydrogen production, water electrolysis, incurs higher costs due to the use of expensive catalysts and the expenses associated with compression and liquefaction for long-term storage. The use of noble metal catalysts in

Table 4
Summary of hydrogel-mediated hydrogen production.

Hydrogels used	The technique of using hydrogels	Mechanisms	Target substances	H_2 production rate	Current and/or power density	Advantages	Remarks
PVA [12]	- Liquid PVA hydrogel was poured into a porous melamine foam - Then it was frozen and immersed in KOH solution	- Melamine-incorporated hydrogel stacked cell acted as a self-dampening electrolyte - A self-driven transition mechanism simulta-neously separated pure water and produced H_2	- Human urine	-32.48 mL/min	- 212.60 mA/cm^2	- Simultaneous production of hydrogen, pure water, and liquid fertilizer from human urine	-Successful proof of hydrogen production from human urine
PVA [19]	- KOH solution was mixed with PVA hydrogel - PVA-KOH hydrogel acted as a self-dampening electrolyte (SDE)	- The SDE provided a driving force across hydrophobic membrane in the electrolyzer for continuous water migration within the hydrogel for continuous electrolysis	- Seawater	–	- 250 mA/cm	- Direct H_2 generation from seawater - Extended effective operating time - Less expensive than industrial alkaline water electrolysis	- A H_2 generation system direct from seawater with eliminating corrosion and side reaction problems - Promising spatiotemporal adaptability
PEI [114]	Crosslinked PEI was coated on flat Pt and porous Ni foam electrodes	- Superaerophobicity of PEI hydrogel repelled the gas bubbles from the electrodes - Prevented blocking of active surface area, thus enhancing hydrogen evolution reaction	- H_2O	–	- Current density requirement was tested	- Enhanced hydrogen evolution - Outperforms the electrodes modified with electrocatalysts - Inexpensive - Stability of hydrogels is suitable for practical water electrolysis	- Insights can be used for other gas evolution reaction - Can be scaled up
Cellulose [13]	- Wheat straw cellulose (WSC) added with PVA and crosslinker to form hydrogel - WSC hydrogel was used as template for Ni and Cu nanoparticles - Metal ion containing hydrogel was mixed with $NaBH_4$ solution	-WSC hydrogel-nanometal composite acted as a catalyst to produce hydrogen from hydrolysis of $NaBH_4$	- $NaBH_4$	- Hydrogel-Ni composite: 129.96 mL/g.min - Hydrogel-Cu composite: 27.84 mL/g.min	- no current is required for hydrolysis	- Eco-friendly hydrogel - The hydrogel prevented the aggregation of metal nanoparticles and maintained full dispersion - Better catalytic efficiency - Can be used up to 5 times with 100 % conversion	- Better performance than other catalysts - Applicable for catalytic conversion
Poly (3-sulfopropyl acrylate) [115]	- Pd (II) ions were adsorbed into p (SPA) hydrogel and reduced by $NaBH_4$ to make Pd (0) nanorod containing hydrogels - Pd (0) nanorod containing hydrogel turbulently mixed with EDAB	- Hydrogel supported the Pd nanoparticles, which acted as the catalyst for the hydrolysis of EDAB	- Ethylene diamine bisborane (EDAB)	-Turnover frequency (TOF): 5.90 mol H_2/mol Pd (0). min	- No current is required for hydrolysis	- Good catalytic activity - Promising catalytic system - Capable of long-term storage	First demonstration of hydrogen production from EDAB using Pd(0) nanoreactor containing hydrogels.
PAA [116]	- P(AAc) hydrogel was synthesized via free radical polymerization - Cryogels were formed in very low temperature - Co^{2+} and Ni^{2+} ions were reduced into the hydrogel and cryogel matrices	- Co and Ni containing hydrogel and cryogel served as the porous catalysts system for catalytic hydrolysis of $NaBH_4$	- $NaBH_4$	-TOF-Hydrogel: 0.62 mol H_2 / (mol Co.min) -Cryogel: 4.1 mol H_2/ (mol Co.min)	- No current is required for hydrolysis	- High catalytic activity	- Controllable H_2 Production by magnetic field

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Table 4 (*continued*)

Hydrogels used	The technique of using hydrogels	Mechanisms	Target substances	H_2 production rate	Current and/or power density	Advantages	Remarks
Graphene [112]	Phosphorus-doped graphene quantum dots were incorporated into graphene hydrogel to prepare composite	- The hydrogel composite had dual role of photocatalyst and electrocatalyst to produce H_2	- Seawater and freshwater	-20.47 mmol/g.h	- Photocatalysis: 100 mW/cm^2 Electrocatalysis: 15 mA/cm^2	- Excellent photocatalytic activity - Excellent electrochemical HER and OER activity of hydrogel composite - Excellent stability	- Photocatalytic H_2 production using visible light absorption - Operable in real conditions - The hydrogel composite is a promising corrosion-resistant material
PVA + Chitosan [14]	- PVA and Chitosan solution were filled into photocatalyst-loaded delignified wood - This hybrid system endowed highly hydrated cellulose network with CdS-$MoSe_2$ photocatalyst	- Wood-based hydrogel served as a multifunctional energy conversion system for hydrogen generation and freshwater production	- Wastewater and seawater	-9.7 mmol/g.h	- Light source: 300 W Xe-lamp	- Simultaneous photocatalytic H_2 production and solar evaporation - Highly efficient - Highly improved - Intercepted VOCs to purify water	- Excellent energy efficiency - Practical applicability
Agarose [117]	TiO_2/Pt nanoparticles were added to agarose hydrogel via in-situ gelation method	- The hydrogel matrix held TiO_2 nanoparticles which acted as the catalyst during photocatalysis	- Heavy metal-containing solution	–	- Light source: 300 W Xe lamp	- Enhanced photocatalytic H_2 generation from metal-containing-industrial wastewater - Solved the catalyst poisoning effect caused by metal contamination	- The sulfurization treatment of absorbed heavy metals achieved synergistic enhancement of photocatalytic H_2 generation
Poly (Propylene Glycol) [20]	- PPG and hydrophilic polyurethane (HPU) were mixed to form a gel phase - The gel phase was swollen with water to form an HPU-PPG elastomer-hydrogel hybrid	- N_2 fixing bacteria was incorporated in the hybrid hydrogel, which can produce H_2 via N_2 fixation mechanism - The hydrogel floats above the water surface and produces H_2 from the water	- H_20	-104 mmol/h.m^2	- Uses solar energy	- Scalable, environmentally friendly solar H_2 production system - Very high H_2 production rate	- First demonstration of using N_2 fixing bacteria for solar H_2 generation
Fluorenyl methyloxy carbonyl-diphenylalanine (FmocLL) (FmoCFF) [118]	- FmocLL 3d hydrogel was formed by a self-assembling mechanism in an aqueous solution - [FeFe] Hydrogenase enzymes were encapsulated into the hydrogel	- Hydrogenase enzymes immobilized and uniformly dispersed in 3D hydrogel system provided enzyme-mediated H_2 production	- H_2O	–	-3.5 mA at -0.54 V	- Highly efficient enzymatic H_2 production	- Successful enzyme-mediated electrochemical H_2 production

the hydrolysis of $NaBH_4$ for enhanced performance also results in high production costs. Recent research efforts aim to address these challenges, with some focusing specifically on the use of hydrogels. Novel hydrogels have successfully addressed wide variety of limitations in hydrogen production, paving the way for innovative technologies that extend beyond laboratory-scale applications. However, employing hydrogel based hydrogen production systems in real-world conditions with long term scalable features still remains a limitation which can be subject to future research endeavors.

3.2. Osmotic energy harvesting hydrogels

Considering the rising energy crisis throughout the world, osmotic energy is regarded as one of the renewable energy sources. It is clean, renewable, and can be harvested from abundant sources [119]. Osmotic energy can be harvested from a gradient in salinity concentration, particularly in places where rivers and oceans converge. An estimation of as much as 2.6 TW available energy from salinity gradients in estuaries, which is equivalent to about 17 % of the global electricity demand [15]. Reverse electrodialysis (RED) technology is one of the techniques to effectively harness osmotic energy [119]. This technology employs ion-selective membranes to convert osmotic energy into electrical energy [15,119]. Hydrogel-based membranes can effectively work as ion-selective membranes in RED technologies. Table 5 summarizes the utilization of hydrogels and their mechanisms in osmotic energy harvesting technologies.

The development of osmotic energy harvesting technologies faces various operational challenges. Prior research endeavors have been

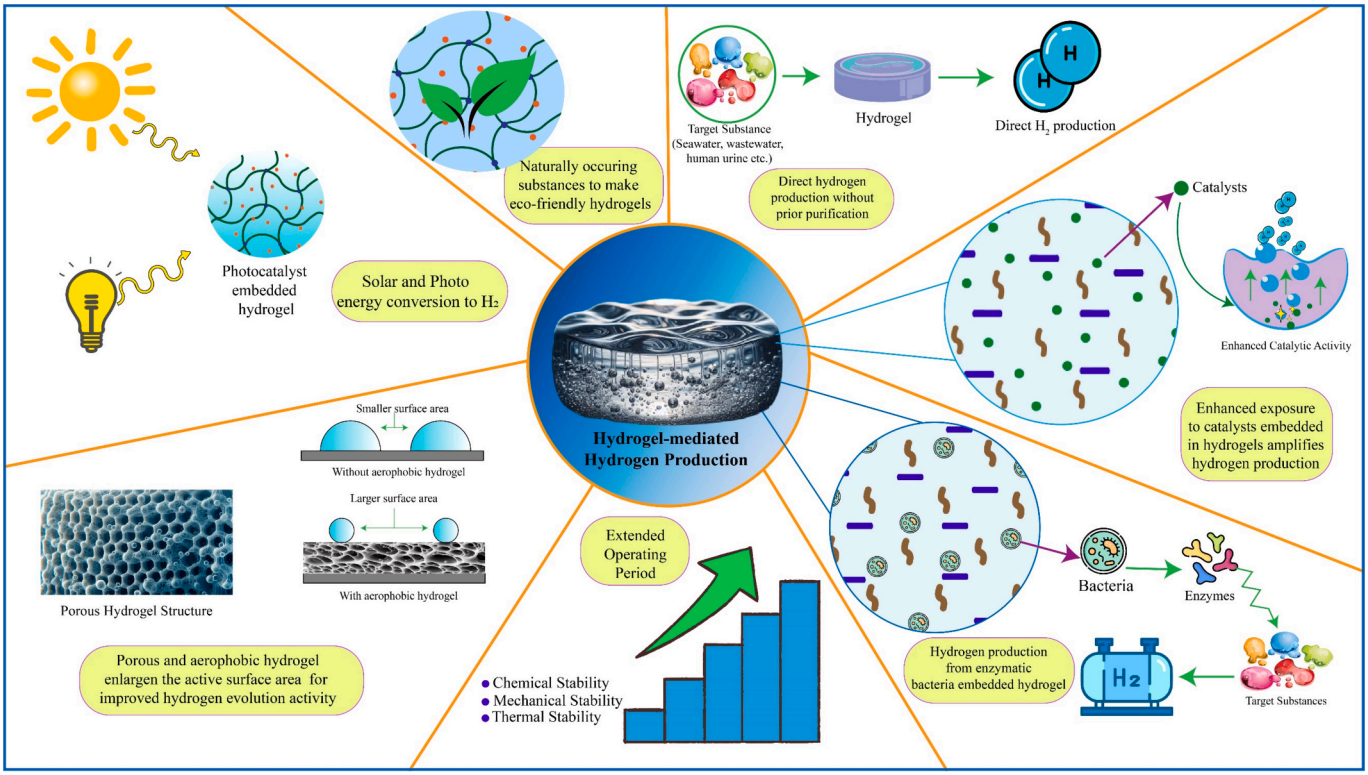

Fig. 3. Hydrogel-mediated hydrogen generation. Hydrogels serve as templates for catalysts and enzymes in hydrogen production reactions. Their larger active surface area dramatically improves the activity of catalysts, and the stability of hydrogels extends the operating period.

dedicated to effectively overcoming these obstacles, aiming to enhance the performance of osmotic energy harvesting systems. Among the potential solutions investigated, polyelectrolyte hydrogels emerge as promising candidates for augmenting the efficiency of osmotic energy conversion. This is attributed to their ability to offer rapid ion selectivity and transport capabilities. Gradient pore structure in hydrogels can be employed to induce a diode-like environment that directs ions in one direction [119] (Fig. 4a).

This modification of hydrogel can potentially be used to drastically reduce the biofouling of hydrogels (Fig. 4a). Hydrogels can be modified using stable substances like cellulose to enhance the mechanical stability of hydrogels for longer operating cycles. A recent study developed a biopolymer hydrogel derived from bacterial cellulose (BC) and acrylic acid-acrylamide (AAc-AAm) that demonstrated excellent cation selectivity while channeling ions to function as an osmotic generator [15]. The BC is a promising low-cost double network (DN) membrane. The functional groups present in a polymeric network of wood cellulose can be utilized to form a polyelectrolyte hydrogel. For instance, Chen and associates used hydroxyl (-OH) functional group wood cellulose in conjunction with carboxyl (-COO-) of PANa to form a stable polyelectrolyte hydrogel to generate osmotic energy [18]. The approach utilized by the researchers prevented excessive swelling of the hydrogel, thereby maintaining mechanical stability, and showcased a notable conversion efficiency of 25.5 %. Cellulose-derived hydrogels represent a cost-effective and environmentally friendly option within the aspect of hydrogel materials.

Two primary bottlenecks hinder the mechanism of energy harvesting from osmotic energy using polyelectrolyte hydrogels: reduced ion selectivity and excessive swelling of the hydrogel induced by high ion concentrations in saline water. However, modifications to hydrogels have proven effective in addressing these bottlenecks to some extent. A recent study focused on copolymerizing sulfopropyl acrylate potassium (SPAK) with phenoxyethyl acrylate (PEA) to create a novel type of hydrogel [120]. This research demonstrated superior salt-tolerant properties and excellent ion selectivity, offering promising solutions to overcome these challenges. Furthermore, this hydrogel had a power density (PD) of 16 W/m^2 showcasing its promise for practical applicability in osmotic power generators. Incorporating GO and CNTs into hydrogels has been demonstrated to enhance their structural integrity in salinity gradients, thereby sustaining power output even after repeated usage [121].

Recent research endeavors exploring hybrid osmotic power generators integrated with simultaneous solar evaporation or desalination have shown promising results in leveraging hydrogel utilization. Alginate hydrogel dispersed by carbon nanoparticles was employed to generate electricity in salinity gradients while synchronously evaporating water [50]. In a separate study, PAAm hydrogel was interpenetrated with 2-arylamido-2-methyl-1-propanesulfonic acid sodium salt (NaAMPS), introducing electronegative charges that facilitated the selective passage of ions in salinity gradients [49]. This innovative approach enabled the generation of electricity during nighttime while producing desalinated water during daytime operations (Fig. 4b).

3D hydrogels can induce a diode effect within the matrix that can be utilized to harness the osmotic energy. The first demonstration of 3D hydrogels for osmotic energy harvesting was done with agarose hydrogel [110]. A thermally stable Bis [2-(methacryloyoxy) ethyl] phosphate (BMAP) hydrogel was utilized for harnessing salinity gradient energy. This hydrogel featured 3D interconnected nanochannels, enabling the efficient utilization of thermal differences in water, including low-grade heat from industrial sources to harness salinity gradients. The resulting PD reached as high as 18.99 W/m^2, showcasing the effectiveness of this approach with an easy scale-up property [122].

Hydrogels can be modified to locally enhance the charge concentration to improve energy harvesting efficiency, facilitating faster energy extraction from salinity gradients [123]. Salinity gradients can be harnessed to generate power through an alternative mechanism involving hydrogel swelling and shrinking in response to changing salinity levels, which in turn drives a piston to generate mechanical

Table 5
Summary of osmotic energy harvesting hydrogels.

Hydrogels used	Techniques to employ hydrogels	Working mechanisms	Output	Benefits	Highlights
2-acrylamido-2-methyl propane sulfonic acid [119]	- A zwitterionic gradient double network hydrogel was synthesized by a two-step photo-polymerization process. - Two-step photopolymerization gave a continuous pore gradient to the hydrogels.	- The pore-gradient structure of the hydrogel induces diode-like ionic rectification, enabling a one-way flow of ions	- 5.44 W/m^2 at 50-fold NaCl gradient -49.6 W/m^2 at 500-fold NaCl Gradient	- Outstanding anti-biofouling property - Biocompatibility - Output PD exceeds the commercial benchmark	- The novel hydrogel membrane can potentially be used in wearable power harvesting devices
BC + Acrylic acid-co-acrylamide-co-methyl methacrylate [15]	- A precursor solution was prepared from AAc, AAm, methyl methacrylate (MMA), *N*, *N*′-methylenebis (MBAA) and diethoxyacetophenone (DEOP). - Precursor solution was poured onto BC hydrogel. - The composite hydrogel was passed through UV photopolymerization to get a 3D double network (DN) structure.	- The presence of acrylic acid in the hydrogel imparts a negative charge that develops excellent cation selectivity - The hydrogel functions as an ion transport channel that acts as an osmotic generator	- 7.63 W/m^2 at 50-fold salinity gradient (pH 11) - 45.5 W/m^2 (acid-base neutralization reaction) - 28.4 W/m^2 (wastewater-seawater mix)	- BC does not require additional complex separation, purification, mechanical and chemical treatment - Low-cost DN membrane	- Promises to construct high-performance nanofluidic devices from natural biopolymers
PANa [18]	- PANa hydrogel was infiltrated into wood vessel channels - The (-OH) of wood cellulose and (-COO-) of PANa engaged in hydrogen bonds to form a stable polyelectrolyte hydrogel.	- Confining the hydrogel within wood prevented it from over-swelling - Mechanically stable hydrogel showed high ion selectivity that enhanced the power output	- 8.5 W/m^2 at 50-fold concentration gradient	- High conversion efficiency of 25.5 % - High ionic selectivity - Low-cost and eco-friendly - Excellent mechanical strength and stability	- Excelled both membrane selectivity and internal resistance
Poly (sulfopropyl acrylate potassium) + Phenoxyethyl acrylate (SPAK-PEA) [120]	- The monomers of SPAK and PEA were added with crosslinkers to form a hydrogel. - The hydrogel was immersed in KCl solution.	- UV-induced copolymerization of SPAK and PEA imparted a gradient due to the presence of anionic monomers and aromatic monomers.	- 16.4 W/m^2	- Superior salt-tolerant property. - Ultrahigh PD. - Excellent cation selectivity. - Excellent anti-swelling performance.	- Highly efficient and stable osmotic energy conversion system with promise for practical application
Poly (acrylic acid-*co*-acrylamide) (PAAM) [121]	- GO and CNT were added to PAAM hydrogel by in situ radical solution polymerization method	- The addition of GO and CNT into the hydrogel enhanced the swelling ability and mechanical strength for efficient osmotic energy harvesting	- 7.07 J/g at 35 g/L NaCl solution	- Enhanced swelling property and mechanical strength	- Excellent reproducibility of hydrogel's osmotic energy recovery due to stable structural integrity
Alginate [50]	- Carbon black particles were dispersed into calcium alginate hydrogel. - Calcium alginate hydrogel was tightly compounded within nickel foam.	- The carbon black within the hydrogel matrix absorbed solar energy that induced evaporation. - The difference in salinity in a hydrogel matrix and seawater caused a salinity gradient for electricity generation.	- 5.3 W/m^2	- Synchronous solar evaporation and salinity gradient power generator	- A novel pathway to generate evaporation-induced salinity gradient energy.
PAAm [49]	- PAAm hydrogel was interpenetrated by NaAMPS. - The SO_3^- in NaAMPS imposes electronegative charges in hydrogel matrices that repel anions and transport cations.	- The ion selectivity of hydrogel imposes a salinity gradient redox reaction that is utilized to generate electricity	–	- Night-time salinity gradient energy harvesting with daytime solar desalination. - High evaporation rate of 2.9 Kg m^{-2} h^{-1} with 95.6 % efficiency.	- Demonstration of ionization engineering of hydrogels with effective utilization
Agarose [110]	- Agarose was added with PSS solutions from composite hydrogel. - The composite hydrogel was uniformly coated on the membrane	- Coating of 3D hydrogel on the aramid nanofiber membrane employed a stable ionic diode effect that facilitated the cation transport within the two layers	~ 5.06 W/m^2	- Excellent PD - Enhanced interfacial transport efficiency	- First use of 3d hydrogel interface for osmotic energy harvesting
(BMAP) [122]	- BMAP hydrogel has a self-crosslinking property that was initiated by 2,2-diethoxy aceto-phenone (DEAP). - No other monomers were added to ensure high-density space charge.	- 3D interconnected nanochannels with negative space charge efficiently weaken the concentration polarization for improved salinity gradient energy conversion.	- 18.99 W/m^2	- Thermally stable hydrogel - Anti-swelling property. - Enhanced energy conversion with thermal difference.	- Utilization of low-grade heat with salinity gradient. - Easy scale-up property enhances the possibility of practical application.

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Table 5 (*continued*)

Hydrogels used	Techniques to employ hydrogels	Working mechanisms	Output	Benefits	Highlights
(3-acrylamidopropyl) Trimethylammonium chloride (APTMA) [123]	- P(APTMA) hydrogel was placed at the surface of the APTES-modified anodic aluminum oxide (AAO) membrane	- The hydrogel on the surface of the AAO membrane served as positively charged layer that stored mobile counter anions that were attracted towards AAO surface. - This concentration gradient ions enabled more energy extraction	- ~220 mW/m^2	- Improved energy harvesting efficiency.	- A demonstration of local concentration enhancement by charge storage mechanism in the membranes.
PAA + poly (4-styrene sulfonic acid-*co*-maleic acid) sodium [24]	- AAc hydrogel was interpenetrated with poly (4-styrene sulfonic acid-*co*-maleic acid) sodium. - The hydrogels were mixed with different amounts of crosslinkers for different densities.	- The hydrogel was contained in a cylinder with piston which moved with the swelling and deswelling of the hydrogel.	- 36 J/cycle	- Converts entropic mixing energy of solutions to mechanical energy.	- A scaled-up osmotic engine that produces mechanical energy from salinity gradients.
PAA [23]	- A series of PAA hydrogels were developed by free radical polymerization with polyvinyl sulfonic acid (PVSA) and poly (4-styrene sulfonic acid-*co*-maleic acid) (PSSA).	- Introducing sulfonic acid improved the swelling ratio of the hydrogel for efficient energy conversion.	- 0.6 J/g (PAA) - 9.5 J/g (PAA/PVSA) - 13.3 J/g (PAA/PSSA-MA)	- Maintains swelling recovery for 30 repeated cycles. - Enhanced mechanical properties for efficient recovery of mechanical energy.	- A comparative study of a series of hydrogels for harvesting mechanical energy from salinity gradients.

energy [23,24] (Fig. 4c).

