Check for updates

Organogels versus Hydrogels: Advantages, Challenges, and Applications

Mariia A. Kuzina, Dmitrii D. Kartsev, Alexander V. Stratonovich, and Pavel A. Levkin*

Organogels are an important class of gels, and are comparable to hydrogels owing to their properties as liquid-infused soft materials. Despite the extensive choice of liquid media and compatible networks that can provide a broader range of properties, relatively few studies are reported in this area. This review presents the applicability of organogels concerning their choice of components, unique properties, and applications. Their distinctive features compared to other gels are discussed, including multi-stimuli responses, affinity to a broad range of substances, thermal and environmental stability, electronic and ionic conductivity, and actuation. The active role of solvents is highlighted in the versatility of organogel properties. To differentiate between organogels and other gels, these are classified as gels filled with different organic liquids, including highly polar organic solvents and binary solvent systems. Most promising applications of organogels as sophisticated multifunctional materials are discussed in light of their unique features.

1. Introduction

A gel is a non-fluid colloidal or polymer network that is expanded throughout its whole volume by a fluid (IUPAC Gold Book).[1] Organogels are gels with organic liquids as the fluid phase. They can swell and retain large (up to 99%) quantities of the liquid phase, absorb and release substances, and respond to various physical and chemical stimuli, such as tem-

M. A. Kuzina, P. A. Levkin
Institute of Biological and Chemical Systems – Functional Molecular Systems (IBCS-FMS)
Karlsruhe Institute of Technology (KIT)
Hermann-von-Helmholtz-Platz 1
76344 Eggenstein-Leopoldshafen, Germany
E-mail: levkin@kit.edu
D. D. Kartsev, A. V. Stratonovich
SCAMT institute
ITMO University
9 Lomonosova str. 191002, Saint Petersburg, Russia
P. A. Levkin
Institute of Organic Chemistry (IOC)
Karlsruhe Institute of Technology (KIT)
Fritz-Haber Weg 6, 76131 Karlsruhe, Germany

perature, light, pH, and mechanical deformation. The broad functionality range of organogels arises from the combination of properties given by the organic liquid and gelator components. Given the wide choice of organic liquids and compatible gelators, the properties and functionalities of organogels are varied. Despite their potential and increased research interest in this field,[2] there have been few studies, as indicated by the number of related publications (2253) during the period 2012–2021 (Scopus), compared with the number of papers mentioning hydrogels (70956) during the same period (Scopus). Hydrogels have been immensely popular because they retain large amounts of water and aqueous solutions, often making them biocompatible with natural tissue-mimicking properties. Nevertheless, hydrogels have limitations with respect to the choice of gelators,[3] poor thermal and environmental stability,[4,5] and poor affinity to hydrophobic compounds.[3,6,7] One of the main advantages of organogels is that the choice of a liquid phase is much wider than that for hydrogels, enabling more applications of organogels compared with hydrogels. As an example, the thermal stability of a gel can be manipulated using the boiling point of the chosen solvent.[8,9] Environmentally stable gels can be created by swelling the gels with lubricants, providing hydrophobicity,[10,11] ice-phobicity,[12,13] and anti-biofouling properties.[14,15] However, the application of organogels is not limited to achieving orthogonal properties of hydrogels, and new combinations of the whole range of available gelators with all kinds of organic liquids reveal the potential for the application of multifunctional organogels.

One of the obstacles in understanding the potential of organogels includes existing biases about their properties ("hydrophobic," "toxic," and "non-conductive") owing to the ambiguity regarding the classification of gels. Therefore, it is important to establish a precise classification system for gels, specifically organogels. To achieve this, the existing gel-related terminology should be defined by the nature of the liquid (gas) media or the active components of the gel, where gels containing the organic fluid phase are classified as organogels.

Recent reviews about organogels have focused primarily on physically cross-linked oil-based organogels applied as carriers for transdermal drug delivery[16–19] or to replace trans-fat in various food products.[20–22] Zeng et al. performed a detailed study on organogels, including the relationship between applied solvents and gelators and gel behavior and applications,

**ADVANCED
SCIENCE NEWS**

www.advancedsciencenews.com

**ADVANCED
FUNCTIONAL
MATERIALS**

www.afm-journal.de

and highlighted areas, such as i) anti-icing, ii) anti-fouling, iii) droplet manipulation, iv) drug delivery, v) food processing, and vi) other applications.[2] However, with the recently increased research in this field, new functional materials have been developed, opening new directions for the application of organogels, such as actuators, supercapacitors, and oil–water separation membranes. Besides, organogels are among the most versatile and promising classes of multi-phase materials, having the potential to outscore hydrogels and dominate in particular areas.

This review presents research progress in organogels with distinctive, unconventional properties. As the focus is to highlight new perspectives on organogels, we exclude previously well-reviewed areas of organogels for the food industry and transdermal drug delivery. To overcome existing biases and ambiguity in the terminology of gels, we overview the existing classification of gels and propose a precise definition of organogels as a separate class. To further promote organogel development, we focus on recent progress regarding their unique capabilities, accentuating the active role played by organic solvents in gel functionality. As distinctive perspective features for the application of organogels, we consider the: i) multi-stimuli response, ii) durability and long-lasting environmental stability, iii) ability to absorb and release substances, iv) special wettability, and v) actuation.

2. Gel Classification

Owing to the wide range of gel-type materials and their complex composition, it remains challenging to organize and classify gel types. We summarize various principles for classifying gels into classes and sub-classes (**Figure 1**). Except for the general term

proposed by the IUPAC for gels (given at the beginning of this review), there are several recommendations for types of gels according to gel composition, namely "hydrogel," "alcogel," "aerogel," and "xerogel." Hydrogels are classified as gels "in which the swelling agent is water,"[1] and alcogels are gels "in which swelling agent consists predominantly of an alcohol or a mixture of alcohols."[1] The polymer networks where the "dispersed phase is a gas," are called aerogels,[1] whereas a xerogel is an "open network formed by the removal of all swelling agents from a gel."[1] Therefore, both aerogels and xerogels are conventionally formed from initially wet gels. They are distinguished as follows: aerogels preserve most of the gel network after removing the liquid phase, e.g., by the fast replacement of the liquid with the gas, whereas xerogels are obtained via conventional drying methods, causing significant shrinkage and a collapse of the original gel network.[23]

The recent trend in gel terminology is to differentiate gels based on their main functional component. Many standard terms have thus been established regarding the active species giving special gel functionality: a liquid phase, a network, or additional components. Examples of such terms include "ionogels" (functionality derived from the ionic liquid as a dispersed phase),[24,25] "PEGgels" (functional phase is poly(ethylene glycol)),[26,27] "magnetogels" (gels containing magnetic nanoparticles),[28] and "fluorogels" (fluorinated compounds in the solid or liquid state).[29] The continuous updating of terms helps to organize the gained common knowledge about particular gels, but significantly limits their characterization as a class and hinders clear and efficient scientific communication.

With increasing progress in gel research, existing terminologies do not include many developed functional gels. Importantly, for organogels, which are the second largest type of gels after hydrogels, there is no standard terminology, and many
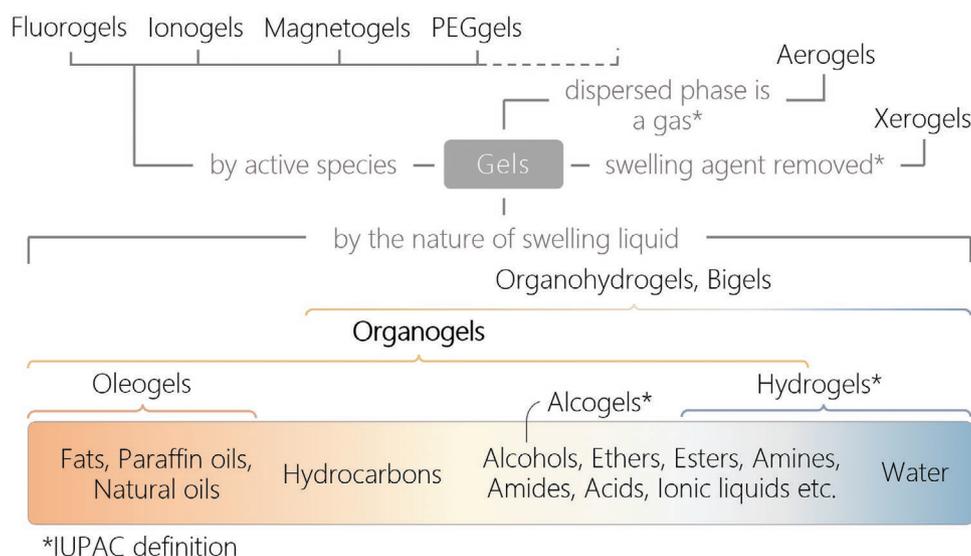
Figure 1. Classification of gels based on their content. Gel types refer either to the nature of the swelling phase (gas, organic liquids, and water) or to the most functional component (e.g., fluorinated monomers, ionic liquids, magnetogels, and PEG). Organogels and hydrogels can overlap when the liquid phase is a mixture of aqueous and organic solvents. Gels, where liquid phases and/or 3D networks of opposite polarities are interconnected or have a clear interface, are called organohydrogels or bigels. When classified based on the nature of the swelling liquid, organogels are gels filled with organic liquids, including polar organic solvents and their aqueous mixtures, ionic liquids, fats, and oils, among others. This definition reflects the most active role of the liquid phase in organogel functionality and their versatility as a separate class of gels.

definitions currently co-exist. In recent publications, organogels have been classified as follows: i) "Organogels (or oleogels) contain apolar liquids, such as organic solvents or mineral or vegetable oils, as their continuous phase;"[30] ii) "Organogels (hydrophobic polymer gels) are soft materials based on polymeric networks swollen in organic solvents;"[31] and iii) Zheng et al. reported that "Organogels are distinguished from hydrogels by adopting organic filling liquids."[2]

Among those, iii) has the closest description for the whole range of organogels, whereas i,ii) are limited to apolar or hydrophobic solvents only and specify only polymer or colloidal (a liquid is the only continuous phase) networks. Therefore, we define organogels as colloidal, supramolecular, or polymer 3D networks filled with organic liquids. Such a definition gives the flexibility required to classify the broad range of gels and to combine existing organogels as a separate class. For example, it does not limit organogels by the exact content of the liquid phase, its complexity, or its specific function. Accordingly, narrower groups of gels, such as ionogels or magnetogels, can be classified as either hydrogels or organogels, while oleogels, PEGgels, and alcogels are sub-classes of organogels (see scheme in Figure 1). There will always exist a noticeable overlap between organogels and hydrogels in the case of binary solvent systems.[5,32,33] For such cases, we define a gel according to the nature of the predominant or the most functional solvent. For gels, where 3D networks and solvents of opposite polarity can co-exist in one material with a clear interface, the term "bigel" or "organohydrogel" is appropriate (Figure 1).

3. Role of Solvent Choice in Gel Versatility

When a gel system is designed, the solvent plays a critical role, first with the choice of a gelator, then during the gel formation process, and after the gelation is complete. For a supramolecular network, the solvent structure may determine optimal gelation parameters, such as initiation[34] or impede stable gelation.[35–37] The self-assembly of gelator molecules, which is the core process in supramolecular gelation, depends upon reversible non-covalent interactions, such as van der Waals forces, π–π stacking, and hydrogen bonding. Consequently, flexible and stable gelation and higher gelation numbers are achieved in solvents possessing limited non-covalent interactions with the gelator. Zhu et al. demonstrated this principle using the example of trehalose diester gelators. Strong gelator–solvent interactions caused the clustering of gelator assemblies, resulting in the formation of thick unstable gels with lower gelation numbers.[38] Therefore, when designing a supramolecular organogel, it is important to fine-tune the solvent or chemical environment of the specific gelator.

The choice of a solvent also affects the process of polymer network formation. For instance, the co-monomer reactivity ratio in radical copolymerization varies in different solvent systems,[39–42] which is described as the "bootstrap" effect. However, the solvent effect is closely linked to specific copolymerization systems.[39] As well as in supramolecular networks, the knowledge of solvent–gelator interaction for specific networks is not fully transferrable and has to be collected for new solvent systems, such as deep eutectic solvents.[43]

The structure of the resulting gel is affected by the properties of the solvent. Therefore, specific fabrication methods that consider the solvent effect, are required to produce gels with defined properties. Whereas most of the existing knowledge is attributed to hydrogels, the methods developed for hydrogels are not reproducible with non-polar media owing to complete changes in solvent–gelator and solvent–network interactions. Meanwhile, by introducing new solvents to different networks, greater functionality can be achieved, which we will discuss later.

After gelation is complete, the gel acts as a quasi-solid material, where the bulk and surface properties are defined by the 3D network, the liquid phase, and their interaction in the material. According to the polarity of the gelator and the entrapped solvent, the gel may be hydrophilic,[44] hydrophobic,[31] or amphiphilic.[45] The surface of a gel does not behave as an ordinary solid surface. As the solvent extrusion occurs, the liquid surface layer defines the wettability properties. The properties of the bulk, such as its affinity to different substances, ionic conductivity, stimuli responsiveness, and stability are defined both by the nature of the solvent and the 3D network.

