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Functional biomaterials towards flexible electronics and sensors

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ABSTRACT

Biomaterials have gained increasing attention in the fabrication of a variety of flexible electronics due to their tunable solubility, robust mechanical property, multi-active binding sites, and excellent biocompatible and biodegradable characterization as well. Here, we review the recent progress of bio-based materials in flexible sensors, mainly describe nature biomaterials (silk fibroin, cellulose and chitin) and chemical-synthesized biomaterials as well as their applications in health monitors, biosensor, human-machine interactions (HMIs) and more, and highlight the current opportunities and challenges that lay ahead in mounting numbers of academia and industry. Furthermore, we expect this review could contribute to unveiling the potentials of developing outstanding and eco-friendly sensors with biomaterials by utilization of printing techniques.

1. Introduction

Flexible wearable electronic devices which can tolerate or withstand the stretching or bending forces, or other large deformation have been widely investigated in past decades (Wu et al., 2010). The flexible, extensible electronic device normally consists of traditional rigid board systems and stretchable materials: the former typically shows good mechanical properties and the lateral can be stretched like a rubber band (Zhang et al., 2014) and folded like paper (Yu et al., 2014). Particularly, the ever-growing demands for flexible electronic devices will dedicate to a rapid population growth. Meanwhile, the disposal of abandoned electronic devices has already received considerable concerns around the globe in recent years, which may cause adverse impact on the environment. That is because majority of previous developed electronic devices that are made up of non-biodegradable raw materials, like some plastics, cannot meet the trend of frequent updating of consumer electronics. Therefore, intensive studies are striving to challenge with this issue.

With the rapid development in electronic biomaterials and the relevant manufacturing techniques, researchers tend to combine bio-based materials with flexible electronic devices for sustainable development. The flexible sensor, one of the most essential parts of flexible electronics has its potential applications in monitoring human and robot motion (He et al., 2017; Jeong et al., 2013; Jung et al., 2014) and personal healthcare (Cheng et al., 2017; X. Wang et al., 2017), detecting overall hygiene in the food system (Dubal et al., 2018) as well as analyzing pesticide residues and toxic substances (Mishra et al., 2017). Furthermore, strategies to construct bio-based sensors with high mechanical durability, high sensitivity to deformations and responsive conductivity are also well addressed in numbers of published works (Kaushik et al., 2008; Zhu et al., 2013; Yuqing Liu et al., 2016; Yang et al., 2017).

Herein, we focus on the work of predecessors who committed to studying biomaterials and bio-based sensors. First, we summarize various biomaterials, including natural and conventional chemical-synthesized ones, and introduce fundamental processing properties of biomaterials and their great potential for bio-based sensors. Moreover, we highlight several applications with the example of respective biomaterials in individual healthcare, wellness pressure, sensing-based electronic skin (E-skin), biomedical diagnosis, food safety and environment governance. In addition, the future perspectives of bio-based sensors and the technological issues involved in applying biomaterials and practical devices are considered and prospected with the hope to improve the development and application of biomaterial-based flexible sensors in our daily life.

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2. Biomaterials

Biomaterials, also known as bio-based materials are produced from renewable resources. Specifically, those biomaterials are made from raw materials such as cereals (Diouf-Lewis et al., 2017), legumes (Voisin et al., 2014), straw (Paranthaman et al., 2009), bamboo powder (Hsieh et al., 2006) and other raw materials through biosynthesis, bioprocessing and biological refining process. Besides that, biomaterials also include bio-based plastics and fibers, sugar engineering products, and bio-based rubber made from biomass thermoplastic processing, which are degradable when exposed to microorganisms, carbon dioxide (aerobic) processes, methane (anaerobic processes), or water (aerobic and anaerobic processes) (Nair and Laurencin, 2007; Kumar et al., 2018; Stagner, 2016; Rivas et al., 2016). Polymer materials play irreplaceable roles in human daily life. However, except natural rubber and few other materials, most polymer materials are highly dependent on fossil resource (mainly oil and coal), which have induced lots of problems on environmental pollution, human health issues and destruction of the entire eco-environment (Brunner and Rechberger, 2016). To bring a significant reduction in greenhouse gas emissions and saving of fossil energy, sustainable and eco-friendly polymers have become increasingly important. Many efforts have been taken to fabricate various biodegradable materials from bio-based chemicals (Sheldon, 2014; Ummartyotin and Pechyen, 2016).

2.1. Chemical-synthesized biomaterials

The first generation of bio-based polymers focused on deriving polymers from agricultural feedstocks such as corn, potatoes, and other carbohydrate feedstocks. Natural biopolymers are the other class of biomaterials, such as proteins, nucleic acids, and polysaccharides (cellulose, chitin) (Badawy and Rabea, 2009). In addition, biodegradable polymers can be produced by chemical synthesis (da Silva et al., 2018), such as polylactic acid (PLA), polyurethanes (PUs), polydopamine (PDA) and polyhydroxyalkanoates (PHAs) with the chemical structures shown in Fig. 1.