Osmotic energy was previously considered a non-viable domain for energy generation. However, the application of hydrogels, with their diverse modifications, has paved the way for effectively harnessing osmotic energy. Nanocomposite hydrogels paired with hybrid systems have demonstrated impressive ion selectivity, reduced internal resistance, and durable stability. Furthermore, membranes modified with hydrogels have exhibited performance that surpasses commercial benchmarks. Numerous research efforts have been undertaken to address the persistent challenges of utilizing engineered and modified hydrogels in salinity gradient energy harvesting. These challenges include excessive swelling, low ion permeability, low PD or output, biofouling, and mechanical and thermal instability. The results of these research initiatives show significant promise. Consequently, this domain of renewable energy harvesting holds the potential for transitioning into practical applications, revolutionizing the energy sector with the aid of hydrogels.

3.3. Bioenergy harvesting hydrogels

In the pursuit of sustainable energy, researchers are increasingly exploring innovative materials and technologies capable of efficiently harnessing renewable sources. Microbial fuel cell (MFC) represents a promising technology for tapping into biologically derived energy, which is chemically stored in organic matter, waste, wastewater, benthic mud, and other sources [124–126]. It is an innovative way of resource recovery and an environmentally friendly technology [125,127,128].

Nevertheless, the practical applicability of MFCs still encounters challenges. These hurdles are being potentially addressed with the introduction of novel hydrogels and their composites as building materials for MFCs. Table 6 summarizes the application of hydrogels in bioenergy harvesting. Miniature MFCs offer several advantages over their larger counterparts, including a larger surface area-to-volume ratio, reduced diffusion resistance, and quicker response times. However, miniature MFCs with air cathodes often face the challenge of evaporative water loss. This issue can be effectively addressed through the utilization of hydrogel, which mimics plant transpiration, as reported by Wu et al. [124]. The 3D structure of hydrogels facilitates the resolution of several limitations encountered in MFCs, such as diminished performance, decreased electrochemical activity, and reduced conductivity [25,125,126]. Biobased polymer like alginate, derived from brown algae, is one of the most promising materials for bioanodes of MFCs [125,128,129]. Modified sodium alginate (SA) presents a compelling alternative to other materials for electrodes in MFCs, given its enhanced environmental friendliness and biocompatibility. A recent study demonstrated the development of a composite hydrogel comprising polyaniline-sodium alginate-graphene oxide (PANI-SA-GO), offering synergistic advantages from diverse materials and achieving a high-PD (PD) [128]. In another recent study, SA hydrogel was employed to produce electricity from concentrated waste leachate [129]. The proposed MFC featured a modified hydrogel anode, which conferred adaptability to varying environmental conditions, particularly fluctuating salinity levels. This innovative design achieved an impressive PD of up to 289 W/m^3.

Graphene oxide (GO) nanosheets are a suitable candidate for MFCs for their high surface area [26]. Reports indicate that reduced graphene oxide (rGO) hydrogels featured biocompatibility, reduced resistance, and enhanced capacitance owing to their high surface area, facilitating microbial attachment in microbial fuel cells (MFCs) [26,107,130]. Simultaneous hydrogen production and microbial electricity generation utilizing GO hydrogel were reported [26]. Hydrogels can also be used as a medium to embed microbes within, which optimizes the performance of MFCs by the reduction of internal resistance [107]. A pseudocapacitive material polypyrrole (PPy), can be used to store electrical energy through a reversible redox reaction. Despite PPy's low solubility, its hydrogel form can be modified to enhance stability, rendering it a self-supporting conducting material suitable for MFCs [25]. Recent research has successfully generated electricity from simulated benthic sea mud as a source, employing MFCs equipped with modified PPy and polyaniline anode [126]. Extracellular electron transfer (EET) stands as a critical performance criterion for MFCs, where the microbes transfer electrons generated during their metabolic processes to an external electrode. Reports suggest that combining Mxene with PPy has emerged as a promising strategy to enhance the EET kinetics and PD of MFCs by improving the close contact of microbes with electroconductive materials [131]. Zhong et al. synthesized a PEDOT: PSS hydrogel incorporated with nitrogenated MXene (N-Mxene) to enhance EET kinetics and optimize the performance of MFC [132].

The integration of catalysts in MFCs substantially enhances their performance. Hydrogel matrices serve as suitable hosts for embedding catalysts, allowing for the maintenance of electrical conductivity and microbial activity [25,126,127]. MFCs can be made more cost-effective by substituting expensive catalysts with alternative catalyst-embedded hydrogels. The large specific surface area of these hydrogels facilitates

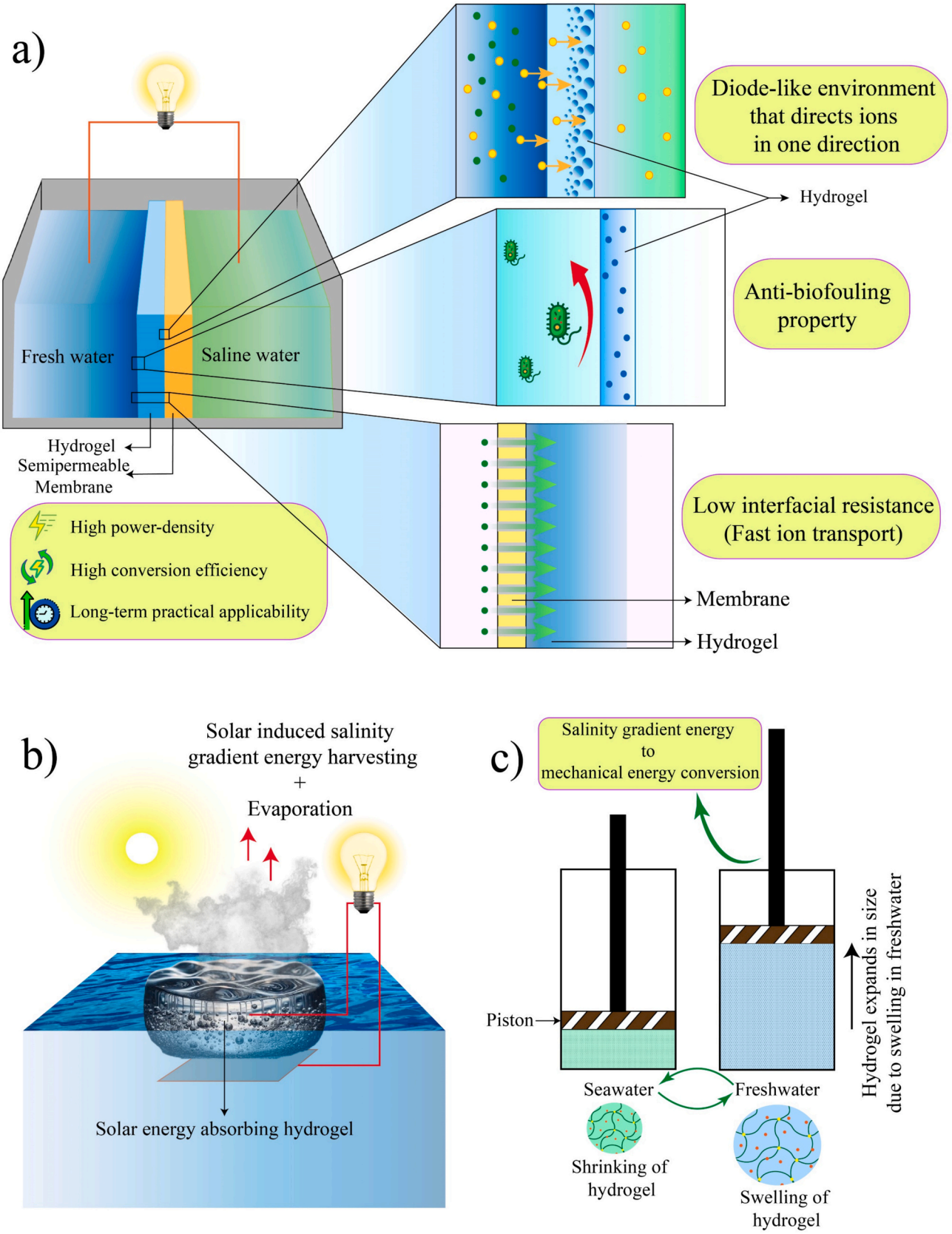

Fig. 4. Hydrogels for osmotic energy harvesting (a) hydrogels for electricity generation from salinity gradient. (b) solar-induced electricity generation coupled with evaporative water desalination using floating hydrogels. (c) salinity gradient to mechanical energy conversion by hydrogels.

Table 6
Summary of hydrogel applications in bioenergy harvesting.

Hydrogel types	Targeted bioenergy sources	Harvesting mechanisms	Energy output	Advantages	Remarks
PANa [124]	Pre-grown electroactive biofilm	- The hydrogel electrolyte is the conductor medium of generated electricity. - The hydrogel automatically draws water due to evaporative loss, thus maintaining a consistent auto-feeding.	- (PD): 1182 ± 115 mW/m^2 and 295.5 ± 28.8 W/m^3; -Voltage: 0.55 V	- Self-sustaining. - 3 MFCs can power an LED bulb - Portable MFC.	- A mimic of transpiration mechanism in plants.
Carboxymethyl Cellulose (CMC) [25]	Synthetic nutrient solution	- Hydrogel was doped with TiN and PPy which strengthened the hydrogel. - The 3D microporous structure of the hydrogel provided large surface for microorganism attachment.	-PD: 14.11 W/m^3; -Voltage: -0.3 V	- Improved energy output. - Improved biocompatibility. - Large surface area.	- First PPy-CMC-TiN carbon brush composite hydrogel.
PANI + PPy [126]	Sea mud and nutrient solution	- CNTs and Fe_3O_4 were added to the hydrogel. - Modified hydrogels were applied as anodes. - Microporous 3D structure of hydrogel provided attachment site for electroactive microbes.	-PD: 5901.49 mW/m^3; -Current density (CD): 0.170 mA/cm^2	- Improved electrochemical activity. - High biocompatibility. - Good PD.	- New insights into benthic microbial fuel cells (BMFC).
SA [125]	Synthetic nutrient solution	- Modified hydrogel was used as an anode in the MFC. - Hydrogel embedded by polyaniline demonstrated a highly conductive self-supporting 3D structure.	-PD: 515 mW/m^2; -Voltage: 0.783 V	- Enhanced performance. - Can function as bio-capacitors.	- New insights to improve efficiency and energy storage.
PANI + PANa + GO [128]	Synthetic nutrient solution	- PANI-SA-GO hydrogel composite was directly coated on the anode. - The hydrogel-modified anode obtained advantages of different materials.	-PD: 4970 mW/m^2; -CD: 4.66 A/m^2; -Stored Charge: 6378.41C/m^2	- Simultaneous power generation and energy storage capability. - Enhanced biomass content in anode.	- Environment-friendly hydrogel to enhance MFC's performance.
SA [129]	Waste leachate	- Bacteria embedded inner hydrogel layer provided better bio-electrochemical performance. - Highly crosslinked outer layer provided salt tolerance and mechanical stability.	-PD: 289 W/m^3;	- Improved adaptability to various environments - Enhanced energy storage capacity. - Better mechanical properties.	- Electricity production from waste leachate.
GO [26]	Xylose	- GO hydrogel was reduced via yeast to form Reduced 3D GO (rGO) hydrogel. - The rGO hydrogel was assembled on the surface of the carbon felt anode of the MFC. - Conductive and porous rGO induced more efficient charge transferring	-PD: 152 ± 2 mW/m^2; -CD: 301 ± 32 mA/m^2	- Simultaneous bioenergy harvesting and H_2 production. - High conductivity. - Enhanced cell loading. - Improved capacitance. - Reduced resistance.	- Novel strategy to improve the performance of MFCs.
GO+ PPy [107]	Lysogeny broth	- Bacteria-containing solution was mixed with hydrogel precursor which later self-assembled. - rGO was formed due to bacterial activity that demonstrated superior electrochemical activity.	-PD: 3366 ± 42 mW/m^2	- Extremely high-PD. - Biocompatible hydrogel. - Reduced resistance.	- A promising performance optimizer for bio-electrochemical systems
GO [130]	Lactate media	- The modification of rGO provided a large specific surface area with pores. - This large porous area served as the attachment site for bacteria and mass transfer.	-PD: 905.1 mW/m^2; -CD: 2.68 A/m^2	- Significant performance improvement. - Long-term stability.	- A stable, biocompatible anode material for practical application in MFCs.
Poly (acrylamide-co-acrylonitrile) [127]	Activated sludge + nutrient solution	- Fe-embedded hydrogel was pyrolyzed to form a catalyst containing carbon foam powder and used as a cathode in MFC - The hydrogel-derived cathode exhibited excellent conductivity.	-PD: 736.06 mW/m^2; CD: 132.04 mA/m^2	- Less expensive - Excellent ORR activity. - More stable. - Reduced resistance.	- A promising substitute of Pt-doped catalysts in MFCs
PPy + PAAm [131]	CH_3COONa	- Incorporation of MXene nanosheets into hydrogels for high power output and efficient EET kinetics. - The composite hydrogel provided enhanced mechanical and electric performance.	-PD: 6.71 W/m^3; -CD: 0.26 mA/cm^2	- Enhanced extracellular electron transfer (EET) - Enhanced microbial activity	- Promising high-PD material for MFCs.
PEDOT:PSS [132]	Synthetic nutrient solution	- Modified hydrogel composite exhibited porous morphology and larger specific surface area for better formation of biofilms.	-PD: 4.78 W/m^2; -CD: 7.83 A/m^2	- Enhanced electrocatalytic activity. - Improved efficiency of EET.	- A high-performance material for the anode in MFCs.

electron transfer pathways, thereby enhancing electrocatalytic activity and improving waste-to-energy conversion efficiency. Future research should focus on addressing the challenges of increasing the stability of hydrogels and preventing the accumulation of salts and metabolites to further enhance the performance and sustainability of MFCs. Additionally, research should place greater emphasis on the practical feasibility of these technologies. Hydrogels, with their diverse advantages, hold promise as suitable materials for MFCs. By introducing features such as

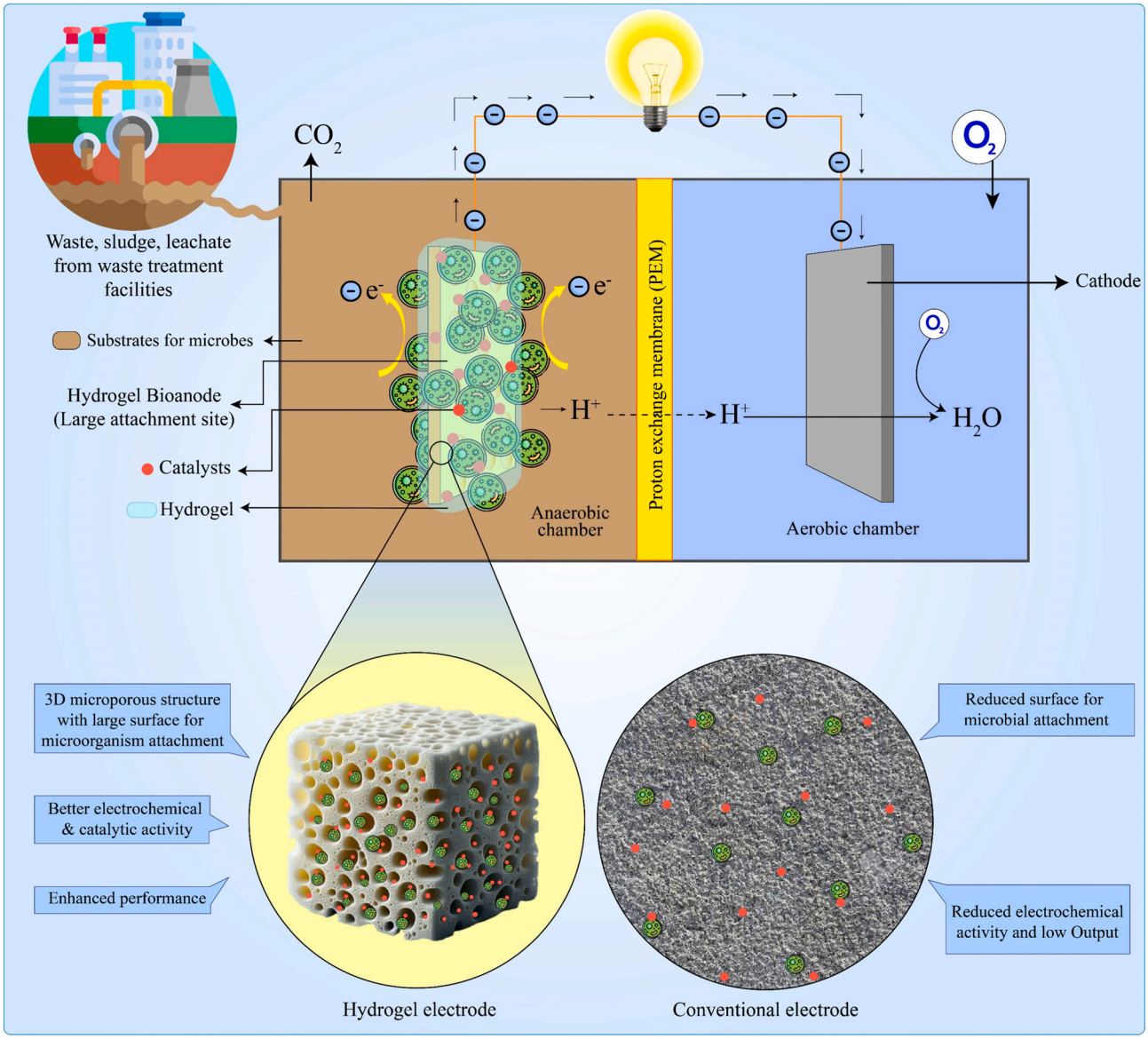

Fig. 5. Hydrogels in microbial fuel cell. Hydrogels serve as microbe-embedded bio-anodes, and their large active surface area facilitates electricity generation due to enhanced microbial activity.

high PD, enhanced energy storage capacity, and improved biochemical activities, hydrogels offer a potential solution to the practical applicability of MFCs (Fig. 5). This approach can pave the way for a waste-to-resource strategy, leveraging enhanced performance, improved stability, and cost-efficiency.

3.4. Hydrogel-based uranium extraction

Nuclear energy stands as a carbon-free energy source, offering reliability and high productivity. Globally, nuclear energy contributes to approximately 13 % of total power usage [133]. Uranium-235 (U_{235}) is the primary raw material of nuclear energy. Nonetheless, it is a non-renewable energy source and is limited in quantity, as it is unevenly dispersed in land-based sources [134]. But oceans contain over a hundred times more uranium with an estimation of 4.5 billion tons [22,133,134]. Among various uranium recovery approaches such as ion exchange, membrane electrodialysis, solvent extraction, co-precipitation, and photocatalysis, the adsorption technique is widely regarded as the most effective [22,133]. Hydrogels hold promise as adsorption materials for extracting uranium from seawater and nuclear wastewater to increase the availability of fuel for nuclear energy.

Despite its effectiveness, adsorption for uranium extraction still encounters bottlenecks such as biofouling, low and slow adsorption capacity, and interference by other ions [106,133,134]. Intense research efforts have been devoted to developing highly efficient hydrogel adsorbents aimed at overcoming these hurdles. Table 7 highlights the recent research endeavors for extracting uranium from seawater and nuclear wastewater.

One of the barriers to extracting uranium from seawater is the interference of vanadium ions, which attach to the active sites of hydrogels, thereby drastically reducing the available adsorption sites for uranium and consequently leading to reduced uranium adsorption capacity. This challenge has been effectively addressed by Fe-diffused polydopamine (PDA) endowed poly amidoxime (PAO) photothermal hydrogel (Fe@PDA-PAO) [134]. This hydrogel employs the absorption of solar radiation to elevate the temperature of the material. This temperature increase promotes the endothermic coordination of uranium, leading to selective adsorption of uranium while mitigating interference from thermoneutral adsorption of vanadium. In other studies, various other hydrogels were employed to increase the selectivity of uranium

Table 7
Summary of applications of hydrogels for uranium recovery.