Different applications require different polarities to obtain the best performance. For biocompatibility,[46] pH-sensitivity,[47] and ionic conductivity,[48–55] polar liquid media are essential (**Figure 2**A). These applications are covered by hydrogels, but can still be reached by organogels with polar media,[56] whereas only non-polar low-surface-tension liquids introduce hydro- and omniphobic properties to the gel surface,[10,11,57–64] providing functions, such as self-cleaning[65] or anti-fouling,[10,60] and water droplet manipulation.[57–59,61,66,67]

The solvent also plays a significant role in thermal and environmental stability, as well as the dynamical properties of a gel (Figure 2B). In the case of lower boiling point solvents, despite being entrapped in the 3D network, the solvent evaporates from the gel, leading to gel shrinkage. This may be useful in cases where rapid solvent elimination is needed,[68] whereas being a disadvantage for gel mechanical and thermal stability.[5] For applications where temperature stability is required, high-boiling-point organic liquids contribute to gel stability.[69] In the case of low temperatures, the freezing point of the solvent is a key factor to be considered. Liquid phase solidification leads to changes in visco-elastic parameters and negatively affects ionic conductivity[53,70,71] and is common for conductive hydrogels, limiting their tolerance to temperature and pressure changes. On the contrary, conductive organogels exhibit great potential in maintaining functionality in extreme conditions.[5,50,51,53,55,72,73] A commonly used approach to achieving environmentally stable conductive gels is the use of a binary water–organic liquid phase.[9] Hydrogen bond disruption by polar organic molecules in water–organic solvent mixtures results in a higher tolerance to freezing and evaporation compared with pure water. For instance, water–dimethyl sulfoxide (DMSO) binary solvent systems can have freezing points as low as −140 °C. Owing to the functional role of organic solvents, we refer to gels possessing such solvent mixtures as organogels, despite some water content. Rong et al. introduced supramolecular organohydrogels based on poly(3,4-ethylene-dioxythiophene):polystyrene sulfonate (PEDOT:PSS) self-assembly.[52] The use of a binary water–ethylene glycol (EG) solvent system produced a gel that retained mechanical stability

**ADVANCED
SCIENCE NEWS**
www.advancedsciencenews.com

**ADVANCED
FUNCTIONAL
MATERIALS**
www.afm-journal.de
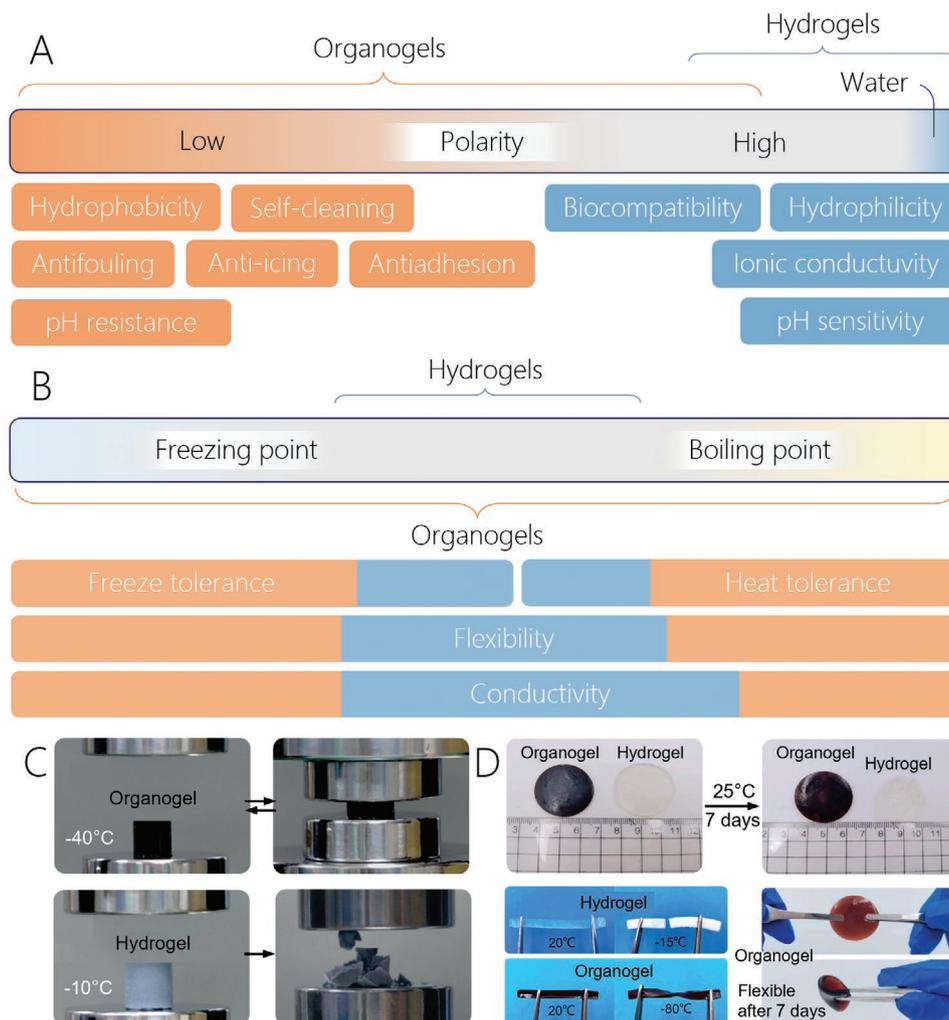
Figure 2. The role of solvents in organogels. A) Typical applications of gels with different polarity of the media. B) Thermal stability of the gels. C) Anti-freezing, conductive self-healing organogels with stable strain sensitivity at subzero temperatures. Reversible compression test at −40 °C (top, organogel swollen in a water–ethylene glycol binary solvent), at −10 °C (down, hydrogel). Reproduced with permission.[52] Copyright 2017, Wiley-VCH. D) Comparison of PVA-LN organogels and PVA hydrogels after storing for 7 days at 25 °C and 50% humidity. Reproduced with permission.[5] Copyright 2022, Elsevier.

at −40 °C (Figure 2C). Feng et al. introduced poly(vinyl alcohol)-lignin (PVA-LN) organogel with a water–DMSO mixture as a solvent. The formation of strong hydrogen bonds between water and DMSO molecules disrupted the hydrogen bond structure of water, preventing crystallization even at −80 °C (Figure 2D). The binary solvent system also provided for 88% weight retention under the ambient environment. PVA-LN organogel retained the mechanical properties after storing at 25 °C for 7 days, in contrast to PVA-LN hydrogel, which eventually shrank and became fragile.[5] The low vapor pressure of polar organic solvents, such as EG, ethylene carbonate, or propylene carbonate provides for weight retention at ambient and extreme conditions. Lee et al. demonstrated the synthesis of the PEDOT:PSS acrylamide organogel with EG as a solvent. The corresponding organogel retained 60% of EG after 22 h in vacuum (0.013 atm).[4]

An analysis of recently reported studies about organogels revealed several distinctive properties that make them prom-

ising candidates to supplement hydrogels in various fields of materials sciences (**Figure 3**). These properties include: 1) the ability to absorb and release substances having various polarities, 2) stimuli-responsive behavior, 3) ionic conductivity, 4) thermal and environmental stability, and 5) special wettability. In many cases, a combination of these properties can be achieved within one organogel, resulting in superior multifunctional behavior (examples are given in **Table 1**). For example, a significant advantage of organogel-based wearable electronics comes from the combination of solvent-induced conductivity and enforced mechanical stability (up to self-healing properties) due to the chemical and physical connection of a solvent with a polymer network. Another significant advantage resulting from the high boiling point and tolerance of the solvent is long-term stability under environmental and extreme conditions, which is highly desirable for many gel-based devices. On the contrary, chemically active solvents enhance the extraction, sensing, and
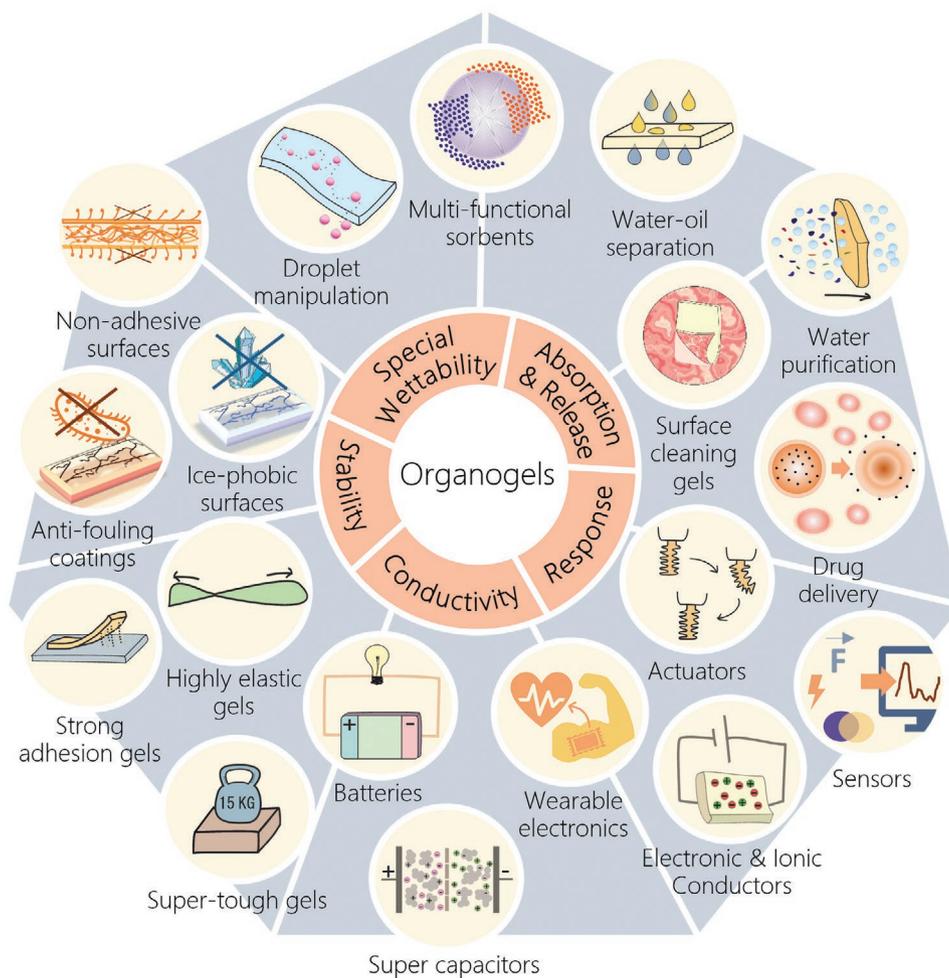
Figure 3. Distinctive features and related applications of organogels. Defined distinctive properties include: 1) mechanical, thermal, and environmental stability, 2) special wettability, 3) absorption & release of liquids and substances, 4) response to various stimuli, and 5) electronic and ionic conductivity. Real applications usually require combinations of several features, e.g., conductivity, responsiveness, and stability for wearable electronics.

absorbing properties of the gel.[74,75] Based on the proposed classification (Figure 3), we will discuss in detail recently developed strategies for achieving advanced functional organogels.

3.1. Durability of Organogels

In their swollen state, gels usually become less flexible, less tough, and more brittle owing to tension forces caused by large volumes of a confined solvent and reduced interactions between network chains.[78] However, the presence of strong interactions of the network with the swelling liquid may result in the reinforcement of the gel´s mechanical properties. Thus, Park et al. reported the use of non-polar aromatic interactions of polystyrene networks swollen in hexyl, octyl, and dodecyl benzene (**Figure** 4A).[79] Dynamic aromatic interactions enhanced stretchability ≈11 times and enabled a high photoluminescence efficiency of ≈99.8% throughout the volume of the gel while bending, folding, stretching, and twisting (compared with non-swollen polystyrene networks).

Dynamic non-covalent interactions between a network and a solvent can lead to self-healing effects and recovery of elastic properties. Guo et al. recently reported that amidoximated polyacrylonitrile-based gel swollen in *N, N*-dimethylformamide (DMF) showed 80 times higher stretchability with a strain of ≈800% compared to pure PAN (Figure 4B).[80] Such polymer plasticization is attributed to hydrogen bond formation among the network amidoxime groups and between the network and DMF molecules. Moreover, after re-dissolving gel fragments in DMF, owing to the reversibility of physical entanglement and dynamic hydrogen bonds of polymer chains, the amidoximated polyacrylonitrile gel could fully restore its original mechanical strength. Another example of an organogel demonstrating self-healing and recovery through the dynamic interaction of the network and the swelling solvent is DNA-based dynamic organogel in DMSO, as demonstrated by Meng et al.[81]

Another important property that is dependent on the choice of solvents in organogels is adhesion. In organogels, as opposed to hydrogels, various solvents can be used, including those possessing stronger, multivalent interactions with surfaces.

Table 1. Applications of organogels with a combination of advanced properties.

