



INSA



In Mold Electronics on Poly(lactic acid)

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Smart Plastics Congress 2022

BIOANTENNA project

PhD work
at the INSA Lyon labs

Project partners



3D Plastronics lab (AMPERE), Villeurbanne
Plastics processing site (IMP), Bellignat



Antennas development
and characterization



3D Plastronics



Polymer materials processing and characterization,
study of screen printing inks on polymers



Funding from AURA Region

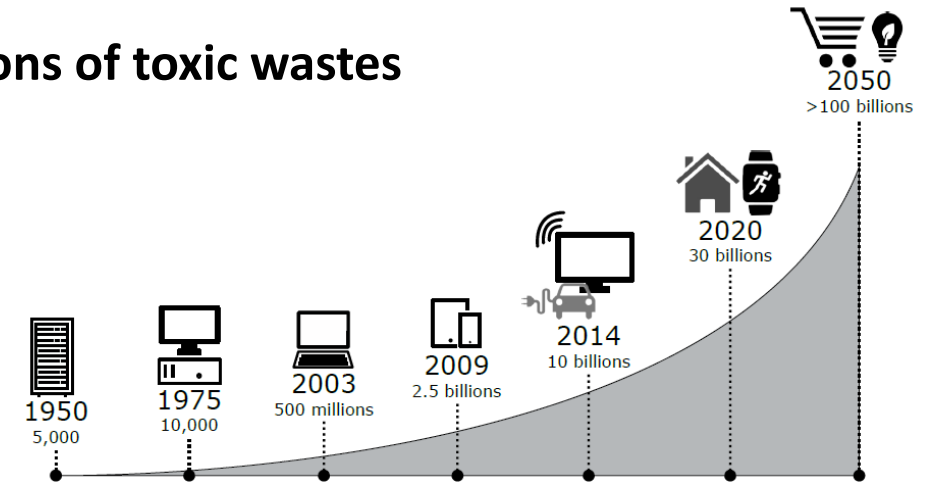
1. Context and objectives of the BIOANTENNA project
2. Polymer materials and IME technology
3. Scientific challenges
4. Experimental results
 - Materials characterization
 - Thermoforming
 - Injection molding
5. Conclusion and perspectives

- IoT (Internet of Things) revolution towards direct interaction between objects thanks to an internet network
 - 30 billions IoT worldwide in 2020¹
- **Why plastronic IoT antennas for energy storage and harvesting ?**



2019, France: 1,5 billion batteries \approx **32 500 tons of toxic wastes**

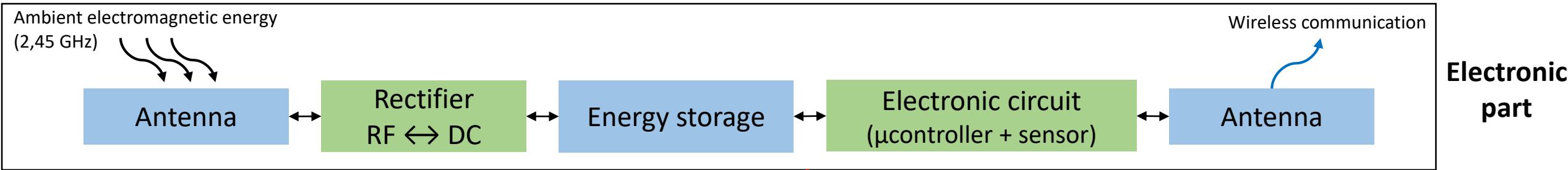
Less than **50% collected**
35% recycled
30% thrown in domestic wastes
 → increasing environmental pollution



[1] Agence de la transition écologique (ADEME), *Piles et Accumulateurs – Rapport annuel 2020– Données 2019*, accessed 13/05/2021

[2] Maurizio CAPRA et al., *Edge Computing: A Survey On the Hardware Requirements in the Internet of Things World*, Department of Electrical, Electronics and Telecommunication Engineering, Politecnico di Torino, 10129 Torino TO, Italy, accessed 13/05/2021

