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The marriage of Xenes and hydrogels: Fundamentals, applications, and outlook
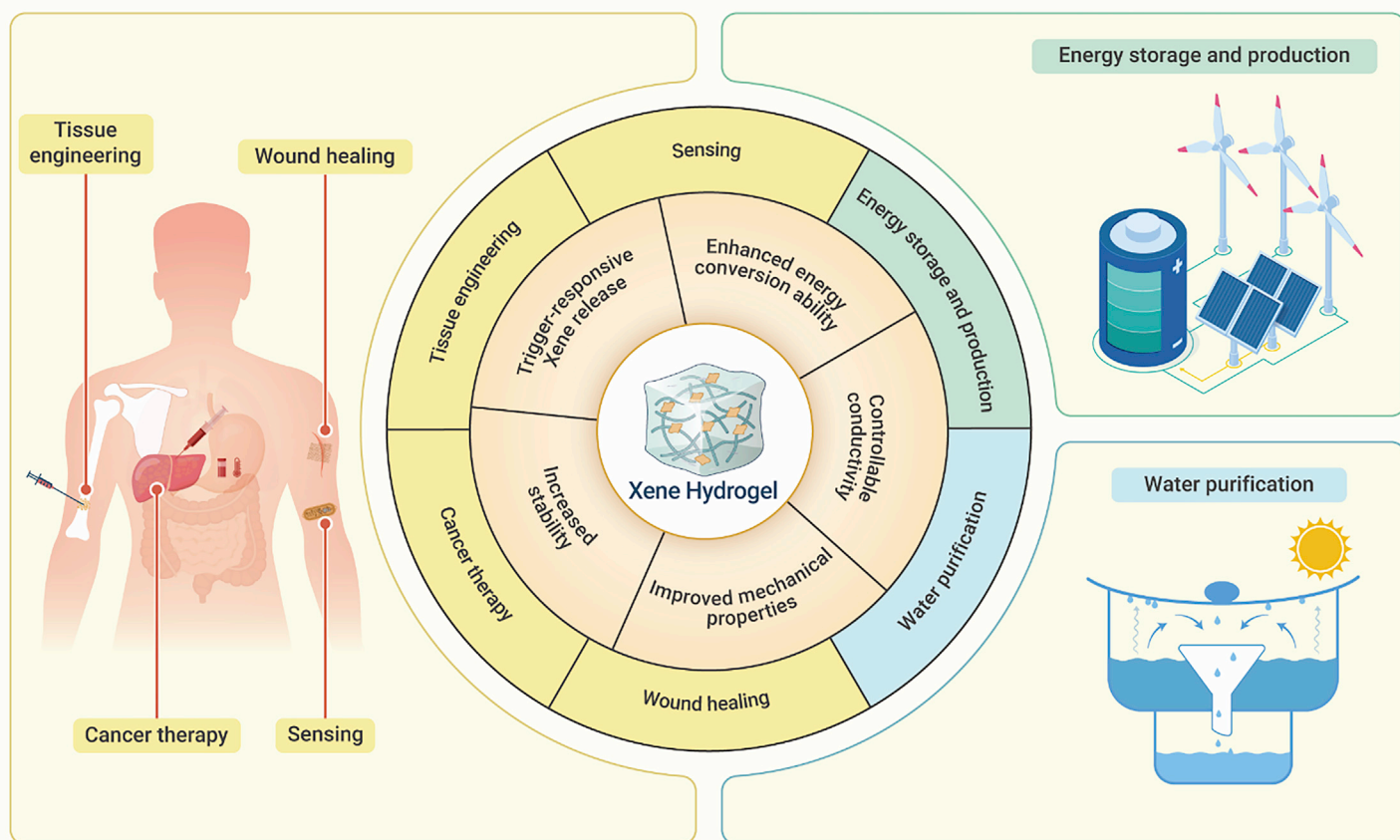
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GRAPHICAL ABSTRACT

PUBLIC SUMMARY

- Two-dimensional mono-elemental nanosheets (Xenes) endow tunable application

- Synthesis mechanism, properties, and applications of Xene hydrogels are summarized

- Expanding performance and applications of both hydrogels and Xenes are presented

The marriage of Xenes and hydrogels: Fundamentals, applications, and outlook

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Hydrogels have blossomed as superstars in various fields, owing to their prospective applications in tissue engineering, soft electronics and sensors, flexible energy storage, and biomedicines. Two-dimensional (2D) nanomaterials, especially 2D mono-elemental nanosheets (Xenes) exhibit high aspect ratio morphology, good biocompatibility, metallic conductivity, and tunable electrochemical properties. These fascinating characteristics endow numerous tunable application-specific properties for the construction of Xene-based hydrogels. Hierarchical multifunctional hydrogels can be prepared according to the application requirements and can be effectively tuned by different stimulation to complete specific tasks in a spatiotemporal sequence. In this review, the synthesis mechanism, properties, and emerging applications of Xene hydrogels are summarized, followed by a discussion on expanding the performance and application range of both hydrogels and Xenes.

INTRODUCTION

Hydrogels are three-dimensional (3D) networks composed of gelators and water and are suitable for applications in the water phase.[1,2,3] The gelators of typical hydrogels are hydrophilic polymers exhibiting distinguishing properties of swelling, transmittance, softness, biocompatibility, and ease of modification.[4] Both market and scientific research data confirmed the practical value of hydrogels in daily life and the biomedical fields, i.e., diapers and wound dressing.[5,6] However, the interactions between hydrogel polymers are weaker to maintain the ductility and stability. Interacting with other materials, hydrogels mainly offer attachment points and spaces rather than disposing of adsorbed cargos due to inertness, limiting hydrogels' applications in established fields.[7] To overcome these limitations, different materials are incorporated into the hydrogel matrix.[8–13] The increased crosslinking points can improve the mechanical properties of the matrix. In addition, functional materials such as molecules, polymers, and inorganic particles offer other exceptional features, such as nourishment, disease treatment, dye degradation, sterilization, and electrical conductivity.[14–17]

The involvement of two-dimensional (2D) materials is an effective method to improve the properties and expand the hydrogel applications, owing to the confined carrier migration and heat diffusion in a 2D plane.[18,19] A class of 2D mono-elemental materials with a similar structure to graphene is defined as Xenes. Xs are elements of the IIIA−VIA groups in the periodic table (Figure 1) and the corresponding Xenes are graphene, arsenene, germanene, boronene, antimonene, silicene, and stanene. Although X can be nonmetals (carbon, phosphorus, and selenium), metalloids (boron, silicon, germanium, arsenic, antimony, and tellurium), and metals (gallium, tin, and bismuth), most Xenes exhibit a metallic property, not exactly corresponding to their bulk materials. As 2D materials, Xenes show outstanding physical and chemical reactivity due to their atomic out-of-plane thickness and attain special electrical, photonic, magnetic, and catalytic properties. Due to their mono-elemental composition, Xenes show superior performance and a wide range of applications, compared with other 2D materials. First, the hybridized forms of X atoms in Xenes are sp^2 or sp^3, i.e., indicating limited covalent bonding modes between identical atoms, making Xenes structures simple, predictable, controllable, and appreciable. Second, Xenes properties can be tuned by changing the structure; i.e., layer number[20] or transformation to another allotrope.[21] Also, the simple composition ensures limited and innocuous degradation products, contributing to high environmental and biological compat-

ibility. Further investigations on these simple Xenes can lead to some fascinating research on other 2D materials.

The restrained reactions in two dimensions and lack of stability of Xenes in an embedded 3D matrix are some limitations; therefore, several strategies have been designed for improved dispersion and extended applications.[22] Recently, emerging 2D Xenes are introduced in hydrogel networks. Three-dimensional hydrogels can serve as a scaffold for 2D Xenes and enhance the properties and applications of hydrogels. In this review, the gelation mechanism, properties, and emerging applications of Xene-based hydrogels are summarized, focusing on the expanding performance and application range of hydrogels and Xenes in different fields.

XENE HYDROGELS: DESIGN AND ASSEMBLY

The synthesis of Xenes and hydrogels has been well developed and discussed as the basic steps for Xene hydrogel preparation.[23,24] Figure 2 shows the simplified process for the synthesis of hydrogel and Xene. Sonication-assisted liquid-phase exfoliation is used to obtain phosphorene,[25] arsenene,[26] and germanene,[27] from the layered bulk materials. Other top-down methods include chemical exfoliation[28] and mechanical exfoliation.[29] In addition, liquid exfoliating and thermal oxidation etching are coupled to prepare ultrathin borophene.[30] Conversely, the construction of Xenes via bottom-up strategies refers to the preparation from atoms/ions, and the methods include physical vapor deposition and chemical vapor deposition. These methods increase Xene categories without referring to the lattice of bulk materials.[31–34] Similarly, methods for hydrogel preparation are based on covalent linkages or noncovalent interactions (hydrogen bonding, electrostatic, and hydrophobic forces) among polymers, including freeze-drying, freeze/thaw cycling, self-assembly, small molecule/ionizing radiation/free radical-induced covalent crosslinking, etc.[35,36] In the last few years, hydrogels are also fabricated by modern technologies such as electrostatic spinning and 3D bioprinting.[37–39]

During Xene hydrogel fabrication, precise control of the interactions between Xenes and hydrogels is required to bring out the desired advantages of both components. For instance, in photocatalytic degradation, the adsorption ability of graphene hydrogel for target pollutants increases with the addition of graphene oxide (GO). However, a high concentration of GO severely restricts light penetration and limits photocatalytic efficiency.[40] Also, the stability of Xenes during the preparation process is also maintained by coating other materials on the surface.[41] Xene hydrogels are also fabricated by incorporating two components in different orders and methods, displaying fundamental differences in the interaction between polymers and Xenes, the architectural organization of the final Xene hydrogel network, and their practical applications. Accordingly, there are two main procedures: (1) Xene incorporation in crosslinked hydrogels (Figure 3A), and (2) hydrogel crosslinking in the presence of Xenes (Figure 3B). Xenes can also gelatinize without polymers, i.e., self-gel of Xenes (Figure 3C). The basic steps of the three manufacturing processes with their principles, as well as the advantages and challenges, are also discussed in the following sections.

