REVIEW

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Recent progress in gel catalysts: boosting efficiency for sustainable energy applications

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Achieving carbon neutrality and mitigating global warming necessitate a shift from fossil fuels to renewable energy sources. This review explores the pivotal role of polymeric gels in advancing energy conversion and storage technologies, highlighting their potential in reducing CO_2 emissions. Gels exhibit unique properties such as thermal conductivity, mechanical resilience, and catalytic efficiency, making them promising candidates for energy applications like photovoltaic cells, batteries, and electrocatalytic systems. Their flexible structure, large surface areas, and porous nature significantly improve redox reaction efficiency and energy storage capacity. Recent innovations, especially hybrid gels combining conducting polymers and nanoparticles, have enhanced catalytic performance, electrical conductivity, and durability, offering more sustainable energy solutions. This review thoroughly examines the synthesis methods, structural properties, and performance metrics of gel materials, focusing on their applications in fuel cells, batteries, and supercapacitors. It also addresses the mechanisms behind energy conversion facilitated by these materials and discusses challenges related to scalability and long-term durability. By providing a comprehensive overview of recent advancements, this review aims to guide future research and drive technological progress in the field of sustainable energy, positioning gel catalysts as key components in the transition to cleaner, more efficient energy systems.

1. Introduction

Scientists widely agree that substantially reducing human-caused CO_2 emissions to a net zero level is crucial to stop global warming. To achieve a carbon-neutral society, it is essential to explore renewable energy sources, especially considering the exhaustion of fossil fuels and their harmful effects on the atmosphere.[1,2] Fossil fuels have significantly degraded the global atmosphere and are also nearing exhaustion, projected to be depleted within the coming few decades. This urgent scenario compels energy researchers to investigate and develop new renewable energy sources. A fundamental property of energy states that it can neither be created nor destroyed but only converted from one form to another. For example, photovoltaic cells convert solar radiation into electrical energy.[3,4] This interconversion requires energy, initially in one form, to be stored as chemical energy *via* a redox reaction. The core concept involves

comprehensively understanding these redox reactions and scaling them to novel materials that facilitate convenient electron transfer, thereby enabling efficient energy conversion into electrical energy and storage for future use.[5,6] This focus on interconversion reactions and electrochemical processes is rapidly garnering attention within the research community as a potential solution to mitigate global warming. Innovative materials and technologies that can effectively convert and store energy are crucial. In this context, gels have emerged as a significant area of interest due to their unique properties and potential energy conversion and storage applications. This review aims to highlight the use of gels in various energy conversion and storage reactions, emphasizing their role in advancing sustainable energy technologies and contributing to the global effort to achieve carbon neutrality.

The versatility of gel materials, including their ability to incorporate different active components, tune physical properties, and provide structural stability, positions them as promising candidates in developing next-generation energy systems.[7] Through a detailed examination of recent advancements and future directions, this review provides a comprehensive understanding of how gel materials can be leveraged to enhance electrochemical energy conversion efficiency and stability, thereby playing a crucial role in

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Catal. Sci. Technol., 2025, 15, 1357–1389 | 1357

addressing our time's pressing energy and environmental challenges. Energy conversion is crucial in advancing sustainable and environmentally friendly energy sources. It plays a vital role in converting environmental pollutants and excess energy into valuable chemical or electrical forms, which helps mitigate climate change and improve energy storage capabilities.[8,9] Electrocatalytic conversion of energy systems can transform environmental pollutants, such as CO_2, into energy molecules that benefit the environment in environmentally friendly activities. The efficiency of this energy exchange method is essential for satisfying the demands of practical applications, taking into account elements such as affordability, dependability, and adaptability in operation.[10,11] Due to this requirement, researchers have been compelled to explore alternative renewable sources, particularly in converting energy forms. One such example is the conversion of solar radiation into electrical energy through photovoltaic cells. This technique involves energy storage through chemical redox reactions.[12,13] Current advances in gel materials have successfully resolved these issues by improving catalytic sites of action, changing shape, and changing electrical characteristics. Most current research is on gels, especially polymeric gels, which are flexible semi-solid materials that can change from solid to liquid. Their versatile features, such as their ability to conduct heat and resist mechanical stress, make these gels extremely promising for energy-related uses, such as improved batteries and innovative materials for storing and collecting energy.[14] Gel catalysts, which consist primarily of polymeric or colloidal structures that enclose a liquid state, have attracted considerable interest because of their unique characteristics. These characteristics include an enormous surface area, a porous structure that facilitates the movement of substances, and a versatile framework that may be chemically modified to meet specific catalytic requirements. These features are essential for increasing the speed of electrochemical processes and raising the reliability and sustainability of the catalysts during operation.[15,16] Gel materials are highly versatile in their applications within the conversion of energy and storage. They can be used as 3D binders, substances for electrode electrolytes, structures for active components, and self-supported energy collectors. Prominent applications include the electrochemical removal of carbon dioxide to produce extremely valuable commodities, oxygen reduction processes, and the oxide-evolved mechanisms in batteries made of metal–air.[17] The production of these newly created gel materials results from either chemical or physical interactions between the individual components or between the individual components and crosslinking agents. The development of gels is primarily influenced by many elements, such as forces of electrostatic bonds, π–π forces of interactions, forces of hydrogen bonds, hydrophobic interactions, and covalent bonds.[18] To enhance the efficiency of catalytic processes by increasing the density of active sites and facilitating easy access to micro- and nanostructural features.

Furthermore, upholding exceptional electrical conductivity to provide efficient electrocatalytic activity in real-world scenarios is crucial. Electrocatalysts often show some negative characteristics, such as clumping together, reduced active sites, inefficient movement of electrons and mass, insufficient conductivity of electricity, and lack of ability to safely attach particles to a typical conductive base. These often lead to issues such as delamination, aggregation, and disintegration. Gel materials exert a highly advantageous impact on the design of catalysts. They offer increased design flexibility, facilitate direct catalytic pathways, enhance catalyst durability, allow for inexpensive processing considerations, and lead to significantly reduced environmental problems.[19] The present condition of polymeric catalysts emphasizes several categories, including synthetic hydrolases, which immobilize enzymes, and other types, such as phase-transfer catalysts and photosensitive substances. These catalysts possess several benefits: they are insoluble, rendering them easy to distinguish from responses and capable of reusing; they shield active centers from removal; and they may enable multiple reactions within a single reactor.

Moreover, the characteristics of these catalysts may be altered by adding polymeric carriers, allowing for pH adjustment to achieve optimum activity in immobilized enzymes.[20] Conductive polymeric gels (CPGs) are nanostructured substances, bioelectronics, and energetic devices. Their microscopic polymer features remain intact as they create a network with three dimensions, leading to the acquisition of novel characteristics. Researchers enhance the electrical conductivity of these gels by employing specialized molecules that form connections with one another. CPGs may be precisely regulated by altering the constituents, including monomers and cross-linkers, and adjusting parameters like temperature and solvents. Additionally, these gels can be combined with other substances, resulting in hybrid gels that exhibit enhanced mechanical and electrical characteristics.[21] They play a role in sophisticated electronic devices, biosensors, and renewable energy systems and possess unique attributes such as self-repairing capabilities and liquid resistance as shown in Fig. 1.

Polymer gels, especially those that conduct electricity, provide several benefits, including high specific surface area, flexibility, and impressive electrical characteristics. These materials can integrate different nanoparticles, significantly enhancing physical and electrical properties. As a result, they are highly suitable for a wide range of energy applications. This article will explore several hybrid polymer gels, such as dihybrid and trihybrid topologies, specifically developed to generate energy (such as solar cells) and store it (like robust capacitors and batteries). The gels are created by combining conducting polymers, such as polyaniline, polythiophene, and PEDOT:PSS, with components like graphene oxide and silver nanoparticles, using supramolecular cross-linking agents like folic acid and dibenzoyl-L-cystine. The hybrid hydrogels formed have improved electrical and electrochemical characteristics by securely attaching
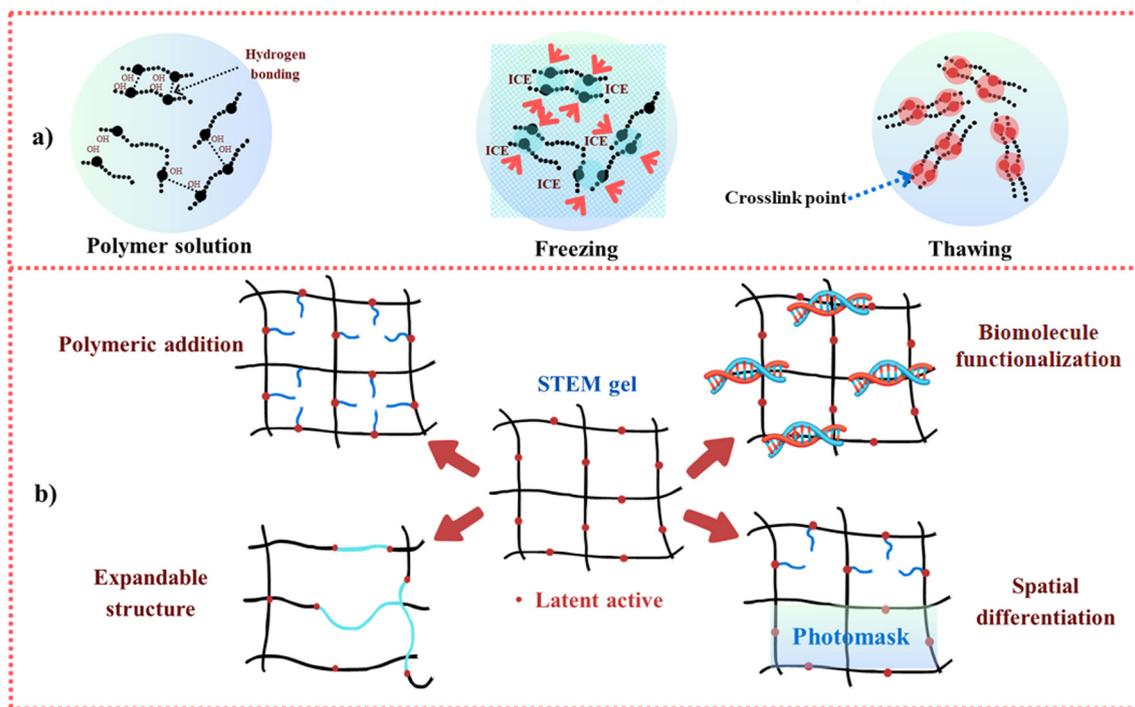
Fig. 1 a) Hydrogel cross-linking mechanism. b) STEM gel enhancement *via* post-synthesis modifications at active sites.

nanoparticles to the network's fibrils. Energy gadgets like dye-sensitized solar cells and batteries mainly utilize hybrid xerogels. In addition, we will investigate the utilization of several synthesized and organic polymer gels in energy-related fields, providing insights into their characteristics, practical uses, and potential advancements.[22]

A notable innovation discussed in the review is the development of gels that combine conducting polymers with nanoparticles. This hybrid approach has significantly improved the catalytic performance, electrical conductivity, and durability of gel materials in energy applications. These advancements make polymeric gels more suitable for sustainable energy solutions, such as batteries, supercapacitors, and electrocatalytic systems. The hybrid gels leverage the unique properties of both polymers and nanoparticles, leading to a flexible structure, enhanced redox reaction efficiency, and increased energy storage capacity, positioning them as promising materials for reducing CO_2 emissions and advancing carbon-neutral technologies.

2. Exploring the physical attributes of gels

Exploring gels' diverse applications and physical attributes reveals a dynamic field rich in scientific exploration and practical use. Gels play pivotal roles across multiple disciplines, from soft matter physics to cutting-edge biomedical engineering. These adaptable materials are central to innovations such as drug delivery systems, tissue engineering frameworks, and even culinary techniques. By analyzing their rheological properties, mechanical strength,

and swelling capabilities, researchers can better understand gel behavior. Gels are identified by their stability and ability to maintain their shape under stress, embodying complex structural and behavioral traits that are crucial for advanced material design. Originating from the work of P. J. Flory, the predominant definition of gels describes them as possessing a continuous, analytically stable structure, accompanied by solid-like rheological properties.[23] Key to their identity are several defining features: a fibrillar network structure, a storage modulus that remains constant across frequencies and exceeds the loss modulus, and a capacity for reversible compression.[24] This solid-like behavior stems from a three-dimensional fibrous network that encases the liquid phase from which the gel originates, hindering flow and conferring its semi-solid nature. Gels thus occupy an intermediary state between liquid and solid states, with their formation being facilitated by a plethora of physical or chemical interactions among constituent building blocks, sometimes aided by cross-linking agents. Chemical gels, formed through covalent bonds, stand in contrast to physical gels, which arise from non-covalent interactions like coulombic forces, hydrogen bonding, and hydrophobic interactions. Resulting fibrils, often nanostructured, endow gels with elasticity, adjustable mechanical strength, and a multitude of desirable electronic and optoelectronic properties, contingent upon the nature and concentration of the building blocks.

The components of gels encompass a wide array, ranging from natural polymers such as gelatin, chitosan, and agarose, to synthetic counterparts like poly(ethylene glycol), poly(acrylic acid), and poly(vinyl alcohol). In energy applications, conducting polymer gels take precedence,

leveraging the responsiveness of conjugated chains to external stimuli such as light, voltage, and pH for device fabrication. Ion-conducting gels, vital for energy devices, facilitate ion mobility and conductivity, while supramolecular gels, demonstrating conductivity through materials like carbon nanomaterials and metallogels, play crucial roles in energy material fabrication. Challenges in dissolving conducting polymers necessitate the use of supramolecular crosslinkers, which not only aid in gel formation but also act as dopants, creating three-dimensional network structures with enhanced electrical conductivity and hierarchical porosity conducive to rapid mass and charge transport.[25,26] Within the electrochemical domain, the hierarchical pores of gels facilitate interconnected pathways for ion transport, mitigating volume changes and damage to transport paths during mass flow. These intricate networks, characterized by their elastic properties, underscore the critical role of gels in enabling efficient electrochemical processes, with shared interactions between ions and neighboring chains ensuring seamless conductivity and transport within the fibrillar matrix. Thus, gels not only stand as a testament to the ingenuity of materials science but also serve as indispensable facilitators of technological advancement, positioned at the forefront of innovation across diverse fields.

2.1 Fundamentals of gel materials

Gel materials encompass a broad class of materials characterized by a three-dimensional network structure that immobilizes a solvent phase within its matrix. This network structure, often referred to as the gel framework, imparts mechanical integrity to the material while retaining the ability to swell and undergo structural rearrangements in response to external stimuli.[27] The properties of gel

materials, including their mechanical strength, porosity, swelling behavior, and responsiveness to stimuli, are intricately linked to the synthesis process and the structural characteristics of the gel network.[28] Depending on the nature of the interactions involved in gel formation, gel materials can be classified into chemical gels, physical gels, or hybrid gels.[29] Chemical gels are formed through the covalent crosslinking of polymer chains or small molecules, while physical gels rely on reversible non-covalent interactions such as hydrogen bonding, hydrophobic interactions, or electrostatic interactions. Hybrid gels combine elements of both chemical and physical gelation, offering enhanced properties and functionalities. The fundamental classification of gel materials into chemical gels and physical or hybrid gels, along with their synthesis methods, is shown in Fig. 2. This figure further highlights the energy-related applications of gel materials, categorized into energy storage, energy conversion, and environmental remediation & energy harvesting applications.

2.1.1 Chemical gelation. Chemical gelation represents a versatile approach to synthesizing gel materials by initiating crosslinking reactions between precursor molecules to form a three-dimensional network structure. One of the most widely employed chemical gelation methods is the sol–gel process, which involves the hydrolysis and condensation of metal alkoxides or other precursors to form an inorganic gel network. The sol–gel process offers precise control over the composition, porosity, and morphology of the resulting gel materials, making it a valuable technique for applications such as catalyst supports, membranes, and optical devices.[30,31] In addition to sol–gel chemistry, chemical gelation can also be achieved through the crosslinking of polymer chains *via* various polymerization reactions, including radical polymerization, condensation
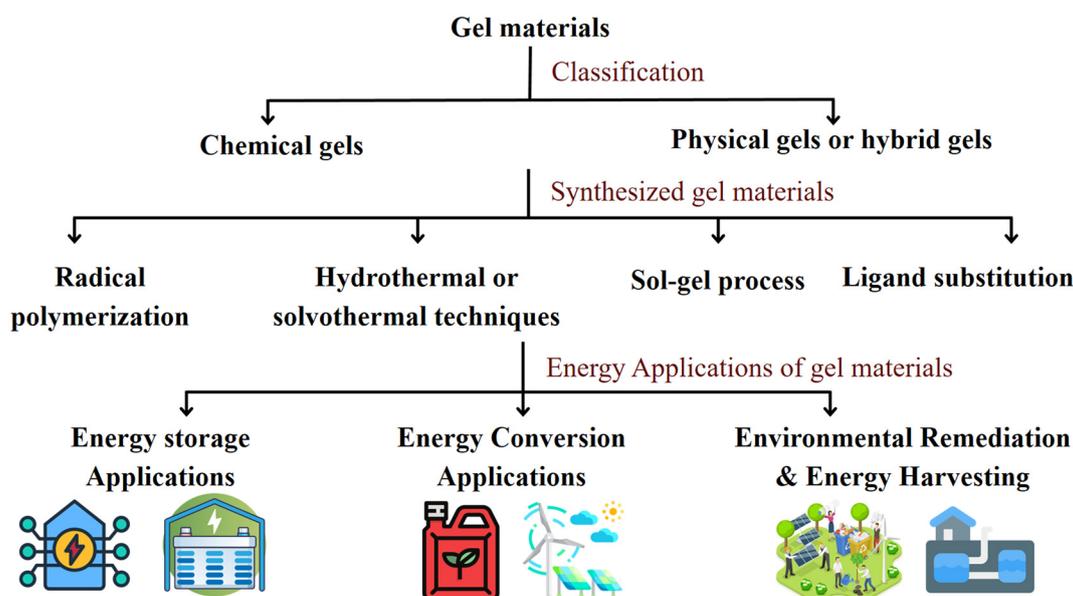

Fig. 2 Overview of gel materials: classification, synthesis methods, and energy applications.

polymerization, and ring-opening polymerization. A simplified illustration of the gelation process for chemically cross-linked protein hydrogels is provided in Fig. 3.[32] While the diagram may not accurately depict true protein hydrogel networks, it serves as a visual guide. Protein monomers are engineered with chemical cross-linking sites, and cross-linking is activated, creating a covalently bonded network, forming a viscoelastic hydrogel in water. These polymer-based gels exhibit tunable mechanical properties, swelling behavior, and degradation kinetics, making them suitable for applications such as tissue engineering scaffolds, drug delivery carriers, and hydrogel actuators.[33] Moreover, chemical gelation strategies can be tailored to introduce specific functionalities into the gel matrix, such as bioactive molecules, stimuli-responsive moieties, or conductive additives, further expanding the potential applications of gel materials.