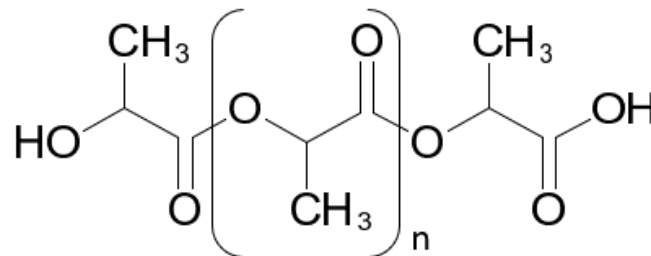
- Replacing small batteries with energy harvesting device integrated in 3D plastronic object



In Mold Electronics (IME) on Poly(lactic acid) (PLA)

Discussed here

- Why PLA ?
 - Biosourced (corn starch) and biodegradable (industrial compost)
 - Very high rigidity
 - Great chemical reactivity

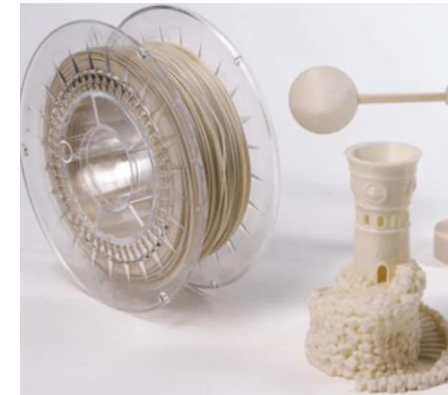


- Two thermoplastic polymers:
 - **Poly(lactic acid) (PLA)**, sustainable alternative for the environment
 - **PolyCarbonate (PC)**, reference material in IME

- PLA already used for:

- Extrusion
- 3D printing
- Injection molding

→ Interesting choice for IME



- Main objective: determine the compatibility of PLA at each step of the IME process

Pictures sources accessed 28/03/2022: <https://www.3dnatives.com/asa-impression-3d-26082020/>

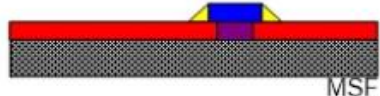
<https://french.alibaba.com/product-detail/high-quality-stretch-PLA-wrapping-film>

<https://french.alibaba.com/product-detail/100-biodegradable-compostable-plastic-pla-resin>

The IME technology

- Used to give a 3D shape to the device which enables better integration of the desired electronic functions to the object
- Technology advantages: innovation in design, features, reliability

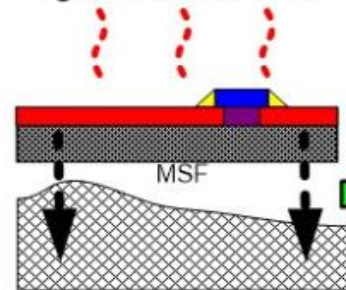
Serigraphy step: Patterning of a PC sheet with conductive ink and soldering of electronics SMT devices



- Polycarbonate MSF
- Conductive ink
- Surface Mount Device

(a)

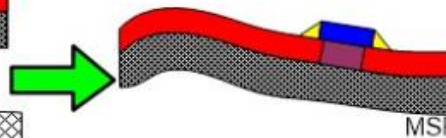
Hot embossing against a 3D tool



- Solder paste
- Embossing tool

(b)

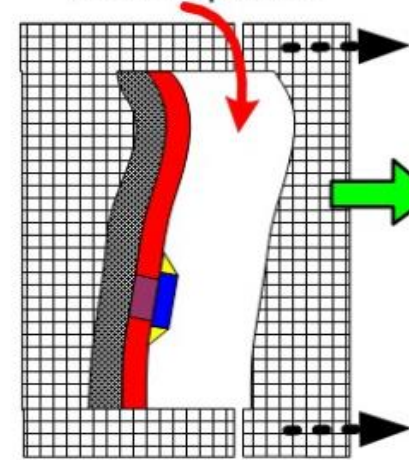
Thermoformed part



- Injection mold
- Glue

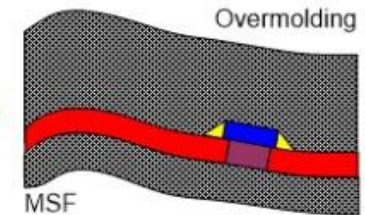
(c)

