

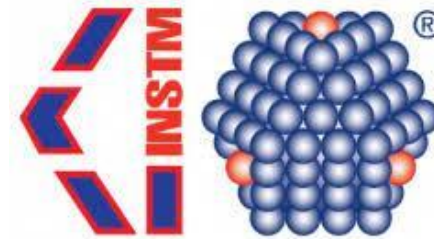
Sustainable coatings for plastic and cellulosic substrates derived from agro-food biomass

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1st October 2026



Projects

We participated as UNIPI-INSTM



Biomass valorization
Protein, cutin, natural fillers



Chitin and chitosan
Packaging, health care



Protein for coating and
packaging



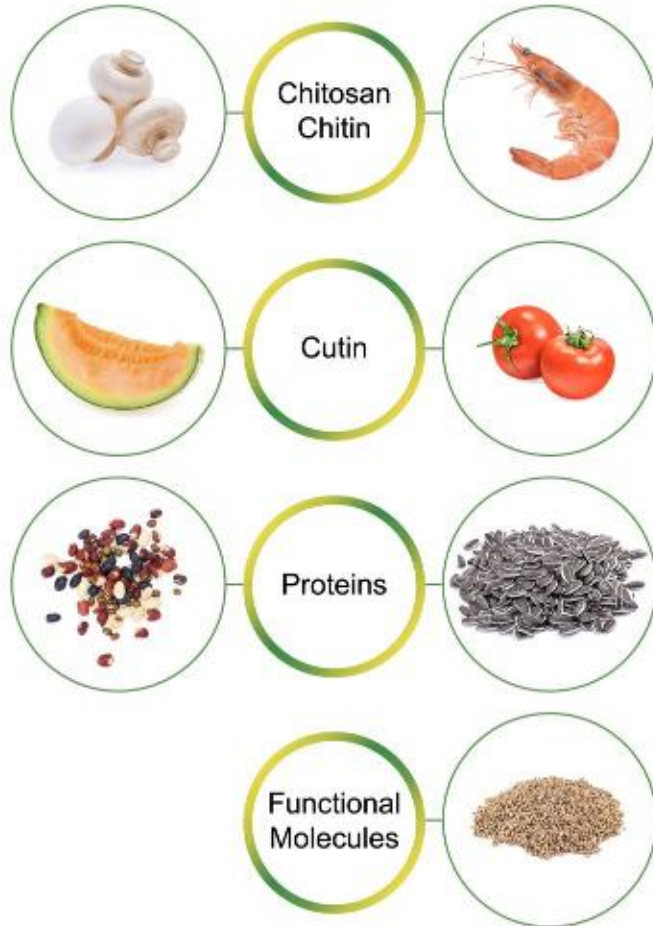
Polyethylene Furanoate



PLA, PHA, Packaging

Strategy

Food and Agro Discarded Biomass



Plastic and Cellulose
+
ecofunco

ecofunco



Personal Care

Anti-Microbial,
Anti oxidant



Disposable Products

Barrier to water and
grease



Multilayer Packages

Barrier to oxygen,
moisture and grease

**ECO sustainable multiFUNctional biobased COatings with enhanced performance and end of life options
GA 837863.**

Products with Biobased Coatings

Barrier Function

Aroma

Gas

Vapour

Sustainability

Packaging Materials

Material Function

Mechanical

Optica

Thermal

Antimicrobial

Processability



Contribution to the bioeconomy

Active biomolecules and macromolecules ,such as microbial cellulose, cutin, protein, chitin and chitosan, are extracted or produced from by products or waste of the agro-food sector. Innovative and green technology for extraction and production.

Celabor (Belgium) produced nano chitins fibrils from commercial chitin (crustaceous, funghi).

Biomass suppliers seeing new opportunities for their waste and by-products

Production of paper-based materials with enhanced barrier and hydrophobic properties, connecting paper industries with agriculture, food-feed sectors

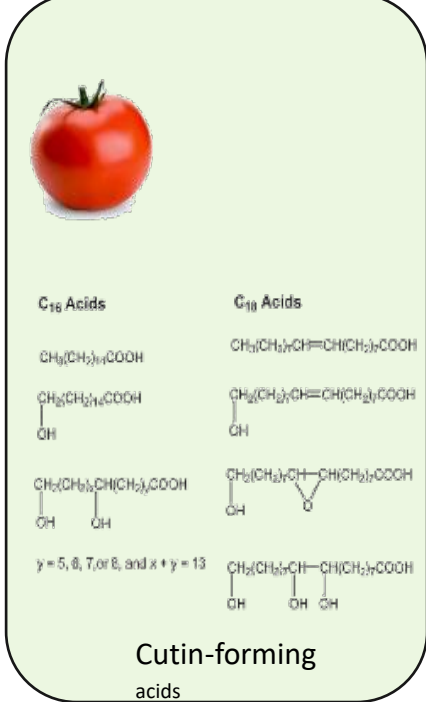
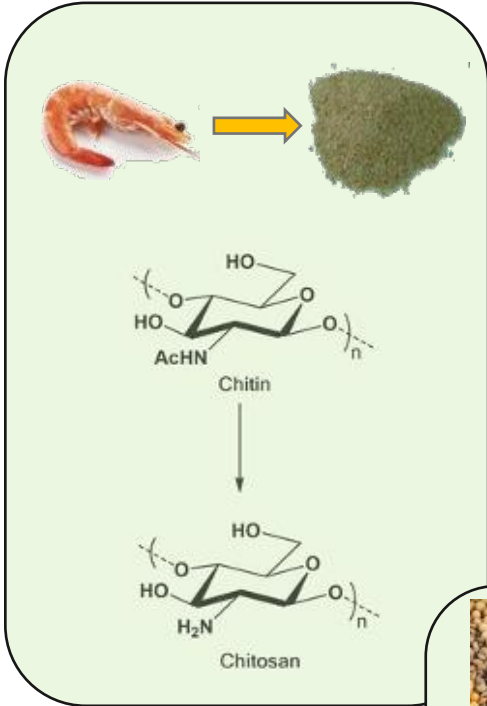


KPI2. New cross-sector interconnection in bio-based economy

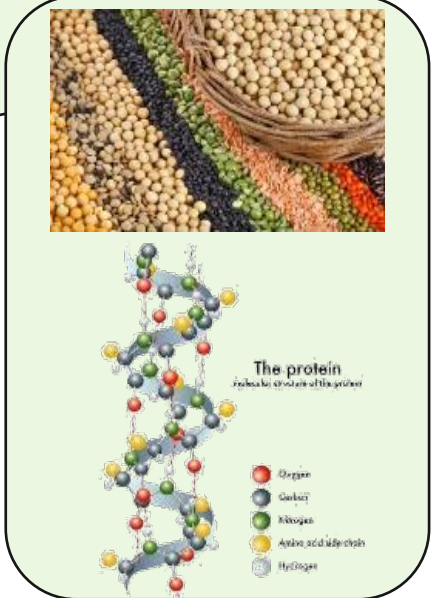
Food residues (shrimps, mushrooms for chitin, chitosan) or the crop residues used in the project:

Tomatoes, legumes, sunflower seeds, melon, leading to valuable molecules for the production of functionalized materials.

Products are being obtained, leading potentially to the demonstration of a high number of individual circular value chains.



