This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

Document Information

Deliverable number	**D1.2 Report on market survey**
Type of deliverable	**Report**
Associated WP	**WP1 Feedstock selection, Biomolecules extraction and functionalization**
Associated Task	**Task 1.1. Updated Inventory of providers and acquisition of raw materials, considering availability, cost and sustainability of feedstock**
Dissemination level	**PU**
Due date for deliverable	**31-07-2019 (M3)**
Actual submission date	**04-03-2021 (M23) Revised file**
Lead beneficiary	**INSTM**

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

Document Control page

Author	**Patrizia Cinelli (INSTM), Maria Beatrice Coltelli (UNIPI), Andreas Staebler (FRAUHOFER), contributes from all partners.**
Version number	**1.0**
Date	**29-07-2019**
Modified by	**[Patrizia Cinelli / patrizia.cinelli@unipi.it]**
Comments	**[Revised as indicated in review report]**
Status	✓ **Draft** ☐ **Accepted**
Action requested	**Deadline for action: 04-03-2021**

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

Executive Summary

The overall objective of project ECOFUNCO is to select, extract, functionalise molecules (proteins, polysaccharides, cutin, polyphenols, carotenoids, fatty acids) from highly available, low valorised biomass such as tomato peels and seeds, melon seeds, legumes, sunflower press cake etc for the development of new bio-based coating materials to be applied on two different substrates respectively cellulosic and plastic based. These coatings will provide improved performances compared to currently available products and at the same time have more sustainable end of life options. The present document addresses the objectives of WP1 and in particular of Task 1.1. "Updated Inventory of providers and acquisition of raw materials, considering availability, cost and sustainability of feedstock".

ECOFUNCO products to be developed in the project are in particular protein-based coatings combining barrier and adhesive properties, antimicrobial-antioxidant coatings based on chitin nano-fibrils, and/or chitosan, microcrystalline cellulose (MC) to be used as primer on both plastic and cellulose substrates as well as medium chain polyhydroxyalkanaotes produced in the project. The coatings will be applied on cellulose tissues (personal care), paper and cardboard (packaging for fresh products like pasta, tableware), woven and nonwoven (sanitary products) and plastic substrates (bio-polyesters) for active packaging. Thus in the project advanced, sustainable, innovative technologies for extraction of functional molecules from agro-food biomass is also promoted, with a green chemistry approach.

Thereby cutin-based formulations will provide water repellent properties (paper cups, service paper etc.), water vapour barrier (packaging) and protective properties (non-food packaging). The protein-based coating will be used as barrier adhesive for multilayer food packaging (bio polyesters based) thus combining adhesive properties and an oxygen barrier in one single layer. The chitin coatings will, in combination with functionalised microcrystalline cellulose enhance the shelf life of food due to their antimicrobial properties. Due to their biodegradability response, and the possibility to easily remove the coatings from the substrate all materials will enhance the end of life options (composting, recyclability) of the structures.

The ECOFUNCO project aims to deliver a number of impacts, such as:

Create a number of new cross-sector interconnections within the bio-based economy between different sectors such as the agro-food industry and the coatings and biopolymer sectors. This in turn will accelerate the development of several new value chains and will:

Set the basis for new value chains; the ECOFUNCO process technologies will enable new product applications, deriving from food and agro-waste biomass as raw materials. The new value chains will be based on applying the bio-based coatings to a range of so-called Fast-Moving Consumer Goods.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

Develop three new types of bio-based materials in the form of coatings: an antimicrobial coating for sanitary tissues and food packaging, a water-repellent coating for food containers and utensils and a barrier adhesive for multi-layered packaging.

Reduce the environmental impact of packaging and increase sustainability by replacing the current non-renewable coatings with renewable ones. Sustainable sourcing of the raw materials will further contribute to these benefits.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

Glossary

AFH	Away from home
CNF	Chitin nano-fibres
CNs	Chitin nano-fibrils
EVOH	Ethylene vinyl alcohol
HIS	Hyper spectral image
mcl-PHAs	Medium chain length polyhydroxyalkanoates
PE	Polyethylene
PET	Polyethylene terephthalate
P(3HB)	Poly(3-hydroxybutyrate)
PPHBV	Poly(3-hydroxybutyrate-*co*-valerate)
PHAs	Polyhydroxyalkanoates
PVdC	Polyvinylidene chloride
SEM	Scanning electron microscopy
TEMPO	(2,2,6,6-tetramethylpiperidine-1-oxyl radical) mediated oxidation
Tg	Glass transition temperature
WP	Work Package

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

Table of contents

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

1 INTRODUCTION

The overall objective of ECOFUNCO project is the selection, extraction, functionalization of biomolecules (proteins, polysaccharides, cutin, etc) from highly available, low price and underutilized biomass such as tomato processing residues, legumes, sunflower press cakes, etc. to develop new bio-based coating materials to be applied on two different substrates (cellulosic and plastic based), with improved performances compared to currently available products and at the same time with more sustainable end of life options.

Cellulose is the most available renewable material on Earth and it would be beneficial for environment to use it in consumer products, such as in personal care and disposable products for food (tableware), where hardly recyclable petro-based plastics are currently used. The limits to be overcome are the hydrophilic and low barrier properties typical of a non-woven fibrous system. For these reasons versions of cellulose combined with poly(ethylene) are widely present on the market, and petro-based version of these products are currently preferred, despite of their negative environmental impact.

In the last decade effort was dedicated by researchers to investigate and formulate new bioplastics, but their effective presence in the market, which is increasing, needs to be promoted in a wide spectrum of applications and petro-based plastics should be more and more replaced with renewable counterparts mainly employing bioplastics and natural polymers.

Coating of cellulose or bioplastics with proper functional coatings, based on biopolymer and functional materials coming from agro-food waste streams, will allow improving their performances allowing them for replacing effectively petrochemical products in the personal care, tableware and food packaging sectors.

To achieve these challenging objectives the project will build on previous research conducted by the participating, on respectively cutin as hydrophobic-water and grease repellent coating, polysaccharides such as chitosan-chitin as antimicrobial coating, and proteins as gas barrier coating and adhesive.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

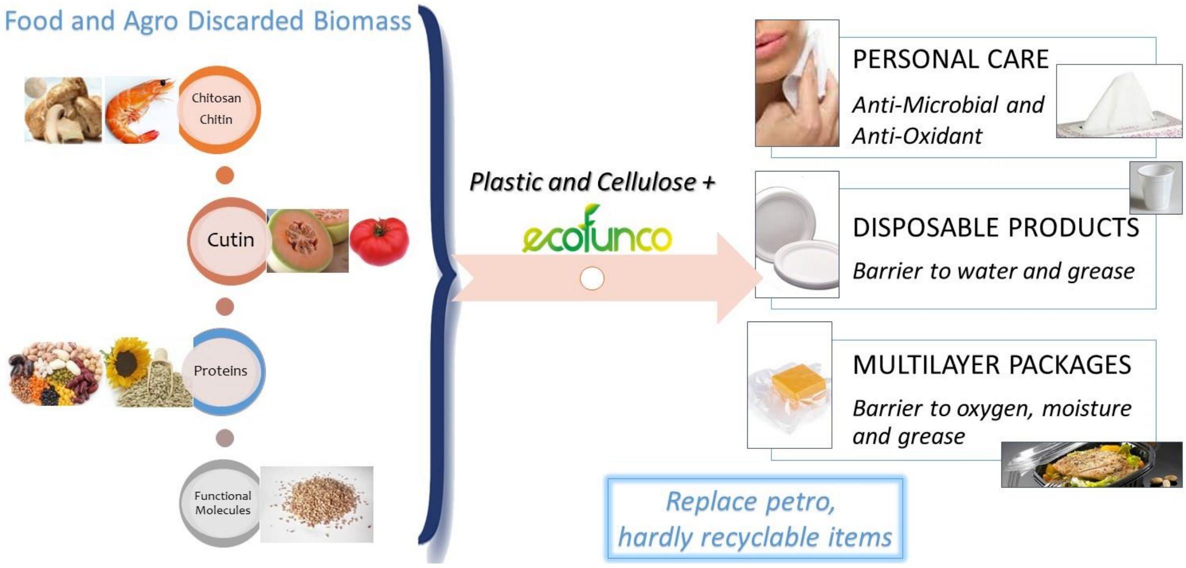

Figure 1. ECOFUNCO scheme

1.1. Description of the document and pursue

In the present document the state of art on materials that are addressed in the ECOFUNCO project is being reported and updated. The literature and market search have been analysed critically with particular focus on the objectives of the ECOFUNCO project that are those of obtaining sustainable, bio-based, biodegradable coatings to be applied respectively on plastic and cellulose substrates to confer properties of barrier to oxygen and water vapour, hydrophobicity and valuable antioxidant or antimicrobial properties. The target applications of the coated materials range from packaging to health care products.

This document will be the source for review papers and review chapters to be published in open access journals such as express polymers letter, coating, materials etc since the content of this public document refers to state of art, published papers, market analysis, published patents etc thus no confidential information from ECOFUNCO project will be reported here.

1.2. WPs and Tasks related with the deliverable

The present deliverable refers particularly to Task 1.1. "Updated Inventory of providers and acquisition of raw materials, considering availability, cost and sustainability of feedstock" (Month1-Month3). The present deliverable is related to all WP1 Tasks, Task 1.2 "Optimization of production processes by biotechnology"; Task 1.3 "Optimization of Cutin extraction (M3-M15); Task 1.4. Extraction process of

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

biomolecules from tomato seeds"; Task 1.5 "Production of chitin nanofibers" as well as to all the following ECOFUNCO WPs.

2 ADVANCES IN USE OF RENEWABLE SOURCES TO OBTAIN POLYMERS AND MOLECULES FOR BIOCOATING

2.1 State of the art on availability and costs of feedstocks for protein extraction

Protein rich by-products are abundantly available from agro-food industry. The most important ones are press cakes from vegetable oil extraction. In 2016 almost 30 Mio tons of suchlike by-products have been produced in the EU with sunflower press cake being the third most common one (3.7 Million tons)[1]. Currently these press cakes are used as animal feed mostly for cattle.

Due to the comparable low nutritional value, in particular low energy and lysine content as well as its high fibre content sunflower meal can only be used as partial replacement for other protein meals[2,3], therefore prices for sunflower meal (~0.20€/kg) are significantly lower than for other press cakes such as rape seed (~0.28€/kg) and soy bean (~0.33€/kg)[4]. Due to its limited use as animal feed and its comparatively low price, sunflower meal is thus an ideal source for proteins to be used in ECOFUNCO materials.

Legumes are in general rich in proteins and have the ability to fix atmospheric nitrogen in the soil thus improving soil structure and soil fertility for subsequent crops[5], therefore legumes are a promising crop to reduce fertilizer use in European agriculture.

Many legumes such as beans and lentils have been a mayor protein source in traditional European diets. There are several non-edible legumes such as bitter lupines (e.g. *lupinus luteus)* that can be cultivated as catch crop to improve soil quality. As these crops are currently only used for green manure the protein rich seeds are not harvested and valorised to a large extent, therefore these crops are an ideal protein source for ECOFUNCO project.

[1] FEDIOL 2016 annual statistics, www.fediol.eu

[2] Nassiri Moghaddam, Hassan, et al. "Evaluation of the nutritional value of sunflower meal and its effect on performance, digestive enzyme activity, organ weight, and histological alterations of the intestinal villi of broiler chickens." Journal of Applied Poultry Research 21.2 (2012): 293-304.

[3] https://www.sunflowernsa.com/wholeseed/sunflower-as-a-feed/

[4] https://www.clal.it/en/?section=conf_semi, access 18.07.2019

[5] http://www.pgro.org/downloads/LEGATO-POSTER-FINAL-2014.pdf

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

At the moment legumes cultivation in Europe is low and only 1.5% of the arable land is used for these crops[6], however this year the European Parliament has adopted a strategy for the promotion of protein crops. By adding value to these crops ECOFUNCO can contribute to the success of this strategy[7].

Several processes can be applied to extract proteins from raw materials such as seeds and press cakes. The most common ones are to solubilize the protein by adding low concentrations of salt (salting in) or by adding a base to adjust an alkaline pH value. After this treatment the solid residue, mostly fibres are removed by centrifugation or filtration. The proteins are then obtained from the supernatant either by increasing the salt concentration (salting out), by isoelectric precipitation, by ultrafiltration or by combinations thereof[8,9,10,11]. With these processes usually result in protein isolates (protein content >90%dm) with most of the proteins being present in their native structure.

In ECOFUNCO however a different approach will be followed. To obtain the so-called micellar proteins, the protein will be extracted from the raw material by applying the salting-in methodology and thus by using low salt concentrations. [12,13,14] Precipitation after removal of the fibre fraction will be conducted by drastic reduction of the ionic strength in the protein solution (dilution precipitation).

---

[6] Watson, Christine A., et al. "Grain legume production and use in European agricultural systems." Advances in Agronomy. Vol. 144. Academic Press, 2017. 235-303.