2.1.2 Physical gelation. In contrast to chemical gelation, physical gelation relies on reversible non-covalent interactions to form a network structure within a gel material. Physical gels can be formed through a variety of mechanisms, including thermo-reversible gelation, pH-triggered gelation, and self-assembly of amphiphilic molecules. Thermo-reversible gels, for example, undergo gelation upon cooling below a critical temperature due to the formation of physical crosslinks, such as hydrogen bonds or hydrophobic interactions, which stabilize the gel network.[34]

Physical gelation occurs through non-covalent interactions, driven by forces like hydrophobic interactions, van der Waals forces, or hydrogen bonding, rather than chemical cross-linking. For example, in the case of PX 407, a triblock copolymer, its physical gelation mechanism in water involves hydrophobic and hydrophilic segments as shown in

Fig. 3 Chemical gelation: a simplified illustration of the gelation process for chemically cross-linked protein hydrogels is provided. (a) Protein monomers are engineered with chemical cross-linking sites, and (b) cross-linking is activated, (c) creating a covalently bonded network, forming a viscoelastic hydrogel in water (reproduced from ref. 32 © IOP Science 2020).

Fig. 4a-(i). The hydrophobic segments cluster together to form micelles, while the hydrophilic portions extend outward. As the concentration or temperature increases, these micelles arrange into a structured network, resulting in gelation. This *in situ* physical gelation is initiated by hydrophobic interactions, which cause the polymer to self-assemble into a viscoelastic gel as shown in Fig. 4a-(ii). The process is reversible and can be controlled by adjusting environmental factors, making it valuable in drug delivery and biomedical applications.

These gels exhibit reversible gel–sol transitions upon heating and cooling, making them suitable for applications such as drug delivery, tissue engineering, and ionic conductors. pH-triggered gels, on the other hand, undergo gelation in response to changes in pH, resulting in conformational changes or electrostatic interactions that promote gel formation. pH-responsive gels find applications in controlled release systems, sensors, and actuators.[37,38] Additionally, physical gelation can be achieved through the self-assembly of amphiphilic molecules, such as surfactants or block copolymers, which form micellar or vesicular structures that coalesce into a gel network. These self-assembled gels offer unique properties such as high porosity, surface functionality, and stimuli responsiveness, making them attractive for applications in drug delivery, cosmetics, and nanotechnology.

2.1.3 Hybrid gelation. Hybrid gelation approaches combine elements of both chemical and physical gelation to achieve synergistic effects and enhanced properties in gel materials. By incorporating covalent crosslinks alongside reversible non-covalent interactions, hybrid gels exhibit improved mechanical strength, stability, and responsiveness to external stimuli. One common example of hybrid gelation is the incorporation of physical crosslinking mechanisms, such as hydrogen bonding or host–guest interactions, within a polymer network formed *via* chemical crosslinking. These hybrid hydrogels combine the mechanical robustness of chemical gels with the stimuli responsiveness of physical gels, making them suitable for applications such as tissue engineering, drug delivery, and soft robotics. In Fig. 4b-(i) and (ii), hybrid gelation occurs during the synthesis of an injectable bioadhesive using GelMA (gelatin methacryloyl) and Gel-Phe (gelatin phenylalanine). Upon injection, UV light initiates chemical cross-linking, solidifying the GelMA/GelPhe network, making it effective for hemostasis. In this hybrid process, pure chemical gels form through chemical cross-linking, while physical-*co*-chemical gels incorporate both physical and chemical interactions. Cluster formation occurs in both types, but the growth in physical-*co*-chemical gels is constrained by triple-helix junctions, creating a more structured network with enhanced mechanical stability. This dual mechanism offers improved versatility for applications such as wound healing and tissue engineering.

Another approach to hybrid gelation involves the integration of inorganic nanoparticles or nanofibers within a
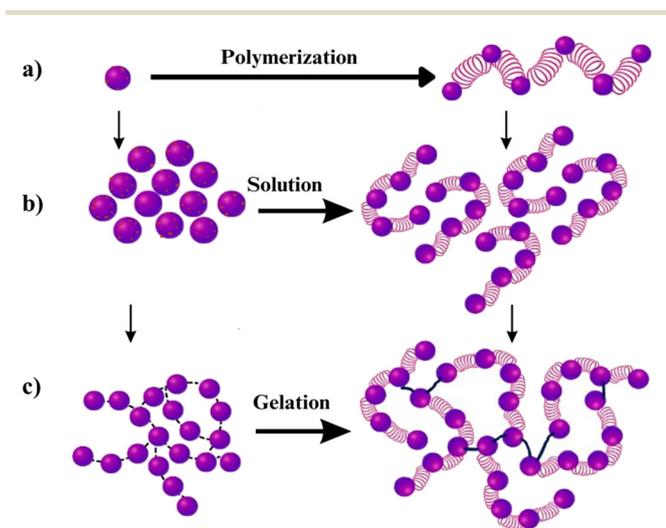
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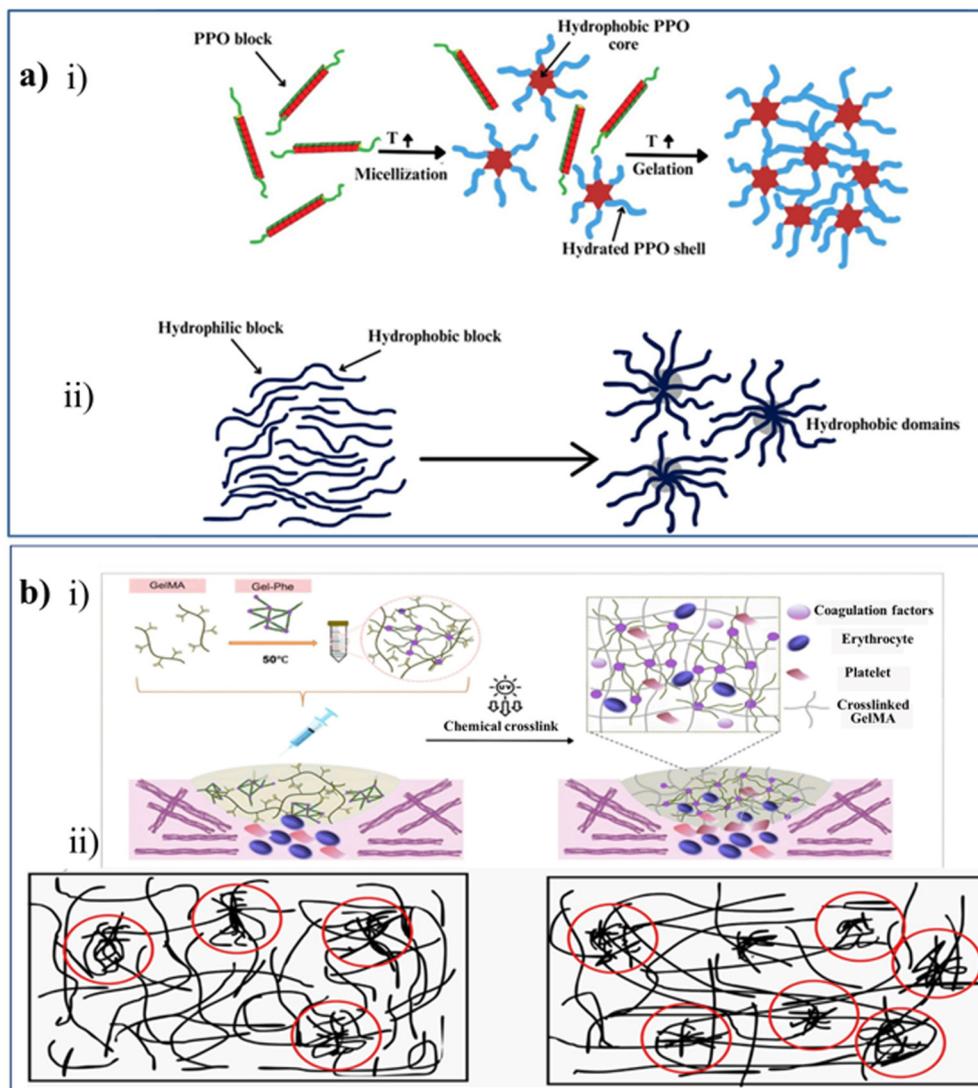*Catal. Sci. Technol.*, 2025, **15**, 1357–1389 | **1361**

Fig. 4 (a) Physical gelation: (i) a schematic illustration depicting the physical gelation mechanism of PX 407 in water and (ii) the mechanism of *in situ* physical gelation initiated by hydrophobic interactions (reproduced from ref. 35 © MDPI 2021). (b) Hybrid gelation: (i) depiction of the synthesis process for GelMA and Gel-Phe, and the injectable GelMA/GelPhe bioadhesive, cross-linked with UV light, designed for hemostasis. ii) Diagram illustrating: pure chemical gels and physical-*co*-chemical gels. In both scenarios, gelation occurs through the formation of clusters (outlined in red), with the growth in physical-*co*-chemical gels being limited by the presence of triple-helix junctions (reproduced from ref. 36 © The Royal Society of Chemistry 2013).[35,36]

polymeric matrix to reinforce the gel structure and impart additional functionalities. These composite gels exhibit tailored mechanical, electrical, or optical properties, depending on the nature and concentration of the incorporated nanoparticles, enabling applications in areas such as sensors, actuators, and energy storage devices.[39]

3. Process of synthesizing gel materials

In this exploration, we navigate through the complex landscape of gel material synthesis, investigating the plethora of methods utilized to craft these adaptable materials. Ranging from chemical cross-linking to physical gelation, we

scrutinize the diverse strategies employed to tailor gels for multifaceted applications. Our examination spans both traditional and cutting-edge synthesis techniques, offering insight into the fundamental principles and critical factors governing gel formation. The synthesis of gel materials involves a diverse toolkit of approaches, each carefully chosen to achieve specific material properties and functionalities. Researchers meticulously select and combine precursor materials, fine-tune reaction conditions such as temperature and pH, and manipulate parameters like stirring speed and solvent composition to orchestrate the formation and structure of gels. The methods used to synthesize gel materials can be classified as follows: (i) radical polymerization, (ii) hydrothermal/solvothermal techniques,

(iii) sol–gel processes, and (iv) ligand substitution. Each of these approaches presents distinct benefits in customizing the characteristics of the resulting gel material to suit particular applications.[40]
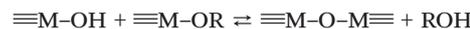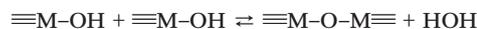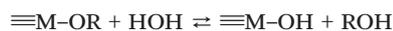
3.1 Free-radical polymerization

This method involves initiating polymerization reactions using free radicals, resulting in the formation of cross-linked networks characteristic of gels. Investigating the intricacies of this method, we analyze variables like initiator choice, reaction conditions, and monomer composition, all of which significantly impact the properties of the resulting gel materials. By elucidating these fundamental principles and exploring recent advancements in radical polymerization techniques, our review aims to provide essential insights crucial for understanding and refining gel synthesis through this versatile method. Radical polymerization plays a fundamental role in creating organic polymeric gels under mild conditions. Through the use of oligomers, polymeric hydrogels are produced *via* radical polymerization, involving processes such as oxidation, deprotonation, or coupling reactions. The process of free radical polymerization occurs in three stages: initiation, propagation, and termination.[41] Employing single crosslinker-type polymerization leads to hydrogel polymers with abundant hydrophilic groups (Fig. 1). In the formation of hydrogels using the F–T method, physical cross-linking occurs through the crystallization of the polymeric solution. This mechanism relies on the interaction between hydroxyl groups in the polymer chain and water molecules, facilitating the formation of hydrogen bonds. The freezing process induces crystal growth, establishing cross-linking points between polymer chains, while subsequent thawing relaxes the polymer chains, allowing their mobility. The iterative F–T cycle generates additional cross-link points, perpetuating the cross-linking process. Crosslinking agents such as acrylic acid, acrylamide, and vinyl acetate are utilized to promote monomer chemical bonds through UV radiation, temperature, or redox initiator mixing, providing the three-dimensional structure of hydrogels and allowing for adjustments in swelling capacity and elastic properties.[42,43] The performance of hydrogels is influenced by the type of monomer, regulation of composition, and modification of structural crosslinking agents. However, improper regulation may lead to the production of inconsistent hydrogel products.[44,45] Additionally, radical polymerization encounters technical challenges due to its rapid and difficult-to-control nature. The exothermic nature of radical polymerization results in gel effects, releasing significant heat suddenly and posing safety concerns. These gel effects represent a significant drawback when scaling up polymerization processes for industrial applications.[46]

3.2 Sol–gel methods

The sol–gel method is widely adopted in wet chemical processes for crafting diverse nanostructures, especially metal oxide nanoparticles. Initiated by precursor solution hydrolysis, it generates colloidal particles that transition into a gel. The ensuing wet gel necessitates proper drying, typically achieved through alcohol combustion for alcohol-containing solutions. Subsequent pulverization and calcination produce gels with controlled chemical composition, thanks to the method's low reaction temperature and cost-effectiveness. Such characteristics make sol–gel materials indispensable in various industries like surface coating and building insulation. Within this method, precursors transition into sols at low temperatures, enabling uniform doping at the nanoscale, ensuring precise control over the material structure, and yielding high purity with uniform particle size distribution. This economic viability underscores its commercial feasibility in large-scale production. Fig. 5 provides a summary of the processes achievable through the sol–gel method and the resulting products. Particularly noteworthy is the sol–gel method's aptitude for producing highly active inorganic hydrogels.[47–49]

The process involves precursor solution solvation, hydrolysis, and partial condensation, culminating in sol formation. Subsequent polycondensation yields gels.[50,51] During drying, solvent evaporation causes porous network collapse, yielding dense xerogels, while supercritical drying produces aerogels. Calcination further stabilizes the products. Various parameters like the precursor type, concentration, solvent, pH, additives (*e.g.*, surfactants, catalysts), and heat treatment significantly impact material properties. The sol–gel process entails three key reaction steps: hydrolysis of alkoxy groups, condensation of –OH-containing species, and mixed condensation of –OH and alkoxy group-containing species, as delineated in below equations.[52–54] The sol–gel technique presents a flexible and economical method for creating gel materials, allowing meticulous management of the composition and structure. Its ability to generate highly functional inorganic hydrogels, coupled with its sequential reaction stages and the multitude of factors affecting material characteristics, highlights its importance in materials synthesis across various sectors.

$$\equiv M\text{--}OR + HOH \rightleftharpoons \equiv M\text{--}OH + ROH$$

$$\equiv M\text{--}OH + \equiv M\text{--}OH \rightleftharpoons \equiv M\text{--}O\text{--}M\equiv + HOH$$

$$\equiv M\text{--}OH + \equiv M\text{--}OR \rightleftharpoons \equiv M\text{--}O\text{--}M\equiv + ROH$$

3.3 High temperature/high pressure method

The versatility and efficiency of the high temperature/high pressure method for gel material synthesis are underscored by its ability to precisely regulate the composition and structure. By subjecting precursor solutions to elevated temperatures and pressures within sealed vessels, this
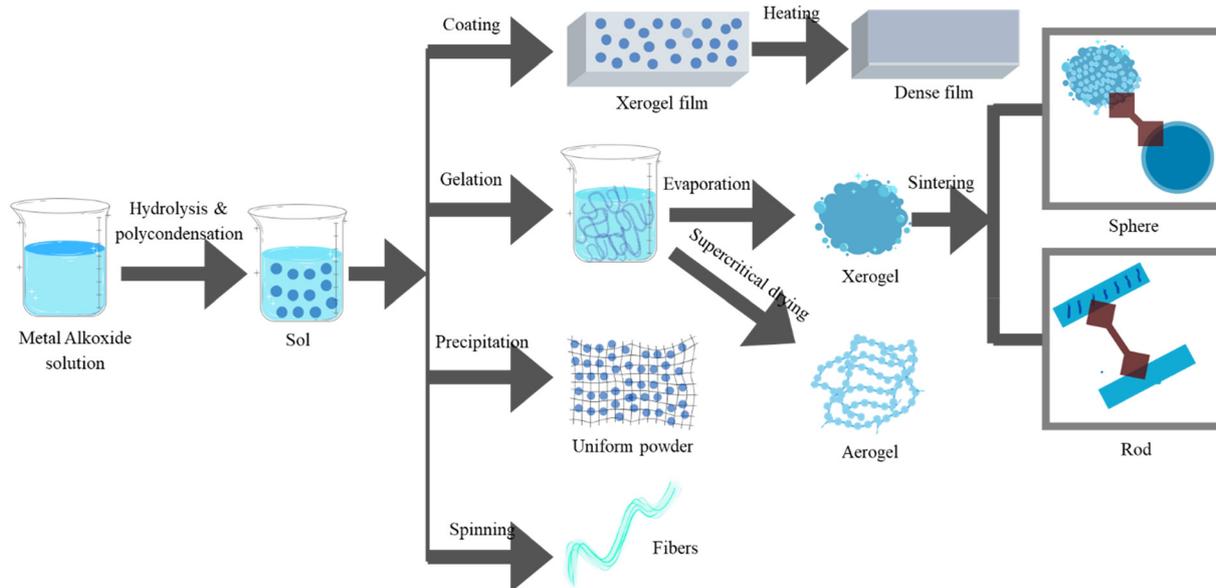
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Catal. Sci. Technol., 2025, **15**, 1357–1389 | **1363**

Fig. 5 Summary of sol–gel processes and their resulting products (reproduced from ref. 48 © Springer Nature 2020).

technique facilitates gel formation through hydrothermal or solvothermal reactions. Operating within specific temperature and pressure parameters, heterogeneous reactants dissolve in water or organic solvents, making this method particularly suitable for components and material phases resistant to high temperatures. A notable advantage is its capacity to generate new products and intermediates at lower temperatures compared to alternative approaches.[55,56]