Gel composition	Combination of properties	Application
Poly(vinyl alcohol)-lignin nanoparticle organogel with a binary DMSO-H_2O solvent	The ionic conductivity, strain sensitivity, mechanical and thermal stability	Wearable electronics with enhanced flexibility, extreme freezing tolerance and anti-dehydration ability[5]
4-Acryloylmorpholine-based cross-linked 3D network; sodium perchlorate's propylene carbonate solution as conductive media	Ionic conductivity, strain sensitivity, mechanical and thermal stability, good adhesion to PDMS	Electrodes with good adhesion to triboelectric elastomers for the construction of triboelectric nanogenerators[55]
3-trimethoxysilylmethacrylate-capped fumed silica-based 3D network, choline chloride's propylene glycol or PEG200 solution as conductive media;	Ionic conductivity, strain sensitivity, mechanical and thermal stability	Mechanically durable strain-sensitive organogel conductors for sensorized tissue analogs[49]
Poly(vinylidene fluoride-co-hexafluoropropylene) (MW ≈ 455000) as a gelator; 1-ethyl-3-methylimidazolium tetrafluoroborate solution in acetone, acetonitrile, methyl formate, and propylene carbonate mixture as conductive media	Ionic conductivity, strain sensitivity, mechanical and thermal stability	Organogel-electrolyte-based supercapacitors for low-temperature energy storage[53]
4-acryloyl morpholine-based cross-linked 3D network; bistrifluoromethanesulfonimide lithium solution in propylene carbonate as conductive media	Ionic conductivity, strain sensitivity, mechanical and thermal stability	Organogel ionic conductors as strain sensors or as electrolytes for lithium-ion batteries[50]
Poly(vinyl alcohol) and poly(3,4-ethylenedioxythiophene):polystyrene sulfonate as conductive gelators; ethylene glycol–water binary solvent	Strain-sensitive electronic conductivity, mechanical and thermal stability	Anti-freezing organogel conductors for strain sensors[52]
Poly(3,4-ethylenedioxythio- phene):polystyrene sulfonate as conductive gelator; acrylamide-based cross-linked 3D network;	Strain-independent electronic conductivity, nonionic media, mechanical and thermal stability	Organogel electronic conductors with predominant electronic conductivity[4]
Dodecyl methacrylate and styrene-based cross-linked 3D network; Fe_3O_4 as nanomagnetic particles	Absorption of hydrophobic solvents and oils, magnetic response	Magnetic sorbents for oil spill removal[76]
N-Lauroyl-L-alanine methyl ester as gelator; oleic acid modified Fe_3O_4 nanoparticles as magnetic component; peanut oil as a solvent	Oleophilicity, thermo-, chemo-, magnetic responsive	Micro-organogel carrier for targeted hydrophobic drug delivery[77]
PDMS-based 3D network; trimethylsilane-modified carbon black nanoparticles as a photothermal-responsive material; paraffin wax (melting point 54–56 °C) as a solvent	Hydrophobicity, photothermal responsiveness, anti-icing, corrosion protection	Organogel coatings for programmable liquid adhesion and transfer; Droplet manipulation[57]
Double 3D network containing poly(1-butyl-3-vinyl imidazolium fluoborate) (PIL-BF4) cross-linked with benzene tetracarboxylic acid (BTCA); 1-propyl-3-methylimidazolium fluoborate as solvent	Ionic conductivity, mechanical and thermal stability, strain sensitivity, transparency, nonflammability	promising applications in various electrical devices, including flexible sensors, energy storage, and electronic skin[24]

* PDMS – poly(dimethylsiloxane).

Therefore, these solvents can give rise to adhesive properties of produced organogels. For example, Ge et al. reported that double-solvent-based glycerol–water organogels showed improved adhesion to wood and other surfaces compared with hydrogels having the same composition (without glycerol).[82] The adhesive effect of glycerol was attributed to the formation of multiple hydrogen bonds with chemical groups on the surface, as well as the penetration of the solvent into the microstructure of the surfaces, causing an interlocking bonding effect. Adhesive gels are important for soft composite electronics, e.g., in wearable applications, such as flexible skin sensors for rapid continuous health monitoring. A recent successful approach to creating both durable and sensitive skin sensors is the use of strain sensors comprising a dielectric organogel layer sandwiched between two ionically conductive hydrogel layers. Mo et al. presented a stable mechanically tough gel sensor with an adhesive organogel-based dielectric layer. Ethylene glycol (the liquid phase of the organogel) behaved as both a plasticizer and cross-linking agent for the double-polymer network

via hydrogen bonds formed both with the adjacent hydroxyl of polyacrylic acid (PAA) and poly(vinyl alcohol) (PVA) chains (Figure 4C).[56] Further, it acted as a source of dynamic hydrogen bonding for dissipating the stress-energy (supported by complexation with borate bonds). This organogel demonstrated strong adhesion to various surfaces, including rubber, glass, metal, and poly(methyl methacrylate) (PMMA), and served as a dielectric layer. The adhesion strength of the organogel toward the hydrogel sensing layer was as high as 28.4 kPa. Other reported ethylene glycol-based organogels showed improved anti-freezing and anti-heat stability,[83] conductivity,[9,84] stretchability, and stress retention.[85,86] Furthermore, poly(ethylene glycol)-based gels (PEGgels) have recently shown great potential as highly conductive, tough, stretchable, and self-healing organogels.[27] Other examples of reported dielectric organogels include propylene carbonate : ethylene carbonate-based dielectric gels[87] and 4-cyano-4'-pentylbiphenyl-based photoelastic organogels.[88]

Apart from mechanical toughness, stretchability, or adhesion ability, gel durability is also assessed by the robustness to

**ADVANCED
SCIENCE NEWS**

www.advancedsciencenews.com

**ADVANCED
FUNCTIONAL
MATERIALS**

www.afm-journal.de
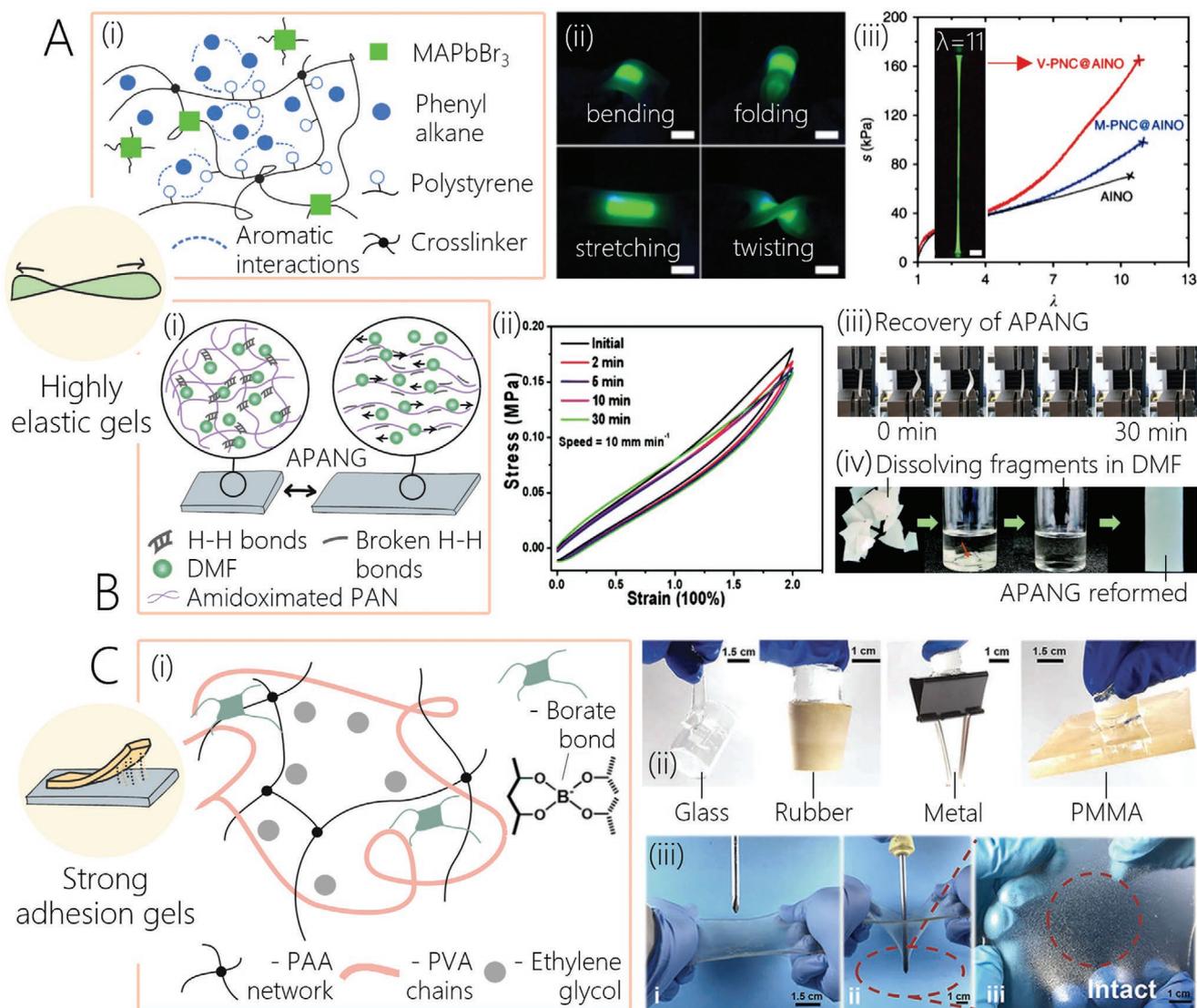
Figure 4. Mechanically durable organogels. A) Highly soft and transparent polystyrene-based organogel perovskite emitters i) show stable fluorescence while stretching or bending, and improved stretchability ii,iii) owing to strong aromatic interactions between the polymer network and alkylbenzenes as swelling solvents. Reproduced with permission.[79] Copyright 2020, Springer Nature. B) Because of dynamic H−H bonds formed with DMF i), amidoximated polyacrylonitrile-based gel exhibits self-recovering ii,iii) and full recyclability iv). Reproduced with permission.[80] Copyright 2020, Royal Society of Chemistry. C) PVA/PAA-borax organogel formed by entangled PAA and PVA networks with embedded ethylene glycol and borate bonds i) show strong adhesion to various materials and ii) high mechanical durability iii). Reproduced with permission.[56] Copyright 2021, Wiley-VCH.

various negative environmental exposures, which is critical for real applications. For hydrogels, there remain many limitations in this regard, such as biofouling, freezing, and formation of ice crystals on the surface, sensitivity to acidic pH and polarity, etc. Organogels have already been proven to show better performance in harsh environmental conditions. For instance, the use of binary DMSO–water systems was shown to both maintain high sensitivity for gel strain sensors and improve their temperature resistance down to –80 °C owing to the lowering of the freezing point of the liquid phase.[5] The same effect of adding anti-freezing and anti-drying properties to a gel by adding an organic solvent has been reported for glycerol–water organogels.[82] The choice of a non-volatile non-polar solvent could enable robustness of the gel to air and water exposure,

humidity, acid, base, heat, and light, as demonstrated for alkylbenzene-based organogels.[79]

Another negative effect of environmental exposure that significantly reduces material durability is biofouling. Owing to the slippery lubrication layer formed by oils on the gel's surface, organogels could be applied as protective coatings to inhibit bacterial settlement, as demonstrated for organogels based on nature-derived fatty oils,[89] silicon oils,[60] and paraffin oils.[15] Furthermore, certain solvents, such as 2-phenylethanol,[90] N-methyl pyrrolidone (NMP), dimethyl sulfoxide (DMSO),[91] and DMF[92] can function as active antimicrobial agents in organogels.

Durability is a key feature that boosts the performance of gel-based devices, such as sensors and flexible electronics.

2301421 (7 of 19)

**ADVANCED
SCIENCE NEWS**

www.advancedsciencenews.com

**ADVANCED
FUNCTIONAL
MATERIALS**

www.afm-journal.de

Organogels exhibit great potential as mechanically and environmentally durable gels whose durability is largely attributed to the properties of swelling media, where various organic solvents have been shown to 1) broaden gels' functionality at low and high temperatures, 2) improve flexibility and stretchability owing to specific strong interactions with the polymer network, 3) enable self-healing by the mobile restoration of the broken bonds, 4) improve adhesion to various surfaces, 5) enable gels' tolerance against harsh environmental exposure, such as bacterial activity, humidity, and acidic or basic pH.

3.2. Organogel-Based Sensors

Organogels have been used to develop sensors where organic solvents' properties participate in supporting, varying, and enhancing sensing performance. For example, contrary to water in hydrogels, organic solvents have much larger versatility in polarity, and organogels can thus be loaded with small organic molecules carrying detection or sensing properties. Liu et al. presented an ionic polyacrylamide (PAAm)-based organogel acting as a sensitive electrochromic sensor (**Figure 5**A,i).[93] To
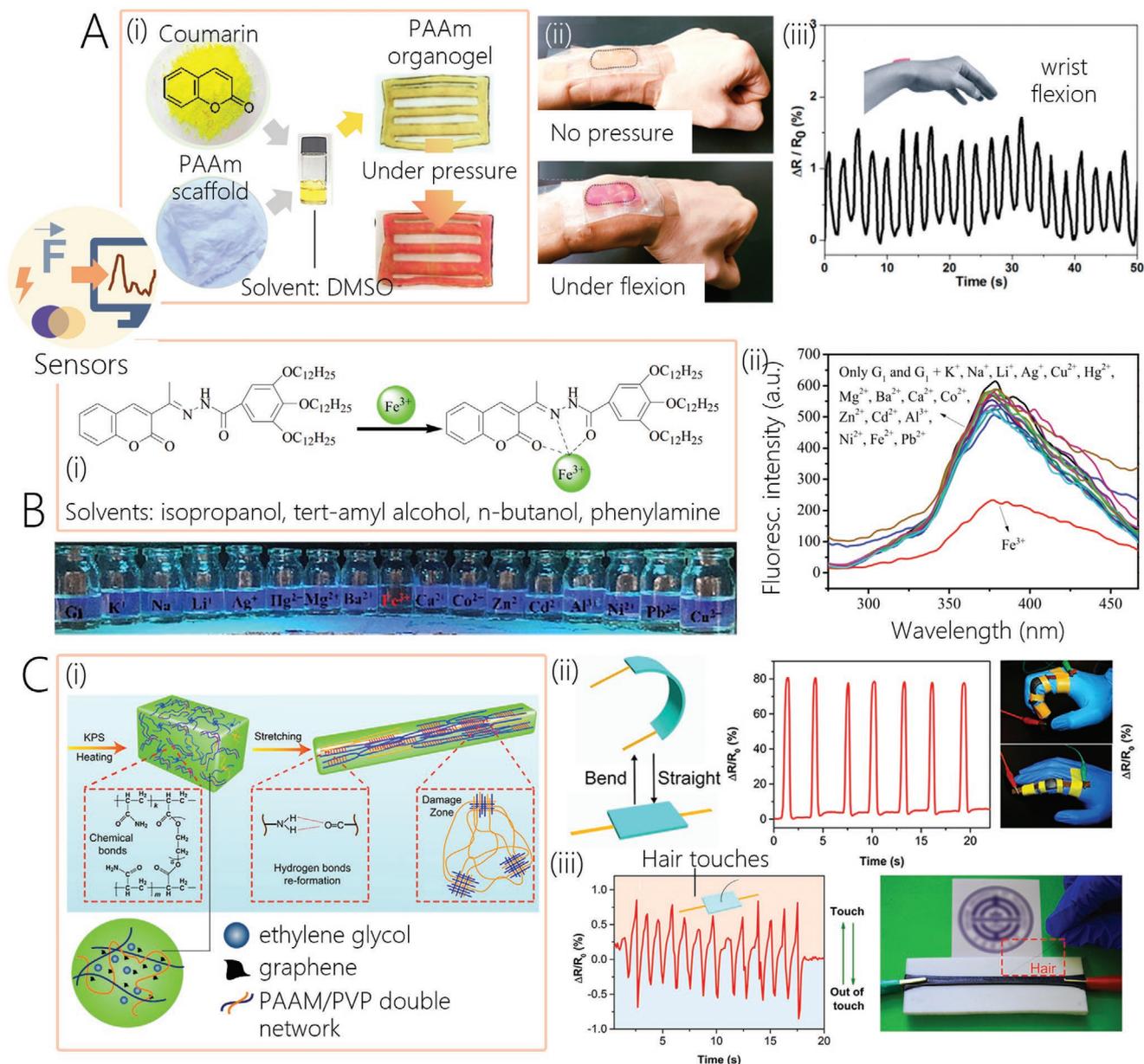
Figure 5. Organogel-based sensors. A) Bioinspired ionic polyacrylamide PAAm-based organogel in DMSO with coumarin i) works by the electrochromic response ii) and can be used as a wearable pressure sensor iii). Reproduced with permission.[93] Copyright 2020, American Chemical Society. B) Coumarin-derived acylhydrazone Schiff-base fluorescent organogel i) for the selective and sensitive detection of Fe^{3+} ions by the drop of fluorescence intensity ii,iii). Reproduced with permission.[35] Copyright 2020, Elsevier. C) Highly stable double-network-based ethylene glycol (EG) organogel i) shows the reliable detection of joint movements, such as finger bending ii) and extreme sensitivity, as demonstrated by the detection of hair touches after stretching 2.5 times iii). Reproduced with permission.[86] Copyright 2018, American Chemical Society.