Xenes incorporation in crosslinked hydrogels

Benefiting from the hydrogel's porous structure and its ability to incorporate smaller-sized particles, the pre-prepared Xenes can be loaded to crosslinked hydrogel networks (Figure 3A). The most common procedure relies on hydrogel
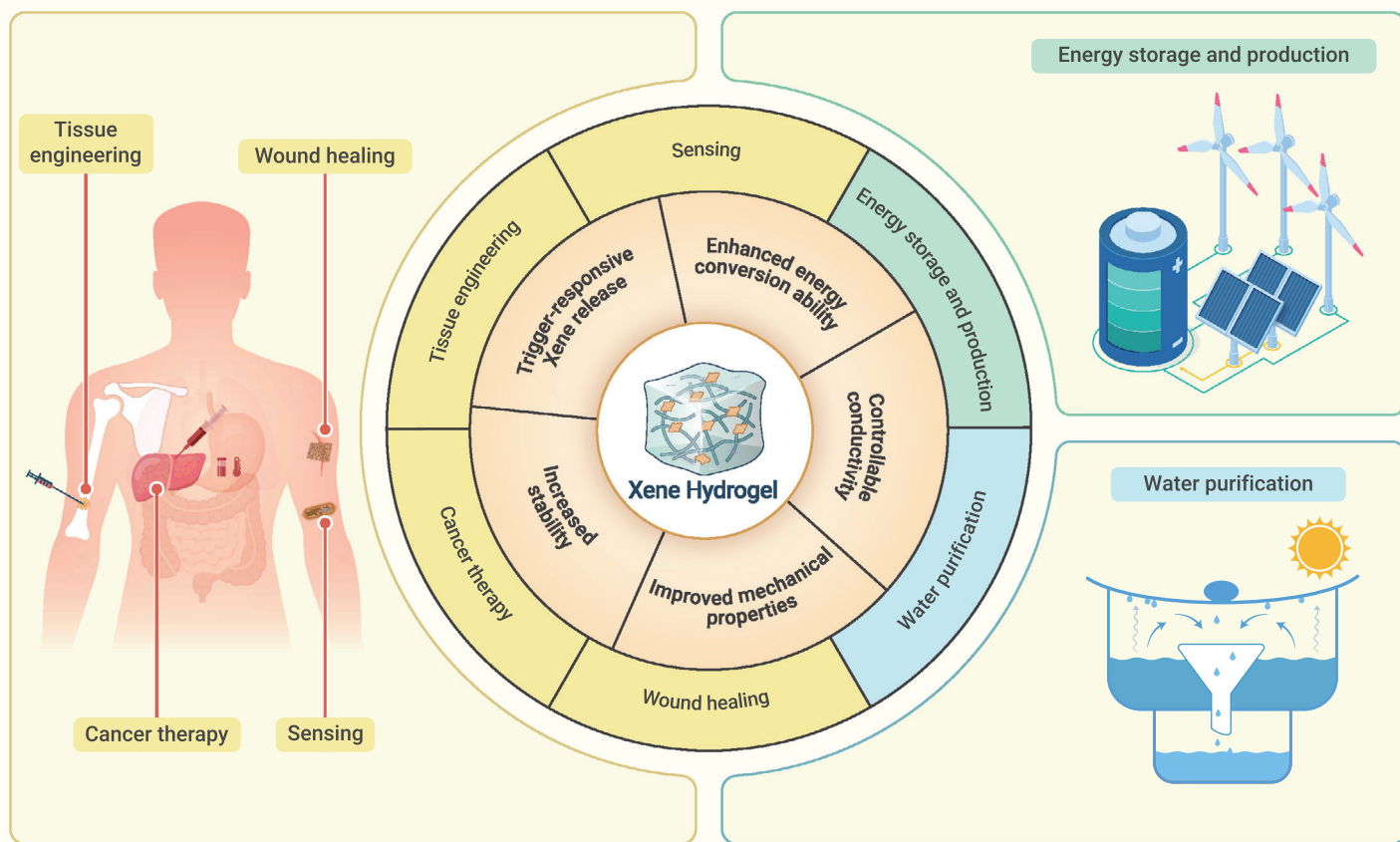
Figure 1. Reported elements (with color background) that can form Xenes Blue background, metals; green background, metalloids; yellow background, nonmetals. White background, unreported yet.

swelling in concentrated Xene solutions where the gelatinous composite is nonresponsive to the Xene embedding conditions. Xenes can diffuse into the injectable hydrogel pores in the solution. For example, hydrogels containing black phosphorus (BP) via hypodermic injection, constructed with hyaluronic acid (2 wt%) and pluronic F-127 (25 wt%), are beneficial to inhibit tumor recurrence after surgery.[42] Germanene-based hydrogels are developed by mixing germanene with hydrogel composed of chitosan and agarose.[27] Hydrogels can also have a fixed shape. Li et al. reported a sandwiched BP hydrogel by synthesizing a layer of chitosan-based hydrogel and then loading BP nanosheets onto the chitosan layer with a syringe filter. Subsequently, another hydrogel layer is equipped with the BP-coated chitosan hydrogel.[43]

These methods are not suitable for Xenes with strong interactions, since strategies for increasing dispersibility may impair some properties of Xenes. For example, graphene is highly hydrophobic that is not evenly dispersed in an aqueous solution and hydrogels. In practical applications, a uniform distribution of graphene hydrogels is achieved by anchoring the surface with polar functional groups, including hydroxyl and carboxyl, through chemical modification of oxidation or reduction. Unfortunately, the covalent functionalization can destroy the original lattice of graphene, resulting in altered properties, especially electric conductivity, and both reduced graphene and GO exhibit lower conductivity than original graphene. Although GO is reduced again, only one-third of the conductivity can be restored.[44] *In situ* Xenes formation after hydrogel crosslinking can significantly avoid undesired self-aggregation of Xenes.

Hydrogel crosslinking in the presence of Xenes

One of the most preferred methods to manufacture hybrid Xene hydrogels is to combine Xene suspensions with hydrogel precursors before hydrogel formation (Figure 3B). Typically, this process can realize relatively homogeneous incorporation of Xenes in hydrogel compared with other techniques, allowing the incorporation of various Xenes since Xenes are pre-synthesized before hydrogel crosslinking. The challenges in this approach involve the requirement of a good uniform dispersion ability and the requirement of an appropriate concentration

of Xenes affecting the distribution of Xenes and hydrogel network formation. The simplest gelation method is dissolving Xenes into polymer solutions and removing the solutions by heating[45,46] or freezing-thawing cycles.[47−49] Also, thermal,[50−52] radiation,[53] pH variation,[54,55] or chemical cross-linkers[56−60] are commonly used to fabricate Xene hydrogels. For example, BP@hydrogel was prepared by mixing the agarose solution with BP sheets under mechanical stirring for 30 min at 90°C and stored at 4°C for 30 min.[61] Liang et al. prepared homogeneous graphene/PVA solution by gradually adding GO/H_2O solution to the PVA solution at 40°C and then sonicating for 30 min. Then, the mixture was poured into a Teflon petri dish and dried at 60°C for GO/PVA hydrogel film formation.[45]

Xenes are also suitable crosslinking nodes in 3D platforms. Benefiting from the advances in polymer chemistry, the selective functional groups with target complementary moieties are engineered onto the Xenes' surface, thus achieving augmented interactions in hydrogel precursors, then preparing nanocomposite hydrogels via interdependent assembly. Meanwhile, such Xene hydrogel interactions possess high flexibility and adaptability to encode nanocomposites in the final hybrid structure. For example, polydopamine-coated BP was attached to polymer hydrogel via noncovalent interactions with GelMA, including hydrogen bonds and $\pi-\pi$ stacking.[41] Liu et al. synthesized graphene peroxide (GPO), which decomposed into free radicals (GO•, OH•) on heating. They dissolved acrylamide in GPO aqueous dispersion, sealed the sample into glass molds, and heated it in a water bath at 45°C to form hydrogels.[62]