Hydrothermal technology employs an aqueous solution as a dispersion medium, while solvothermal technology relies on a non-aqueous dispersion medium, both attracting interest due to their ability to produce high-quality crystal powders *via* single-step reactions at low temperatures. Typically, raw materials are dissolved and dispersed in a solvent before encapsulation in an autoclave reactor (Fig. 6), followed by heat treatment and high pressure to initiate
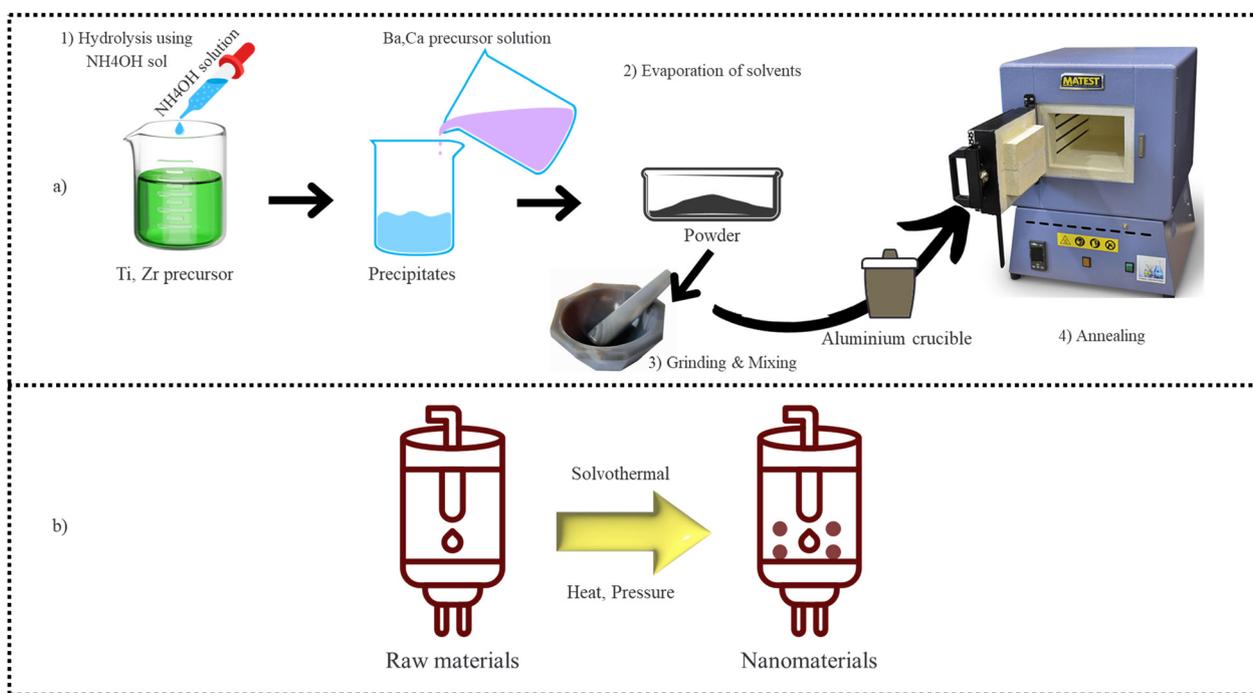

Fig. 6 Diagram showcasing the intricate pathways of (a) sol–gel synthesis and (b) hydrothermal techniques, unveiling the artistry behind material formation products (reproduced from ref. 59 © ACS Publications 2016).

1364 | *Catal. Sci. Technol.*, 2025, **15**, 1357–1389

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crystallization.[56–58] This methodology finds utility in fabricating hierarchical carbon or graphene-based hydrogels, where three-dimensional structures arise from synergistic π–π stacking and hydrophobic interactions, creating extensive porous networks conducive to hosting materials across vast surface areas.[59,60] As a result, the resulting gels exhibit distinctive properties tailored to specific reaction conditions, making them invaluable across various applications spanning catalysis to materials science. Through meticulous management of reaction parameters such as temperature, pressure, and solvent composition, the hydrothermal/solvothermal method enables the production of gel materials customized to suit diverse industrial and scientific needs. Overall, the hydrothermal/solvothermal method represents a versatile and efficient approach for manufacturing gel materials, providing precise control over their composition and structure, with widespread utility across industries and scientific domains.

3.4 Replacement of ligands

The ligand replacement technique offers a promising avenue for manufacturing gel materials, providing a unique method to tailor their composition and structure. In this process, ligands within metal complexes undergo substitution, resulting in the formation of gels with customized properties. By carefully selecting and manipulating ligands, researchers achieve precise control over the characteristics of the resulting gel materials. Unlike oxide-based inorganic gels typically synthesized through sol–gel methods, cyanogels are commonly created via ligand-substitution reactions between chlorometalates ($[M'Cl_4]^{2-}$) and cyanometalates ($[M''(CN)_n]^{2-/3-}$) (Fig. 7a). In this mechanism, chloride ligands from chlorometalates are replaced by cyanide ligands in cyanometalates, forming cyanide bridges ($M'-N\equiv C-M''$). Continuous cyano-reactions lead to the development of

cyano-linked gels with an interconnected structure. For example, Sn–Fe cyanogels can be generated by blending $SnCl_4$ and $K_4Fe(CN)_6$ aqueous solutions, resulting in Sn–N≡C–Fe bridges due to the substitution of chloride ligands and solvent ligands on the Sn(IV) center with cyano ligands on the Fe(III) center.[61,62] Besides these synthetic pathways, gelation processes can also be initiated by alternate reactions, including cyclic freezing–thawing for fabricating poly(vinyl alcohol) (PVA) hydrogels, the Pechini method for producing metal/organic gels, flocculation–precipitation of inorganic oxides, and self-assembly of metal nanoparticles via van der Waals or hydrogen bonds.[63–66] Regardless of the process leading to the final gel, the gel scaffold is fully immersed within solvents, presenting a 3D interconnected framework.[67]

Catalysts derived from ligand substitution methods are acknowledged for their exceptional electrocatalytic properties, facilitated by well-coordinated water and lattice vacancies. The incorporation of multiple metals is evenly dispersed, enhancing hydrophilic characteristics. A 3D porous structure endowed with hydrophilic properties encourages electrolyte penetration, thereby boosting electrocatalytic performance, such as the OER.[68] The highly permeable conductive polymeric gel within an inorganic polymer gel ensures outstanding and efficient mass and charge transfer capabilities, thereby making significant contributions to potential energy storage and conversion processes.[69] Table 1 describes gel catalysts, classified by their composition, structure, and functional properties. These include polymer gels, metal–organic gels (MOGs), and inorganic gels. Polymer gels provide flexibility and conductivity, while MOGs offer tunable catalytic sites. Inorganic gels act as robust supports. Hybrid gels, combining both, exhibit multifunctionality, making them suitable for electrocatalytic reactions such as the ORR, HER, and CO_2 reduction.

Gel electrocatalysts play a critical role in enhancing the oxygen evolution reaction (OER) and oxygen reduction
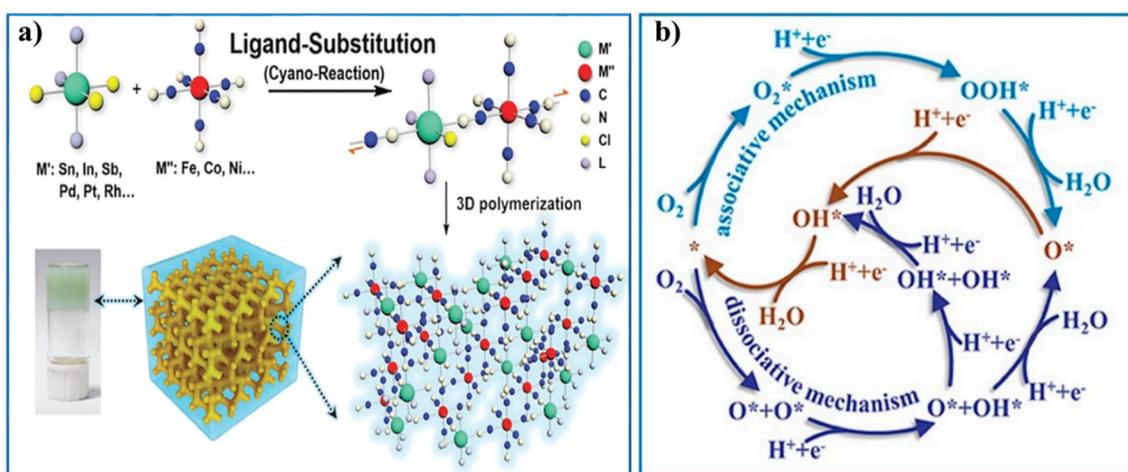

Fig. 7 (a) Illustration outlining the innovative pathway for fabricating gel materials: crafting cyanogels through intricate ligand substitution synthesis (reproduced from ref. 64 © ACS Publications 2012). (b) ORR within associative and dissociative mechanisms (reproduced from ref. 67 © Frontiers in Materials 2019).

Table 1 Comparative analysis of gel catalyst types in electrocatalytic applications: classification, challenges, and advantages

Categories	Example	Challenges	Advantages
Metal–organic gels	Co/Ni-containing organic gels	Potential leaching of metal ions, sensitivity to temperature	Efficient charge and mass transport, rich in active defect sites
Transition metal oxides/OH/complex gels	Fe porphyrin, MnO	Poor wettability, self-agglomeration, susceptibility to intermediate poisoning	Multiple oxidation states, high pseudocapacitance, excellent energy storage, high theoretical activities
Nanostructured supramolecular gels	Guanosine-based supramolecular gels	Limited structural stability during repeated cycling	Improved wettability, electrolyte access, tunable materials, 3D network, efficient electron transport
Noble metal gels	Au, Ru, Ir, Pd, Au–Pd	Low gelation kinetics, difficulty in controlling the microstructure, high material cost	Excellent conductivity, efficient electron/mass transfer, strong stability, plasmonic properties
Bimetallic gels	Noble-transition metal alloys (Pd–Cu) Noble–noble metal alloys (Au–Pd) Non-transition metal alloys (Bi–Sn) Transition–transition metal alloys (NiFe, NiCo, FeCoRu) Transition–non-transition metal alloys (Cu–Bi)	Challenging metal interactions, concerns with reproducibility, stability issues, difficulties in homogeneous alloying and scaling up	Facilitated mass transport, selective catalysis, synergetic effects, lower energy barriers, noble metal cost-efficiency, higher intrinsic polarity
Carbon/graphene with heteroatom-doped gels	FeCo-N-dual-network carbon, Ag–GO	Structural degradation under high current densities or prolonged operation, competing side reactions	Large surface area, optimized porosity, cost-effectiveness, stability, hierarchical structure, enhanced wettability, dual-network for improved contact
Single-atom-doped carbon gels	(Fe, Co, Ni, Cu)-doped N-containing carbon	Optimization challenges for universal catalysts, scaling-up, aggregation, catalyst poisoning	Industrial-level current densities (>100 mA cm^{-2}), enhanced activity
Transition metal carbide gels	Fe$_3$C	Aggregation leading to crystallization growth	Expanded d-orbital interactions for high catalytic activity

reaction (ORR), which are essential for metal–air batteries, fuel cells, water electrocatalysis, and photocatalytic water splitting.[70] The ORR is limited by significant overpotential at the cathode, while the OER experiences rate-limiting steps in metal–air batteries and water-splitting applications, both of which slow down reaction kinetics. Understanding the mechanisms behind these reactions from Fig. 7(b) is essential for designing efficient electrocatalysts to improve the performance of energy storage and conversion devices. Transition metals are promising, low-cost catalysts for the ORR and OER due to their abundance and activity; however, in practical systems like Zn–air batteries, their limited conductivity reduces efficiency, highlighting the need for further development.

3.5 Other methods

In addition to traditional methods like sol–gel processes and hydrothermal/solvothermal techniques, several alternative approaches have emerged for synthesizing gel materials, each offering unique advantages and enabling precise control over material properties. Emulsion polymerization stands out as a versatile method for producing gels with tailored characteristics.[71] In this process, monomers are dispersed in an aqueous phase containing surfactants to form micelles, where polymerization occurs. The resulting gel structure is influenced by factors such as the monomer concentration, surfactant type, and reaction conditions, allowing for fine-tuning of porosity, surface area, and functionality. Electrospinning, on the other hand, involves the application of a high electric field to a polymer solution or melt, resulting in the formation of ultrafine fibers.[72] By adjusting parameters like polymer concentration, solution viscosity, and processing conditions, researchers can control the morphology and properties of the resulting gel fibers. This method offers advantages in terms of scalability and the ability to create gels with high surface area and mechanical strength. Self-assembly techniques represent another promising avenue for synthesizing gel materials with precisely controlled structures.[73] By leveraging non-covalent interactions such as hydrogen bonding, π–π stacking, and van der Waals forces, molecules can spontaneously organize into ordered structures, forming gels with unique properties. Templated self-assembly approaches further enhance control over the gel structure by using pre-designed templates to guide the assembly process. Overall, these alternative methods for synthesizing gel materials offer researchers a diverse toolkit for tailoring material properties to meet specific application requirements, ranging from biomedical applications to energy storage and beyond.

1366 | Catal. Sci. Technol., 2025, 15, 1357–1389

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3.6 Synthesis of modified gel materials

The production of derivatives emerges as a crucial approach to augment functionality and expand utility across various sectors. Derivative synthesis entails a range of chemical and physical procedures geared towards altering the chemical composition or physical attributes of gels. This may encompass the introduction of specific molecules or polymers to confer desired characteristics like heightened mechanical strength, biocompatibility, or responsiveness to external stimuli. Moreover, derivatives may integrate nanoparticles or nanomaterials to introduce innovative functionalities such as catalytic activity or electrical conductivity. Surface modification methods further facilitate the customization of gel surface chemistry, thereby influencing interactions with the surrounding environment or enabling targeted molecular recognition events. Gels possessing interconnected nanostructures can seamlessly transition into hierarchical functional catalysts, encompassing carbon, metal oxide, and metal/alloy-based materials, capitalizing on their inherent large surface areas and open channels for mass/charge transport. By leveraging molecular or nanoparticle-level mixing of initial materials, modified gels serve as ideal precursors for fabricating intricate materials like binary/ternary/quaternary metal oxides, complex alloys, and carbon materials doped with uniform heteroatoms. The synthesis of gel derivatives typically involves a drying phase followed by heat treatment, with techniques such as freeze-drying and supercritical drying often utilized to maintain the hierarchical gel structure.[74,75] By meticulously controlling annealing parameters such as time, temperature, and atmosphere, heat treatment can finely adjust the physical and chemical properties of the resulting products. This includes modifications to crystallinity and particle size in metal oxides or alloys, graphitization of carbon frameworks, regulation of porosity and surface areas in final materials, and integration of heteroatom dopants within gel-derived networks. The resulting derivatives amalgamate beneficial attributes such as highly porous structures with extensive surface areas, uniformly dispersed active species, efficient mass/charge transfer, and precisely controlled dopants, making them promising materials for diverse applications including the hydrogen evolution reaction (HER), oxygen evolution reaction (OER), oxygen reduction reaction (ORR), carbon dioxide reduction reaction (CO_2RR), and beyond. In essence, the synthesis of gel derivatives epitomizes a flexible strategy for tailoring materials to specific needs, thereby unlocking avenues for innovative applications across fields such as biomedicine, environmental remediation, and advanced materials science.

3.6.1 Metals and alloys derived from gels. The rise of metals and alloys derived from gels represents a significant shift in material synthesis methodology, providing a flexible and effective means to customize advanced materials based on specific requirements. This article delves into the complexities of employing gel-based techniques to create metallic and alloyed structures, elucidating the underlying principles, synthesis methodologies, and resulting material properties. By harnessing the distinctive characteristics of gels, such as their ample surface area and adjustable pore structures, scientists have achieved notable enhancements in the mechanical, electrical, and catalytic properties of metallic materials. Metals and alloys, compounds comprised of metal or non-metal elements, demonstrate enhanced attributes like strength, toughness, corrosion resistance, and conductivity. Traditional methods like high-temperature melting and electrochemical deposition often yield dense solids with limited surface areas and active sites. In contrast, gel-derived alloys, blending the advantages of metal alloys and aerogels, emerge as promising electrocatalysts due to their rapid electron transfer rates, smaller particle sizes with increased defect sites, and highly porous structures facilitating efficient mass transfer. A recent study presented a wet-chemical reduction approach for synthesizing Sn–Ni alloys, utilizing a cyanogel/graphene oxide double-network hydrogel as the precursor.[76] The resultant Sn–Ni/C dual framework seamlessly integrated Sn–Ni alloy frameworks and graphene architectures, ensuring uniform alloy incorporation into the graphene network (Fig. 8).[74] While carbon-supported catalysts offer ample surface areas, they are prone to corrosion under elevated potentials, compromising their durability. In contrast, unsupported pure metallic aerogels exhibit satisfactory durability due to direct contact between electroactive species and conductive substrates. Another research group reported hierarchical aerogels comprised of multimetallic Ni–Pd$_x$Pt$_y$ nano-building blocks with precisely engineered shapes and compositions.[77] By employing a sol–gel method and galvanic replacement, the team developed aerogel monoliths with controlled morphologies and compositions. The resulting Ni–Pd$_x$Pt$_y$ aerogel showcased a hierarchical structure with abundant exposed edges and a high surface area, promising enhanced catalytic performance. These examples underscore the potential of gel-derived metals and alloys in advancing electrocatalysis and other applications, emphasizing their versatility and efficiency in material design and synthesis.