[7]http://www.europarl.europa.eu/sides/getDoc.do?pubRef=-%2F%2FEP%2F%2FTEXT%2BREPORT%2BA8-2018-0121%2B0%2BDOC%2BXML%2BV0%2F%2FEN&language=EN

[8] C. Pickardt, S. Neidhart, C. Griesbach, M. Dube, U. Knauf, D. R. Kammerer, et al., "Optimisation of mild-acidic protein extraction from defatted sunflower (Helianthus annuus L.) meal," Food Hydrocolloids, vol. 23, pp. 1966-1973, 10// 2009.

[9] K. Mueller, P. Eisner, Y. Yoshie-Stark, R. Nakada, and E. Kirchhoff, "Functional properties and chemical composition of fractionated brown and yellow linseed meal (< i> Linum usitatissimum</i> L.)," Journal of food engineering, vol. 98, pp. 453-460, 2010.

[10] L. Jervis and W. Pierpoint, "Purification technologies for plant proteins," *Journal of Biotechnology,* vol. 11, pp. 161-198, 1989.

[11] N. A. Deak, P. A. Murphy, and L. A. Johnson, "Effects of NaCl Concentration on Salting-in and Dilution During Salting-out on Soy Protein Fractionation," *Journal of Food Science,* vol. 71, pp. C247-C254, 2006.

[12] T. A. El-Adawy, E. H. Rahma, A. A. El-Bedawey, and A. F. Gafar, "Nutritional potential and functional properties of sweet and bitter lupin seed protein isolates," Food Chemistry, vol. 74, pp. 455-462, Sep 2001.

[13] M. A. H. Ismond, E. D. Murray, and S. D. Arntfield, " The role of non-covalent forces in micelle formation by vicilin from Vicia Faba.- The effect pf pH variations on protein inter actions," Food Chemistry, vol. 20, pp. 305-318, 1986.

[14] M. A. H. Ismond, E. D. Murray, and S. D. Arntfield, "The role of non-covalent forces in micelle formation by vicilin from Vicia Faba.2. The effect of stabilising and destabilising anions and protein inter actions," Food Chemistry, vol. 21, pp. 27-46, 1986.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

This dilution precipitation causes an alteration in the tertiary structure of the proteins and hydrophobic functional groups of the amino acids usually hidden inside the protein become exposed on the surface [15].

Because of high electrostatic attraction of these hydrophilic amino acids micellar aggregates are formed (Figure 2)[16].

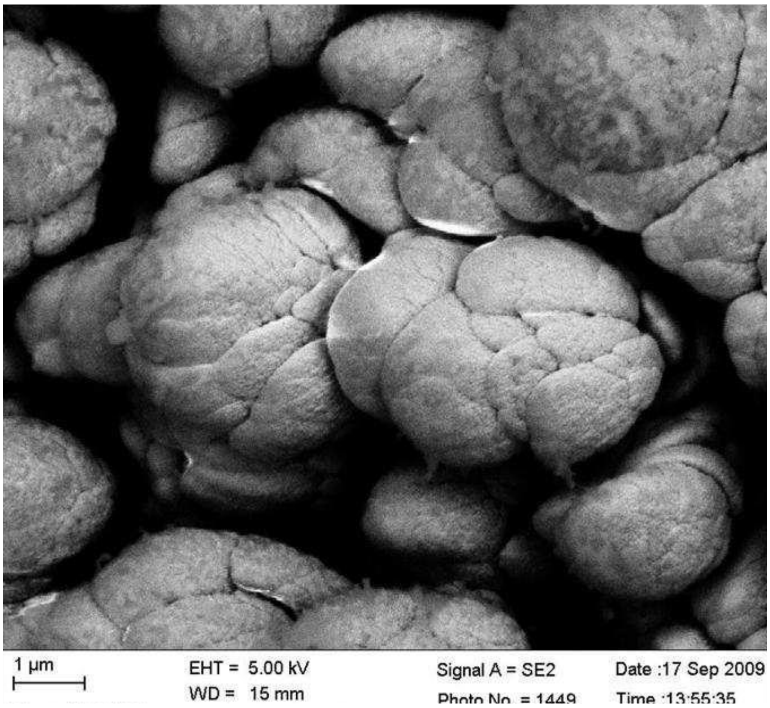

Figure 2: Scanning electron microscopy (SEM) image of Micellar lupin proteins

This change in the protein structure also has an effect the protein properties. Thus, micelle proteins show a low water solubility, a fat-like structure, a strongly structure-viscous behavior as well as a high

[15] M. Y. Cordero-de-los-Santos, J. A. Osuna-Castro, A. Borodanenko, and O. Paredes-Lopez, "Physicochemical and functional characterisation of amaranth (Amaranthus hypochondriacus) protein isolates obtained by isoelectric precipitation and micellisation," Food Science and Technology International, vol. 11, pp. 269-280, Aug 2005.

[16] I. S. Muranyi, C. Otto, C. Pickardt, P. Koehler, and U. Schweiggert-Weisz, "Microscopic characterisation and composition of proteins from lupin seed (< i> Lupinus angustifolius</i> L.) as affected by the isolation procedure," Food Research International, vol. 54, pp. 1419-1429, 2013.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

stickiness[17,18]. Their applicability as a coating in paper based multilayer structures providing both a high adhesive and thus a good bond strength as well as good barrier properties against oxygen has already been proven[19].

2.2 State if the art in availability of Chitosan and Chitin

With an annual production of approximately 1010 – 1020 tons, chitin is considered as one of the most abundant biopolymers on the Earth. Chitin can be present in several materials (Figure 3) belonging to exoskeleton of insects (e.g. tubeworms), arthropods (e.g. mollusc, shell oysters, squid pen) and crustaceans ((e.g., crabs, lobsters and shrimps). Moreover, chitin is also known as a structural component of cell walls of fungi (mushrooms).

Chitin can be present in several materials belonging to insects, molluscs, crustaceans and fungi, hence it is possible to consider several sources, as recently evidenced by Yadav et al.[20]

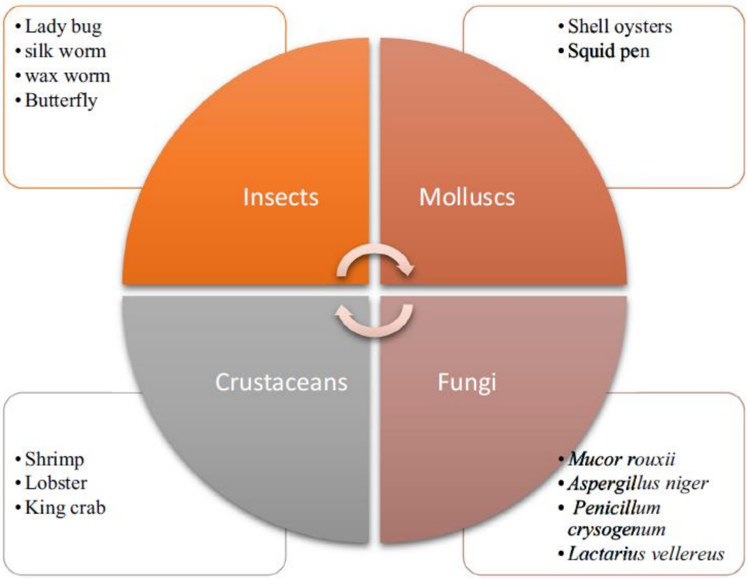

Figure 3: Sources of chitin [by ref 8]

[17] D. Sussmann, T. Halter, C. Pickardt, U. Schweiggert-Weisz, and P. Eisner, "An Optimization Approach for the Production of Fatlike Protein Isolates from Different Leguminous Seeds Using Response Surface Methodology," Journal of Food Process Engineering, pp. n/a-n/a, 2013.

[18] D. Sussmann, C. Pickardt, U. Schweiggert-Weisz, and P. Eisner, "Sensory evaluation of food products containing lupin protein isolate as innovative fat replacer," EFFoST–European Federation of Food Science & Technology, pp. 10-12, 2010.

[19] Eibl, I., von der Haar, D., Jesdinszki, M., Stäbler, A., Schmid, M., & Langowski, H. C. (2018). Adhesive based on micellar lupin protein isolate exhibiting oxygen barrier properties. Journal of Applied Polymer Science, 135(25), 46383.

[20] Yadav M, Goswami P, Paritosh K, Kumar M, Pareek N, Vivekanad V, seafood waste: a source for preparation of commercial employable chitin/chitosan materials

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

Pure chitin is obtained as powder or flakes by a three-step extraction method comprising an alkaline extraction for the removal of proteins (deproteinization) and an acidic treatment to remove calcium carbonate (demineralization) followed by a final decolorization step[21,22,23]. Depending on the orientation of the micro-fibrils, 3 different allomorphic forms of chitin are distinguished: alpha-chitin, beta-chitin and gamma-chitin. α-chitin with the antiparallel configuration is the most abundant form and it can be found in krill, lobsters, shrimps and crabs. β-chitin with the parallel configuration is less common and[24] found in squid pens and tubeworms. ϒ-chitin is a mixture of both mentioned forms
Currently the chitin coming from sea- food waste is widely available and was mainly considered up to now as raw material source.

Chitosan, the deacetylated form of chitin, is soluble in diluted acidic solutions (with degree of deacetylation over 50%). Deacetylation of chitin is typically realized using 40 – 50% NaOH at 120 – 150 °C. However, deacetylation is almost never complete. In fact, chitin is considered as chitosan with a minimum degree of deacetylation of 70% (or with degree of acetylation between 0.05 – 0.3).

Chitosan and chitin nano-fibrils can be produced by the same process based on the deacetilation of raw chitin coming from shrimp shells, as explained by Kumar et al.[25] and Van Toan et al[26] (Figure 4). The formation of chitin nano-fibrils can be obtained by controlling the deacetylation step, thus avoiding the full conversion to chitosan. In any case, the chitin nano-fibrils represent the crystalline part of chitin. The amorphous part of chitin is transformed in any case in chitosan by deacetylation.
MAVI SUD is an Italian company that patented a methodology to obtain chitin nano-fibrils (CNs) in water suspension thanks to an acidic process that resulted in the achievement of CNs with a high content of -NH_2 groups on the surface[27].

Chitin nano-fibrils can be considered a reference for the ECOFUNCO project, because it was demonstrated to have an anti-microbial activity of cellulosic board in the framework of the

[21] D. Elieh-Ali-Komi, M. R. Hamblin, Int. J. Adv. Res. 2016.
[22] Nguyen Van Toan, Production of Chitin and Chitosan from Partially Autolyzed Shrimp Shell Materials, the Open Biomaterials Journal, 2009, 1: 21-24
[23] Elieh-Ali-Komi, Daniel, and Michael R. Hamblin. "Chitin and chitosan: production and application of versatile biomedical nanomaterials." International journal of advanced research 4.3 (2016): 411.
[24] S. Ifuku, in *Handb. Polym. Nanocomposites. Process. Perform. Appl. Vol. C Polym. Nanocomposites Cellul. Nanoparticles*, 2015.
[25] MNV Ravi Kumar, A review of chitin and chitosan applications, Reactive and Functional Polymers, 46, 1, 2000, 1-27
[26] Nguyen Van Toan, Production of Chitin and Chitosan from Partially Autolyzed Shrimp Shell Materials, the Open Biomaterials Journal, 2009, 1: 21-24
[27] (WO2006048829) Preparation of chitin and derivates thereof for cosmetic and therapeutic use.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

INNOCARTOPACK project. The results of this study were recently described by Panariello et all[28]. The treatment of cellulosic board with chitin nano-fibrils or with chitosan was effective in increasing the shelf life of fresh pasta packed in treated trays.

The work of Cinelli et all [29] on the basis of Life Cycle Assessment study, outlined that the process to obtain CNs starting from chitin and using acidic reagents has a high impact related to the use of large amount of water and chemical solvents. This aspect would deserve to be considered in further research, optimizing more sustainable processes to obtain CNs.

