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Laser-induced stretchable bioelectronic interfaces by frozen exfoliation

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Highly stretchable laser-induced graphene—hydrogel film interfaces in flexible electronic materials are fabricated by frozen exfoliation, and exhibit high stretchability, durability, and design flexibility. This technology offers an advanced technological pathway for manufacturing highly flexible substrates. They can be utilized in numerous complex surface applications, providing an advanced technological pathway for manufacturing highly flexible substrates in the future.

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In recent years, the rapid advancement of flexible and stretchable electronics has been fueled by their soft and thin characteristics, biocompatibility, and long-term stability[1-4]. The stretchability of flexible materials[5-7] is critical to the mechanical and electrical performance stability of flexible electronics when conformally adhered to free-form surfaces, and this has attracted widespread attention. Graphene, due to its exceptional electromechanical properties, has been widely applied in flexible electronic systems[8–11]. Recently, the synthesis and processing method of inducing graphene from aromatic polymers through laser irradiation has made the one-step fabrication and patterning of graphene devices possible[12–14]. By controlling the temperature value of the irradiating laser, selective polymer graphitization can be achieved, resulting in laser-induced graphene (LIG) with porous conductive characteristics, which possesses high sensitivity and low resistance. They can conformably and compliantly adhere to irregular organs such as the skin[15, 16], brain[17, 18], and heart[19, 20], and have been extensively applied in multiple scenarios of bioelectronics.

Although LIG has been used in diverse flexible electronic devices, it still remains bound to the precursor polymers, limiting its stretchability and conformability in wearable and implantable applications. This issue has limited their broader industrial applications. Researchers have been devoted to achieving a highly mechanically stable interface between highly conductive electrode materials and highly flexible materials, which is a central challenge in the manufacturing of flexible electronics.

In a recent paper published in *Nature Electronics*[21], Prof. Kaichen Xu and Prof. Zhen Gu et al. from Zhejiang University proposed a thin elastic conductive nanocomposite based on LIG micropatterns, which is realized by a cryogenic transfer approach with an ultrathin and adhesive polyvinyl alcohol (PVA)–phytic acid (PA)–honey (PPH) hydrogel with the thickness of 1–1.5 μm. This approach has achieved 110% intrinsic stretchability and 220% structural optimization, which realize an improvement of more than fivefold over the previously reported flexible electrode materials composed of brittle LIG and polydimethy-lsiloxane (PDMS). The graphene-hydrogel nanocomposite films prepared in this way possess excellent sensing performance, antibacterial properties, biocompatibility, and high stretchability, which are

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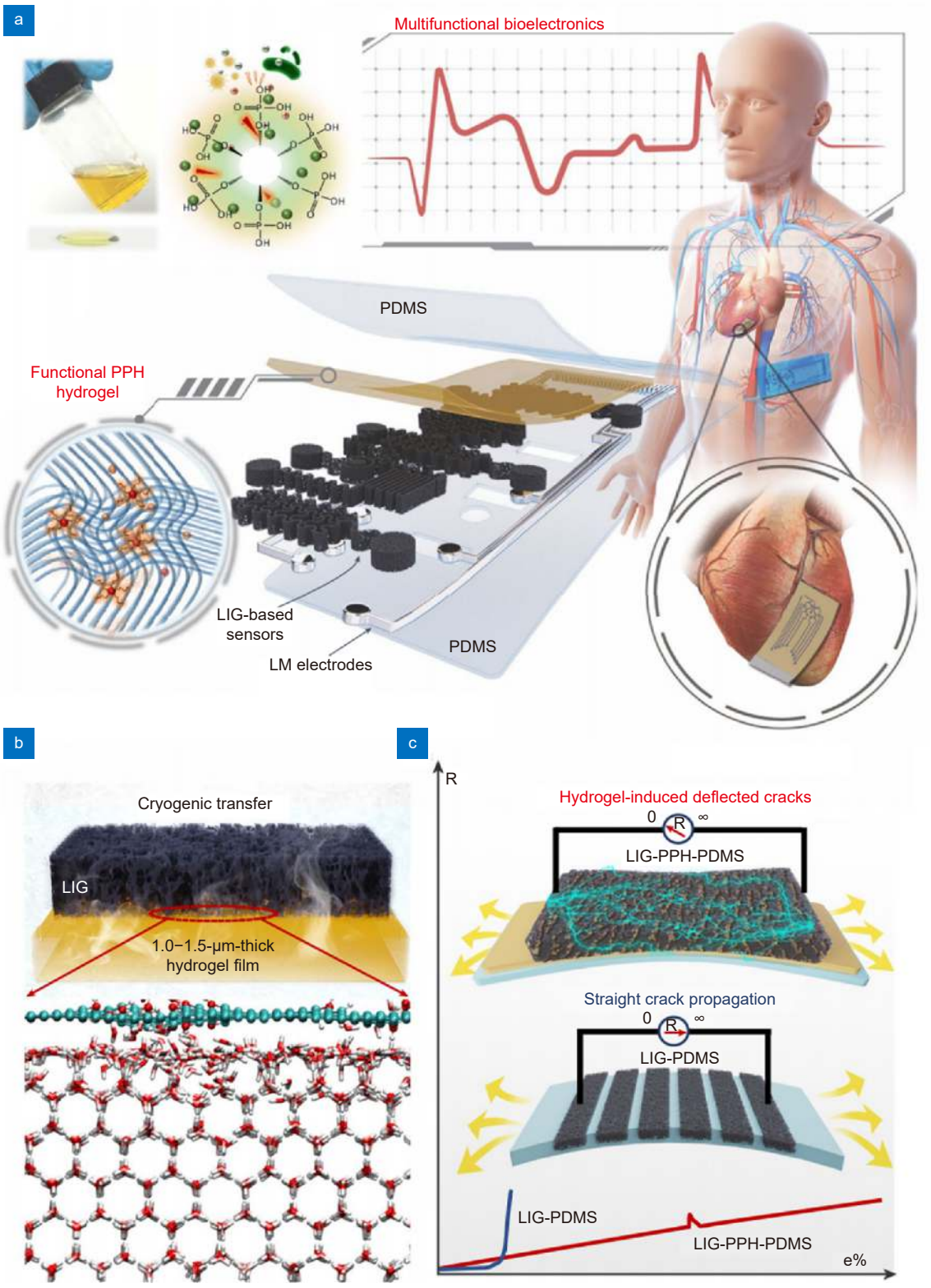