Self-gel of Xenes

In 2008, Li et al. reported a chemically converted graphene (CCG) that could stably disperse in water without any surfactants (Figure 3C).[63] Exfoliated graphite oxide dispersions were first synthesized and then directly converted to stable graphene colloids through hydrazine reduction under specific conditions. Later in 2011, the same group found that CCG sheets can self-gel at the solid-liquid interface via the face-to-face method during filtration when the vacuum was applied; therefore, a layer of water molecules was tightly captured by the oxygen-containing groups on the CCG surface. This kind of directional
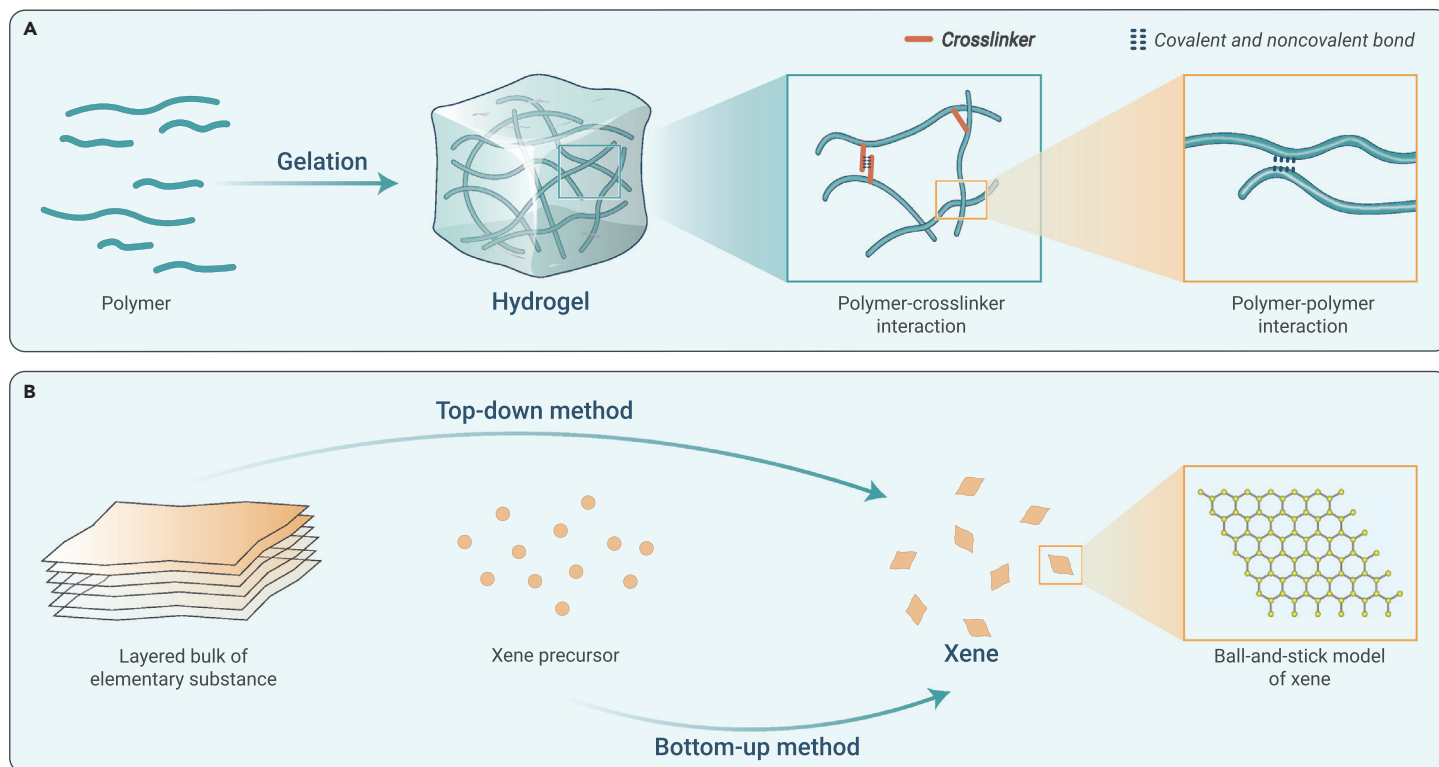
Figure 2. Brief synthesis process and structure of hydrogel and Xene, respectively (A) Traditional hydrogels are fabricated by polymers and water via a process called gelation. Crosslinkers can be used to connect polymer chains. And covalent and noncovalent bonds also contribute to the 3D mesh development. (B) Xenes can be synthesized via two methods. The top-down method always requires a layered structure of bulk materials and uses techniques to break the interlayer forces. The bottom-up method takes no account of the bulk structure, synthesizing Xenes from their precursors. Most Xenes have a two-dimensional honeycomb lattice.

conductive hydrogel film demonstrated unprecedented electrical, mechanical, and anisotropic stimuli-responsive properties.[64] Moreover, during self-gelation, a second Xene can *in situ* grow on the original Xene to form the composite hydrogel.[65]

PROPERTIES AND INTERACTIONS OF XENES AND HYDROGELS

The integrated Xene hydrogels display the properties of both components and additional features associated with the interactions between Xenes and hydrogels (Figure 4). In this section, the most fundamental properties of polymer hydrogels and Xenes are discussed, based on their interactions. Furthermore, the influence of hydrogels on Xenes and the increased/new features of Xenes-based hydrogels for further applications are described.

Fundamental properties of hydrogels and Xenes

For hydrogels. *Hygroscopicity and swelling property.* Hydrogels contain several hydrophilic groups, such as -OH, -CONH-, -CONH$_2$, and -SO$_3$H; therefore, obtained hygroscopicity and swelling properties promise their utilization in diapers, wound dressing, and swelling-related sensors.

Mechanical properties. The water content and crosslinking concentration can influence the mechanical behaviors of hydrogels, especially the plasticity and toughness, endowing them with various shapes to adapt to different applications. Pure hydrogels always show brittleness, which limits their practical applications.

Biocompatibility. Good biocompatibility mainly depends on two aspects. First, many hydrogels rapidly degrade in the body, eliminating the risk of any serious hepatotoxicity and nephrotoxicity. For example, Li et al. developed a biodegradable photothermal hydrogel for photothermal therapy (PTT). The hydrogel was composed of aldehyde-modified dextran through imine bond formation. After injecting it into the tumor tissue, it degraded over time, demonstrating good biocompatibility.[66] Second, the unique characteristics of hydrogel also determine its good biocompatibility. The low interfacial tension between hydrogels and aqueous fluids, due to high-water content reaching 99% in some cases, leads to weak adhesion of hydrogels to proteins and cells. It also imparts softness properties to living tissue; hence, showing good biocompatibility with blood, body fluid, and human tissue. Many studies have confirmed that most hydrogels are biocompatible.[67]

Porosity. The 3D mesh structure of hydrogels offers anchors and spaces for other materials, to deliver or adsorb cargos, such as loading substances with specific functions, like therapeutics,[68,69] enzymes,[70,71] nanoparticles,[72] and nanosheets.[73,74]

Transparency. Most hydrogels are so transparent that they hardly affect the optical properties of their contents. Therefore, several concentration-related hydrogel sensors were developed due to photorefraction variation.[75]

Adjustability. The hydrogel structure and properties can be altered by the water content, crosslinking concentration, additives, and environment. Additives can impart more features, further extending the applications of hydrogels.

Stability. When the crosslinking of hydrogel networks is through noncovalent physical interactions, the change in nanoscale structure and water content occurs with environmental change.[76,77]

For Xenes. Seven properties of Xenes influencing their application are listed below. Since Xenes contain a wide range of 2D mono-elemental materials, therefore, several unique properties of these materials are observed, such as high stability, tunable band gap, photothermal conversion efficiency, catalytic ability, and electrical properties (Table 1).

Layered structure. Controlling the synthesis of Xenes can effectively change the layer number of obtained Xenes, which alters the band gap size and conductivity of Xenes.[20] Due to layered structure, Xenes have large surface areas with exposed atoms, physically or chemically interacting with other materials. These interactions can be used for further modification, cargo delivery, and several other applications.

Stability. Xenes exhibit variable stability. Borophene,[30] gallenene,[83] and antimonene[114,115] are relatively stable under atmospheric conditions. However, the active surface can damage the stability of Xenes by aggregation or degradation in the environment. With a layered structure, the bare 2D nanosheets are likely to stack together through strong *van der Waals* interactions and π-π stacking, thereby, covering the surfaces, decreasing active sites, and even impairing electrical, optical, and other properties of graphene and stanene.[86,100] Germanene can degrade rapidly.[95,96] Similarly, silicene,[90] phosphorene,[25,127] arsenene,[110,128] bismuthene,[119] selenene,[123] and tellurene[125] are biodegradable, further affecting the properties and even functions.
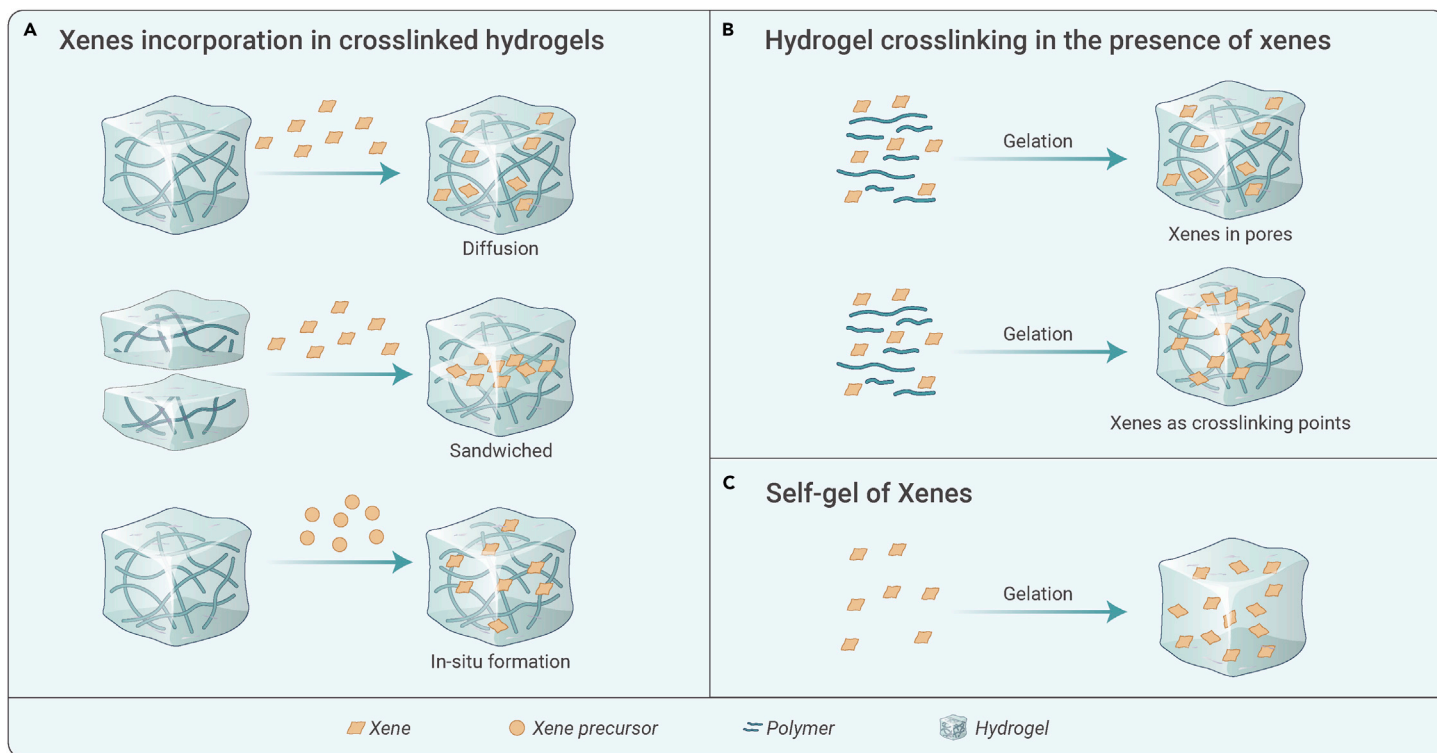
Figure 3. Synthesis of Xene hydrogels via three different methods (A) Xenes incorporation in crosslinked hydrogels, (B) hydrogel crosslinking in the presence of Xenes, and (C) self-gel of Xenes.