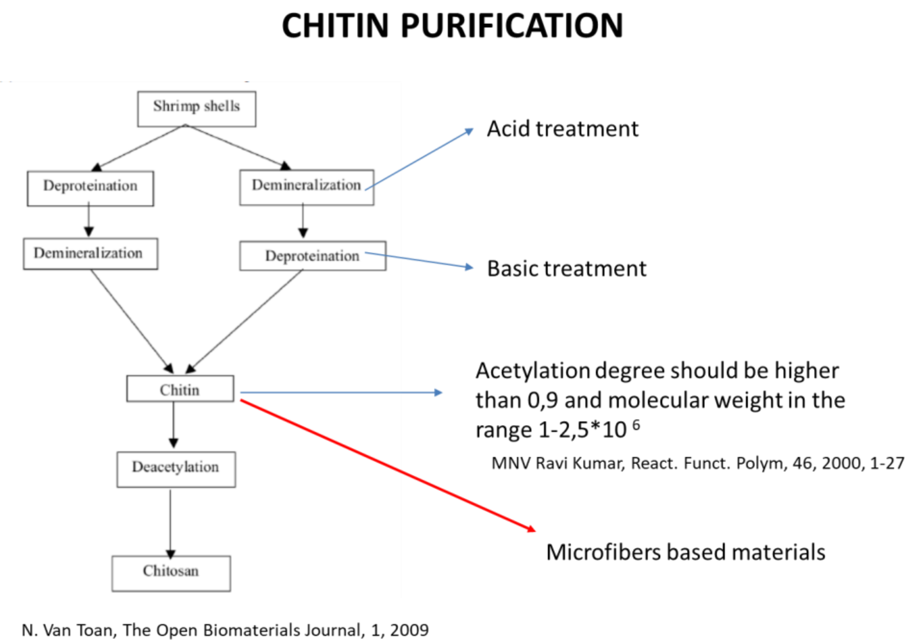

Figure 4: Scheme of the general industrial purification scheme adopted for purifying chitin and chitosan

One of the most common chemical–mechanical approach is to modify the chitin fibre surface via carboxylation reactions to introduce negative charges onto the micro-fibril surface allowing easy separation; the modified fibres are subjected to mild to intensive mechanical treatments to liberate

[28] Panariello L, Coltelli MB, Buchignani M, Lazzeri A, Chitosan and nano-structured chitin for biobased anti-microbial treatments onto cellulose based materials, Eur. Polym. Jour., 113, 2019, DOI: 10.1016/j.eurpolymj.2019.02.004

[29] Cinelli P, Coltelli MB, Mallegni N, Morganti P, Lazzeri A, Degradability and Sustainability of Nanocomposites Based on Polylactic Acid and Chitin Nano Fibrils, Chem. Eng. Trans, 60, 2017, ISBN 978-88-95608- 50-1; ISSN 2283-9216

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

CN's from the raw materials [30]. It was shown that this process had the lowest environmental impact compared to the other processes for energy and global warming potential (based on LCA study). These CNs will be considered too within ECOFUNCO project.

2.3 State of the art in availability of Cutin

Cutin is present in tomato peels (Table 1) and in other vegetables such as watermelon/melon and apple peels. All of these by-products too are largely available in EU where 3.21 million tons of watermelons and 1.77 million tons of melons are produced (FAOSTAT 2017 data) per year; these crops lead to large amount of wastes, besides about 20% of watermelons and 9% of melons remains in fields. EU production of apple has reached 10.1 million tons in 2017 (FAOSTAT 2017 data); the solid waste, discarded after the industrial processes, represents 20-35% of the fresh weight of the apple fruit, about 12.00 million tons worldwide. Tomato production in the EU-28 amounted to 18 million tons in 2017 (FAOSTAT 2017 data), of these 18 million tons, 10% are lost as they remains in the field while, on the processed tomato, the percentage of waste is included between 10 and 30%, in particularly the 2% is made up of skins and seeds.

Table 1. Fraction and component content of tomato pomace by-product[31].

Pomace Fraction	Content (%, w/w)	Components (*)	Calculated Content in Pomace (%, w/w)
Fiber	15	Polysaccharides (cellulose, hemicelluloses) and lignin (100%)	15
Seed	55	Polysaccharides (cellulose) (19%)	10
		Proteins (29%)	16
		Lipids (unsaturated:saturated ~80:20) (26%)	14
		Others (26%)	14
Skin	30	Cutin (fatty acids, including minor contributions of flavonoids and phenolic compounds) (65%)	20
		Polysaccharides (cellulose, hemicelluloses, pectin) (32%)	9
		Waxes (3%)	1

* Percentages between brackets are from literature.

[30] Yin, Jin, et al. "Effect of surface chemistry on the dispersion and pH-responsiveness of chitin nanofibers/natural rubber latex nanocomposites." Carbohydrate polymers 207 (2019): 555-562.

[31] Valorization of Tomato Processing by-Products: Fatty Acid Extraction and Production of Bio-Based Materials José J. Benítez, Paula M. Castillo, José C. del Río, Manuel León-Camacho, Eva Domínguez, Antonio Heredia, Susana Guzmán-Puyol, Athanassia Athanassiou, José A. Heredia-Guerrero Materials 2018, 11, 2211

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

3 ADVANCES IN INNOVATIVE COATINGS

3.1 Coatings for bioplastics

Plastic and packaging market

The combined plastic and food sector form an important part of the EU economy, accounting for 15 million jobs (7.6% of total employment) and 4.4% of GD. Unlocking the innovation potential in the field of packaging will significantly contribute to job creation and competitiveness.

Should ECOFUNCO reach a minute (0.1%) penetration in the packaging EU turnover (€675 billion per annum) the project will generate annually €168 M and ~1400 new jobs in Europe within 5 years post project (to allow reaching full commercial scale) and will then grow further as the production capacity, visibility and policy awareness and support to such solutions increase.

In addition, the project will develop the combination of the protein-based oxygen barrier with cutin to offer **a bio-based barrier both against oxygen and humidity**. Consequently, the protein-cutin based coatings used in multilayer laminates can completely substitute synthetic oxygen-barrier layers currently used packaging sector. The combination of both adhesive as well as barrier properties in a single layer could result in significant material savings in multilayer structures, thus improving both economic as well as ecologic effectiveness.

The combined use of protein-based coating from bio-based allow us to deliver smart food packaging with bio-based barrier coating, biodegradable films. In addition, protein, polysaccharides and cutin coatings are both **bio-based** (valorising an agro-food by-product), **highly biodegradable for protein and polysaccharides** (they even accelerate biodegradation), to be quantified but still based on a natural produced material when cutin is used, thus ECOFUNCO full compliant with EC Regulation No 10/2011[32].

Several companies, such as BASF, LIMAGRAIN, NOVAMONT, etc., have developed polymer resins that are home compostable. Further improvements are needed to provide cost effective solutions with high bio-based content and suitable performances to meet for example the target of the newly enforced law in France[33] that requires some disposable items such as **tableware** to be home compostable from 2017 with a minimum bio-sourced content of 30% (increasing progressively in subsequent years to 60% in 2025). In ECOFUNCO, the target bio-based content will be above 80% for all applications.

[32] Commission Regulation (EU) No 10/2011 of 14 January 2011 on plastic materials and articles intended to come into contact with food Text with EEA relevance, http://eur-lex.europa.eu/legal-content/EN/ALL/?uri=CELEX%3A32011R0010
[33] Décret n° 2016-1170 du 30 août 2016 relatif aux modalités de mise en œuvre de la limitation des gobelets, verres et assiettes jetables en matière plastique

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

3.2 Coatings for cellulose-based products

Cellulose is widely used in personal care applications, such as wipes for skin cleaning, and in disposable tableware and items in contact with food, both considered in the ECOFUNCO project. Regarding the application in personal care the experience developed in the framework of the ongoing POLYBIOSKIN project, participated by INSTM, IRIS and UOW, allowed addressing different treatments showing high compatibility with skin[34],[35]. In particular chitin nano-fibrils and its derivatives, also in the presence of active molecules, were kept into account for preparing bioplastic active surfaces.

The research developed in the frame of project INNOCARTOPACK, funded by Cassa di Risparmio di Lucca, Tuscany, Italy, gave the possibility to INSTM and LUCENSE of applying CNs and chitosan on paperboard and antimicrobial properties of these coatings were observed [36]. These findings will be also considered a starting point for the activity regarding the coating for personal care applications in ECOFUNCO project.

3.3 State of the art and innovation for protein-based coatings

Proteins are natural polymers synthesized by all living organisms for a wide range of reasons. The monomer units are 20 different so called proteinogenic amino acids whereas the structure and properties of a specific protein is determined by the number, sequence and types of amino acids. The use of different proteins as oxygen barrier layer has received quite some attention in literature[37,38,39,40,41] and first whey protein-based coatings developed in EU funded projects[42,43] are about to be commercialized.

34 Morganti, P, Coltelli, MB, Danti, S, Bugnicourt, E, The Skin: Goal of the EU Polybioskin Project, Global Research Journal of Pharmacy and Pharmacology, Vol. 2(1): pp 007-013, October, 2017

35 Coltelli M.B, Gigante V., Panariello L., Aliotta L., Morganti P., Danti S., Cinelli P., Lazzeri, A., Chitin nanofibrils in renewable materials for packaging and personal care applications, Advanced Materials Letters, 2019, Advanced Materials Letters

[36]Panariello L., Buchignani M, Coltelli MB, Lazzeri A., INNOCARTOPACK- Innovative nano-structured treatments for biodegradable cardboard packaging, 4th international Conference and Exhibition on natural products, June 11-12, 2018, Rome, proceedings p. 84

[37] Jost, V., et al., Influence of plasticiser on the barrier, mechanical and grease resistance properties of alginate cast films. Carbohydrate Polymers, 2014. 110: p. 309-319.

[38] Pommet, M., et al., Study of wheat gluten plasticization with fatty acids. Polymer, 2003. 44(1): p. 115-122.

[39] Mo, X. and X. Sun, Plasticization of soy protein polymer by polyol-based plasticizers. Journal of the American Oil Chemists' Society, 2002. 79(2): p. 197-202

40 Coltelli, M.-B., et al., State of the Art in the Development and Properties of Protein-Based Films and Coatings and Their Applicability to Cellulose Based Products: An Extensive Review. Coatings, 2015. 6(1): p. 1.

[41] Schmid, M., et al., Storage time-dependent alteration of molecular interaction–property relationships of whey protein isolate-based films and coatings. Journal of Materials Science, 2015: p. 1-9.

[42] WHEYLAYER. FP7, Grant Agreement Nr. 218340-2, European Commission.

[43] WHEYLAYER 2. FP7, Grant Agreement Nr. 315743, European Commission.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

The excellent barrier properties of protein-based films are due to covalent and non-covalent intermolecular interactions caused by free functional groups of the amino acids in the polypeptide chain. These causing the formation of a protein network acting as efficient barrier for oxygen [44],[45],[46] However as a result of these interactions protein-based films and coatings are usually brittle and require the formulation with plasticisers[47]. These plasticisers on the other hand cause an increase in oxygen permeability due to the increased free volume in the protein network[48]. It is of utmost importance to develop suitable protein-based formulations combining both good barrier as well as mechanical properties.

The properties of protein obtained by dilution precipitation differ significantly from protein isolates obtained by other processes. The micellar protein exhibits a very smooth and fat like texture and a high stickiness.

In a national funded project[49] micellar proteins obtained from different sources were used to develop a lacquering adhesive having the unique property of combining both a high adhesive strength as well as an excellent barrier against oxygen. The oxygen permeability of the coating was as low as 0.93 cm^3 (STP)/(m^2*d*bar) (normalized to 100 µm) and thus comparable to the whey protein-based coatings. The adhesive strength could not be quantified as rupture of the paper substrate occurred before the protein coating failed. This is however indicating that the bond strength of the coating was exceeding the cohesion strength of the substrate[50].

This combination of two properties, usually provided by different materials, in a single layer offers the opportunity to significantly reduce the number of layers and thus material used in a multilayer structure while improving biodegradability and recyclability likewise. Suchlike coating is so far only available from DOW Chemicals (Adcotem L85-800, presented 2017), however this material is fossil based, not biodegradable and only suitable for medium barrier applications.

[44] Schmid, M., Prinz, T. K., Stäbler, A., & Sängerlaub, S. (2017). Effect of sodium sulfite, sodium dodecyl sulfate, and urea on the molecular interactions and properties of whey protein isolate-based films. Frontiers in chemistry, 4, 49.

[45] Schmid, M., Sängerlaub, S., Wege, L., & Stäbler, A. (2014). Properties of transglutaminase crosslinked whey protein isolate coatings and cast films. *Packaging Technology and Science*, *27*(10), 799-817.

[46] Schmid, M., Hinz, L. V., Wild, F., & Noller, K. (2013). Effects of hydrolysed whey proteins on the techno-functional characteristics of whey protein-based films. *Materials*, *6*(3), 927-940.

[47] Schmid, M. (2013). Properties of cast films made from different ratios of whey protein isolate, hydrolysed whey protein isolate and glycerol. *Materials*, *6*(8), 3254-3269.

[48] Schmid, M., Dallmann, K., Bugnicourt, E., Cordoni, D., Wild, F., Lazzeri, A., & Noller, K. (2012). Properties of whey-protein-coated films and laminates as novel recyclable food packaging materials with excellent barrier properties. *International Journal of Polymer Science*, *2012*

[49] Micellar Adhesive, 2015- 2017, Federal Ministry for Education and Research, IBÖM01

[50] Eibl, I., von der Haar, D., Jesdinszki, M., Stäbler, A., Schmid, M., & Langowski, H. C. (2018). Adhesive based on micellar lupin protein isolate exhibiting oxygen barrier properties. Journal of Applied Polymer Science, 135(25), 46383.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

The currently available micellar protein adhesive has some drawbacks that need to be overcome. This is a limited process-ability resulting in and inadequate coating speed. In addition, and more important, the barrier adhesive can only be used for paper-polymer laminates. When applied in polymer-polymer multilayer structures the water used as solvent cannot evaporate and no adhesion between the coating and the plastic layer is achieved at all. So far trials on using other solvents such as ethanol or ethyl acetate have failed. This is mainly due to the low solubility of proteins in non-aqueous media. Many organic solvents such as ethanol act as denaturants thus altering micellar structure of the protein needed to achieve both desired properties, however the use of the protein barrier adhesive in plastic multilayers is highly desirable due to market volume but mainly due to the fact that plastic multilayers pose the biggest problem regarding leakage into ecosystems and recyclability.