Types of hydrogels	Used sources	Uranium adsorption capacity	Equilibrium adsorption time	Advantages
Fe@PDA-PAO [134]	- Natural seawater	- 12.31 mg g^{-1} (in light) - 9.42 0.32 mg g^{-1} (in dark)	- 20 min	- Excellent photothermal performance. - Selective extraction of U from seawater.
PVA + Bacterial debris [133]	- Natural seawater, - Simulated seawater	- Simulated Seawater: 134.07 mg g^{-1}; - Uranium solution: 263.38 mg g^{-1}; - Unfiltered seawater: 1.18 mg g^{-1}; - Filtered seawater: 1.31 mg g^{-1}	- Simulated seawater: 10 min - Unfiltered seawater: 28 h	- Excellent mechanical robustness. - Environment friendly. - Highly antibiofouling. - Suitable for the ocean environment.
Cs-PIDO [138]	- Natural Seawater, - Synthetic U solution	- 415 mg g^{-1} (maximum in synthetic solution) - 5.75 mg g^{-1} (in natural seawater) - Adsorption rate: 0.35 mg g^{-1} d^{-1} (1st 2 weeks)	- Synthetic solution: 20 min - Simulated seawater: 2 h	- Rapid and efficient U adsorption. - Outstanding regeneration performance. - Negligible performance loss in repeated cycles.
Soy protein isolate (SPI) [21]	- Simulated nuclear wastewater, - Natural seawater	- Simulated nuclear wastewater: 53.94 mg g^{-1} - Natural seawater: 5.29 mg g^{-1} - Simulated seawater: 139.45 mg g^{-1}	- Simulated seawater: 16 h - Natural seawater: 21 d	- Low-cost natural U adsorbent. - High U selectivity. - High salinity tolerance.
PAO/PMPC-SH [137]	- Natural seawater, - Synthetic U solution	- Natural seawater: 6.26 mg g^{-1} - Synthetic solution: 168.75 mg g^{-1}	- Natural seawater: 24 d - Synthetic Solution: 20 h	- Excellent resistance to salt shrinkage. - Excellent anti-biofouling property.
$PAOI_B$-PTC [136]	- Filtered Natural seawater - Synthetic solution	- 974.8 mg g^{-1} (maximum in synthetic solution) - Adsorption rate: 88.6 mg g^{-1} d^{-1} (synthetic solution) - 2.1 mg g^{-1} d^{-1} (natural seawater)	- Synthetic solution: 11 h	- Excellent salinity tolerance. - Highly selective U adsorption. - Excellent recycling performance.
$(GO\text{-}PHMB)_x$@Gelatin-PAAm [22]	- Simulated seawater	- 181.16 mg g^{-1} (theoretical capacity) - 625 mg g^{-1} (Maximum)	- 30 min	- Antifouling capability against bacteria and algae. - Excellently selective U extraction. - Excellent theoretical adsorption capacity.
AL-PNIPAM [106]	- Simulated seawater	- 285.5 mg g^{-1} (at 298 K) - 719.42 mg g^{-1} (maximum at 308.15 K)	- 240 min	- High U selectivity. - High adsorption capacity.
PAO + Gelatin Methacryloyl [135]	- Synthetic U solution	- 386.61 mg g^{-1} (at 298 K) - 540.85 mg g^{-1} (at 303 K) - 695.25 mg g^{-1} (at 308 K)	- 60 min	- Fast and selective U extraction. - High adsorption capacity. - Enhanced mechanical robustness. - Environment friendly.
PVA + CBCS [136]	- Synthetic U solution	- 496.049 mg g^{-1} (theoretical maximum at 288 K) - 457.824 mg g^{-1} (at 298 K) - 424.812 mg g^{-1} (at 308 K)	- > 3 h	- Excellent U selectivity. - Excellent adsorption. - Reusability.
PEI + Chitosan (PEI/CTS) [105]	- Synthetic wastewater	- 291.75 mg g^{-1}	- ~ 3 h	- Good selective adsorption. - High adsorption capacity. - Superior mechanical stability. - Easy preparation and eco-friendly.

adsorption from solution with other available ions. For instance, soy protein isolate (SPI) hydrogel [21], perylene tetra carboxylate crosslinked PAO hydrogel ($PAOI_B$-PTC) [11], poly (hexamethylene biguanide) hydrochloride modified GO doped gelatin acrylamide ($(GO\text{-}PHMB)x$@Gelatin-PAAm) hydrogel [22], aminated lignin-based PNIPAM (AL-PNIPAM) hydrogel [106], gelatin methacryloyl functionalized PAO (GMPAO) hydrogel [135], carbon nanotubes containing chitosan crosslinked PVA based hydrogel (PVA-CBCS) [136] etc., were employed to significantly enhance the selectivity of uranium adsorption (Fig. 6).

The equilibrium adsorption time, essential for achieving a balance between adsorption and desorption mechanisms, is paramount for effective uranium adsorption. It can be defined as the duration required for the adsorption process to reach a steady-state where no significant increase in uranium uptake occurs, marking the threshold of the uranium adsorption capacity [21,105]. A shorter time to reach equilibrium adsorption facilitates faster ion transfer. This duration can vary from a few minutes to weeks, depending on the environment and type of solution utilized to extract uranium from [21,105,136–138]. Hydrogel adsorbents offer the flexibility to modify the hydrogels to facilitate faster ion transfer. As natural seawater is endowed with salinity, and microorganisms, like bacteria and algae and hydrogel adsorbents, can be susceptible to salinity and biofouling. Several studies have been employed to develop hydrogels with high salinity tolerance and antibiofouling property, including PVA with bacterial debris [133], poly methacryloyloxyethyl phosphorylcholine crosslinked PAO hydrogel (PAO/PMPC-SH) [137], ($PAOI_B$-PTC) [11], and $(GO\text{-}PHMB)x$@Gelatin-PAM [22].

The reusability of hydrogel adsorbents is crucial, as it can significantly reduce the operational costs and complexity associated with uranium adsorption processes. Zhang and colleagues developed a novel chitosan-poly(imide dioxime) (Cs-PIDO) hydrogel that exhibited outstanding regeneration performance and incurred minimal performance losses across repeated cycles [138]. Modified PAO hydrogel and PVA hydrogel were also reported to have excellent recycling performance [11,136].

Recent research has investigated the applicability of hydrogels for recovering uranium from various solutions. To explore practical applicability, studies have utilized natural seawater, simulated seawater, nuclear wastewater, and synthetic seawater solutions. The results of these studies have demonstrated the excellent viability of hydrogels for uranium recovery. However, future research endeavors should focus on assessing the environmental impacts of deploying hydrogels for uranium extraction from seawater. Additionally, the economic viability and scalability of integrating hydrogel adsorption techniques into nuclear waste management should be thoroughly evaluated. Hydrogels represent a promising avenue for revolutionizing nuclear energy availability

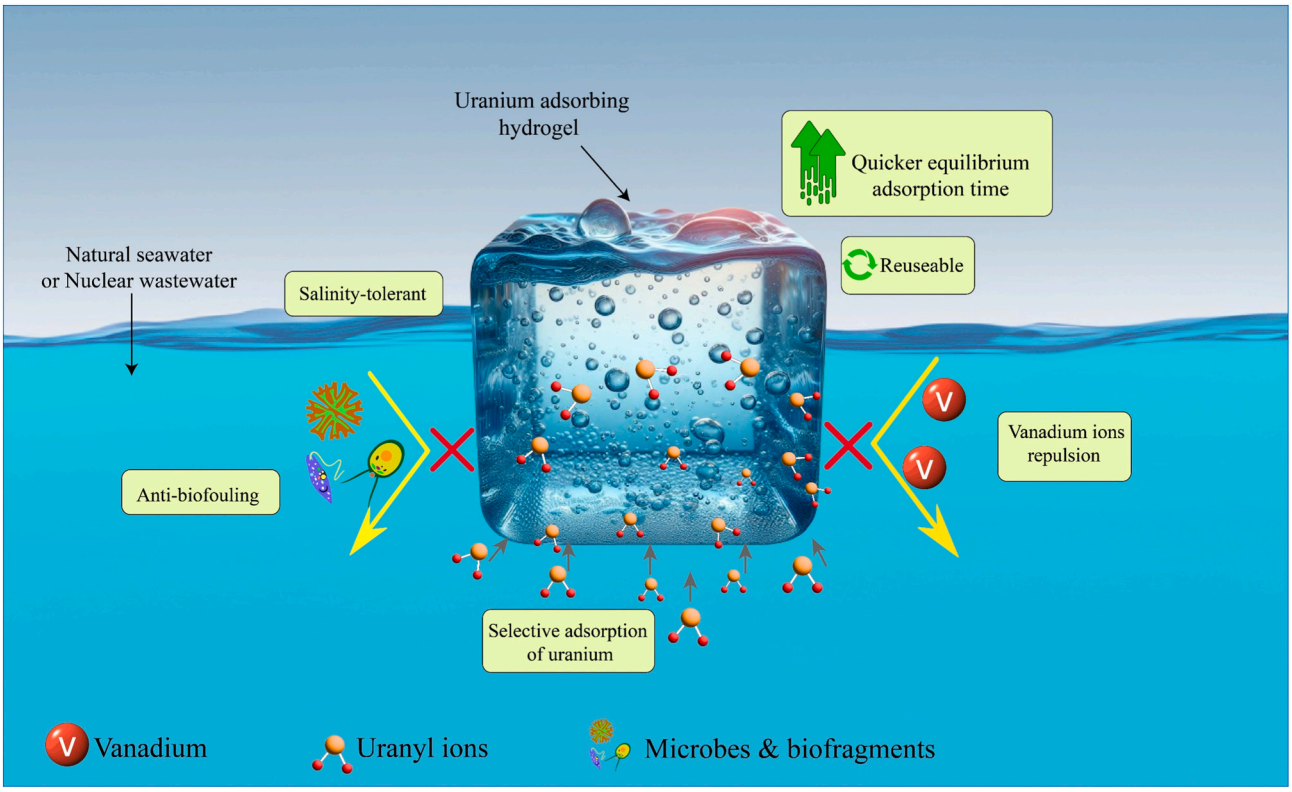

Fig. 6. Hydrogel-mediated uranium extraction. Hydrogels are excellent adsorbents for uranium recovery due to their superior selectivity. Modified hydrogels with salt tolerance and limited interference of vanadium ions prove to be highly efficient in uranium recovery.

on a global scale due to their cost-effectiveness, eco-friendliness, high adsorption rates, reusability, mechanical robustness, anti-biofouling properties, and salt tolerance.

3.5. *Hydrogels for thermal energy*

Approximately two-thirds of the total energy is dissipated as low-grade waste heat, typically below 200 °C, from various devices or machines, and industrial processes during their regular operation [111,139]. Indeed, in addition to devices or machines and industrial processes, the human body also generates heat, a portion of which is lost to the environment [27,140]. This waste heat can indeed be harnessed to develop sustainable green energy and improve the efficiency of devices and machines, particularly in response to the globally expanding demand for energy. Hydrogels, with their thermoelectric effects, hold promise for effectively harnessing this lost waste heat. The Seebeck effect observed in thermoelectric hydrogels results in the generation of an electric voltage in response to a temperature difference [111]. The Seebeck effect in thermoelectric hydrogels is the phenomenon wherein a temperature gradient induces a net ionic flux, resulting in an electrical potential difference between the hot and cold junctions of the hydrogel. This potential difference arises from the differential diffusion of ions due to their varying thermal energies at different temperatures [27,28,139]. The efficiency of thermal to electric conversion depends on the Seebeck coefficient (SCo), which is defined as the proportionality constant between the induced thermoelectric voltage and the temperature gradient. Table 8 summarizes the use of hydrogels in thermal energy while highlighting the advantages and drawbacks.

The thermoelectric generator (TEG) functions by converting heat energy within a temperature gradient into electrical power. Recent studies have indicated the potential practical application of hydrogels within TEG systems to efficiently recover energy from waste heat. These TEGs offer advantages such as self-healing properties, high mechanical strength, and excellent Seebeck effects [28,141,142]. Thermoelectric hydrogels can also be engineered for flexibility, offering the advantage of potential use in wearable electronic devices [27] and as human heat harvesters [140] (Fig. 7). Wu et al. utilized a printing strategy to fabricate highly efficient flexible thermoelectric hydrogels [27]. These flexible hydrogels demonstrated excellent mechanical stability.

The thermogalvanic effect, while based on a similar mechanism as the thermoelectric effect, exhibits slight differences. Thermogalvanic hydrogels engage in electrochemical reactions within temperature gradients to induce a potential difference across the electrodes. They have demonstrated considerable effectiveness in energy generation [111,139,143]. These hydrogels can also find application in reducing the heating of devices and machines through evaporative cooling. [139] Previous research endeavors have explored thermocapacitive storage, revealing the thermal energy storage capacity of hydrogels [111]. The composite hydrogel of polyethylene glycol (PEG) mixed with PVA and PAAm, AAc, and eicosane were also employed for thermal energy storage [111,144,145]. The effect of phase changing of PEG/PVA/PAAm hydrogel composite was utilized to store thermal energy within hydrogel [144].

Photothermal electricity generation represents another mechanism for harnessing thermal energy. Solar energy can be utilized to induce the photoelectric effect within hydrogels, resulting in the generation of electric current [146,147]. Modification of hydrogels implies efficient and stable photothermal conversion (Fig. 7). Light energy can alternatively be converted into thermal energy by storing it as latent heat in phase change hydrogels [148].

Hydrogels possess inherent properties for harnessing thermal energy, offering immense promise for utilization in the energy sector. This significant attribute holds the potential for more efficient and environmentally friendly approaches to resource recovery. However, challenges remain in harnessing waste heat to converting it into usable energy. Particularly, thermal cycling performance of thermal storage devices

Table 8
Hydrogel's application in thermal energy.

Technology	Hydrogels	Performances	Advantages	Drawbacks
Thermoelectric generator	PVA/PEDOT/Poly(2-acrylamido-2-methyl-1-propanesulfonic acid)PAMPS [28]	- SCo: -25.0 mV K^{-1} - Ionic power factor: 9.94 mW m^{-1} k^{-2} - Ionic figure of merit: 7.2 (RH 80 %, Room temp) - Thermovoltage of a prototype: -2.75 V ($\Delta T = 5.5$ K)	- Self-healing property. - High performance.	- Hygroscopic PAMPS can readily absorb water and hinder thermoelectric conversion.
	PEDOT: PSS/PVA [141]	- SCo: 787 μV K^{-1} - Voltage output: 138 mV	- Outstanding thermoelectric performance. - High tensile value.	- Limited conductivity and swelling stability.
	PAAm + CMC [142]	- SCo: 11.58 mV K^{-1} - Ionic conductivity: 18.4 mS cm^{-1} - Thermal conductivity: 0.47 Wm^{-1} k^{-1} - Ionic power factor: 198.2 mW m^{-1} K^{-2}	- Excellent tensile strength. - Eliminates electrolyte leakage.	- Hydrophilicity can cause excessive water uptake.
Flexible thermoelectric generator	PVP + CMC [27]	- Performance of 70 flexible thermoelectric couples: ~ 500 mV voltage ($\Delta T = 70$ K), PD: 1.278 W m^{-2} ($\Delta T = 50$ K) - Electrical conductivity: ~ 145.1 S cm^{-1} (n-type) ~ 734.6 S cm^{-1} (p-type)	- Applicable in flexible and wearable electronics. - Good SCo.	- Hygroscopic CMC can account for excessive water uptake.
Human heat harvester	PAAm + PEG [140]	- SCo: 1.43 mV K^{-1} - Retention rate: ~95 %	- Excellent mechanical stability. - Antifreezing property.	- Limited thermal conductivity.
Thermogalvanic hydrogel	PAAm [139]	- Cooling: 20 °C - Electricity retrieval: 5 μW at 2.2C discharging rate.	- Low-grade heat recovery. - Synchronous evaporative cooling and heat harvesting.	- Brittleness. - Toxicity concerns.
	PAAm + PVP [143]	- PD: 2227.5 $\mu Wm^{-2} K^{-2}$ - Thermopower: 4.5 mVK^{-1}	- Stretchable high-performance hydrogel. - Excellent thermoelectric properties.	- Long-term stability concerns.
Thermogalvanic power + Thermocapacitive storage	AAc [111]	- Thermogalvanic PD: 95 mW m^{-2} ($\Delta T = 20$ K) - Thermocapacitance: 220 F cm^{-2}	- Good thermogalvanic PD. - Excellent thermocapacitance. - Mechanical robustness.	- Potential to self-discharging.
Thermoelectro-chemical cells (TECs)	BC [9]	- SCo: 4.5 mV K^{-1} (n-type TEC) 0.72 mV K^{-1}(p-type TEC) - 3 pairs of TEC: 0.82 V voltage 4.5 μW peak power	- BC is commercially available. - Longer durability. - Practically scalable. - High mechanical strength.	- Degrades at temperatures higher than 200 °C
Thermal energy storage	PEG + PVA + PAAm [144]	- Thermal conductivity: 0.64 w m^{-1} K^{-1} - Photothermal conversion efficiency: 88.9 % - Phase change enthalpy: 124.8 J/g	- Excellent thermal energy storage capacity. - Reliable physical stability. - Impressive thermal management.	- Relatively high cost in scalability.
	Eicosane + CNT [145]	- Latent heat: 204.8 J/g - Conductivity: 620.3 S/m - Energy storage efficiency: 91.3 %	- Good thermal stability. - Good energy storage efficiency.	- Costly materials. - Potential nanoparticle release concerns.
Photothermal electricity	PAAm [146]	- Electricity production: 124 mW m^{-2} at 1 kW m^{-2} solar intensity - Water production: 0.84 kg m^{-2} day^{-1}	- Simultaneous electricity generation and water production.	- Limited light absorption range.
	PAAm + CMC + PEI [147]	- Power output: 6.67 mW m^{-2} - SCo: -1.40 mV K^{-1}	- Stable photothermal conversion - Good mechanical stability.	- Limited light absorption range.
Light-to-thermal conversion	AAm + AAc	- Latent heat: 202.4 J/g - Efficiency: 87.1 %	- Good thermal physical integrity. - High light-to-thermal conversion efficiency.	- Limited biocompatibility. - Susceptible to over-swelling.

which is caused by gradual decreasing of chemical stability, poses a major challenge. Furthermore, long term storage of thermal storage is challenging due to lack of proper insulation. Most of the current thermoelectric hydrogels have narrow operating temperature. Also, this technology still lacks in comparatively lower energy conversion efficiency. Improving the SCo of thermoelectric hydrogels can effectively address these challenges. The utilization of thermoelectric hydrogels has the potential to revolutionize the energy sector by enhancing the efficiency and environmental friendliness of everyday items, including electronics and devices, wearables, industrial processes, solar panels, solar evaporators, and building materials.

3.6. Mechanical energy harvesting hydrogels

Harnessing mechanical energy from various sources has emerged as a promising avenue in the quest for sustainable energy solutions.

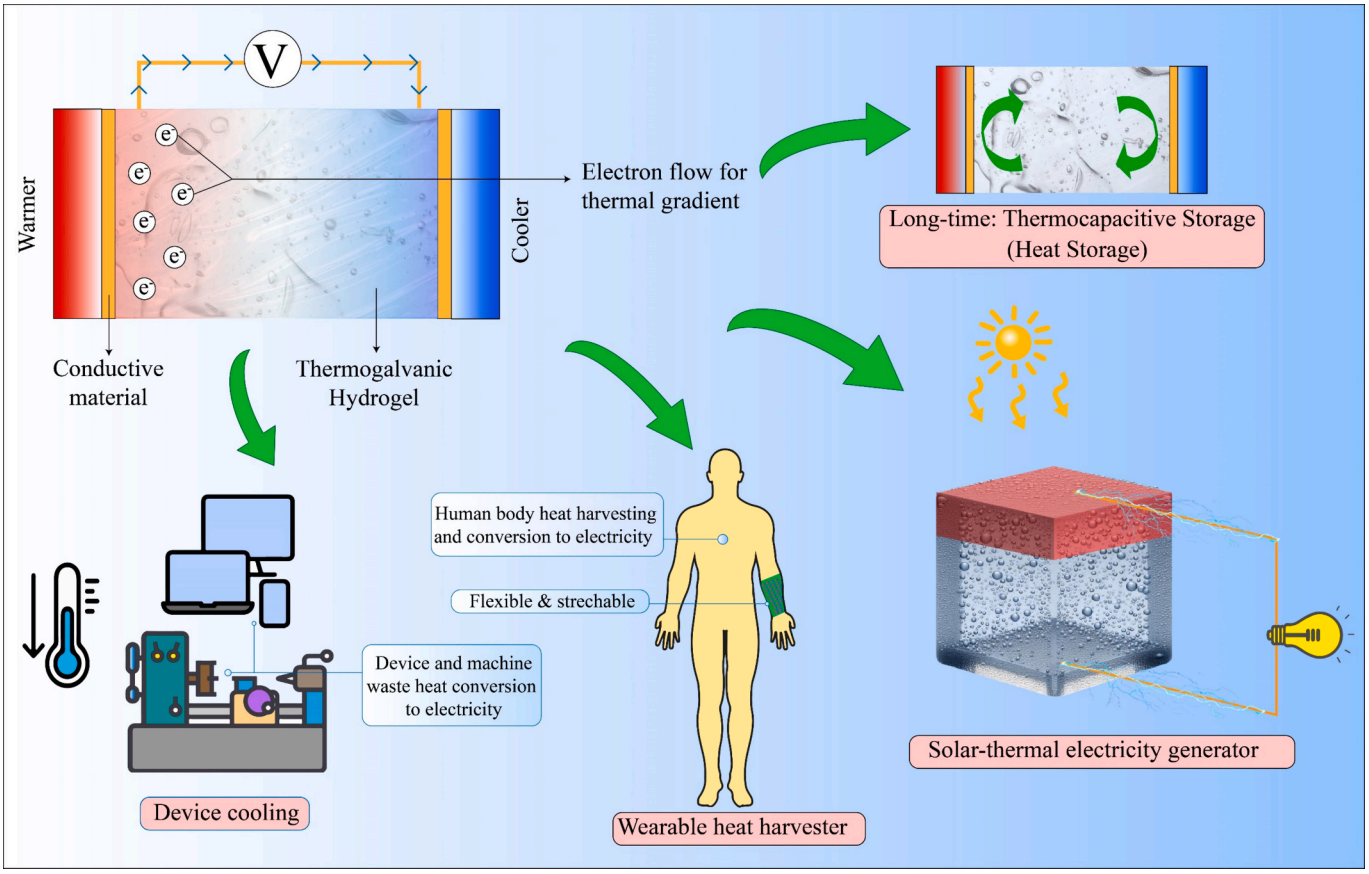

Fig. 7. Thermal energy harvesting by hydrogels. Under a temperature gradient, ions in hydrogels move from warmer to cooler regions, generating an electric current. This phenomenon is utilized to harvest electricity from the waste heat produced by devices, machines, and the human body.

Hydrogels have gained significant attention for their unique properties and versatility among the diverse materials explored for this purpose. Hydrogels are particularly convenient for their application in flexible wearable devices [149], electronic skins [150], smart electronic textile [151] etc., for their stretchability, transparency and stimuli-responsiveness. These hydrogels possess the capability to harness energy from the physical movement of the devices to which they are applied. As the pace of modern life evolves, wearable smart devices are becoming integral parts of daily life. These devices can become more efficient and capable of generating power with the introduction of mechanical energy-harnessing hydrogels.

Mechanical energy harvesting with hydrogels can be realized with technologies like triboelectric nanogenerators (TENGs) [31,149,152], piezoelectric nanogenerators (PENGs) [32,153], electrostictive hydrogels [154] and magnetostrictive hydrogels [109]. Table 9 presents a comprehensive overview of hydrogels utilized in mechanical energy harvesting, emphasizing their advantages and recent advancements.

TENGs generate electricity through charge transfer within materials. When hydrogels come into contact with other materials, they generate static charges as electrons are transferred. This static charge differential creates a potential difference, resulting in the generation of electric energy [152,155]. Hydrogels and their composites, such as PVA, PAAm, PEDOT: PSS, GO, Polydimethylsiloxane (PDMS), and cellulose nanofibers, are employed in triboelectric nanogenerators (TENGs) due to their ion-conducting properties and stretchable mechanical integrity [31,149–152,155–158]. Hydrogels offer distinct advantages over other deformable mechanical energy harvesting materials, including high efficiency, short response time, high sensitivity, compressibility, stretchability, durability, and versatility [159,160]. Additionally, hydrogels can be tuned for specific applications and possess promising self-healing abilities and eco-friendliness.