3.6.2 Metal oxides derived from gels. The investigation into metal oxides derived from gels presents promising opportunities in material synthesis, offering a dynamic and efficient means to tailor advanced materials with precise characteristics. This manuscript conducts a comprehensive analysis of the methodologies and core principles involved in synthesizing metal oxides using gel-based approaches, elucidating the fundamental mechanisms and resulting material traits. Nanostructured metal oxides have gained significant attention from researchers and practitioners alike due to their wide-ranging applications in energy conversion, storage, catalysis, and separation processes. For instance, titanium dioxide (TiO_2) and zinc oxide (ZnO) nanocrystals, functioning as carriers for photon-generated electrons, are extensively utilized in dye-sensitized solar cells and
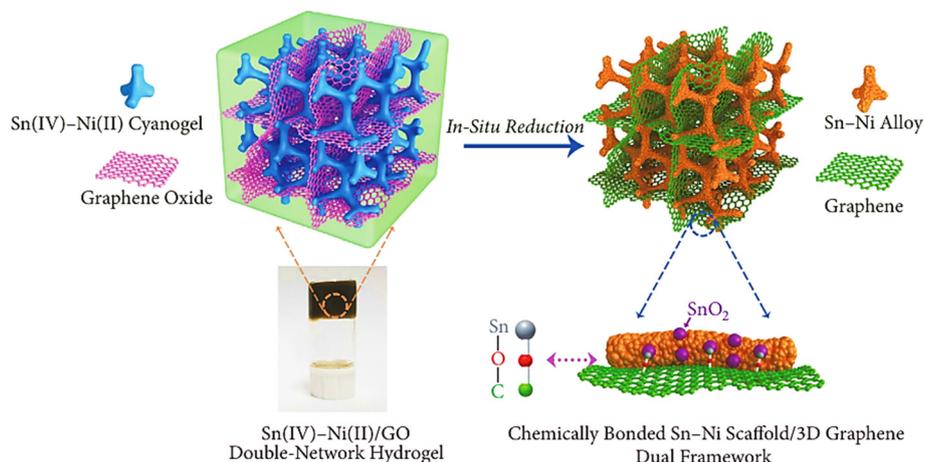
Fig. 8 Pioneering the synthesis of Sn–Ni/graphene gels through innovative routes (reprinted from ref. 74 © Springer Nature 2011).

photoelectrochemistry.[78–81] Transition metal oxides, known for their distinctive 3D electronic structures, are extensively employed in energy storage devices such as lithium-ion batteries and supercapacitors, as well as efficient electrocatalysts for the oxygen evolution reaction (OER) and oxygen reduction reaction (ORR).[82–86] Within the plethora of metal oxide nanomaterials, hierarchical metal oxides derived from gels demonstrate exceptional thermal, electronic, mechanical, and optical properties owing to their high specific surface area, porosity, and low density, thus significantly improving performance across various applications. Notable research, exemplified by the work of Yan *et al.*, showcases innovative methods for fabricating hierarchical metal oxide structures. Their investigation into

stacked ultrathin cobalt oxide (Co_3O_4) nanosheets with precisely controlled interlayer spacing and surface functionalization, achieved through a scalable sol–gel technique, illustrates the potential of gel-derived metal oxides in tailoring materials with specific properties.[87] In this process, the nano/microstructure efficiently absorbs a substantial amount of water, facilitating the dissolution of lithium lanthanum titanate (LLTO) precursors within the hydrogel matrix. Subsequent annealing in air removes polyvinyl alcohol (PVA), resulting in the formation of interconnected LLTO frameworks in a three-dimensional arrangement. This innovative approach underscores the adaptability and effectiveness of gel-derived metal oxides, paving the way for advancements across diverse technological
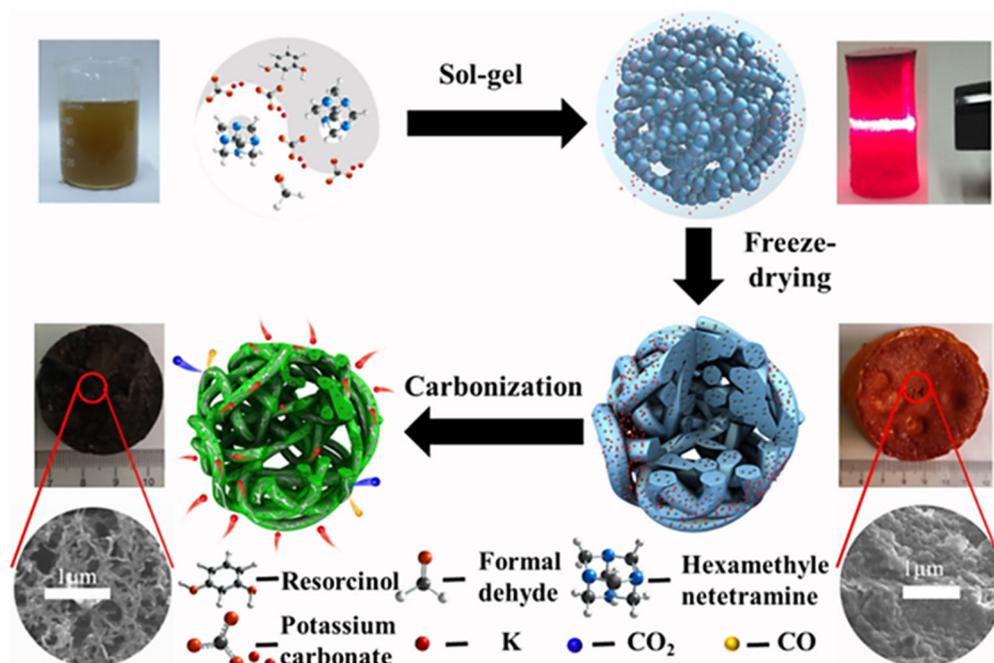

Fig. 9 Crafting carbon aerogels: a visual roadmap through synthesis (reproduced from ref. 92 © Elsevier 2017).

1368 | *Catal. Sci. Technol.*, 2025, **15**, 1357–1389

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3.6.3 Carbon-based materials derived from gels.
Carbon-based materials derived from gels have garnered significant attention due to their remarkable structural and functional attributes, which are advantageous across various advanced applications. These materials are synthesized through a sol–gel process followed by carbonization, producing a highly porous structure with a substantial surface area and tunable pore sizes.[88,89] Such features endow them with high electrical conductivity, thermal stability, and robust mechanical properties, making them highly suitable for energy storage devices like supercapacitors and lithium-ion batteries.[90] For instance, carbon aerogels are prepared (Fig. 9) by the sol–gel method, followed by freeze-drying and subsequent carbonization stages.[91,92] In a typical synthesis, resorcinol and formaldehyde polymerize in the presence of K_2CO_3 as a catalyst and hexamethylenetetramine as a crosslinking agent, resulting in a hydrogel precursor. Adjusting the mass ratio of K_2CO_3 to resorcinol significantly influences the gelation time and pH of the precursor solution, which in turn affects the polymerization rate and the formation of the cross-linked structure, as evidenced by the Tyndall effect. The subsequent freeze-drying step transforms the hydrogel into a carbon aerogel, with excess K_2CO_3 crystallizing and immobilizing within the structure. During carbonization, K_2CO_3 melts and reacts with the carbon, creating abundant micropores. As the temperature increases, K_2CO_3 decomposes into K_2O and CO_2, further etching the material to form mesopores and macropores. This method contrasts with traditional approaches for preparing single-atom materials, which often require complex post-treatments. Instead, the gel-driven strategy offers an effective alternative by confining polymer chains and fostering strong interactions between the support and active sites. Additionally, the large surface area and customizable surface chemistry of gel-derived carbon materials enhance their performance in catalysis, environmental remediation, and sensing applications.

Gel catalysts achieve enhanced active site accessibility and diffusion rates through specific structural modifications and experimental techniques that quantify these effects. Incorporating heteroatoms like nitrogen or sulfur into the carbon matrix further improves their electrochemical and catalytic properties. For example, nitrogen-doped carbon gels synthesized *via* sol–gel methods exhibit increased electrochemical performance, as confirmed by cyclic voltammetry (CV) measurements, which show higher current densities at lower overpotentials.[93] This indicates reduced activation energy, directly attributed to the nitrogen doping and the porous gel structure that facilitates electron and ion transport. Furthermore, electrochemical impedance spectroscopy (EIS) is often used to demonstrate decreased charge transfer resistance in gel-derived catalysts compared to non-porous counterparts, showing enhanced ion mobility and faster reaction kinetics.[94] Another example is the use of scanning electron microscopy (SEM) and nitrogen adsorption–desorption analysis to measure the pore structure and specific surface area of gel-based catalysts, which directly correlate with increased diffusion rates. In one study, incorporating metal oxides into carbon gels created hierarchical porosity, resulting in superior oxygen reduction activity, evidenced by linear sweep voltammetry (LSV) that showed lower onset potentials.[95] The development of hybrid composites that combine gel-derived carbon materials with metals or metal oxides is a promising avenue, yielding synergistic effects that boost material performance.[92,96] As a result, gel-derived carbon-based materials are poised to make significant contributions to sustainable technologies and high-performance devices.

3.7 Recent advances in gel material synthesis

Recent advancements in gel material synthesis have focused on expanding the repertoire of available gel precursors, optimizing synthesis conditions, and developing advanced characterization techniques to elucidate gel structure–property relationships. In the realm of gel precursors, researchers have explored new monomers, crosslinkers, and initiator systems to enable the synthesis of gels with enhanced properties and functionalities. For example, the development of bio-based monomers and crosslinkers has garnered interest due to their sustainability, biocompatibility, and tunable properties.[97] Moreover, advances in controlled polymerization techniques, such as living polymerization and controlled radical polymerization, have facilitated the synthesis of well-defined polymer architectures with precise control over molecular weight, dispersity, and end-group functionality. These controlled polymerization methods enable the synthesis of gels with tailored mechanical properties, degradation kinetics, and stimuli responsiveness, opening up new avenues for applications in drug delivery, tissue engineering, and regenerative medicine.[98] Furthermore, the integration of responsive moieties, such as pH-sensitive groups, temperature-responsive polymers, or photoresponsive molecules, into gel networks has enabled the development of smart gels that exhibit programmable behaviors in response to external stimuli.

In addition to advancements in gel precursor design, researchers have focused on optimizing synthesis conditions to control the gel structure and properties. Strategies such as template-assisted synthesis, microfluidic fabrication, and 3D printing techniques offer precise control over the gel morphology, pore size distribution, and spatial organization, enabling the design of gels with tailored properties for specific applications. Template-assisted synthesis involves the use of sacrificial templates or molds to create hierarchical structures within the gel matrix, leading to enhanced mechanical properties, cell adhesion, or mass transport properties. Microfluidic fabrication techniques leverage the precise control over fluid flow and mixing at the microscale to produce gels with controlled composition, morphology, and functionality.[99] These microstructured gels find

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Catal. Sci. Technol., 2025, **15**, 1357–1389 | **1369**

applications in tissue engineering, drug screening, and biosensing. Furthermore, 3D printing technologies enable the direct fabrication of complex gel structures with spatially controlled properties, offering new opportunities for personalized medicine, tissue engineering, and biomedical devices. By optimizing synthesis conditions and leveraging advanced fabrication techniques, researchers can tailor gel materials to meet the demanding requirements of specific applications, ranging from regenerative medicine to environmental remediation.

4. Classification of gels

Gel classification is a crucial component of materials science and bioengineering, serving as a foundation for understanding the vast array of gel structures and their respective properties. The classification of gels is typically based on several criteria, including their origin, physical properties, and chemical composition. This systematic categorization aids in the precise application of gels across various fields, from medical therapeutics to industrial processes. In addition to these basic criteria, the classification of gels can be further refined by considering their network structure, rheological behavior, and responsiveness to external stimuli. This systematic categorization aids in the precise application of gels across various fields, from medical therapeutics to industrial processes.

4.1 Based on origin

One primary method of classifying gels is based on their origin, distinguishing between natural and synthetic gels. Natural gels are derived from biological sources such as polysaccharides (*e.g.*, alginate, agarose, and carrageenan) and proteins (*e.g.*, gelatin and collagen). These gels are often biocompatible and biodegradable, making them suitable for biomedical applications such as drug delivery and tissue engineering. For example, alginate, extracted from brown seaweed, forms gels in the presence of divalent cations and is widely used for encapsulating cells and drugs due to its gentle gelling conditions and biocompatibility.[100] Alginate gels, formed through ionotropic gelation with calcium ions, create a mild environment for encapsulated cells, preserving their viability and functionality, which is crucial for tissue engineering and regenerative medicine applications. Additionally, agarose, another polysaccharide derived from red algae, is frequently used in electrophoresis for DNA and protein separation due to its robust gel matrix and ease of preparation.[101] Carrageenan, a sulfated polysaccharide extracted from red seaweed, forms thermoreversible gels and is widely used in the food industry for its gelling, thickening, and stabilizing properties.[102] Carrageenan's ability to form gels in the presence of potassium or calcium ions is also exploited in drug delivery systems, where controlled release of active ingredients is required. Gelatin, a protein derived from collagen, exhibits thermoreversible gelation and is used

extensively in food, pharmaceuticals, and medical devices. Gelatin gels are known for their biocompatibility, biodegradability, and ability to form hydrogels at physiological temperatures, making them suitable for wound dressings, scaffolds in tissue engineering, and as carriers for drug delivery.[103] Collagen, the primary structural protein in the extracellular matrix of connective tissues, forms gels that mimic natural tissue environments, promoting cell adhesion, growth, and differentiation, which is crucial for regenerative medicine applications.[104]

On the other hand, synthetic gels are engineered from synthetic polymers such as polyacrylamide, polyethylene glycol (PEG), and polyvinyl alcohol (PVA). These gels offer tunable mechanical properties, stability, and the ability to incorporate a variety of functional groups, making them ideal for industrial applications like wastewater treatment and as scaffolds in regenerative medicine.[105] Polyacrylamide gels, for example, are widely used in electrophoresis for the separation of proteins and nucleic acids due to their stable and reproducible gel matrix.[106] The mechanical properties of polyacrylamide gels can be easily adjusted by varying the concentration of acrylamide and cross-linkers, allowing for the customization of gel stiffness for different applications. PEG-based hydrogels are particularly notable for their biocompatibility and low immunogenicity, making them suitable for drug delivery systems and tissue engineering scaffolds. PEG hydrogels can be designed to degrade under physiological conditions, providing a controlled release of encapsulated drugs or growth factors, which is essential for therapeutic applications.[107] Furthermore, PEG hydrogels can be functionalized with bioactive molecules to enhance cell adhesion and proliferation, facilitating tissue regeneration. PVA, known for its excellent film-forming, emulsifying, and adhesive properties, forms hydrogels through physical or chemical cross-linking. PVA hydrogels are used in medical applications such as wound dressings, artificial cartilage, and contact lenses due to their high water content, flexibility, and biocompatibility.[108] The versatility of PVA hydrogels in forming stable and elastic networks makes them suitable for applications requiring mechanical resilience and durability.

4.2 Based on the physical state

Another vital classification criterion is the physical state of the gels. Hydrogels, which are water-swollen polymer networks, represent a significant class of gels. Their highwater content and porosity make them suitable for applications requiring biocompatibility and mimicking biological tissues, such as wound dressings and contact lenses. Hydrogels can further be classified into conventional hydrogels, which remain in a swollen state once formed, and stimuli-responsive hydrogels, which can change their properties in response to environmental factors such as pH, temperature, and ionic strength. For instance, poly(*N*-isopropylacrylamide) (PNIPAM) hydrogels exhibit a lower critical solution temperature (LCST) of around 32 °C, above

1370 | *Catal. Sci. Technol.*, 2025, **15**, 1357–1389

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which they undergo a reversible phase transition from a swollen to a collapsed state, making them useful for drug delivery systems.[109] These stimuli-responsive hydrogels, also known as "smart hydrogels", are particularly advantageous in biomedical applications where controlled release or specific responses to environmental changes are required. Conventional hydrogels, typically formed by polymerizing hydrophilic monomers such as acrylamide, methacrylate, and their derivatives, are extensively used in medical and pharmaceutical fields. Their stable and consistent swelling behavior makes them ideal for applications like contact lenses, wound dressings, and drug delivery systems. For instance, polyacrylamide hydrogels, due to their non-toxic nature and excellent water absorption capabilities, are often employed in electrophoresis for the separation of biomolecules.[110] The uniform network structure of these hydrogels allows for predictable mechanical properties and diffusion rates, which is crucial for their performance in clinical and laboratory settings. Stimuli-responsive hydrogels, on the other hand, exhibit significant changes in their physical properties in response to external stimuli. This category includes pH-responsive hydrogels, temperature-responsive hydrogels, and ion-sensitive hydrogels. PNIPAM hydrogels, as a prime example, demonstrate temperature-sensitive behavior with an LCST of around 32 °C. Below this temperature, they remain hydrophilic and swollen, but above it, they become hydrophobic and collapse, expelling water. This reversible transition makes PNIPAM hydrogels highly useful in drug delivery systems where a temperature change can trigger the release of a therapeutic agent.[109] Similarly, pH-sensitive hydrogels, typically composed of polymers like poly(acrylic acid) or chitosan, respond to changes in the pH environment by swelling or deswelling, which can be exploited for targeted drug delivery in different parts of the gastrointestinal tract where pH varies significantly.[111]

In contrast, organogels are gels composed of a three-dimensional network of organic molecules dispersed in an organic solvent. These gels can be formed from low molecular weight gelators (LMWGs) or polymer-based gelators and are extensively used in applications where water-based gels are unsuitable, such as in the cosmetic and pharmaceutical industries. For example, LMWGs like cholesterol derivatives and sorbitan monostearate form organogels through non-covalent interactions, providing a matrix that can entrap lipophilic drugs, enhancing their stability and bioavailability.[112] Organogels are particularly beneficial in delivering hydrophobic drugs, which are otherwise challenging to administer due to their poor water solubility. The gelation mechanism in organogels typically involves non-covalent interactions such as hydrogen bonding, van der Waals forces, and π–π stacking, which provide the gel with a delicate balance of stability and responsiveness. Moreover, organogels can be tailored to exhibit stimuli-responsive behaviors similar to hydrogels. For instance, thermo-responsive organogels that transition between solid and liquid states at specific temperatures can be utilized in

topical drug delivery systems. The ease of incorporating various functional groups into the gelator molecules allows for the customization of organogels to meet specific application requirements. This versatility has led to the development of organogel-based systems in cosmetics, where they serve as carriers for active ingredients, and in pharmaceuticals, where they enhance the bioavailability and stability of drugs.

4.3 Based on the gelation mechanism

The cross-linking mechanism within the gel matrix is another critical factor in gel classification, influencing mechanical strength, swelling behavior, and responsiveness. Cross-linking can occur through physical or chemical means, each imparting distinct properties to the resulting gel.