3.4 State of the Art and Innovation on Chitosan and Chitin

Chitosan is a cationic polysaccharide deriving from the partial deacetylation of chitin and it can have animal (shells of crustaceans) or vegetal (fungi such as *Aspergillus niger)* origin. Chitosan is characterized by non-toxicity, biodegradability, film-forming capacity, antimicrobial and antioxidant properties and good barrier properties of chitosan films against oxygen. [51] The main advantage of chitosan application is the possibility to realize films and coatings with intrinsic antimicrobial properties and is just this characteristic that differentiates chitosan from other common antimicrobials (for example, ethanol, sorbic acid, bacteriocins as nisin, lysozyme, essential oils etc.).

The properties of chitosan can vary in relation to chitosan type, origin and its physio-chemical characteristics. Referring to chitosan films and coatings, antimicrobial and barrier properties depend on chitosan molecular weight and deacetylation degree, its concentration, the solvent used for its solubilisation, the pH and possible plasticizers or other additives added in the formulations. Antimicrobial activity is possibly owing to the positive charges which can interact with negatively charged residues of macromolecules on the microbial cell surface, causing membrane leakage[52]. It is thus possible to find many examples of coatings application by dipping technique, spraying etc and use of films produced by casting technique for fruit and vegetables, meat, cheese and fish. Antimicrobial properties of chitosan were largely investigated even combined with other antioxidants and antimicrobials substances, as essential oils, or with other film forming materials, as proteins and gelatine.

[51] A. Verlee, S. Minckle, C.V. Stevens, Recent developments in antibacterial and antifungal chitosan and its derivatives, Carbohydrate Polymers, Vol. 164, pp. 268-283, 2017; M. Aider, Chitosan application for active bio-based films production and potential in the food industry: Review, LWT-Food Science and Technology, Vol. 43, pp. 837-842, 2010; M. Kong, X.G. Chen, K. Xing, H.J. Park, Antimicrobial properties of chitosan and mode of action: A state of the art review, Vol. 144, pp. 51-63, 2010

[52] Zheng, L.Y., and Zhu, J.F. 2003. Study on antimicrobial activity of chitosan with different molecular weights. Carbohydrate Polymers, 54, 527-530

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

SSICA tested chitosan-based cast film on fresh anchovies. It was possible to maintain microbial load at low levels until 15 days of storage, with a great improvement in shelf-life with respect to control samples (not in contact with chitosan), that reached high microbial concentrations, even antioxidant effects were observed. Both types of fish were preserved in the presence of chitosan coating [53].

Chitin nano-fibres (CNF) have recently attracted significant interest because of their high aspect ratio, high mechanical strength and high crystallinity. Due to the strong hydrogen bonds between chitin nanofibers, large amounts of energy are needed to disintegrate chitin fibrils into nanofibers via mechanical treatments, such as high-pressure homogenization[54] and disk milling circumvent the problem of high energy consumption during mechanical processes, chemical pre-treatment by TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl radical)-mediated oxidation was used to weaken the bonds that hold the chitin chains together making it easier to break them into CNF [55]. Partial deacetylation associated with partial mechanical scission of the fibrils during disintegration was also used to obtain CNF[56]. In addition, the esterification of hydroxyl groups of chitin by carboxylate groups can significantly improve the mechanical disintegration of chitin using a grinder[57]. Unfortunately, most of these techniques require the use of hazardous solvents that strongly limit the environmental benefit of the use of CNF.

Regarding the preparation of poly(lactic acid) (PLA)-based nanocomposites containing Chitin nano-fibrils (CNs), a dispersion at the nanoscale was achieved thanks to the preparation of pre-composites, as described by Coltelli et al. [58,59] This strategy can be considered to disperse CNs in bio-polyester formulations or hot-melt oligo-polyesters for producing functional film or coatings.

[53] Video realized by AGERiretti (soon available on web)
[54] 10.1021/bm501416q
[55] https://doi.org/10.1016/j.carbpol.2010.02.006
[56] https://doi.org/10.1016/j.carbpol.2009.10.044
[57] https://doi.org/10.1016/j.carbpol.2016.07.060
[58] Coltelli MB, Cinelli P, Gigante V, Aliotta L, Morganti P, Panarielo L, Lazzeri A, Chitin Nanofibrils in Poly(Lactic Acid) (PLA) Nanocomposites: Dispersion and Thermo-Mechanical Properties, Int. J. Mol. Sci. 2019, 20, 504; doi:10.3390/ijms20030504
[59] Coltelli MB, Gigante V, Panariello L, Aliotta L, Morganti P, Danti S, Cinelli P, Lazzeri A, Chitin nanofibrils in renewable materials for packaging and personal care applications, Advanced Materials Letters, 2019, 10(6), 425-430

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

Interestingly CNs were found cytocompatible, showing anti-inflammatory activity and may serve for the delivery of biomolecules for skin care and cells regeneration[60]. These findings suggest applications in the personal care sector, because of the good compatibility of the CNs with skin[61,62].

3.5 State of the art and innovation for cutin and valuable biomolecules production

Cutin is a crosslinked polyester formed mainly by condensed polyhydroxylated acid[63] and it is the main component of plant cuticles. The main role attributed to plant cuticles is to avoid water loss from internal tissues[64], acting as a gas barrier and thermal regulator, as defence against pathogens, reduction of nutrient leaching and protection from mechanical injuries and UV damages.[65,66]

Tomatoes are a particularly interesting source for cutin, due to the high concentration in the peels[67]. The world tomato production is 182 M tons (FAOSTAT 2017 data) mainly concentrated in the northern hemisphere, of which 38 M tons are processed in different products, tomato paste, diced and peel tomato[68]. The wasted peels and seeds, produced each year at industrial level, are significant, about 1.0 million tons, 300000 tons at EU level.

Cutin is generally extracted from plant material after cuticle isolation. Enzymes such as pectinases and cellulases, in acetate buffer, are usually employed for cuticle isolation.[69,70] Isolated plant cuticles are

[60] Danti S, Trombi L, Fusco A, Azimi B, Lazzeri A, Morganti P, Coltelli MB, Donnarumma G, Chitin Nanofibrils and Nanolignin as Functional Agents in Skin Regeneration, Int. J. Mol. Sci. 2019, 20(11), 2669; https://doi.org/10.3390/ijms20112669

[61] Morganti P, Danti S, Coltelli MB, Chitin and lignin to produce biocompatible tissues, Research in Clinical Dermatology (2018) Volume 1, Issue 1, 2018

[62] Morganti P, Coltelli MB, A New Carrier for Advanced Cosmeceuticals, Cosmetics 2019, 6(1), 10; https://doi.org/10.3390/cosmetics6010010

[63] Heredia A. 2003. Biophysical and biochemical characteristics of cutin, a plant barrier biopolymer. *Biochimica et Biophysica Acta (BBA) – General Subjects* 1620, 1–7.

[64] Riederer M, Schreiber L. 2001. Protecting against water loss: analysis of the barrier properties of plant cuticles. *Journal of Experimental Botany* 52, 2023–2032.

[65] Heredia A. 2003. Biophysical and biochemical characteristics of cutin, a plant barrier biopolymer. *Biochimica et Biophysica Acta (BBA) – General Subjects* 1620, 1–7.

[66] Martin LB, Rose JK. 2014. There's more than one way to skin a fruit: formation and functions of fruit cuticles. *Journal of Experimental Botany* 65, 4639–4651.

[67] Järvinen, R., Cuticular and suberin polymers of edible plants. Analysis by gas chromatographic-mass spectrometric and solid state spectroscopic methods. 2010.

[68] WPTC World production estimate of tomatoes for processing – Date of last update: 16/05/2019

[69] Kallio, H., et al., Cutin Composition of Five Finnish Berries. *Journal of Agricultural and Food Chemistry*, 2006. 54 (2), 457-462.

[70] Järvinen, R., et al., Solid state 13C CP-MAS NMR and FT-IR spectroscopic analysis of cuticular fractions of berries and suberized membranes of potato. *Journal of Food Composition and Analysis*, 2011. 24 (3), 334-345.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

dewaxed by immersion in organic solvents[71] and polysaccharides are removed by acid hydrolysis.[72,] Cutin can be depolymerized by cleaving the ester bonds using alkaline hydrolysis, with NaOH or KOH in water, transesterification with methanol containing BF_3 or $NaOCH_3$, reductive cleavage by exhaustive treatment with $LiAlH_4$ in THF, or with trimethylsilyl iodide in organic solvents.[73] These methodologies are not adequate for large-scale cutin extraction due to the number of steps involved and the cost of solvents and chemicals. The method patented by SSICA and CTAEX in the European project FP7 BIOCOPAC[74], is solvent-free and does not use a pre-treatment for cuticle isolation. This innovation allowed the design of a pilot plant, during the project LIFE+ BiocopacPlus, that extracts cutin from tomato by-products at a semi-industrial scale. The method developed for the cutin extraction allows the isolation of 10,16-dihydroxyhexadecanoic acid and its oligomers at a purity of about 80%. Currently the main and patented[75] application of the cutin obtained from the LIFE+BiocopacPlus plant us the use as coating in metal packaging. For this the process to obtain cutin also comprises the synthesis of the bioresin by homopolymerization in solvent at medium temperature and the formulation of the bio-lacquer for metal materials application. However other protocols have been studied and compared by SSICA[76] in order to enhance the purity and the reproducibility of the extracted products.

In the literature, the cutin was extracted and characterized in vegetable by-products different from tomato as watermelon peels[77], tea residues[78] and apple peels[79].

---

[71] Luque, P., S. Bruque, and A. Heredia, Water Permeability of Isolated Cuticular Membranes: A Structural Analysis. *Archives of Biochemistry and Biophysics*, 1995. 317 (2), 417-422.

[72] J. A. Heredia-Guerrero, A. Heredia, E. Dominguez, R. Cingolani, I. S. Bayer, A. Athanassiou, J.J. Benitez, Cutin from agro-waste as a raw material for the production of bioplastics, *Journal of Experimental Botany*, accepted 12 July 2017; Chefetz, B., Decomposition and sorption characterization of plant cuticles in soil. Plant and Soil, 2007. 298 (1), 21-30; Shechter, M., et al., Competitive Sorption−Desorption Behavior of Triazine Herbicides with Plant Cuticular Fractions. *Journal of Agricultural and Food Chemistry*, 2006. 54 (20), 7761-7768

[73] Kolattukudy PE. 2001. Polyesters in higher plants. In: Babel W, Steinbüchel A, eds. Biopolyesters. Berlin, Heidelberg: Springer Berlin Heidelberg, 1–49.

[74] I.M. Cigognini, A. Montanari, R. De La Torre Carreras, M. Gomez-Cardoso Bernet "Extraction method of a polyester polymer or cutin from the wasted tomato peels and polyester polymer so extracted", WO 2015/028299A1, 2015 (Pending)

[75] C.M. Cedri, L. Cioni, X. Gomez, A. Montanari, V. Orlandi, D. Vareckova, T. Vlcek ,"Lacquer composition having cutin based resin, method for obtaining said lacquer and said cutin based resin", Patent n° 0001426490, 2014

[76] A. Cifarelli, I. Cigognini, L. Bolzoni, A. Montanari, Physical-chemical Characteristics of cutin separated from tomato waste for the preparation of bio-lacquers, *Advances in Sciences and Engineering,* 2019, 11:1, 33-45

[77] S. A. Chaudhari, R. S. Singhal, Cutin from watermelon peels: a novel inducer for cutinase production and its physicochemical characterization, *International Journal of Biological Macromolecules*, accepted 10 may 2015

[78] S. Tsubaki, H. Iida, M. Sakamoto, J. Azuma, Microwave heating of tea residue yields polysaccharides, polyphenols, and plant biopolyester*, Journal of Agricultural and Food Chemistry*, 2008, 56, 11293-11299

[79] Eglinton, G., Hunneman, D.H, Gas chromatographic-mass spectrometric studies of long chain hydroxy acids-I. The constituent cutin acids of apple cuticle, *Phytochemistry*, 1968, 7(2), 313-322

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

In addition, tomato, other sources for plant cutin such watermelon peels[80], tea residues[81] and apple peels[82] have been described in literature. Tomatoes possess a high nutritional value due to its content of different types of micronutrients: vitamins (C and E), folates, carotenoids and phenolic compounds (Table 2), which are well known for their antioxidant/antimicrobial properties. Tomato is the main source of lycopene in the western diet and this carotenoid confers the characteristic red colour[83].

The tomato cutin was the main component of a bio-lacquer developed for food metal packaging in the European project LIFE+BiocopacPlus. This application was patented[84] and consists in the synthesis of the bioresin by homopolymerization in solvent at medium temperature and the formulation of the bio-lacquer for metal materials application.