Piezoelectric nanogenerators (PENGs), on the other hand, induce a polarization effect in response to mechanical stress. This mechanically induced polarization leads to the generation of electrical potential, resulting in the production of electricity. Research endeavors have explored the viability of PENGs for mechanical energy harvesting by implementing them in various applications such as floors, wave energy conversion systems, shoe insoles, wearable devices, and body implants [32,108,153,161].

Electrostrictive hydrogels contain polar molecules that align or reorient themselves upon the application of an electrical field. This phenomenon results in the mechanical deformation of the hydrogel, which can subsequently be reversibly converted into electrical energy when the mechanical deformation is reversed. This kind of hydrogel is particularly applicable to self-powered sensors and self-powered actuators [154,162]. Magnetostrictive hydrogels undergo physical deformation when subjected to a magnetic field. Typically, these hydrogels are composite materials containing magnetic particles that respond to changes in magnetic fields [109]. Magnetostrictive hydrogels have not yet been established for energy harvesting applications, but they hold potential for use in mechanical energy harvesting systems.

Hydrogels, with their unique capability to convert mechanical stimuli into electrical energy, represent a promising frontier for a sustainable future. Mechanical energy harvesting using hydrogels presents challenges such as material durability, low energy output, and environmental sensitivity. Triboelectric, piezoelectric, magnetostrictive, and electrostrictive hydrogels each face specific issues like mechanical degradation, low efficiency, and complex synthesis. Future research should focus on developing advanced hydrogel composites, improving stability, enhancing energy conversion efficiency, and creating scalable manufacturing techniques that is viable with changing environmental conditions. Integration with wearable electronics and emphasizing

Table 9
Summary of mechanical energy harvesting hydrogels.

Harvesting mechanisms	Hydrogels used	Applications	Advantages	Recent Advancements
Triboelectric nanogenerator (TENG) [31,149–152,155–158]	- PVA/MXene - PVA/Agarose - PAAm/PEDOT: PSS/ gelatin - PAAm - GO - PDMS - CNF composites - Poly(ionic liquids) (PILs) for self-healing properties	- Wearable electronics (watches, fitness trackers). - Smart electronic textiles. - LED sensors. - Self-powered devices. - Shoe insoles for energy harvesting while walking. - Harvesting human motion energies. - Detecting human motions - Keyboard-integrated TENGs for powering portable devices. - Promising smart devices and artificial intelligence.	- High efficiency at low frequencies (can harvest energy from everyday movements). - Short response time. - High sensitivity. - Compressibility and stretchability. - Durability and reproducibility. - Versatile material selection for tailoring properties. - Can be integrated into various forms (fabric, thin films) for unobtrusive energy harvesting. - Promising in-vivo application. - Promising self-healing mode. - Promise to be eco-friendly.	- Development of self-healing TENG hydrogels with microscopic cracks that self-repair upon pressure, maintaining long-term performance. - Incorporation of liquid metal electrodes (e.g., gallium-based alloys) for stretchability and improved efficiency due to high electrical conductivity.
Piezoelectric nanogenerators (PENG) [32,108,153,162]	- PVA - PVA-PVDF - PAN - PEO - PAAm - PVP - SA - Barium titanate ($BaTiO_3$) nanoparticle hydrogel composites - PEDOT composites	- Shoe insoles for energy conversion from walking. - Pressure sensors for wearable health monitoring devices. - Implantable energy harvesting devices (pacemakers). - Wave energy conversion and solar interfacial vapor generation [108].	- High energy conversion efficiency from pressure or strain. - Biocompatible options available for in vivo applications. - Customizable functional layers. - Transparency. - Flexibility and stretchability. - Stimuli-responsiveness. - Biocompatibility and biodegradability.	- Improved biocompatibility of piezoelectric fillers like biocompatible polymers or natural materials (e.g., silk fibroin) for implantable devices. - Development of stretchable and transparent piezoelectric hydrogels using double network structures or by incorporating conductive fillers with high aspect ratios.
Electrostrictive Hydrogels [154,161,163]	- PAA - PVA - Ionic liquid incorporated hydrogels. - Stimuli-responsive hydrogels for controlled actuation	- Microfluidic devices with controlled movement for manipulating fluids (e.g., drug delivery) - Artificial muscles for soft robotics with biomimetic motions	- Large actuation strain for significant movement. - Biocompatible options available for in vivo applications	- Design of self-charging electrostrictive hydrogels using light as the energy source. Light irradiation triggers a photochemical reaction generating ions, leading to volume change and energy conversion. - Development of muscle-inspired hydrogels with enhanced actuation force by incorporating contractile nanofibers or by mimicking the hierarchical structure of natural muscles.
Magnetostrictive Hydrogels [109,162]	- Alginate - CNF - Fe_3O_4 or other magnetic nanoparticle composites. - DN hydrogels for enhanced mechanical strength	- Not yet widely established for energy harvesting, but has potential applications	- Potentially high energy conversion efficiency due to the magnetostrictive effect (material changes dimension in response to a magnetic field)	- Limited research on using hydrogels for magnetostrictive energy harvesting. However, studies on core-shell hydrogels with a magnetic core for targeted drug delivery offer promising strategies. These hydrogels can be responsive to an external magnetic field, allowing for controlled movement and potential energy conversion.

sustainability can expand their applications, making hydrogel-based technologies more viable and effective in energy fields.

3.7. *Hygro-gradient energy harvesting hydrogels*

A versatile strategy to harness energy from the environment is currently needed to address the energy crisis. One novel idea gaining widespread attention is the harnessing of hygroscopic gradient energy from the environment [29,164]. The moisture in the air can be harnessed to generate electricity through a moistureelectric generator (MEG) [165,166]. Hydrogels are utilized in MEGs to harness energy. MEGs convert the chemical potential energy induced by moisture gradients into electric energy (Fig. 8). This is based on the principle that different species of ions have varying effects on the properties of a solution. As the moisture level in the surrounding environment changes, the ions present in hydrogels may alter their solubility, resulting in a chemical potential that leads to the generation of electric energy [29,165–167]. Table 10 presents a comprehensive overview of hydrogels used in hygro-gradient energy harvesting.

PAAm stands as one of the most commonly utilized hydrogels for harnessing moisture-driven energy. PAAm has been modified with additives such as carbon black (CB), poly (acrylic acid-*co*-styrene sulfonate) (PAAS), lithium chloride (LiCl), and MXene to develop MEGs, as reported in previous studies [30,164,165]. In addition to individual usage, these MEGs are stacked in series to investigate their capability to power devices (Fig. 8). A composite hydrogel derived from (PAAm) incorporating MXene hydrogel has also been reported to assess its suitability for human skin moisture [30]. The MEGs are also deployed in indoor and simulated environments to evaluate their adaptability to changing atmospheric conditions and assess their applicability in real-world scenarios [29,166–169]. Hydrogel-based MEGs have practical applicability for their excellent performance, long-term stability, cost-efficiency and versatility.

The MEGs exhibit sufficient energy output to power electronic devices such as watches, smart helmets, calculators, self-powered sensors, and wearable electronics. The mechanism of energy harnessing using MEGs is straightforward and practical. With changing atmospheric conditions throughout the day, these electronic devices can self-power, eliminating the need for separate charging. However, maintaining sustained output performance and structural integrity over long periods in a wide range of atmospheric conditions remains a challenge that needs to be addressed. Future advancements in hygrogradient energy harvesting hydrogels must prioritize improving efficiency, durability, and adaptability to diverse environmental conditions to maximize their application potential.

The exploration of advanced hydrogel-based energy harvesting

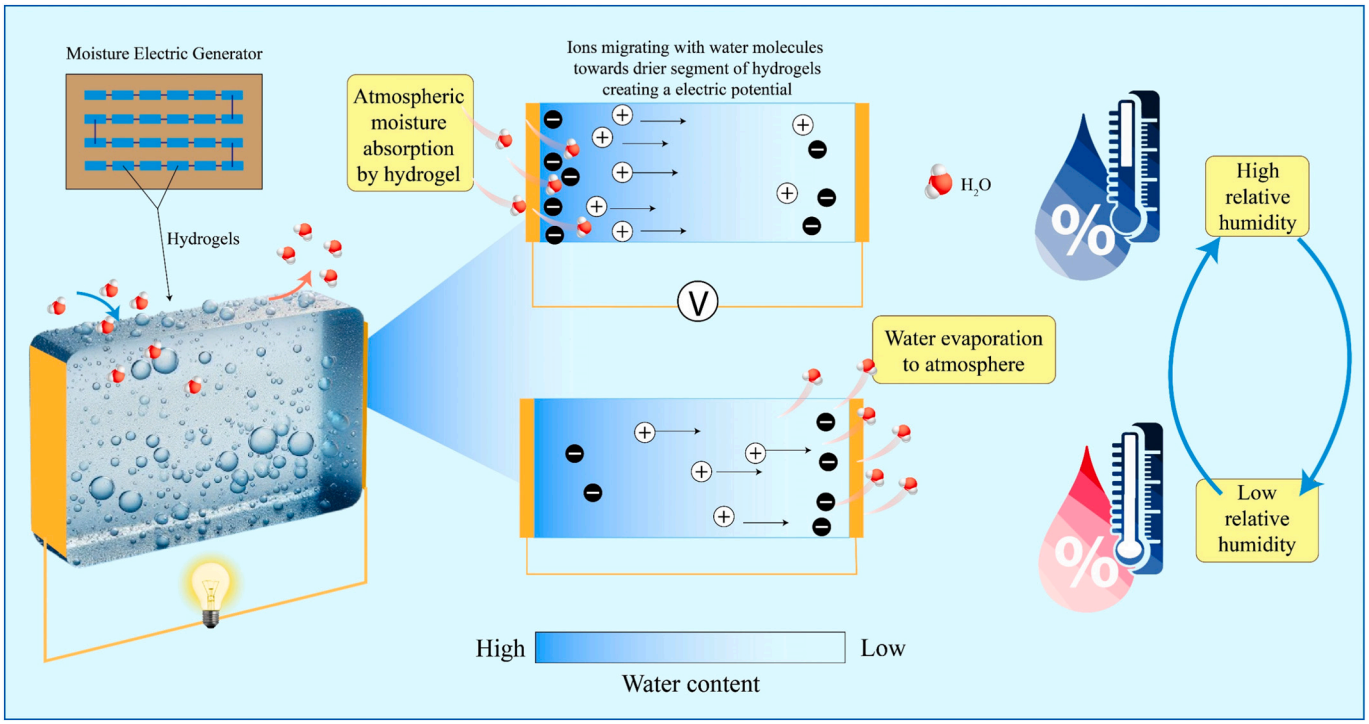

Fig. 8. Hydrogel in moisture electric generator (MEG) for hygro-gradient energy harvesting. Upon absorbing moisture from the atmosphere, water molecules and ions migrate to the drier end of the hydrogel, creating an electric current. This potential difference is utilized to generate electricity from atmospheric hygro-gradients.

technologies underscores their pivotal role in revolutionizing the energy sector. Hydrogels offer multifaceted solutions for sustainable energy generation from enabling hydrogen production to harnessing salinity gradients, bioenergy, uranium extraction, thermal, mechanical, and hygro-gradient energy. Their adaptable properties and impressive efficiency make hydrogels a cornerstone of the energy revolution. Continued research and development in this field promise to unlock even more innovative applications, further solidifying hydrogels' position as a key driver of the transition to renewable energy. With their potential to address pressing environmental challenges and meet growing energy demands, hydrogels represent a beacon of hope for a greener, more sustainable future. By leveraging the remarkable capabilities of hydrogels, we can pave the way towards a cleaner and more resilient energy landscape, benefitting both current and future generations.

4. Hydrogels in energy storage systems

Some unique characteristics make hydrogels highly significant and preferable in energy storage systems. For example, hydrogels boast high ionic conductivity and wettability, unlike conventional electrolytes. These properties facilitate intimate electrode contact and optimize ion transport paths [170]. This enhances the electrode-electrolyte interface area, which enhances the theoretical and practical capacity limitations in batteries and supercapacitors [171]. Some important and unique characteristics of hydrogels, such as their capacity to encapsulate and immobilize active materials within their matrices, prevent aggregation, maximize utilization, and enhance both the capacity and cycling stability make the storage devices more preferable and user-friendly [172]. Their inherent flexibility and self-healing properties further contribute to their effectiveness in various applications, marking them as versatile and advanced materials in the realm of energy storage [173–176]. Hydrogels based devices are enable to minimize electrode delamination and structural breakdown during charge/discharge cycles [173]. As a result, the storage devices are stable with a prolonged lifespan and sustained capacity retention, surpassing expectations and making them a perfect choice for various energy storage devices [171,177]. These multipronged approaches pave the way for next-generation energy storage systems with significantly increased capacities and enhanced efficiency.

4.1. Hydrogel-based batteries

Hydrogel-based batteries are one of the most innovative approaches that offer a sustainable solution [178,179] to the inherent limitations of traditional batteries such as lithium-ion, lead-acid, and nickel-metal hydride systems. Traditional batteries operate through electrochemical reactions where ions move between the anode and cathode via an electrolyte, facilitating energy storage and release [180,181]. Over time, repeated charge-discharge cycles degrade their capacity, reducing lifespan and efficiency having environmental concerns [182]. Besides, traditional batteries pose safety risks due to thermal runaway. Building instruments of these types of traditional batteries pose significant environmental challenges when they are degraded and disposed of, as many traditional batteries contain toxic substances that, if not managed properly, can contaminate the environment [183,184]. Table 11 provides a comparative overview of hydrogel-based and traditional batteries.

In contrast, the core of the hydrogel-based batteries lies in hydrogels, which are three-dimensional networks of polymer chains capable of retaining substantial amounts of water or other solvents. These hydrogels act as the electrolyte, providing a medium for ion transport between electrodes while maintaining mechanical stability [173]. In addition to being highly conductive, hydrogels are flexible, lightweight, and thermally stable, which enhances the overall safety and adaptability of the batteries [170]. Unlike traditional batteries, hydrogel-based systems often use non-toxic, biodegradable materials that minimize environmental impact during production and disposal [185–187].

The basic mechanism of hydrogel-based batteries revolves around the integration of advanced materials within the hydrogel matrix e.g.,

Table 10
Summary of hygro-gradient energy harvesting hydrogels.

Hydrogel materials	Target water vapor sources	Conversion methods	Performances	Advantages
PAAm + CB + LiCl [165]	- Atmospheric moisture	- Hofmeister effect	- Open-circuit voltage (OCV): 0.3 V (ambient environment) - Short-circuit current (SCC): 50 μA (ambient environment)	- Excellent output performance. - Excellent moisture capture capacity. - Quick ion transport. - Toughness and flexibility. - Can be easily scaled up. - Applicable in self-powered wearable electronics.
Poly (sodium-4-styrene sulfonate) (PAAS) + PAAm [164]	- Atmospheric moisture	- Engagement of dual ion with opposite charge.	- Power supply: 0.36 nW cm^{-2} and -50 mV, up to 40 h. - Voltage up to 400 mV by connecting 20 cells in series.	- Power delivery even under extremely wet conditions. - Can also harvest fresh water from the atmosphere. - Operates in a wide RH range (20-100 %). - Cost-effective and eco-friendly.
PAAm + MXene [30]	- Atmospheric moisture - Skin Moisture	- Ionic and proton transport	- OCV: 600 mV - SCC density: 1160 μA cm^{-2} - PD: 24.8 μW cm^{-2}	- High electric output. - Long-term stability. - Operable in wide temp range (-20 to 80 °C). - Operable in RH of 20-95 %.
PVA + Graphene [168]	- Indoor moisture	- Directional transport of ions through nanochannels.	- OCV: 0.34 V - SCC: 1 μA - Water capture rate: 1.32 mg cm^{-2} h^{-1} (at 98 % RH)	- Increased output performance. - Good output stability. - Excellent electrical conductivity. - Applicable for self-powered sensors and low-powered electronic devices.
TiO_2/Co [169]	- Indoor moisture	- Electric double-layer effect	- OCV: 0.95 V (20 °C and RH of 50–80 %) - Current: 0.1 mA	- Remarkable output performance. - Long-term stability. - Applicable for LED lights and calculators.
PVA + Phytic acid (PA) [29]	- Simulated moist air	- Electrochemical	- PD: 35 μW cm^{-2} - OCV: ~ 0.8 V - SCC density: 0.24 mA cm^{-2} - Stable DC current: 9 μA (over 20 h)	- Highly cost-efficient. - Fast ion transportation. - Good moisture-absorption capability. - Easy to scale up. - Applicable in a wide range of temp (-24 to 60 °C) and relative humidity-RH (10-85 %) - Sufficient energy generated to operate electronics like calculators, watches, etc.
PAAm + AMPS + LiCl [166]	- Simulated moist air	- Hofmeister effect	- OCV: 0.81 V - SCC density: 480 μA cm^{-2} - Ionic conductivity: 14.5 S/m,	- Good performance. - Highly stretchable and flexible. - Applicable for smart helmets, protective clothing, and respiration monitoring masks. - Operates in RH range 30-100 %
PAAm + PAMPS + LiCL [167]	- Simulated environment	- Electric double-layer effect	- At 98 % RH, 25 °C Temp: OCV: 1.25 V SCC: 300 mA cm^{-2} Output power: 71.35 mW cm^{-2} - At 50 % RH, -20 °C Temp: OCV: 1.11 V SCC: 15 mA cm^{-2}.	- High power output. - High surface charge density. - Antifreezing - Even applicable in electronics in extremely cold and high humidity.

nanoparticles, carbon nanotubes, or metal oxides can be embedded into the hydrogel to increase ionic conductivity and energy density [188]. The hydrogel structure allows efficient ion movement while preventing the issues associated with traditional liquid electrolytes, such as leakage or thermal instability [189]. Additionally, hydrogels can absorb stress and deformation, making these batteries highly durable under various mechanical and thermal conditions. As a result, hydrogel-based batteries exhibit longer lifespans and greater resistance to capacity degradation over time. These advancements make hydrogel-based batteries a compelling alternative to traditional systems [190,191]. They not only improve energy storage capabilities but also address critical issues such as environmental sustainability, safety, and cost-effectiveness. By eliminating the reliance on scarce materials and hazardous components, hydrogel-based batteries pave the way for a cleaner and more efficient energy storage future. Their potential applications range from wearable electronics and medical devices to large-scale energy storage solutions, proving their versatility and superiority over traditional battery technologies.

4.2. Hydrogel-based supercapacitors (HBSCs)

Energy storage can be achieved in various ways, including thermal, battery-based, electrochemical, and supercapacitor energy storage devices. Recently, supercapacitor-based energy storage devices have gained attention. This is because supercapacitors not only have a wide range of applications, such as in hybrid vehicles, smartphone components, and energy harvesting devices, but they also deliver superior performance [202]. Besides, supercapacitors are renowned for their rapid charge and discharge capabilities. Though traditional batteries are more prominent in this field they have several drawbacks that hinder their widespread adoption and limit their effectiveness in various applications [182] e.g., capacity degradation over time due to electrode material degradation and electrolyte decomposition, leading to reduced performance and lifespan, etc. [180,181]. Besides, they often suffer from limited life-cycle, safety concerns such as thermal runaway and electrolyte leakage, slow charging rates, and environmental impact from toxic materials and manufacturing processes [183,184]. These limitations create a demand for innovative solutions. However, HBSCs with some unique properties could be able to fix the challenges [203]. Table 12 provides a comprehensive summary of HSBCs with their advantages over traditional batteries.

Hydrogels, with their enhanced stability and resistance to degradation, self-healing and regenerative capabilities, improved safety features such as non-toxic and non-flammable materials, rapid charging rates,

Table 11
Comparative overview of hydrogel-based batteries.

Features	Traditional batteries	Hydrogel-Based batteries	Advantages of hydrogel-based batteries
Ionic Conductivity	Limited by solid electrolytes	High due to water-rich, porous hydrogel structures	Enhanced ion transport, faster charge-discharge cycles [180,181].
Flexibility	Rigid and inflexible	Soft, stretchable, and flexible	Suitable for wearable and flexible electronics [192].
Safety	Risk of flammability and leakage	Non-flammable, non-toxic	Improved safety, reduced hazard risks
Environmental Impact	Toxic materials and complex recycling processes	Biodegradable, renewable materials	Eco-friendly and sustainable [187,193].
Self-Healing Ability	Absent	Can autonomously repair mechanical damage	Longer lifespan, reduced maintenance needs [194–196].
Energy Density	Higher in traditional lithium-ion batteries	Moderate, with ongoing improvements	Potential for optimization with hybrid materials [197,198].
Temperature Stability	Performance affected at extreme temperatures	Stable across diverse temperature ranges	Reliable in various environmental conditions [199,200].
Cost of Production	High, with expensive materials and processes	Potentially cost-effective with scalable techniques	Economical large-scale production [187,193].
Applications	Limited to rigid devices	Wearables, biomedical devices, flexible electronics	Broader application scope [182].
Longevity	Degradation over repeated cycles	Prolonged lifespan due to self-healing and anti-fouling properties	Reliable for long-term use [201].

and reduced environmental footprint, are revolutionizing sustainable energy storage systems in the modern era of energy use [185–187]. Unlike traditional electrolytes, hydrogels display high ionic conductivity and wettability, ensuring a smooth interaction of contact and ion pathways as well as enhance energy density, enhancing stored energy for enduring power. [188] Hydrogels do more than just conduct electricity, they become like a protective agent for the active materials in electrodes, keeping them safe. This stops them from clumping together making the storage capacity and stability even better [7].

Moreover, their natural flexibility and ability to heal themselves protect electrodes from breaking down like sturdy shields, giving a supercapacitor a long life filled with strong energy storage [215]. Self-healing is a smart solution to combat mechanical damage and extend the lifespan as well as remain the significant energy and power density after self-healed (Fig. 9) [216]. Through dynamic interactions, the self-healing hydrogel electrolyte can effectively repair its structure after experiencing mechanical harm and restore its excellent electrochemical performance [185]. Besides, self-healing can shield the hydrogel electrolyte from mechanical harm, but the supercapacitor may still malfunction if it suffers from collapse or dehydration of the hydrogel electrolyte. Combined with excellent water retention, this guarantees improved physical stability even during heavy use (Fig. 10) [217]. Their intrinsic resilience capabilities contribute to improved heat stability, enabling effective performance across a broader temperature range and seamlessly adapting to the environmental rhythm [188,218,219].

