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Editorial

Sustainable Polymer Technologies for a Circular Economy

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We inhabit a defining moment in history. It is a moment in which the scientific community has united to agree on an ambitious framework with which to resolve the environmental issues associated with plastic waste. Plastics are the "workhorse" material of the modern economy, offering multiple functions that help tackle several challenges facing our society. Plastics production has increased from 15 million tons in the 1960s to 311 million tons in 2014 and is expected to triple by 2050 as plastics come to serve increasingly more applications. Packaging is the dominant sectoral use of plastics globally, representing nearly 40% of the plastic market. However, after an initial short use cycle, most of the economic value of plastic packaging material is lost. Furthermore, hundreds of millions of tons of plastics escape collection systems, ending up in the environment either as microscopic particles or surviving in a recognizable form for hundreds of years. Therefore, it is high time to implement the principles of Circular Economy in the plastics industry. The Circular Economy has recently become a widespread concept, emerging in the past few decades from several schools of thought, such as "Cradle2Cradle" and "Biomimicry. It envisages a novel economic model that is restorative and regenerative by design, wherein its key aims are to eliminate the concept of waste, rebuild natural capital, and create economic value by using—not consuming—resources effectively. However, the current economic system of the plastics industry still follows the linear model 'make, use, and dispose', which erroneously assumes that economic growth can be based on the abundance of resources and unlimited waste disposal. According to the model outlined in the New Plastics Economy and other recent sustainable initiatives, strategies of Circular Economy regarding the plastics industry entail promoting sustainable polymer technologies that decouple plastics from fossil feedstocks, drastically reduce the leakage into and the effects of plastics on natural systems, and create an effective after-use plastics economy. This Special Issue focuses on recent research studies devoted to enabling better economic and environmental advances in the plastic packaging value chain that can successfully accelerate the transition of the plastics industry from its traditional linear economy to a more valuable and sustainable model.

The polymers used to produce plastics, together with the corresponding additives, should not require petroleum or other fossil fuels as feedstocks. In this context, pectin, one of the most abundant polysaccharides in nature, can be applied to produce biodegradable, biocompatible, and water-soluble films as well as coatings or interlayers in biopolymer-based structures of interest in food packaging. In this regard, Akinalan Balik et al. [1] prepared and characterized glycerol and polyethylene oxide-2000 (PEO_{2000})-containing pectin films via electrospinning and the subsequent annealing of the resultant fiber mat at temperatures below 160 °C. These newly developed pectin films were fully characterized and thereafter applied as interlayers between two external layers of poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) (PHBV). The resultant fully bio-based and biodegradable multilayer film presented a strong barrier to water and limonene vapors, revealing their promising potential in food packaging applications. Similarly, Aldas et al. [2] developed fully bio-based materials using thermoplastic starch (TPS) that was obtained from corn plasticized with water and glycerol, which was blended with up to five different pine resin derivatives as

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a kind of natural additive. The authors reported that all the gum-rosin-based additives were able to improve the thermal stability and mechanical performance of TPS, particularly in the case of the gum resins with higher amounts of carbonyl groups in their chemical structures due to their interaction with the hydroxyl groups of starch and glycerol. The developed TPS/pine resin blends were finally proposed for rigid packaging and disposable applications in hot foods, such as lids for containers and cups for beverages. Some relevant physical improvements in TPS films were also obtained by Freitas et al. [3] through the incorporation of cellulose microfibers (CMFs) obtained from rice straw (RS) waste. Fractions of 1, 3, and 5 wt% CMFs were incorporated into TPS by melt mixing, whereas starch was also subjected to dry heating (DH) modification, yielding TPS modified via dry heating (TPSDH). It was observed that both DH modification and 3 and 5 wt% fiber loadings interfered with starch gelatinization, leading to non-gelatinized starch granules in the biopolymer matrix. This represents a remarkable green physical methodology for enhancing the water barrier capacity and tensile strength of starch films. Bio-based polymers also include thermoset resins, as demonstrated by Lascano et al. [4]. In this research work, green composites were developed using partially bio-based epoxy resin reinforced with lignocellulosic particles obtained from by-products or wastes of the flax industry. Different particles of flaxseed flour, with sizes ranging between 100–220 μm and 40–140 μm, were incorporated at different concentrations, that is, 10, 20, 30, and 40 wt%, into the bio-based epoxy resin during molding and thereafter characterized. The most promising results in terms of mechanical properties and water absorption were obtained for the lowest reinforcement content and with the finest particle size, yielding highly environmentally friendly composites with a wood-like appearance and potential use in furniture or automotive sectors. Another example of the development of the biorefinery concept achieved by means of the valorization of agricultural and food wastes was presented by Ortiz-Barajas et al. [5]. In this study, the authors performed the torrefaction of coffee husk flour at 250 °C and then incorporated it into polylactide (PLA) pieces produced by injection molding. A content of 20 wt% of torrefied coffee husk flour (TCHF) successfully yielded pieces with balanced mechanical properties and improved hardness, thermal degradation, and thermomechanical resistance, which can be of great interest in in the design of compostable rigid packaging (e.g., food trays and containers), beverage cups, and food contact disposables (e.g., cutlery and plates).