Environmental and biological compatibility. The degradation of the single element Xenes is more obvious in the environment or *in vivo* than in other 2D materials, and some Xenes are biodegradable with innocuous metabolites, enhancing their potential in environmental and biomedical applications.[129]

Tunable band gap. The band gaps of Xenes can be adjusted by the number of layers, surface modification, and external physical triggers, influencing the carrier mobility, thermal conductivity, and other electrical properties.[120] Among these materials, phosphorene,[78] arsenene, antimonene,[111] and telllurene[126] have layer-dependent band gaps. For example, BP nanosheets have thickness-dependent band gaps, in the range of 0.3 eV and 2 eV, obtained by adjusting the thickness of nanosheets.[130] Some Xenes have zero or near-zero band gaps,[78] such as borophene, gallenene, graphene, and silicene. The band gap of silicene can be tuned under a vertical electric field[91] and that of borophene can be increased by hydrogenation/fluorination or a shear strain.[79] Similarly, the band gap of germanene and stanene can be tuned under a perpendicular electric field.[91,101]

Photothermal conversion ability. Some 2D Xenes have a direct and adjustable bandgap with light conversion efficiency.[91,104,131,132] Due to the supereminent light absorption and conversion efficiency, Xenes are applied in photoacoustic imaging, PTT, and photodynamic therapy (PDT). For example, borene with distinguishing photothermal conversion efficiency has been reported to attain 42.5% much higher than that of Au nanoparticles, and 13.1% for PDA.[30,133] In addition, graphene,[87] stanene,[100] antimonene,[116] and tellurene[125] have 40% higher photothermal conversion efficiency, which makes these materials excellent photothermal agents for PTT.

Catalytic ability. Owing to a suitable band gap, some Xenes respond to external triggers (light or mechanical stress) to catalyze adsorptive substances, such as consuming H_2O to produce O_2 and CO_2 to formate. Except that the catalytic ability of gallenene, selenene, and tellurene has not been reported yet, other Xenes can catalyze energy generation reactions and other applications.[80,88,89,92,93,97,102,105–108,112,117] For example, Cao et al. prepared an atomic thin layer of bismuthene with an average thickness of 1.28–1.45 nm. The obtained materials had a large number of exposed active centers, which significantly improved the CO_2 reduction performance and showed long-term stability in the electrochemical conversion of CO_2 to formate.[121]

Electrical property. As a honeycomb lattice structure, most Xenes are topological insulators to harness a quantum-mechanical property of spins to conduct electricity.[134] Borophene,[81,82] gallenene,[84,85] graphenes,[78] and silicene[94] exhibit high electrical conductivity, and other Xenes are semiconducting as well.[98,103,109,113,118,122,124] Some Xenes possess a metallic-to-semiconductor transforming character with the change in thickness,[111] applied stress,[135] or annealing temperature.[99] For example, Chen et al. fabricated germanium sheets, revealing the low electrical conductivity of germanium, consistent with the larger band gap. After heat treatment, the electrical conductivity increased sharply and the material was converted from insulator to metal.[99] As a result, the conductivity and semi-conductivity of different Xenes can be used in hydrogel structures.

Influence of gelation on Xene properties

Increased stability. The well-sized holes and attachable groups in hydrogels can keep additives at a specific distance to maintain the structural and functional stability of hybrid hydrogels, thus reducing Xene aggregation and displaying steady and continuous function.[67]

Hydrogel-controlled Xene release. The structure of some hydrogels can change with the external environment, resulting in cargo release. This aspect is particularly suitable for drug delivery applications.[136]

Increased conductivity. Self-gel can integrate scattered Xenes with increased electrical conductivity and initial discharge capacity.[65]

Xene-induced/enhanced properties in hydrogels

Increased toughness and stiffness. On account of abundant water content, conventional hydrogels have the disadvantages of lower mechanical properties, limiting their applications in many fields. The adulteration of Xenes improves the tensile strength of the hydrogel.[137,138] For instance, Ye et al. introduced GO sheets into the poly(acrylic acid) hydrogels (BIS-gel) and increased the mechanical performance.[139] The hydrogel without GO sheets readily broke and its elongation was just 70%. However, the GO-BIS-gel showed excellent ductility and could even be knotted with elongation at break was close to 300%. Zhang et al. fabricated graphene oxide/polyacrylamide (GO/PAM) hydrogels as actuator devices via *in situ* polymerization.[140] The compressive strength of as-synthesized hydrogels increased significantly with GO content, and it was about six times stronger than pure PAM hydrogel at 1% GO content.

Increased conductivity. The conductive properties of hydrogels are not particularly high, and these can be increased by the addition of Xenes. For example, the electrical impedance values dramatically decreased to 12 kΩ after BP loading, while that of GelMA hydrogel was 46 kΩ.[41] Compared with
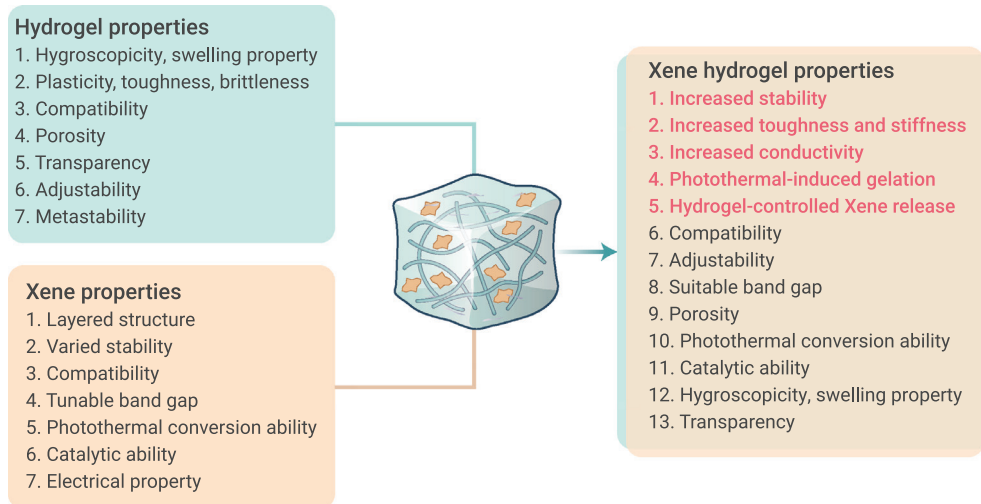
Figure 4. Properties of hydrogel, Xene, and Xene hydrogel

Hydrogel properties
1. Hygroscopicity, swelling property
2. Plasticity, toughness, brittleness
3. Compatibility
4. Porosity
5. Transparency
6. Adjustability
7. Metastability

Xene properties
1. Layered structure
2. Varied stability
3. Compatibility
4. Tunable band gap
5. Photothermal conversion ability
6. Catalytic ability
7. Electrical property

Xene hydrogel properties
1. Increased stability
2. Increased toughness and stiffness
3. Increased conductivity
4. Photothermal-induced gelation
5. Hydrogel-controlled Xene release
6. Compatibility
7. Adjustability
8. Suitable band gap
9. Porosity
10. Photothermal conversion ability
11. Catalytic ability
12. Hygroscopicity, swelling property
13. Transparency

electroconductive hydrogels obtained from the conductive polymers and highly hydrated hydrogels, Xene hydrogel exhibited poor conductivity due to the simple embedding of Xenes into the hydrogel while conductive polymers were blended with the hydrogel matrix to form a continuous conducting 3D structure. In addition, Xenes are not always evenly distributed in hydrogels, which can impair the conductivity of hybrid hydrogels.[141]

Photothermal conversion ability. Conventional hydrogels show distinct energy conversion characteristics after the incorporation of Xenes. Wang et al. constructed CS/AM NSs composite hydrogels with distinguished photothermal properties by introducing antimonene nanosheets (AM NSs) into chitosan.[142] Under 808-nm laser irradiation with the intensity of 1.5 W cm^{-2}, the temperature of the CS/AM NSs hydrogel increased by 30°C within 10 min, while the temperature of that without AM NSs almost remain unchanged.

Photothermal-induced gelation. The photothermal function of Xenes also provides a stimulus resource to design stimuli-responsive hydrogels. Gao et al. fabricated an injectable thermo-responsive BP NSs-based chitosan hydrogel, which was liquid at room temperature.[143] Under the laser irradiation (wavelength 808 nm and intensity 1.0 W cm^{-2}), BP NSs converted light energy into heat energy and rapidly solidified the thermosensitive hydrogel.

APPLICATIONS OF XENE HYDROGELS

Although Xenes and hydrogels have been already used in many fields,[144–148] the combination of these two materials expands their applications due to the increased and enhanced properties. Specifically, Xene hydrogels have been used in biomedical, sensing, energy, and environment, as shown in Table 2. In this part, we explore the current applications of hybrid Xene hydrogels.

Biomedical application

Due to adjustable mechanical properties, exceptional electrical and optical features, biocompatibility, and biodegradability, Xene hydrogels are widely used in the biomedical field, including cancer therapy, tissue engineering, and wound healing (Figure 5). In Xene hydrogel, Xenes are always functional components, and hydrogels are scaffolds. In addition, Xene hydrogels are suitable for designing delivery systems owing to their excellent cargo-loading capacity and controllable release. The pores in hydrogels provide ample spaces and Xenes are characterized by broad surface area and unique surface properties.[161] In addition, due to their excellent biocompatibility, soft structure, and appropriate mechanical strength, the hydrogels are tuned into various dosage forms for drug delivery and controlled drug release through nasal delivery, oral delivery, transdermal and subcutaneous injection, and so on.[162] For example, BP has a conjugate structure with a negative zeta potential of about −25 mV. These surface properties are suitable for loading positively charged or π-π stacked matters like small molecular drugs and small interfering RNA.[163] Excellent properties of BP nanosheets and the structural changes in thermosensitive hydrogels are useful in achieving the purpose of controlled drug release.[150,164] In addition, the drug release rate and release amount can also be tuned by changing the water content of hydrogel, the concentration of nanosheets, light intensity, and irradiation time

of the laser.[165] GO nanosheets contain numerous -OH, -CHO, and -COOH groups on their surface, offering massive active sites to bind with biomacromolecules such as protein and nucleic acids.[166] In addition, the Xenes could convert light into heat that further softens and melts drug-loaded platforms.[48] The drug release rate can also be optimized by laser irradiation duration, laser intensity, the concentration of Xene, and the composition of hydrogels.

