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Novel bio-based polymers and biomolecules for sustainable packaging developments

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Abstract:

Only 31% of plastic is currently recycled and plastic packaging still have a deficient end of life. Thus, improvements are needed to provide cost effective solutions with high bio-based contents and suitable performances for demanding packaging applications, while still achieving compostability in mild conditions.

In this sense, **BIOnTOP** project, funded by the Bio Based Industries Joint Undertaking under the European Union's Horizon 2020 research and innovation program under grant agreement No 837761, aims to deliver recyclable-by-design cost competitive packaging solutions that can be mechanically recycled, industrially/home composted or even suitable for anaerobic digestion using sustainably sourced comonomers, additives and fillers to formulate novel PLA copolymers and compounds. The barrier properties of delivered bio-packaging trays, films and derived packaging, will be enhanced using removable protein-based coatings and a novel fatty acid grafting technology to decrease permeability and compete with fossil packaging. Moreover, in the field of textile packaging, most used coatings are not bio-based and of different nature from the coated fibers, making organic recycling extremely difficult, so new PLA coatings or fatty acid grafted PLA will allow reprocessing without significant loss of properties. In summary, the main goal of BIOnTOP is to deliver novel bio-based biodegradable packaging based on versatile copolymers and coatings that optimally preserve the packed products but also our resources (recyclable packaging based on significantly >85% renewable resources, partly produced from by-product biomass). The demonstrators that will be produced are the following:
• Recyclable, home-compostable monomaterial trays & films for F&V (fruit and vegetables).
• Recyclable, multilayer trays & films compatible with MAP (modified atmosphere packaging), e.g. dairy and personal care products.
• Home compostable & organically recyclable nets for F&V.
• Home compostable & organically recyclable coated textiles.
• Recyclable, reusable coated woven fabrics, e.g. food wraps.
• Recyclable, reusable secondary packaging from SRM (secondary raw material): extruded blown bags and non-woven bags.

A related project is **PERCAL**, funded also by the Bio Based Industries Joint Undertaking under the European Union's Horizon 2020 research and innovation program under grant agreement No 745828, which aims to obtain high added value chemical building blocks and products through the valorization of the organic fraction of municipal solid waste (OFMSW). Thus, among other products, PERCAL is producing PLA based hot melt adhesives using a cascade approach. In the EU over 100 million tons of biowaste are thrown away each year. Currently, 75% of this biowaste is landfilled or incinerated, causing major environmental problems, since biowaste produces greenhouse gases when it decomposes and contaminates soil and groundwater. Furthermore, landfilling of biowaste is against the principle of a circular economy and is a waste of nutrients, energy and potential resource for biobased products. For that reason, PERCAL proposes the development of innovative solutions to transform OFMSW into high value-added products and complementary routes to the bioethanol (current PERSEO technology) production are being developed. The three main pathways are focused on the one side, on lactic acid to produce eco-friendly ethyl lactate solvents by reactive distillation from lactic acid & bio-ethanol to be used in cleaning products and

inks, and PLA hot melt adhesives which have showed good adhesion and lap shear strength similar of above standard EVA based ones for cardboard and other non-food applications in combination with maleic anhydride by reactive extrusion. On the other side, on succinic acid as an intermediate building blocks to production of polyols for the polyurethane industry. And finally, on the production of biosurfactants by chemical and/or microbiological modification of protein and lipid fraction from remaining fraction of MSW fermentation, for its use in cleaning products.

Addressing the goal of reducing the use of multimaterials to provide the functionality and barrier properties desired for a specific packaging product, as BIOnTOP does, **REFUCOAT** project, funded by the Bio-Based Industries Joint Undertaking under the European Union's Horizon 2020 Research and Innovation programme, works on this line. Currently, most barrier containers are based on co-extruded or laminated multilayer structures since there is not a single material that has all the necessary properties. The main drawback of these multilayer structures is their high carbon footprint that must be reduced to make more sustainable products. The main challenge for sustainable materials is that they do not have the necessary properties of food protection, so it is necessary to introduce new alternatives to increase the product self-life. In REFUCOAT project, totally recyclable and biodegradable materials and functional coatings with antioxidant, antimicrobial and oxygen and water vapor barrier properties have been developed. The final objective of REFUCOAT project is to develop different packages for chicken meat, cereals and savory products. These final packages will have better mechanical, thermal and barrier properties as the current metallised and non-bio-based alternatives. The biodegradable polymers developed in this project are middle chain polyhydroxyalkanoate (PHA) which shows a low moisture permeability, making them a good alternative for fresh meat packaging, and PGA (polyglycolic acid) synthetized through a more efficient, ecological and economic process which will be applied as barrier coatings, due to its very low permeability to O_2 and CO_2 as well as largely insensitiveness to humidity conditions, making it ideally suited for beverage and perishable food packaging applications. In addition, hybrid polymers will be produced from it, through its reaction with silanes, to obtain high barrier biodegradable coatings that will be applied by conventional printing methods and not by expensive methods such as chemical vapor deposition as well as hybrid coatings with unexpected opportunities for new innovative surfaces using sol-gel approaches which have many advantages associated to due to the use of mild and low-temperature conditions. Active packaging including antioxidant and/or antimicrobial substances further contributes to the over-ridding goal of the REFUCOAT project to provide gas barrier layer based on renewable resources for food preservation.

Emerging active packaging solutions for food preservation have also been addressed in **DAFIA** project, funded by European Union's Horizon 2020 research and innovation program under grant agreement n° 720770. Thus, marine rest raw materials (MRRM) such as salmon skin and backbones are harnessed to develop cost-efficient isolated and purified gelatin and hydrolysates. Gelatin is a denatured polypeptide extracted by hydrolysis from pre-treated collagen sources, mainly animal skins and bones, which is mostly affected by its amino acid composition and molecular weight distribution. The fractionation and lipids separation pre-treatment protocols have been optimized in mild conditions and the bio-based coating has been formulated comparing different natural plasticizers, pH and temperature as well as drying conditions. Furthermore, the mechanically separated salmon muscle has been hydrolysed with proteases as a potential antioxidant and antimicrobial actives. The addition of hydrolysates has shown positive antioxidant activity. The coating application and lamination have been successfully carried out at the pilot scale resulting in laminates with promising oxygen barrier. This shows that bio-macromolecules from marine sub-products have a great potential to be used as high added value active barrier coatings for multilayer packaging or edible coatings directly applied to food.
Other biomacromolecules and bioplastics for high added value applications have been developed in DAFIA project. Thus, different technical and economic conversion routes of MSW and MRRM have been explored in order to obtain high added value products, such as flame retardants and chemical building blocks (dicarboxylic acids and diamine) to produce biopolyamides for a wide range of industrial applications. The new halogen-free flame retardant (FR) is based on nucleic-acid, gelatine (from fish waste) and lignosulfonates (as P and N source). Vertical flammability tests of the bio-based containing 20-30% dose rate V0. The development of a robust microbial strain with high productivities by fermentation of pre-treated MSW has produced the targeted chemical building blocks (diacids and diamines), which are being used for the synthesis of different biopolyamides with a bio-content close to 100%. At the end of the project, DAFIA will show the viability to provide added value to the waste at pilot plant scale.