木质素原位组装制备具有仿生多相结构离子导电弹性体

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摘　要:【目的】利用木质素通过仿生结构设计制备基于低共熔溶剂的具有优异机械性能、自修复性能、良好导电性及环境适应性的木质素基离子导电弹性体（ICs）。【方法】将改性的木质磺酸钠大分子单体（LSM）作为交联剂与氯化胆碱（ChCl）、丙烯酸（AA）及香草醛衍生单体（VAM）组成的可聚合低共熔溶剂通过一步自由基聚合制备具有仿生多相结构的木质素基离子导电弹性体，通过拉伸试验、电导率测试及传感性能测试等实验方法测试木质素基离子导电弹性体的机械性能、导电性、环境适应性及可穿戴传感性能。【结果】1）利用 LSM、ChCl、AA 和 VAM 组分中存在的多重非共价相互作用成功制备了具有仿生多相结构的木质素基离子导电弹性体；2）多重非共价相互作用及软-硬相结构赋予离子导电弹性体优异的机械性能，拉伸强度和韧性分别可以达到 7.14 MPa 和 23.3 MJ/m^3，同时离子导电弹性体还具有优异的弹性；3）离子导电弹性体具有良好的导电性及耐候性，在-20 ℃和 25 ℃下的电导率分别为 0.25×10^{-4} S/cm 及 0.33×10^{-4} S/cm；4）基于离子导电弹性体制备的应变传感器具有高的灵敏度（0.358-0.653）、快速响应特性（386 ms）以及优异出色的传感稳定性（1300 次循环），并且该应变传感器可用于实时监测人体不同部位的运动。【结论】利用木质素制备了基于可聚合低共熔溶剂的具有仿生多相结构的离子导电弹性体，并实现了其在多功能可穿戴传感器领域的应用。

关键词：木质素；可聚合低共熔溶剂；离子导电弹性体；仿生设计；多功能传感器

Lignin In-situ Self-assembly Facilitates Biomimetic Multiphase Structure for Fabricating Ionic Conductive Elastomer

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Abstract:【Objective】The modification of natural lignin that preserves its original structure for constructing ionic conductors (ICs) with soft–hard multiphase structures integrated with multiple noncovalent interactions is a highly desirable but challenging process. Herein, we present a new lignin mediated biomimetic multiphase structure integrated with multiple noncovalent interactions for fabricating strong, tough, and conductive ICs with high elasticity and solvent resistance, as well as good anti-freezing and self-healing properties. 【Method】In this study, ChCl, AA, a renewable vanillin-derived monomer (VAM), and a lignin-derived macro-crosslinker (LSM) are easily polymerized in one-pot to generate an IC via UV-induced radical polymerization. And the mechanical properties, conductivity, environmental adaptability and wearable sensor properties of ICs were analyzed by tensile tests, electrical conductivity measurement and sensor tests. 【Result】1) Owing to the polymerization of acrylic acid, the solubility of lignin in the system gradually decreases. Therefore, lignin starts to in-situ self-assemble into rigid nanosphere structure crosslinkers via noncovalent interactions, such as π-π interaction and hydrogen bonds. This structure is essential to realize the microscale biomimetic design of soft–hard two-phase structures. 2) The ionic conductor possessed high tensile strength of 7.14 MPa, toughness of 23.3 MJ/m^3 and maintained the high elasticity, owing to the increase in the rigidity of polymeric chains and larger number of noncovalent interactions, such as hydrogen bonds and dipole–dipole interactions. 3) The conductivity of ICs was 0.33 × 10^{-4} S/cm at 25 °C and 0.25 × 10^{-4} S/cm at −20 °C, suggesting a potential applicability of the studied ICs in cold environments. 4) The high-performance strain and pressure sensors manufactured from these materials demonstrated satisfactory sensitivity (S = 0.358–0.653), relatively fast response (386 ms), and long-term reliability (1300 cycles), which allow their potential applications in multifunctional wearable sensors for monitoring human motions, facial expression, speaking, and handwriting and pressure sensor arrays. 【Conclusion】A lignin mediated ICs with biomimetic multiphase structure based on polymerizable deep eutectic solvents paves the way for the fabrication of sustainable and multifunctional wearable sensors suitable for harsh environments.

Keywords: lignin; polymerizable deep eutectic solvent; ionic conductor; biomimetic design; multi-functional sensor.