EURECA-PRO Conference
on Responsible Consumption and Production 2023

Processing and optimizing polyhydroxyalkanoates: circular materials of the future for use as innovative food packaging material

Prof. dr. ir. Mieke Buntinx
Chania, 28 Sep 2023

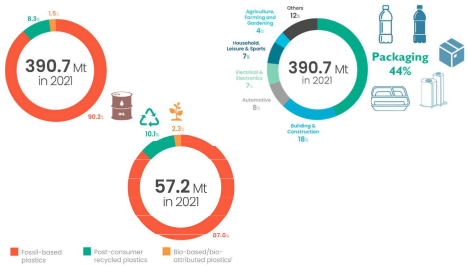
MPR&S Materials and Packaging Research & Services
IMO-IMOMEC UHASSELT imec

Overview

1. Polyhydroxyalkanoates: circular materials of the future for use as innovative food packaging material
2. Development of nanocomposite films to achieve active packaging materials

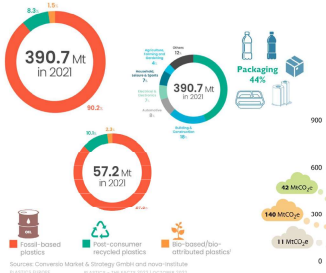
Plastics production, applications and consequences

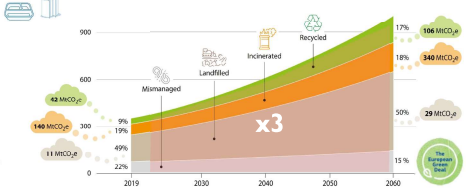
World and European Plastics production (2021)



Plastics production, applications and consequences

World and European Plastics production (2021)





Source: Global Plastics Outlook: Policy Scenarios to 2060 (OECD, 2022)

Packaging in a circular economy

Policy goals

- All (plastic) packaging should be reusable or recyclable in an economically viable way in 2030
- Packaging and Packaging Waste Regulation (Nov 2022)
- …

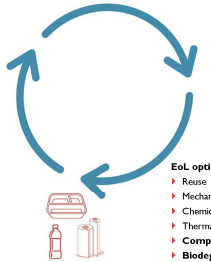
BIOPLASTICS

Bioplastics can significantly contribute to tackle the challenges of the Green Deal, BUT:

- They require a transparent and reliable regulatory environment
- Biodegradability must be seen as an extra means of recovery at EoL
- Compostability should fall within the definition of (organic) recycling
- Defossiling the economy by biobased plastic should be acknowledged
- Biobased content should be considered equal to recycled content
- Innovations in biobased and biodegradable plastics must be promoted

Polyhydroxyalkanoates are promising circular plastics

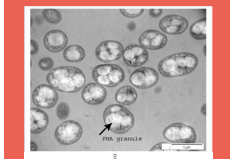
Biobased and biodegradable polymers

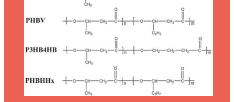


EoL options

- Reuse
- Mechanical recycling
- Chemical recycling to monomers
- Thermal recycling
- **Composting**
- **Biodegradation**

POLYHYDROXYALKANOATES





Reguart P, Buntinx M, et al. Polyhydroxyalkanoates for Food Packaging Applications in Reference Module in Food Science, Elsevier (2019).

Challenges of polyhydroxyalkanoates

Research strategy

- Using bioplastics such as PHAs, could address various SDGs.
- However, **cost, processability and functional performance** remain crucial factors to compete with fossil-based plastics.
- To improve their competitiveness, a promising and popular strategy is to incorporate nanoparticles, creating **advanced nanocomposite materials.**

IMO-IMOMEC UHASSELT imec

Tiekstra S, Dopico A, Koivula H, Lahti J, Buntinx M. Holistic Approach to a Successful Market Implementation of Active and Intelligent Food Packaging. Foods (2021).

Nanocomposites for use as active packaging materials

The final objective

- Today, the world wastes and loses around a third of the food it produces while almost 690 million people go hungry.
- **Active packaging concepts** interact with the packaged product or the atmosphere inside the packaging to protect valuable nutritional components, prevent spoilage or loss of quality, and prolong shelf life [6].
- How can we incorporate ZnO or Ag NPs to obtain safe antimicrobial packaging materials made from biobased and biodegradable PHAs?