ECOFUNCO PRODUCTS

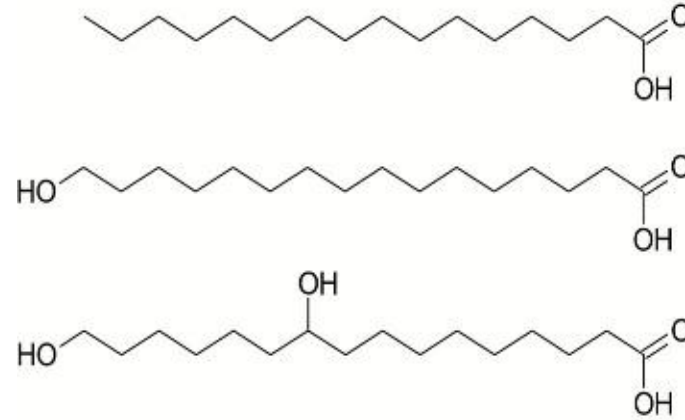


CUTIN for Coatings

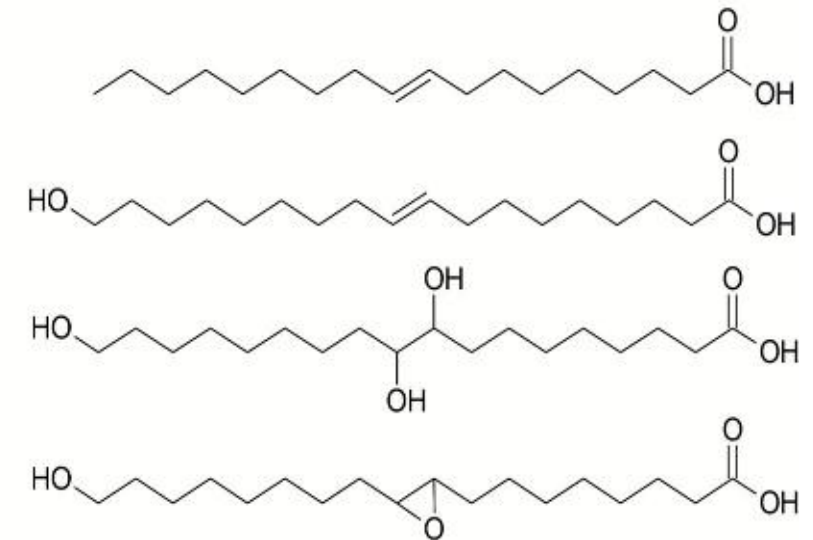
The main component of the cuticle is cutin (up to 80% w/w), a nontoxic, biodegradable, hydrophobic, insoluble and infusible amorphous biopolyester mostly made of interesterified C16 polyhydroxy acids.



C₁₆ monomers:



C₁₈ monomers:

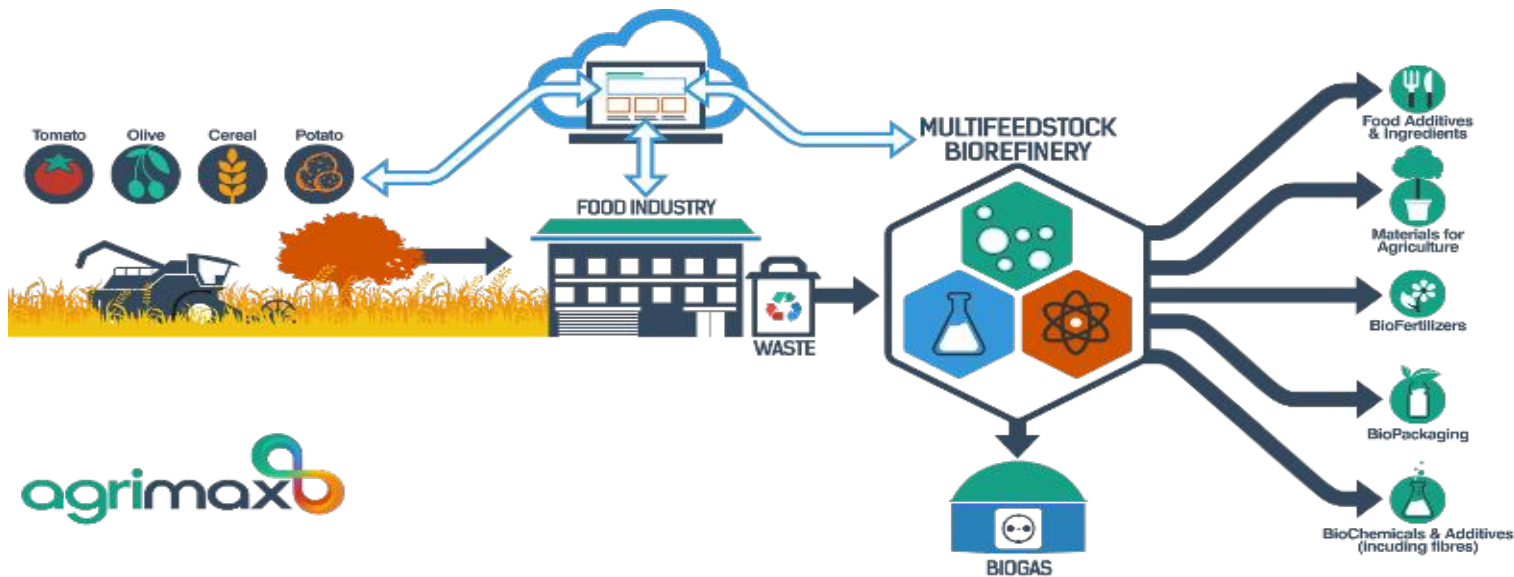


Benítez J.J. et. al, Bio-Based Coatings for Food Metal Packaging Inspired in Biopolyester Plant Cutin, Polymer, 2020, 12, 942; doi:10.3390/polym12040942

Biorefinery success story

Food waste providers of European origin were identified,
For example Cutin mainly produced in Italy (Virginio Chiesa farm) from
tomato skin (BBIJU Demo AGRIMAX GA 720719)

*Agri & food waste valorisation co-ops based on flexible multi-feedstocks biorefinery
technologies for new high added value applications, BBI.VC3.D5. DEMO*
Starting date: 01.10.2016 (5 years)



Toma paint spin off <https://www.it.tomapaint.com/>

<https://www.ssica.it/>



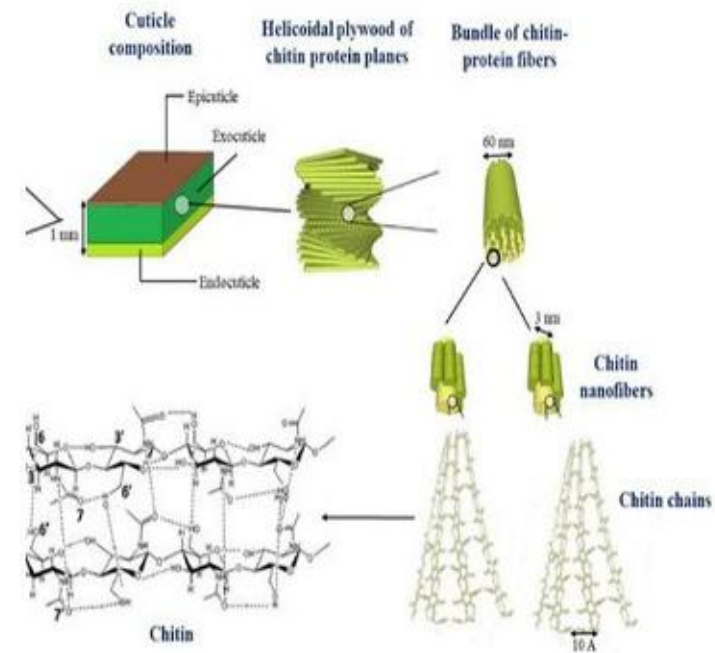
Stazione sperimentale per l'industria delle conserve alimentari, Parma, Italy



