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SPECIAL ISSUE: Flexible Intelligent Materials

Marching towards flexible intelligent materials

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Our society is transforming into more intelligent eco-systems that seamlessly merge human beings, machines, the internet, and energy. The unprecedented momentum in such dramatic transformation inevitably relies on innovation in smarter materials. Flexible intelligent materials are eagerly demanded in such evolution. Flexible intelligent materials are soft materials whose properties can be altered by external stimuli, such as chemicals, mechanical forces, temperature, light, humidity, pH, and electric or magnetic fields. The mechanical flexibility enables the adaptability to directly interface with human beings, while various responsiveness to different stimuli provides a rich recipe for constructing different intelligent systems. Recently, flexible intelligent materials have been applied in diverse interdisciplinary fields such as flexible and wearable electronics, smart sensing, human-machine interfaces, soft robotics, drug delivery, information security, energy storage, etc. To show the leading-edge research in the field of flexible intelligent materials, a special issue entitled *Flexible Intelligent Materials* has been organized by *Science China Materials*, including high-quality reviews and research articles from this emerging and interdisciplinary field.

This special issue covers topics such as intelligent organic/polymer materials and their applications in various fields, including energy storage and conversion devices, ionic e-skins, smart sensors, light-emitting and fluorescent devices, actuators, neuromorphic devices, and information encoders. From the materials perspective, smart polymer/organic materials and their composites have been rationally constructed, and have been applied in various fields. Yu *et al.* [1] summarized the recent developments in the field of organic crystal-based flexible smart materials, including the derivatives of azobenzene, diarylethene, anthracene, and olefin, which can bend, curl, twist, deform, or respond otherwise to external stimuli, such as heat or light. Such a collection of crystal-based flexible smart materials can be applied in photoswitches, artificial muscles, and robots that can be remotely triggered by light. Leveraging chemical interactions to design desired smart material properties presents huge opportunities for sustainable materials development. Dynamic covalent bonds are a powerful toolkit for such representatives. Xu *et al.* [2] reviewed recent research efforts on dynamic covalent bonds-containing polymer materials with different functions, including self-healing, chemical recycling, and shape controlling. Smart display materials are another emerging category of intelligent materials that allows secure information encoding and decoding at the material property level. Zhao *et al.* [3] reported dynamic stimulus-responsive long-lived room-temperature phosphorescent with various emission colors by doping organic chromophore molecules into newly developed oxygen-consuming polymer matrices, pushing the room-temperature phosphorescent material a step closer to information security applications. Hydrogel material for ionotronics is getting more and more functional. Wu *et al.* [4] developed a multifunctional ionic skin based on ionic conductive and light-managing hydrogels *via* a facile one-step locally confined polymerization. It can block UV light to protect skin from damage, shield thermal IR light for stealth, dynamically tune the transmittance under solar light for heat insulation, and enables intelligent dialogue in the form of Morse codes between humans and machines.

High-performance energy storage and conversion devices have been developed to meet the ever-growing demands for powering flexible consumable electronics, electric vehicles, and hybrid electric vehicles. Lithium metal batteries are boosting energy density and are considered an ideal candidate for next-generation flexible power supply. Wang *et al.* [5] provided a critical discussion on the developments, opportunities, and challenges of high-performance and highly flexible lithium metal batteries. Apart from energy or power density, smarter batteries are highly demanded as wearable applications grow. Batteries that can autonomously respond to various stimuli are emerging. Cui *et al.* [6] focused on the development of polymer-based stimulus-responsive materials (heat, pH, moisture and pressure, electric fields, etc.) and their applications in the field of energy storage. As wearable batteries that directly interface with human beings, battery safety is under more and more stringent scrutiny recently. Zhou *et al.* [7] proposed a nonflammable multi-functional Janus separator with self-extinguishing capability, high thermal stability and conductivity, good electrolyte infiltration, uniform lithium deposition, and efficient polysulfide shuttling inhibition. In addition, batteries are also made more stable, especially at the electrode-electrolyte interface. Tang and Hong *et al.* [8] introduced a strong Lewis acid, AlF_3, into poly (ethyleneoxide) (PEO) for the first time to increase the ion transport kinetics and improve interface stability of the PEO-based electrolyte in all-solid-state Li-metal batteries. Meanwhile, the mechanism for the construction of stable composite elec-