IMO-IMOMEC UHASSELT imec

Use of PHA as packaging material

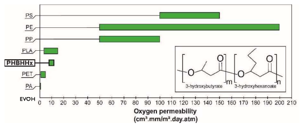
Characterization of gas permeability and other physical-mechanical properties of PHBHHx films

MPR&S Materials and Packaging Research & Services IMO-IMOMEC EURECA-PRO

Use of PHBHHx as packaging material

Gas permeability properties of PHBHHx films

- **PO_2** = 8.3 ± 0.2 $cm^3 \cdot mm \cdot m^{-2} \cdot day^{-1} \cdot atm^{-1}$ at 23°C; 0% RH ★★
- **PH_2O** = 1.42 ± 0.04 $g \cdot mm \cdot m^{-2} \cdot day^{-1}$ at 23°C; 0% RH
- **PCO_2** = 54 ± 1 $cm^3 \cdot mm \cdot m^{-2} \cdot day^{-1} \cdot atm^{-1}$ at 23°C; 0% RH

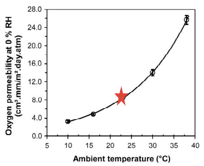


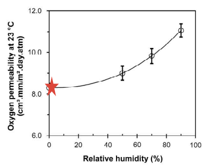
IMO-IMOMEC UHASSELT imec

Use of PHBHHx as packaging material

Gas permeability properties of PHBHHx films

- **PO_2** = 8.3 ± 0.2 $cm^3 \cdot mm \cdot m^{-2} \cdot day^{-1} \cdot atm^{-1}$ at 23°C; 0% RH
- **PH_2O** = 1.42 ± 0.04 $g \cdot mm \cdot m^{-2} \cdot day^{-1}$ at 23°C; 0% RH
- **PCO_2** = 54 ± 1 $cm^3 \cdot mm \cdot m^{-2} \cdot day^{-1} \cdot atm^{-1}$ at 23°C; 0% RH





IMO-IMOMEC UHASSELT imec

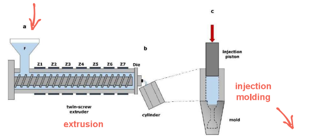
Vandewijngaarden J, Murariu M, Dubois P, Carleer R, Yperman J, Peeters R, Buntinx M. Gas Permeability Properties of PHBHHx. J Polym Environ. (2014).

Strategies to enhance PHA processing and properties

Optimization of melt processing parameters for PHBHHx

PHBHHx
base material, (Kaneka)

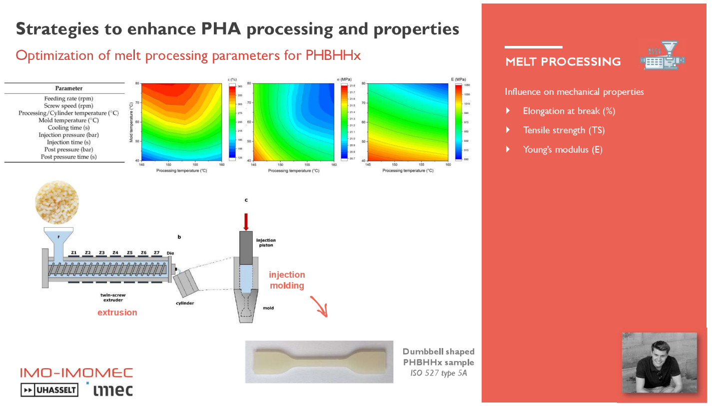
Parameter	Value
Feeding rate (rpm)	4
Screw speed (rpm)	Variable
Processing/Cylinder temperature (°C)	Variable
Mold temperature (°C)	Variable
Cooling time (s)	Variable
Injection pressure (bar)	200
Injection time (s)	15
Post pressure (bar)	150
Post pressure time (s)	10

