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EDITORIAL

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Smart polymeric hydrogels

In nature, most biological tissues exist in a quasi-solid hydrogel state, adapting their properties in response to environmental changes via natural evolution. Encouraged by these natural hydrogels, significant research efforts over the past two decades have been devoted to creating their synthetic counterparts, known as smart polymeric hydrogels (SPHs). Nowadays, SPHs have attracted a global research focus due to their unique and adaptable properties; however, creating SPHs that meet specific demands remains one of the major scientific challenges. Through rational designs, SPHs can be engineered to exhibit remarkable adaptability in both structure and function beyond imagination for many unprecedented applications. In light of these advancements, this special issue of *SmartMat* hosted by Prof. Tao Chen, Prof. Feng Zhou, Prof. Ximin He, and Prof. Jie Zheng, featuring the topic of "Smart Polymeric Hydrogels (SPHs)" selectively collects one Review and 11 Research Articles from leading experts. This collection aims to highlight recent several key aspects of material design and synthesis, functional advancement, and diverse applications of SPHs.

1 | CONDUCTIVE SPHs

In the modern era, there is a growing interest in flexible wearable electronics that can mimic the remarkable mechanical properties of biological tissues and skin-like sensations. These devices have huge potential in health monitoring, soft robotics, prosthetics, and more. Polymeric hydrogels, which mimic human tissues in mechanical modulus, softness, and moisture, are believed to be promising materials for these applications. The key step in creating SPH-based flexible electronics lies in the design of SPHs with responsive conductive properties. To this end, Jie Zheng and coworkers (https://doi.org/10.1002/smm2.1160) introduced conductive polypyrrole (PPy) into polyvinyl alcohol (PVA) matrix to produce a type of double-network PVA/PPy hydrogels via in situ ultrafast gelation followed by a freezing/thawing process. Peiyi Wu and colleagues (https://doi.org/10.1002/smm2.1228) proposed to leverage the rapid self-initiated polymerization of conductive MXene nanomaterials with abundant hydrophilic functional –OH/–F groups and cationic methyl chloride quarternized *N*,*N*-dimethylamino ethylacrylate monomers to produce a new MXene-based nanocomposite hydrogel through both hydrogen bonds and electrostatic interactions. Benefiting from the incorporation of PPy and MXene as conductive additives, which have been extensively tested for their sensitivity to strain, pressure, or temperature changes, the resultant SPHs have achieved excellent conductivity for diverse applications, including the continuous monitoring of human respiration under physiological conditions, limb movement, and body temperature. Furthermore, electroencephalography, a critical physiological signal, has been addressed by Jianxin Tang et al. (https://doi.org/10.1002/smm2.1173), who reported a self-adhesive and low-contact impedance PVA/polyacrylamide (PAAm) double-network hydrogel used as a promising semidry electrode for real-world brain–computer interfaces.

Another important strategy for creating smart conductive polymeric gels involves the integration of ionic liquids or deep eutectic solvents into the voids of three-dimensional (3D) cross-linked polymer networks. This integration forms 3D ion channels within the polymer matrix, enabling ions to directionally migrate and redistribute under external electric fields, thus endowing the polymeric gels with intrinsic ionic conductivity. Unlike traditional ionic hydrogels, these polymeric ionogels can effectively address the long-standing issues of water evaporation, offering the potential for enhanced long-term measurement stability in flexible sensory devices. To highlight the recent progress on polymer ionogels and their applications in flexible ionic devices, Tengling Ye and coworkers (https://doi.org/10.1002/smm2.1253) were invited to contribute a review article. This article thoroughly presents the design principles, working mechanisms, and representative examples of polymer ionogel-based flexible ionic devices. It also explores current challenges and future directions in this dynamic field, aiming to inspire more future research and bring

polymer ionogel electronics much closer to real-world applications.

2 | FLUORESCENT SPHS

Fluorescent polymeric hydrogels, emerging as innovative luminescent materials, combine the properties of classic fluorescent materials with polymeric hydrogels, inheriting the advantages of both to offer many promising applications in optical sensors, soft actuators, and robotics. Facile modulation of their fluorescent features (e.g., intensities and colors) is crucial for these promising applications. In this context, Benzhong Tang and Xiaofan Ji (https://doi.org/10.1002/smm2.1184) copolymerized a special solvent polar-responsive aggregation-induced emission monomer that presents a twisted intramolecular charge transfer into the chemically cross-linked poly(*N*-isopropylacrylamide) (PNIPAm) network, successfully producing smart fluorescent hydrogels capable of changing with emission colors in response to different solvents. Michael J. Serpe and coworkers (https://doi.org/10.1002/smm2.1254) chemically grafted hydrophobicity-sensitive Nile blue fluorophore as the pendent group of PNIPAm network, creating smart fluorescent microgels. These microgels exhibited enhanced fluorescence upon the introduction of cucurbit[7]uril (CB[7]) and significantly reduced brightness with further addition of polyamine to remove CB[7], suggesting their potential for optical polyamine detection. Tao Chen and colleagues (https://doi.org/10.1002/smm2.1190) explored the use of fluorescent lanthanide complexes to create pH-sensitive emission color-changeable polymeric hydrogels, followed by the fabrication of bilayer hydrogel actuators with unique synergistic color and shape changes. The hydrogel actuators replicate the dual responsiveness (both shape deformation and color change) of living creatures to environmental variations, making a significant step toward developing biomimetic intelligent soft robotics.