2.1.1. Polylactic acid (PLA)

The original materials for synthesizing PLA mainly include corn, soybeans, beets, potatoes and other biological materials which are made of starch and sugar. One of the most important raw materials is corn, so that PLA resin is also known as "corn resin". Based on different optical rotation, PLA has three typical optical isomeric forms (Tsuji, 2013): optically active and crystalline form (poly (L-lactide) (poly (L-lactic acid) (PLLA)) and poly (D-lactide) (poly (D-lactic acid)) (PDLA)), optically inactive and amorphous poly (DL-lactide) (poly (DL-lactic acid)) (PDLLA). PLA is nowadays economically competitive and has been widely used in many day-to-day applications, mainly used in food packing (Carrasco et al., 2014) on the account of its outstanding advantages: ease of processing, superior transparency and environmentally benign characteristics. Furthermore, oriented PLLA materials exhibit unique piezoelectric properties (Ando et al., 2017), and the piezoelectric constant of PLLA increases with the draw ratio of the molecule and reaches the maximum at a draw ratio of around 4–5 (Ikada et al., 1996), which make PLLA a good candidate for piezoelectric sensors.

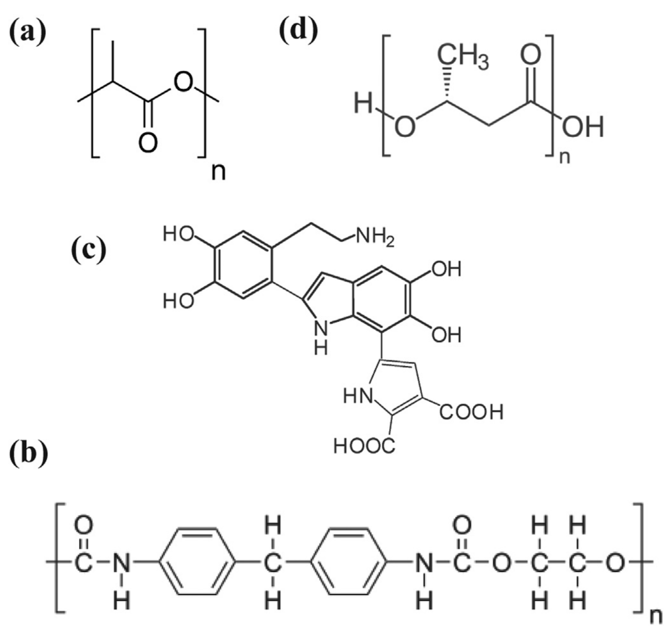

Fig. 1. The chemical structure of chemical-synthesized biomaterials: (a) PLA, (b) PU, (c) PDA, (d) PHAs.

2.1.2. Polyurethanes (PU)

PU refers to a type of polymer which contains a urethane feature unit in the main chain. PU is a block copolymer obtained by a stepwise addition reaction of isocyanate (NCO) and active hydrogen groups (Narayan et al., 2006), followed by chain extension and cross-linking. By changing the proportion of raw materials and the ratio of active hydrogen to NCO, properties of PU can be adjusted within a wide range: from soft sponges to elastomers, coatings to sealants, and shoe soles to elastic fibers. In particular, polyurethane (PU)-based materials have been used in different forms, such as in thermoplastic polyurethane (TPU), PU sponges, and PU yarn (Z. Wang et al., 2016). They are known for the favorable tensile properties and thus appropriate for preparing strain or pressure sensors (Huang et al., 2018; Xu et al., 2018). The sensors based on the above-mentioned materials can demonstrate high stretchability, little hysteresis, and long-term durability.

2.1.3. Polydopamine (PDA)

PDA is a principal pigment of natural melanin (eumelanin), which possesses many striking properties in optics, magnetics, electricity and biocompatibility (Liu et al., 2014). In addition, the multi-functional groups of PDA, such as amine, imine and catechol, act as the covalent/noncovalent binding of desired molecules and loading of transition metal ions, which further realize the emergence and extend the application of various PDA-based hybrid materials. Besides, PDA could be simply synthesized through various methods, such as simple polymerization of dopamine monomers in a mild condition (Hong et al., n.d.), enzymatic oxidation process (Tan et al., 2010), and electropolymerization (J. Wang et al., 2014). With these advantages, PDA has been widely used in energy, biomedical science, water treatment, and sensing applications (E. Lynge et al., 2011; Liu et al., 2014; G. Wang et al., 2017).

PDA is a promising functional material in the application of flexible electronics as the adhesive substrate and semiconductor, which has been inspired separately by the adhesive nature of catechols and amines and electron transition through π-system formed by aromatics and conjugated molecules (Chen et al., 2016; Ryu et al., 2018). Furthermore, incorporation with other materials via cross linking (Bie et al., 2016), in-situ deposition (Tan et al., 2010), and tethering (Xiaoling Wu et al., 2015) improves the mechanical and electrical properties of PDA complex, which further develops the connection between PDA-based materials and flexible electronics.

2.1.4. Polyhydroxyalkanoates (PHAs)

PHAs are a family of polyesters produced by bacterial fermentation with the potential to replace conventional hydrocarbon-based polymers, since they are thermoplastic, biodegradable, biocompatible, and nontoxic (Valentini et al., 2014). The most common PHAs includes poly-β-hydroxybutyrate (PHB), hydroxybutyrate copolyester (PHBV), poly(3-hydroxybutyrate-*co*-3-hydroxyhexanoate) (PHBH), and poly(3-