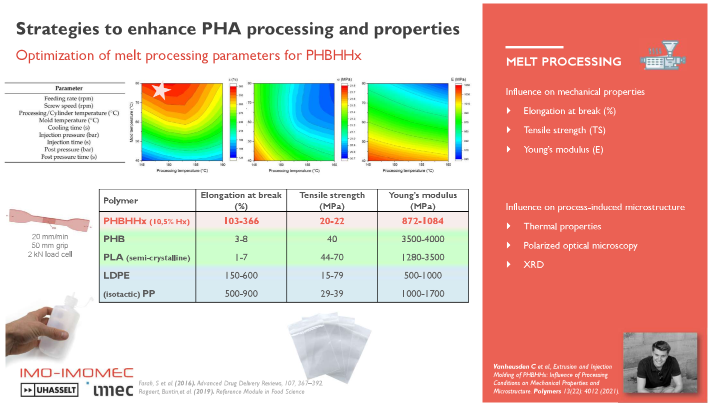


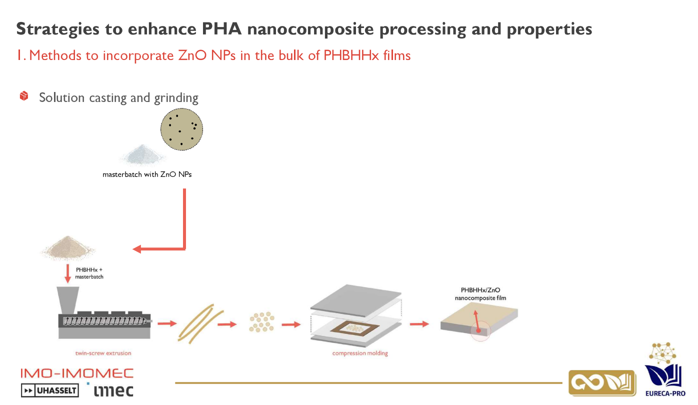
Dumbbell shaped PHBHHx sample
ISO 527 type 5A