Physical gels are formed through non-covalent interactions such as hydrogen bonding, ionic interactions, and van der Waals forces. These interactions create reversible and stimuli-responsive networks, making physical gels highly adaptable to changes in their environment. For example, gelatin forms physical gels through hydrogen bonding and helical aggregation. When heated, the gelatin molecules dissolve, breaking these bonds and transitioning to a sol state. Upon cooling, the helical structures re-form, resulting in a gel state. This thermoreversible property makes gelatin gels particularly useful in food products, where a smooth, melt-in-the-mouth texture is desired, and in biomedical applications, such as drug delivery and wound dressings, where the gel must conform to body temperatures.[113] Ionic gels are another type of physical gel where gelation occurs through ionic interactions. Alginate, a natural polysaccharide, forms gels in the presence of divalent cations such as calcium. The calcium ions bridge the guluronic acid blocks of alginate chains, creating an ionically cross-linked network. This process is mild and does not involve harsh chemicals or high temperatures, preserving the biological activity of encapsulated cells or drugs. Consequently, alginate gels are extensively used in cell encapsulation, tissue engineering, and drug delivery systems where biocompatibility and gentle processing conditions are crucial.[114]

Chemical gels, in contrast, are formed *via* covalent bonding, resulting in permanent and robust networks. These gels are characterized by their stable mechanical properties and resistance to environmental changes. Polyacrylamide gels, a common example, are chemically cross-linked using N,N'-methylenebisacrylamide. The covalent bonds formed during polymerization create a stable and resilient gel matrix that can withstand mechanical stresses and maintain its structure over extended periods. Polyacrylamide gels are widely used in electrophoresis for the separation of proteins and nucleic acids due to their uniform pore sizes and consistent performance, which are critical for reproducible analytical results.[115] Another example of chemically cross-linked gels is polyethylene glycol (PEG) hydrogels. PEG hydrogels are synthesized through the covalent cross-linking

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Catal. Sci. Technol., 2025, **15**, 1357–1389 | **1371**

of PEG chains, often using photopolymerization techniques. The resulting hydrogels are biocompatible and can be engineered to degrade under physiological conditions, making them suitable for drug delivery and tissue engineering applications. The mechanical properties and degradation rates of PEG hydrogels can be finely tuned by adjusting the molecular weight of the PEG and the density of cross-links, allowing for customized performance tailored to specific medical needs.[116] The mechanical properties of gels are directly influenced by the nature and density of cross-links. Physically cross-linked gels tend to be softer and more flexible due to the reversible nature of their interactions. These gels can swell significantly in the presence of water, which is advantageous for applications requiring high water content and porosity, such as in wound dressings and contact lenses. In contrast, chemically cross-linked gels typically exhibit higher mechanical strength and lower swelling ratios. The permanent covalent bonds provide structural integrity and resistance to deformation, which is essential for applications like scaffolds in tissue engineering where mechanical support and stability are required. Stimuli-responsive gels, a subset of physically cross-linked gels, can change their properties in response to external stimuli such as temperature, pH, and ionic strength.

4.4 Based on their structural organization

Moreover, gels can also be classified based on their structural organization into homogeneous and heterogeneous gels. This classification impacts their mechanical properties, diffusion behavior, and potential applications, particularly in fields such as drug delivery and tissue engineering.

Homogeneous gels exhibit a uniform network structure throughout their matrix, which results in consistent mechanical and diffusion properties. These gels are typically formed through well-controlled polymerization processes that ensure even distribution of cross-linking points and polymer chains. As a result, homogeneous gels display uniform mechanical strength and elasticity, which are crucial for applications that require predictable and reliable performance. For instance, homogeneous hydrogels are widely used in contact lenses, where consistent optical and mechanical properties are essential for user comfort and visual clarity.[117] The uniform network structure of homogeneous gels also leads to predictable and consistent diffusion pathways. This characteristic is particularly advantageous in drug delivery applications, where the controlled release of therapeutic agents is required. In such systems, the diffusion of drugs through the gel matrix follows a predictable pattern, enabling precise control over the release kinetics. Polyethylene glycol (PEG) hydrogels, known for their homogeneous network, are often employed in drug delivery due to their biocompatibility and ability to release drugs at a controlled rate.[116] The homogeneous nature of these gels ensures that the encapsulated drugs are evenly distributed, providing a steady and sustained release over time.

In contrast, heterogeneous gels have phase-separated domains within their structure, resulting in varied mechanical properties and heterogeneous diffusion pathways. These gels are typically formed through processes that lead to the segregation of different polymer components or the incorporation of particulate fillers, creating distinct regions within the gel matrix. The presence of these phase-separated domains can impart unique mechanical properties to heterogeneous gels, such as anisotropy and enhanced toughness. This structural heterogeneity can be beneficial in applications that require materials with varying mechanical properties, such as tissue engineering scaffolds that mimic the complex architecture of natural tissues.[111] The heterogeneous structure of these gels also influences their diffusion behavior. Unlike homogeneous gels, heterogeneous gels exhibit varied diffusion pathways, which can be exploited for controlled release applications. The presence of distinct domains within the gel matrix can create regions of differing diffusivity, allowing for the targeted release of encapsulated agents. For example, in drug delivery systems, heterogeneous gels can be designed to have regions with faster or slower diffusion rates, enabling a more complex release profile that can better match the therapeutic needs of patients. This capability is particularly useful for the delivery of multiple drugs or the staged release of a single drug, where different release rates are required at different times.[80] Additionally, the structural organization of gels can be tailored to create hybrid gels that combine the properties of both homogeneous and heterogeneous gels. These hybrid systems can offer the benefits of uniform mechanical strength and controlled diffusion while incorporating phase-separated domains that provide additional functionality. For instance, hybrid gels can be engineered to have a homogeneous network for structural support, with embedded nanoparticles or microdomains that release drugs or other therapeutic agents in response to specific stimuli.[118]

In conclusion, the classification of gels based on their origin, physical state, cross-linking mechanism, and structural organization provides a comprehensive framework for understanding and utilizing these versatile materials. This nuanced classification not only facilitates the targeted application of gels in diverse fields but also drives the development of novel gels with tailored properties for specific industrial, medical, and environmental uses.

5. Energy applications of gel materials

Gel materials have garnered significant attention in the field of energy applications due to their versatile properties, including high surface area, tunable porosity, flexible mechanical characteristics, and the ability to encapsulate functional materials. These unique attributes make gels ideal for a wide range of energy-related uses, from energy storage and conversion to environmental remediation. This review explores

1372 | *Catal. Sci. Technol.*, 2025, **15**, 1357–1389

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the various roles of gels in energy applications, detailing their benefits, challenges, and future research directions.

5.1 Energy storage applications

5.1.1 Lithium-ion batteries (LIBs). Lithium-ion batteries (LIBs) have revolutionized energy storage technologies, powering a wide range of applications from portable electronics to electric vehicles. However, the performance, safety, and longevity of LIBs are highly dependent on the electrolytes used within them. Traditional liquid electrolytes, though effective, pose significant challenges such as leakage, flammability, and limited operational temperature ranges. Gel polymer electrolytes (GPEs) mitigate these issues by blending the ionic conductivity of liquid electrolytes with the mechanical stability of solid polymers. Poly(vinylidene fluoride-hexafluoropropylene) (PVDF-HFP) based gels have garnered extensive attention due to their high ionic conductivity and good mechanical properties. The integration of nanoscale fillers like silica nanoparticles into these gel electrolytes has further enhanced their performance by improving both ionic conductivity and mechanical strength.[119,120] One significant advantage of GPEs is their tunability. By modifying the polymer composition and degree of cross-linking, and incorporating various additives such as plasticizers and nanoparticles, researchers can enhance the ionic conductivity, mechanical strength, and thermal stability of GPEs. For example, adding nanoscale fillers like silica (SiO_2) or titanium dioxide (TiO_2) can improve the ionic

conductivity by creating additional pathways for ion transport and reinforcing the polymer matrix, thereby enhancing the overall mechanical properties.[121] A new polymer electrolyte with enhanced lithium-ion conductivity has been developed to address the issue of low ionic conductivity in conventional polymer electrolytes (in Fig. 10a and b).[122,123]

This electrolyte incorporates a fluorinated ionic liquid, which strengthens anion coordination while weakening cation coordination, thereby improving lithium transport and preventing dendrite formation. Using living radical polymerization, a gradient copolymer of styrene (St) and methyl acrylate (MA) is synthesized. The gradient structure ensures a smooth change in monomer composition along the polymer chains, eliminating sharp domain boundaries and facilitating efficient ion pathways. This gel electrolyte reaches an ionic conductivity of 1.20×10^{-3} S cm^{-1} at room temperature, making it suitable for lithium-ion batteries. In Fig. 10c, the preparation methods for gel polymer electrolytes include: *in situ* polymerization directly on cell components for application in lithium-ion cells and the creation of a self-standing membrane, utilized for both conductivity and LSV evaluations.[124]

The performance of LIBs heavily depends on the ionic conductivity of the electrolyte. GPEs can achieve ionic conductivities comparable to those of liquid electrolytes through careful selection of polymer matrices and the incorporation of conductive fillers. PEO-based GPEs, for example, when doped with lithium salts and plasticizers, exhibit high ionic conductivities and good compatibility with

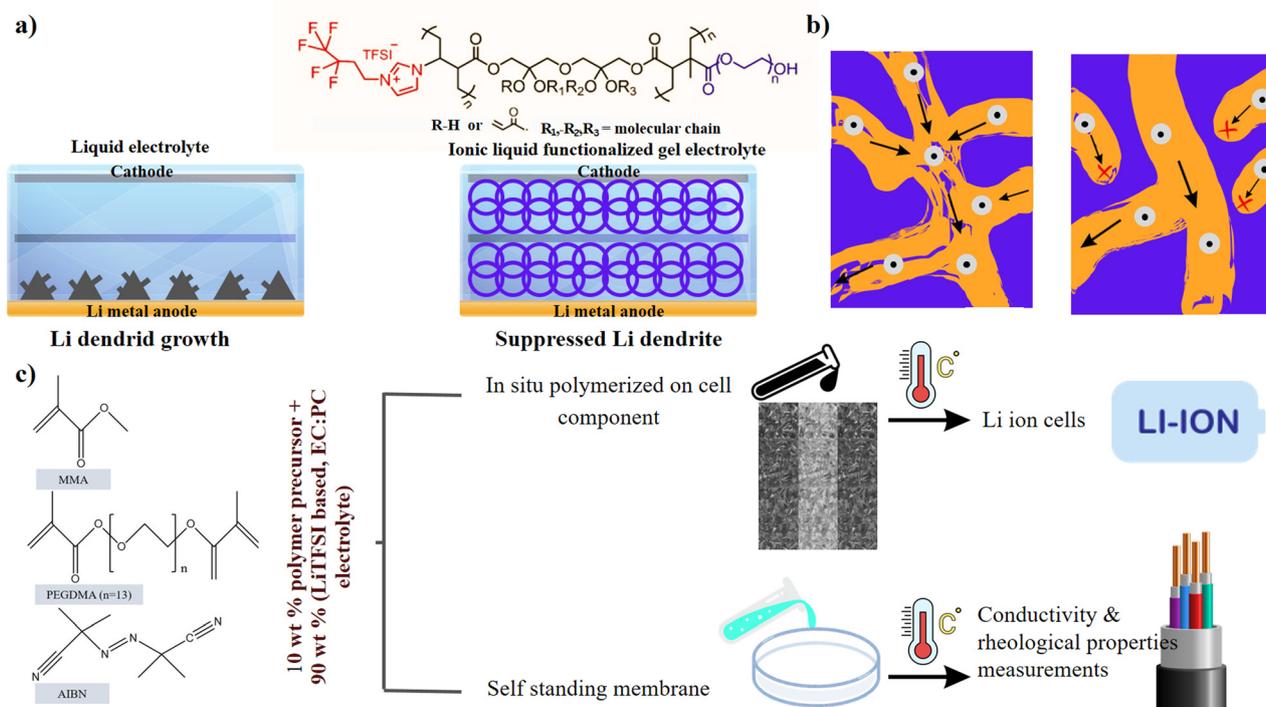**Fig. 10** Schematic of gradient copolymer gel electrolytes for lithium-ion batteries: (a) ionic liquid-functionalized precursors, (b) ion conduction mechanism, (c) lithium metal battery setup, and (d) synthesis process with monomers (MMA, PEGDMA, AIBN) and electrolyte incorporation.

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Catal. Sci. Technol., 2025, **15**, 1357–1389 | **1373**

Table 2 Evaluation of various gel polymer electrolyte compositions, including their synthesis methods, ionic conductivities, charge capacities, and cycle stability performance in lithium-ion batteries

Components of gel polymer electrolytes	Synthesis method	Ionic conductivity (S cm^{-1})	Capacity (mA h g^{-1})	Cycle stability	Ref.
E2BADMA/PEGDA/LiTFSI	*In situ* polymerization	4.4×10^{-4}	170 (at 0.1 °C)	82.35% after 100 cycles	126
P(VDF-HFP)/PEGDGE/EMITFSI	*In situ* polymerization	1.69×10^{-3} at 25 °C	162 (at 0.1, 60 °C)	96.1% after 50 cycles (at 60 °C)	127
Methacrylate monomer/POSS nanoparticles	*In situ* polymerization	7.71×10^{-4} at 30 °C	126 (at 0.5 °C)	Stability > 960 cycles	128
PVDF-HFP/Li$_{10}$GeP$_2$S$_{12}$	*In situ* polymerization	2.5×10^{-3} at 20 °C	142 (at 0.5 °C)	85% over 500 cycles	129
PETEA/cystine/ICEM/LiTFSI	*In situ* polymerization	1.1×10^{-3} at 25 °C	135.4 (at 0.1 °C)	Stability > 1200 h	130
PEGMEA/LiTFSI/liquid carbonates	*In situ* polymerization	1.73×10^{-4} at 60 °C	158.5 (at 0.5 °C)	98.87% after 100 cycles	131
PEGDMA/TFMA/LIF	*In situ* thermal polymerization	3.1×10^{-3}	170 (at 0.5 °C)	90% after 1000 cycles	132
PVDF-HFP/LLZTO	UV curing process	3.0×10^{-4} at 25 °C	126 (at 0.5 °C)	Stability > 960 cycles	133
PDOL/LiTFSI in PAN/PLLA matrix	Electrospinning/immersion	1.2×10^{-3}	151.79 (at 1 °C)	98.19% after 100 cycles	134
PVDF-HFP/CA/SN/ZIF-8 NPS	Electrospinning/solution casting	3.44×10^{-3}	257.5 (at 0.2 °C)	84.6% after 500 cycles	135
Polyacrylonitrile (PAN)/LiTFSI	Electrospinning	4.2×10^{-3} at 25 °C	300 (1 °C)	87% after 200 cycles	136
Single-ion conducting siloxane-based GPE	Electrospinning/immersion	1.5×10^{-3}	315 (at 0.1 °C)	97.5% after 300 cycles	137
PVDF-HFP/ZIF-8	Solution casting MOF addition	4.2×10^{-4} at 30 °C	138 (at 0.2 °C)	Stable up to 200 cycles	138
PVDF-HFP/LiTFSI	Solution casting	8.0×10^{-4} at 25 °C	870.3 (at 0.1 °C)	48.28% after 20 cycles	139
PVDFHFP/LiTFSI	Solution casting	3.11×10^{-5} at 25 °C	87.5 (at 0.5 mA cm^{-1})	93.26% after 830 cycles	140
PVDF-HFP/PP13TFSI/LiTFSI	Solution casting	—	128.7	Stability > 400 cycles	141
MXenes/PVDF-HFP/LiTFSI	Solution casting	8.1×10^{-4}	900 (at 0.5 °C)	74% after 500 cycles	142
PVDF-HFP/SN-based electrolyte	Solution casting	1.5×10^{-3} at 25 °C	200 (0.5 °C)	85% after 300 cycles	143
3D crosslinked PVDF-HFP/PMMA	Solution casting	1.23×10^{-3}	152 (at 0.2 °C)	91% after 100 cycles	144
PEO/PMMA/LiTFSI	Solution casting	1.0×10^{-3} at 25 °C	160 (1 °C)	95% after 100 cycles	145
PVDF-HFP/SiO$_2$ nanocomposite	Solution blending	3.6×10^{-4} at 25 °C	135 (at 0.1 °C)	95% after 600 cycles	146
PVDF-HFP/metal–organic framework	Gel casting	3.8×10^{-4} at 25 °C	145 (at 0.1 °C)	Stability over 500 cycles	147
PVDF-HFP/DMMP	Solvent evaporation	2.4×10^{-3} at 25 °C	250 (0.2 °C)	90% after 400 cycles	148
PVDF-HFP/PEA/PP13TFSI	Photopolymerization	9.5×10^{-4} at 25 °C	153.7 (0.5 °C)	92.7% after 500 cycles	149
PEO/PA/TEGDME/LiTFSI	Photopolymerization	6.4×10^{-4} at 25 °C	140 (at 0.5 °C)	88.57% after 100 cycles	150

lithium metal anodes, making them suitable for high-energy-density batteries.[119] In LMBs, solid-state gel polymer electrolytes (GPEs) have demonstrated remarkable efficacy in inhibiting the formation of lithium dendrites, resolving this important issue and opening the door for their practical use. A thorough comparison of the different GPE compositions for Li-metal batteries is given in Table 2, which also includes information on the preparation process, specific capacity, cyclic stability, and ionic conductivity. Recent advancements include polymer-in-salt electrolytes, where a high concentration of lithium salt is dissolved in a minimal amount of polymer. This approach reduces the crystallinity of the polymer matrix, facilitating ion transport and significantly improving battery performance. Moreover, the development of single-ion conducting polymers, where only lithium ions are mobile, minimizes side reactions and enhances battery efficiency by reducing ion pairing and increasing the transference number.[120] Furthermore, incorporating ionic liquids into gel electrolytes can enhance thermal stability and electrochemical performance, making them suitable for high-temperature applications.[125]

5.1.2 Supercapacitors. Supercapacitors are renowned for their rapid charge and discharge capabilities, which make them suitable for applications requiring high power density. These characteristics are crucial for a range of modern technologies, including regenerative braking systems in electric vehicles, power backup systems, and portable electronic devices. One of the key advancements in supercapacitor technology involves the use of gel materials, particularly hydrogels, which serve as both electrodes and electrolytes. Hydrogels, with their high water content and porous structure, facilitate efficient ion transport, significantly enhancing the capacitance of supercapacitors.