Cancer therapy. Photonic therapy (PTT and PDT) is a promising cancer treatment with minimal invasiveness. Xene hydrogels also exhibit excellent photonic performance and good biodegradability. Research revealed that BP-incorporated hydrogels serve as controlled drug release platforms for PTT.[167] Emetine loading in the BP hydrogel system can weaken tumor resistance to BP-mediated PTT, owing to the near-infrared light-controlled release, thus, inhibiting stress granule formation involved in the resistance process.[149] Qin et al. prepared an injectable thermosensitive hydrogel containing BP nanoparticles loaded with gemcitabine. Under near-infrared (NIR)-laser irradiation, the temperature rise in BP material was induced to achieve the intratumoral controlled release of gemcitabine.[150] Similarly, Li et al. encapsulated BP nanosheets and docetaxel micelles in an injectable thermally reversible hydrogel. By controlling the drug release in the tumor tissue, the drug system showed an exceptional therapeutic effect.[49] In addition, tumor cell membrane-coated BP quantum dots, together with PD-1 antibody and immunogenic adjuvants, were also embedded in hydrogels to function as a tumor vaccine.[42] After subcutaneous injection of vaccine hydrogel, the NIR irradiation recruited DC and aided T lymph cell activation, while the immune modulators crippled immune suppression of tumor cells. As a result, the photothermal hydrogel systems activated multi pathways to boost the immune system, realizing the inhibition of tumor recurrence.

Tissue engineering. Tissue engineering is a promising strategy to treat injuries by replacing damaged tissue with manufactured implants.[168] To date, several Xene hydrogels have been used in bone and cardiac tissue engineering. Bone tissue engineering requires materials that promote bone generation and repair.[169,170–172] Studies indicated that graphene and its derivatives promote osteogenic differentiation by acting as a preconcentration platform for osteogenic inducers due to the aromatic structure and strong noncovalent interactions.[173–176] Graphene hydrogel films show cell adhesion, spreading, and proliferation, and also stimulated osteogenic differentiation of stem cells after being implanted into subcutaneous sites of rats. The in vivo swell and crack of the hydrogel indicate its biocompatibility.[151] BP-based hydrogels promote osteogenesis through biomineralization. BP gradually degrades to phosphates, an important constituent of bone, filling injured bone and mineralizing with calcium ions to promote bone generation.[152] Lu et al. fabricated BP and CNT co-loaded hydrogels for bone injury repairment, and the marriage of these materials witnessed better bone tissue engineering effects due to the increased electrical conductivity of the carbon tube under electrical stimulation.[153]

Cardiac cells are non-proliferation cells. Replacing defective cardiac tissue with manufactured hydrogels provides an efficient pathway for severe cardiac injury patients. The contractibility and conductivity of cardiac cells make cardiac tissue engineering very picky about the similar properties of materials,[177] such as graphene hydrogels.[178,179] Ali Khademhosseini's group constructed several graphene hydrogels for connecting cardiac tissues. They designed an anionic plasmids-loaded GO hydrogel for acute myocardial infarction treatment. The loaded DNA efficiently transfected damaged cardiac tissues, and the soft nature of hydrogel reduced the injury of injected materials.[154] Another example was methacryloyl-substituted recombinant human tropoelastin (MeTro)/GO hybrid hydrogel. GO served as the hydrogel network connector under compression. The addition

Table 1. The comparisons between different Xene materials

Periodic group	Element	2D form	Stability	Tunable band gap	Photothermal conversion efficiency (%)	Catalytic ability	Electrical property
IIIA	B	borophene	Highly chemical stable, photostable[30]	Zero or near-zero bandgap;[78] opened by hydrogenation/fluorination or a shear strain (in theoretical calculations)[79]	42.50%[30]	Hydrogen evolution;[80] oxygen reduction; oxygen evolution; CO_2 electroreduction[79]	High electrical conductivity (in theoretical calculations)[81,82]
IIIA	Ga	gallenene	stable[83]	zero or near-zero bandgap[78]	no report	no report	metallic;[84] superconductivity[85]
IVA	C	graphene	aggregating in saline[86]	zero or near-zero bandgap[78]	49.13% (single-layered graphene); 46.26% (GO)[87]	nitrobenzene hydrogenation (GO);[88] oxidation of sulfides, olefins and various hydrocarbons (GO)[89]	high electrical conductivity (in theoretical calculations)[78]
IVA	Si	silicene	biodegradable[90]	zero or near-zero bandgap;[78] opened under vertical electric field (in theoretical calculations)[91]	36.09%[90]	CO_2 hydrogenation;[92] nitrobenzene reduction[93] (in theoretical calculations)	resistance: 224~823Ω under different conditions[94]; high electrical conductivity[78]
IVA	Ge	germanene	rapid degradation[95,96]	opened under vertical electric field (in theoretical calculations)[91]	38.40%[95]	picric acid violet light-assisted photodegradation (hydrogenated germanene)[97]	semiconducting (in theoretical calculations);[98] from insulator to metal after heating[99]
IVA	Sn	stanene	aggregating in saline[100]	tuned under vertical electric field (in theoretical calculations)[101]	48.60%[100]	CO_2 reduction (in theoretical calculations)[102]	semiconducting (in theoretical calculations)[103]
VA	P	phosphorene	biodegradable (black)[25]	layer-dependent band gap[78]	19.8°C increased after irradiation (BPQD, 10 ppm, 10 min, 1 W cm^{-2})[104]	oxygen evolution;[105,106] $^1O_2$ generation;[107] ·OH and ·O_2^- generation[108]	semiconducting (in theoretical calculations)[109]
VA	As	arsenene	biodegradable[110]	layer-dependent band gap (in theoretical calculations)[111]	14 °C increased after irradiation (100 μg/mL, 5 min, 2 W cm^{-2})[110]	Chlorine evolution[112]	Semiconducting (in theoretical calculations)[113]
VA	Sb	antimonene	Highly stable in the atmosphere[114,115]	layer-dependent band gap (in theoretical calculations)[111]	44.60%[116]	Ammonia synthesis under ambient conditions (oxidized antimonene)[117]	Semiconducting (in theoretical calculations)[118]
VA	Bi	bismuthene	Biodegradable[119]	Shrank with nerve agents assimilated on the surface[120]	21.3%[119]	CO_2 reduction;[121] CO generation[119]	Semiconducting: electronic conductivity reached up to 10^8 S m^{-1} (in theoretical calculations)[122]
VIA	Se	selenene	biodegradable[123]	no report	no report	no report	semiconducting (in theoretical calculations)[124]
VIA	Te	tellurene	biodegradable[125]	layer-dependent band gap (in theoretical calculations)[126]	55%[125]	no report	semiconducting[78]

Table 2. The applications of Xene hydrogels

Field	Application	Xene	Polymer/Other Linker
Biomedical	Cancer therapy	BP nanosheets	Low-melting-point agarose[149] Pluronic F-127[150] Pluronic F-127 and hyaluronic acid[42]
	Bone regeneration	graphene	no polymer[151]
	Bone regeneration	BP nanosheets	Gelatin methacrylate and U-Arg-PEA[152] oligo(poly(ethylene glycol) fumarate)[153]
	Cardiac tissue engineering	graphene	low methacrylated gelatin methacrylate[154] methacryloyl-substituted tropoelastin[155]
	Neural differentiation	BP nanosheets	gelatin methacryloyl[41]
	Wound healing	BP nanosheets	fibrin[156]
	Wound healing	antimonene	chitosan[142]
	Wound healing	germanene	chitosan and agarose[27]
Sensing	Biosensor	graphene	L-cysteine[157]
Energy	Battery	graphene and BP nanosheets	no polymer[65]
	Battery	graphene	no polymer[46]v
	Supercapacitor	graphene	polyaniline[158]
	Hydrogen production	graphene	no polymer[159]
Environment	Water purification	BP nanosheets	chitosan[43]
	Water purification	graphene and BP nanosheets	no polymer[160]

of GO enhanced the torsion tolerance and conductivity of protein hybrid hydrogels.[155]

The conductivity of Xene hydrogel (GelMA–BP@PDA) is also applied for neural-like cell differentiation from mesenchymal stem cells under electrical stimulation.[41] The immunofluorescent staining revealed an increased level of neuronal-specific marker β-Tubulin III (Tuj1) and MAP2 of GelMA–BP@PDA, compared with the GelMA hydrogels under electrical stimulation, and a slight decrease in glial marker GFAP content.