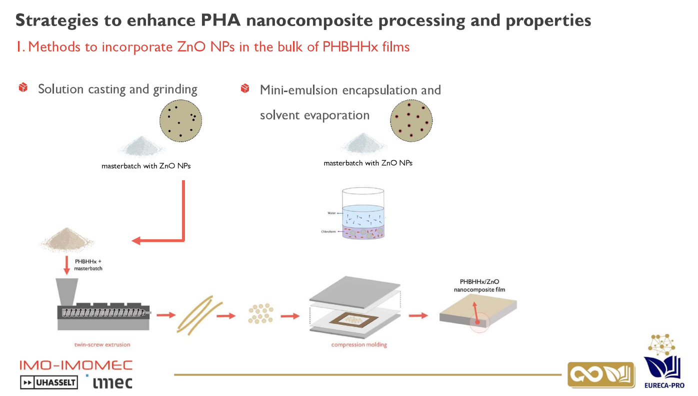
IMO-IMOMEC UHASSELT imec

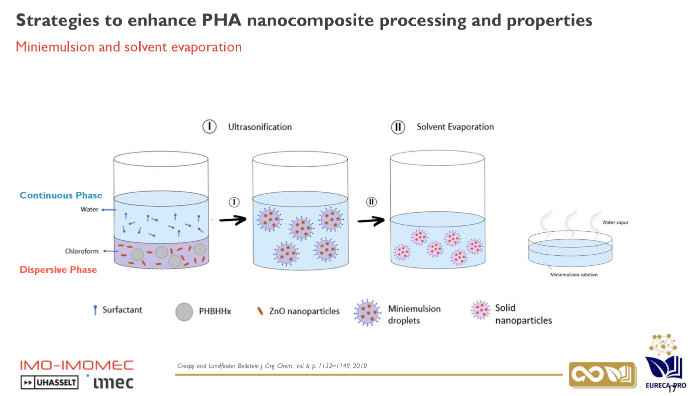
Mieke Bunitnx

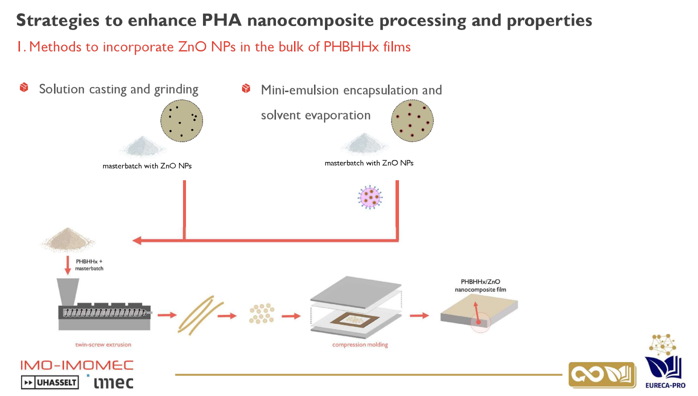

Strategies to enhance PHA processing and properties
Optimization of melt processing parameters for PHBHHx
Parameter
Feeding rate (rpm)
Screw speed (rpm)
Processing/Cylinder temperature (°C)
Mold temperature (°C)
Cooling time (s)
Injection pressure (bar)
Injection time (s)
Post pressure (bar)
Post pressure time (s)
extrusion
injection molding
Dumbbell shaped PHBHHx sample
ISO 527 type 5A
IMO-IMOMEC
UHASSELT
imec
MELT PROCESSING
Influence on mechanical properties
Elongation at break (%)
Tensile strength (TS)
Young's modulus (E)


Strategies to enhance PHA processing and properties
Optimization of melt processing parameters for PHBHHx
20 mm/min
50 mm grip
2 kN load cell
Polymer | Elongation at break (%) | Tensile strength (MPa) | Young's modulus (MPa)
PHBHHx (10,5% Hx) | 103-366 | 20-22 | 872-1084
PHB | 3-8 | 40 | 3500-4000
PLA (semi-crystalline) | 1-7 | 44-70 | 1280-3500
LDPE | 150-600 | 15-79 | 500-1000
(isotactic) PP | 500-900 | 29-39 | 1000-1700
IMO-IMOMEC
UHASSELT
imec
Farah, S et al. (2016). Advanced Drug Delivery Reviews, 107, 367–392.
Ragaert, Buntin,et al. (2019). Reference Module in Food Science
MELT PROCESSING
Influence on mechanical properties
Elongation at break (%)
Tensile strength (TS)
Young's modulus (E)
Influence on process-induced microstructure
Thermal properties
Polarized optical microscopy
XRD
Vanheusden C et al., Extrusion and Injection Molding of PHBHHx: Influence of Processing Conditions on Mechanical Properties and Microstructure. Polymers 13(22): 4012 (2021).


Strategies to enhance PHA nanocomposite processing and properties
1. Methods to incorporate ZnO NPs in the bulk of PHBHHx films
Solution casting and grinding
masterbatch with ZnO NPs
IMO-IMOMEC
UHASSELT
imec
EURECA-PRO


Strategies to enhance PHA nanocomposite processing and properties
1. Methods to incorporate ZnO NPs in the bulk of PHBHHx films
Solution casting and grinding
Mini-emulsion encapsulation and solvent evaporation
masterbatch with ZnO NPs
masterbatch with ZnO NPs
IMO-IMOMEC
UHASSELT
imec
EURECA-PRO

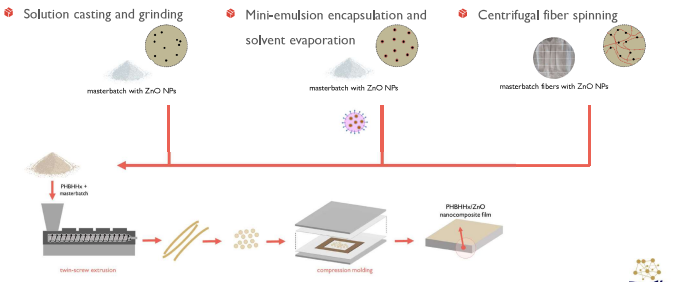

Strategies to enhance PHA nanocomposite processing and properties
Miniemulsion and solvent evaporation
Ultrasonification
Solvent Evaporation
Continuous Phase
Water
Chloroform
Dispersive Phase
Surfactant
PHBHHx
ZnO nanoparticles
Miniemulsion droplets
Solid nanoparticles
IMO-IMOMEC
UHASSELT
imec
EURECA-PRO


Strategies to enhance PHA nanocomposite processing and properties
1. Methods to incorporate ZnO NPs in the bulk of PHBHHx films
Solution casting and grinding
Mini-emulsion encapsulation and solvent evaporation
masterbatch with ZnO NPs
masterbatch with ZnO NPs
IMO-IMOMEC
UHASSELT
imec
EURECA-PRO