Polyacrylamide-based hydrogels embedded with conductive polymers such as polyaniline (PANI) or polypyrrole (PPy) have demonstrated notable improvements in supercapacitor performance. These conductive polymers are well known for their high electrical conductivity and excellent electrochemical stability, which are essential for energy storage applications. For instance, Zhang *et al.* (2016) demonstrated that hydrogels incorporating polyaniline exhibited enhanced specific capacitance and better cycling stability compared to traditional electrode materials.[151] The
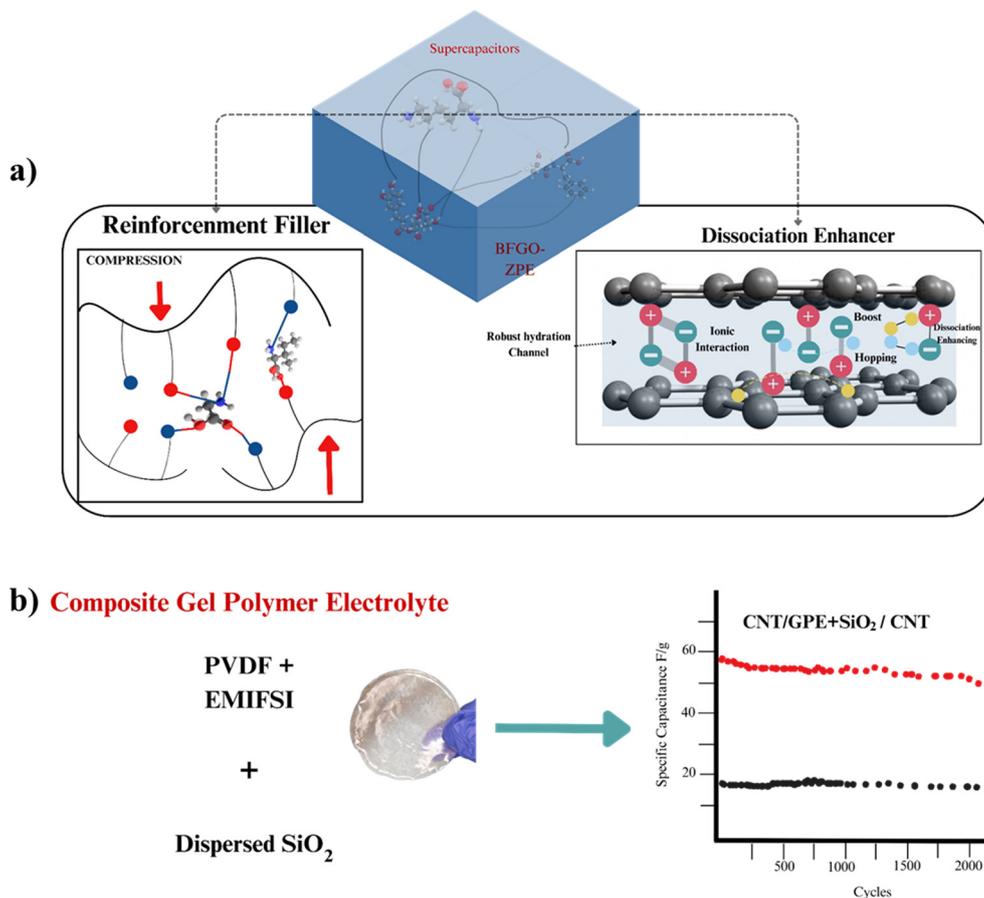
1374 | *Catal. Sci. Technol.*, 2025, **15**, 1357–1389

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Fig. 11 Composite gel polymer electrolyte incorporating reinforcement fillers and dissociation enhancers (a) with dispersed SiO$_2$ (b) for enhanced ionic conductivity, mechanical stability, and improved supercapacitor cycling performance.

polyacrylamide matrix provides a robust and flexible scaffold that supports the conductive polymer network, ensuring efficient electron and ion transport during the charge–discharge cycles. Research has also focused on developing hybrid hydrogels that combine conductive materials with traditional hydrogel matrices to further improve supercapacitor performance. For example, graphene oxide (GO) and carbon nanotubes (CNTs) have been incorporated into hydrogel matrices to enhance their electrical conductivity and mechanical strength. A. Watanabe (2021) reported that incorporating graphene oxide into hydrogels resulted in a significant increase in electrical conductivity and mechanical robustness.[152] A new strategy to improve both ionic conductivity and mechanical strength of zwitterionic polymer electrolytes incorporates betaine-functionalized graphene oxide. This approach introduces a composite gel polymer electrolyte (CGPE) that combines a reinforcement filler and a dissociation enhancer. The reinforcement filler provides compression resistance and mechanical durability, while the dissociation enhancer facilitates ion hopping and interactions, establishing robust hydration channels that enhance ionic conductivity as shown in Fig. 11(a).[153] This advanced electrolyte is highly effective for flexible and high-performance supercapacitors, enhancing

device stability and capacitance. Furthermore, Fig. 11(b) shows a gel nanocomposite polymer electrolyte consisting of nanostructured SiO$_2$, PVDF, and imidazolium which further elevate supercapacitor performance, offering an innovative solution for energy storage systems.[154] These hybrid hydrogels exhibited superior electrochemical properties, including high specific capacitance, excellent rate capability, and long cycle life.

The incorporation of graphene oxide into hydrogels leverages the exceptional electrical, thermal, and mechanical properties of graphene. Graphene oxide's high surface area and excellent electrical conductivity make it an ideal material for improving the performance of hydrogel-based supercapacitors. When integrated into the hydrogel matrix, graphene oxide provides additional conductive pathways and structural reinforcement, leading to enhanced ion transport and mechanical stability.[155] Similarly, carbon nanotubes, known for their high electrical conductivity and tensile strength, contribute to the overall performance and durability of the hybrid hydrogel supercapacitors.[156] Moreover, hybrid hydrogels can be designed to respond to environmental stimuli, such as pH, temperature, and light, making them suitable for a wide range of applications. Stimuli-responsive hydrogels can undergo reversible changes in their structure

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Catal. Sci. Technol., 2025, **15**, 1357–1389 | **1375**

and properties, allowing for tunable performance characteristics in supercapacitors. For instance, temperature-responsive hydrogels can exhibit changes in swelling behavior, affecting ion transport and capacitance depending on the operating conditions. This tunability can be particularly advantageous in applications where environmental conditions vary significantly.[157] The development of these advanced hydrogel-based materials aligns with the growing demand for flexible, lightweight, and high-performance energy storage devices. Flexible supercapacitors, which can be integrated into wearable electronics and other portable devices, benefit greatly from the mechanical flexibility and high ionic conductivity of hydrogel-based materials. Poly(vinyl alcohol) (PVA)-based hydrogels, for example, are highly flexible and can maintain their electrochemical performance under mechanical deformation, making them ideal for use in flexible and wearable supercapacitors.[158]

5.1.3 Metal–air batteries. Metal–air batteries, particularly lithium–air batteries, are gaining significant attention due to their high theoretical energy densities, which are substantially greater than those of conventional lithium-ion batteries. This makes them highly attractive for applications that demand long-lasting energy storage, such as electric vehicles and renewable energy systems. However, the practical implementation of lithium–air batteries faces several challenges, primarily related to electrolyte leakage and the formation of lithium dendrites, which can lead to short circuits and battery failure. Gel electrolytes have emerged as a promising solution to these issues, providing both ionic conductivity and mechanical stability to inhibit dendrite growth.

Gel electrolytes based on polymers such as polyethylene oxide (PEO) and polyvinylidene fluoride (PVDF) have been extensively studied due to their potential in lithium–air batteries. PEO, a well-known solid polymer electrolyte, offers high ionic conductivity at elevated temperatures and good mechanical properties, which help to suppress dendrite formation. PVDF, on the other hand, is valued for its chemical stability and electrochemical performance. The use of these polymers in the gel form combines the benefits of solid and liquid electrolytes, maintaining ionic conductivity while providing the necessary mechanical support to prevent dendrite penetration.[159] To enhance the performance of gel electrolytes, researchers have focused on incorporating various additives that can improve ionic conductivity and stability. For instance, the addition of lithium salts such as lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) can significantly increase the ionic conductivity of the gel electrolytes. LiTFSI is known for its high dissociation in polymer matrices, leading to a greater number of mobile lithium ions. Moreover, the incorporation of ionic liquids (ILs) into gel electrolytes has shown promising results. ILs, which are salts in the liquid state at room temperature, provide excellent ionic conductivity and thermal stability, and they can also enhance the electrochemical stability of the gel electrolytes, making them more suitable for practical applications.[159]

Another approach to optimizing gel electrolytes involves the use of nanofillers, such as silica nanoparticles, carbon nanotubes, and graphene. These nanomaterials can create a more uniform and stable structure within the gel, enhancing both mechanical properties and ionic conductivity. For example, silica nanoparticles can help to create a more rigid framework that supports the polymer matrix, reducing the risk of dendrite formation. Carbon nanotubes and graphene, with their high electrical conductivity and mechanical strength, can further improve the overall performance of the gel electrolytes, making them more robust and efficient for use in lithium–air batteries. The development of advanced gel electrolytes is also focused on achieving a balance between ionic conductivity and mechanical stability. An ideal gel electrolyte should possess high ionic conductivity to ensure efficient ion transport and low internal resistance, as well as sufficient mechanical strength to withstand the formation of lithium dendrites. This balance can be achieved through careful selection of polymer matrices, optimization of additive concentrations, and incorporation of nanomaterials to reinforce the gel structure. Recent studies have also explored the potential of hybrid gel electrolytes, which combine the properties of different polymers and additives to achieve superior performance. For instance, a hybrid gel electrolyte composed of PEO and PVDF, combined with lithium salts and ionic liquids, can offer enhanced ionic conductivity, thermal stability, and mechanical strength. This synergistic approach leverages the advantages of each component, resulting in a more effective and durable electrolyte for lithium–air batteries.[160]

5.2 Energy conversion applications

Energy conversion applications have seen significant advancements with the introduction of gel materials, which offer unique structural and functional benefits. Gels, including hydrogels, organogels, and aerogels, provide a versatile platform for energy technologies, acting as electrolytes, separators, or electrode materials. Their high ionic conductivity, flexibility, and thermal stability make them ideal for various systems, such as lithium-ion batteries, fuel cells, and supercapacitors, where they enhance electrochemical performance by facilitating ion transport.[161] Gels can also be engineered with nanoparticles, conductive polymers, or carbon-based materials to improve energy storage and transfer capabilities. In solar energy conversion, photoactive gels are used in dye-sensitized solar cells (DSSCs) for superior light absorption and charge transport, while their soft, porous structures make them suitable for flexible, wearable energy devices—vital for advancements in portable electronics and renewable energy.[162] The accompanying Fig. 12 demonstrates how conductive self-healing gels apply to energy and electrochemical devices, highlighting their role in solar cells, thermoelectric devices, rechargeable batteries,
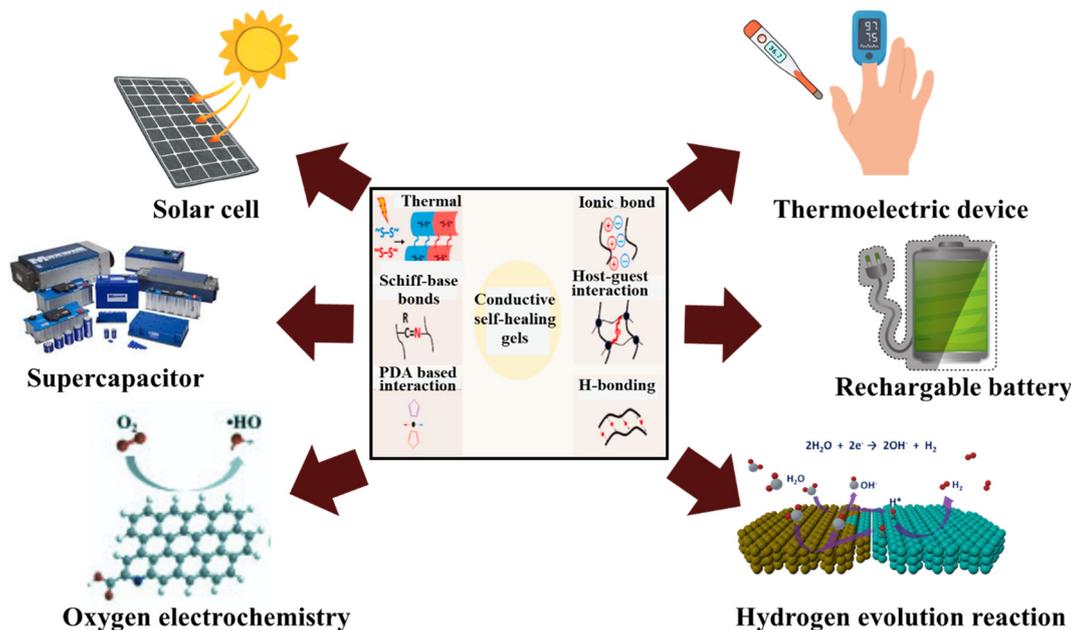
1376 | *Catal. Sci. Technol.*, 2025, **15**, 1357–1389

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Fig. 12 Applications of conductive self-healing gels in energy conversion and electrochemical devices.

supercapacitors, hydrogen evolution reactions, and oxygen electrochemistry. These gels leverage various bonding mechanisms, such as thermal bonds, ionic bonds, and H-bonding, to provide self-healing properties that enhance the durability and efficiency of these systems.[19] Overall, gel materials are critical to the future of energy conversion technologies, contributing to improved efficiency, sustainability, and adaptability.

In energy conversion processes like those in fuel cells and CO_2 reduction reactions, gels act as scaffolds that enhance electron transfer by creating a supportive environment for redox-active species. These polymer-based matrices incorporate functional groups or metal sites that improve electron pathways and mobility. Gels facilitate electron movement through semi-structured, conductive pathways formed by embedded nanoparticles, polymers, or ionic clusters, which support faster electron transfer. Additionally, gel structures increase electron density and influence localization by redistributing active sites, enhancing electron delocalization, which benefits reaction kinetics. Specific interactions within the gel, like hydrogen bonding and coordination with metal ions, further stabilize charge carriers or create migration pathways, making gels highly effective in boosting electron transfer and reaction efficiency in energy conversion applications.

5.2.1 Fuel cells. A fuel cell is a highly efficient and eco-friendly energy conversion device that operates through electrochemical reactions occurring at two primary electrodes: the anode and the cathode. These reactions generate electricity by facilitating the movement of charged ions between the electrodes. Central to this process is the electrolyte, which not only supports ion flow but also contains catalysts that expedite the surface reactions at the

electrodes. The principal fuel for fuel cells is hydrogen, complemented by the requirement for oxygen or air. One of the foremost advantages of fuel cells is their minimal environmental impact, as the primary by-product of the hydrogen and oxygen reaction is water, resulting in significantly reduced pollution compared to traditional combustion-based power generation methods. The architecture of a fuel cell includes an electrolyte layer flanked by a porous anode and cathode. For practical applications, multiple fuel cells are often assembled into a stack to produce sufficient direct current (dc) to power various devices, such as electric motors or lighting systems.[163] In a dual-chamber fuel cell, the fuel, typically hydrogen, enters the anode compartment, while an oxidant, such as oxygen, enters the cathode compartment. These two compartments are selectively separated by the electrolyte, which permits the conduction of charge carriers such as protons (H^+), carbonate ions (CO_3^{2-}), oxide ions (O^{2-}), and hydroxide ions (OH^-) between the electrodes. This selective separation is crucial for maintaining the efficiency and directionality of the electrochemical reactions within the cell.

Gel materials play a pivotal role in enhancing the performance of various types of fuel cells, particularly proton exchange membrane fuel cells (PEMFCs) and microbial fuel cells (MFCs). In PEMFCs, gel electrolytes are essential for providing high proton conductivity while maintaining the necessary flexibility and durability for prolonged operation. Nafion-based gels are particularly notable for their ability to retain hydration and proton conductivity under varying environmental conditions, which is critical for the stable operation of PEMFCs.[164] Recent advancements in this field have led to the development of composite gels that integrate Nafion with other polymers or inorganic fillers, thereby
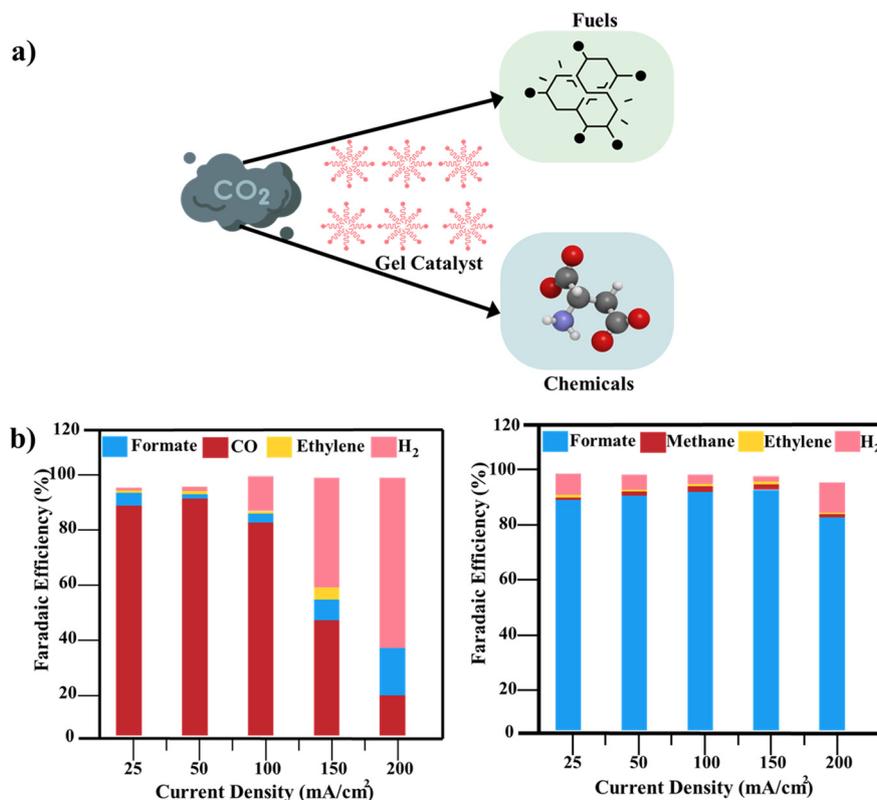
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Catal. Sci. Technol., 2025, **15**, 1357–1389 | **1377**

Fig. 13 CO_2 reduction using gel catalysts: (a) pathways for fuel and chemical production, (b) faradaic efficiencies for various products at different current densities, highlighting selective and efficient catalytic performance.

enhancing both the mechanical strength and proton conductivity of the electrolyte.[165] In the realm of microbial fuel cells, hydrogels serve an important function by immobilizing microorganisms, thus stabilizing and enhancing the bioelectrochemical processes involved in energy conversion. Hydrogels derived from natural polymers such as alginate and chitosan are particularly advantageous due to their biocompatibility and ability to maintain the activity of encapsulated microorganisms.[166] Furthermore, the incorporation of conductive materials such as carbon nanotubes or graphene into these hydrogels can significantly improve electron transfer rates and overall cell performance.[167]

The advancements in gel materials for fuel cells highlight ongoing innovation aimed at enhancing both efficiency and durability. The incorporation of composite gels and conductive hydrogels demonstrates the trend toward improving the mechanical and electrochemical properties of fuel cell components. These advancements not only elevate fuel cell performance but also broaden their applicability in diverse sectors, including portable power, stationary power generation, and transportation.[19] As illustrated in Fig. 13(a), gel catalysts facilitate CO_2 reduction pathways, enabling the production of fuels and valuable chemicals. In Fig. 13(b), the faradaic efficiency of different reduction products across varying current densities showcases how gel catalysts influence the distribution of compounds like formate, CO, methane, ethylene, and hydrogen. The left chart emphasizes

CO and hydrogen production at higher current densities, while the right chart highlights formate as the main product at lower densities, underscoring the effectiveness of gel catalysts in selectively producing desired compounds through CO_2 reduction. The pursuit of innovations in fuel cell technology is driven by the need for sustainable and clean energy solutions. Fuel cells, known for their high efficiency and low environmental impact, present a promising alternative to traditional fossil fuels. The use of advanced materials, such as Nafion-based gels, composite electrolytes, and conductive hydrogels, shows potential for significant technological improvements.[168] The potential of fuel cells to revolutionize energy generation and usage cannot be understated. As research progresses and new materials and technologies are developed, the efficiency, reliability, and affordability of fuel cells are expected to improve, paving the way for their widespread adoption. The development of fuel cells that are not only efficient but also cost-effective and environmentally friendly is a critical step towards achieving a sustainable energy future.