Wound healing. The sprayed gel can cover the wound tissue in a large area, and Xene incorporation can promote wound repair. Therefore, an intelligent BP-based gel was developed by our group with rapid formation and NIR response to solve chronic diabetic ulcers.[156] Figures 6A−6C show that the *in situ* sprayed BP-based gel can act as a temporary bionic "skin," accelerating the healing of chronic wounds by promoting endothelial cell proliferation, and angiogenesis, temporarily shielding tissue from contact with the external environment. In addition, the hydrogel is also used as a drug "repository" to store bactericidal BP and painkiller lidocaine hydrochloride (Lid). Similarly, Wang et al. incorporated AM NSs into the chitosan network structure to fabricate a chitosan/AM NSs hydrogel composite, in a first attempt to apply AM NSs in the antibacterial field. The hybrid hydrogel collected bacteria through electrostatic interaction of chitosan with their cell membrane and eliminated them by outstanding photothermal effect of AM NSs. During the *in vitro* experiment, the AM-based hydrogel demonstrated a broad-spectrum antibacterial property against *E. coli* (97.1%) and *S. aureus* (100%).[142]

The sprayed Xene hydrogel can induce rapid gelation to form a gelled membrane on tumor resection wounds under a rapid photo-induced sol-gel transition upon photoirradiation. Complications such as the infection occurring after surgery can be addressed by Xene hydrogels owing to the good photonic antibacterial performance of the proposed synthesized membrane. Furthermore, germanene hydrogels can serve as post-surgical treatment agents to prevent tumor recurrence and avoid wound infection.[27]

Sensors

The biosensor is a device consisting of an identification and signal conversion element, responding to target variations (e.g., concentration changes of materials), with high selectivity and analytical sensitivity.[180–184] The basis for Xene hydrogels as sensors originates from the optical/electrical performance and their modifiable surface, stimuli responsiveness of hydrogels, and their adsorption capacity.[185–187] In other words: (1) the Xene and hydrogel can capture targeting materials via several interactions; (2) the stimulus-sensitive hydrogels can change the gelatinous state with the external environment; and (3) Xenes can improve the sensitivity and reaction time of hydrogel biosensors due to abundant sp2 bonds and increased electrochemical reactivity and mass transfer of electrons. Singh et al. fabricated a dual-modality microfluidic biosensor based on graphene-based hydrogel to quantify human cardiac myoglobin (cMb) (Figure 6D).[157] They used reduced GO with an l-cysteine amino acid to connect the cMb antibody via covalent interactions (-COOH on cysteine and -NH$_2$ on antibody), as shown in Figures 7E–7G. The CV responses of RGO, Cys, and Cys-RGO hydrogel electrodes and cMAb/Cys-RGO bioelectrode indicated that Cys-RGO hydrogels offered a larger porous surface and higher electron transfer rate than Cys alone, thus improving the biosensor efficiency. Differential pulse voltammetry responses were measured by a microfluidic chip (CMAB/Cys/Au) without RGO hydrogel and different concentrations of standard cMb were introduced. The response peak current decreased with the increase in cMb concentration, indicating the effectiveness of the designed sensor.

Energy storage and production

In recent years, the construction of efficient energy storage devices has gained tremendous attention due to the increasing industrialization and growing population.[188] Among them, supercapacitor with excellent energy storage capacity and power density has been highly demanded, which stores energy either by surface adsorption of electrolytic ions or fast surface redox reactions.[189] Xenes exhibit exceptional properties as electrode materials.[189] For example, Mei et al. prepared an interesting BPNs@TiO$_2$@G hydrogel with an exceptional 2D-TiO$_2$-2D structure. Compared with traditional hydrogels, this 2D-TiO$_2$-2D structure exhibits outstanding high specific surface area, abundant hierarchical porous, and excellent conductivity to store and release Li ions during charging and discharging. High discharge capacity significantly improved cyclic capacity and superior rate capacity. Such a BPNs@TiO$_2$@G hydrogel provides a valuable research direction for the development of other metal oxide nanomaterials with efficient energy storage, inspired by the interfacial chemistry of energy storage materials (Figures 7A–8C).[65] This team also successfully fabricated a highly phosphorus-doped 3D graphene hydrogel through a simple one-step hydrothermal process to achieve increased Li-ion storage.[46] Extremely high amounts of
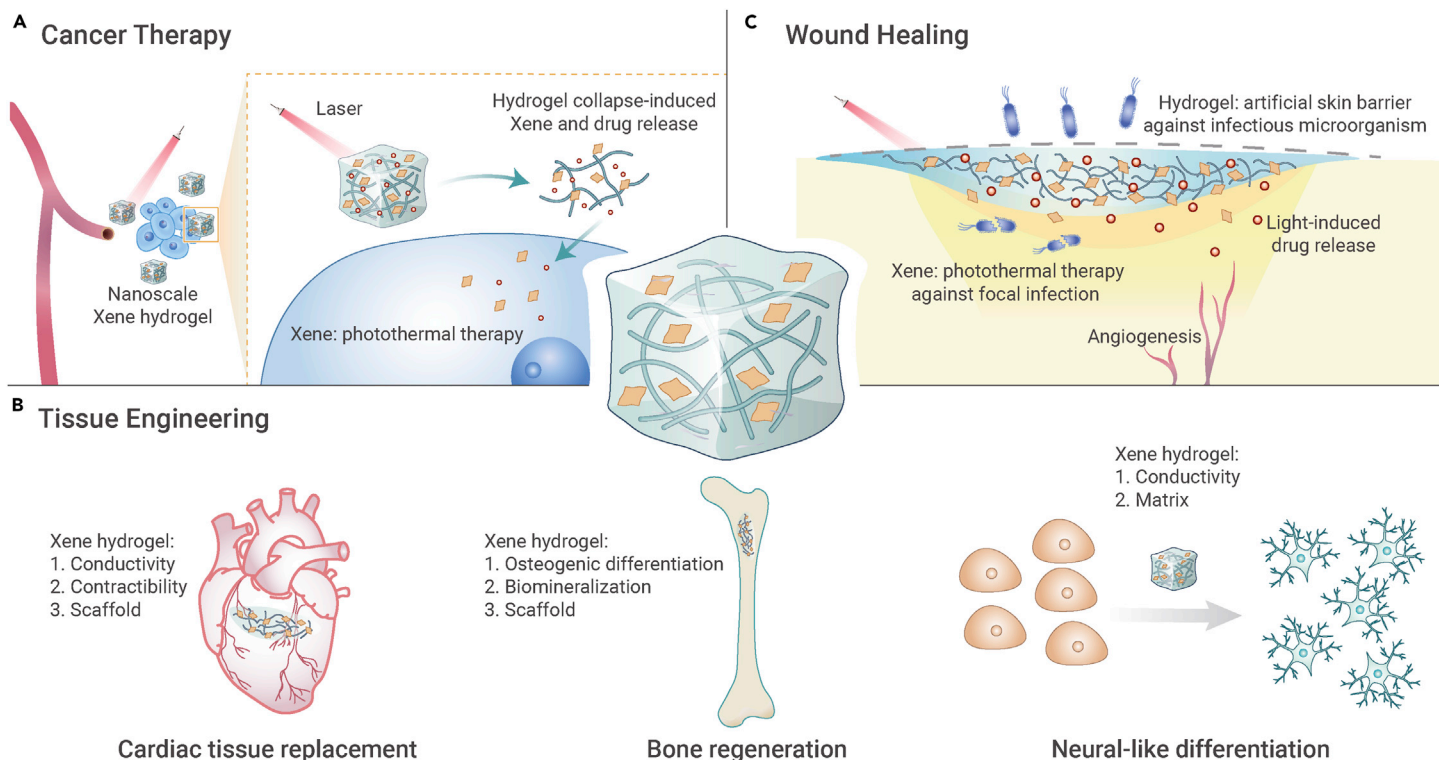
Figure 5. Medical applications of Xene hydrogels (A) Loaded with therapeutic drugs, nanoscale Xene hydrogel can be delivered to the tumor site and be decomposed in tumor cells upon light irradiation to release the cargos. Xenes can function as photothermal agents against cancer cells. (B) Xene hydrogel can be utilized in cardiac tissue replacement, bone generation, and neural-like differentiation. (C) Xene hydrogel can be applied to protect wounds from infectious microorganisms. Similar to the case in the tumor site, Xene can convert light energy into heat to kill focal bacteria. Also, released cargos can exert different functions such as promoting angiogenesis.

phosphorus (even up to 4.84%) and various sizes of apertures (ranging from 1.7 to 17.5 nm) were used in phosphorus-doped 3D graphene hydrogel for construction of heterostructured 2D BP on graphene. Therefore, Li-ion storage performance was further boosted by abundant and stable C-P bonds, significantly shortening the circulation time and are convenient to obtain and lose Li-ion. This is a potential strategy for the future development of phosphorus-doped hydrogel networks while avoiding using highly toxic organophosphates.

Moreover, the conductive and stretchable properties of the hydrogel make it particularly suitable for flexible electronic materials. Xiao et al reported a 3D polyaniline/graphene hydrogel-based conceptual all-gel-state fibrous supercapacitor with excellent plasticity and electrochemical properties. The supercapacitor is designed through the self-assembly of GO and carbon nanotube macromolecular structure (Figure 7D).[158] Due to the strong macromolecular interaction between carbon fiber and graphene, the 3D hybrid hydrogel exhibited uniform connectivity and enhanced mechanical properties, thereby greatly reducing aggregation during fiber formation. In addition, a large strain (up to 40%) was achieved and the volumetric energy density reached up to 8.80 mWh cm^{-3} at a power density of 30.77 mW cm^{-2}.

Solar photocatalytic hydrolysis is an important way to alleviate energy and environmental pressure to produce hydrogen. Ho et al. prepared graphene and TiO$_2$ hybrid NGH hydrogel to produce hydrogen under light.[159] In the absence or presence of UV-visible, 3D NGH, and NGH-Au generated hydrogen more efficiently than 1D or 2D TiO$_2$ and RGO-TiO$_2$, attributed to the 3D hydrogel structure. which is more conducive to charge quick separation.