Strategies to enhance PHA nanocomposite processing and properties

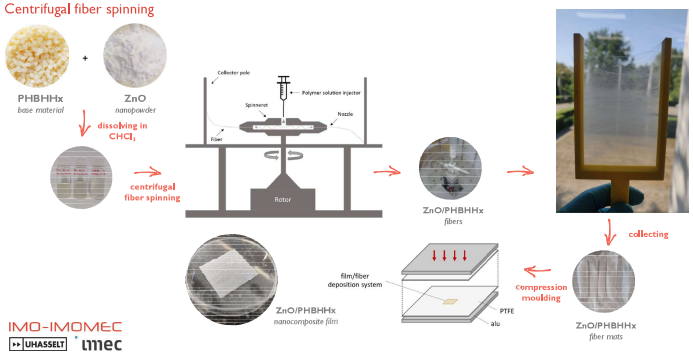
1. Methods to incorporate ZnO NPs in the bulk of PHBHHx films

- Solution casting and grinding
- Mini-emulsion encapsulation and solvent evaporation
- Centrifugal fiber spinning



Strategies to enhance PHA nanocomposite processing and properties

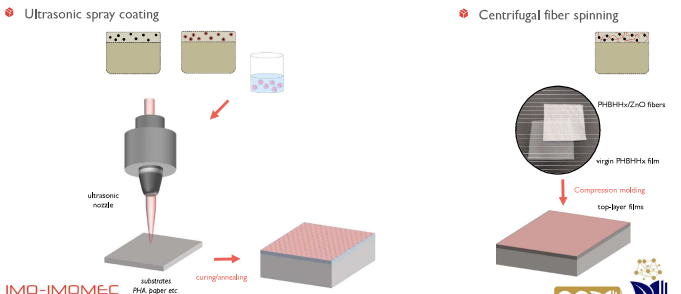
Centrifugal fiber spinning



Strategies to enhance PHA nanocomposite processing and properties

2. Methods to deposit ZnO NPs on top of PHBHHx films

- Ultrasonic spray coating
- Centrifugal fiber spinning



Strategies to enhance PHA nanocomposite processing and properties

Incorporation of ZnO NPs | Ag NPs

- Melt blending – compression molding
- Melt extrusion – injection molding
- Melt extrusion – compression molding
- Centrifugal fiber spinning
- Ultrasonic spray coating
- Mini-emulsion

- Dispersion quality
- Thermal stability
- Crystallization behavior
- Color & opacity
- UV-VIS transmittance
- Gas permeability
- Tensile properties
- Antimicrobial properties

- Potential migration in consumer and postconsumer conditions

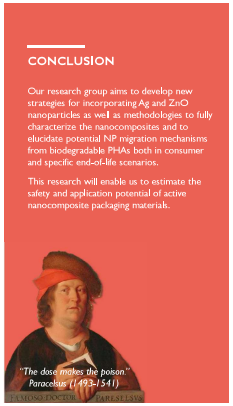
?

Safe use of biodegradable nanocomposite materials

SP-ICP MS to measure potential migration

- Public concern about the potential risks related to migration of NPs from packaging into food is associated with insufficient knowledge about their safety and toxicity, especially if the host material is a biodegradable polymer. This drives authorities to use precautionary principles and handle the issue conservatively.
- Therefore, the value-chain of PHA products from design through processing, value enhancement, and disposal should be strategic, considering **safety** as well as **legislation** → taking into account the European Green Deal, regulation with regard to packaging and packaging waste, and (active) food contact materials is currently very dynamic in the EU!

European Green Deal: Putting an end to wasteful packaging, boosting reuse and recycling https://ec.europa.eu/commission/presscorner/detail/en/ip_22_7155 (2022)
Revision of EU rules on food contact materials. https://ec.europa.eu/info/law/better-regulation/have-your-say/initiatives/12497-Revision-of-EU-rules-on-food-contact-materials_en (2021)



Thank you for your attention!

mieke.buntinx@uhasselt.be

Special thanks to: Chris Vanheusden, Dries Hermans, Mohsin Abbas, Jens Vandewijngaarden Wouter Marchal, Wim Deferme, Pieter Samyn, Naveen Reddy, Anitha Ethirajan, Roos Peeters Jan D'haen, Louis Pitet, Robert Carleer, Jan Yperman & co-workers of MPR&S and imo-imomec