5.2.2 Solar cells. Gel materials have been extensively explored to improve the efficiency and stability of solar cells, particularly DSSCs. Gel electrolytes are beneficial in DSSCs as they help maintain the necessary redox reactions while preventing issues associated with liquid electrolytes, such as leakage and evaporation. Poly(ethylene oxide) (PEO)-based gels, doped with iodine/iodide redox couples, enhance DSSC

performance by providing a stable ionic conductive medium.[169] These PEO-based gels offer a practical solution by ensuring a steady flow of ions, which is critical for the continuous operation of the solar cell. The gel state of these electrolytes eliminates the risks associated with liquid electrolytes, such as spillage and degradation over time, thus extending the operational lifespan of DSSCs. The incorporation of nanomaterials into gel electrolytes can significantly improve the photovoltaic performance of DSSCs. Nanomaterials such as quantum dots, graphene, and metal nanoparticles have been employed to enhance light absorption and charge transfer properties. For instance, quantum dots can be tuned to absorb a broad spectrum of light, increasing the amount of captured solar energy and thus boosting the power conversion efficiency (PCE) of DSSCs. Graphene, with its exceptional electrical conductivity and high surface area, facilitates faster electron transport, reducing recombination losses and enhancing overall efficiency.[170] Metal nanoparticles, such as gold and silver, have also been integrated into gel electrolytes to utilize their plasmonic properties. These nanoparticles can increase light scattering within the cell, allowing more light to be absorbed by the dye molecules, which directly contributes to a higher PCE.[171] Additionally, the synergistic effects of combining various nanomaterials can further optimize the performance of gel electrolytes. For example, a hybrid gel electrolyte containing both graphene and quantum dots has shown improved stability and efficiency compared to gels with single nanomaterial additives.[172]

Recent research has also explored the use of ionic liquids in gel electrolytes to enhance the performance of DSSCs. Ionic liquids offer high ionic conductivity, thermal stability, and a wide electrochemical window, making them ideal candidates for incorporation into gel electrolytes. When combined with PEO-based gels, ionic liquids can create a more efficient ion-conducting network, further improving the DSSC's overall performance.[173] These ionic liquid-based gel electrolytes also exhibit lower volatility and higher thermal stability, which are crucial for the long-term operation of solar cells under varying environmental conditions. Furthermore, the development of solid-state gel electrolytes has gained attention due to their potential to provide even greater stability and durability for DSSCs. Solid-state electrolytes eliminate the issues related to liquid or semi-liquid electrolytes, such as leakage and crystallization. Advances in polymer chemistry have led to the creation of solid-state gels with enhanced ionic conductivity and mechanical strength, which are critical for the robust performance of DSSCs in real-world applications.[174] In addition to enhancing the efficiency and stability of DSSCs, gel electrolytes have shown promise in improving the environmental sustainability of solar cells. Many gel materials can be synthesized from renewable resources, and their use can reduce the reliance on volatile organic solvents traditionally used in liquid electrolytes. This shift towards more sustainable materials aligns with the broader goals of green chemistry and sustainable energy.[175]

5.2.3 Thermoelectric devices. Thermoelectric devices, which convert temperature differences into electrical energy, have seen significant advancements through the incorporation of gel materials. Conductive polymer gels, such as those based on poly(3,4-ethylenedioxythiophene) (PEDOT) and polyaniline, are particularly notable for their potential in forming flexible and efficient thermoelectric materials. These gels offer a unique combination of mechanical flexibility and electrical conductivity, making them ideal for applications where traditional rigid thermoelectric materials fall short. Flexible thermoelectric generators (TEGs) constructed from these gels can efficiently harvest waste heat from various sources, including industrial processes and wearable electronics.[176] This ability to convert otherwise wasted thermal energy into useful electrical power presents a promising route for enhancing energy efficiency in a range of applications. Recent research has focused on improving the performance of these polymer gels by incorporating inorganic thermoelectric materials. Hybrid gels that integrate materials such as bismuth telluride (Bi_2Te_3) or silicon nanowires into the polymer matrix have demonstrated enhanced thermoelectric properties. These hybrid gels benefit from the high electrical conductivity and setback coefficient of the inorganic components while maintaining the flexibility and processability of the polymer matrix. The result is a material that not only improves the efficiency of heat-to-electricity conversion but also offers practical advantages in terms of device fabrication and application versatility.[177,178]

For instance, the incorporation of bismuth telluride into a PEDOT gel matrix has been shown to significantly enhance the power factor of the resulting material. Bismuth telluride is one of the most efficient thermoelectric materials at room temperature, and its integration into a polymer gel matrix leverages its superior thermoelectric properties while mitigating issues related to brittleness and mechanical rigidity. Such hybrid materials can be processed into thin films or coatings, which can be applied to various surfaces to create flexible TEGs capable of conforming to complex geometries and dynamic environments. Moreover, the inclusion of nanostructured materials like silicon nanowires into polymer gels has opened new avenues for optimizing thermoelectric performance. Silicon nanowires exhibit unique size-dependent properties that can enhance the thermoelectric efficiency by increasing the density of electronic states and reducing thermal conductivity through increased phonon scattering.[179] When these nanowires are embedded within a conductive polymer gel, the resulting hybrid material can achieve a higher thermoelectric figure of merit (ZT), indicating improved efficiency in converting heat to electricity. Another promising direction in the development of thermoelectric gel materials is the use of organic–inorganic composite gels. These materials combine the advantageous properties of both organic conductive polymers and inorganic thermoelectric particles. For

example, the integration of carbon nanotubes or graphene into PEDOT or polyaniline gels has been shown to further enhance electrical conductivity and thermoelectric performance. Carbon-based nanomaterials provide excellent electrical pathways and can improve the overall charge transport within the gel matrix, leading to higher efficiency in thermoelectric applications.[180] The advancements in thermoelectric gel materials not only improve the efficiency and functionality of TEGs but also pave the way for new applications. Flexible and stretchable TEGs can be integrated into wearable electronics, providing a sustainable power source by harvesting body heat. Additionally, these materials can be applied to industrial machinery and exhaust systems to capture waste heat and convert it into usable electricity, contributing to overall energy efficiency and sustainability.

5.3 Environmental remediation and energy harvesting

5.3.1 Water splitting and purification. Water scarcity and limited access to clean water are critical issues affecting over 1.1 billion people globally, driven by factors such as population growth, poverty, pollution, and climate change.[181–183] To effectively manage waste and control pollution, including microbiological hazards in water, it is essential to consider the cyclical movement of matter and energy in the environment, maintaining balance through the atmosphere and the hydrosphere. In the field of energy conversion and storage, waste electrical and electronic equipment (WEEE) presents a significant challenge due to its hazardous components, especially pollutant metals like palladium, nickel, gold, and lithium. These metals can drastically impact access to freshwater, as global warming increases their solubility and concentration in water, worsening toxic conditions and disrupting biogeochemical cycles.[183] While sustainable and responsible water management and conservation are vital, scientific advancements are also focusing on creating efficient water treatment methods that not only sequester metal ions but also repurpose them as new gelling nanomaterials for energy applications. Hydrogels, with functional groups such as carboxyl and amino groups, have shown great promise in adsorbing heavy metals and organic pollutants from water. This capability contributes to cleaner water supplies essential for industrial processes, including those involved in energy production, such as hydraulic fracturing and thermal power plants where water purity is critical.[184]

Recent advancements in hydrogel-based water purification have seen the development of smart hydrogels that respond to environmental stimuli like pH or temperature changes. These smart hydrogels can selectively adsorb and release pollutants, providing a more efficient and sustainable method for water treatment.[185] For instance, pH-responsive hydrogels can undergo significant changes in their swelling behavior and ion adsorption capacity when exposed to different pH levels, allowing for targeted removal of contaminants. Temperature-responsive hydrogels, on the other hand, can change their physical state or permeability at specific temperatures, enabling controlled release of adsorbed pollutants. The integration of nanomaterials into hydrogel matrices has further enhanced their water purification capabilities. Nanomaterials such as graphene oxide, carbon nanotubes, and metal–organic frameworks (MOFs) can be incorporated into hydrogels to improve their mechanical strength, adsorption capacity, and selectivity for various pollutants. For example, graphene oxide-based hydrogels have demonstrated superior adsorption capabilities for heavy metals and dyes due to their large surface area and functional groups that interact with contaminants.[185] Similarly, MOF-based hydrogels offer tunable porosity and high surface areas, which are advantageous for capturing specific pollutants from water. Another innovative approach in hydrogel technology is the development of multifunctional hydrogels that combine different purification mechanisms. These hydrogels can simultaneously remove a broad spectrum of contaminants, including heavy metals, organic pollutants, and microorganisms. For instance, hydrogels embedded with antimicrobial agents can not only adsorb heavy metals but also inhibit the growth of pathogenic bacteria, providing a comprehensive water purification solution.[186] The application of hydrogels in water purification also extends to environmental monitoring and remediation. Hydrogels can be used as sensors to detect the presence of specific contaminants in water bodies. By incorporating responsive dyes or conductive polymers into the hydrogel matrix, these sensors can provide real-time monitoring of water quality, facilitating timely intervention and remediation efforts.[187]

Metal–organic gels (MOGs) have shown great potential for the oxygen evolution reaction (OER) in hydrogen generation due to their ability to form 3D hierarchical structures under mild synthesis conditions. These coordination-driven supramolecular polymers possess functional metal-centered sites and are created through self-assembly, which results in fibrous, flake-like, or granular structures held together by intermolecular forces. Singh *et al.* developed a $Ni_{0.6}Fe_{0.4}$–MOG that enhanced OER catalytic activity with an overpotential of 289 mV at 10 mA cm^{-2} and a Tafel slope of 33 mV dec^{-1}, outperforming traditional catalysts like RuO_2.[19]

In another study, the Li group introduced a P-doped Ni–Mo bimetal aerogel as a bifunctional electrocatalyst for water splitting. Phosphorus doping enhanced the electronic structure, improving hydrogen and oxygen desorption, leading to excellent performance in both the HER (69 mV at 10 mA cm^{-2}) and OER (235 mV at 10 mA cm^{-2}) with efficient water splitting at a low cell voltage of 1.46 V.[188] Additionally, Saha *et al.* used a supramolecular Ni(II)–triazole gel to synthesize a bifunctional electrocatalyst. This approach formed Ni(0) nanoclusters within heteroatom-doped carbon onions, resulting in efficient catalytic performance with overpotentials of 360 mV for the OER and 250 mV for the HER as shown in Fig. 14. These developments highlight the
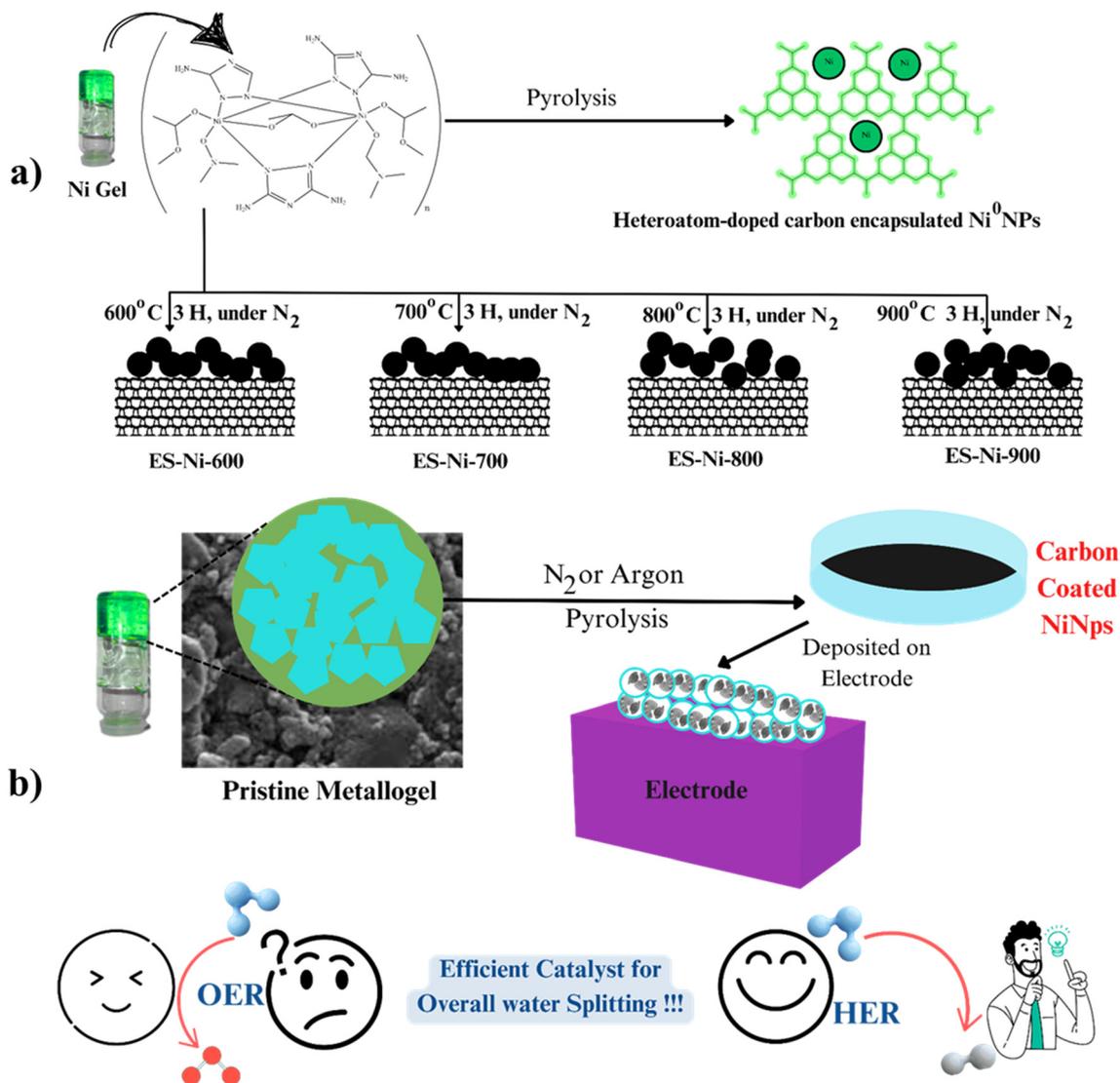
1380 | *Catal. Sci. Technol.*, 2025, **15**, 1357–1389

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Fig. 14 (a) Diagram depicting the synthesis of Ni(0) nanoclusters encapsulated within heteroatom-doped carbon onions originating from Ni-based gels and (b) application of the synthesized material as a bifunctional electrocatalyst for water splitting (reproduced from ref. 189 © Elsevier 2023).

potential of advanced gels and aerogels in catalysis for sustainable energy solutions.[189]