**ADVANCED
SCIENCE NEWS**

www.advancedsciencenews.com

**ADVANCED
FUNCTIONAL
MATERIALS**

www.afm-journal.de

do this, coumarin, which is an organic electrochromic compound, was dissolved in DMSO and uniformly distributed via swelling within a PAAm network, resulting in a highly (over 1600%) stretchable electrochromic film. The organogel could detect localized mechanical stimuli by a fast and reversible color change and was applied as a wearable sensor for the electrical monitoring of small motions, such as wrist or finger flexion.

For selective chemical sensing, low-molecular-weight gels have been extensively studied and used.[68] The emission or absorption spectrum of such structures can quench noticeably upon exposure to vapors or ions, making them promising candidates for applications in air pollution and aqueous system monitoring and control. Recent studies have shown that organic solvents have good advantages for low- and supramolecular-weight chemosensors. In the presence of organic solvents, various gelators are able to form stable organogels, metallogels, and xerogels owing to the self-assembling of π-conjugated networks, as well as hydrogen and van der Waals forces between the moieties and the solvent.[94,95] However, matching these conditions requires the testing of a wide range of solvents with each supramolecular network to describe its functionality, as well as stability conditions and limitations. As an example, Yang et al. reported that a coumarin-based Schiff-base gelator forms low-molecular-weight organogel networks in isopropanol, *tert*-amyl alcohol, *n*-butanol, and phenylamine, and acts as an ion detector (Figure 5B).[35] The obtained organogels show stable fluorescence owing to the electrochemical properties of the hydrophobic coumarin-based gelator. Such a gelator is unable to form a gel and is therefore used in aqueous solutions because of its hydrophobicity. Furthermore, it was demonstrated that using isopropanol as the solvent caused a reversible organogel phase transition in response to heating, irradiation, and vigorous agitation, which illustrates how the use of a certain organic solvent can lead to the unique behavior of the same organogel.

Solvent-enhanced fluorescence behavior has been also demonstrated by Zhang et al. on organogels based on triphenylamine-substituted acylhydrazone derivative.[95] The organogels showed the polarity-affected solvatochromic effect with the emission color changing in solvents from nonpolar cyclohexane to polar dimethyl sulfoxide (DMSO). In other reported studies, organogel chemosensors demonstrated ultra-selective fluorescence detection of Pd^{2+} and $H_2PO_4^-$ ions,[96] volatile acid vapor sensing,[97] multi-responsive sensing and enrichment of Cu^{2+} and Co^{2+},[98] and the selective detection of F^- ions.[99]

Interestingly, EG was shown to preserve the protein-ligand binding and enzymatic activity of 2DPC bovine serum albumin protein, which is normally suppressed in aqueous media.[7] This property was used to create an organogel to sense ligands, such as ibuprofen and fatty acids. The shelf-life of the bovine-serum-albumin-based organogel was over nine times longer compared with the corresponding protein hydrogels owing to the low volatility of EG used as a solvent and its antimicrobial activity, preventing the digestion of the protein.

One of the key challenges for real-life applications of strain sensors is maintaining both ultra-high sensitivity and superior mechanical stability. Owing to the mechanical durability of some organogels and the compatibility of many organic solvents with both ionic and electronic conductivity, organogels

are often applied as strain sensors. Thus, Zhang et al. reported an organogel based on a polyacrylamide-polyvinylpyrrolidone double-network in EG with incorporated graphene, which showed high stretchability (up to 10500%). The resulting organogel was applied for the sensitive detection of motions from large (e.g., finger bending, ii) to mild (e.g., breathing) and subtle (touches of the single hair, in the stretched state, iii) (Figure 5C).[86] EG as a swelling agent enabled the uniform dispersion of graphene, and thus stable conductivity throughout the gel; it also contributed to the stretchability of the sensor.

Recent progress in the development and application of conductive organogels, including other examples of strain sensors, will be discussed in the following chapter.

3.3. Conductive Organogels

Gels are widely applied as quasi-solid-state electrolytes. This kind of electrolyte combines ionic conductivity (comparable to liquid electrolytes) with distinctive features of solid-state electrolytes, such as the absence of leakage, long-term stability, and mechanical strength.[73]

Most studies on the conductivity of gels have been attributed to ionic conductivity. This type of conductivity is provided by the gel media capable of ion transport, commonly aqueous ionic solutions.[72,100,101] Because of their aqueous nature, hydrogel ionic conductors have faced limitations in thermal and environmental stability, hindering their practical use.[48,50–55] On the contrary, organogels demonstrate better thermal and environmental stability combined with high ionic conductivity by using appropriate polar non-volatile liquids[50] or ionic liquids[102] as solvents.

Except for water, there are several other solvents capable of sustaining mobile ions and thus supporting ionic conductivity. For instance, propylene carbonate (PC) salt solutions are applied in lithium-ion batteries as electrolytes owing to their high ionic conductivity.[103] PC was used as swelling media in a 4-acryloylmorpholine-based organogel ionic conductor.[50] It demonstrates extraordinary performance, including high transparency (93%) in the visible region, extreme-temperature tolerance (from −100 to 100 °C), stretchability (elongation at break of 1219%), high ionic conductivity (7.9×10^{-4} S cm^{-1} at 25 °C), wide voltage window (5.0 V), and perfect chemical stability (**Figure 6**A).

The environmental and mechanical stability of gels is especially valuable in prolonging the lifespan of strain sensors for wearable electronics. Feng et al. introduced a polyvinyl alcohol-lignin-nanoparticle-based organogel strain sensor (PVA-LN) with anti-freezing (−80 °C) and anti-dehydration (88% weight retention after 7 days) properties. As a co-solvent, DMSO enabled the simultaneous dissolution of both the gelator (PVA) and lignin as a reinforcing agent. The binary solvent medium initiated lignin self-assembly into nanoparticles, which dispersed gradually throughout the final gel without special treatment. The lignin nanoparticles served as both the physical cross-linkers and reinforcing fillers, strengthening the resulting gel. Meanwhile, due to hydrogen bonds formed in binary water–DMSO mixture, the processes of crystallization and solvent evaporation were hindered simultaneously. The PVA-LN
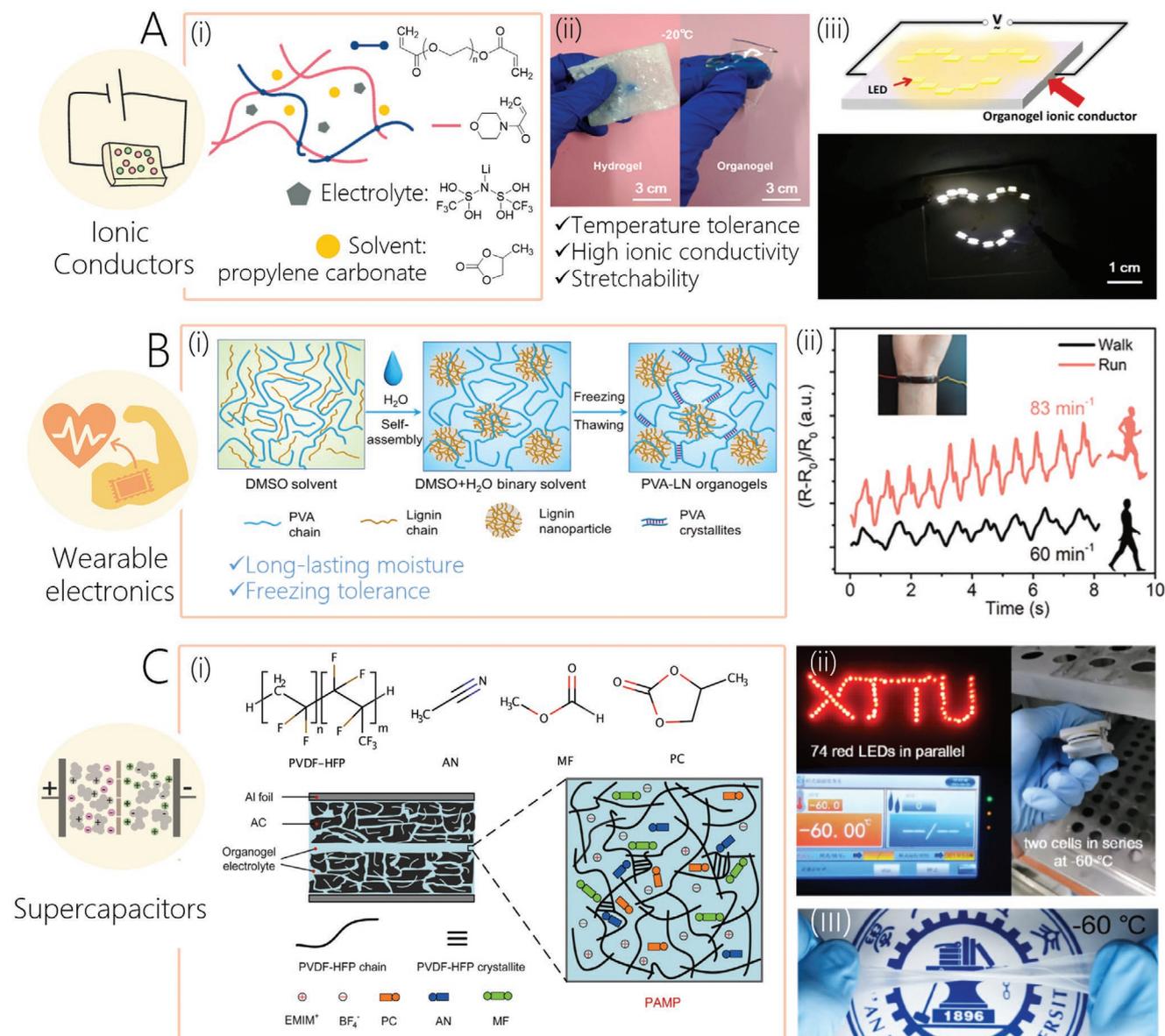
Figure 6. Applications of ionically conductive organogels. A) Stretchable organogel ionic conductors with extreme temperature tolerance. i) Molecular structures of reactants used for gel preparation. ii) Photographs of the hydrogel (left) and organogel (right) at −20 °C showing the water crystallization problem of hydrogels. iii) Schematic illustration (top) and photograph (bottom) of an organogel used as a flexible conductive substrate. Reproduced with permission.[50] Copyright 2019, American Chemical Society. B) Solvent-induced in situ self-assembled lignin nanoparticles to reinforce conductive nanocomposite organogels as anti-freezing and anti-dehydration flexible strain sensors. i) The schematic preparation process of anti-freezing conductive PVA-LN organogels. ii) Real-time monitoring of human motions by tracking the relative change of the resistance of PVA-LN strain sensors. Reproduced with permission.[5] Copyright 2022, Elsevier. C) High-performance solid-state supercapacitors based on conductive organogel electrolyte functional at low temperatures. i) Schematic diagram of an organogel polymer electrolyte applied to activated carbon (AC) electrode. ii) Two cells supply light-emitting diodes for >3 min at −60 °C. iii) Snapshot of the low-temperature mechanical performance of organogel-based electrolyte. Reproduced with permission.[53] Copyright 2022, Elsevier.

organogel retained stable mechanical flexibility and ionic conductivity over a wide temperature range (−80 °C to room temperature) (Figure 6B).[5]

Achieving stability at extremely low temperatures is critical for applications of energy storage devices. The use of hydrogel-based electrolytes is unsuitable under such conditions owing to the solidification of electrolytes, and thus a reduction of ionic conductivity <0 °C. The exchange of aqueous systems to

organic anti-freezing solvents can improve low-temperature stability, but may reduce the voltage window of conductive gels, limiting the energy density of supercapacitors. To combine low-temperature stability with the highest electrochemical performance, Zheng et al. optimized the solvent composition of the organogel ionic conductors.[53] The best performance was demonstrated by using a mixture of acetonitrile (AN), methyl formate (MF), and PC (AN : MF : PC as 2 : 2 : 1). The developed

gel electrolyte exhibited an ionic conductivity of 2.95 mS cm^{-1}, mechanical strain rate of 350%, and a voltage window of 0–4 V at –60 °C (Figure 6C).