The HBSCs are increasingly favored for energy storage due to their unique properties. Graphene-based hydrogels are exemplary for their outstanding capabilities, ultrahigh energy density, and durability [220]. Carbon-based hydrogels also excel with their remarkable mechanical strength, high electrical conductivity, specific surface area, and hierarchical structure [221]. Poly (2-acrylamido-2-methyl-1-propanesulfonic acid) (PAMPS) combined with ammonium molybdate demonstrates effective super capacitive performance, achieving a high capacitance of 698.8 F g^{-1} at 5 mV/s. Polyaniline (PANI), another standout, often combined with polyvinyl alcohol (PVA), reaches 237 mF/cm^2 at a current density of 0.5 mA/cm^2, showcasing excellent charge-discharge cycles. Poly(3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT) is notable for its high electromechanical properties, water dispersibility, and environmental stability. When cross-linked with $AlCl_3$, PEDOT hydrogels achieve a capacitance of 158 F g^{-1} at a scan rate of 50 mV/s with 84.9 % retention after 2000 cycles [222]. Chitosan, a biocompatible and biodegradable polymer, is gaining attention for its use in supercapacitors, achieving an aerial capacity of 1 mF/cm^2 at 1.8 mA/cm^2, withstanding over 10,000 cycles [223]. Sodium alginate hydrogels, doped with conductive polymers like polypyrrole, reduced graphene oxide, and Ag nanoparticles, offer excellent electrochemical performance [224]. Table 13 demonstrates some exceptional hydrogel materials, including their performances and capacitance.

Thus, HBSCs could revolutionize the future of energy storage with their remarkable advancements, addressing our next-generation energy challenges effectively. These HBSCs are not only feasible for large-scale production but are also environmentally friendly due to their sustainable fabrication materials. HBSCs offer unique benefits, including high flexibility, excellent conductivity, and biocompatibility, making them ideal for a variety of applications. As we seek sustainable and efficient energy solutions, HBSCs stand out as a promising technology that combines practicality with ecological responsibility. Despite significant progress in the development of HBSCs, several challenges remain. A major issue is selecting a suitable electrolyte that is nonflammable, noncorrosive, biocompatible, and safe, but hydrogels made from organic building blocks often fail to meet these criteria [222]. The major hydrogels used in HBSCs are lignin-based hydrogels, which face drawbacks like self-aggregation, low mechanical properties, and limited functionality [225]. Composite hydrogels are sometimes good players for HBSCs, but producing these composites is often lengthy, complex, and expensive [172]. Another challenge for hydrogels is peeling and loss of conductivity; flexing the device can cause cracks in the conductive track, and adhesion issues can lead to the hydrogel peeling off the substrate [226]. Polymer binders can address adhesion but also reduce conductivity. Hydrogels also face stability challenges, such as maintaining performance in high temperatures and ensuring oxidative stability, cyclic performance, and charge-discharge ability [222,225,226]. Although there has been substantial progress in developing flexible supercapacitors, enhancing their energy and power density remains an ongoing effort.

4.3. *Hydrogel electrodes in energy storage device*

Traditional electrodes commonly utilized in supercapacitors (SCs) consist of porous carbon, transition metal oxides, conductive polymers, and their combinations [227]. However, these conventional electrodes exhibit numerous drawbacks. Mechanical stress on these electrodes presents significant hurdles in storage devices, leading to issues like fracture, delamination, electrode deformation, and loss of contact, thereby compromising their structural integrity [228,229]. Moreover, limitations such as restricted energy density, slow charge/discharge rates, and a finite cycle life hinder overall device performance by reducing active surface area, impeding ion movement, and diminishing energy storage capacity [230,231]. Additionally, conventional flexible electrodes may impede the electrochemical performance of SCs due to reduced diffusion of the insulating polymer gel, inhibiting efficient contact between the electrodes and electrolytes [232,233]. These

Table 12
Summary of the advantages of hydrogel-based supercapacitors.

Current challenges	Traditional batteries	Solution provided by HBSCs	Recent advancements in HBSCs	Remarks
Capacity degradation	Electrode material degradation, electrolyte decomposition, and the formation of a solid-electrolyte interface (SEI) layer.	High porosity and mechanical robustness mitigate capacity degradation, resulting in prolonged cycle life and minimal capacity loss over numerous charge-discharge cycles.	Nanostructured hydrogel electrodes exhibit improved electrochemical performance and enhanced stability under prolonged cycling conditions [204–206].	- Long-lasting energy supply with prolonged cycle. - Enhanced durability and performance stability. -Reduced maintenance and replacement frequency.
Limited life-cycle	Experience limited life-cycle, with capacity loss occurring over repeated charge-discharge cycles.	Can autonomously repair mechanical damage, prolonging device lifespan and ensuring reliable operation over thousands of cycles.	Hybrid hydrogel-electrolyte systems with improved mechanical properties and electrochemical stability [190,191].	- High electrochemical performance. - Reliable operation in various conditions.
Safety concerns	Pose safety risks such as thermal runaway, short circuits, and electrolyte leakage, particularly those containing flammable or toxic materials.	HBSC offer improved safety features, including non-toxic, non-flammable, and biocompatible properties.	Implementation of self-extinguishing hydrogel electrolytes with improved fire-retardant properties, minimizing the risk of thermal runaway and enhancing device safety [193,207].	- Safety with thermal stability. - Reduced risk of hazardous incidents.
Slow charging	Exhibit slow charging rates, limiting their applicability in fast-charging scenarios.	Advanced electrolytes with higher ionic conductivity enable rapid ion transport, resulting in reduced charging times and improved energy efficiency.	Engineering of ion-selective hydrogel membranes and interfaces to facilitate rapid ion diffusion and charge transport [193,208].	- Quick ion transfer. - Improved energy efficiency.
Voltage limitations	Batteries have voltage limitations, restricting their application in high-voltage systems.	HBSC can operate over a wide voltage range, addressing voltage limitations associated with traditional batteries.	Development of multi-layered hydrogel architectures and electrode designs to extend the operational voltage window of supercapacitor devices [209,210].	- Extended voltage window. - Enhanced performance in high-voltage systems.
Cost	Expensive materials and complex manufacturing processes hinder widespread applications.	HBSC offers cost-effective production processes.	Manufacturing techniques such as 3D printing and roll-to-roll processing for the production of HBSC [187,193].	- Low cost for installation. - Easy to economical large-scale production.
Environmental impacts	Traditional batteries use toxic materials that contribute to environmental footprint.	HBSC reduces environmental footprint by using biocompatible and renewable materials.	Utilization of eco-friendly hydrogel precursors and bio-based additives [187,193].	- Environment friendly. - Reduced ecological impact.
Self-discharge	Batteries can experience self-discharge over time, leading to energy loss and reduced performance.	HBSC exhibits low self-discharge rates, contributing to enhanced energy retention and improved long-term stability.	Development of self-regenerating hydrogel materials with intrinsic anti-corrosion and anti-fouling properties [211,212].	- Self-healed capacity. - Consistent long-term performance.
Maintenance requirements	Batteries require regular maintenance.	Self-cleaning and self-healing mechanisms mitigate the need for frequent maintenance, ensuring prolonged device lifespan and consistent performance.	Integration of self-monitoring and diagnostics systems in supercapacitor devices [186,212].	- Affordable to operate. - Minimal intervention is needed.
Scalability	Complex manufacturing processes and limited availability of raw materials.	Standardized fabrication techniques and readily available precursors contribute to streamlined production and cost-effective scaling.	Integration of automated assembly and additive manufacturing technologies [181,183,213].	- Easy to manufacture. - Cost-effective large-scale deployment.
Flexibility and conformability	Rigid and inflexible.	HBSC offer flexible and conformable designs.	Development of stretchable and shape-memory hydrogel materials for supercapacitor electrodes and substrates [184,186,214].	- Flexible for use. - Suitable for wearable electronics. -Enhanced adaptability.
Efficiency	Efficiency losses due to internal resistance, heat generation, and chemical reactions.	Advanced electrode designs and optimized electrolyte formulations minimize internal resistance and parasitic reactions.	Integration of advanced materials and manufacturing techniques [176,180].	- Able to resist internal issues. - Reduced energy losses.

challenges underscore the necessity for advancements in electrode materials and design. Hydrogel electrodes represent a revolutionary breakthrough in the material science of batteries, offer a promising solution and hold immense promise for the future of energy storage. Table 14 summarizes the application of hydrogel electrodes [234]. Their inherent flexibility and resilience mitigate mechanical stress, ensuring long-term structural integrity [235]. Besides, hydrogels provide a larger surface area and efficient ion diffusion pathways, addressing limitations in energy density and charge/discharge rates [236–238]. Enhanced electrode-electrolyte contact improves electrochemical performance [239,240]. These advantages highlight the potential of hydrogel electrodes in revolutionizing SCs technology. Table 15 summarizes the advantages of hydrogel electrodes over traditional electrodes. Studies utilizing electrochemical impedance spectroscopy (EIS) and scanning electron microscopy (SEM) have revealed that hydrogels enhance electrodeposition on both cathodic and anodic surfaces, leading to the formation of electrodeposited polysaccharides that increase the electrode's surface area and flexibility [241,242]. This enhancement is particularly encouraging for applications in functional films and flexible devices [243]. Beyond their environmental sustainability, hydrogel electrodes significantly enhance overall battery performance [244,245]. Firstly, hydrogel electrodes represent a departure from conventional batteries that rely on rigid metal or carbon electrodes. Despite their soft, gel-like nature, these hydrogels exhibit unique properties that give rise to new possibilities [246].

Secondly, the polymer framework of hydrogel electrodes, primarily composed of polyethylene glycol (PEG) and polyacrylamide (PAAm), contributes to their excellent ionic conductivity and customizable

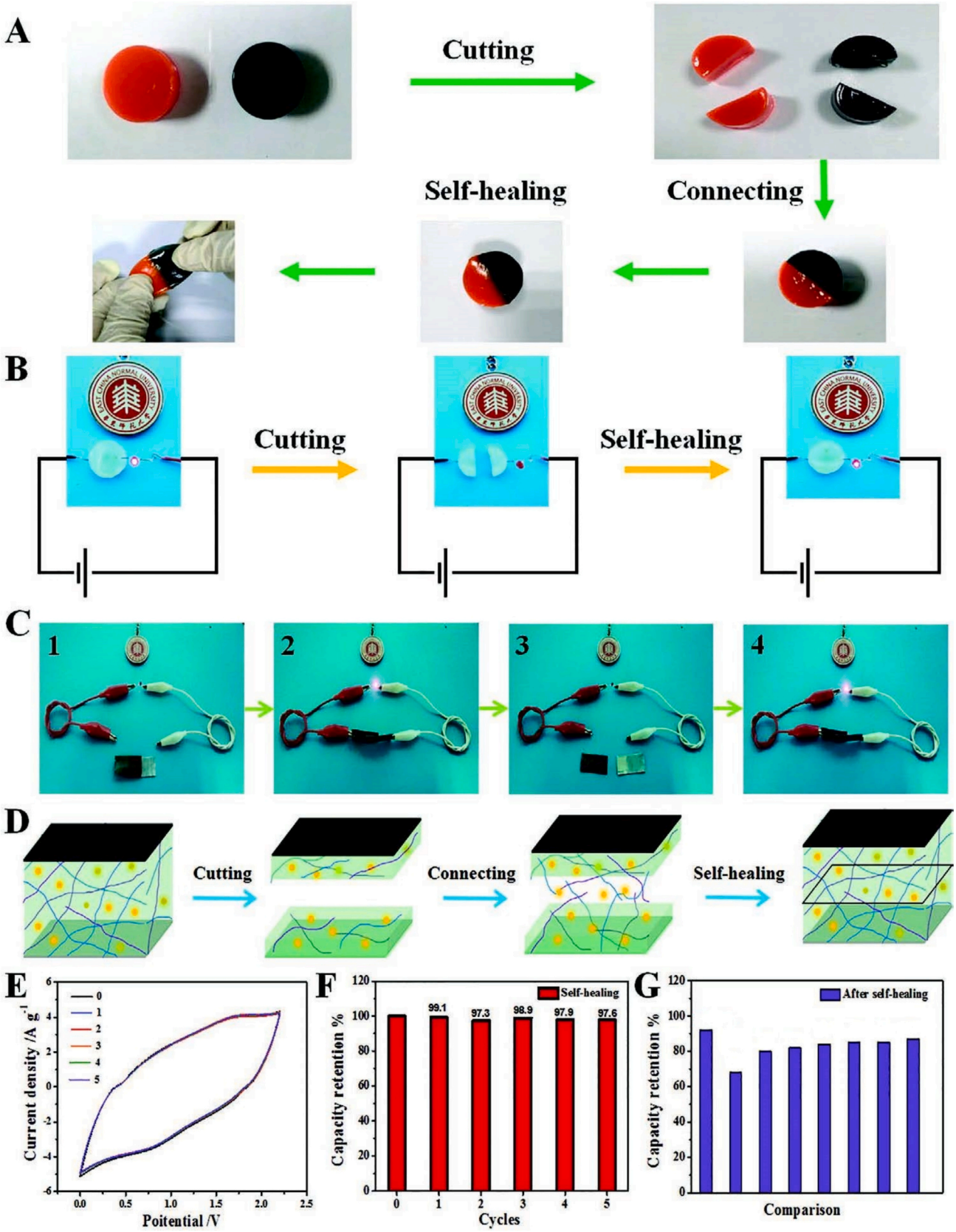

Fig. 9. A) Self-healing processes of hydrogels; B) Luminance properties of hydrogels after self-healing; C) Self-healing proof; D) Electrochemical characterization of 5-cutting self-healed hydrogels; E) Cell-voltage curves at 10 mVs^{-1}. F, G) Comparative study of different self-healed hydrogels in HBSCs [216].

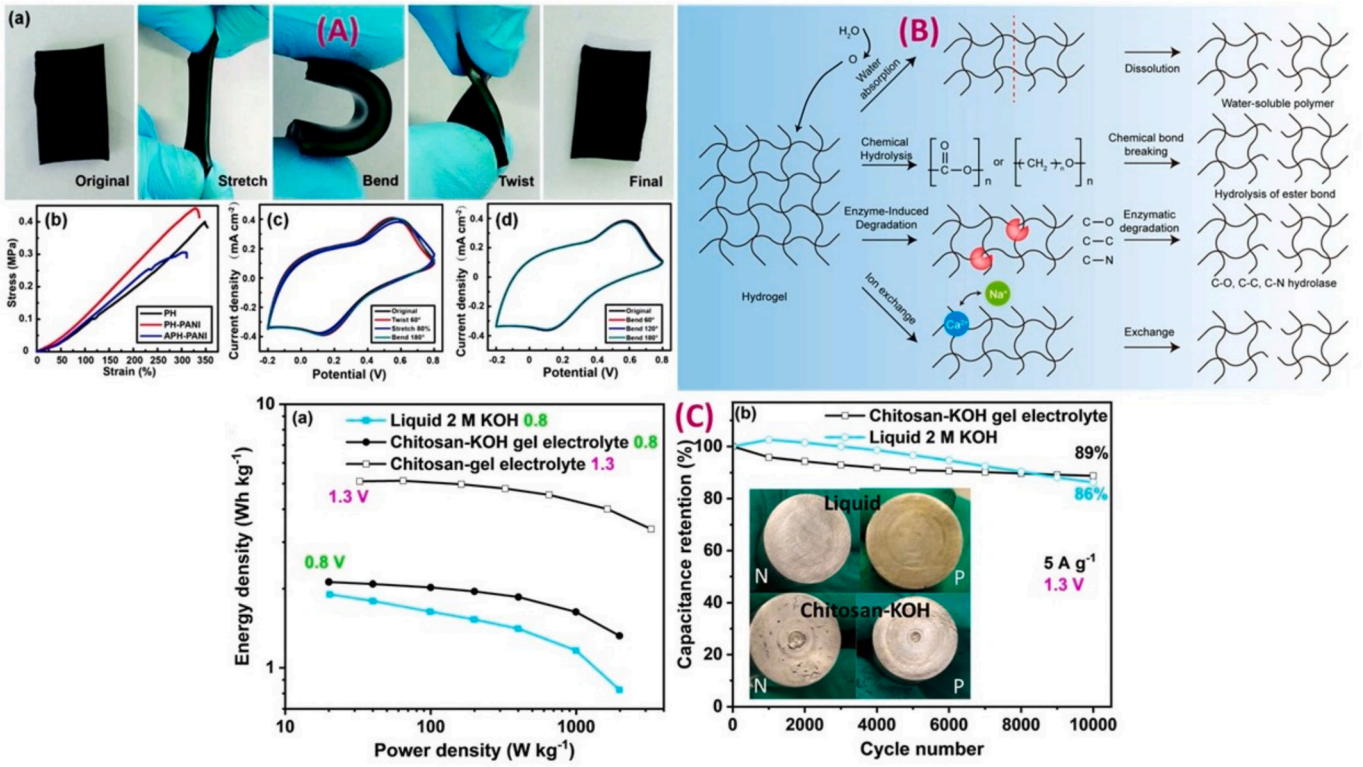

Fig. 10. A) Mechanical strength of hydrogels; B) Biodegradability mechanisms; C) Power and energy density enhancement [271–273].

Table 13
Exclusive hydrogels and their performance in HBSCs with their capacitance [222].

Hydrogel electrolytes	Electrodes	Capacitance	Remarks
PVA/H_2SO_4	$Ti_3C_2T_x$/Ag/nylon	328 mF cm^{-2}	-High ionic conductivity
PVA/H_2SO_4	PPy/PVA/H_2SO_4	13.06 F cm^{-2}	-Good mechanical properties
Al^{3+}/alginate/PAAm	PPy/carbon nano-tube paper	394.6 mF cm^{-2}	-Biocompatible
PVA/ H_2SO_4	PANI/PVA/ H_2SO_4	488 mF cm^{-2}	-Easy to synthesize
PVA/ H_2SO_4	PVA/PEDOT	128.9 mF cm^{-2}	-High ionic conductivity
BC/PAAm/ H_2SO_4	PANI/RGO	148 mF cm^{-2}	-Biocompatible
PVA-g-TMAC/KCl	PANI/RGO/polyester	546 mF cm^{-2}	-Good electrochemical stability
PVA/TA/H_3PO_4	PVA-g-TMAC/AC/AB/KCl	89.0 F g^{-1}	-High ionic conductivity
Borax/PVA/GO/KCl	PANI/carbon cloth	102.7 F g^{-1}	-Good mechanical properties
VHSNPs-PAA/H_2SO_4/ARS	PVA/PEDOT	119.2 F g^{-1}	-Biocompatible
PAD/ H_2SO_4	AC	248 F g^{-1}	-High ionic conductivity
PA/KOH	PANI/PAD/ H_2SO_4	430 mF cm^{-2}	-Biocompatible
F/MMT/PVA/ H_2SO_4	BC/RGO	221.3 F g^{-1}	-High mechanical strength
PVA/ H_2SO_4	Ti3C2Tx/ANF/ H_2SO_4	295 F cm^{-3}	-High ionic conductivity
EG/PVA/ H_2SO_4	RGO/PEDOT/ PVA	296.9 mF cm^{-2}	-Biocompatible

properties [264]. The hydrogel's porous structure emerges as a key aspect, enabling the accommodation of substantial amounts of active materials, such as lithium metal or metal oxides [265]. This characteristic directly correlates with an increase in theoretical capacity, a crucial metric for evaluating a battery's efficiency and effectiveness. Moreover, hydrogel electrolytes play a pivotal role in interacting with and stabilizing active materials like lithium and metal oxides ($LiFePO_4$ and $LiCoO_2$) [266]. This interaction occurs within the hydrogel environment, ensuring reliable battery performance and an extended life-cycle [267]. Matching the hydrogel electrolyte with the electrode and active material becomes imperative for efficient ion transport and the reduction of interfacial resistance [238]. Additionally, the polymer chemistry and composition of hydrogels can be finely tuned to accommodate specific active materials. This flexibility results in electrodes that are customized for desired battery characteristics and exhibit tunable physical appearances [268]. Their ability to bend and deform without compromising performance opens up possibilities for flexible and wearable electronics [243].

The modification of the hydrogel surface with conductive or stabilizing elements further improves electrode-electrolyte contact and enhances the adhesion of active materials. This modification contributes to prolonged life-cycle and capacity retention. Moreover, the inherent softness of hydrogels prevents electrode delamination and capacity loss during the volume changes those active materials undergo during charging and discharging [269]. To prevent the insufficient interfacial adhesion between electrode and electrolyte and the frozen hydrogel matrices at subzero temperatures, the incorporation of $ZnCl_2$ in hydrogel electrolyte prevents the freezing effect and develops the mechanical flexibility, which is fabricated in in-situ polymerizing onto hydrogel electrodes [267]. The capacitors are able to work with flexibility in 20-60^0C. Also, energy density and cyclic stability were very suitable.

Additionally, the presence of water within hydrogels reduces the risk of fire and thermal runaway, addressing a significant safety concern associated with conventional batteries [227]. The interconnected water

Table 14
Hydrogel electrodes for the energy storage system.