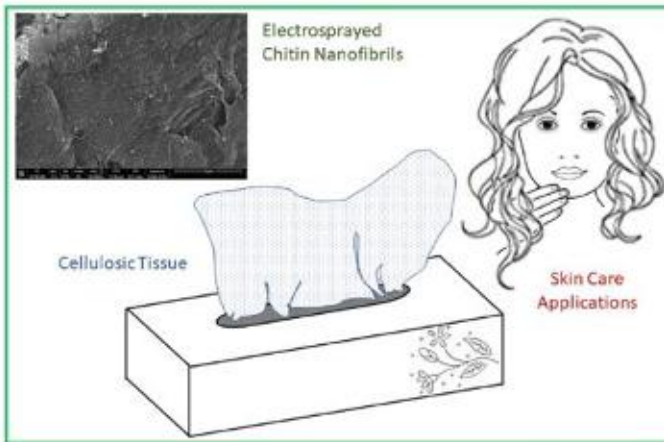
Chitin and chitosan

Nano chitin fibrils

Chitin is the second most abundant polymerized carbon-based macromolecular material found in nature and can be converted to innovative **high-value bio- and eco-compatible materials**. Chitin consists of both **crystalline** and **amorphous** domains, but the elimination of the amorphous phase results in **chitin nanofibrils (CNs)**, which lose their **pro-inflammatory** and **allergenic** character and obtain anti-inflammatory, anti-microbial properties.



Cosmetics have recently focused on **biobased skin-compatible materials**. Materials from natural sources can be used to produce **more sustainable skin contact products** with enhanced bioactivity.



Panariello, L., Coltelli, M.-B, Giangrandi, S., Garrigós, M. C., Hadrach, A., Lazzeri, A., Cinelli, P. (2022) Influence of Functional Bio-Based Coatings Including Chitin Nanofibrils or Polyphenols on Mechanical Properties of Paper Tissues. *Polymers*, vol. 14, n. 11, June 2022, [pag. 2274](https://doi.org/10.3390/polym14112274).
<https://doi.org/10.3390/polym14112274>

KPI5. New bio-based materials: bio-based coatings applied on plastic and cellulose substrates showing improve barrier and anti-microbial properties.

Coatings on Plastics and cellulose

Coating on cellulose substrates tissue paper or cardboard with suspensions of chitin, chitosan, nano fibrils of chitin for anti microbial properties.

Coating on cellulose with cutin for hydrophobic properties

Coating on plastic films, based on compostable biodegradable substrates (biopolyesters based)

Coating with lupine extracted proteins

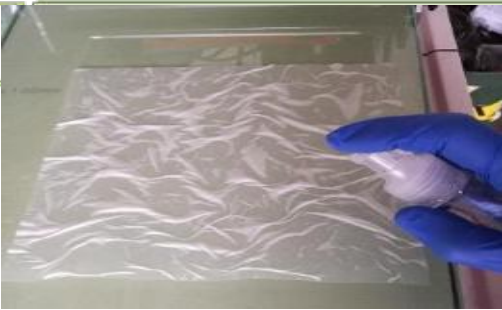
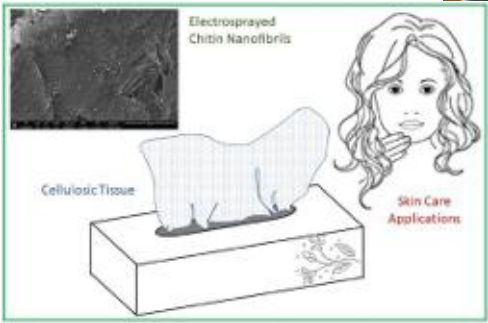
Coating with cutin

Coating with chitin

Assembling of multi layers



Substrate	Without treatment	Pure chitin (Celabor) treatment	Chitin + OLA 1350 treatment	Substrate	Without treatment	Pure chitin (Celabor) treatment	Chitin + OLA 1350 treatment
Biopla I - PESA/PIBV		 Continuous film, slightly adhesive	 Continuous film, excellent adhesion	Biopla I - PLA/PIBV		 Not continuous film, poor adhesion	 Continuous film (but concentration of treatment in the center of the film), slightly adhesive
Biopla II - PLA/PBAT		 Not continuous film, excellent adhesion	 Continuous film, excellent adhesion	Biopla II - PBA/PIBV		 Continuous film, excellent adhesion	 Continuous film, slightly adhesive, presence of sticky material only not treated layer
Tipa C32N - PESA/PBS		 Not continuous film, excellent adhesion	 Continuous film, excellent adhesion	Biopla III - PBA/PIBV		 Not continuous film, excellent adhesion	 Continuous film, excellent adhesion
Tipa C32N - PESA/PBS		 Not continuous film, poor adhesion	 Continuous film, excellent adhesion	Biopla IV - PLA		 Not continuous film, excellent adhesion	 Continuous film, excellent adhesion



Hot Melt Coatings (HMC) on Cellulose Substrate

- Paper substrate: A 150x150 mm sheet of 50 gsm cellulosic board supplied by Huhtamaki (Espoo, Finland).

HMC name	PBSe (wt. %)		OLA 2 (wt. %)	206/3 NL (wt.%)	Chitin (wt.%)	Chitosan (wt.%)	Cutin (wt.%)
HM	100		-	-	-	-	-
HM chitosan	90		9	-	-	1	-
HM chitin	90		9	-	1	-	-
HM cutin	80		-	15	-	-	5

Cutin 20% by weight in water/Ethanol (50/50)
Chitosan: EN GP 8523 (Glentham Life Science, Corsham, UK)
Polyadipate ester with trade name of Glyplast® 206/3 NL provided by Condensia Química (Barcelona, Spain)
Lactic Acid Oligomer (OLA): Marketed as Glyplast® OLA 2
Poly(butylene sebacate) (PBSe), Technipol® 709, SIPOL (Mortara, Italy),
Chitin nanofibrils by CELABOR (Verviers, Belgium), 1.5% by weight water suspension

Hot Melt Coatings on Paper and Plastic



Processing workflow of the dispersion of biomolecules, production of Hot Melt Coatings granules and application onto paper and plastic substrate

Cobb test on HM cutin coated sample, at 1 min, 30 min and 12 h.¶

Sample¶	Cobb at 1 minute¶	Cobb at 30 minute¶	Cobb at 12 hours¶
HM <u>cutin</u> on paper¶	0 g/m²¶	2 g/m²¶	< 3 g/m²¶

Hot Melt Coatings Anti Microbial

Total Bacteria – Contact Plate (PCA Agar)							
Sample	Colonies in Test 1	Colonies in Test 2	Colonies in Test 3	Colonies in Test 4	Colonies in Test 5	Area (cm²)	UFC/100cm²
HM chitosan on paper	0	1	0	2	0	23.5	2.6
HM chitin on paper	0	0	2	0	0	23.5	1.7
Total Moulds – Contact Plate (SDA Agar + bacterial inhibitor)							
Sample	Colonies in Test 1	Colonies in Test 2	Colonies in Test 3	Colonies in Test 4	Colonies in Test 5	Area (cm²)	UFC/100cm²
HM chitosan on paper	0	1	3	0	0	23.5	3
HM chitin on paper	0	0	0	0	0	23.5	0

HMC with chitin showed no mould colony growth and an exceptionally low UFC/cm³ count for bacterial contamination. Similarly, total moulds contact plate tests demonstrated a highly inhibiting effect on mould colony growth for HM chitosan, with samples exhibiting near-zero microbial growth. This strong antimicrobial and antimould activity observed in both HM chitin and HM chitosan can be attributed to the intrinsic properties of the biomolecules.

Coatings on Plastic

Oligomeric lactic acid (OLA) with a Mw = 1350 Da by Condensia Quimica (Barcelona, Spain).