Table 2. Typical composition (mg 100 g^{-1} fresh weight) in tomato ripe fruits of carotenoids and polyphenols[85]

Carotenoid	Concentration	Polyphenol	Concentration
Lycopene	7.8–18.1	Naringenin chalcone	0.9–18.2
Phytoene	1.0–2.9	Rutin	0.5–4.5
Phytofluene	0.2–1.6	Quercetin	0.7–4.4
β-Carotene	0.1–1.2	Chlorogenic acid	1.4–3.3
γ-Carotene	0.05–0.3	Caffeic Acid	0.1–1.3
δ-Carotene	0–0.2	Naringenin	0–1.3
Lutein	0.09	Kaempferol-3-rutinoside	0–0.8
Neurosporene	0–0.03	*p*-Coumaric acid	0–0.6
α-Carotene	0–0.002	Ferulic acid	0.2–0.5
Neoxanthin	-	Kaempferol	0–0.2
Violaxanthin	-	Myricetin	-
Anteraxanthin	-	Cyanidin	-
Zeaxanthin	-	Pelargonidin	-
		Delphinidin	-

80 S. A. Chaudhari, R. S. Singhal, Cutin from watermelon peels: a novel inducer for cutinase production and its physicochemical characterization, *International Journal of Biological Macromolecules*, accepted 10 may 2015

81 S. Tsubaki, H. Iida, M. Sakamoto, J. Azuma, Microwave heating of tea residue yields polysaccharides, polyphenols, and plant biopolyester*, Journal of Agricultural and Food Chemistry*, 2008, 56, 11293-11299

82 Eglinton, G., Hunneman, D.H, Gas chromatographic-mass spectrometric studies of long chain hydroxy acids-I. The constituent cutin acids of apple cuticle, *Phytochemistry*, 1968, 7(2), 313-322

83 Stéphane Georgé, Franck Tourniaire, Hélène Gautier, Pascale Goupy, Edmond Rock, Catherine Caris-Veyrat, Changes in the contents of carotenoids, phenolic compounds and vitamin C during technical processing and lyophilisation of red and yellow tomatoes Food Chemistry 124 (2011) 1603–1611.

84 C.M. Cedri, L. Cioni, X. Gomez, A. Montanari, V. Orlandi, D. Vareckova, T. Vlcek ,"Lacquer composition having cutin based resin, method for obtaining said lacquer and said cutin based resin", Patent n° 0001426490, 2014

85 Raúl Martí, Salvador Roselló, Jaime Cebolla-Cornejo, Tomato as a Source of Carotenoids and Polyphenols Targeted to Cancer Prevention, Cancers 2016, 8, 58

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

3.6 State of the art and progresses in the field of bio-based oxygen barrier

Protection against oxygen is a key factor for sensitive/fresh food, cosmetic and pharmaceutical products to achieve a sufficient shelf-life. Otherwise colour or taste deviation, oxidation of grease, formation of micro-organisms or degrading nutrients will take place. To achieve low oxygen permeability of food packaging usually several different materials are combined in so called multilayer structures by co-extrusion or lamination.

Multilayer plastic films are widely used in the packaging industry whereby mineral oil derived ethylene vinyl alcohol (EVOH), or polyvinylidene chloride (PVDC), are the oxygen barrier materials mostly used. However, theses polymers are neither biodegradable nor bio-based. Therefore, research into sustainable packaging materials that maintain the performance of their composite structures has been recently intensified. In recent European resp. national funded projects oxygen barrier coatings based on whey protein were developed with participation of IRIS, FRAUNHOFER and INSTM.

For this coating protein extracted from whey, a by-product from cheese production is formulated with water and a bio-plasticiser. The formulation is then applied on a plastic substrate in a wet coating process and dried. Thereby protein denaturation is taking place, a step required to obtain a crosslinked protein network and thus an oxygen permeability as low as 1.5 $cm^3/(m^2*d*bar)$ (Q100).

Finally, the coated film is laminated into the final structure and eventually additional conversion processes such as thermoforming, sealing etc. can be carried out to obtain the packaging items. Application of both standard and thermos-formable whey coating formulations has been proved in pilot scale in roll-to-roll lacquering and lamination lines. Resulting films were reported to have excellent mechanical as well as barrier properties, beyond most existing biopolymers (Figure 5).

Whey protein has a rather high prize of >10€/kg. In addition, framework conditions concerning the use of whey have changed recently.

When the research projects were conducted, whey was a widely underutilized by-product from cheese production, whereas today most cheese manufacturers use whey to produce high value food products.

In addition, the use of an animal derived protein in a packaging is questionable with respect to sustainability, and consumer acceptance (vegetarian, vegan etc).

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

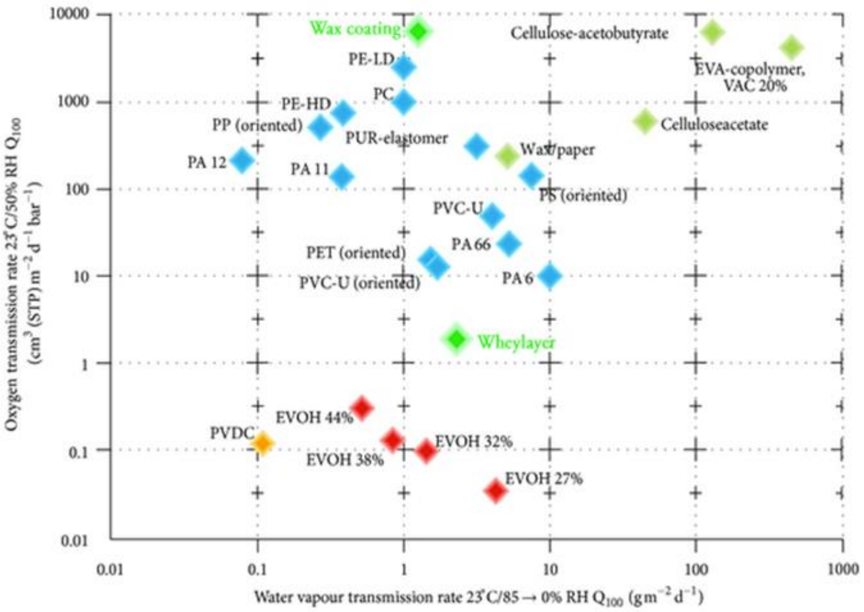

Figure 5: *Barrier properties of whey-based layer as well as wax coating vs. other plastics commonly used in food packaging industry (normalized to 100 µm)*

Therefore novel, plant residue derived proteins have to be identified to tap the potential of a bio-based oxygen barrier coating in combination with high sustainability and low price. Low value and underutilized by-products from agro-food industry will be explored by FRAUNHOFER and SSICA regarding their utilization as protein source for oxygen barrier coatings.

4 ADVANCES IN DEVELOPMENT OF KEY TECHNOLOGIES AND DEMONSTRATORS

4.1 State of art and Innovation on Tableware

The global market for disposable tableware was worth 27.37 billion US$ in 2016 and is believed to reach 29.7 billion US$ by 2020. With an annual growth rate of almost 6%, disposable cups are the most important market driver in this segment. In 2016, they accounted for around 43% of sales of disposable tableware, this will grow to more than 50% by 2020.

Bio-based products still represent a niche solution in relation to the overall disposable tableware market. For instance, the global market for disposable cutlery was worth around 2.6 billion US$ in 2017. Biodegradable cutlery had a share of 1.17% resp. a value of 30.5 Million US$. Although the biodegradable solutions are expected to grow at a rate above the overall market, they will remain at

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

a market share below 2 % over the next five years. Considering that disposable tableware is among the 10 most widespread items of beach litter on Italian beaches in 2018, this represents a significant environmental issue.

A number of bio-based materials are available on the market, from companies spread worldwide, based mainly on starch or polylactic acid (PLA) (Novamont, Greenday, Plantatableware, Bioevelop, Earthshell, Naturework, Evercorn,[86] Corbion, BASF etc). These address products produced mainly by injection moulding for cutlery, by thermoforming for plates or cups, then by thermo-foaming for foamed trays or shells, this latter enabling to save up to 60% less plant-based raw material than traditional plastic or paper products. In general, the plant starch utensils are made from 70% renewable resources and 30% fillers. Non-biodegradable (polyethylene PE, polypropylene PP) or no bio-based additives (inorganic fillers such as talc and calcium carbonate) extensively used in PLA based materials should be progressively reduced and replaced by bio-based additives, such as natural fillers.

For most of these products, compost ability is claimed, but the issue of meeting conditions for home compost ability is relatively new and fulfilled by standard PLA-based materials. Another main issue in products such as trays is the low properties of barrier to gas, and in the case of starch-based materials the low resistance to water and any juicy food. For starch, and sometime even for trays with minor PLA amount, coating of the tray with a second material is performed to enhance resistance to gas and moisture.

4.2 Technology for improvement of coating adhesion to the substrates

There are many physical and chemical processes employed for activating the surface of materials, the most common of which are plasma treatment, irradiation, crosslinking and chemical treatment with weak acids or alkali in order of grafting smaller molecules on the surface. In the Durawood project (IRIS' website), plasma was used as a pre-treatment before wood coating. Plasma treated wood presented a substantially better adhesion with the coating leading to an increased durability, as well as a reduced attack by blue stain fungi.

As opposed to chemical treatment, plasma does not require consumables and does not generate by-products. It will be tested in the project in terms of obtaining coated materials with optimal properties, for surface decontamination and finally for process intensification as it is expected to speed up the impregnation of the applied liquid.

[86] http://www.plantableware.com/; https://www.papstar.com/en/once-tableware/pure/; Xiao, L.; Wang, B.; Yang, G.; Gauthier, M. Poly(Lactic Acid)-Based Biomaterials: Synthesis, Modification and Applications. Biomedical Science, Engineering and Technology. 2012, pp. 247-282.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

4.3 Monitoring

The application of a coating in consistent thickness and even distribution is also important to warrant eg. constant barrier properties while not using more resources than required. It is envisaged that coatings of several microns thickness will be applied to reach the multifunctional requirements of our applications, possibly in subsequent step. As such, monitoring a range of materials both laterally and across the samples is needed. A number of monitoring techniques exist for thin printed coatings in the sub-micro/ micro ranges. Most of them are in fact implemented off line and require sample preparation. Nevertheless, according to a recent review paper by IRIS selected combined optical technologies have potential for such in-situ analysis.[87] Spectral Reflectance is the most widely used technique that provides absolute, quantitative data. A white light source is directed at the sample surface, and the reflected light is collected and analysed with a spectrometer. Thickness is calculated by identifying the wavelengths of the interference peaks in the Reflectance spectrum, where the thickness of the layer is a function of the peak wavelength and refractive index of the material[88, 89]. The technique is ideally suited for the thickness range of 1- 50 micrometres.

Recent work in the EU project THIME[90] has led to the development of an imaging spectral reflectance system, validated as an on-line quality control system for monitoring thin film thickness in roll-to-roll organic photovoltaic (OPV) production processes. Furthermore, OptiNanoPro (see IRIS profile) developed a system suited for monitoring the application of transparent nanocomposite whey protein coatings on transparent plastic films at high speed[91] as well of micro-textured polymers[92]. Near Infrared spectroscopy and hyperspectral imaging (HIS) allow mapping composition in real time as well as to warranty suitable drying[93].

In ECOFUNCO project will be applied an approach where, thanks to the tailored monitoring solutions, 1.2 % of accuracy was reached, by partner IRIS, on a system running at up to 180 m/min, which was the upper limit of the winding machine used for testing. Similar systems in imaging modes could be further trained to detect further defects in coating production such as pinholes and scratches, which also jeopardise barrier properties.

Indeed, it has been demonstrated, that hyperspectral imaging technique is capable to detect the

[87] E.Bugnicourt et al., Recent Prospects in the Inline Monitoring of Nanocomposites and Nanocoatings by Optical Technologies, Nanomaterials 2016, 6(8)
[88] M. Merklein, "High resolution measurement of multilayer structures," Applied Optics, vol. 29, pp. 505-511, 1990/02/01 1990.
[89] F. Inc. (2016, 18th January 2016). Tutorial: Advanced Thin-Film Measurement Systems. Available: http://www.filmetrics.com/technology
[90] THIME, "Final Report Summary," 2016.
[91] http://optinanopro.eu/optinanopro01/files/2018/06/ApplicationNote-Texture.pdf
[92] http://optinanopro.eu/optinanopro01/files/2018/06/ApplicationNote-Thickness.pdf
[93] Ya Su, Min Zhang, and Arun S. Mujumdar, Recent Developments in Smart Drying Technology, Drying Technology Vol. 33 , Iss. 3,2015

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

distribution of actives or coatings when are applied on a substrate, providing the real time, in-line monitoring of the coating processes to assure the quality of the final product. Near Infrared spectroscopy and hyperspectral imaging allow mapping composition in real time as well as to warranty suitable drying[94].

4.4 Barrier packaging

One of the most important features of packaging materials is to ensure protection of the packed good. Especially in case of sensitive products such as food, this requires certain barrier properties against gases such as oxygen and water vapour accelerating deterioration of the packed good. In most cases adequate barrier properties cannot be achieved by a single material and thus modern packaging consist of different layers and coatings combined in a multilayer structure. Global transparent barrier films reached an annual market of 1.5 million tonnes in 2015, with annual growth of 3.9%[95]. Barrier films are used in all formats (flexible and rigid packaging obtained by lacquering and lamination, coextrusion or multi-material injection, etc.) by numerous packaging converters and food packers, both large and small companies, all over the world.