Hydrogel electrodes	Mechanisms of efficient energy storage	Advantages	Drawbacks
Cellulose nanofibril (CNF) hydrogel electrode [247].	Separators and electrolyte matrices	Promote efficient ion transport and exhibit good mechanical stability	Limited electronic conductivity.
The alginate-lithium metal composite electrode [248].	Prevention of dendrite growth	Improving safety and cycle life	Complex manufacturing processes
PEG-based gel polymer electrode (GPES) [249].	Facilitate rapid ion transport	High-rate cycling	Mechanical properties are less robust.
PAAm-graphene oxide composite electrode [250].	Enhancing electron transport	Promoting high-capacity lithium-sulfur batteries	Expensive and complex to produce
Cellulose- LiFePO4 composite electrode [251].	Hold LiFePO4 (Lithium Iron Phosphate) cathode particles	Offering good stability and long cycle life	Reducing performance
3D-printed alginate-carbon nanotube scaffold electrode [252].	Robust host for various active materials	High surface area, mechanical stability	Resource-intensive and costly
Self-healing polyacrylamide electrode [195].	Self-healing	Repair internal damage, extending battery lifespan and reliability	Not fully restore original properties
Hydrogel electrode with conductive interphases [253].	Improvement of electrode-electrolyte contact	Improved electrical conductivity	Challenging to manufacturing process
Microfluidic-patterned hydrogel electrode [254].	Controlled distribution of active materials	Optimizing battery performance	Precise fabrication techniques
Polyvinyl alcohol (PVA)-sulfur composite hydrogel electrode [257].	Enhances sulfur utilization	High capacity and energy density	Sulfur dissolution and loss during cycling
Chitosan-based hydrogel electrode [255].	Provides structural integrity and ionic pathways	Biodegradable and renewable material	Limited electronic conductivity
Polyacrylamide (PAAm)-CNT composite hydrogel electrode [256].	Facilitates electron and ion transport	High conductivity and mechanical flexibility	Potential environmental impact
Silica nanoparticle-reinforced hydrogel electrode [257].	Improves mechanical strength and stability	High surface area and robust structure	Complexity in synthesis
Agarose hydrogel electrode [258].	Provides a biocompatible matrix	Low cost and environmentally friendly	Limited electronic conductivity
Polyvinyl alcohol (PVA)-LiFePO4 composite hydrogel electrode [259].	Supports uniform dispersion of LiFePO4 particles	Enhanced cycle stability and safety	Possible mechanical degradation over time
Poly(3,4-ethylenedioxythiophene) (PEDOT)-PVA hydrogel electrode [260].	Conductive polymer matrix	High conductivity and flexibility	PEDOT synthesis can be costly and complex
Graphene hydrogel electrode [261].	Provides a highly conductive and porous structure	High surface area, excellent conductivity	Graphene production can be expensive
Cellulose acetate hydrogel electrode [262].	Acts as a separator and electrolyte matrix	Biodegradable and low-cost	Limited electronic conductivity
Hyaluronic acid hydrogel electrode [263].	Provides biocompatible and biodegradable matrix	Environmentally friendly and renewable	Limited electronic conductivity

channels within the hydrogel network further provide short diffusion pathways for ions, minimizing ionic transport limitations and enabling rapid charge-discharge cycles [243]. Additional components like cellulose nanofibrils and alginate further enhance biocompatibility and sustainability [270]. The inherent flexibility and stretchability of hydrogels make them ideal for powering flexible and implantable electronics [264].

While challenges remain, particularly concerning long-term stability and interface optimization, hydrogel electrodes represent a monumental leap in battery technologies. Their ability to seamlessly integrate enhanced capacity with sustainability and versatility paints a vibrant picture of a greener, more potent future for energy storage.

4.4. Hydrogels for energy and power density enhancement in storage devices

The HBSCs are a promising advancement in energy storage technology, offering a blend of high electrical power and high energy capacity [197,198]. They are highly regarded for their superior energy and power density and reliable cycling stability, making them top contenders for energy storage systems (Fig. 9) [283]. However, liquid electrolyte-metal electrodes and solid electrolyte-metal electrode supercapacitors are prone to some major issues, e.g., mechanical deformations, leakage, corrosion and mostly their moderate energy and power density with low voltage windows [284,285]. Hydrogels are the perfect candidate for energy and power density enhancement, as well as they can serve as both electrodes and electrolytes, offering advantages such as high ionic conductivity and resilience against mechanical deformation, which play a crucial role in enhancing overall performance [175,286]. Table 16 comprehensively summarizes hydrogel's application in energy and power density enhancement.

Active materials doping with hydrogel electrolytes and electrodes play a significant role in enhancing ion and electron transport efficiency, leading to improved energy density [175,283]. For instance, a hydrogel with activated microwave-expanded graphene oxide boasts a large surface area and efficient ion diffusion pathways, resulting in impressive energy and power density. Graphene oxide (GO) is also a promising electrode material due to its high surface area and excellent electrical conductivity [287]. The two-dimensional layered structure of GO promotes wettability and electron transportation ability between the electrolyte and electrode [288]. Hydrogel electrode-electrolyte e.g., conjugated polyelectrolyte (CPE) hydrogels, self-doped conjugated polyelectrolyte (CPE—K) hydrogel, cobalt sulfide/graphene composite hydrogel, MXene hydrogels, polyvinyl alcohol (PVA)/HQ/H_2SO_4, poly (3,4-ethylenedioxythiophene) (PEDOT) and active carbon materials with large surface areas have been extensively recorded with increase specific capacitance as well as energy and power density of SCs for energy storage devices [287,289–291]. In addition, the development of new hybrid hydrogels consisting of active materials and a polymeric matrix presents an opportunity to enhance SCs performance [292]. These hybrid hydrogels boast an inherent porous structure, excellent conductivity, and flexibility [175].

Enhancing the specific capacitance of electrode materials is another effective strategy to boost energy and power density [293]. Various carbon materials with large surface areas have been extensively studied for this purpose [174,294]. Additionally, innovative approaches, such as operating positive electrodes in a potassium iodide (KI) aqueous solution and negative electrodes in a potassium hydroxide (KOH) solution, show promise in expanding the working voltage of supercapacitors, have been reported to increase up to to 1.5 V, significantly improved the calculated

Table 15
Advantages of hydrogel electrodes over traditional electrodes.

Different metrics	Traditional electrodes	Hydrogel electrodes	Remarks
Structural composition	Traditional electrode materials mainly include porous carbon transition metal oxides, conductive polymers and composites thereof.	Conductive hydrogels are a hybrid material produced from a conductive polymer (CP) and a hydrogel, with a mechanical modulus more than three orders of magnitude below that of platinum (Pt), a conventional bioelectrode material [274,275].	-Hybrid structure with more than three orders of different conductive polymers.
Mechanical properties	Low ductility and brittleness, weight and bulk, high stiffness mismatch with soft tissues	Hydrogels boast remarkable elasticity and deformability, allowing them to conform to irregular surfaces and withstand large strains without compromising their functionality [199,200].	-Mechanically elastic and deformable
Physical stability	Being rigid and solid in nature and physical stability is very poor	Able to operate in bending, folding, twisting, and stretching conditions but also restore the original properties after damage [276,277].	-Able to bend, fold, twist, and stretch etc.
Functions	Difference in electrical potential between the metal electrode and its electrolyte interface	Hydrogels work by absorbing and storing ions from the surrounding electrolyte during charging and releasing them when the device is discharged. This process occurs at the interface between the hydrogel and the electrolyte solution, where ions are exchanged, allowing for rapid charge and discharge cycles and efficient energy storage [278,279].	-Create an interface between the hydrogel and the electrolyte solution
Electrical conductivity	Conductivity may depend on the material used (e.g., metal or carbon), and they may have a lower conductivity compared to hydrogel electrodes.	The hydrogel composition enhances electrical conductivity, ensuring better signal quality.	-Smart electric conductivity with ensure high storage quality
Charging-discharging capacity	Poor charging-discharging capacity	Sound and high charging-discharging capacity [274,277,280].	-Very high and perfect charge-discharge cycle
Electron-ion transduction	Suffers from high impedance, signal distortion, and poor voltage tolerance	Electron-ion transduction occurs as ions migrate through the hydrogel structure, facilitating the storage and release of electrical energy [199,279,281].	-Enhance charging quality as well as energy storage capacity
Self- healing property	Unable to self-healing	The abundant polymer chemistry and polymer engineering enable the integration of reversible dynamic bonds into conductive polymer hydrogels, leading to excellent mechanical stretchability and self-healing ability [194–196].	-Able to self-healed
Adhesive properties	They use adhesive materials or straps to secure the electrode to the skin.	The hydrogel itself serves as an adhesive, making it easier to attach and remove the electrode without additional adhesives [282].	-Make more robust for mechanical performance
Performances	Side reaction, weight loss provides poor electrical performance	Conductive hydrogels have porous microstructure, large surface area, superior conductivity, and excellent electrochemical activity [230,274].	-Very good conductivity, and excellent electrochemical activity.
Applications	Mainly in conventional batteries	Tissue engineering, environmental engineering, sensors biomedical devices, drug delivery systems, and flexible energy storage devices [235,274].	-Diverse applications for different advantages

energy density [287], thereby improving energy density. Solid-state electrolytes offer advantages over liquid electrolytes, simplifying assembly and enhancing performance [291].

However, achieving significant areal performance requires a large loading of active materials (typically exceeding 4 mg cm^{-2}) while ensuring high compactness of the electrode structure is crucial for achieving good volumetric energy density [295]. Optimizing the performance of supercapacitors involves fine-tuning the ionic conductivity of the electrolyte by adjusting the content of the lithium bromide salt [176]. In flexible zink hydrogel supercapacitor (ZHS) structures, the flexible polymer hydrogel electrolyte plays a pivotal role. Researchers like Peng et al. have investigated zwitterionic gel electrolytes for solid-state supercapacitors, achieving a volume capacitance of 300.8F cm^{-3} at 0.8 A cm^{-3} within a voltage range of 0–1.0 V [296]. These hydrogel electrolytes offer long cycling stability, excellent mechanical strength, and water retention capabilities, along with controlled ion migration to improve energy density. Similarly, Wang et al. have reported on a zwitterionic hydrogel supercapacitor with a high working voltage range of 0–2.1 V and an energy density of 26.5 $Whkg^{-1}$. The zwitterionic structure facilitates the formation of dynamic 'molecular cages' within the hydrogel, interacting with water molecules to minimize electrochemical activity and widen the operating voltage [296].

Solid-state electrolytes offer unique advantages for implementing electrolytes with different pH values compared to liquid-state electrolytes. Important hydrogels like PEG-PAMA-P(C6Ax-C7Ay-DPAz-DBAm) (EAASh), Poly(Di- methylamino ethyl methacrylate) (PDMAEMA), N-(Hydroxypropyl) Methacrylamide (HPMA), Polyethylene glycol Dimethacrylate (PEGDMA), Methacrylic acid (MMA), carboxymethyl chitosan (CMCh), and Hydroxy- ethyl- cellulose (HEC)/hyaluronic acid (HA), to name but a few, are such kind of hydrogel which are very responsive for different pH ranges and play the role for increasing energy and power density in energy storage devices [289,297]. Their stabilized forms make it easier to separate solid-state electrolytes during assembly into a SCs cell. Effective separation of the catholyte and anolyte is essential for achieving a wide voltage window and low self-discharge.

Thus, hydrogels present a solution due to their flexible network and high hydrophilicity, offering advantages like rapid charge-discharge rates and high energy and power density. Hybrid hydrogels and solid-state electrolytes further enhance SCs performance, offering long cycling stability and increased energy density. However, existing hydrogel electrolyte SCs face significant challenges and potential research can focus on shortcomings in improving electrochemical performance, including a limited working voltage range, low power density, energy density, and unstable life-cycle under high current conditions [284,298].

Efforts to overcome these limitations and improve the energy density of SCs are ongoing. Research is primarily focused on developing electrodes and electrolyte materials with diverse chemical compositions and structures, and in this matter, polymeric hydrogels could be one of the most suitable candidate [299].

4.5. Revolution in hydrogel-based energy storage systems

Hydrogel, a pivotal driver in the realm of energy storage devices which, has revolutionized the energy storage system with some amazing

Table 16
Summary of remarkable applications of hydrogels in enhancing power and energy density of energy storage devices.

Hydrogels	Energy density	Power density	Life cycle	Remarks	Challenges
Boron, nitrogen, and phosphorus ternary-doped holey graphene hydrogel (BNP-HGH) [300].	38.5 Wh/Kg	83 kWh/Kg	81.3 %	˙Stable physical stability as well as good energy density	-High production cost -Complex fabrication process
Nitrogen-doped graphene hydrogel (N-RGOH) [301].	94.5 Wh/Kg	640 Wh/Kg	87.0 %	˙Storing electrical energy. -Improving charge transfer kinetics and increasing the specific capacitance.	-Complex synthesis process -Potential environmental and health risks.
(PVA-Na_2SO_4) hydrogel [302].	13 Wh/Kg	106 Wh/Kg	78.0 %	-Efficient ion adsorption and desorption, enabling rapid charge-discharge cycles.	-Limited mechanical strength -Potential for ion leakage
(PVA-H_2SO_4) hydrogel [303].	6.5 Wh/Kg	287.4 Wh/Kg	75.0 %	-Rapid charge-discharge cycles -Increasing the specific capacitance	-Corrosive nature -Short cycle life
PVA-H_2SO_4-ARS (Alizarin red S) [304].	25 Wh/Kg	1581 Wh/Kg	88.0 %	˙Enhances the ionic conductivity of the hydrogel. -Improving charge transfer kinetics.	-Stability issues -Potential toxicity
PVA-H_2SO_4-KI-$VOSO_4$ hydrogel [305].	25.4 Wh/Kg	190 Wh/Kg	78.0 %	˙Effective ion adsorption and desorption. -Facilitating rapid charge-discharge cycles.	-Complex chemical interactions -Potential for ion leakage
PVA-PANI hydrogel [306,307].	18.7 Wh/Kg	107 Wh/Kg	86.0 %	˙Enhances the electrical conductivity -High specific capacitance and rate capability.	-Limited mechanical durability -Potential degradation of PANI
PVA-BC-LA-LiBr hydrogel [308].	16.3 Wh/Kg	932.6 Wh/Kg	93.4 %	-Mechanical strength. -Enhances flexibility. -Very good cyclic stability.	-Expensive and complex synthesis -Potential for ion leakage
SPMA-Zn: $ZnSO_4$/sodium alginate/polymethyl acrylic acid [309].	164.13 Wh/Kg	1283.44 Wh/Kg	95.3 %	-Structural integrity and flexibility. -High conductivity with very good cyclic stability.	-Limited availability -Potential environmental impact
PEGMA hydrogel [310].	356.6 Wh/Kg	2647.4 Wh/Kg	99.0 %	-Biocompatibility, flexibility, and high-water absorption capacity. -Very high energy density. -Self-healing capacity.	-High production cost -Potential for mechanical degradation
(H-ZHS) used zwitterionic natural polymer hydrogel [311].	286.6 Wh/Kg	220 Wh/Kg	95.4 %	-Resistance to fouling -Long-term stability.	-Limited scalability -Potential environmental impact
Zn-ion hydrogel with Fe^{3+} ionic cross-linked anionic copolymer formed by AMPSZn (2-acrylamido-2-methyl-1-propane sulfonate zinc) and AAZn (zinc acrylate) in the presence of $ZnCl_2$ [312].	205.3 Wh/Kg	1010 Wh/Kg	81.2 %	-Enhancing its mechanical strength and stability. -High energy and power density.	-Synthesis complexity
Polyaniline (PANI)/PVA/ATMP/CNTs (PPAC) composite hydrogel. [313]	12.8 Wh/Kg	125.0 Wh/Kg	77.31 %	-High conductivity -Improved electrical properties.	-Limited mechanical stability -Lower cyclic lifespan
Chitosan–KOH hydrogel [314].	5.1 Wh/Kg	32.5 Wh/Kg	86.0 %	-Good cyclic stability. -Biocompatibility and mechanical strength	-Low energy density -Limited scalability
3D defective RGO (DRGO) hydrogel [307].	85.23 Wh/Kg	960.31 Wh/Kg	98.86 %	- High surface area. -Very high-power density and cyclic rate.	-Complex fabrication process -Potential environmental impact
KCl-CH_2 = CH-SiO_2/PAAm hydrogel [315].	40 Wh/Kg	519 kWh/Kg	96.0 %	-Increasing swelling behavior. -Very good energy density.	-Limited mechanical strength -Potential for ion leakage
PVA/H_2SO_4/$FeCl_3.6H_2O$ (PHF) based redox-active polymer hydrogel [315].	120 Wh/Kg	4300 Wh/Kg	99.7 %	-Enhances the conductivity. -High cyclic stability.	-Corrosive nature -High-cost ingredients
Polysaccharide-enhanced hydrogel electrolyte (PBXHE: PVA/Borax/Xylan/Zn $(OTf)_2$) with OH-rich xylan [316].	82.1 Wh/Kg	193.7 Wh/Kg	90.0 %	-High hydrophilicity and improved ion conductivity. - High power density.	-Limited mechanical strength -Potential for ion leakage
Nitrogen and sulfur co-doped graphene hydrogel (N, S-GH) [317].	8.6 Wh/Kg	2400 Wh/Kg	89.4 %	-Surface area and enhanced catalytic activity. -Very good power density.	-Limited stability -Complex synthesis
Water-in-bisalt (WIBS)-soaked poly(acrylic acid) hydrogel [317].	30.1 Wh/Kg	1507 Wh/Kg	78.7 %	-Providing sustained hydration to the hydrogel matrix. -Very high-power density	-Stability issues with the water-in-salt solution -Potential for ion leakage

properties e.g., high surface area, interconnected porous structure, and tunable polymer chemistry, hydrogel achieves high specific capacitance in storage devices [200,318]. Some advanced techniques, such as synthesizing nanostructured hydrogels and incorporating nano-sized additives, can be employed for high ionic conductivity [319]. Besides, the Structural variation of hydrogels at the nanoscale and the introduction of nanoparticles into their composition significantly accelerate the charge-discharge rates of hydrogel-based energy storage systems [320]. This improves the efficiency of charge and discharge processes, making hydrogel-based systems even more effective. Energy density is a pivotal factor for energy storage devices; for the energy density enhancement of hydrogel-based energy storage devices, various strategies, e.g., incorporating conductive additives, doping with active materials, and engineering hierarchical structures, are taken into grant for the capacitance and energy density enhancement to meet specific application requirements.

Table 17
The key factors influencing the revolution in the energy storage system by hydrogels.

Revolutionary issues	Description	Applications	Advantages
High capacitance	Hydrogels have high capacitance due to their large surface area and porous structure [200,318].	- Mainly in supercapacitors.	- Shows very high energy storage capacity.
Fast charge and discharge rate	Facilitating fast charge and discharge rates in energy storage devices due to rapid ion transport [277].	- Supercapacitors, general batteries, and fuel cells.	- Rapid energy storage and release.
Long cyclic life	Hydrogel energy storage systems show strong durability and stability during extended use [348,349].	- Supercapacitors, general batteries, and fuel cells.	- Increased device lifespan with proper cell function.
Self-healing and self-regeneration	Hydrogels possess self-healing and self-regenerating abilities, extending device durability and reliability [321].	- Wearable electronics - Biomedical implants	- Autonomous repair of mechanical damage
High thermal stability	Ensure safe operation over a wide temperature range for energy storage devices [329,350].	- Electronics-automotive	- Safe operation under varying temperature conditions
Selective ion transport and separation	Hydrogel membranes selectively transport ions, useful for desalination and water purification [331].	- Desalination - Water purification	- Efficient and selective removal of specific ions or contaminants from water
Biocompatibility and biodegradability	Hydrogels are perfect for biomedicine and the environment because they're biocompatible and biodegradable [351,352].	- Implantable medical devices - Drug delivery systems	- Reduced risk of adverse reactions in biological systems
Flexibility and conformability	Hydrogels are ideal for wearable electronics due to their flexibility [280,353].	- Wearable electronics - Flexible electronics	- Lightweight and conformable energy storage solutions.
Multifunctionality and versatility	Hydrogels are versatile and used for energy storage, sensors, actuators, and drug delivery [281].	- Sensors - actuators - Drug delivery systems	- Integration of multiple functionalities into a single device
Scalability and cost-effectiveness	Hydrogels can be made in large quantities using cheap and widely available materials [354].	- All energy storage applications	- Economical production and scalability

Table 17 represents a comprehensive summary of the key factors of hydrogel that play the actual role in the revolution in the energy storage systems. Self-healing is an extraordinary property of hydrogels, allowing them to autonomously repair structural defects, cracks, and fractures [321]. When subjected to mechanical damage or deformation, hydrogels have the remarkable ability to undergo self-repair, ensuring that the device maintains its integrity and performance over time [322]. This means that even with prolonged use and wear and tear, hydrogel-based devices can continuously mend themselves, preserving their functionality and reliability. Additionally, the durability of hydrogels is another standout feature. Their robustness, coupled with the ability to withstand mechanical stress and environmental conditions, contributes to their extended life-cycle [323]. This durability ensures that hydrogel-based energy storage systems remain reliable and functional over an extended period [324]. Techniques such as surface modification, interface engineering, and encapsulation play a crucial role in mitigating degradation mechanisms in hydrogel-based energy storage devices [325,326]. By addressing issues such as instability at the electrode-electrolyte interface and dissolution of active materials, these strategies effectively prolong the life-cycle of such devices [327]. This ensures that hydrogel-based energy storage systems maintain their reliability and performance over the long-term. Thermal stability is another critical factor for energy storage devices and their users. Without adequate thermal stability, these systems may not last long, rendering them less competitive in the market [328].

Fortunately, hydrogels offer a solution to this issue. They exhibit stability within a suitable temperature range, making them the preferred option for energy storage applications [329]. The thermal stability of hydrogels prevents thermal runaway reactions and reduces the risk of device failure or degradation, even under high temperatures [330]. This characteristic enhances the reliability and longevity of hydrogel-based energy storage systems, further solidifying their appeal in the market. Hydrogels offer long-term reliability and safety, making them an excellent choice for energy storage applications. Selective ion transportation makes hydrogel more adaptable for users' choice; the selective ion transport mechanism enables hydrogel to efficiently remove specific ions or contaminants from water or electrolyte solutions [331]. Hydrogels's selectively remove targeted ions or contaminants and play a crucial role in various processes, such as improving water quality, facilitating resource recovery, desalination, water purification, and electrochemical separation [279,331]. This capability enhances the efficiency and effectiveness of these processes, contributing to environmental sustainability and resource management Fig. 10 [332]. The mechanical, biodegradability, energy and power density increasing techniques are presented in Fig. 10.

Recently, developed countries are scaling up hydrogel-based energy storage devices as well as different hydrogels for specific capacity enhancement; various fabrication techniques, e.g., solution casting, 3D printing, and roll-to-roll fabrication, enable efficient scaling of hydrogel-based energy storage technologies for mass production and deployment [333,334]. Additionally, common hydrogel precursors, including monomers and crosslinkers, are commercially available at a relatively low cost, and the synthesis processes are simple and scalable [335].

While hydrogel has revolutionized energy storage systems with some supercapacitances, it also presents several challenges. Mechanical stability is the most significant issue, as hydrogels often lack the necessary strength and durability, making them prone to deformation and mechanical failure under stress [336,337]. Additionally, their tendency to swell in the presence of water or other solvents can alter their physical properties and reduce the efficiency of energy storage devices [338]. Despite efforts to enhance conductivity, hydrogels still exhibit lower conductivity compared to traditional materials, which can limit their performance [339]. The complex and costly synthesis and fabrication processes, particularly for doped hydrogels, further complicate their widespread adoption [340]. Ion leakage is another concern, as it can diminish the efficiency and lifespan of hydrogel-based devices [341,342]. Environmental and health risks associated with certain hydrogels and their precursors pose additional challenges [343,344]. Limited thermal stability affects their performance in high-temperature environments, while scalability issues hinder their application in large-scale energy storage systems [337,345]. Long-term stability is also problematic, with hydrogels prone to degradation over time, impacting device performance [346]. Furthermore, complex interactions with electrolytes can lead to issues with ion transport and overall efficiency [347].