Certainly, the best way to prevent the leakage of plastics into natural systems and its associated negative consequences is to support the growth of robust waste infrastructure. Indeed, plastic waste from land sources is continually flowing into world oceans via rivers due to accidents or carelessness, resulting in a high level of accumulation of plastics in the environment, the so-called "white pollution". Even though temperatures in nature are relatively low, different oligomers are often detected in the sea water and sand of coastal areas due to polymer degradation. In this regard, the study of Kimukai et al. [6] analyzed the formation of styrene oligomers (SOs) from polystyrene (PS) decomposition in the thermal range from 30 to 150 °C. In this temperature range, 2,4,6-triphenyl-1-hexene (styrene trimer, ST) was the dominant product, which was an intermediate yielding 1,3-diphenylpropane (styrene dimer, SD_1), 2,4-diphenyl-1-butene (styrene dimer, SD_2), and styrene (styrene monomer, SM). Based on a simulation, the authors also predicted that over 400 million metric tons of SOs would be present in the ocean by 2050, including those capable of thermal decompose at 30 °C. In addition, compostable and biodegradable polymers, which can disintegrate in the environment, represent a sustainable alternative to non-biodegradable polymers derived from petroleum. Compostable plastic articles can be collected and thus treated in municipal/industrial facilities, thereby capturing and delivering organic residues and diverting organic waste from landfills. More importantly, biodegradable polymers can reduce the impact of leaked plastics on the environment since some of them can also disintegrate in natural environments and thus be reincorporated into the natural cycle. This is the case of polyhydroxyalkanoates (PHAs), which are also renewable since they are synthesized by bacteria and other microorganisms (mainly using sugars). However, these and other thermoplastic biodegradable polymers usually lack sufficient physical properties to be properly applied in food packaging. In this context, Freitas et al. [7] prepared microstructures of microfibrillated cellulose (MFC) via atomization to reinforce films of PHBV, a PHA copolymer. The authors also explored the capacity of two reactive compatibilizers, namely, a multi-functional epoxy-based styrene-acrylic oligomer (ESAO) and a combination of triglycidyl isocyanurate (TGIC) with dicumyl peroxide (DCP), to improve the properties of the PHBV/MFC composites by enhancing filler–matrix adhesion. It was observed that the incorporation of MFC via reactive extrusion with TGIC and DCP led to green composite films with contact transparency, enhanced thermal stability, mechanical strength, and ductility, and high barrier performance with respect to aroma vapor and oxygen, thus unveiling new opportunities in the food packaging field. The environmental impacts of plastic packaging can also be mitigated by the use of paper in combination with biodegradable polymers, which are both derived from natural resources and can be easily composted together. Hernández-García et al. [8] proved that PHBV is an excellent material for paper-coating applications by developing PHBV/paper/PHBV multilayer sheets using heat-sealing technology. The authors observed that double coatings of PHBV successfully improved mechanical resistance and ductility, protected from moisture, and also reduced the aroma and oxygen permeances of paper, inducing a minimal effect on its optical and thermal properties and provoking a slight reduction in the aerobic biodegradation and disintegration of paper.

Finally, plastic materials can be successfully sorted and recycled by means of different strategies of waste management. While waste collection systems are evolving, novel postconsumer recycling technologies are being developed, including mechanical (secondary) recycling or reprocessing and chemical (tertiary) recycling or depolymerization–repolymerization. All these advanced recycling technologies can contribute to building an effective after-use plastics economy, but other options such as pre-consumer (primary) recycling in plastic factories and thermochemical (quaternary) recycling can also favor the development of a more sustainable plastics industry. In this regard, the research carried out by Klejnowska et al. [9] focused on determining the influence of the process temperature on the composition of gases produced during the pyrolysis of pre- and postconsumer waste pharmaceutical blisters (WPBs) in order to enhance waste valorization. Pyrolysis, a thermal decomposition process that occurs in the absence of oxygen, could effectively separate plastic from the metal fraction of WPBs in order to recover aluminum and generate gaseous fuel. The authors demonstrated that high temperatures favor the production of larger amounts of process gas rather than oil and wax. In addition, the calorific value of the gas at the maximum temperature (450 °C) was higher (21.96 MJ/m^3) for the mixed postconsumer waste than that for the clean pre-consumer material (20.14 MJ/m^3). Ascertaining the key factors of reducing costs and energy consumption during production can also represent another strategy to improve the economy and waste management of plastics. In this regard, Chen et al. [10] successfully analyzed the most influencing aspects during the extrusion-molding quality control of food-grade polypropylene (PP) packaging products. The study revealed that four key technical factors are involved: (1) extrusion sheet production; (2) extrusion line design; (3) forming and mold manufacturing; and (4) mold and thermoforming line equipment design. Moreover, according to their research results, these key factors are not only applicable to classical PP extrusion sheet and thermoforming production but are also related to processes of extrusion and thermoforming techniques used for other plastic products, such as expanded polypropylene (EPP) and PLA sheets. These results are considered to be capable of providing a key technical reference with which enterprises can improve quality in order to enhance the competitiveness of products, reduce production costs, and achieve sustainable development, energy savings, and carbon reductions.

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