Macromolecular Reorganization as a basis for Converting Cellulosic Hydrogels into Sustainable Plastics

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The development of lignocellulose-derived sustainable plastics is an important strategy for a greener future. Cellulosic hydrogels, which are readily generated from a cellulosic source (*e.g.,* wood pulp), can be converted into high-strength plastics by hot-pressing. In this process, cellulose macromolecules are fluidized and reassembled, leading to significant change of bonding interactions and structural characteristics. This interesting concept would open the door for new possibilities of bioproduct design.

Keywords: Cellulosic hydrogels; Pulp and paper industry; Bioplastics; Hot-pressing

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Definition of Plastics

Plastic materials are widely used in packaging products, buildings, automobiles, household items, and toys, among others. Basically, plastics are polymer-based materials characterized by plasticity, a term correlated to the deformation of materials in response to applied forces. As described in ISO 472 (2013), a plastic is "a material that contains as an essential ingredient a high polymer and which, at some stage in its processing into finished products, can be shaped by flow." However, elastomeric materials (also shaped by flow) are not generally considered to be plastics. It is worth noting that in some countries, particularly the United Kingdom, the term "plastics" is used as the singular form as well as the plural form.

Converting Cellulosic Hydrogels into Bioplastics by Hot-Pressing

Most plastics are derived from unsustainable petroleum reserves. During the processing of these plastics, mineral fillers and other additives may also be added (Hubbe 2016). The use of bioplastics (*i.e.,* plastics derived from renewable resources) as alternatives to conventional plastics has much potential in terms of a greener economy (Dusselier *et al.* 2015). Interestingly, a simple concept of converting cellulosic hydrogel into bioplastics by hot-pressing has recently been proposed and demonstrated by Chinese researchers (Song *et al.* 2017; Wang *et al.* 2013, 2015, 2016).

In accordance with this concept, cellulosic hydrogel is formed from cellulosic pulp that is dissolved in an aqueous solution containing an alkali hydroxide and urea. A non-solvent is used for cellulose regeneration, and the subsequent washing with water results in the formation of cellulosic hydrogels. A key feature of the concept is the unique role of hot-pressing in reorganizing the macromolecules of the hydrogels. The impacts that would occur during hot-pressing include: (1) breakage of physical crosslinks of cellulosic hydrogels, (2) stretching and rearrangement of cellulose chains, (3) reduction in the crystallinity of cellulose, (4) strengthening of intermolecular bonding interactions, and (5) plastic deformation of cellulosic hydrogels (due to rapid removal of water).

Capability of being shaped by flow is a critical feature of plastics. Cellulose macromolecules are somehow fluidized and reorganized during hot-pressing, which is evidently similar to melt processing of conventional plastics. However, the transparent bioplastics generated from cellulosic hydrogels are distinct from conventional plastics in terms of processability. The change of aggregated structure of cellulosic hydrogels is a pronounced impact of hot-pressing. Encouragingly, the tensile strength, flexural strength, and thermal stability of cellulosic bioplastics has been found to be superior to some conventional synthetic plastics (acrylonitrile-butadiene-styrene copolymer, polycarbonate, polyethylene, polyvinyl chloride, and polybutylene terephthalate) and regenerated cellulose films. Also, functional materials (*e.g.* graphene) can be readily incorporated into cellulosic hydrogels, which leads to the construction of functional bioplastics for a wide range of end-use applications.

Possibilities

Essentially, the concept of converting cellulosic hydrogels into high-strength bioplastics could be readily applied to hydrogels from diversified sources, including microcrystalline cellulose, paper-grade pulp, and dissolving pulp. Comprehensive implementation of this concept would facilitate the development of new bioproducts for tailored applications, opening the door for numerous envisaged possibilities.

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