Despite these challenges, ongoing research and development aim to mitigate these drawbacks and enhance the applicability of hydrogels in energy storage systems, ensuring they remain a promising material for future energy innovations.

5. Hydrogels for energy efficiency and conservation

The hydrogel's revolution extends beyond energy production and storage systems. Hydrogels play a pivotal role in sustainable and efficient energy utilization. In previous sections, we highlighted the revolutionary impact of hydrogels on energy production and storage. Now, let's delve into their contribution to energy efficiency and conservation, particularly in housing systems. The housing systems are significant

contributors to global energy consumption, accounting for 40 % of the total, and HVAC systems are responsible for half of this usage [355]. In HVAC systems, air-cooling systems alone account for 25 % of the total HVAC energy consumption [356,357]. To transform our energy landscape, we must adopt higher energy efficiency and conservation technologies within housing systems. A paradigm shift towards hydrogel-based building materials can significantly reduce HVAC energy demands, marking a decisive step to sustainable building practices. Through the adoption of revolutionary hydrogels in making high energy-efficient housing technology, we can ensure the best energy conservation and efficiency practice over half of the generated energy.

Along with enhancing energy conservation and efficiency, hydrogel-based building materials can be very effective in reducing GHGs release of housing HVAC systems. Housing systems contributors nearly 40 % of the global total CO_2 emissions [358,359], where air-cooling systems alone contribute 7 % to global GHGs emissions [356,357]. By incorporating hydrogels into building materials, we can significantly reduce the reliance on GHG-producing technologies. Although hydrogel itself is a non-GHG-generating material, there is currently no scientific evidence of GHG generation from hydrogels. Consequently, it emerges as a promising candidate for adoption in building technologies aimed at achieving higher energy efficiency and conservation.

In this section, we explore into the energy efficiency and conservation performance of hydrogels when integrated into building' s windows

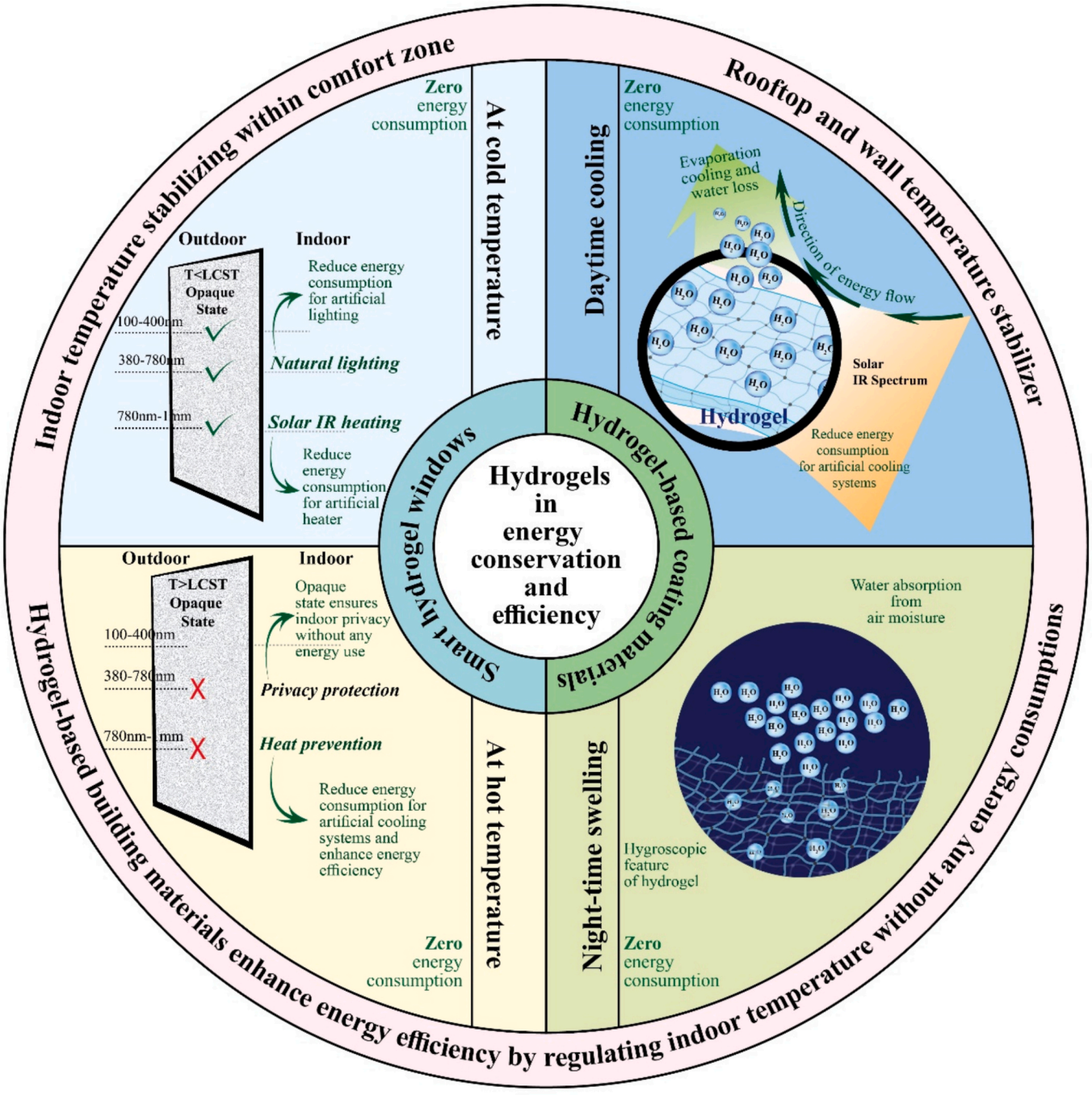

Fig. 11. Hydrogels in energy conservation and efficiency: indoor temperature stabilization by hydrogel-based smart windows and rooftop and wall temperature stabilization by hydrogel coating. The energy efficiency performance under different weather conditions (temperature variations) during both day and night is described.

and coating materials. We shall also discuss the associated challenges. The mechanisms and performance behind hydrogel-based wall and rooftop temperature stabilization, as well as the performance of hydrogel-based smart windows in modulating indoor temperature and lighting, are depicted in Fig. 11. Remarkably, these processes operate with zero energy consumption for temperature stabilization and exhibit cyclic capabilities, ensuring long-term sustainable performance.

5.1. Advanced hydrogel windows for indoor energy efficiency

Windows emerge as a critical factor in a building's energy inefficiency; they not only escalate cooling requirements by converting solar energy into heat during hot weather but also facilitate significant heat loss during cold weather, highlighting their significant impact on a building's energy balance. Traditional HVAC technologies are effective but carry high energy consumption and GHGs emissions, along with substantial manufacturing and maintenance expenses. Thermochromic smart (TS) windows have garnered significant interest due to their cost-effectiveness, responsive behavior to stimuli, and negligible energy requirements. Constructed by encapsulating phase change materials within a "sandwich structure," these TS-windows operate without additional energy input. TS-windows present a promising substitute for conventional HVAC systems.

Recent investigations have demonstrated that thermochromic hydrogels possess an exceptional ability to enhance the energy efficiency of TS-windows, thereby solidifying their role as energy-conserving materials in sustainable building design. Vanadium dioxide (VO_2) has been the primary phase-changing material for TS-windows; its high lower critical solution temperature (LCST) far exceeds the human comfort zone (68 °C), presenting a limitation for its use in temperature regulation [360]. Moreover, VO_2's substantial light absorption at both high and low temperatures complicates the optimization of solar transmittance (T_{sol}) and luminous transmittance (T_{lum}), hindering its practical application in TS-windows [361]. Thermochromic hydrogels surpass traditional VO_2 in smart window applications, offering a lower transition temperature, superior visible light transmittance, and enhanced infrared (IR) blocking above the LCST, along with robust solar modulation. HBPS [362], AEMR-PNIPAM [363], PNIPAM [364], PAM-PNIPAM [365], HPC [366], HPC–CTO [367], HPC-PVA [368], HPC/PAA [369], HPMC [370], P(VdF-HFP) [371], PAANa [372], and PVB [373] are hydrogels commonly used in smart window applications. Among them, PNIPAM [363–365,374–385], HPC [366–369,386,387] and HPMC [370,377] are the most frequently employed hydrogels. They share a common feature of having low and adjustable LCST, which makes them suitable for use in smart windows.

Exploring the energy-saving potential of thermochromic hydrogels, we gather information on key parameters presented in Table 18 that reveal the efficiency of smart window applications. The LCST is crucial; ideally, it should be proximate to human comfort temperatures to minimize HVAC system usage, thereby enhancing energy efficiency. A hydrogel with an optimal LCST facilitates autonomous modulation of solar influx, remaining transparent to transmit sunlight for heating below the LCST, and turning opaque to reflect heat and cool the interior above it [388,389]. The solar modulation capacity (T_{sol}) is vital for reducing solar heat gain in hot weather, [390] thus enhancing energy efficiency by reducing air conditioning reliance and maintaining indoor comfort. Visible transmittance (T_{lum}:380-780 nm) is significant as well [391]; a higher T_{lum} below the LCST ensures better natural illumination, which can curtail the need for artificial lighting [392]. The IR transmittance (780 nm^{-1} mm) is a determinant of a hydrogel's energy-efficient performance [391], as it must effectively block IR radiation above the LCST to prevent unwanted solar heating [393]. The switching speed of the hydrogel indicates the rapidity of transition from transparent to opaque, crucial for managing indoor temperature shifts [394]. Lastly, durability is a measure of a hydrogel's long-term performance and energy-saving potential in TS-windows, with a preference for those that sustain high efficacy over time [395,396]. For a comparative analysis of the energy efficiency across various hydrogel-based thermochromic smart windows, the different parameters used in hydrogels have been mentioned in Table 18.

The behavior of a thermochromic hydrogel-based smart window at temperatures below and above LCST is depicted in Fig. 12. It also delineates a generalized method for fabricating composite hydrogels for thermally switchable TS-windows. To reduce LCST of hydrogel to human comfort temperature or to achieve an adjustable phase transition temperature, composite hydrogels are synthesized with various ionic compounds such as sodium dodecyl sulfate (SDS), sodium shloride (NaCl), potassium chloride (KCl), lithium chloride (LiCl) and sodium metasilicate (Na_2SiO_3) [370,373,376,380]. These ionic compounds attenuate the hydrogel's bonding strength, facilitating phase transitions at lower temperatures. Additionally, another type of compounds is incorporated to increase the heat absorption capacity of the hydrogel's surface. This modification leads to a quicker temperature rise, enabling rapid switching between transparent to opaque state for superior energy efficiency.

The LCST values of the fabricated composite hydrogels, ranging from 20 to 40 °C as shown in Table 18, are ideally suited for energy-efficient smart window applications. Furthermore, it is fascinating to note that the LCST of these hydrogels can be tuned through changes in pH, pressure, or the application of applied voltage [369,372,381]. Table 18 also indicates that the majority of thermochromic composite hydrogel windows exhibit a high solar modulation capacity, with the apex value reaching 89 %. At temperatures equivalent to or below the threshold of human comfort, these hydrogel glass perform in a transparent state and transmit most of the visible light (65-99 %) for indoor lighting. The use of natural light for indoor illumination, instead of energy-consuming artificial light, can make a huge difference in energy consumption. Moreover, as temperatures exceed the comfort threshold, these hydrogel windows effectively obstruct 80-99 % of the visible spectrum, thereby providing privacy at no additional energy cost. Most of the composite hydrogel reduces the IR transmittance by >95 % through which it makes a huge temperature difference from the other types of glasses. To control temperature rise, these hydrogel smart windows take 3 to 5 min. Notably, certain hydrogels, such as PHC-Gel, demonstrate an ultra-fast response time of merely 6 s [397]. Supplementary voltage can further hasten the response time of these hydrogels, optimizing their performance in dynamic environmental conditions. Due to all these features, thermochromic composite hydrogels-based smart windows make a 2 to 13 °C indoor temperature difference compared to regular glass windows.

PNIPAM-based hydrogels find widespread use in smart window applications. Research indicates that smart window incorporating PNIPAM-KCA-Na_2SiO_3 hydrogel can effectively reduce indoor temperatures by an impressive 12.3 °C. Moreover, this hydrogel boasts an exceptionally high emissivity of 0.962. However, it's crucial to recognize that these performance measurements were conducted in a model house, which is 50,000 times smaller than a typical 400 sq. ft. residence [376]. Ensuring consistent efficiency in a standard-sized human room under varying humidity conditions remains a critical objective. Another noteworthy study explored PVA-PNIPAM with LimCsnWO_3 composite hydrogel, revealing its remarkable thermal insulation capacity. This hydrogel exhibited a substantial 40 °C difference between external and internal temperatures [375]. In a separate investigation, scientists impregnated PNIPAM-PAM hydrogel into transparent-delignified balsa wood, enhancing its mechanical properties. While achieving robust mechanical strength and low thermal conductivity for hydrogel windows, this material fell short in terms of indoor temperature reduction only 4.3 °C compared to quartz glass during hot weather [378]. The PNIPAM-PAM hydrogel, in synergy with SDS and NaCl, introduces a sophisticated three-stage optical modulation system for smart windows: opaque, transparent, and translucent. By applying voltage, rapid phase shifts occur. Notably, this hydrogel window retains an additional 1.3 °C of heat in cold conditions and effectively reduces temperatures by 2 °C

Table 18
Comparative energy-efficient performance analysis of the thermochromic hydrogels in smart windows. LCST value near human comfort temperature, high solar modulation capacity, high visible transmittance below LCST, faster switching speed, and low IR transmittance above LCST enhance energy efficient impression for composite hydrogel-based TS-windows. Higher cyclic performance attributes its durability and suitability in smart windows applications.

Fabricated hydrogels	LCST value	T_{sol}	T_{lum}		T_{IR}	Switching speed, S	Switching energy/ voltage	Durability (Cycle number of stable performance)	Significant outcomes	Remarks
			Below LCST	Above LCST	Above LCST					
HPMC/ NaCl/ water [370] *Ratio: 2:5:100*	-40 °C	–	100 %	1 %	<1 %	39 s	~0.2 W/ cm^2	$n > 100$	- Compared to regular quartz glass, it can reduce indoor temperature by 10 °C more in hot weather.	- LCST 20 °C lower than traditional VO_2 based smart windows. - Ecofriendly, cost effective, durable and provides highly stable performance.
HPC/ KCl [366] *HPC = 20 g/L, KCl = 1.0 M*	-30 °C	57.52 %	73.01 %	–	<3 %	30 s	8 V	$n = 250$	- Can lower indoor temperatures by 13.3 °C compared to traditional glass, showcasing its energy-saving efficiency.	- Below LCST, it's visibility can be changed within a range from 75.53 % to 15.70 % by applying voltages.
PNIPAM+ 30 % PAM + Delignified balsa wood [378]	22.9 °C	89 %	82.7 %	39.8 %	–	120-180 s	0	$n = 100$	- Compared to regular quartz glass, it can reduce indoor temperature by 4.3 °C more in hot weather.	- Thermal conductivity is 63 % lower than quartz glass, can save 2.6 %-10.2 % building energy.
HPC/PAA [369]	20 -35 °C	80 %	90.1 %	<5 %	<50 %	–	0	n = 100	- Smart window maintains a consistent 12 °C difference for 35 min compared to peak outdoor temperatures.	- Cost-effective and maintain a high T_{lum} and T_{sol}, - HPC/PAA hydrogel exhibits a lower LCST range from 20 to 35 °C across the pH of 4.5–6.3.
PNIPAM/Ppy/ KCl [381] *Ppy = 0.125 mg/mL KCl = 0.1 mol/ L*	20-32 °C	53.7 % (Regular) and 63.0 % (T-ECD)	65.6 %	0.2 %	3.0 % (T-ECD)	180 s (Regular)/ 90 s (T-ECD)	-1.0 to 5 V	$n > 7$	- Can reduce 3.5 °C more than normal windows in hot weather. - Can adjust visible transmittance by applying voltage -1 to 5 V	- KCl significantly reduces LCST of PNIPAm hydrogel. - Switching speed 3.7 times shorter than pure PNIPAm.
KCA/Na_2SiO_3/ PNIPAM [376]	27.2 °C	69.65 %	87.38 %	0.30 %	Very low	210 s	0	n > 100	- Can reduce 12.3 °C more than regular quartz glass.	- High emissivity (0.962) - Na_2SiO_3 ionic compound reduces LCST value - KCA enhances energy efficiency by shedding IR spectrum.
PVA-PNIPAM+ $LimCsnWO_3$ [375]	32 °C	–	67.48 %	16.76 %	0.23 %	30 s	0	$n > 150$	- Makes 40 °C difference between outside and inside surface where normal glass's thermal insulation capacity is almost zero.	- Bidirectional fast self-responsive hydrogel windows. - $LimCsnWO_3$ boosts the hydrogel's NIR blocking, while PVA lowers its LCST.
AEMR-PNIPAM/ $Cs_{0.33}WO_3$ [363]	27.4 °C	53.6 %	73.6 %	15.7 %	2.3 %	–	0	–	- Can reduce 7 °C more, due to excellent thermal insulation capacity and significant NIR shedding ability.	- AEMR is a good composite material, and reduces LCST of pNIPAM. - AEMR and $Cs_{0.33}WO_3$ both have the feature of shedding IR spectrum. - A promising material for flexible smart windows.

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Table 18 (*continued*)

Fabricated hydrogels	LCST value	T_{sol}	T_{lum}		T_{IR}	Switching speed, S	Switching energy/ voltage	Durability (Cycle number of stable performance)	Significant outcomes	Remarks
			Below LCST	Above LCST	Above LCST					
HPC–CTO [367] *CTO: 0.7 wt%*	48 °C	–	80 %	<6 %	<3 %	8.8 s	0	–	- Having excellent capacity of shedding NIR, can effectively stop temperature rise during hot weather.	- 0.7 wt% CTO absorbs NIR and releases heat to reach LCST of the HPC hydrogel and accelerates phase transition switching speed.
PHPA [399]	12 -34 °C	83.9 %	93.7 %	3.2 %	2.6 %	<60 s	0	n = 100	- This window's indoor-outdoor temperature gap reached 15 °C, notably less than double-glazed windows at 4.9 °C.	- Transition temperature and transition speed of hydrogel can be adjusted by temperature change. - LCST from 12 to 34 °C could be controlled by simply adjusting the HPA water ratio.
PNIPAM-PAM/ SDS/NaCl [380]	24.9 °C	72.9 % (at temp. 15 – 18 °C) and 42.7 % (at temp. 18 – 35 °C)	80.3 % at 15 – 28 °C	0.07 % (at temp. < 15 °C) and 8.3 % (at temp. 40 °C)	0.07 % (at temp. < 15 °C) and 44.6 % (at temp. 40 °C)	–	16 V for 60s	*n* = 80	- The hydrogel window maintains 1.3 °C more heat in the cold and reduces temperatures by 2 °C in the heat compared to standard glass. - Voltage application enables quick phase shifts.	- SDS and NaCl adjust PNIPAM hydrogel's LCST for comfort; PAM hastens opacity change. - Three-stage optical modulation: Opaque below 15 °C at night, transparent from 15 – 28 °C for lighting, and translucent above 28 °C to mitigate midday heat.
PHC-Gel [397]	20–35 °C	57.2 %	95.2 %	Very low	Very low	6 s	0	n = 100	- with a maximum cooling effect of ≈4 °C compared to ordinary windows and significant energy savings.	- The PHC-Gel is conductive, temperature-sensitive, and durable, stretching up to 300 % without damage. - The hydrogel becomes transparent in acidic conditions and opaque in base pH.
TRS + AgNWs/ cellulose PET film [400]	33 °C	17.9 %	67.7 %	19.8 %	7.2 %	–	5 V	*n* = 15	- Energy efficient than regular glass windows. - It requires additional electric energy.	- The hydrogel's efficiency is reduced by the need for added electrical energy, despite its adaptable light transmission properties around the LCST.
PNIPAM/HPMC [377]	32 °C	81.52 %	90.82 %	0.10 %	0.11 %	–	0	n = 100	- This smart window consistently maintained indoor temperatures below 32 °C, proving its effectiveness in reducing air conditioner use during heat, thereby saving energy.	- HPMC increases transparency of the hydrogel, enables higher percentage of visible light passing at low temperature. - Consistent after 100 heating/ cooling cycles between 60 and 25 °C.

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Table 18 (*continued*)

Fabricated hydrogels	LCST value	T_{sol}	T_{lum}		T_{IR}	Switching speed, S	Switching energy/ voltage	Durability (Cycle number of stable performance)	Significant outcomes	Remarks
			Below LCST	Above LCST	Above LCST					
HPC/PAA [386]	26.5 °C	47.5 %	90.1 %	31.17 %	43.05 %	84 s	0	n = 100	- The HPC/PAA hydrogel window outperformed standard glass windows and water-filled double-glazed windows, keeping the house cooler by 9.1 °C and 6.2 °C, respectively.	- Safe, sustainable, and affordable. - PAA reduces HPC's LCST to a cozy 26 − 28 °C for smart windows. - LCST adjusts with pH: 44 to 10 °C as pH drops from 6.0 to 1.0.
PNIPAM with glycerol–water (GW) [385] NIPAM =6.35 wt% Glycerol = 15 wt%	20–40 °C Auto tuned	60.8 %	90 %	–	–	10 s	0	*n* = 10	- have the fastest thermal response. - Good energy-saving option for indoors, provides high visible transmittance below LCST.	- outstanding freezing tolerance (-18 °C). - Have excellent solar modulation capacity and visible transmittance.
PVB/ LiCl [373]	28.5 °C	80.8 %	90 %	15 %	18.7 %	–	0	*n* = 500	- could reduce energy consumption by 36 % compared to conventional glass windows in warm seasons.	- The hydrogel's 700 % stretchability and consistent performance across 500 cycles highlight its sustainability. - excellent anti-freezing properties (stable performance at -15 °C).
HPC-PVA (4 wt %) [368]	40 °C	82.8 %	91.3 %	80 %	>19.4 %	15-20 s	0	stable performance after 30-days test	- Its LCST is close to the human comfort zone, and its high visible light transmittance reduces the need for electrical lighting.	- It's phase transition changes state from transparent to translucent. - Shows excellent visible transmittance properties at both hot and cool temperatures.
PS hydrogel + ITO film [401] PS = PTH + SDS + LiCl+ HMA + AAm + KPS+ TMEDA [401]	28-37 °C	–	85.4 %	11.7 %	14.6 %	–	3-8 V	n = 100	- High visible and IR spectrum shedding indicates its effectiveness will be higher than regular glass in warm conditions.	- Adjusting the voltage between 3 and 8 on the hydrogel window allows for control over its light transmission speed and energy efficiency.
Cu_7S_4/ PNIPAM [364]	32 °C	71.9 %	79 %	41.1 %	43.0 %	180 s	0	*n* = 11	- This hydrogel's efficient filtering of IR and UV light offers superior indoor temperature regulation compared to standard glass.	- This hydrogel smart window can be used for both thermal insulation and self-production of heat.
NaCl/ NSG40-γ-MPS/ (PNIPAM) [384] NaCl = 0.4 mol/L	27.2	89.93 %	95.15 %	1.30 %	2.53 %	–	0	n = 100	- Indoor temperature rises more slowly with this hydrogel window compared to regular glass.	- Higher NaCl concentration lowers LCST and visibility, with 0.4 mol/L being ideal for energy efficiency.
PNIPAM-AEMA [383]	31 °C	35.5 %	88 % -90 %	53.0 -60.7 %	0.9 -5.4 %	–	0	*n* ≥ 1000	- In Tucson, AZ, this hydrogel smart window can save up to 8.1 % in cooling energy annually, amounting to 30.6 kWh/m^2.	- This hydrogel's high thermal capacity (4.35 kJ/kg·K) helps cut down on heat loss and energy use.