Injection molding with PC pellets



(d)

Final part



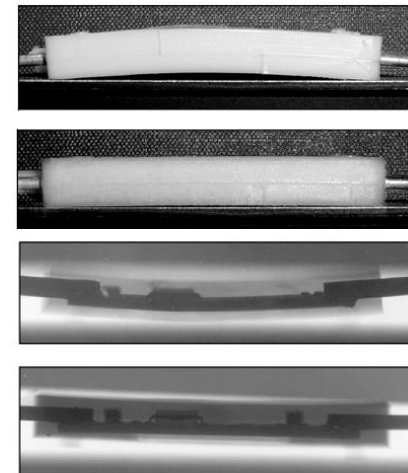
(e)

IME process steps

- Process in development with amorphous PC
- Not studied with semi-crystalline PLA

- Compatibility of inks with PLA films
 - Adhesion?
 - Low transition temperature T_g of PLA → During curing after inking?
- Adhesion “polymer/ink+electronic components” during thermoforming?
- Adhesion “polymer/polymer” and “circuit/polymer” during injection molding?
- Thermoformability of PLA with screen printed inks and glued components (cracks, delamination of inks on PLA)?
- Injection molding: thermal shrinkage inducing warping, early deterioration, ink delamination or component shearing?

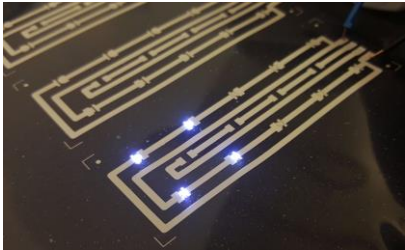
Example of warping after injection molding³



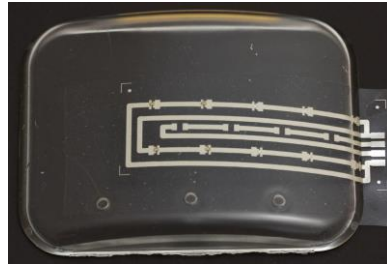
[3] N. J. Teh et al., “Statistical optimisation of thermoplastic injection moulding process for the encapsulation of electronic subassembly,” *J. Electron. Manuf.*, Sep. 2000

- Independent works published since the beginning of the project:

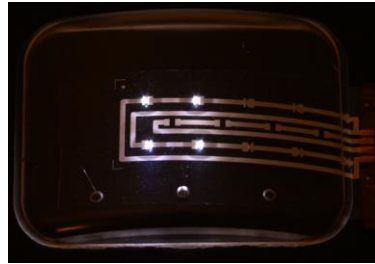
- Välimäki et al. (sept. 2020)



LED foil prepared on PLA film



LED foil prepared on PLA substrate overmoulded with PLA



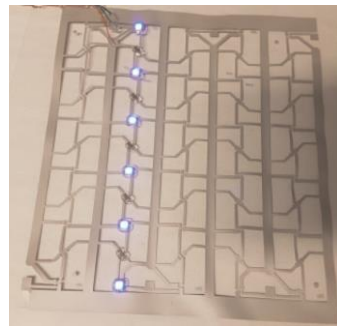
Amorphous PLA (Luminy LX175, Total Corbion, film thickness: 60-70 μm)

Screen printing and injection molding
No thermoforming

- Luoma et al. (2021)



(a)



(b)

Sheet-to-sheet screen printed silver on (a) unannealed stereocomplex PLA film dried at 100°C (left) and 80°C (right) and (b) annealed stereocomplex PLA film dried at 80°C.