Fig. 1 | The schematic of the design scheme of stretchable graphene–hydrogel nanocomposites[17]

crucial for skin patches and implantable electronic devices.

Figure 1 shows the schematic of the highly stretchable flexible electronic material preparation technology based on high-strength bonding of LIG and PPH hydrogel. The key aspects of this technology mainly include two parts. First, the high-stability transfer scheme allows for low-temperature peeling of LIG onto the PPH hydrogel with

low Young's modulus, high adhesiveness, and antibacterial performance. On the one hand, low-temperature interfacial electrostatic interactions enhance the interfacial binding energy between crystalline water molecules in the hydrogel. At the same time, the low temperature causes the gel layer to expand in volume, allowing the porous LIG part to embed into the PPH film. This leads to the formation of interlocking structures with high mechanical stability, which improves the robustness of the composite material. Second, compared with traditional LIG-PDMS conductive materials, the ultra-thin PPH provides the higher adhesion due to the dipole effect of −P=O, −C=O, −C–F and $-CF_3$ groups, which results in the deflected fractures with ductile behaviours, and can provide out-of-plane points of contact to connect the fractured LIG film, thus enhancing the intrinsic conductive stretchability of the nanocomposite material. Based on the aforementioned fabrication technology, a further development involves constructing integrated multifunctional sensing skin patches that include strain sensors, temperature sensors, humidity sensors, and electrocardiogram (ECG) sensors, as well as an array of strain sensors adhered to the surface of the heart for in-body applications. This facilitates the monitoring of near-death signals caused by arrhythmias, demonstrating practical applications of the sensors. It also proves the enhanced conformability of the fabricated flexible sensors to complex surfaces and the improvement of their signal stability.

Based on the low-temperature transfer scheme for nanocomposite material preparation, there is a significant advantage in terms of stretchability and mechanical strength, and there is also great potential for further development. The design and fabrication of three-dimensional micro/nanostructures and circuit morphologies can further enhance the sensitivity of the sensing system[22-24]. Moreover, laser fabrication methods, such as LIG, necessitate further investigation into their scalability for the production of large-area electronics and bioelectronics. Advancing this research could elevate these technologies to the scalability of other fabrication strategies, such as printed electronics using two-dimensional materials[25]. For example, spatial-temporal shaping femtosecond laser can achieve flexible control of the surface micro-nano structures and efficient preparation. It has been reported that femtosecond laser-induced graphene possesses the smaller dimensions and higher electrical conductivity, allowing for the preparation of LIG with higher precision[12, 26]. In summary, the scheme presented in this paper for low-temperature transfer of LIG to ultra-thin flexible substrates exhibits high stretchability, high robustness, and high design flexibility, providing an advanced technical pathway for the future fabrication of highly flexible substrates towards many complex surface applications, such as smart brain patches, soft wound patches, and electrical stimulation for muscle treatment.

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