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Editorial

Alternative Building Blocks and New Recycling Routes for Polymers: Challenges for Circular Economy and Triggers for Innovations

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Circularity often seems to be synonym of recycling and valorization of waste (Massardier-Nageotte 2014; Massardier and Quitadamo 2018), but associating alternative building blocks routes for the eco-design of polymer materials (Hachemi, Belhaneche-Bensemra, Massardier, 2014; Boubekour, Belhaneche-Bensemra, Massardier, 2015; Delamarche, Massardier, et al. 2020; Boubekour, Belhaneche-Bensemra, Massardier, 2020) made from biomass can be a relevant challenge. The present research topic gives a few answers relative to valorization of PLA (polylactic acid) based materials through (bio)degradation as well as to potential alternative routes to produce building blocks for sustainable polymer materials, compatible with mechanical and chemical recycling, considering not only technological but also socio-economical points of view.

Recent families of polymers, such as (bio)degradable PLA, made of starch and able to be processed again in a perspective of mechanical recycling, could satisfy criteria of circularity from a technological point of view (Payne, McKeown, and Jones 2019; Cosate de Andrade et al. 2016; Bouti, Irinislimane and Belhaneche-Bensemra, 2022). Unfortunately, PLA still corresponds only to a few percent of the polymer world production, which compromises the development of recycling chains for it, when considering socio-economical aspects. Biobased polymers, such as PLA, are in early stages of development and production and suffer from a lack of learning and economies of scale that can trigger their development. This new trajectory, which currently represents only 1% of the world market, is a niche dedicated to specific needs and applications (European Bioplastics 2021). Main challenges reside both in the improvement of technological innovations and their organizational adaptation in the current socio technological system, (collecting, sorting, and recycling). This novelty

is also perceived as a disturbing element for recyclers and as a new deal for consumers who do not know in which type of bin to put these new “items” (Ansink, Wijk, and Zuidmeer 2022).

In this context, why not elaborating conventional polymers highly recycled in existing chains, such as PET (Polyethylene terephthalate), from building blocks obtained by alternative routes? In the present research topic, the short review of Sandei, that includes socio-economical points of view, potentials and limits, deals with this question (Sandeï, Massardier, and Brunel 2022). Nowadays, PET is synthesized from ethylene glycol and terephthalic acid, which can represent a double challenge. Thankfully, bio-based ethylene glycol is already produced with bio-ethanol but represents only 20 % of the carbon of PET. The 80 % remaining part come from terephthalic acid which still remains petroleum-based. Even if discoveries are still at laboratory or pilot scale, some alternative routes for building block production are emerging at industrial scale with still small quantities produced. One of the most notable alternatives is the synthesis of p-xylene, necessary to synthesize terephthalic acid, from isobutanol. The technology to produce isobutanol from glucose is well known since it is used as biofuel but not established yet to produce building blocks for polymers. Transposition to another sector can be not so easy but one of the main problems associated with the valorization of biomass for producing PET seems to be the access to resources that would imply local models adapted to the resources of the territory. According to them, ethylene *via* second generation bio-ethanol, based on green waste seems to be relevant for producing the ethylene glycol monomer implied in the synthesis of PET. For terephthalic acid monomer, p-xylene seems the priority synthon of interest, with a production from isobutanol to be favored, basing on the technological aspect, at least. Considering a more local scale, synthesis of terephthalic acid from p-cymene and terpenes is also an interesting alternative for using citrus wastes from food industry, which can be considered as a good example of industrial ecology.

If mechanical or chemical recycling often seem the end-of-life to be privileged, for some applications, such as agriculture welsh films, ability for biodegradation seems to be a relevant property to achieve. Two research articles of the present research topic deal with the formulation and processing of PLA based materials associating use properties and ability for biodegradation in defined scenarii.

The first article deals with the «impact of Ionic Liquids on the (bio)degradability of Poly(butylene succinate)/Poly(lactic acid) blends» (Delamarche et al. 2022) and follows a previous paper dealing with the influence of a deep eutectic solvent on the (bio)degradability of those blends (Delamarche, Mattlet, et al. 2020). Both properties and degradation of a single polymer seem to be rather difficult to adjust, whereas polymer blends offer the possibility to tune both properties and stability. The study shows that elaborating multiphase polymer materials enables to combine properties of both rigid PLA and PBS (Polybutylene succinate) for which degradation processes are preferentially located in a “predegraded” dispersed phase, can be a way to tailor macroscopic (bio)degradation. In the study, morphologies, mechanical properties as well as (bio)degradability were adjusted by adding small amounts of ionic liquids. Degradation experiments carried out at 58°C show that the formulations degrading faster correspond to the blends having undergone the most important degradation of the PLA dispersed phase during processing, with decrease of melting temperature, of weight percentage of PLA associated with fragmentation. Finally, the work shows that ionic liquids can be used to tune mechanical properties of polymer blends as well as their (bio)degradability.

The second article deals with the « impact of microcrystalline cellulose extracted from walnut and apricots shells on the biodegradability of Poly (lactic acid) » (Mahmoud, Belhaneche-Bensemra, and Safidine 2022) and follows a previous paper dealing with the extraction and characterization of microcrystalline cellulose (MCC) from walnut and apricot shells (WS and AS) as agricultural wastes in order to use it as reinforcement in polymer composites (Mahmoud, Belhaneche-Bensemra, and Safidine 2021). To the best of our knowledge, there is no single report made in literature on the use of MCC extracted from walnut (WC) or apricot shells (AC) for the preparation of PLA composites. Various microcrystalline celluloses, extracted from apricots and walnut shells by alkaline treatments combined with hydrogen peroxide blanching, were blended with PLA, to make composites such as ones containing 7% AC and referred to as PLA/7% AC and PLA/7% WC. PLA blended with microcrystalline cellulose from apricot shells displays an increase in Young's modulus from 802.6 MPa for neat PLA to 1145.6 MPa for PLA composites containing 7% WC (PLA/7% WC). Then, the biodegradation was evaluated by a soil burial test and displayed weights losses of 14% for PLA, 38% for PLA/7% WC, 13% for PLA/7% AC after 12 months within soil burial, the best compromise between mechanical properties, crystalline structure and biodegradability rate being achieved for PLA composites containing 7% WC.

Finally, the research topic treats of various and complementary aspects of circularity for polymer. It explains the main challenges for the development of new trajectories and the phase of exploration between various technological options. It shows why the combination of quite new (PLA) and older options (such as PET), potentially bio-based, can be also considered in a transition phase (Quitadamo, Massardier, and Valente 2017) before the development of fully bio sourced polymers at a larger scale. Thus, circular economy encounters many opportunities and challenges at this early stage. The critical issue for PLA resides not only at a technological level but also at the organizational level (waste management) for going beyond a niche market.

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