0.5g of OLA in ethanol for each gram of dry chitin mass, then mixed with chitin suspension

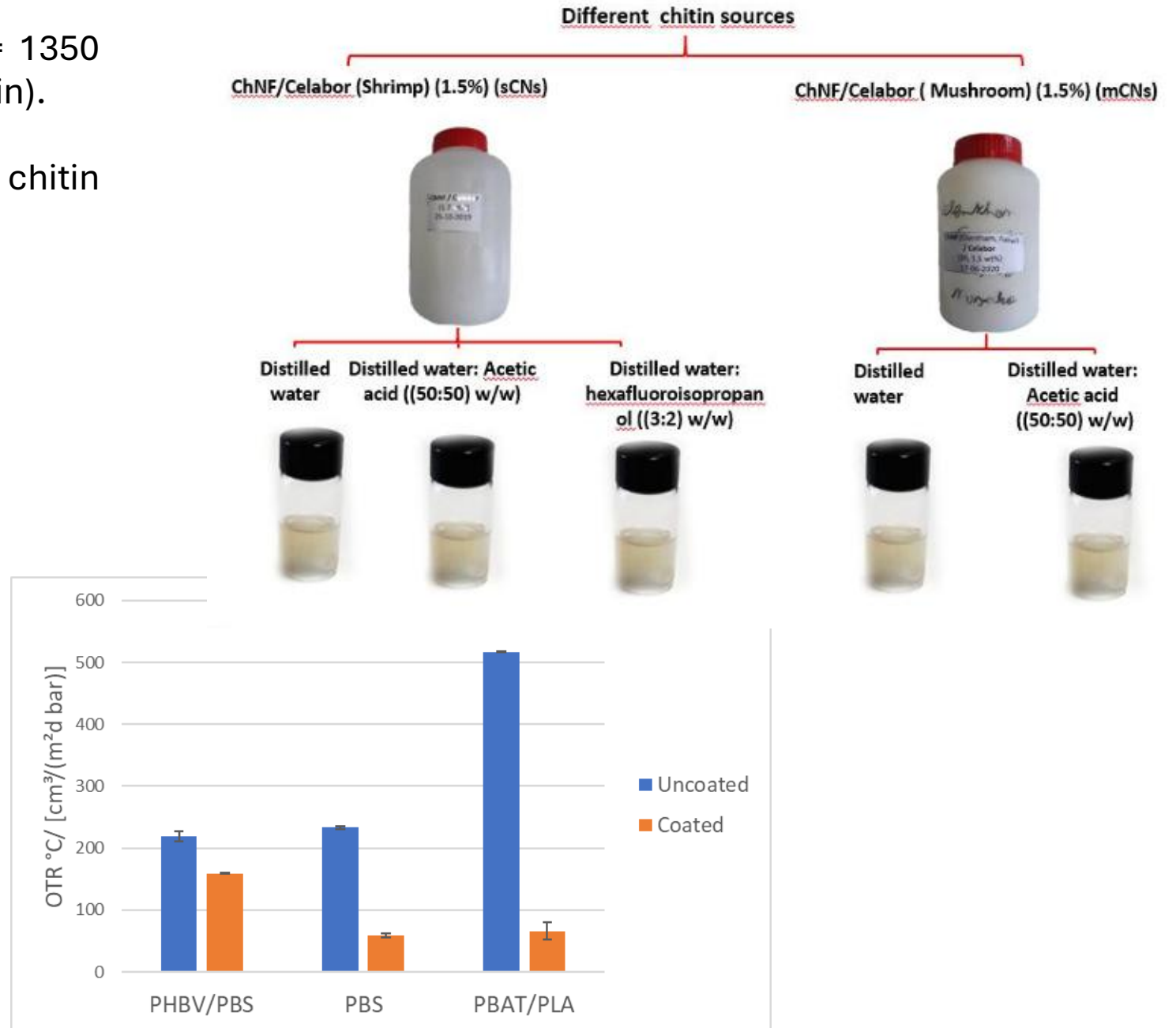
Plastic substrates: V Planet Bioplastic s.r.l. (Pisa, Italy), film based on PBSA/PHBV

TIPA (Hod Hasharon, Israel) PBS based film

Bio-mi Ltd. (Put Brdo, Croatia) films based on PBAT/PLA

Automatic applicator Zehntner ZAA 2300

Application rate was set to 10 mm/sec, the width of applicator was 20 cm and the thickness was set to 150 μm



Anti Microbial on Plastic Substrates

Results of antibacterial test on untreated films used as reference¶

Sample¶	Recovery after 24 h incubation (cells/cm²)¶	
	E. coli¶	S. aureus¶
PHBV/PBS¶	15600¶	687¶
PBS¶	5938¶	156¶
PLA/PBAT¶	6130¶	325¶

Results of antibacterial tests on films treated with chitin (Aldrich) and chitin (Aldrich) + OLA1350¶

Sample¶	Recovery after 24 h incubation (cells/cm²)¶		Antibacterial activity (R)¶	
	E. coli¶	S. aureus¶	E. coli¶	S. aureus¶
PBSA/PHBV + Chitin shrimp 3h 1.5%¶	12500¶	356¶	0.09¶	0.29¶
PBSA/PHBV + Chitin shrimp 3h 1.5% + OLA ¶	6875¶	13¶	0.35¶	1.73¶
PBSA/PHBV + Chitin fungi 3h 1.5% + OLA ¶	10625¶	58¶	0.16¶	1.08¶
PLA/PBAT + Chitin shrimp 3h 1.5% + OLA ¶	350¶	38¶	1.25¶	0.93¶
PBS + Chitin shrimp 3h 1.5% + OLA¶	0.33¶	3¶	4.25¶	2.79¶
PBS + OLA¶	17500¶	1875¶	0¶	0¶

L. Panariello, M.B. Coltelli , A. Harich, F. Braca4 S. Fiori, A. Haviv, F. Miketa, Lazzeri, A. Staebler , V. Gigante, P. Cinelli, Polymer, “Antimicrobial and gas barrier crustaceans and fungal chi-tin-based coatings on biodegradable bioplastic filmsPolymer, MDPI, 2022, under revision.

Formulations based on hot melts (UNIPI/INSTM)

HMC matrix

Technipol®707

PolyButylene Sebacate

Tm: 64°C

Biodegradable

Biobased content: 60% up to 100%



Plasticizers

GLYPLAST OLA2

Lactic Acid Oligomer

Fully biodegradable



GLYPLAST 206/3 NL

Polyester of adipic acid

Insoluble in water



Active Molecules



Celabor Chitosan
from Shrimps



Celabor Glenham Chitin
from Shrimps



SSICA Cutin
from tomato peels

before



after

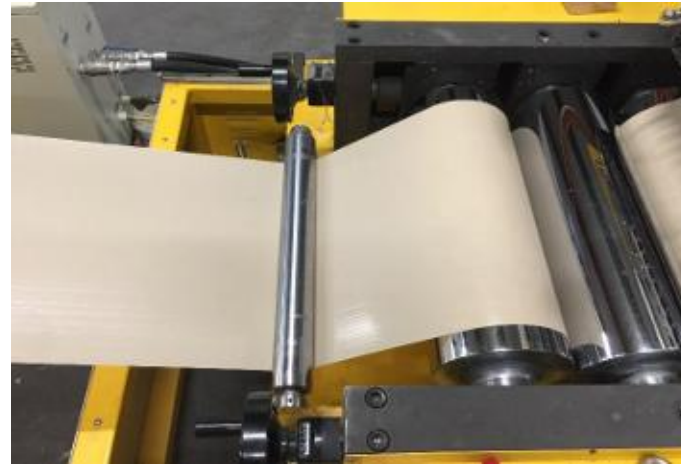


Ultraturrax



Patent applied: Hot melt biobased polymeric formulations for coating applications containing active biomolecules with antimicrobial and water repellent properties, Patent Application Number 102022000016212; 29 July 2022, UNIPI, INSTM, PLABIO.