3 | THERMOCHROMIC SPHS

Thermochromic smart windows, which can spontaneously adjust the transmittance of solar radiation to control indoor temperature, have emerged as a promising strategy for reducing energy consumption in buildings. The traditional materials used in thermochromic smart windows, such as rigid VO_2 coatings, however, face challenges such as limited flexibility, high activation temperature (~68 °C), and low luminous transmittance, all of which hinder their practical applications. In contrast, thermoresponsive polymeric hydrogels, known for their excellent flexibility, high luminous transmittance, and tunable activation temperature, are considered ideal candidates for next-generation thermochromic smart windows. Leveraging this potential, Junqi Sun and coworkers (https://doi.org/10.1002/smm2.1256) have developed polyelectrolyte complex-based thermochromic hydrogels by combining polyallylamine hydrochloride, polyacrylic acid, and carbonized polymer dots in aqueous NaCl solution. These hydrogels showed a unique combination of rapid transition time in seconds, ultrahigh luminous transmittance, and desirable critical temperature, suggesting their wide potential uses for practical thermochromic smart windows.

4 | BIOMEDICAL SPHS

Owing to their unique water-swollen 3D cross-linked hydrophilic polymer networks, polymeric hydrogels usually have a tissue-like, soft, wet nature, intrinsic biocompatibility, and efficient permeability to small ions/molecules. Such advantages make polymeric hydrogels excellent candidates as drug/antibody carriers, enabling controlled release to minimize drug accumulation in healthy tissues and improve therapeutic efficacy at tumor sites. Therefore, the development and application of biomedical SPHs are becoming a significant focus in current research. In this special issue, Yanyan Jiang and colleagues (https://doi.org/10.1002/smm2.1148) report on a new in situ-forming hybrid hydrogel system, combining gold nanorods with sodium alginate, followed by the encapsulation of the chemotherapeutic drug doxorubicin hydrochloride and the molecular-targeted drug Herceptin for synergistic tumor therapy. The resulting hydrogels have featured several properties such as injectability, photothermal ablation, dual-stimuli-responsive drug release, and synergistic drug–antibody therapy, suggesting their potential as universal carriers for localized delivery of multiple drugs, aiming to address the challenges in treating HER-2 overexpressing cancers.

5 | SMART SURFACES/INTERFACES OF SPHS

Surficial and interfacial properties play essential roles in the study of SPHs. For example, rational surface functionalization can facilitate the assembly of simple polymeric hydrogel blocks into sophisticated macroscopic network structures. Zhihai Ke (https://doi.org/10.1002/smm2.1251) introduced the synthesis of alkynes- and azides-functionalized PAAm hydrogel blocks and demonstrated their selective covalent assembly via macroscale interfacial click reactions.

This strategy is generally applicable for rapidly connecting various soft materials to create macroscopic hydrogel assembly. Compared with stable click covalent bonds, dynamic covalent bonds provide SPHs with reconfigurable surficial/interfacial features because of their reversible dissociation/recombination nature in response to environmental changes. Qian Zhao and colleagues (https://doi.org/10.1002/smm2.1190) employed 2,2′-dithiodiethanol dimethacrylate with dynamic disulfide bond as a cross-linker to create a dynamically cross-linked amphigel, enabling orthogonal reconfiguration of both surface structures and stiffness patterning. Feng Zhou and coworkers (https://doi.org/10.1002/smm2.1235) designed and synthesized coumarin-functionalized photoreversible polymeric materials, showing both long-range reversible maneuverability and UV light-triggered adhesive strength owing to their dynamic cross-linking through reversible coumarin dimerization and cycloreversion. This study holds promise for advancing the development of remotely controllable smart underwater adhesives.

In summary, studies on SPHs are currently experiencing significant global research interest due to their rapid development and proposed applications. SPHs are poised to become essential materials and devices for the future needs of soft electronics/robots, optical sensing, energy management, tumor therapy, and smart adhesion, among others. We hope that the readers will find this special issue on SPHs both informative and inspiring. Finally, we extend our heartfelt gratitude to the *SmartMat* editorial team for their professional editing, and to all the coauthors and reviewers for their invaluable contributions to this special issue.

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