Multilayer structures can be easily adapted to the specific requirement of the packed food and are usually thinner and thus more resource efficient than monolayers. This complies with one of the general trends in packaging which is the decrease of packaging weight and therefore thickness to reduce the environmental impact due to resources and transport savings and leading sometimes to the capacity for flexible packaging to substitute rigid ones. In terms of multilayer barrier films, the market is dominated by fossil materials applied either by lacquering e.g. PVdC, or extrusion EVOH in terms of oxygen barrier or by PE in terms of humidity barrier.

One main drawback of multilayer structures is their low recyclability. The multilayer structures used in packaging cannot efficiently be separated in different material fractions of sufficient purity and thus not be reused in a circular approach. Therefore only 31.3% of post-consumer plastic waste is recycled while the rest is combusted (41.6%) or ends up in landfills[96]. In addition, it is estimated that globally approximately 25 Million tons of plastic material are leaking into aquatic and terrestrial ecosystems[97].

4.5 State of the art and Innovation on biodegradable packaging

The majority of the currently used bioplastic-packaging (86% according to EUBP data) are not biodegradable. These are mainly bio-based PE and PET. There are undoubtedly applications where this

[94] Ya Su, Min Zhang, and Arun S. Mujumdar, Recent Developments in Smart Drying Technology, Drying Technology Vol. 33 , Iss. 3,2015
[95] http://www.smitherspira.com/news/2016/february/key-trends-for-barrier-packaging-market
[96] PlasticsEurope (2017). Plastics - The Facts 2017. Brussels.
[97] The New Plastic Economy- Rethinking the future of plastics, 2016, Allen MacArthur Foundation

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

feature is desirable (eg. when food rest or material structure make recycling non-feasible, or when collection is not properly handled). Food packaging takes up more than 70% of the globally produced bio-based plastic which is predicted to grow at over 18% CAGR globally from 2013 to 2019 to reach a value of $8,415.20million[98]. As opposed to previously commented very low presence of EU in the overall bioplastics manufacturing, countries like Germany have a very good position in the smaller segment of bio-based biodegradable packaging. One main concern regarding the use of biodegradable plastics in applications such as packaging is their inadequate properties such as an insufficient barrier. The material has to be applied in greater thickness thus increasing costs and contradicting sustainability. The multifunctional solutions developed in ECOFUNCO project will help to overcome this issue by improving functional properties of bioplastics while maintaining resp. improving biodegradability and recyclability.

5 MARKET

5.1 Innovation and Economic Impact:

The project, through a **systemic and multidisciplinary approach, will test and demonstrate possibilities to increase the efficiency of bio-based value chains** and to establish new supply and value chains in the bio-based economy. The project will involve actors covering the whole value and supply chains in order to promote a better management of resources, increase the added-value of food and agro by-products and waste stream ultimately improving the cascading use of bio-based resources.

Market for innovative bio-based products in Europe possesses immense potential of growth up to 2021 and beyond especially when they replace fossil-based products (chemicals, plastic, etc.) mostly driven by the rising awareness of consumers about sustainable environment. ECOFUNCO project is aiming at boosting of the competitiveness of the industries in these sectors and impact on the European economy and society **while increasing sustainability of the industrial production improving the efficiency in the transformation of sustainable, EU-source primary and secondary raw materials.**

Bio-based polymers produced in ECOFUNCO are targeting **various end-use applications including the packaging, sanitary products and disposable products.** Hence, the ECOFUNCO project potentially addresses an extremely wide range of industries including polymer chemistry, plastics converting and manufacturing, packaging, etc. Thereby ECOFUNCO is not only aiming at a mere substitution of fossil-

[98] "Biodegradable Packaging Market by Packaging Type (Plastic and Paper), by Applications (Food Packaging, Beverage Packaging,Pharmaceuticals Packaging, Personal & Home Care Packaging, and Others), and by Geography (North America, Europe, Asia-Pacific, and ROW) – Global Trends & Forecast to 2019" published at RnRMarketResearch.com

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

based coating solutions but also **to provide improved functionalities thus outperforming existing systems.**

The polymer chemistry, biopolymers producers and subsequent end-users in various sectors will benefit from the development of new, environmentally friendly solutions based on renewable, low cost raw materials with high functionality. Over 8 billion tons of materials per year are used in the EU, from which 20% is imported. The size and growth of EU economy is generating pressure also on other countries: new tools to reduce this pressure are required. Every kilogram of imported products leads to 3 to 6 kg's of more material outside the EU. Improvement of the re-use of raw materials could save 1.4 billion Euros a year and generate 1.6 Billion Euros in sales. In 2009, 0.75 – 1.5 Million tons of bio-based plastics were produced worldwide with an estimated growth to as high as 4.5 Million tons by 2020. Today, the European bio-economy sectors are worth €2 trillion in annual turnover and account for 22 million jobs in the EU, which is 9% of the EU's workforce[99]. Bioplastics have been designated a lead market by the European Commission. In line with the green movement, we are seeing manufacturers increasingly shift from traditional plastics to bioplastics, and analysts are expecting the bioplastic market to grow at CAGRs of 17.5% or more in 2016-2020[100]. Respectively, the European bio-plastics market is estimated to grow from 0.13 Million tons in 2008 to 0.90 Million tons by 2020. The sector represents only about 0.4% of the overall global polymers industry volume. At the current stage of technology, it is estimated that 5-10% of the current plastics could be substituted by bio-based plastics, whereas by 2030 as high as 70-100% share could be achieved.

ECOFUNCO developed materials will stay in this category[101], replacing petro hardly recyclable products.

The sales of compostable and biodegradable plastic products (end products) reached around 100,000 tonnes in Europe in 2015. The market of compostable and biodegradable plastic products could grow to beyond 300,000 tons in 2020 if the legal framework were to be set more favourably. This estimation is based on an assumed annual growth rate of about 27% for the "5 years" period. Disposable short-life products marketed as waste management solutions and made of compostable polyester-based films are today's bestsellers. However, functional products such as barrier packaging or fibre products and many outdoor uses have a significant potential for market breakthrough.

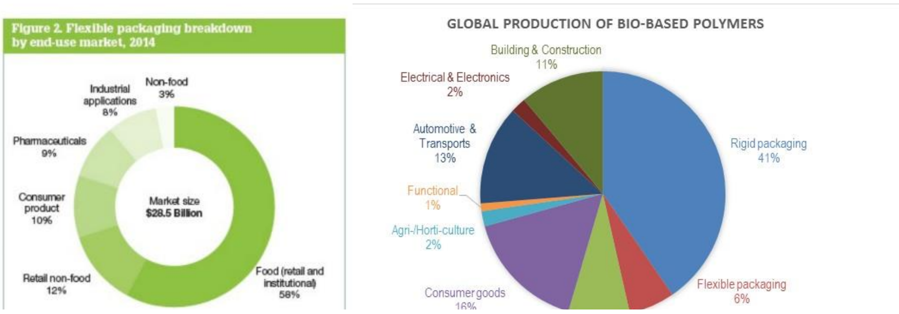

Figure 6: Breakdown of the biobased polymers industry per application

99 Source: European Bioplastics Association
100http://energyandgold.com/2016/03/10/an-enormous-investment-opportunity-in-a-rapidly-growing-space/
101 H.Kaeb, F. Aeschelmann, L. Dammer, M. CarusMarket study on the consumption of biodegradable and compostable plastic products in Europe 2015 and 2020 from NOVA Institute.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

In packaging, transparent barrier films are replacing traditional materials such as flexible foil for various applications due to consumer demand for packaging transparency, feasibility and use in microwave ovens. Smithers Pira predicted that global demand for transparent barrier films reached 1.5 million tonnes for 2015, and is forecast to grow at an annual average rate of 3.9% over the five-year study period, reaching 1.9 million tonnes in 2021 (Figure 6).[102]. Such growth would generate many positive effects and benefits, i.e. new jobs and prosperous SMEs and start-ups in sectors such as innovative plastics, food service and biotechnology thus causing a further positive effect in health, derived by a promotion of balanced diet with fish fruit and vegetables, and in higher quality and safety of food.

Consumer goods (the target of the ECOFUNCO project) share of the total bioplastics market sits at 16%[103] (Figure 5). Assuming that this share remains at about 16%, the segment would be worth $ 3.36Bn in 2019, growing to $ 5.2 Bn by 2030. In Europe**,** a market of 28.5 Billion $ was estimated in 2014 for flexible packaging, of whom 16.4 Billion for food packaging[104]. Thus, just a small percentage of this being covered by ECOFUNCO films will result in huge increase of SMEs turnover and consequently new plant for production and new workers to run them can be converted in many new job positions, considering an average of one new job position every 230 KEur.[105]

5.2 Bio-based Coatings Market:

For Europe's coatings industry a key objective is sustainability so that the sector sharply reduces its CO_2 emissions by switching to low carbon materials with a minimum carbon footprint and helps to conserve resources through reuse and recycling of products and their ingredients. The European coatings supply chain is stepping up its efforts to ensure that a much higher proportion of its products are derived from bio or renewable sources. Much of the impetus is coming from climate change. The European Union has set a target of 30 percent of existing petrochemical-derived chemicals and materials being replaced by bio-based and biodegradable alternatives by 2030. This objective is in line with the long-term aim in Europe of establishing low-carbon, circular economies with substantial reductions in CO_2 emissions and greater reliance on its own raw materials with much more recycling and reuse of materials to achieve zero waste. Demand for bio-based coatings and its corresponding European and global market is on the rise. The global packaging coatings market was estimated at

102 http://www.smitherspira.com/industry-market-reports/packaging/the-future-of-transparent-barrier-films-to-2021
103 http://www.prnewswire.com/news-releases/global-bioplastic-packaging-market-2014-2018-237009271.html
104 https://www.alliedmarketresearch.com/press-release/polylactic-acid-market.html
105 Based on a new job being created for every €230k of growth, taking into account figures by PlasticsEurope, which reports that in 2014 the turnover generated was 350 billion € and the number of employees was ~1,45 millions. http://www.plasticseurope.org/

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

around USD 2.9 billion in 2017, and is expected to reach $3,865 million by 2023, registering a CAGR of 4.6% from 2017 to 2023 (Figure 7). The major factors driving the growth of the market are the increasing demand for packaged foods and the rapidly expanding e-commerce sector around the world. **Antimicrobial Coatings Market** size was worth over USD 2 billion revenue in 2015 with forecast to grow over 10% Compound annual growth rate (CAGR) up to 2024.

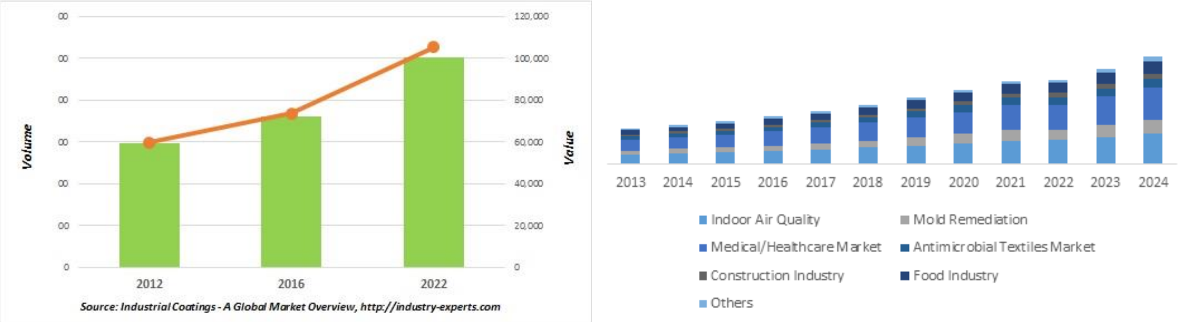

Figure 7 a): Global Industrial Coatings Market [106], **b)** Europe Antimicrobial Coatings Market Revenue, by Application, 2013 - 2024 (USD Million)[107]

Food Packaging is largely the most used application for polymers (approximately a 45% of the worldwide polymer production goes into these applications). Since most of the polymers used in packaging come from oil-derived sources, the consumption of these sources only in this field is enormous. Global, the packaging industry is estimated to expand at CAGR of 4% over the forecast period. The growing scope of packaging in various products, especially in consumer goods and food and beverages industry, is estimated to further drive growth of the globally packaging coatings market in the coming years 2016 – 2024 (Figure 8). It is in that framework that the introduction of new bio-based coatings, obtained from food and agro-wastes represent such an attractive alternative to conventional polymers, both in the aspects of a non-ending source and even with better and improved properties and a minimal environmental impact.

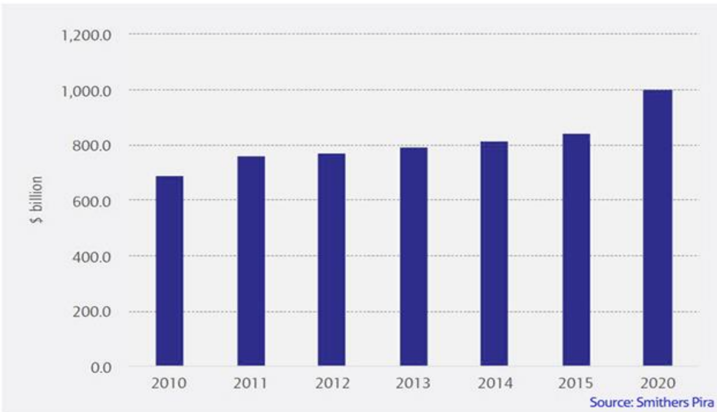

Figure 8: Global packaging market forecasts[1]

Packaging of ham is quite important in this proposal as CPP is a partner. On 2016, 7900 tons of packed product were sold, mainly for export. Assuming that the package is about 24 grams per unit, 1580

106 http://industry-experts.com/verticals/chemicals-and-materials/industrial-coatings-a-global-market-overview

107 https://www.gminsights.com/industry-analysis/antimicrobial-coatings-market-report

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

tons/year of trays and 316 tons/year of films are obtained. This is the amount of packaging necessary only for this consortium.