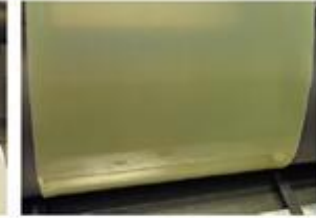
Prototypes production



Billerud / MLP



Biomi 7 flexible / MLP



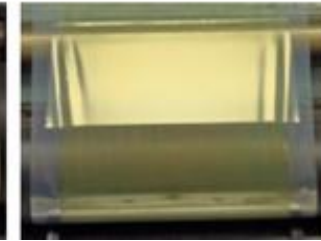
Biomi 7 rigid / MLP



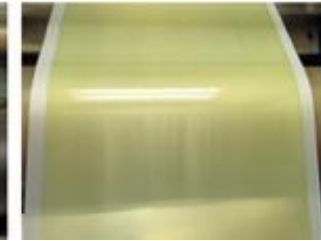
PackPro / MLP



TIPA317 / MLP



PlaBio I / MLP



HM Matrix



HM Chitosan 1



HM Chitin 1



HM Cutin 5

HM= Hot melts

Anti Microbial on Plastic Substrates with Hot Melts Coatings

	Judgement for Antibacterial activity (R)	
	<i>E. coli</i>	<i>S. aureus</i>
BIO-MI 2 Chitin + OLA	MEDIUM	LOW
BIO-MI 3 Chitin + OLA	MEDIUM	HIGH
BIO-MI 7 Chitin + OLA	MEDIUM	MEDIUM
TIPA 302 + Cutin resin	MEDIUM	MEDIUM
TIPA 317 + Cutin resin	MEDIUM	MEDIUM
TIPA 317 + Chitin resin	LOW	MEDIUM
TIPA 302 + Chitin resin	MEDIUM	MEDIUM

- All the coated samples provided relevant antimicrobial properties both for *E. Coli* and *S. Aureus*;
- The **chitin + OLA** coating on BIO-MI substrates was n effective for *E. Coli* than for *S. Aureus*;

	BIOMI7NEW		BM7N + HM Chitin1		BM7N + HM Chitosan1		BM7N + HM Cutin5	
	lab-scale							
	MD	CD	MD	CD	MD	CD	MD	CD
Elastic Modulus (GPa)	1,00	0,89	0,50	0,42	0,45	0,44	0,35	0,23
Stress at Break (MPa)	30,79	26,42	6,37	7,90	12,18	7,62	15,82	12,54
Elongation at Break (%)	12,37	6,70	2,53	2,62	4,89	2,77	12,90	8,68
Critical Tearing Energy (N/mm)	11,61	60,14	4,62	10,44	7,49	11,44	18,71	24,42
Elmendorf Force (N)	0,26	0,39	0,26	0,21	0,31	0,34	0,42	0,39

Project Demonstrators – Multi layers

Compostable substrates:

Plastic: (BIOMI, TIPA, PLABIO)

Layers of:

Platic/Cutin

Protein/Plastic

Chitin

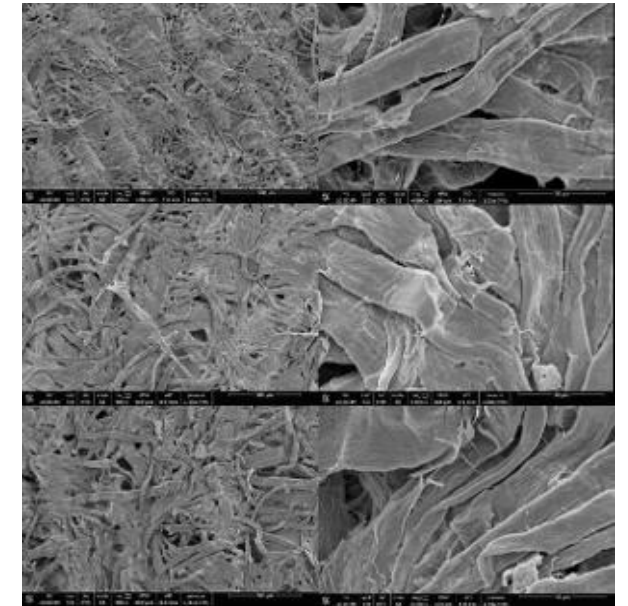


Biodegradable substrates:
Cellulose: (Huthamaki,
Organikimia)

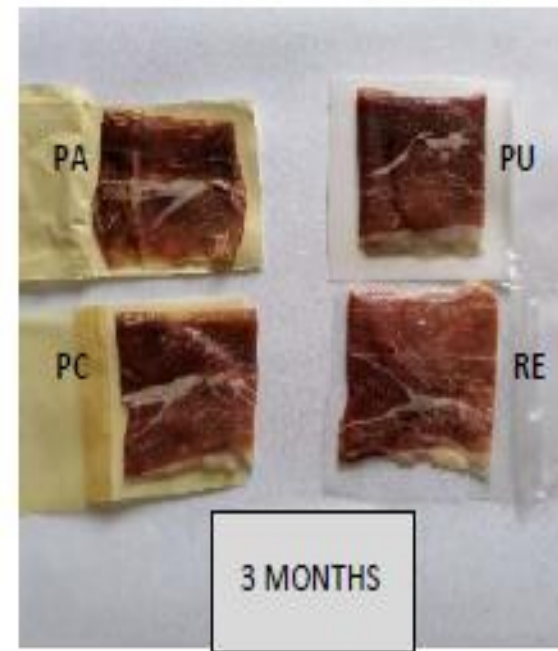
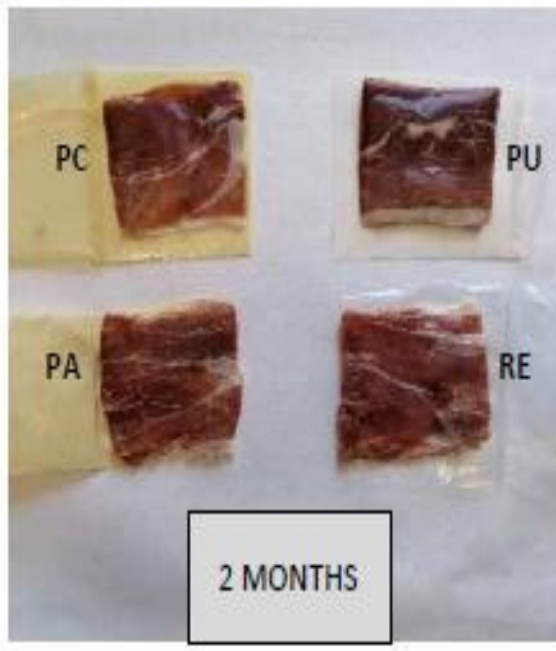
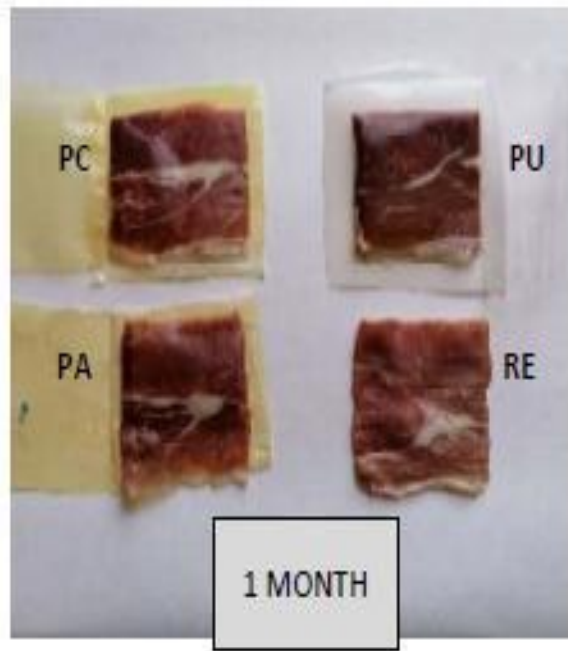
Layers of:

Cutin

Chitin, polyphenols



Micrographs of the three layers of facial tissue, coated with polyphenols



Ham packed in the four experimental materials at 1, 2 and 3 months of shelf life.