5.3.2 Energy harvesting. Gel materials have garnered significant attention in the development of devices that harvest energy from ambient sources, such as piezoelectric and triboelectric nanogenerators. These devices are designed to convert mechanical energy from vibrations, movements, and other mechanical deformations into electrical energy, providing a promising avenue for sustainable energy generation. Fig. 15 illustrates different energy harvesting effects, including the triboelectric effect, piezoelectric effect, thermoelectric effect, and hybrid effect, and their applications in energy harvesters.[190] Energy harvesters can power devices such as LEDs, capacitors, and electronic devices by converting mechanical or thermal energy. The triboelectric effect generates charges through friction between different materials, while the piezoelectric effect

produces an electric charge when mechanical stress is applied. The thermoelectric effect utilizes temperature differences to generate electricity, and hybrid systems combine multiple energy conversion methods to enhance efficiency. Poly(vinylidene fluoride) (PVDF)-based gels are particularly notable for their piezoelectric properties, which can be exploited to generate electricity when subjected to mechanical stress.[191] This makes them ideal for applications where energy can be harvested from ambient mechanical movements, such as in industrial machinery, vehicle operations, or even daily human activities. Moreover, the integration of conductive nanoparticles into hydrogels has been explored for enhancing the performance of triboelectric nanogenerators. These hydrogels can capture energy from human motion or environmental vibrations, leveraging the triboelectric effect, where contact and separation between different materials generate electrical charges. For instance,
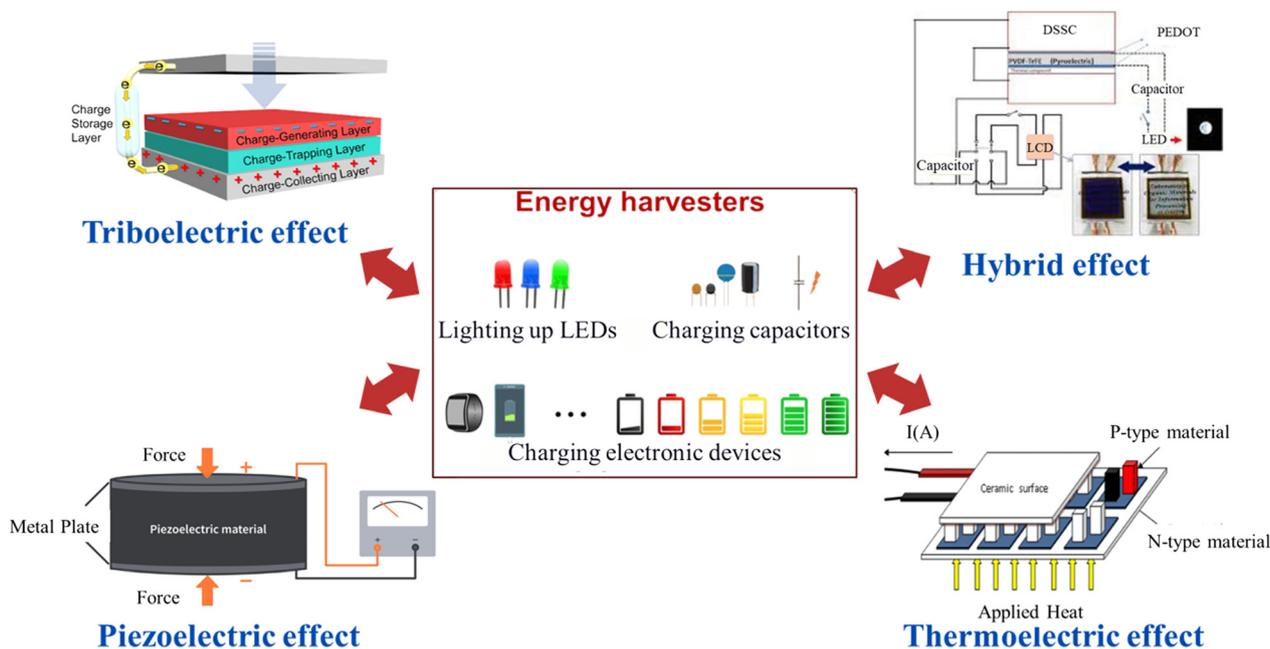
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Fig. 15 Schematic representation of various energy harvesting mechanisms (reproduced from ref. 190 © MDPI 2023).

Mang Gao *et al.* demonstrated the use of hydrogels embedded with conductive nanoparticles in triboelectric nanogenerators, achieving efficient energy conversion from various mechanical inputs.[192] Recent innovations in this field
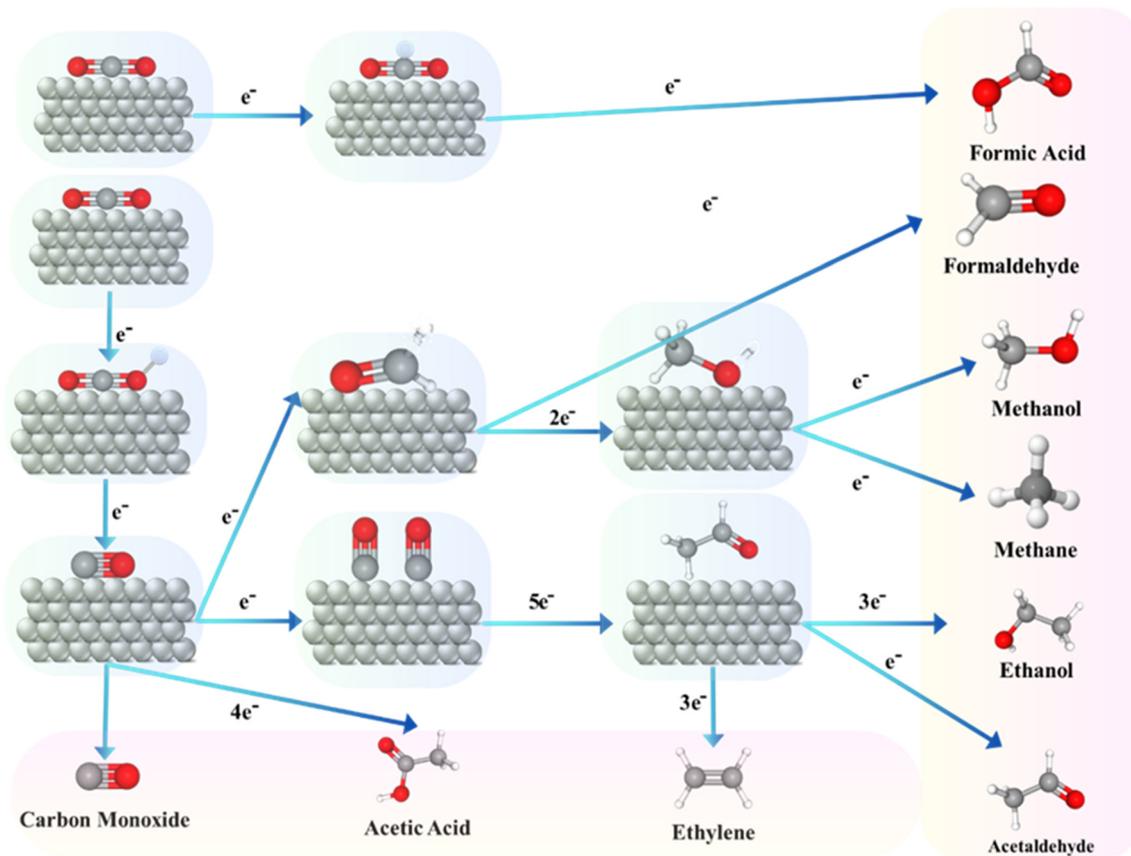

Fig. 16 The pathway for CO_2 reduction reactions leading to various value-added products is illustrated, with blue spheres denoting catalysts, gray spheres indicating carbon, red spheres representing oxygen, and white spheres symbolizing hydrogen (reproduced from ref. 197 © Wiley 2021).

include the development of hybrid gels that combine the properties of both piezoelectric and triboelectric materials, leading to significantly higher energy conversion efficiencies. These hybrid gels synergize the advantages of both mechanisms, capturing a broader range of mechanical energies and converting them into electrical power more effectively. Another group highlighted the potential of these hybrid gels in wearable electronics, where they can provide a sustainable and reliable power source for devices such as sensors, health monitors, and smart textiles.[193]

The application of gel materials in energy harvesting devices extends beyond simple energy capture. These materials are also engineered for flexibility, durability, and biocompatibility, making them suitable for integration into a variety of environments and applications. For instance, flexible piezoelectric gels can be incorporated into clothing or embedded in footwear to harvest energy from walking or other movements, continuously powering wearable devices without the need for external batteries. Similarly, triboelectric nanogenerators utilizing hydrogels can be integrated into infrastructure to capture energy from environmental vibrations, such as those caused by wind or traffic, contributing to the energy supply of smart cities. Further advancements in materials science have led to the exploration of novel composite materials that enhance the mechanical and electrical properties of these gels. By incorporating materials like graphene, carbon nanotubes, or metallic nanowires, researchers have developed gels with improved conductivity, mechanical strength, and sensitivity to mechanical stimuli. These enhancements enable more efficient energy conversion and broader application possibilities. For example, the inclusion of graphene in PVDF gels has been shown to significantly increase the piezoelectric output, making these materials more viable for high-performance energy harvesting.[194] Additionally, the environmental and economic benefits of using gel materials for energy harvesting cannot be overstated. These materials often require less energy to produce and can be derived from renewable resources, aligning with the principles of green chemistry and sustainable development. The ability to generate electricity from ambient mechanical energy also reduces reliance on traditional energy sources, decreasing greenhouse gas emissions and promoting a more sustainable energy ecosystem.

5.3.3 For CO_2 reduction mechanisms. Gel structures contribute to CO_2 reduction mechanisms through unique molecular and electronic properties that enhance reaction efficiency and selectivity. The electrochemical reduction of CO_2 into valuable chemicals like methane, ethanol, and formate is a promising approach to address rising CO_2 emissions.[195] The electrochemical conversion of CO_2 into valuable products, including chemical feedstocks (such as carbonate, polycarbonate, and polymer precursors) and liquid fuels (like CH_4, C_2H_4, C_2H_6, and CH_3OH), is considered a promising approach for enhancing environmental sustainability on our planet as shown in Fig. 16. This process, however, faces challenges due to the stability of CO_2, which demands high overpotentials and competes with the hydrogen evolution reaction (HER). Recent research has focused on developing advanced catalysts to improve CO_2 reduction selectivity and efficiency.[196]

For instance, the Alexander group created Au–Pd core–shell aerogels, achieving nearly perfect faradaic efficiency (99.96%) for CO at −0.5 V.[198] Other 3D gel catalysts, such as Choi's graphene-based hydrogels combined with iron porphyrin, have shown 96.2% efficiency for converting CO_2 to CO, thanks to enhanced surface area and improved wettability.[199] Similarly, Pd–Cu bimetallic aerogels by the Han group achieved 80% efficiency in reducing CO_2 to methanol, leveraging structural porosity and metal synergy.[200]

Moreover, nitrogen-doped graphene aerogels developed by the Shi group enhanced stability and selectivity for CO_2 reduction to CO, demonstrating 86% efficiency.[201] Ni-doped carbon aerogels, developed by Hou's group, reached industrial-level current densities of 300 mA cm^2 and 98% faradaic efficiency for CO_2 to CO conversion.[202] Meanwhile, Bi–Sn and Cu–Bi bimetallic aerogels have shown promising efficiencies (90.9% and 96.5%, respectively) for formate production. To align with sustainability, Kraatz's group developed biodegradable peptide-based aerogels with Ag nanoparticles, achieving up to 88% efficiency.[203] For scalable CO_2 reduction, Abdinejad's team designed 3D Cu–Pd/MXene aerogels with high current densities (150 mA cm^2) and 93% efficiency, indicating potential for industrial CO_2 electroreduction applications.[204]

In energy storage systems, redox reactions are fundamental because they facilitate the transfer of electrons, which enables energy to be stored and released. Gel catalysts, with their distinctive structural properties, have the potential to enhance these reactions by stabilizing them, improving efficiency, and increasing reversibility. By examining the redox mechanisms in gels, researchers can determine whether these catalysts effectively support electron transfer, which is critical for devices like batteries that undergo repeated charge and discharge cycles.[197] Gel catalysts can help stabilize reactions by reducing side reactions that degrade material quality over time. Catalytic efficiency, which indicates how well a catalyst drives a reaction with minimal energy, can be improved with gels, leading to faster reactions and enhanced battery performance. Furthermore, gels may enhance the reversibility of redox reactions by providing a stable, porous structure that allows ions to move smoothly, thus reducing material wear during cycles. Some gels also incorporate active sites that support electron transfer, making each cycle more reliable. Understanding these mechanisms is essential for developing more durable, efficient energy storage technologies.

6. Challenges and future directions

While the use of gels in energy applications is promising, several challenges need to be addressed to fully realize their potential.

6.1 Stability and durability

Ensuring the long-term stability and durability of gel materials under operational conditions is a critical challenge, particularly for hydrogels used in supercapacitors and fuel cells. These materials need to maintain their mechanical integrity and conductive properties over numerous cycles of use. To address these issues, researchers are exploring the incorporation of cross-linkers and reinforcement materials to enhance the mechanical properties of gels without compromising their functional characteristics.[205] One effective strategy involves incorporating nanofillers such as carbon nanotubes, graphene, and silica nanoparticles into gel matrices, which has been shown to significantly improve mechanical strength and stability. For instance, nanocomposite gels that integrate carbon nanotubes provide exceptional tensile strength and electrical conductivity, which enhance the overall durability and efficiency of the gels.[206] Carbon nanotubes are particularly effective due to their high aspect ratio and excellent mechanical properties, which contribute to the formation of a robust network within the gel matrix. Similarly, graphene, known for its superior thermal and mechanical properties, can be incorporated into hydrogels to improve their rigidity and thermal stability. This is crucial for maintaining performance during prolonged operational cycles.[207]

Moreover, silica nanoparticles are also used to enhance the mechanical properties of hydrogels. The addition of silica helps create a more rigid network within the gel matrix, which not only enhances mechanical strength but also improves resistance to environmental degradation.[208] These nanocomposite gels exhibit enhanced mechanical properties and resistance to degradation, making them suitable for long-term energy applications. Recent advancements have also focused on developing hybrid gels that combine organic and inorganic components to further bolster their mechanical and conductive properties. For example, combining organic polymers with inorganic nanofillers can lead to gels with superior mechanical flexibility and enhanced electrochemical performance.[209] These innovations are crucial for creating more resilient and efficient gel materials for use in energy storage and conversion devices, ultimately contributing to the advancement of sustainable energy technologies

6.2 Ionic conductivity

Improving the ionic conductivity of gel electrolytes, particularly at low temperatures, is a significant challenge in the field of energy storage. Recent efforts have focused on developing novel polymer composites and nanocomposite gels capable of maintaining high ionic conductivity across a broad temperature range. One promising approach involves incorporating ionic liquids as additives, which can enhance ionic conductivity and provide better thermal stability due to their unique properties, such as low volatility and high ionic mobility. Another effective strategy is the development of

single-ion conducting gels. These gels are designed so that only the desired ions, such as lithium ions, are mobile within the electrolyte matrix. This selective ion mobility reduces the occurrence of side reactions and improves the overall efficiency and safety of the energy storage device. Additionally, research into the incorporation of nanoscale fillers, such as ceramic nanoparticles, into polymer gels has shown promise in further enhancing ionic conductivity and mechanical stability, making these materials highly suitable for advanced battery applications. These advancements underscore the ongoing innovation aimed at optimizing gel electrolytes for better performance in diverse operational environments.[119,210]

6.3 Scalability

The scalability of manufacturing gel materials for industrial applications presents significant challenges. While many gel materials exhibit excellent performance at the laboratory scale, translating these results to large-scale production requires overcoming obstacles related to cost, reproducibility, and integration with existing technologies.[211] Innovations in 3D printing and additive manufacturing provide promising solutions for the scalable production of gel-based energy devices. These advanced manufacturing techniques enable precise control over the composition and structure of gels, facilitating the fabrication of complex and high-performance energy devices. For instance, 3D printing allows for the layer-by-layer construction of gel materials, ensuring uniformity and consistency in large-scale production. This method also supports the customization of gel properties to meet specific application needs, enhancing the adaptability of gel-based technologies in various industrial contexts.[212] Moreover, the integration of additive manufacturing with automated production lines can significantly reduce production costs and improve the reproducibility of gel materials. As these technologies continue to evolve, they hold the potential to bridge the gap between laboratory-scale innovation and industrial-scale application, driving the widespread adoption of gel-based energy solutions.

6.4 Environmental impact

The environmental impact of gel materials, especially those derived from synthetic polymers, remains a significant concern in the field of energy storage and conversion. Consequently, research is increasingly directed toward developing biodegradable and bio-based gels that can minimize the environmental footprint of these devices.[213] For example, natural polymers such as alginate, chitosan, and gelatin are being investigated as sustainable alternatives to traditional synthetic polymers in various energy applications. These biopolymers offer the advantage of biodegradability and are derived from renewable sources, making them more environmentally friendly. Additionally, the focus on developing recyclable gels that can be easily processed and reused is gaining traction. Recyclable gels can significantly

1384 | Catal. Sci. Technol., 2025, 15, 1357–1389

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reduce waste and diminish the environmental impact of energy devices by facilitating a more sustainable lifecycle.[214] For instance, gels based on thermally reversible networks can be depolymerized and reprocessed, thus offering a practical solution to the disposal issues associated with conventional gels. The integration of such sustainable practices in the design and development of gel materials is crucial for advancing green energy technologies and reducing the overall environmental impact.

7. Conclusions

The development of green energy conversion schemes is vital for ensuring the sustainability of our planet and maintaining acceptable living standards for future generations. Despite progress in energy storage technologies, the reliance on fossil fuels continues to hinder efforts to combat global warming. Emphasizing renewable energy sources like photovoltaics and fuel cells, along with wind, tidal, and hydro energy, is crucial for establishing a diversified and resilient energy mix. The versatility of gel materials presents significant opportunities in energy applications, enhancing the performance and safety of batteries, supercapacitors, and fuel cells. Hydrogels, in particular, offer innovative solutions due to their unique properties, including high water content, tunable mechanical strength, and responsiveness to environmental stimuli. While challenges such as stability, scalability, and material selection remain, ongoing research and technological advancements are likely to overcome these obstacles. The exploration of hybrid, smart, and biomimetic gels is set to propel the next generation of energy storage technologies, aligning with global efforts to achieve carbon neutrality and net-zero emissions. By integrating these advanced gel materials into energy systems, we can advance toward a sustainable future, mitigating environmental impacts and ensuring the livability of our planet for generations to come.

Ethical approval

This article does not contain any studies with human participants or animals performed by any of the authors. As a review manuscript, it synthesizes existing research and does not involve new data collection requiring ethical approval.

Data availability

The data supporting the findings of this study can be obtained from the corresponding author upon request. Due to privacy concerns and other restrictions, the data are not publicly accessible.

Author contributions

Tasmina Khandaker: writing – original draft, review and editing; Md Al-Amin Mia Anik, Ananya Nandi, Tasniqul Islam, Md. Mohibul Islam: editing and drawing of figures for the original draft; Md. Kamrul Hasan, Palash Kumar Dhar, M. Abdul Latif: proof reading and editing; Muhammad Sarwar Hossain: formal analysis, methodology, resources, validation, writing and editing, supervision and project administration.

Conflicts of interest

The authors declare that they have no known financial or personal conflicts of interest that could have influenced the work reported in this paper.

Acknowledgements

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