Water purification

The water purification by Xene hydrogels mainly includes two steps: pollutant capture and disposal. The pollutants such as bacteria, organics, and inorganic particles are adsorbed in hydrogel pores or on the Xene's surface and subsequently sterilized or decomposed via the enhanced functional Xene.[190–194] Compared with traditional physical (UV, heat sterilization by size exclusion, etc.) and chemical methods (chlorine, ozone, and metal ion), Xene hydrogels are less toxic to human health, have low energy consumption, and generate less environmental pollution and global warming.[195,196] Li et al. constructed a sandwich-liked water disinfection system (CSBPP) based on chitosan hydrogel, BP

nanosheets, and layer-by-layer stacking of chitosan hydrogel (Figure 8A). At an appropriate concentration of glutaraldehyde, the removal efficiency of bacteria reached 98%, as shown in Figure 8B. At the same time, BP nanosheets showed excellent photothermal conversion efficiency under NIR radiation, enhancing the CSBPP's ability to kill bacterial cells adsorbed on hydrogel scaffolds through the photothermal effect (Figure 8C). CSBPP circumvented membrane fouling caused by undesirable bacterial growth and was more environmentally benign, revealing an overall potential for potent, sustained, and safe water disinfection.[43]

Many researchers developed highly reusable and superhydrophobic aerogels or hydrogels for photocatalysts to eliminate water waste and air pollutants.[197–199] For sewage treatment, Wang et al. prepared rGO-BP-Pd composite hydrogel using rGO as a catalyst carrier (Figure 8D). The prepared hydrogels showed good catalytic performance for 4-nitrophenol. Specifically, with the participation of rGO-Pd-based composite hydrogels, NaBH$_4$ provided negative hydrogen ions to attack 4-nitrophenol and reduce 4-nitrophenol to less toxic 4-aminophenol (Figure 8E). The reasons for the high efficiency of the catalytic system are revealed in Figure 8F. Due to favorable band structure, the photoexcited electrons in the conduction band of BP NS were transferred to the conduction band of Pd. Moreover, the hole in the valance band of PdNPs was transferred to the valance band of BP NS. Since the electron-hole pairs in BP NS and Pd were relatively independent, the charge carriers exhibited a long lifetime.[160]

CONCLUSIONS AND FUTURE OUTLOOK

Various Xene hydrogels have rapidly emerged in recent years with significant advantages over many other static platforms, establishing new standards for the development of complex and powerful platforms. Tremendous design flexibilities and unique opportunities for soft devices have benefited from the combination of Xenes and hydrogels, and the gain-of-function has enabled those devices with exotic features and augmented application performance. In this review, Xene hydrogels have been discussed in detail, especially the synthesis methods, properties, and applications. The general characteristics, advantages, and disadvantages of each synthesis method are summarized for readers to choose a suitable method according to different needs. Then the general and special properties of hydrogels and emerging Xenes are introduced in detail and compared with traditional hydrogels to illustrate their advantages and challenges.
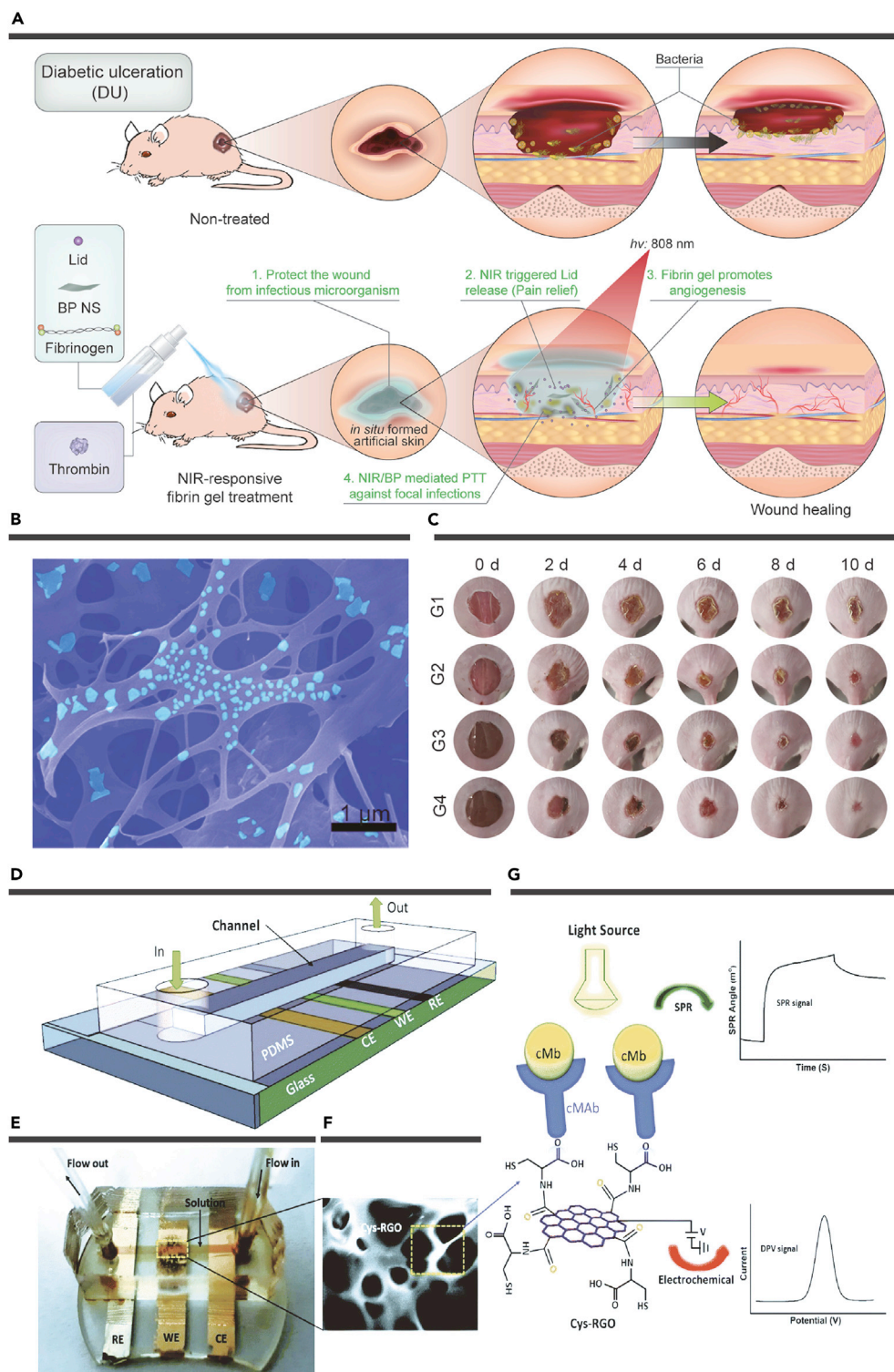
gregation of skeletal muscle fiber bundles are vital to their related functions.[203–205] However, most of the existing Xene-based hydrogels have uniform Xene distribution and disordered structure, hindering their anisotropic mechanical properties and functions. Therefore, the development of highly ordered Xene-based hydrogels can achieve anisotropic mechanical, optical, and biological properties, further promoting the development of Xene-based hydrogels in bioengineering applications. Also, the research on theory and mechanism will provide great guidance for the rational design of Xene hydrogels.

The interaction between additives and macromolecules can provide another direction for the development of anisotropic materials. For example, γ-glycine can self-assemble between two PVA films via hydrogen bonding (O atoms of glycine and -OH of PVA chain). The sandwiched thin film is heterostructured and piezoelectric, beneficial for biomechanical energy (such as leg stretching).[206] Similarly, Xenes can be orderly added into hydrogels to form heterogeneous materials since 2D materials with π bonds and controlled surface chemical modification can react with multitudinous hydrogel polymers.

Also, the Xene distribution can be regulated by external factors. Anisotropic materials are easier to self-align in the desired orientations where the dielectrically/magnetically susceptible axis is parallel to the external, uniform, and static field.[207] For example, exploiting the intrinsic anisotropic and magnetic susceptibility of carbon nanotubes is conducive to developing anisotropically luminescent hydrogels via an external magnetic field.[208] Organic polar nanocrystals are effectively polarized and orientated in polymer matrices by applying a direct-current electric field, functioning as an electro-optic material for fast and high-capacity optical information processing.[209] Therefore, Xene nanosheets with intrinsic anisotropy can be aligned by external electric or magnetic fields.[210] Mechanical strain changes the alignment of additives with a load transfer medium, i.e., the polymer can also increase the ductility for large plastic deformation.[211] Moreover, stretching polymers can direct them into anisotropic hydrogels due to the maximum crosslinking. Moreover, the Xene distribution can be regulated by external factors as well. Anisotropic materials can be self-aligned in specific orientations with dielectrically/magnetically susceptible axis parallel to the external, uniform, and static field.[207] For example, exploiting

Finally, the applications of Xene hydrogels are explored in biomedicine, pollution prevention, and electronic equipment. Our insights on the challenges and future directions of Xene hydrogel in other potential applications are also expressed.