Whereas ionic conductivity is determined by the presence of ions in the liquid phase of the gel, the second type – electronic conductivity – occurs through the gel network. Hydrogels commonly demonstrate mixed electrical conduction. Meanwhile, electrochemical stability in ionic conductors is restricted within the voltage window. Switching to pure electronic conductivity would prevent the unwanted irreversible electrochemical changes in the conductor even at high voltages. Deionized organic solvents, for example, ethylene glycol, demonstrate the ability to prevent ionic conduction in electronic conductors.[4] Lee et al. introduced PEDOT:PSS (poly(3,4-ethylene-dioxythiophene) : polystyrene sulfonate)–PAAm (polyacrylamide) organogel-based stretchable electronic conductors. The organogels were fabricated by swelling a PEDOT:PSS–PAAm network in EG. By replacing water with EG, the unwanted electrochemical reactions could be eliminated, hence hydrogen and oxygen generation did not occur with increasing voltage (for applied voltages ranging from –5 to +5 V). Furthermore, PEDOT:PSS–PAAm organogels have demonstrated stretchability exceeding 350% strain and long-term stability evaluated by weight retention at low pressures (60% weight after 22 h at a pressure of 0.013 atm).

In summary, replacing water with an organic solvent or a mixture of solvents resolves many limitations of gels in the field of electronics. Organogels infused with high-boiling-point polar liquids demonstrate greater mechanical and environmental stability, which enables faster practical use of wearable electronics and energy storage devices. At the same time, the variation of applied solvents provides versatility in terms of conductivity. Thus, deionized organic solvents can be used as inert media to prevent unwanted electrochemical reactions over a broad range of applied voltages in electronically conductive gels. Further studies of conductive organogels in terms of fabrication methods and applicability are required to fully reveal their potential in the field of conductive materials.

3.4. Special Wettability of Organogels

Because organogels are liquid-infused materials, their surface properties are determined by both the solid matrix and the nature of the solvent used to swell the matrix. Such liquid-infused surfaces often exhibit interesting properties, such as slippery effect, omniphobic properties,[104] anti-icing,[10,61] anti-(bio)fouling,[10,60] self-cleaning,[65] and anti-adhesion.[11,62]

Droplet manipulation ability is a complex feature demonstrated by organogel surfaces. Droplet-based systems have found applications in various fields of chemical and biomedical engineering, e.g., microfluidics. The ability to manipulate the flow of droplets can be achieved with the combination of surface tolerance, slipperiness, and stimuli-responsive wettability. In the case of organogels, these features can be provided by the properties of the solvent. For example, solvents show thermo-responsive behavior at their melting point. Based on that, the approach of thermally driven droplet manipulation via paraffin wax infused in a PDMS-cross-linked network was demonstrated

by Gao et al.[57] The water-guiding track could be created on the coating surface by melting the paraffin with locally applied heat. In the initial state, paraffin-infused PDMS demonstrated hydrophobic water-pinning behavior, whereas regions heated to a temperature above the melting point of paraffin became slippery. As a result, water droplets could slide across the surface with the specific pathway (**Figure 7**A). In addition, anti-icing properties were shown by covering the surface with ice and later illuminating it under sunlight. The ice layer began to melt and the generated water slid off the surface easily. As a result, no water was left on the coating surface after melting.

In addition to anti-icing properties, the stable liquid layer at the surface of organogels infused with oils can provide protection against crystallization-induced deposition of nonpolar substances. In the oil industry, the crystallization of wax at the surface of petroleum pipelines often leads to pipe clogging and is an important problem.[105] Yao et al. introduced organogels with significant self-replenishable anti-waxing properties.[61] In this case, the PDMS network was infused with crude oil, which resulted in the low adhesion of paraffin wax to the surface of the organogel. Owing to the much lower melting point of crude oil, it protected the organogel surface by enabling the easy removal of crystallized paraffin. When the liquid paraffin crystallized, nucleation started inside the diffusion layer where the liquid oil and the liquid paraffin had mixed (Figure 7B).

Liquids with a low friction coefficient and chemical tolerance of the solvent have been shown to protect organogel surfaces against various contaminants, including biofouling.[60,65] Contaminants deposed on the organogel can be easily removed with the top solvent layer. In addition, the liquid surface layer can be easily renewed to fully restore lubrication, including when the swelling agent is solidified.[62] Wang et al. reported a solid organogel material that combined anti-graffiti, anti-fouling, and anti-icing properties. Here, the oleophilicity of the PDMS network was used to swell it in alkanes having different carbon numbers (Figure 7C,i). They demonstrated that organogels swollen in solid alkane (carbon number higher than C17) showed much higher mechanical and environmental stability, did not contaminate nearby surfaces, and had a longer shelf life. The alkane sacrificial layer is regenerable within 24 h because of the stress gradient in the polymer network and excess of the alkane in the organogel bulk. Furthermore, the surface of organogel swollen in an alkane with carbon number 24 was tested to determine tolerance to various paints. Water-based paint, nitro lacquer paint, and alkyd paint formed films that could be easily peeled off the organogel, whereas aerosol paint could be wiped off with a cleaning cloth (Figure 7C,ii).

From the demonstrated examples of organogels with special wettability behavior, it is clear that applying various organic liquids to the same conventional polymer networks creates a variety of new functional materials. Thus, reconsidering the reported non-volatile-liquid-swollen organogels could reveal their functionality and new applications. It has been shown that hydrophobic thermally stable solvents not only contribute to gel thermal stability, but can also enable dynamic response, slipperiness, crystal phobicity, self-healing, contamination protection, and anti-fouling. Therefore, we propose the incorporation of organic solvents into conventional polymer networks as a potential route in this field of study.

ADVANCED
SCIENCE NEWS
www.advancedsciencenews.com

ADVANCED
FUNCTIONAL
MATERIALS
www.afm-journal.de
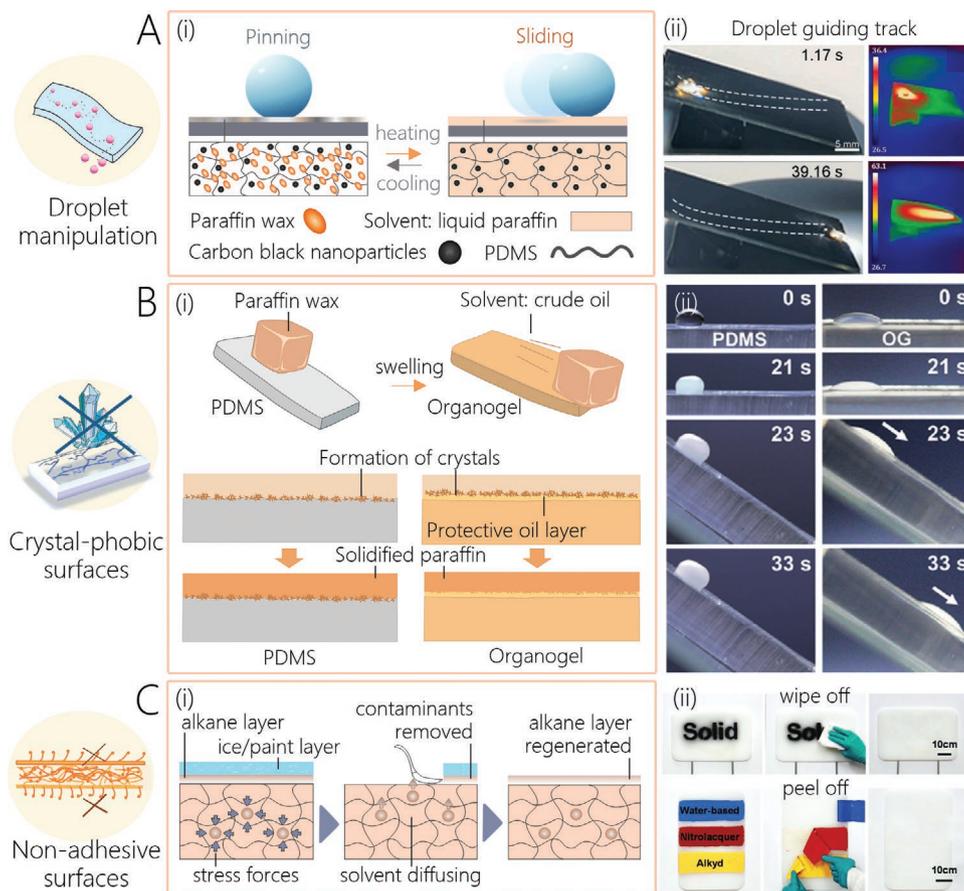
Figure 7. Organogel special wettability. A) A thermal-driven self-replenishing slippery coating. i) Paraffin wax layer on the organogel surface can be switched from a solid to a liquid phase by heating locally. ii) Schematic illustration of the thermo-programmable water motion. Reproduced with permission.[57] Copyright 2021, Elsevier. B) Self-replenishable crystal-phobic organogels. i) Schematic illustration of nucleation sites in crystallization process on the non-swollen PDMS and swollen organogel. ii) Adhesion of crystallized paraffin wax on PDMS (left) and organogel (right) surfaces. Reproduced with permission.[61] Copyright 2015, Wiley-VCH. C) Bioinspired organogels with regenerable sacrificial alkane surface layer. i) Regeneration of the alkane layer. ii) Aerosol paint (top) and different paints (down) can be easily removed from the organogel surface. Reproduced with permission.[62] Copyright 2017, Wiley-VCH.

3.5. Application of Absorption and Release

Soft polymeric networks act as sorbents in the process of swelling in organic solvents depending on their polarity. Pre-organogel networks have several advantages compared with conventional sorbents, such as clays, zeolites, silica,[106] activated carbon,[107] and natural fibers.[108] First, the mass uptake for gel sorbents is practically higher (up to 99 wt.%). Other advantages include the potential for reversible and selective sorption. For instance, non-polar 3D polymer networks have higher affinities toward organic contaminants than water.

An additional benefit of organogels is a wider range of polarity for absorbed substances and selective adsorption of non-polar substances. For example, dynamic covalent organogel networks show a high potential in the selective adsorption of aromatic compounds and separation of toluene from the azeotropic mixtures, combined with the ability to be recycled and re-polymerized.[109] As another example, the hydrophobic cross-linked polymer structure of PDMS is known to swell in organic liquids of various polarities, including methyl-*tret*-butyl ether (MTBE), dichloromethane, toluene, and DMSO. Bayraktaroglu

et al. performed a one-step synthesis of PDMS-based hydrophobic sorbents from tetraethyl orthosilicate (TEOS) and PDMS having various lengths (**Figure 8**A,i). The proposed low-cost synthetic approach did not require any activators, initiators, or catalysts. At the same time, the authors demonstrated the adjustability of the polymer composition. The sorbent produced from PDMS (1500 g mol^{-1}) and TEOS exhibited the best performance in terms of the absorption of organic liquids. The maximum absorbency values in dichloromethane (DCM), tetrahydrofuran (THF), methyl tertiary-butyl ether (MTBE), toluene, xylene, ethylbenzene, and benzene, were shown to be 2119, 1963, 1628, 1611, 1369, 1345, and 1293%, respectively. Moreover, the obtained sorbents had a much lower affinity to water (absorbency in water 40%), enabling the separation of floating organic liquids from the water surface (Figure 8A,ii). Stability tests showed that the PDMS sorbents retained structural integrity and sorption efficiency after being stored in gasoline for 300 days, and could be reused for at least 10 absorption–desorption cycles.[110]