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trolytes by increasing ion mobility and stabilizing the Li/PEO interface was revealed. The application scenario of batteries is also getting extended to more and more harsh environments. Niu *et al.* [9] developed an anti-freezing and anti-drying gel electrolyte based on polyacrylamide and glycerol. The flexible Zn ion batteries based on this gel electrolyte can work at −40°C and retain 98% of their initial capacity after 30 days. Mai *et al.* [10] designed a novel three-dimensional-networked composite of iron vanadate nanosheet arrays/carbon cloths as a binder-free cathode for flexible Mg ion batteries, which exhibited potential for practical applications. Besides batteries, other forms of power sources are developed simultaneously. You *et al.* [11] prepared a stretchable and transparent organohydrogel for a triboelectric nanogenerator, showing an instantaneous peak power density of 262 mW m^{-2} at a load resistance of 10 MΩ and efficiently harvests biomechanical energy to drive an electronic watch and light-emitting diodes (LEDs).

Flexible and wearable sensors and electronics are other hotspots in the wide applications of human-machine interface, healthcare monitoring, and the Internet of Things. Starting from innovations in materials design and fabrication processes, stretchable conductors, strain or pressure sensors, bioelectrodes, neuromorphic devices, displays, and actuators, are discussed in this issue, covering a broad spectrum of flexible and stretchable applications. Zhang *et al.* [12] focused on one class of crystalline materials-based flexible memristors for state-of-the-art data storage and neuromorphic demonstrations. Qi *et al.* [13] reviewed various methods of using liquid metals, a type of highly stretchable and conductive materials, to fabricate stretchable electronic devices. Yang *et al.* [14] reviewed the latest advancements in high-resolution flexible electronics fabrication made by e-jet printing technology, including various materials used in e-jet printing inks, the process control of e-jet printing, and their applications. Chen *et al.* [15] focused on low-dimensional materials with excellent mechanical properties that boost the development of flexible neuromorphic devices. Shen *et al.* [16] constructed an integrated monitoring system based on a liquid metal-thermoplastic polyurethane film-based flexible strain sensor. Liu *et al.* [17] developed stretchable micro electrocorticogram electrodes to investigate penicillin-induced epilepsy in rats. Liu *et al.* [18] fabricated a Fe^{3+} ion-coordinated poly (acrylic acid) ionogel (PAIFe) with high stretchability, extreme temperature tolerance, and self-healing capability by a dynamic ionic cross-linking strategy for a skin-inspired ionic sensor. Dong *et al.* [19] prepared a hydrophobic organogel based on micelle aggregation and metal ion coordination for high-sensitivity underwater sensors. Zhang *et al.* [20] presented a unique poly(acrylic acid)/calcium acetate shape memory hydrogel with cold-induced shape recovery performances as ultrastrong artificial muscles. Hu *et al.* [21] developed a stress-deconcentrated ultrasensitive strain (SDUS) sensor with ultrahigh sensitivity (gauge factor up to 2.3×10^6) and a wide working range (0%–50%) *via* incorporating notch-insensitive elastic substrate and micro-crack-tunable conductive layer. Zhang *et al.* [22] reported high-performance inkjet-printed, flexible, and patterned electrochromic devices based on two-dimensional polyaniline sheets. Feng *et al.* [23] developed stable and low-resistance polydopamine-modified methacrylamide-polyacrylamide hydrogel for the brain-computer interface. The flexible electrode with the hydrogel membrane has been demonstrated to enable a similar function as that of commercial counterparts with a more stable interface.

Intelligent materials are also promising candidates in other fields, such as LED display, electronic mesofliers, and plasmonic encoding. Shi *et al.* [24] reviewed three main categories of mass transfer technologies for micro-LED display (pick-and-place, fluid self-assembly, and laser-enabled advanced placement) and the coupled detection and repair technologies after the transfer, and provided a comprehensive direction for micro-LED manufacturing. Jiang *et al.* [25] proposed a novel strategy based on plasma etching combined with controllable *in-situ* growth on one substrate to prepare unique quasi-three-dimensional Au nano-mushrooms with programmable morphologies, which led to a high-capacity plasmonic encoder. Zhang *et al.* [26] developed a type of morphable 3D mesofliers with shape memory polymer-based electrothermal actuators, capable of a large degree of actuation deformations with a fast response. These various functional devices enriched the eco-system of flexible intelligent materials, playing a paramount role in the next-generation human-oriented electronics.

In summary, this special issue presents state-of-the-art developments of organic/polymer materials and their applications in various fields, covering energy storage and conversion devices, smart sensors, flexible electronics, LEDs, electrothermal actuators, and information encoding devices. This special issue gives a deep insight into the flexible intelligent materials and promotes their applications in different fields. We thank all the authors for their contributions to this special issue entitled *Flexible Intelligent Materials*.

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