Preparing anisotropic hydrogels

Highly ordered Xene-based hydrogels are a new direction in the area of hydrogel bioengineering.[200] In the human body, different tissues such as blood vessels, cornea, and cartilage are natural hydrogels.[201,202] They are composed of a variety of cells and proteins arranged in an orderly manner, essential to achieve corresponding biological functions. For example, the ordered layered structure of the cornea and the directional ag-
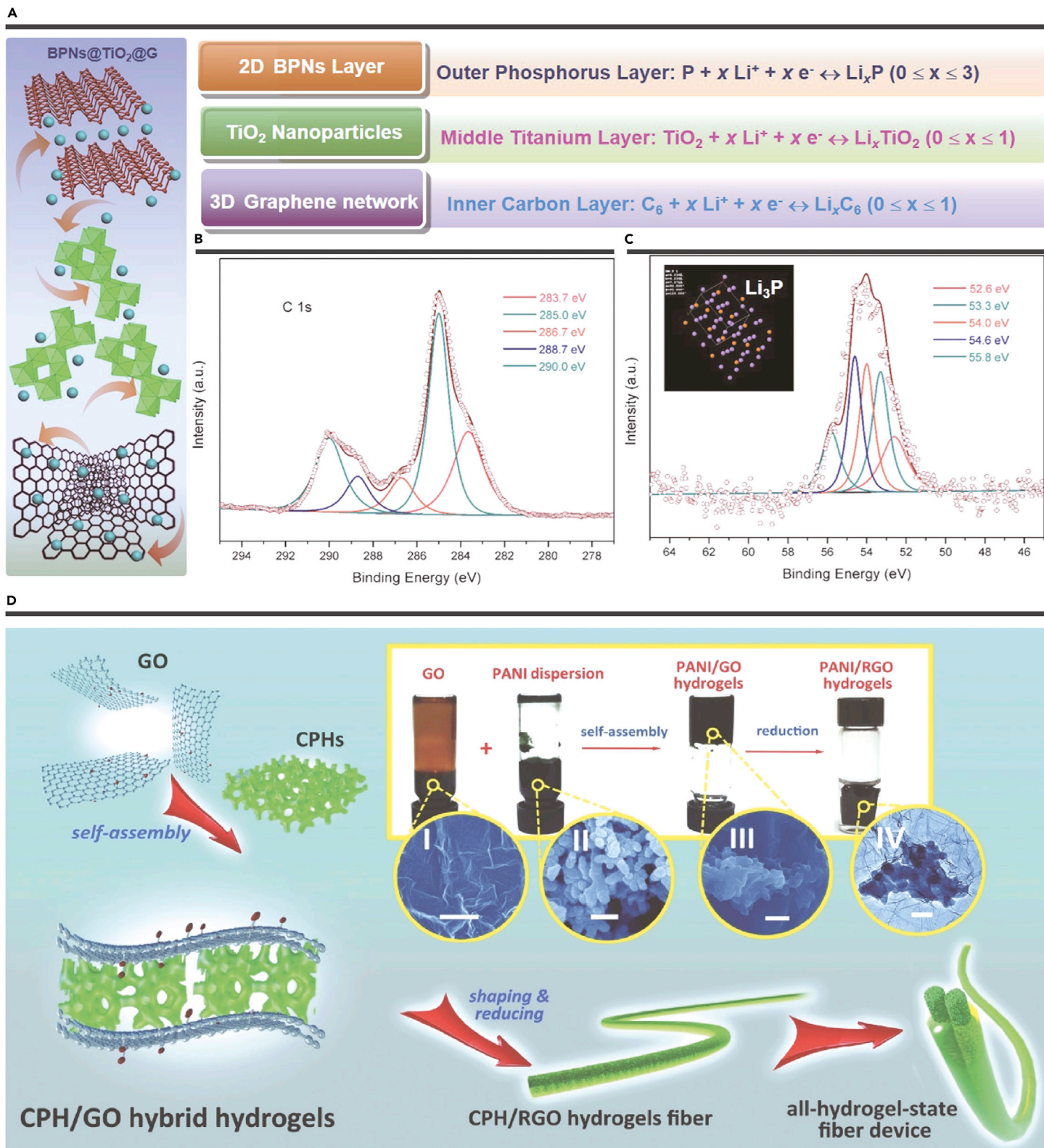
Figure 7. Lithium storage mechanism of 2D-TiO$_2$-2D heterostructured electrode and PANI/GO hybrid hydrogels formation, shaping/reduction process, and their conductivity (A) Schematic illustration on possible diffusion and transfer paths of Li$\dot{p}$ ions in BPNs@TiO$_2$@G composite electrode, (B) C 1s and (C) Li 1s spectra of BPNs@TiO$_2$@G electrode after cycles. Inset in (C) shows the crystal structures of Li$_3$P.[65] (D) Schematic illustration of PANI/GO hybrid hydrogels formation and further shaping/reduction process; photos of the self-assembly of PANI/GO hybrid hydrogels and the reduction of PANI/RGO (the inset) as well as the corresponding SEM and TEM images of samples after freeze-drying[158] (copyright Elsevier, 2019; copyright John Wiley and Sons, 2018.)

the intrinsic anisotropic magnetic susceptibility of carbon nanotubes can develop anisotropically luminescent hydrogels via an external magnetic field.[208] Organic polar nanocrystals can be effectively polarized and orientated in polymer matrices under a direct-current electric field, functioning as an electro-optic material for fast and high-capacity optical information processing.[209] Therefore, Xene nanosheets with intrinsic anisotropy exhibit great alignment potential under external electric or magnetic fields.[210] Mechanical strain can also change the alignment of additives under a load transfer medium, i.e., the polymer can increase the ductility for large plastic deformation.[211] Stretching polymers can produce anisotropic hydrogels due to maximum crosslinking.[212,213]
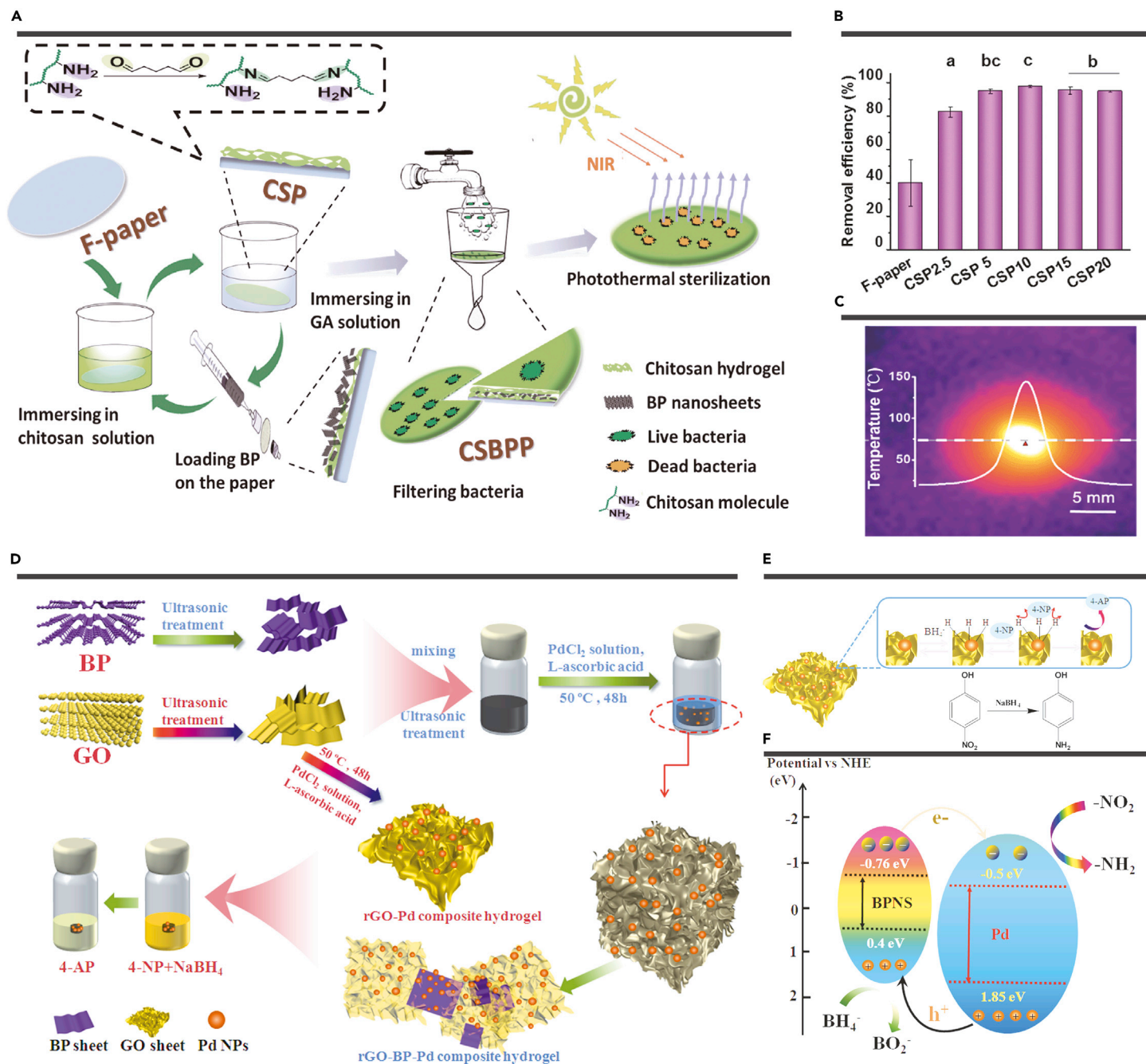
Figure 8. Application of Xene hydrogel in water treatment (A) Fabrication process of the sandwich-structured filter systems and the description of bacteria removal–disinfection. (B) Removal efficiency for *E. coli* treated with the F-paper and CSP with different crosslinking degrees.[43] (C) Thermographic image of CSBPP4 after 10 min of NIR illumination. (D) Schematic illustration of the synthesis process and catalytic performances of the prepared rGO-based composite hydrogels. (E) Schematic of the Langmuir-Hinshelwood model and (F) the catalytic mechanism for 4-NP catalytic reduction[160] (copyright Elsevier, 2020; copyright Royal Society of Chemistry, 2019).

Developing artificial skin

Artificial skin first appeared in skin wound healing with promising efficiency as protective, soft, stretchable, water-rich, and self-healable characteristics.[214] Hydrogels with appropriate mechanical properties (e.g., 0.5–1.95 MPa of Young's moduli, 140%–180% of ductility, and over 70% of water content) have also been listed as potential candidates.[215] Nowadays, artificial skins are more like electronic sensors. Similar to real skin, delivering surrounding variables to the nervous system, the artificial skin always converts mechanical stress or temperature into electrical signals.[216] Xene is the icing on the cake in this field,[216] due to the perfect mechanical properties of hydrogels and increased conductivity, as mentioned above. More importantly, some Xenes are piezoelectric and even pyroelectric, such as BP, imparting outstanding sensing performances to the artificial skin. In addition, the Xene hydrogel-based artificial skin is more sensitive than the ZnO-based skins due to the plane structure, more active sites, and larger electron cloud density.[217] Related research proved that the photocatalytic

performance of additives can change the inner structure and electronic conductivity of hydrogels.[218] Xene hydrogel-based artificial skin can increase the antibacterial ability also under external environments by producing reactive oxygen species.[219]

Clinical translation of *in vivo* applications

Xene hydrogels can be applied *in vivo* via surgical implantation, local injection, or systemic administration. The priority is safety and biocompatibility. Most hydrogels are biocompatible; however, the metabolism of Xenes has not been fully understood yet. Figuring out the interactions between the body and Xenes is conducive to exploring more *in vivo* applications. Stability is another key point to be taken into consideration. Surgical implants (such as tissue scaffolds) always work for a long time and require long-term structure and function stability. In this case, degradable BP is not an ideal choice. However, due to the injectability of most hydrogels, they can be used as drug reservoirs and controlled drug

release, and the type and amount of Xenes should promise the injectability of hydrogels. Although Xene-based nanogel can be developed for intravenous injection, the systemic side effects cannot be ignored. Despite this, Xene hydrogels have significant clinical transformation value due to the combined effect of the two components.

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DECLARATION OF INTERESTS

The authors declare no competing interests.

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