Demirel et al. demonstrated a similar approach. They reported the synthesis of a pre-organogel network by
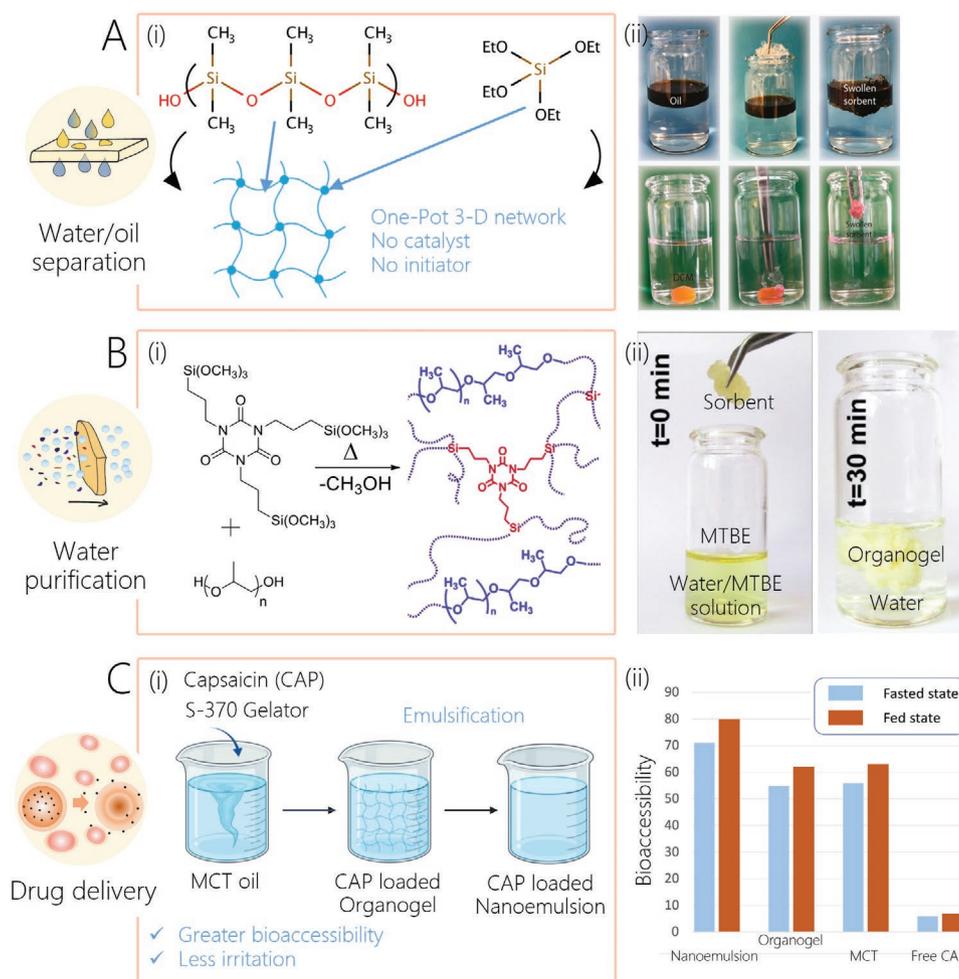
Figure 8. Applications of absorption/release. A) A highly reusable polydimethylsiloxane sorbent for oil/organic solvent clean-up from water. i) Preparation of PDMS sorbents and their expected final 3D structure. ii) Absorption of crude oil (top) and DCM (down) by the sorbent from water. Reproduced with permission.[110] Copyright 2021, Elsevier. B) Clean-up of oily liquids, fuels, and organic solvents from contaminated water fields using poly(propylene glycol)-based organogels. i) Preparation of PPG-ICS organogels and their expected final 3D structure. ii) Absorption of MTBE from water solution. Adding dry organogel to the mixture (left), 30 min later after addition of dry organogel (right). Reproduced with permission.[111] Copyright 2017, Elsevier. C) Development of organogel-derived capsaicin nanoemulsion with improved bioaccessibility and reduced gastric mucosa irritation. i) Schematic illustration of capsaicin- (CAP) loaded organogel nanoemulsion preparation. ii) Comparison of bioaccessibility of CAP for different forms of administration. Reproduced with permission.[117] Copyright 2016, American Chemical Society.

condensation reactions of polypropylene glycols (PPG)s with different molecular weights and tris[3-(trimethoxysilyl) propyl] isocyanurate (ICS) (Figure 8B,i).[111] The authors observed a proportional increase in sorption capacity with a higher molecular weight of PPG. It was also found that the degree of crosslinking decreased with the molecular weight of PPG. Thus, the increase in mass uptake with the molecular weight of PPG can be attributed to the increase in the plasticity of the network. The prepared organogels readily absorbed various organic liquids. The best performance was demonstrated by the sorbent obtained from PPG (4000 g mol^{-1}). The maximum absorbency in crude oil, gasoline, acetone, MTBE, toluene, benzene, THF, and DCM was found to be 233, 770, 686, 714, 1269, 1319, 1439, and 1872, respectively (Figure 8B,ii). The capacity of the synthesized organogels did not change in either pure organic liquids or organic liquid–water mixtures. This was associated with an extremely low affinity of the obtained

sorbents to water. PPG 4000-ICS sorbents were capable of removing MTBE both from the surface of water and the water/MTBE solution. MTBE and gasoline would be fully released after absorption within 2 h without the application of vacuum or heat. Reusability limits included at least 10 sorption–desorption cycles.

Supramolecular gelation represents a widely utilized approach toward the delivery of oleophilic bioactive compounds.[112,113] Nutritional oils and biocompatible organic solvents, such as soybean oil, coconut oil, medium chain triglyceride, glyceryl tributyrate, and glyceryl triacetate create a non-polar media, readily dissolve, and sustain water-insoluble substances. The oils and their solutions can be gelated by adding supramolecular agents. Organogel-based formulations facilitate oral,[114] intravenous,[115] and transdermal[116] routes of administration for hydrophobic medications, such as statins. This approach enables the controlled release of medication by

ADVANCED
SCIENCE NEWS

www.advancedsciencenews.com

ADVANCED
FUNCTIONAL
MATERIALS

www.afm-journal.de

tunable diffusion rate or sol–gel transition.[77] For instance, Lu et al. performed the synthesis of capsaicin-loaded organogel nanoemulsion.[117] The role of organogel was to increase the bioaccessibility of capsaicin relative to its pure form and to overcome irritation issues associated with its oral intake. Capsaicin-loaded organogel was prepared by adding sucrose stearate (S-370) as a gelator to a solution of capsaicin in medium-chain triacylglycerol (MCT). Tween-80 was used as an emulsifier to obtain oil-in-water (O/W) emulsion with organogel as the oil phase (Figure 8C,i). Ultrasonication treatment decreased the droplet sizes of the emulsion to 168 nm, enhancing the dissolution rate and bioaccessibility (Figure 8C,ii). CAP-loaded nanoemulsion demonstrated reduced rat gastric mucosa irritation. Thus, an organogel-based formulation provided a convenient oral route for capsaicin-based treatment.

The absorption ability is a bulk property of the gel that is essential for applications, such as drug delivery, water purification, and water-oil separation. The 3D network of hydrogels has an affinity to water and other polar substances, but it commonly fails to be infused with non-polar substances, such as oils or lipophilic drugs. On the contrary, the polarity of organogel media varies over a wide range, their full potential in drug delivery and oil spill liquidation has not yet been fully discovered.

3.6. Organogel Actuators

Actuators are materials that respond to changes of shape, volume, or complicated movements in response to certain stimuli. The main requirements for advanced actuators include material adaptability and the complexity of its responses to various stimuli, as well as a combination of flexibility and strength. Gels represent flexible, adaptable, and bio-inspired soft actuators.[118] Owing to the large number of compatible organic solvents and polymer networks, organogels can complement existing gel actuators in terms of their environmental tolerance, new types of responses (e.g., swelling in non-polar and shrinking in polar media), or extreme plasticity.

The functionality of the advanced soft actuator is determined by operational characteristics, such as diversity and precision of motion (rotary, linear), speed of response, as well as cyclability. For the advanced soft actuator, the smart design of the gel network can enable a combination of mechanical strength and stretchability with cyclability and self-recovery, whereas the incorporation of responsive fillers contributes to the fast and reliable actuation response. This approach was demonstrated by Meng et al., who fabricated organogels based on the self-assembly of DNA chains functionalized with cationic surfactant in an excess (up to 96%) of organic solvents, including polar (THF, DMSO, ethanol) and nonpolar ($CHCl_3$ and toluene) (**Figure 9**A,i).[81] The actuation was enabled by incorporating oleic acid-stabilized Fe_3O_4 nanoparticles (NPs) to the DNA-surfactant network. The abundance of the alkyl chains in the networks enabled the uniform distribution of functionalized magnetic NPs, which provided a quick and strong response to an external magnetic field (ii, iii). The highest mechanical performance was shown by DMSO-based gel, which demonstrated thermal responsiveness and fast self-healing, elasticity over

3×10^4%, outstanding stiffness (over 20 MPa), and toughness up to 18 MJ m^{-3} appearing as the current record-holder among DNA gels.

Owing to their nature, gels react to an excess of the solvent with swelling or shrinking, which itself is an actuating movement. However, it remains challenging to create a gel actuator with in situ swelling in response to the local absorption of the solvent without diffusion throughout the volume. Furthermore, it is challenging to make such shape-programmed movement reversible and continuous during the application of the liquid. A solution for such actuator was proposed by Wang et al. The elastic organogel films were prepared by solution-casting from a synthesized fluorinated epoxide monomer in dimethyl formamide, where the elasticity of the network was provided by a long-chain covalently bounded crosslinking fluoronetwork (CFN).[68] After depositing a volatile organic solvent (e.g., DCM) on the surface of the organogel, there was rapid local shape deformation of the film (Figure 9B,i). By depositing solvent droplets in different locations of the organogel film, different shapes will be formed and restored to the initial film after full evaporation. The shapes demonstrated excellent repeatability and unchanged response after >100 cycles of swelling–deswelling or upon heating to 70 °C. This robustness made the created gel applicable as a multiple-use chemical sensor.

Liquid crystal (LC) organogels can play a supporting and dynamic role in the creation of 4D LC actuators. Jin et al. fabricated LC organogel by UV polymerization, and further evaporated the solvent phase (DMF) during the uniaxial stretching of the organogel.[119] This created micropores in the gel network, which acted as physical crosslinks and fixed the desired shape (Figure 9C,i). Multiple shapes could be erased and reprogrammed from the same LCN on demand depending on the alignment of the organogel during the solvent evaporation. Such solvent-assisted programming orthogonal to the temperature-activated LC phase transition enabled the LCNs to be stably actuated between the temporary and permanent shapes. Owing to this shape memory effect, a series of 2D-kirigami objects were created switching reversibly to the 3D shapes in heating-cooling cycles (ii). The same principle was applied to 3D-printed bulk organogels that could be preserved in multiple deformed states during the solvent evaporation, and further act with the temperature-responsive shape change (iii). Thus, the combination of rapidly evaporating solvents (DCM, Toluene) and stretchable LC gel networks led to reprogrammable 4D shape morphing actuators.

With respect to actuation toward various types of external stimuli, the reported organogels include durable and temperature-tolerant DMF-based,[48] as well as water-resistant liquid-paraffin-based[120] electric actuators, UV-triggered contraction networks,[121] temperature- and swelling-driven dynamic covalent networks with shape memory,[122] and reversible light-controlled stiffening-softening actuators.[123] Organogel-based actuators have shown several advantages compared to hydrogel-based actuators, such as: 1) environmentally durable long-lasting performance supported by the protective role of apolar non-volatile solvents; 2) ability to perform fast and reversible swelling–deswelling actuation, including in situ and 4D-shaped swelling, when applying small volatile organic solvents.
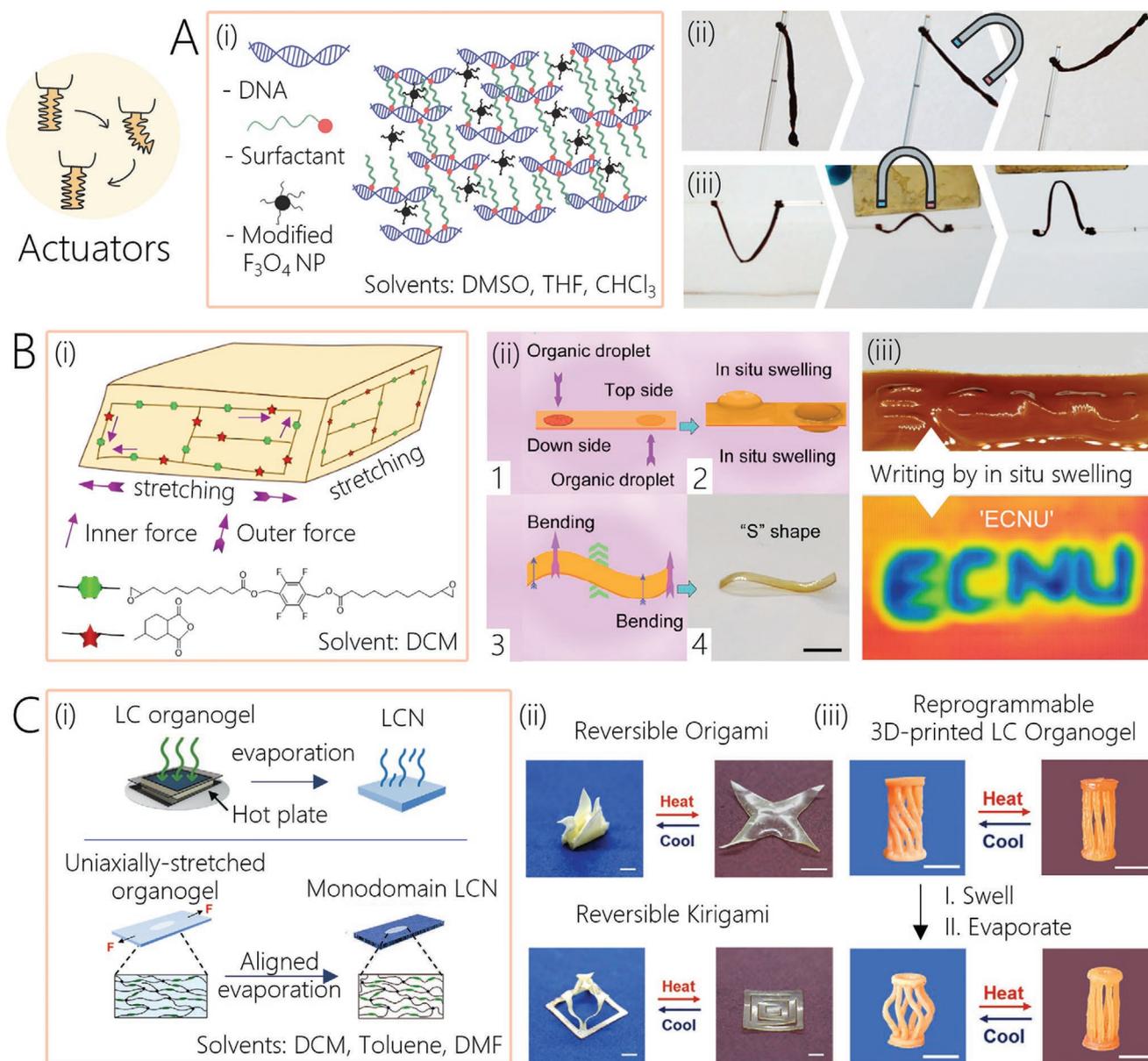
Figure 9. Organogel-based actuators. A) Magnetic field-responsive DNA-based liquid crystalline actuator with endowed Fe$_3$O$_4$ nanoparticles. Reproduced with permission.[81] Copyright 2021, Wiley-VCH. B) In situ swelling-induced reversible bending of organogel films. Solvent-induced swelling and free evaporation lock the deformed state. Reproduced with permission.[68] Copyright 2020, Elsevier. C) Evaporation-induced (re-)programming of liquid-crystalline-network (LCN) actuator combined with a kirigami–origami strategy or DLP 3D-printing. Reproduced with permission.[119] Copyright 2021, Wiley-VCH.