5.3 Tissue personal care products market.

The world tissue consumption has increased at a yearly rate of 3.1% in the past ten years and its growth is expected to continue at a similar rate until 2025, according to the 10-years forecast of Statistas Outlook for world tissue business (Figure 9).

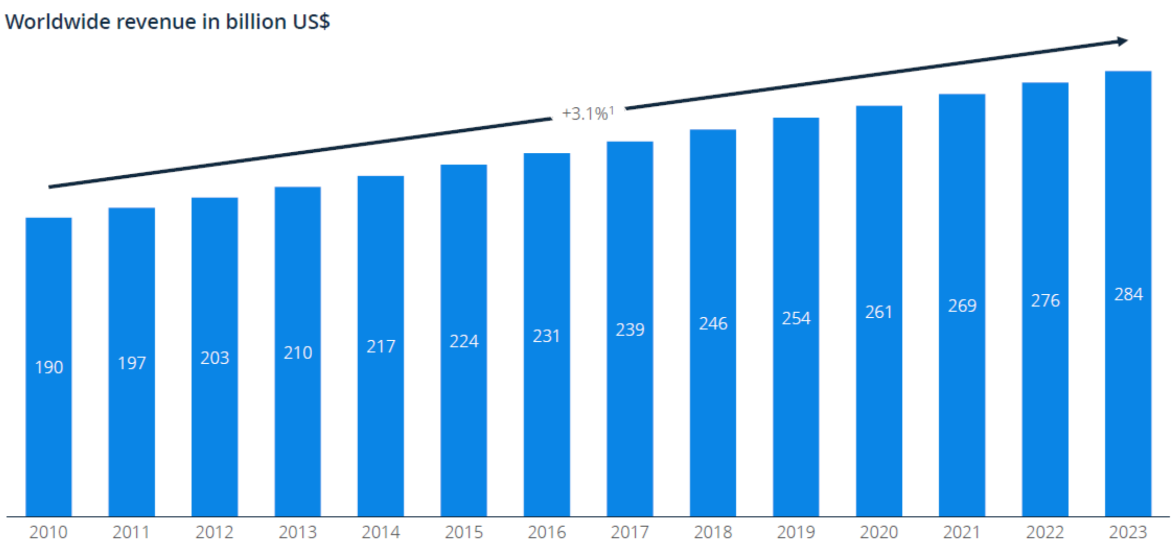

Figure 9: Worldwide revenues from Tissue and hygiene paper from 2010 to 2023

North America is the unchallenged leader in terms of per capita tissue consumption, followed by Western Europe and Japan (Figure 10). The most import and demand drivers are the expected future economic growth, population growth and demographic changes, product penetration level, development in tissue quality and product specifications, substitution effects and AFH (away from home) dispenser developments.

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

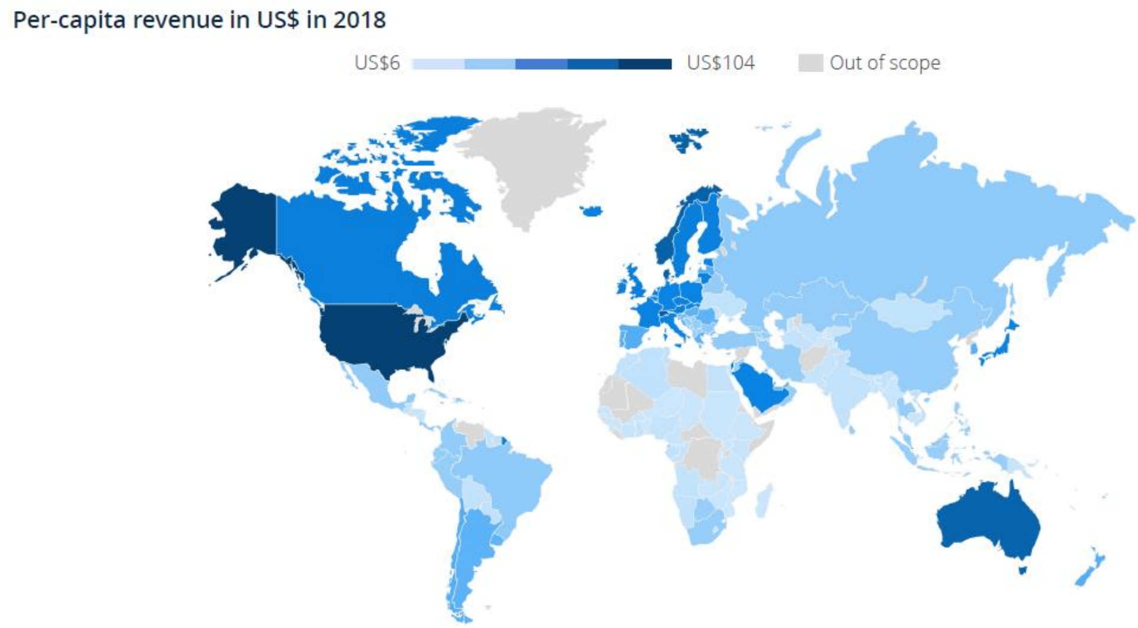

Figure 10: Per capita revenues from tissue and hygiene paper by region[108]

The demand for facial tissue and handkerchiefs has been growing at the even faster yearly rate of 6.2%, with a current total demand of around 6 million tonnes per year and a market share of more than 18% in the consumer sector. The RISI forecast for the coming years estimates an average annual growth of 4.2%, with a further increase of market share and a total production of 7.6 million tonnes per year by 2025.

Western Europe is a mature market, with a yearly growth between 1 and 3.1% (Figure) and a current average per capita consumption of 15.7 kg of tissue products per year[109].

[108] Statista Consumer Market Outlook, 2019, Tissue and Hygiene Paper Report - 2019

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

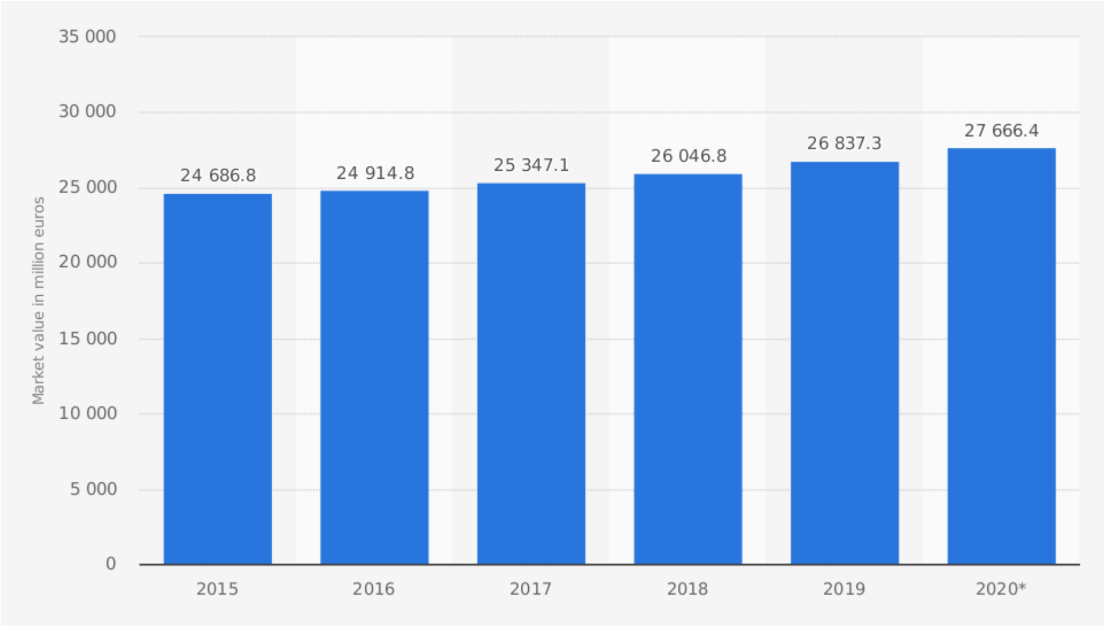

Figure 11: Market value of tissue and hygiene products in Western Europe from 2015 to 2020 (in million euros)[109]

The overall trend in the European consumer tissue sector, is toward higher quality products, with a trend in facial tissue toward upgrading to softer, super premium products.

5.4 The Fine chemicals Market

Chemical market (including pharmaceutical**,** chemicals, plastics and rubber) is projected to grow at a CAGR of 5.7% during the forecast period 2018-2024 (Figure 12). Pharmaceutical segment which has the highest market share expected to generate $140.6billion by 2024 growing at a CAGR of 5.81%. North America is the leading manufacturer of fine chemicals and manufactures around 40% followed by Asian Pacific (APAC) with 26%. Chemical sector accounts for 12% of the EU manufacturing industry's added value and represents a huge source of income to the EU economy and innovation is a pressing need for keeping this sector competitive with other consolidated economies such as Japan and the US. From a European point of view, there is a high interest in 3 research areas related to food packaging. These are new packaging materials that are not derived from oil, the fact that reducing

[109] Euromonitor. (March 4, 2019). Market value of tissue and hygiene products in Western Europe from 2015 to 2020 (in million euros) [Chart]. In Statista. Retrieved July 25, 2019, from https://www.statista.com/statistics/491549/tissue-and-hygiene-market-value-western-europe/

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

packaging waste and environmental impact and that safety and quality of packaged food will be increased compared to nowadays.

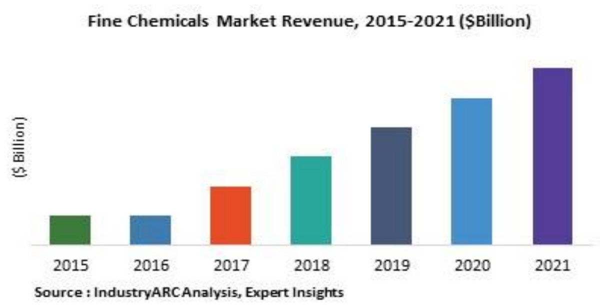

Figure 12. Fine Chemicals Market revenue, 2015-2021[1]

The ECOFUNCO project will promote the development of new bio-based coatings to be used for the formation of biopolymers. These new coatings will help boosting the chemical sector by direct sales and by their take up by the polymer industry sector. With EU directives moving towards the Green Chemistry concept, biopolymers and coatings derived from natural sources are the key factors to promote the evolution of the chemical industry sector and to bring innovation to a highly competitive market. The ECOFUNCO project will contribute to the minimisation of the chemical industry environmental impact by developing biodegradable coatings derived from natural resources which will not have a hazardous effect on the environment. The impact of this project in the Chemical industry will not be only at the technological level; it will set the basis for new processes and new packaging production technologies. These technologies aim at the development of biodegradable and environmentally friendlier material, which minimise the effects on ecosystems not only after use, but also on the production stage, contributing to a greener Europe.

5.5 Disposable food and drink products and relation with agro-food market

Disposable food and drink products items associated with food service industry that are commonly only used once before they are thrown away or recycled. These include, among other items, take-away packaging, straws, plates, cups and cutlery. The world foodservice disposables industry is estimated to expand at a CAGR of 5 to 6% reaching a volume of 21.2 billion US$ by 2021. Around one third of this growth will come from the EMEA region and thus Europe, Africa and Arabia.

The global biodegradable food service disposables market, with paper and paper being the most important ones, reached a value of US$ 2.4 Billion in 2018. Although biodegradable materials do have a noticeable market share in this segment, still around 90% of the items used are neither biodegradable nor bio-based. It is estimated that in 2017 around 346 .000 tonnes of waste were generated from disposable tableware and takeaway packing in Germany alone. This is an increase of 44% compared to 1994.

As disposable is commonly used in outside areas (e.g. for picnics and BBQ) it is not surprising that disposable food service products are a main source for litter in the environment (cf. Figure 13)

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

Impact on the European Agro-food industry: The global agro-food product market is expected to reach $1,148.3 billion by 2018, with good growth over the next five years. With agro-food exports reaching €122 billion in 2014, the EU became the world's number one exporter of agricultural and food products, followed by the US with €121 billion worth of exports. Final products for direct consumption made up most of the EU's exports (Figure 13).