RE= Reference, PU= Plastic uncoated, PC= plastic coated, PA= Paper with coatings.

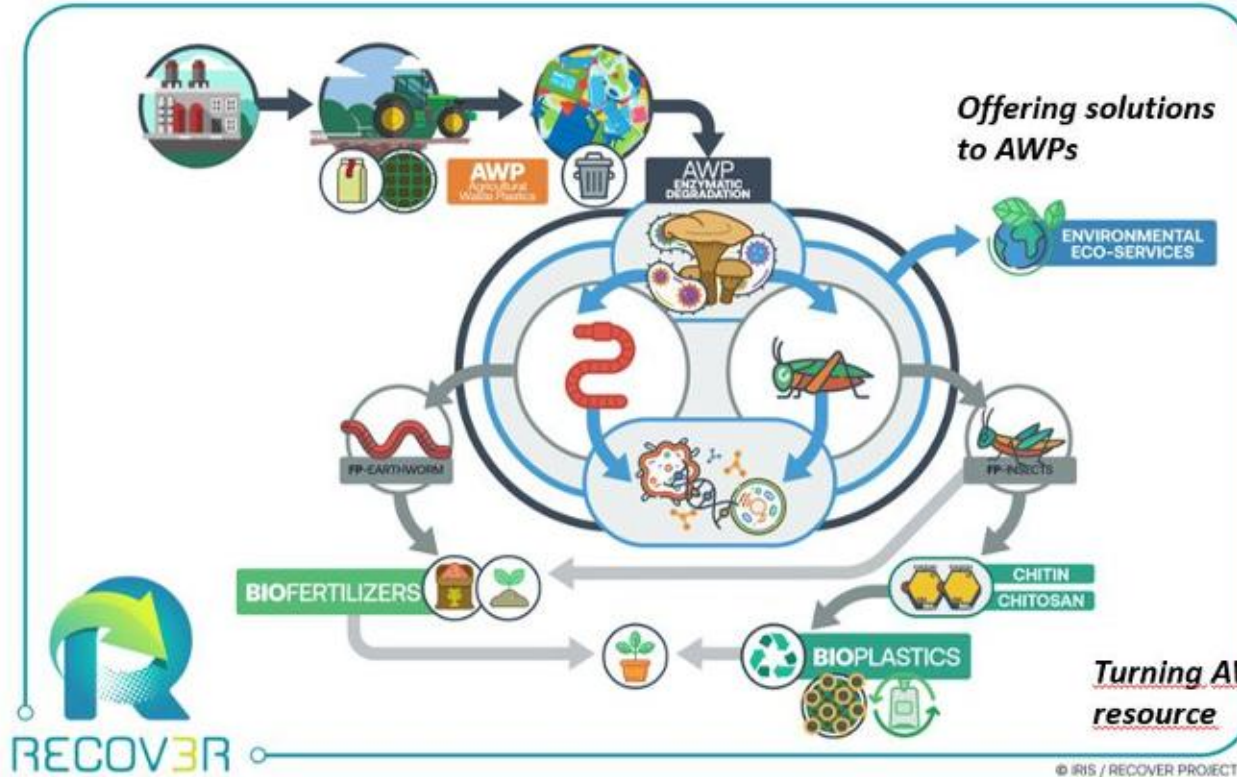
- All ECOFUNCO materials tested were found to be suitable for preserving the quality of Parma Ham up to 1 month shelf-life.
- The oxidation index values and chemical and sensory changes observed at 60 and 90 days suggested that shelf-life should not be extended beyond 30 days for hams packed in PU and PA.
- In the PC packs, the quality of the ham was comparable with the reference materials (RE) at least up to 90 days. Therefore, the presence of coatings allowed the shelf-life to be extended by at least two months compared to materials without coatings.

Recover solutions & innovations

Agri-food Waste
Plastics (AWPs)

Combining new
enzymes,
microorganisms,
insects &
earthworms

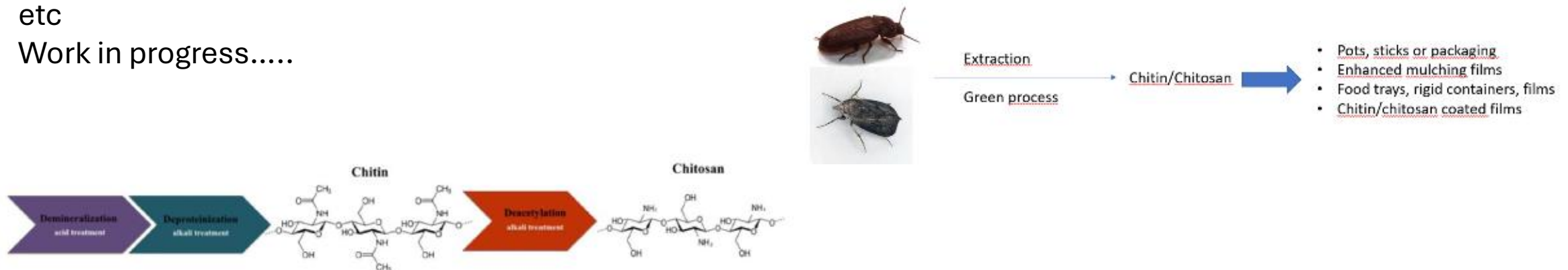
Products



Development of innovative biotic symbiosis for plastic biodegradation and synthesis to solve their end of life challenges in the agriculture and food industries

GA 887648

Chitin and chitosan: Chemical extraction, enzymes, ionic liquids, etc
Work in progress.....



CHITIN/CHITOSAN FROM INSECT



**Mealworm (*Tenebrio Molitor*)
Chitosan**



**Mealworm (*Tenebrio Molitor*)
Chitin**



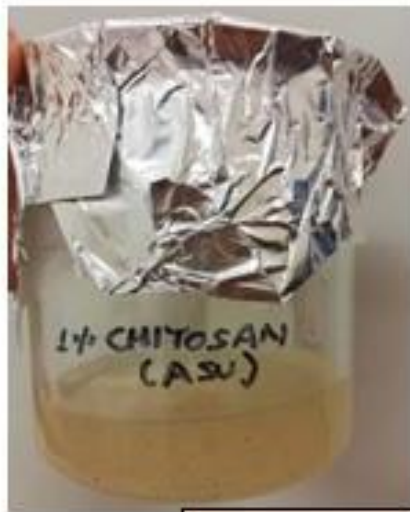
- Insect and larvae protein are important for animal and human feeding and the remaining biomass/ exoskeletons is a no value waste.
- Chitin extracted from exoskeleton has great potential to be used as coating in packaging, or in cosmetic and health care product industries.

Chitin and Chitosan Water Solution

- Chitosan from shrimps (Sigma-Aldrich) and mealworm (*Tenebrio Molitor* Larvae) were dissolved in an aqueous solution acidified with 1% acetic acid and plasticised with 1% (w/v) glycerol to improve filming properties.
- Chitin from shrimps (Sigma-Aldrich) and mealworm (*Tenebrio Molitor* Larvae) were dissolved in a solution of water and heated formic acid (HCOOH) in silicon oil bath until 70°C for 11 hours with different concentrations.