4. Perspectives and Conclusion

Organogels are fascinating materials comprising polymers or supramolecular networks filled with an organic liquid phase. The functional diversity of organogels and their vast, but untapped potential as a functional material for a wide range of applications are exhibited through the numerous conceivable combinations of known organic solvents with different suitable gelators. However, in contrast to the vast literature on hydrogels, there are fewer research articles about organogels. In this review, we covered major advances in organogel research and

the distinctive properties demonstrated by these, intending to highlight and promote organogels as an essential class of functional soft materials. Furthermore, we described progress in the application of organogels and highlighted the active role of solvents in organogel functionality.

The full potential of organogels can only be understood after addressing misconceptions about their composition and properties that limit their current development and applications. We proposed resolving these limitations by applying a broader definition of organogels that includes gels based on hydrophilic, polar solvents, and other functional organic liquids, as well as

hybrid organic solvent-water systems. Following this interpretation, we introduced organogels as a class of gels with a wider variety of properties than previously considered. Furthermore, we confirmed that expanding the scope of physical or chemical parameters of the swelling solvents enables organogels to overcome limitations known for hydrogels.

Reported organogels demonstrate various properties, ranging from conductive to dielectric, from adhesive to slippery, and from hydrophilic to hydrophobic. We highlighted specific features that enable organogels to perform better than hydrogels or show new functionalities. Such features include higher mechanical and environmental stability, a broader working temperature range, potential for ionic and electric conductivity, and affinity to lipophilic substances. In addition, the combinational structure of organogels allows precise adjustment of their properties with composition, thus providing an avenue for further study. At the same time, the current state of the field indicates the lack of both theoretical and experimental research on various organogel systems. As the most promising directions for the further development of organogels, we propose: 1) development of versatile and low-cost fabrication methods, 2) development of new organogelators for specifically targeted interactions with solvents, 3) implementation of organogels in fields currently dominated by hydrogels: implants, drug release and targeted delivery, biomedicine, and biomimicking materials, 4) combination of organogels with hydrogels in multifunctional materials with extreme differences in response behavior. Further, the possibility of incorporating organic solvents in gels made of various nature-derived organic molecules, such as proteins and lipids, and preserving their functionality should be further explored. However, owing to the preserving properties of many organic solvents that are known to form colloidal suspensions, organogels exhibit more potential for incorporating functional nanoparticles in the gel network.

Despite their promising properties, we observed limitations in the practical applications of organogels. The major issue that needs to be overcome for practical applications of organogels relates to their sustainability in terms of toxicity and recyclability. As possible solutions, we see the further development of organogels based on green solvents, as well as new strategies for dynamically degradable and reusable polymer networks. Another factor that limits the rapid growth of the field is the lack of computational investigation combined with empirical testing to describe the interactions between specific gelator systems and various organic liquids. Providing the theoretical basis for the gelation process and properties of the gel would contribute to the smart design of the gel network and successful incorporation of other components.

Considering all the evidence and prospects in this review, we believe that organogels are the broadest class of gels that can still reveal the full potential of soft liquid-infused multifunctional materials.

Conflict of Interest

The authors declare no conflict of interest.

[1] S. J. Chalk, *IUPAC, Compendium of Chemical Terminology (the "Gold Book")*, 2nd ed. (Eds: A. D. McNaught, A. Wilkinson), Blackwell Scientific Publications, Oxford **1997**.

[2] L. Zeng, X. Lin, P. Li, F.-Q. Liu, H. Guo, W.-H. Li, *Prog. Org. Coat.* **2021**, *159*, 106417.

[3] R. Eelkema, A. Pich, *Adv. Mater.* **2020**, *32*, 1906012.

[4] Y.-Y. Lee, H.-Y. Kang, S. H. Gwon, G. M. Choi, S.-M. Lim, J.-Y. Sun, Y.-C. Joo, *Adv. Mater.* **2016**, *28*, 1636.

[5] Y. Feng, J. Yu, D. Sun, W. Ren, C. Shao, R. Sun, *Chem. Eng. J.* **2022**, *433*, 133202.

[6] E. Larrañeta, S. Stewart, M. Ervine, R. Al-Kasasbeh, R. F. Donnelly, *J. Funct. Biomater.* **2018**, *9*, 13.

[7] N. L. Smith, A. E. Coukouma, D. C. Wilson, B. Ho, V. Gray, S. A. Asher, *ACS Appl. Mater. Interfaces* **2020**, *12*, 238.

[8] L. Li, Q. Wang, X. Liang, Z. Li, S. Guo, G. Sun, *Polym. Test.* **2021**, *99*, 107018.

[9] X. Su, H. Wang, Z. Tian, X. Duan, Z. Chai, Y. Feng, Y. Wang, Y. Fan, J. Huang, *ACS Appl. Mater. Interfaces* **2020**, *12*, 29757.

[10] C. Urata, H. Nagashima, B. D. Hatton, A. Hozumi, *ACS Appl. Mater. Interfaces* **2021**, *13*, 28925.

[11] J. Lv, X. Yao, Y. Zheng, J. Wang, L. Jiang, *Adv. Mater.* **2017**, *29*, 1703032.

[12] G. Chen, S. Liu, Z. Sun, S. Wen, T. Feng, Z. Yue, *Prog. Org. Coat.* **2020**, *144*, 105641.

[13] Y. Zhuo, J. Chen, S. Xiao, T. Li, F. Wang, J. He, Z. Zhang, *Mater. Horiz.* **2021**, *8*, 3266.

[14] Y. Wang, X. Yao, J. Chen, Z. He, J. Liu, Q. Li, J. Wang, L. Jiang, *Sci. China Mater.* **2015**, *58*, 559.

[15] B. Eslami, P. Irajizad, P. Jafari, M. Nazari, A. Masoudi, V. Kashyap, S. Stafslien, H. Ghasemi, *Soft Matter* **2019**, *15*, 6014.

[16] V. Tantishaiyakul, P. Ouiyangkul, M. Wajasat, T. Pawisat, N. Hirun, T. Sangfai, *Eur. J. Lipid Sci. Technol.* **2018**, *120*, 1800178.

[17] B. Chaulagain, A. Jain, A. Tiwari, A. Verma, S. K. Jain, *Artif. Cells, Nanomed., Biotechnol.* **2018**, *46*, 472.

[18] J. Jose, K. Gopalan, *Res. J. Pharm. Technol.* **2018**, *11*, 1242.

[19] S. S. Sagiri, B. Behera, R. R. Rafanan, C. Bhattacharya, K. Pal, I. Banerjee, D. Rousseau, *Soft Mater.* **2014**, *12*, 47.

[20] N. Siraj, M. A. Shabbir, T. Ahmad, A. Sajjad, M. R. Khan, M. I. Khan, M. S. Butt, *Int. J. Food Prop.* **2015**, *18*, 1973.

[21] K. F. Chaves, D. Barrera-Arellano, A. P. B. Ribeiro, *Food Res. Int.* **2018**, *105*, 863.

[22] A. Shakeel, U. Farooq, D. Gabriele, A. G. Marangoni, F. R. Lupi, *Food Hydrocolloids* **2021**, *111*, 106190.

[23] C. E. Ibarra Torres, T. E. Serrano Quezada, O. V. Kharissova, B. I. Kharisov, M. I. La Gómez de Fuente, *J. Environ. Chem. Eng.* **2021**, *9*, 104886.

**ADVANCED
SCIENCE NEWS**

www.advancedsciencenews.com

**ADVANCED
FUNCTIONAL
MATERIALS**

www.afm-journal.de

[24] Y. Ren, J. Guo, Z. Liu, Z. Sun, Y. Wu, L. Liu, F. Yan, *Sci. Adv.* **2019**, *5*, eaax0648.

[25] J. Rao, X. Wang, R. Yunis, V. Ranganathan, P. C. Howlett, D. R. MacFarlane, M. Forsyth, H. Zhu, *Electrochim. Acta* **2020**, *346*, 136224.

[26] Q. Li, Z. Liu, S. Zheng, W. Li, Y. Ren, L. Li, F. Yan, *ACS Appl. Mater. Interfaces* **2022**, *14*, 26068.

[27] Z. Wang, H. Cui, M. Liu, S. L. Grage, M. Hoffmann, E. Sedghamiz, W. Wenzel, P. A. Levkin, *Adv. Mater.* **2022**, *34*, 2107791.

[28] S. R. S. Veloso, P. M. T. Ferreira, J. A. Martins, P. J. G. Coutinho, E. M. S. Castanheira, *Pharmaceutics* **2018**, *10*, 145.

[29] E. Kumarasamy, I. M. Manning, L. B. Collins, O. Coronell, F. A. Leibfarth, *ACS Cent. Sci.* **2020**, *6*, 487.

[30] A. Martín-Illana, F. Notario-Pérez, R. Cazorla-Luna, R. Ruiz-Caro, M. C. Bonferoni, A. Tamayo, M. D. Veiga, *Drug Discovery Today* **2022**, *27*, 1008.

[31] J. M. Scheiger, S. Li, M. Brehm, A. Bartschat, P. Theato, P. A. Levkin, *Adv. Funct. Mater.* **2021**, *31*, 2105681.

[32] S. Hou, J. Zhang, B. Huang, X. Wang, P. Xing, *Colloids Surf. A* **2022**, *648*, 129267.

[33] M. J. Zohuriaan-Mehr, K. Kabiri, M. Kheirabadi, *J. Appl. Polym. Sci.* **2010**, *117*, 1127.

[34] M. Lescanne, A. Colin, O. Mondain-Monval, F. Fages, J.-L. Pozzo, *Langmuir* **2003**, *19*, 2013.

[35] Y.-S. Yang, C. Liang, C. Yang, Y.-P. Zhang, B.-X. Wang, J. Liu, *Spectrochim. Acta, Part A* **2020**, *237*, 118391.

[36] X. Ren, Z. Xie, H. Wu, W. Shi, Y. Zhang, *Colloids Surf. A* **2019**, *583*, 124003.

[37] S. Basak, S. Bhattacharya, A. Datta, A. Banerjee, *Chemistry* **2014**, *20*, 5721.

[38] G. Zhu, J. S. Dordick, *Chem. Mater.* **2006**, *18*, 5988.

[39] J. Hao, F. An, C. Lu, Y. Liu, *J. Macromol. Sci., Part A* **2019**, *56*, 1012.

[40] M. Fernández-García, M. Fernández-Sanz, E. L. Madruga, R. Cuervo-Rodriguez, V. Hernández-Gordo, M. C. Fernández-Monreal, *J. Polym. Sci., Part A: Polym. Chem.* **2000**, *38*, 60.

[41] K. Liang, R. A. Hutchinson, *Macromolecules* **2010**, *43*, 6311.

[42] I. Sideridou-Karayannidou, G. Seretoudi, *Polymer* **1997**, *38*, 4223.

[43] J. D. Mota-Morales, R. J. Sánchez-Leija, A. Carranza, J. A. Pojman, F. del Monte, G. Luna-Bárcenas, *Prog. Polym. Sci.* **2018**, *78*, 139.

[44] L. Li, J. M. Scheiger, T. Tronser, C. Long, K. Demir, C. L. Wilson, M. A. Kuzina, P. A. Levkin, *Adv. Funct. Mater.* **2019**, *29*, 1902906.

[45] H. Chang, C. Li, R. Huang, R. Su, W. Qi, Z. He, *J. Mater. Chem. B* **2019**, *7*, 2899.

[46] P. Eslami, F. Rossi, S. Fedeli, *Pharmaceutics* **2019**, *11*, 71.

[47] A. Hendi, M. Umair Hassan, M. Elsherif, B. Alqattan, S. Park, A. K. Yetisen, H. Butt, *Int. J. Nanomed.* **2020**, *15*, 3887.

[48] Y. Zhang, Y. Zhao, Z. Peng, B. Yao, Y. Alsaid, M. Hua, D. Wu, Y. Qiu, Q. Pei, X. Zhu, Z. He, X. He, *ACS Mater. Lett.* **2021**, *3*, 1477.

[49] M. R. Crump, S. L. Bidinger, F. J. Pavinatto, A. T. Gong, R. M. Sweet, J. D. MacKenzie, *npj Flexible Electron.* **2021**, *5*, 7.

[50] Y. Gao, L. Shi, S. Lu, T. Zhu, X. Da, Y. Li, H. Bu, G. Gao, S. Ding, *Chem. Mater.* **2019**, *31*, 3257.

[51] W. Li, J. Zhang, J. Niu, X. Jin, X. Qian, C. Xiao, W. Wang, *Nano Energy* **2022**, *99*, 107359.

[52] Q. Rong, W. Lei, L. Chen, Y. Yin, J. Zhou, M. Liu, *Angew. Chem., Int. Ed. Engl.* **2017**, *56*, 14159.

[53] Q. Zheng, X. Li, Q. Yang, C. Li, G. Liu, Y. Wang, P. Sun, H. Tian, C. Wang, X. Chen, J. Shao, *J. Power Sources* **2022**, *524*, 231102.

[54] Z. Luo, Y. Wang, B. Kou, C. Liu, W. Zhang, L. Chen, *Energy Storage Mater.* **2021**, *38*, 9.

[55] T. Jing, B. Xu, Y. Yang, M. Li, Y. Gao, *Nano Energy* **2020**, *78*, 105373.

[56] F. Mo, Y. Huang, Q. Li, Z. Wang, R. Jiang, W. Gai, C. Zhi, *Adv. Funct. Mater.* **2021**, *31*, 2010830.

