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Review

Electrospinning and electrospray of bio-based and natural polymers for biomaterials development

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ABSTRACT

Over nearly 70 years, polymers have revolutionized the global economy, manufacturing and, mainly, the fields which require biocompatible materials, as food technology and packaging, controlled drug delivery, tissue engineering, regenerative medicine, wound dressing, anti-allergy textiles, and personal care. While new high-performance polymers were produced from fossil-based sources to meet the functional performance demands of new applications, Earth has been polluted by the operation of factories that released CO_2 to the atmosphere during the production of synthetic polymers. At the same time, biocompatible and biodegradable alternatives were being required to meet specific needs of a range of applications. In this paper, we reviewed the use of electrospun/electrospray bio-based and natural polymers in the last ten years in food technology and smart packaging, food additives, antimicrobial packaging, enzyme immobilization, tissue engineering, drug delivery, wound dressing, anti-allergy fibers from milk, and faux meat. Also, we reviewed the use of ionic liquids and click chemistry techniques as alternatives for modification and functionalization of electrospun/electrospray bio-based and natural polymers.

1. Introduction

Polymers are essential materials in the modern society [1]. Over the past 60 years, polymers have transformed the agricultural production and food technology, improved health care through the introduction of functional medical devices, equipment, reduced the fuel consumption due to the fabrication of lighter vehicles, and enhanced the performance of aircraft. [1,2] All of these achievements were thanks to the synthesis of polymers in a diversity of properties, such as high mechanical performance and biodegradability [1]. The feedstocks used for the production of the polymers above mentioned are fuel/fossil-derivatives, primarily because they are easily assessable via petroleum and natural gas [1].

However, two main factors govern the gradual substitution of fossil-derived polymers by bio-based polymers [1,2]:

i. The scarcity of feedstock and the rise in prices of petroleum and natural gas, which impose higher costs for polymers, and
ii. The lack of environmental impact.

From The WWII to a few years ago, the petroleum-based plastics industry has powered the world's economy. Currently, the increasing demand for the petroleum-based raw material have been threatening the global economy and, most important, have been responsible for the world's climate change [1,3]. The release of carbon from petroleum-derived polymers by incineration or other degradation method increases the net greenhouse gasses in the atmosphere, which in turn, contributes to global warming (Fig. 1) [1,3].

Furthermore, it is important to highlight that fossil raw materials are a finite resource and, within a few generations, it will be depleted [1,3]. New resources, synthesis, and processing methods are strongly desirable to contribute to the reduction of the environmental impact generated by the current polymer industry [1].

A more sustainable level of development has to be reached, and one of the ways is to reduce the use of petroleum-based plastics [1,3,4]. Due to the advances in polymer science, the petroleum-based polymers can be replaced in nearly every function by bio-based polymers [1,3].

Bio-based polymers are materials derived from biomass (Fig. 1), according to the European Committee for Standardization (CEN) [5]. It

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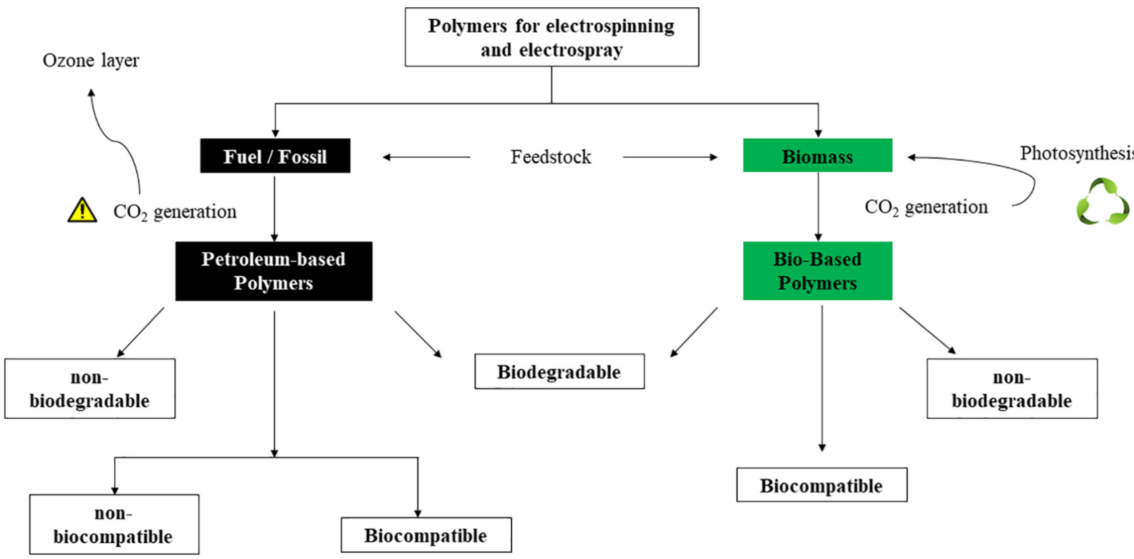

Fig. 1. Sources of polymers used in electrospinning/electrospray and its properties.