Shrimp Chitosan Solutions



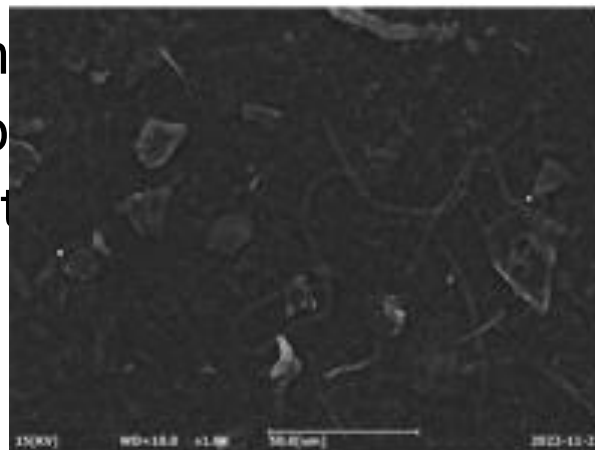
Mealworm Chitosan Solutions



Mealworm Chitin Solution

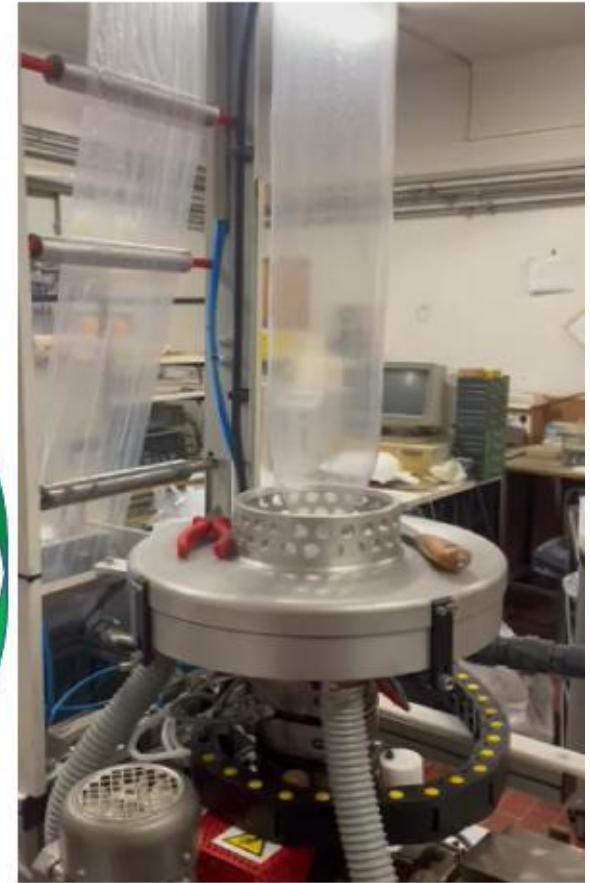
Chitin and Chitosan Anti Microbial

- Functional activity of chitin as **an anti-microbial agent** is known to be **enhanced** when chitin is used in the form of nano-fibrils.
- Chitin nano-fibrils can provide **stable and uniform dispersion coatings**.
- The nano-fibrils are produced by fragmenting chitin and suspending the nano-fibrils in water, this allows **easier application of chitin**, considering the difficulty to achieve chitin solubilization.



https://www.celabor.be/site/index.php?id_surf=&surf_lang=en

Coatings on Films, Anti Microbial



Wet coatings on plastic film substrates

Coating mixture preparation:

1. suspension of OLA 0.25 wt.%/vol.% in H_2O /EtOH 50/50 v/v
2. ChNF (1.0%wt shrimps, 1.0%wt insect)
3. 1.5%wt chitosan in acidic water (1.0%wt Acetic acid)

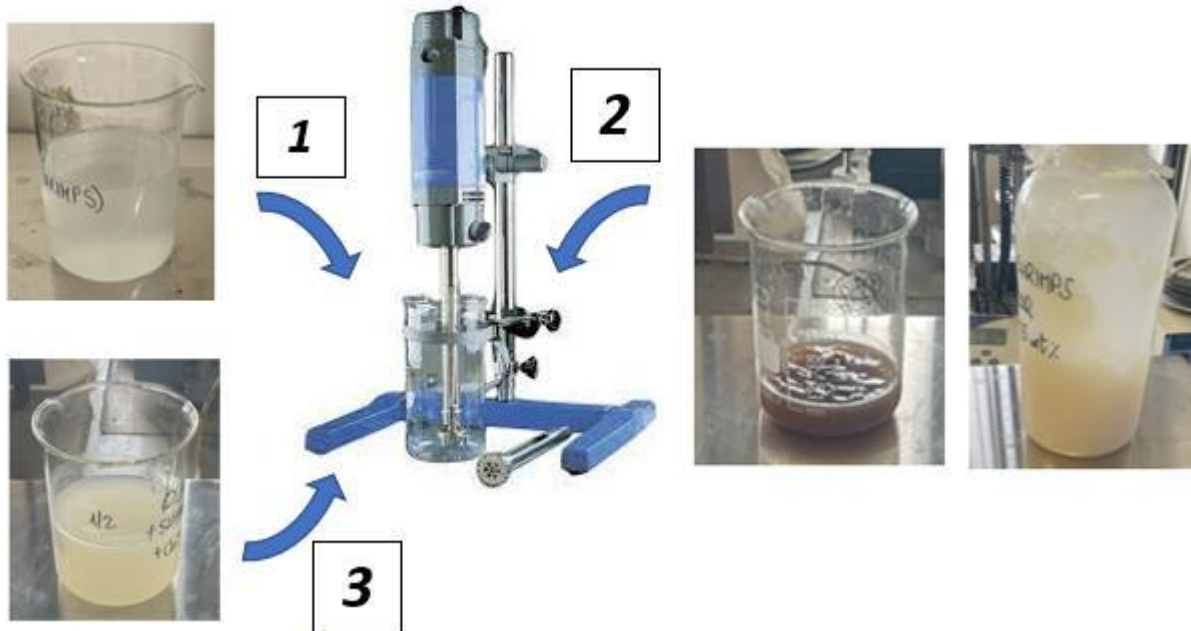
Coating formulation:

OLA/ChNF/Chitosan
0.5/1/1 (wt. ratio)

PBS Food Packaging



PBAT/Starch Mulching



Validated on Perishable Food

Shelf-life study (5 °C, 14 days)

6 samples (100 g breast chicken fillets) per treatment:

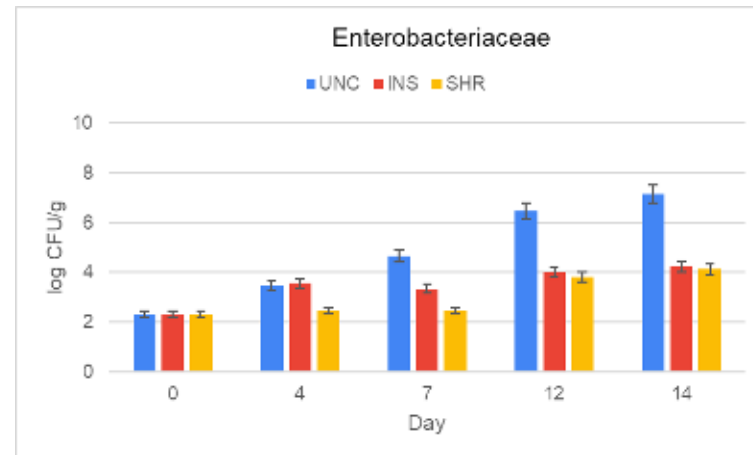
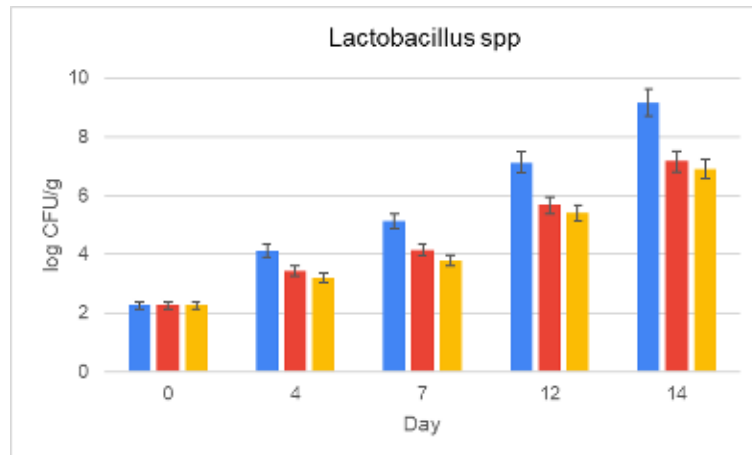
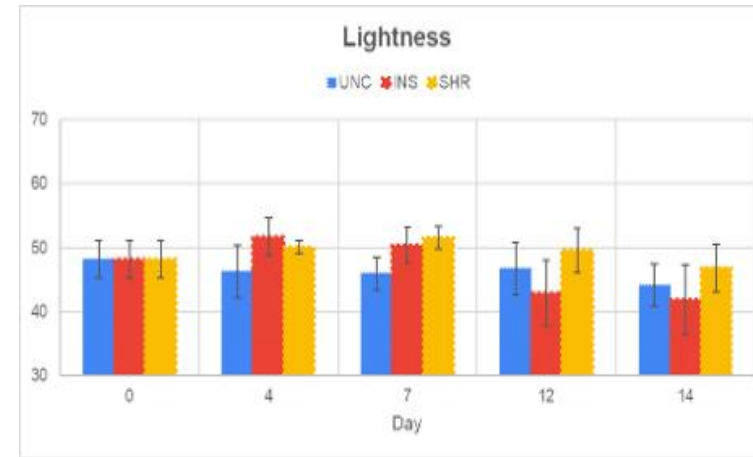
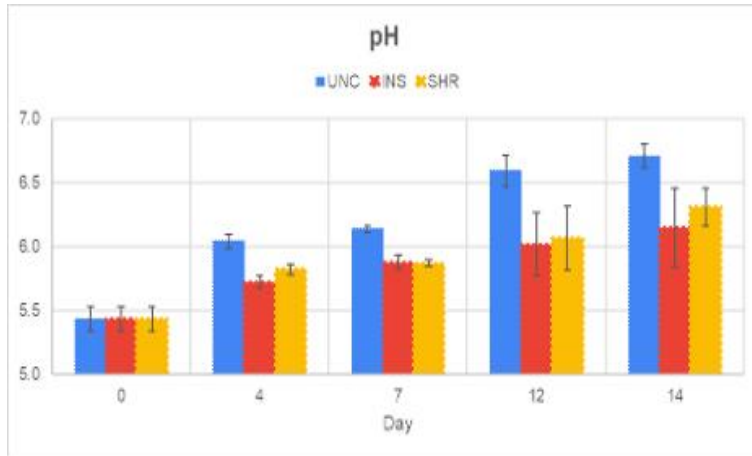
- ***Uncoated***
- ***Coated Nanochitin from Shrimps***
- ***Coated Nanochitin from Insects***



<https://recover-bbi.eu/iris-technology-solutions-s-l-iris-spain/>

- Physicochemical Analysis: pH, colour, TBARS
- Microbial Analysis: Mesophiles, Psychrophiles, Molds & Yeast, Lactobacillus, Enterobacteriaceae

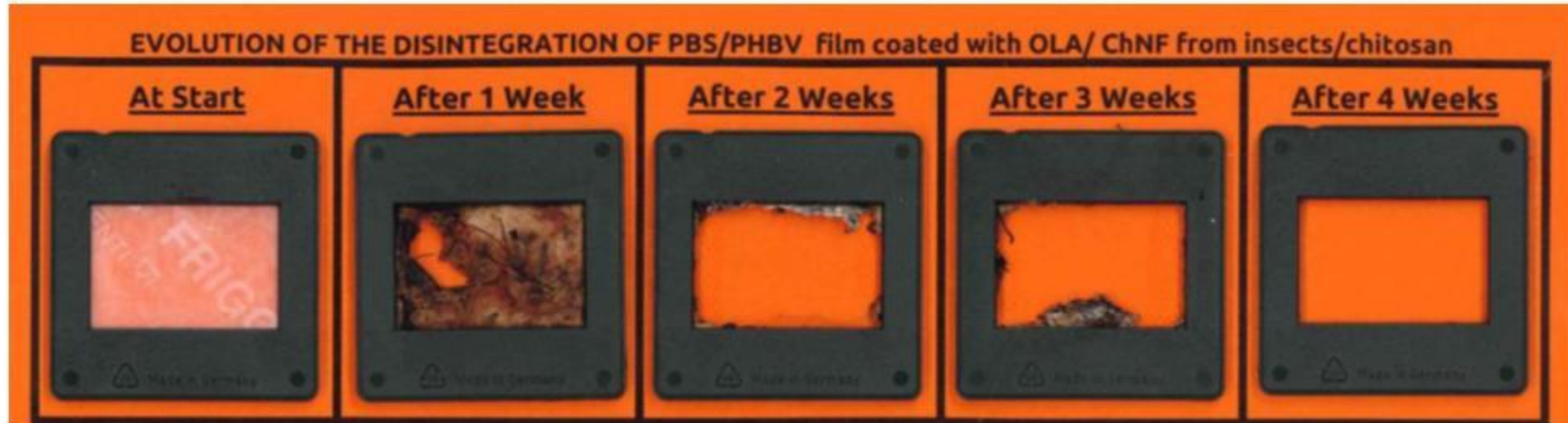
Wet coatings on plastic film substrates



- Nanochitin coatings, derived from insects and shrimps, exhibit potential as effective inhibitors of microbial proliferation and lipid oxidation, thus extending the shelf-life of chicken fillets.

Compost Ability is maintained even with coating

- Chitosan coating did not hinder disintegration under industrial composting (complete disintegration within 4 weeks as for the film without coating)*



Conclusions and Perspectives

- i) Use of biobased resources derived from by-products of food and agri-industrial sector, and of bio-based functional molecules can be sustainable (ethic, environment, cost), with synergies and logistic approach.
- ii) Highly performing, economically and environmentally sustainable bio-based coating for plastic and cellulosic substrates can be prepared with biomass derived molecules (protein, chitin, cutin, polyphenols..) and applied on plastic and cellulose substrates
- iii) Cutin can confer water barrier properties without compromising recyclability and biodegradability
- iv) Chitin and chitosan can be applied on bioplastic and cellulosic substrates for oxygen barrier and antimicrobial properties and is efficient on perishable food, nano chitin is easier to apply and more efficient
- iv) Sustainability and cost depend from substrates, extraction efficiency, and process ability of the biomolecules. Chitin and chitosan, are quite advanced, cutin needs refining and optimization.

Challenges in the transition from fossil-based to bio-based

- Higher cost of innovative processes not yet optimised
- Get a regular, reliable delivery of biomass feedstocks
- Personnel with relevant training and expertise
- Adapt production lines to the bio-based materials
- Achieve comparable performances (or better) versus petro-based with moderate cost increase
- Have proper recognition of the benefits for transition to circular economy (fair LCA, support for extra costs, support from legislations)

THANKS TO ALL RECOVER AND ECOFUNCO PARTNERS



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Thanks to Green ChemLab Team



Maurizia Seggiani



Patrizia Cinelli



Sara Filippi



Miriam Cappello



Damiano Rossi



Irene Anguillesi



Marco Sandroni



Anna Silvia Citernesesi



Chiara Pedrotti



Riccardo Gallorini