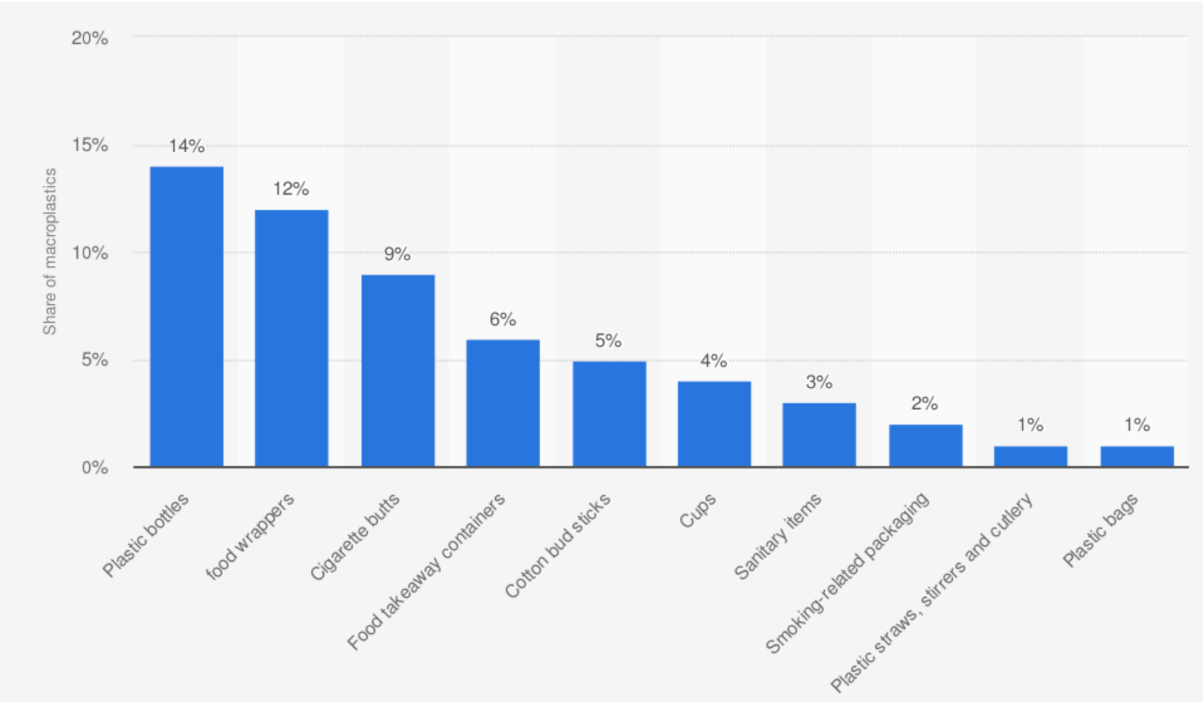

Figure 13: Most prevalent macro-plastics in European freshwater environments as of 2019[110]

Therefore, European Commission is putting rules in place for reducing single-use plastic products, and curbing marine litter. The proposal is focused on the top ten disposable plastic items found on European beaches and in the seas, as well as abandoned or disposed fishing gear. Together, these products account for 70% of waste in Europe's marine environments.

Impact on the European Agro-food industry: The global agro-food product market is expected to reach $1,148.3 billion by 2018, with good growth over the next five years. With agri-food exports reaching €122 billion in 2014, the EU became the world's number one exporter of agricultural and food products, followed by the US with €121 billion worth of exports. Final products for direct consumption made up most of the EU's exports (Figure 14).

[110] Earthwatch Institute, & Plastic Oceans UK. (April 8, 2019). Most prevalent macroplastics in European freshwater environments as of 2019 [Graph]. In Statista. Retrieved July 29, 2019, from https://www.statista.com/statistics/1022984/macroplastics-in-freshwater-europe/

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

The agro-food industry is central to the wider, economic development of Europe as it develops over the next decades. The only possibility for this sector to continue its growth, expansion and technological dominance over the external competing markets is to continuously innovate and rely on R&D as source added value. The main ECOFUNCO project contributions to this sector are:

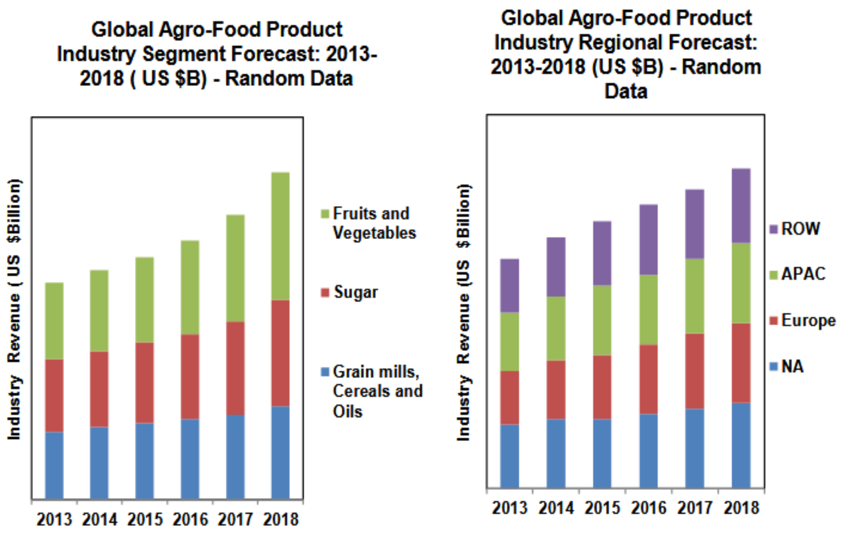

Figure 14: Global Agro-Food Product Industry Segment and Regional Forecast, 2013-2018[1]

- technological development and higher quality of products: better preserved and fresher products will arrive to the consumer as a consequence of improved packaging properties;
- making cultivation of legumes more attractive thus contributing to an improved nitrogen fixation in the soil (less fertilizer use) and helping to close the European protein gap
- increased sustainability: the industry is able to adopt a safe, socially responsible and sustainable approach to food production/commercialisation in an economically-viable way that meets the expectations of society.
- the multidisciplinary and integrated approach to research that will generate knowledge and technologies essential to stimulate the European food and sanitary industry and ensure the industry expands its world-leading position.
- a stronger EU position in world markets for environmental technologies, which will contribute to sustainable consumption, production, delivering sustainable growth and improved competitiveness, while protecting our cultural and natural heritage.

5.6 Impact on consumers and consumer choice.

With the growing sensibilisation of consumers towards greener and wiser choices from the environmental point of view, more attention is paid to products with a lower environmental impact. This concerns the product as a whole and necessarily also includes the packing material,

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

especially critical in the case of food, and sanitary and disposable products. These products are sold in massive quantities and the packaging must be light, resistant and impermeable to gases and liquids, maintain the organoleptic properties of its contents and at the same time present high levels of biodegradability. In terms of impact on consumers and on their choice, the ECOFUNCO project is expected to impact on the following aspects:

1) favour the selection of products that are packed in biodegradable polymeric materials, which per se (or on the monomeric state) do not pose a safety problem to human health due to migration effects and are easily biodegraded upon release on the environment not posing a risk to the ecosystems;
2) consumers will benefit of high-quality products due to the improved preservation properties of the new packaging formulations to the developed on the ECOFUNCO project.
3) consumers' expectations regarding biodegradability and reduction of packaging waste and environmental risk will be met, while at the same time, being able to benefit from improved formulations from the technological point of view. This project is a step forward into the promotion of the sustainability of the growing demand from consumers for packed food and sanitary and disposable products.

5.7 Environmental impact

The growth of the world economy and the rising global population (9 billion by 2050) mean that the Earth's natural resources are being used up fast. Resources need to be managed more efficiently throughout their life cycle, from extraction, transport, transformation and consumption, to the disposal of waste. Resource efficiency is an imperative. This means managing more effectively, producing more value using less material and consuming differently to limit the risks of scarcity and keep environmental impacts within our planet's natural limits. Using resources more efficiently will help us achieve many of the EU's objectives. It will be key in making progress to deal with climate change and to achieve our target of reducing EU greenhouse gas emissions by 80 to 95% by 2050. It is needed to protect valuable ecological assets, the services they provide and the quality of life for present and future generations.

The ECOFUNCO project will contribute to the decrease of the Environmental Impact of packaging goods and disposable products by:

1) developing three new bio-based coating materials (plus coating combinations) to be applied on plastic and cellulose, conferring improved performances compared to currently available products;

2) increasing the recyclability of the packaging polymers versus currently available products;

3) ECOFUNCO will allow to use food and agro-waste biomass: usually these by-products are immediately used for energy recovery while ECOFUNCO will allow for their use in material application

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

pro-longing their life-cycle before final disposal as energy recovery hence adding at least one more step in the cascading use of biomass and improving the overall efficiency of the value chain pursuing the concept of doing more (products) with less (feedstocks).

5.8 Impact on EU policies.

In recent years, the European Union set important economic, environmental and societal goals which have been integrated into European policies. Notably the **European Commission's Roadmap on a resource efficient Europe** (EC, 2011), the **Bioeconomy Strategy** (EC, 2012) and the **EU Action Plan for the Circular Economy** (EC 2015) and the Circular Economy Package launched in 2017. The production of bio-based products fits in all these policies as it is expected to consume less energy and emit less carbon dioxide than fossil-based products.

The new concept of ECOFUNCO takes a significant step towards the efficient use of bio-based material resources and their economically viable production, thus contributing to reach the targets of the above-mentioned policies. In ECOFUNCO, raw materials (agro-food wastes: notably tomato, sunflower, legumes, fish, crustaceous, fungi, cellulose, chitosan and chitin) will be upgraded into renewable alternatives to petroleum-based polymers, with bio-based coatings development technologies that will be based on energy efficient processes and on ecological approaches and green chemistry. The project is also in line with the objectives of the **Eco-innovation Action Plan (EcoAP),** which focus on boosting innovation that results in or aims at reducing pressures on the environment and on bridging the gap between innovation and the market. Considering the **European Environmental Technology Action Plan (ETAP)**, ECOFUNCO contributes to the goals of the action plan by developing eco-efficient ways to convert agro-food wastes into **bio-based coating products**. Environmentally benign technologies developed in ECOFUNCO for efficient agro-food wastes utilization can promote innovation and competitiveness, but also decouple economic growth from environmental impacts. ECOFUNCO contributes to the **Environmental Technology Action Plan** also by supporting the development of eco-technologies and promoting thus foreign investments in environmental technologies leading to increased employment and economic growth within the EU.

The project is also in line with the **European Lead Market Initiative for bio-based products** which points at the need for the fast growth of the industries producing environmentally friendly solutions and approaches, and the necessity for the industry to satisfy various end-user requirements at a competitive cost during their entire life cycle. Finally, the proposal goes in line with Directive 94/62/EC **on Packaging and Packaging Waste**[111] directly contributes to prevent or reduce the impact of packaging and packaging waste on the environment and to ensure the functioning of the Internal

[111] European Parliament and Council Directive 94/62/EC of 20 December 1994 on packaging and packaging waste, *Official Journal L 365 , 31/12/1994 P. 0010 – 0023,* http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:31994L0062:EN:HTML

This project has received funding from the Bio Based Industries Joint Undertaking (JU) under grant agreement No 837863. The JU receives support from the European Union's Horizon 2020 research and innovation programme and the Bio Based Industries Consortium

Market. In addition, the directive contains provisions on the prevention of packaging waste, on the re-use of packaging and on the recovery and recycling of packaging waste.

6 CONCLUSIONS AND FUTURE PERSPECTIVES

ECOFUNCO project comes from previous research experience from the consortium gained in National and European previous projects, focused on biomass valorisation, molecules extraction and cutin, chitin-chitosan and proteins applications. The consortium shared part of the background reported in this document and set the basis for incoming research activity.

The main sources of raw materials to be used in ECOFUNCO project were reviewed with a critical analysis considering benefits and disadvantages of the materials including cost and sustainability, evidencing the presence of reliable providers allowing the acquisition of raw material at a suitable cost. This resulted from a detailed state of the art on the availability, cost, properties of sources from proteins, polysaccharides (chitin, chitosan, microbial cellulose), and cutin was conducted and discussed. The literature survey conducted on bio-based, sustainable coatings to be applied on plastic or cellulose substrates, considering the possible technologies for application and the options for monitoring and consequently control-optimise the processes, outlines the space for innovation targeted by ECOFUNCO consortium.

The survey considers even the strategies for improving compatibility between coating and substrate, evidencing the benefit provided by coatings based on proteins, chitins, and cutin. Sustainability and end of life options for the products developed in ECOFUNCO appear promising considering the challenge of recycling in products currently on the market.

The document even addresses the technical requirements to be met by ECOFUNCO demonstrators to satisfy the expectation of industrial partners, producers and end users of ECOFUNCO products in food packaging, table wares, non-woven tissues, etc

The market for the products targeted in the project (functionalised packaging, table wares, fine chemicals, personal care etc) is huge and there is a wide space for introduction of ECOFUNCO coated materials. The growing awareness of policy makers and consumers versus sustainable, bio-based products, are promising for a positive acceptance of the products on the market, and for ECOFUNCO potential to impact on EU policies.

In the ECOFUNCO project the state of art and market analysis will be regularly updated, with focus on materials, extraction methods, processing, performances that are relevant for the project activities and for objectives achievement. This review work will be valorised by publication in chapters and open access journal and by production of public document to train researchers and consumers versus a more conscious use of packaging and health care products in harmony with our precious environment.