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Table 18 (*continued*)

Fabricated hydrogels	LCST value	T_{sol}	T_{lum}		T_{IR}	Switching speed, S	Switching energy/ voltage	Durability (Cycle number of stable performance)	Significant outcomes	Remarks
			Below LCST	Above LCST	Above LCST					
W-VO_2 (M) with HBPS hydrogel [362]	21-32 °C	34.3 %	73.36 %	68.71 %	5.0-8.5 %	300 s	0	n = 8	- This hydrogel delivers outstanding light-handling capabilities and enhanced efficiency in solar energy regulation.	- LCST value of HBPS hydrogel could be controlled by varying the dosage of BGE
W-VO_2/PAM-PNIPAM [365]	32 °C	46.3 %	72 %	–	–	–	0	*n* = 40	- 100 μL W-VO_2/ PAM-PNIPAM hydrogel achieved the highest energy-saving ability of 46.3 %.	- Its dynamic solar transmission control makes it superior to traditional windows, leading to energy conservation.
PAANa/Agar [372]	Pressure responsive: 8-25 kPa	–	80 %	30 %	–	37.5 ms	Energy is needed to apply pressure.	*n* = 200	- This hydrogel works between 8 and 25 kPa pressure. - Applying pressure turns the hydrogel from opaque to transparent, blocking the light spectrum to cool interiors and boost energy efficiency	- As a pressure-responsive smart window, turns clear instantly when pressure is applied, due to the PAANa film touching the agar film. - The rough surface of agar can scatter about 23 % of visible and NIR light.

T_{sol} = Solar Modulation Capacity, T_{lum} = Luminance Transmittance, T_{IR} = IR Transmittance, S = Switching Speed.

during hot weather, outperforming standard glass [380]. Enhancing the temperature modulation capacity of PNIPAM-PAM-based hydrogels for smart window applications remains a key challenge.

Following PNIPAM, HPC emerges as one of the most prevalent hydrogels for smart window applications. When combined with KCl, HPC demonstrates an impressive ability to reduce indoor temperatures by 13.3 °C compared to traditional glass windows [366]. Similarly, HPC-PAA achieves a substantial reduction of 9.1 °C [386]. In a separate experiment, HPC-PAA consistently maintains a 12 °C temperature difference relative to the external environment [369]. Furthermore, research highlights that HPC-CTO effectively blocks near-infrared radiation due to the presence of CTO [367]. However, a persistent challenge lies in demonstrating the efficacy of these hydrogels across varying weather conditions and in larger room settings. On the other hand, HPMC-based hydrogel windows effectively maintain indoor temperatures below 32 °C, exhibiting a rapid transition period [370,377]. Nevertheless, a significant hurdle emerges: HPMC hydrogel windows are prone to leakage when exposed to temperatures exceeding 80 °C [370]. While HPMC-based hydrogels have undergone testing in lab condition model houses, their performance in mid-sized and large rooms remains unexplored [370,396].

We can manufacture hydrogels with high mechanical strength or, alternatively, create flexible hydrogels, indicating a wide range of applicability for these smart windows [363,378]. These hydrogels demonstrate remarkable durability, with some maintaining stable performance over 1000 cycles, ensuring long-term applicability [383]. Some hydrogels have outstanding freezing tolerance (NIPAM-Glycerol: -18 °C, PVB/ LiCl: -15 °C) [373,385], enabling their application in extreme weather conditions.

These hydrogels exhibit the highest energy efficiency performance with many features and are also characterized by their low cost, non-toxicity, and environmental friendliness. Such attributes underscore the revolutionary potential of hydrogels as materials for smart windows and energy-efficient buildings.

5.2. *Hydrogel-based coating materials for energy-efficient building*

In addition to hydrogel-based TS windows, the application of hydrogel coatings on rooftops and walls can significantly enhance the reduction of HVAC energy consumption as well as GHGs emissions. Owing to the passive cooling properties of hydrogels, these materials emerge as formidable candidates for building coating applications. Compared to other coating materials, hydrogels offer superior thermal stabilization and indoor cooling performance, potentially yielding an indoor temperature difference of 3 to 15 °C relative to ambient conditions, as detailed in Table 18. Consequently, hydrogel coatings on building envelopes present considerable promise as energy-efficient building materials.

To maintain stable temperatures on walls and rooftops, the majority of composite hydrogels coating developed incorporated mechanisms of radiative and evaporative cooling approaches that incur no energy costs and preclude GHGs emissions. This cooling mechanism is illustrated in Fig. 13. Hydrogels are characterized by their high latent heat capacity and substantial water swelling ratios. These properties enable the hydrogels to absorb solar heat effectively as latent heat and facilitate water evaporation, thereby shielding interiors from thermal gain. During this process, the hydrogel coating layer expends most of its water content. However, due to the good hygroscopic nature of hydrogels, they are capable of rehydration through absorption of atmospheric moisture at night, thus rejuvenating the coating for subsequent daytime evaporative cooling. Hydrogel coating for building temperature stabilization is similar to atmospheric water harvesting. For both cases, natural sunlight and radiative cooling mechanism are used [398].

Hydrogel coatings have demonstrated effective thermal management

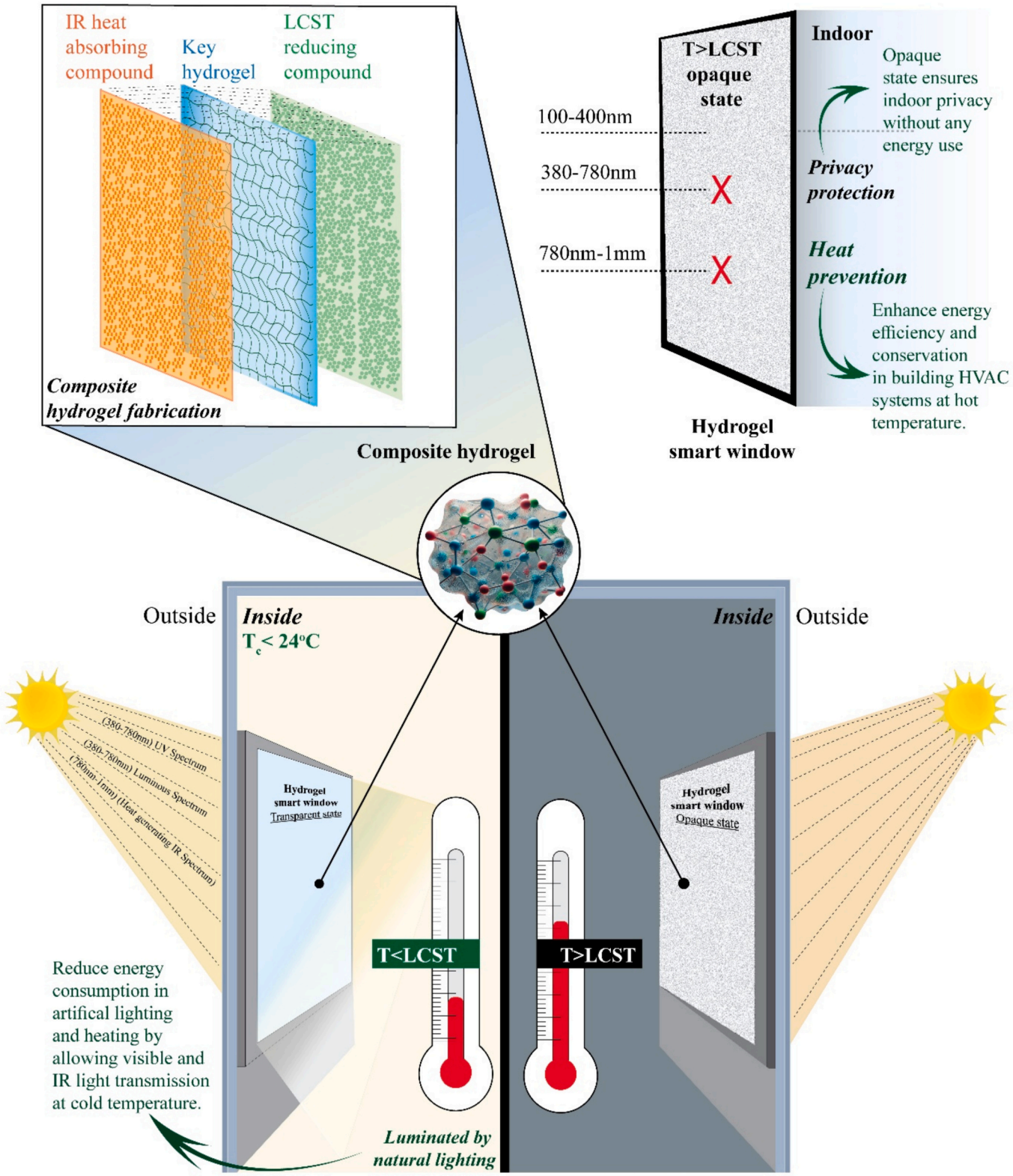

Fig. 12. Thermochromic composite hydrogels for smart windows: fabrication, temperature-responsive transmittance, and solar heat modulation. A generalized composite hydrogel incorporates a key hydrogel along with two distinct types of additives: a compound that lowers the LCST and an IR heat-absorbing compound. At low temperatures (T < LCST), the temperature-responsive light transmittance properties of these composite hydrogels allow high visible light transmittance for indoor illumination. The opaque state (T > LCST) can effectively block both visible and IR light transmittance, thereby providing solar heat protection and privacy for indoor spaces.

in arid desert conditions. PNIPAM [374,402], PAAm [403], PAAm-PVA [404], PAAm-LiBr [7], HPC-NaCl [387], P(VdF-HFP) [371], PNAH [405] to name but a few, hydrogels are used in making different coating materials for building energy efficiency.

A study utilizing polyacrylamide hydrogel as a coating on a specialized desert rooftop revealed that the material's thermal attributes could maintain an indoor temperature at 27 °C, which is notably lower than the external temperature of 32 °C [403]. Such remarkable energy-saving performance positions hydrogels as frontrunners in the future landscape of building materials. For instance, HPC/NaCl composite hydrogel coatings have been shown to achieve 20.3 % greater energy savings compared to conventional coatings and the temperature of the coating was 7- 10 °C lower than conventional coatings [387]. However, it's crucial to thoroughly investigate the long-term stability and durability of both the thermochromic hydrogels. Additionally, DN hydrogel coatings applied to walls and rooftops can significantly reduce electricity usage and GHGs emissions by 51 % [406]. However, addressing the challenges of improving the single hydration charge period remains crucial for further enhancements. Cotton-IPN (Alginate-Ca^{2+}/PNIPAM) hydrogel coatings yield even more superior results, cutting electricity

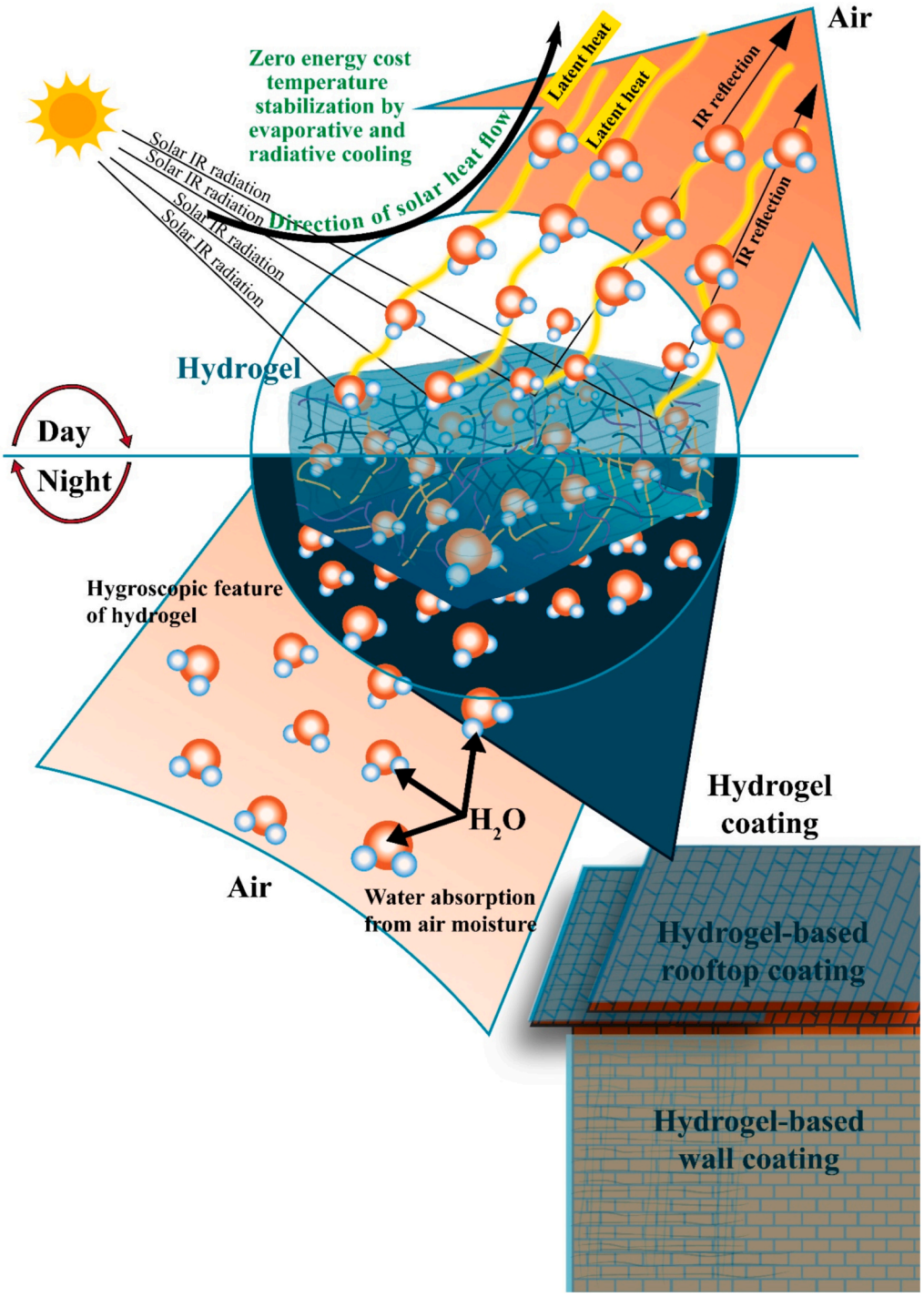

Fig. 13. Passive evaporative cooling mechanism of hydrogel coatings for building rooftop thermal stability. Hydrogel coatings on rooftops absorb solar IR heat as latent heat to ensure thermal heating protection, facilitating water evaporation to stabilize temperature during the day. At night, the hydrogels rehydrate by absorbing water from air moisture, leveraging their hygroscopic properties.

consumption and GHGs emissions by 55 % [379]. Enhancing the hygroscopic properties of the composite hydrogel is a crucial challenge for this work. Another study highlights that ZrO_2@PAAm-LiBr hydrogels possess 18 times the tensile strength and 9 times the compressive strength relative to standard hydrogels, alongside exhibiting high solar reflectance in the IR region [407].

This formidable strength renders them highly suitable as building coating materials. Considering these findings, it is evident that hydrogel coatings stand at the vanguard of energy-efficient building materials, offering unparalleled thermal management and sustainability benefits that herald a new era in architectural design and environmental stewardship.

6. Summary and outlook

The unique tunability of hydrogels has made them promising materials for diverse applications in the renewable energy sectors. Advanced design strategies and cutting-edge innovations have been developed to enhance the mechanical, thermal, and chemical properties of hydrogels. However, challenges in engineering and fundamental understanding still limit their practical applications. The versatility of hydrogel materials demonstrates their potential in innovative technologies for environmental energy harvesting, including hydrogen production [12,20,117], salinity gradient energy [24,120,123], and uranium extraction [11,133,135], to name but a few. Hydrogel works as a most prominent driver to revolutionize the energy storage system, especially with some versatile qualities, e.g., wetness, high ionic conductivity, heat tolerance and selective ion movement, rapid ion-exchange capacity, high life cycle and high charge-discharge interval.

In hydrogel-based supercapacitors, hydrogels enhance the capacitor's stability, enabling them to self-repair, extending the device's lifespan, and ensuring reliable energy storage with safety. Besides, hydrogel electrodes are innovative, reducing mechanical stress, improving structure, enhancing ion flow efficiency, ensuring close contact between electrodes and electrolytes with optimized ion paths, and trapping active materials to enhance the surface area, preventing them from clumping and maximizing usage, thereby boosting storage and stability. Hydrogels increase charge speeds and energy density by refining materials, using solid-state electrolytes, and incorporating active materials, serving diverse fields of energy storage devices. Hydrogel-based building materials and smart windows offer significant energy efficiency and conservation benefits. By incorporating hydrogels, indoor temperatures can be cooled by 2 to 15 °C without relying on energy-intensive HVAC systems. Thermochromic smart windows, utilizing hydrogels as phase change materials, outperform other smart windows by regulating solar light transmission. Additionally, hydrogel coatings on walls and rooftops employ reflective and evaporative cooling to mitigate solar heat gain.

Novel experimental and practical approaches are necessary to enhance the performance and versatility of hydrogels in the energy sectors. The development of new hydrogel materials for energy harvesting is crucial, as it can drive the transition towards a sustainable and secured energy future. PVA-based hydrogel composites, known for their high ionic conductivity and stable chemical and mechanical properties, have demonstrated efficacy in hydrogen production [12,19]. Hydrogels demonstrate superior applicability over other materials in certain domains, for instance, PAO hydrogel composites have been the most used adsorbents to extract uranium from seawater [11,134,135]. Hydrogel composites derived from PAAc, PAAm, cellulose, and PEDOT: PSS exhibit thermal and mechanical stability, making them suitable for thermoelectric conversion [10,27,74], osmotic energy harvesting [49,121], mechanical energy harvesting [151,156,158], and hygro-gradient energy harvesting [165–167].

Additionally, GO hydrogels, with their high electrical conductivity and large surface area, are compatible with microbial fuel cells (MFCs) [26,127,130]. From a fundamental perspective, the tunability of hydrogels offers a promising avenue for further experimental and research innovations. From an engineering standpoint, the development of hydrogels with comprehensive intrinsic properties can diversify energy harvesting technologies with practical applications. Introducing highly conductive and stable hydrogels with compatible properties could be a key strategy for advancing towards a sustainable energy future.

Despite highlighting the profound impact of hydrogels on energy storage systems in this study, achieving a sustainable and efficient energy solution presents some challenges. Especially for advancements in the self-healing capacity of hydrogels, innovative technologies like dynamic covalent chemistry [408] and supramolecular interactions will significantly bolster device durability and lifespan [409,410]. Hybrid hydrogel systems with advanced nanomaterials such as graphene [261], CNTs [258], and MXenes [411] promise to elevate energy storage performances. Optimizing compositions in hydrogel electrodes, such as graphene-based [261] and MXene-based hydrogels [411], will maximize energy and power density. Environmentally sustainable and biocompatible hydrogel formulations like hyaluronic acid [263] and cellulose nanofibril hydrogels [412] will broaden their utility. Finally, scaling up production techniques remains challenging, so it is essential to meet demand and reduce production and maintenance costs effectively to achieve a suitable energy storage supercapacitors and hydrogels-based batteries.

The energy efficiency performance of hydrogels in medium and large-sized rooms remains unknown, making it a significant area for further research. Hydrogel-based building materials exhibit promising anti-freezing properties, as evidenced by several studies [373,385]. However, further research is needed to evaluate anti-freezing properties under real-time weather conditions. Notably, only one paper has investigated hydrogel temperature stabilization in desert climates, highlighting the need for additional research in this area [403]. Integrating photovoltaic hydrogels into smart windows and building coatings for energy generation represents a valuable avenue for future research, requiring collaboration among chemists, material scientists, environmental scientists, and engineers. We must explore next-generation hydrogel-based building material technologies to address global energy conservation and climate change challenges. As the quest for sustainable and efficient energy solutions intensifies, the continued advancement and integration of hydrogel technologies hold the promise to not only revolutionize energy storage and conversion but also to catalyze a new era of green energy innovation.

CRediT authorship contribution statement

Mohammad Mahbub Kabir: Writing the original draft, conceptualization, investigation, designing the experiments, methodology, validation, writing, review and editing; **Golam Md. Sabur**: Writing the original draft, methodology, and validation; **Md. Fazlul Karim Mamun**: Investigation, methodology, validation; **Arman**: Investigation and data analysis; **Leonard Tijing**: Supervision, writing, review and editing; **Yeshi Choden**: Writing, review and editing; **Sherub Phuntsho**: Writing, review and editing; **Ho Kyong Shon**: Supervision, conceptualization, project administration, resources, funding acquisition, validation, writing, review and editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. Ho Kyong Shon serves as a Co-Editor-In-Chief for the Desalination journal, while the editorial handling and review of this manuscript were overseen by a different Co-Editor-In-Chief.

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Data availability

No data was used for the research described in the article.

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