[57] Z. Gao, T. Xu, X. Miao, J. Lu, X. Zhu, Y. Song, G. Ren, Y. Jia, X. Li, *Surf. Interfaces* **2021**, *24*, 101022.

[58] X. Yao, J. Ju, S. Yang, J. Wang, L. Jiang, *Adv. Mater.* **2014**, *26*, 1895.

[59] P. Zhang, H. Liu, J. Meng, G. Yang, X. Liu, S. Wang, L. Jiang, *Adv. Mater.* **2014**, *26*, 3131.

[60] H. Zhang, Y. Liang, P. Wang, D. Zhang, *Prog. Org. Coat.* **2019**, *132*, 132.

[61] X. Yao, S. Wu, L. Chen, J. Ju, Z. Gu, M. Liu, J. Wang, L. Jiang, *Angew. Chem., Int. Ed. Engl.* **2015**, *54*, 8975.

[62] Y. Wang, X. Yao, S. Wu, Q. Li, J. Lv, J. Wang, L. Jiang, *Adv. Mater.* **2017**, *29*, 1700865.

[63] J. Sarma, L. Zhang, Z. Guo, X. Dai, *Chem. Eng. J.* **2022**, *431*, 133475.

[64] X. Wang, J. Huang, Z. Guo, *Adv. Colloid Interface Sci.* **2022**, *301*, 102602.

[65] H. Liu, P. Zhang, M. Liu, S. Wang, L. Jiang, *Adv. Mater.* **2013**, *25*, 4477.

[66] Z. F. Gao, R. Liu, J. Wang, J. Dai, W.-H. Huang, M. Liu, S. Wang, F. Xia, L. Jiang, *Chem* **2018**, *4*, 2929.

[67] S. Kim, D. Yoo, S. Lee, J. Kim, *Adv. Mater. Interfaces* **2022**, *9*, 2200797.

[68] F. Wang, B. Chen, L. Wu, Q. Zhao, L. Zhang, *Cell Rep. Phys. Sci.* **2020**, *1*, 100011.

[69] M. Wang, P. Zhang, M. Shamsi, J. L. Thelen, W. Qian, V. K. Truong, J. Ma, J. Hu, M. D. Dickey, *Nat. Mater.* **2022**, *21*, 359.

[70] F. Mo, G. Liang, Q. Meng, Z. Liu, H. Li, J. Fan, C. Zhi, *Energy Environ. Sci.* **2019**, *12*, 706.

[71] K. Wang, J. Wang, L. Li, L. Xu, N. Feng, Y. Wang, X. Fei, J. Tian, Y. Li, *Chem. Eng. J.* **2019**, *372*, 216.

[72] Y.-S. Kim, Y.-G. Cho, D. Odkhuu, N. Park, H.-K. Song, *Sci. Rep.* **2013**, *3*, 1917.

[73] M. M. Amaral, R. Venâncio, A. C. Peterlevitz, H. Zanin, *J. Energy Chem.* **2022**, *67*, 697.

[74] Y. Jia, G. Sciutto, R. Mazzeo, C. Samorì, M. L. Focarete, S. Prati, C. Gualandi, *ACS Appl. Mater. Interfaces* **2020**, *12*, 39620.

[75] Y. Jia, G. Sciutto, A. Botteon, C. Conti, M. L. Focarete, C. Gualandi, C. Samorì, S. Prati, R. Mazzeo, *J. Cult. Herit.* **2021**, *51*, 138.

[76] H. Ghasemzadeh, M. Dargahi, G. Eyvazi, B. V. Farahani, *Chin. J. Polym. Sci.* **2019**, *37*, 444.

[77] J. Dong, X. Du, Y. Zhang, T. Zhuang, X. Cui, Z. Li, *Colloids Surf., B* **2021**, *208*, 112109.

[78] T. Sakai, T. Fujiyabu, in *Physics of Polymer Gels* (Ed: T. Sakai), Wiley-VCH, Weinheim **2020**, p. 241.

[79] J.-M. Park, J. Park, Y.-H. Kim, H. Zhou, Y. Lee, S. H. Jo, J. Ma, T.-W. Lee, J.-Y. Sun, *Nat. Commun.* **2020**, *11*, 4638.

[80] X. Guo, C. Zhang, L. Shi, Q. Zhang, H. Zhu, *J. Mater. Chem. A* **2020**, *8*, 20346.

[81] Z. Meng, Q. Liu, Y. Zhang, J. Sun, C. Yang, H. Li, M. Loznik, R. Göstl, D. Chen, F. Wang, N. A. Clark, H. Zhang, A. Herrmann, K. Liu, *Adv. Mater.* **2022**, *34*, 2106208.

[82] J. Ge, S. Dai, X. Dong, M. Li, Y. Xu, Y. Jiang, N. Yuan, J. Ding, *New J. Chem.* **2022**, *46*, 4334.

[83] Y. Quan, M. Chen, W. Zhou, Q. Tian, J. Chen, *Front. Chem.* **2020**, *8*, 603.

[84] C. Sun, C. Hou, H. Zhang, Y. Li, Q. Zhang, H. Wang, *APL Mater.* **2021**, *9*, 011101.

[85] J. W. Zhang, D. D. Dong, X. Y. Guan, E. M. Zhang, Y. M. Chen, K. Yang, Y. X. Zhang, M. M. B. Khan, Y. Arfat, Y. Aziz, *Front. Chem.* **2020**, *8*, 102.

[86] H. Zhang, W. Niu, S. Zhang, *ACS Appl. Mater. Interfaces* **2018**, *10*, 32640.

[87] L. Shi, R. Yang, S. Lu, K. Jia, C. Xiao, T. Lu, T. Wang, W. Wei, H. Tan, S. Ding, *NPG Asia Mater.* **2018**, *10*, 821.

[88] Y. Gao, D. Sun, J. Chen, K. Xi, X. Da, H. Guo, D. Zhang, T. Gao, T. Lu, G. Gao, L. Shi, S. Ding, *Small* **2022**, *18*, 2204140.

[89] K. Williams, V. Bertola, D. S. Martin, *Results Phys.* **2016**, *6*, 277.

[90] J. V. Rowley, P. Wall, H. Yu, G. Tronci, D. A. Devine, J. J. Vernon, P. D. Thornton, *ACS Appl. Polym. Mater.* **2020**, *2*, 2927.

**ADVANCED
SCIENCE NEWS**

www.advancedsciencenews.com

**ADVANCED
FUNCTIONAL
MATERIALS**

www.afm-journal.de

[91] T. Phaechamud, O. Setthajindalert, *J. Drug Delivery Sci. Technol.* **2018**, *46*, 285.

[92] Z. I. Kirkwood, B. C. Millar, D. G. Downey, J. E. Moore, *Int. J. Mycobact.* **2018**, *7*, 134.

[93] Y.-F. Liu, Q. Liu, J.-F. Long, F.-L. Yi, Y.-Q. Li, X.-H. Lei, P. Huang, B. Du, N. Hu, S.-Y. Fu, *ACS Appl. Mater. Interfaces* **2020**, *12*, 49866.

[94] L. Zhai, F. Zhang, J. Sun, M. Liu, M. Sun, R. Lu, *Dyes Pigm.* **2017**, *145*, 54.

[95] T. Zhang, F. Chen, C. Zhang, X. Che, W. Li, B. Bai, H. Wang, M. Li, *ACS Omega* **2020**, *5*, 5675.

[96] S. Wu, H. Jiang, Y. Zhang, L. Wu, P. Jiang, N. Ding, H. Zhang, L. Zhao, F. Yin, Q. Yang, *J. Mol. Liq.* **2021**, *327*, 114836.

[97] X. Yang, Y. Liu, J. Li, Q. Wang, M. Yang, C. Li, *New J. Chem.* **2018**, *42*, 17524.

[98] X. Chen, Y. Zhou, M. Yang, J. Wang, C. Guo, Y. Wang, *J. Mol. Struct.* **2022**, *1250*, 131810.

[99] X. Chen, Y. Zhou, G. Zhang, J. Wang, C. Guo, Y. Wang, *Colloid Interface Sci. Commun.* **2021**, *44*, 100489.

[100] L. Wang, T. Xu, X. Zhang, *TrAC, Trends Anal. Chem.* **2021**, *134*, 116130.

[101] Y. Lee, W. J. Song, J.-Y. Sun, *Mater. Today Phys.* **2020**, *15*, 100258.

[102] Y. Gao, J. Guo, J. Chen, G. Yang, L. Shi, S. Lu, H. Wu, H. Mao, X. Da, G. Gao, S. Ding, *Chem. Eng. J.* **2022**, *427*, 131057.

[103] R. Payne, I. E. Theodorou, *J. Phys. Chem.* **1972**, *76*, 2892.

[104] X. Yao, S. S. Dunn, P. Kim, M. Duffy, J. Alvarenga, J. Aizenberg, *Angew. Chem., Int. Ed.* **2014**, *53*, 4418.

[105] P. Singh, R. Venkatesan, H. S. Fogler, N. Nagarajan, *AIChE J.* **2000**, *46*, 1059.

[106] M. O. Adebajo, R. L. Frost, J. T. Kloprogge, O. Carmody, S. Kokot, *J. Porous Mater.* **2003**, *10*, 159.

[107] I. Ali, M. Asim, T. A. Khan, *J. Environ. Manage.* **2012**, *113*, 170.

[108] S. Cao, T. Dong, G. Xu, F. Wang, *J. Nat. Fibers* **2017**, *14*, 727.

[109] S. Debnath, S. Kaushal, S. Mandal, U. Ojha, *Polym. Chem.* **2020**, *11*, 1471.

[110] S. Bayraktaroglu, S. Kizil, H. Bulbul Sonmez, *J. Environ. Chem. Eng.* **2021**, *9*, 106002.

[111] Y. Tezcan Demirel, I. Yati, R. Donmez, H. Bulbul Sonmez, *Chem. Eng. J.* **2017**, *312*, 126.

[112] C. L. Esposito, V. Tardif, M. Sarrazin, P. Kirilov, V. G. Roullin, *Mater. Sci. Eng., C* **2020**, *114*, 110999.

[113] C. L. Esposito, P. Kirilov, V. G. Roullin, *J. Controlled Release* **2018**, *271*, 1.

[114] L. R. Zancan, F. A. Bruinsmann, K. Paese, P. Türck, A. Bahr, A. Zimmer, C. C. Carraro, P. C. Schenkel, A. Belló-Klein, C. I. Schwertz, D. Driemeier, A. R. Pohlmann, S. S. Guterres, *Int. J. Pharm.* **2021**, *610*, 121181.

[115] R. Fracasso, M. Baierle, G. Goëthel, A. Barth, F. Freitas, S. Nascimento, L. Altknecht, V. Olsen, K. Paese, V. D. da Silva, I. Castro, M. Andrades, N. Clausell, A. Pohlmann, S. Guterres, S. C. Garcia, *Toxicol. Res.* **2016**, *5*, 168.

[116] D.-E. Liu, Q. Chen, Y.-B. Long, J. Ma, H. Gao, *Polym. Chem.* **2018**, *9*, 228.

[117] M. Lu, Y. Cao, C.-T. Ho, Q. Huang, *J. Agric. Food Chem.* **2016**, *64*, 4735.

[118] I. Apsite, S. Salehi, L. Ionov, *Chem. Rev.* **2022**, *122*, 1349.

[119] B. Jin, J. Liu, Y. Shi, G. Chen, Q. Zhao, S. Yang, *Adv. Mater.* **2022**, *34*, 2107855.

[120] Z. Zhang, Y. Yu, H. Yu, Y. Feng, W. Feng, *SmartMat* **2022**, *3*, 632.

[121] E. Borré, J.-F. Stumbé, S. Bellemin-Laponnaz, M. Mauro, *Angew. Chem.* **2016**, *128*, 1335.

[122] S. Debnath, C. Upadhyay, U. Ojha, *ACS Appl. Mater. Interfaces* **2022**, *14*, 9618.

[123] M. M. Perera, P. Chimala, A. Elhusain-Elnegres, P. Heaton, N. Ayres, *ACS Macro Lett.* **2020**, *9*, 1552.

Mariia Kuzina graduated from Moscow State University, Faculty of Materials Science, with honors. In 2020, she joined the biofunctional materials systems group of Prof. Levkin as a Ph.D. student. Her research in polymer chemistry is dedicated to 3D printing of functional polymers, organogels, and porous polymers. Her research interests include materials science, surface chemistry, and surfaces with special (patterned) wettability.

Dmitrii Kartsev received his Bachelor's degree in chemical engineering from Saint Petersburg State Technical University and his Master's degree in chemistry from Saint Petersburg State University. In 2021, he joined the biofunctional materials research group at ITMO University as a Ph.D. student. His current research concerns surface chemistry and wettability patterning. His research interests include organic chemistry, surface chemistry, and materials science.

Alexander Stratonovich earned a B.Sc. in Chemical engineering from Saint Petersburg State Chemical-Pharmaceutical University. In 2021, he joined the group of biofunctional materials at ITMO University as a M.Sc. student. His current research is dedicated to the photodegradation of cross-linked polymethacrylate organogels. His research interests include organic synthesis, materials science, and surface chemistry.

Pavel A. Levkin is head of the Biofunctional Materials Systems research group at the Institute of Biological and Chemical Systems (IBCS-FMS) and Institute of Organic Chemistry (IOC) at Karlsruhe Institute of Technology (KIT), Germany. He graduated from the Institute of Fine Chemical Technology, Moscow, and obtained his Ph.D. in Organic Chemistry from the University of Tübingen in Germany, followed by postdoctoral work at the University of California, Berkeley. Pavel Levkin is the cofounder of ScreenFect GmbH and Aquarray GmbH. His research group works, inter alia, on the development of biofunctional, responsive materials, and surfaces; coatings and micropatterns with special wettability; gene delivery; miniaturization of chemistry and biology; as well as 3D printing.