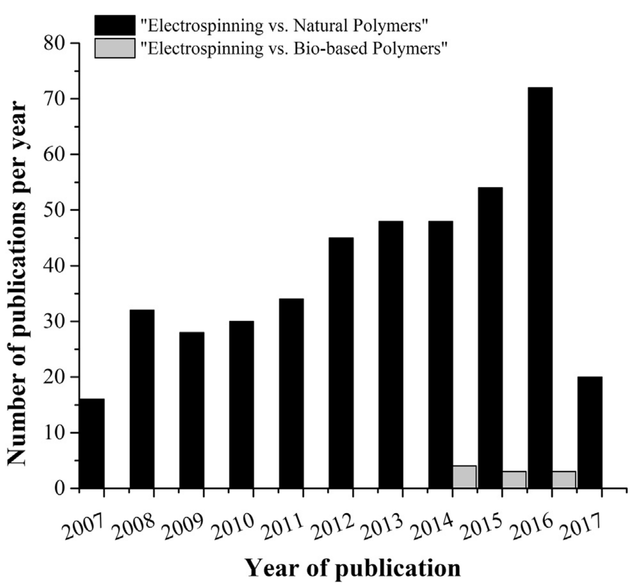

Fig. 2. Schematic representation of the number of publications per year using the expressions "natural polymers" and "bio-based polymers" over the last 10 years following Web of Science.

means that bio-based polymers are mainly composed of non-fossil materials [6] and offer a renewable alternative to traditional petroleum-based polymers. The bio-based raw materials can be found in a variety of biological sources, like agricultural products (corn, potato, or soybeans), micro-organisms, algae, or crustaceans [1]. They can also be partially composed of renewable and synthesized polymers [1]. Some examples of commercialized bio-based polymers are:

- Polylactides (PLA): derived from corn sugar, which is subsequently fermented to lactic acid and, finally, polymerized into poly(lactic acid) [2,7];
- Polyhydroxyalkanoates (PHAs/PHBs): a plant-derived intercellular sugar or lipid is fermented by micro-organisms, giving rise to linear polyesters [2,8];
- Polyols: sugar-alcohols-derived polymers. The most used monomeric polyols are glycerin and ethylene glycol. They have been commercialized and used by urethane industries [2,9].
- Polyamides: Nylon-11 (poly (undecanoic amino acid)) is a polymer derived from castor oil [2,10].
- Bio-PET: the bio-poly (ethylene terephthalate) is a combination of bio-based ethylene and other petroleum-based feedstocks. The ethylene portion is obtained by corn fermentation and is subsequently synthesized in the same process as a traditional PET [2,11].
- Butyl rubber: a renewable version of synthetic butyl rubber that is a copolymer of isobutylene and BioIsoprene™. The genetically modified *E. coli* is used to produce the enzyme isoprene synthase, which polymerizes a sugar solution, giving rise to BioIsoprene™ [2,12].
- Cellulose acetate: obtained from cellulose present in crops and leaves of plants [13].

Natural polymers are macromolecules that can be found in nature. Among them are proteins (e.g., collagen, gelatin, hyaluronic acid, zein, silk), polysaccharides (e.g., starch, cellulose, alginate, chitosan), terpenes (e.g., natural rubber), and lipids [14–16]. The advantage of using natural polymers is their ability to mimic the chemical environment of nature, primarily by giving biocompatibility. In contrast, they present weak mechanical properties, resulting in fragile materials in comparison to synthetic polymers [14,16].

In the last decade, more attention has been given to the use of bio-based and natural polymers as biomaterials for tissue engineering, wound dressing, drug delivery systems, and smart alternatives for biotechnology. It is worth to mention that using a polymer from a renewable source for any application in food, pharmaceutical, or medicine moves towards a more sustainable, affordable, and less dependent technology. Regardless of the area of application, the use of the expression "natural polymers" and electrospinning has constantly been growing in the last decade, unlike the concept of "bio-based polymers" to describe synthetic materials of natural origin, as demonstrated in Fig. 2. We can observe growth in the number of publications per year concerning to electrospinning of natural polymers, while the electrospinning of bio-based polymers remains very low. This result can highlight an urgent need for the development of new polymer molecules from renewable sources which present properties like biodegradability, biocompatibility, superior spinnability, and mechanical properties to be used in electrospinning/electrospray technology.

Given the importance of this issue, the ASTM's Subcommittee D20.96 on Biobased Products and Environmentally Degradable Plastics has published the "ASTM Biobased Content Briefing Paper," which aims to standardize the application and commercialization of bio-based polymers and help with definitions and examples for determining bio-based content [17] (Fig. 3).

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