PLLA Luminy LX175 and PDLA Luminy D120, Total Corbion (Film thickness: 80 μm to 125 μm)

Huge work on the preparation of the PLA films

Only screen printing

No thermoforming or injection molding

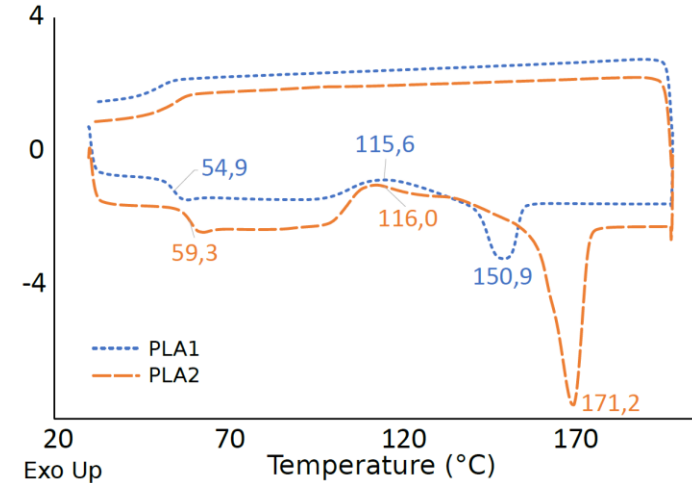
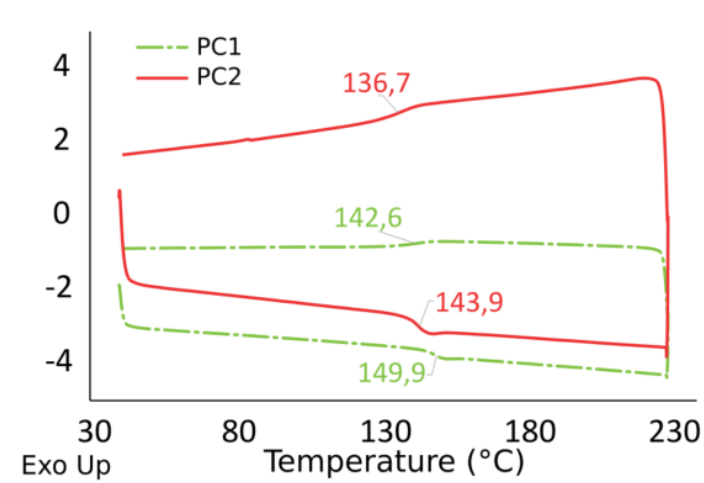
- Prontera et al. (2022), *Fabrication and biocompatibility analysis of flexible organic LED on poly(lactic acid) substrates: toward the development of greener bio-electronic devices*

Materials initial characterization

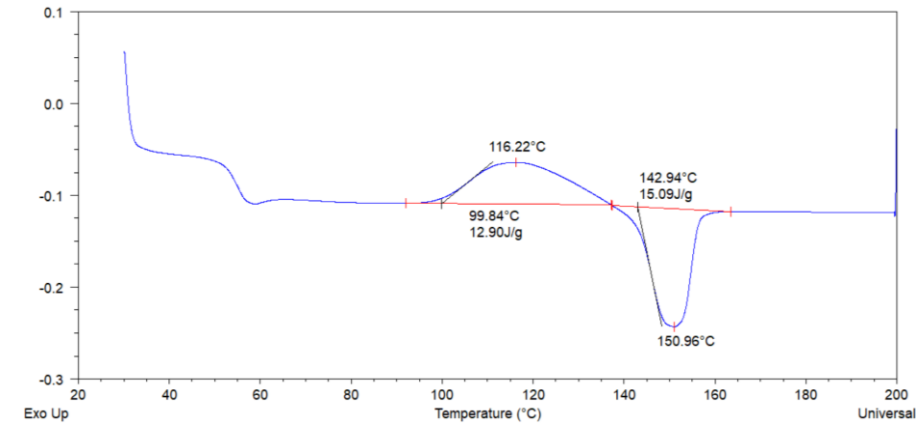
Thermal properties and thermomechanical behavior

PC1: *film DE 1-1 from Covestro (sheet)*
PC2: *grade 2405 from Covestro (injection)*

PLA1: *grade 2003D from NatureWorks (extrusion)*
PLA2: *grade 3251D from NatureWorks (injection)*



DSC curves for PC grades (left) and PLA grades (right)



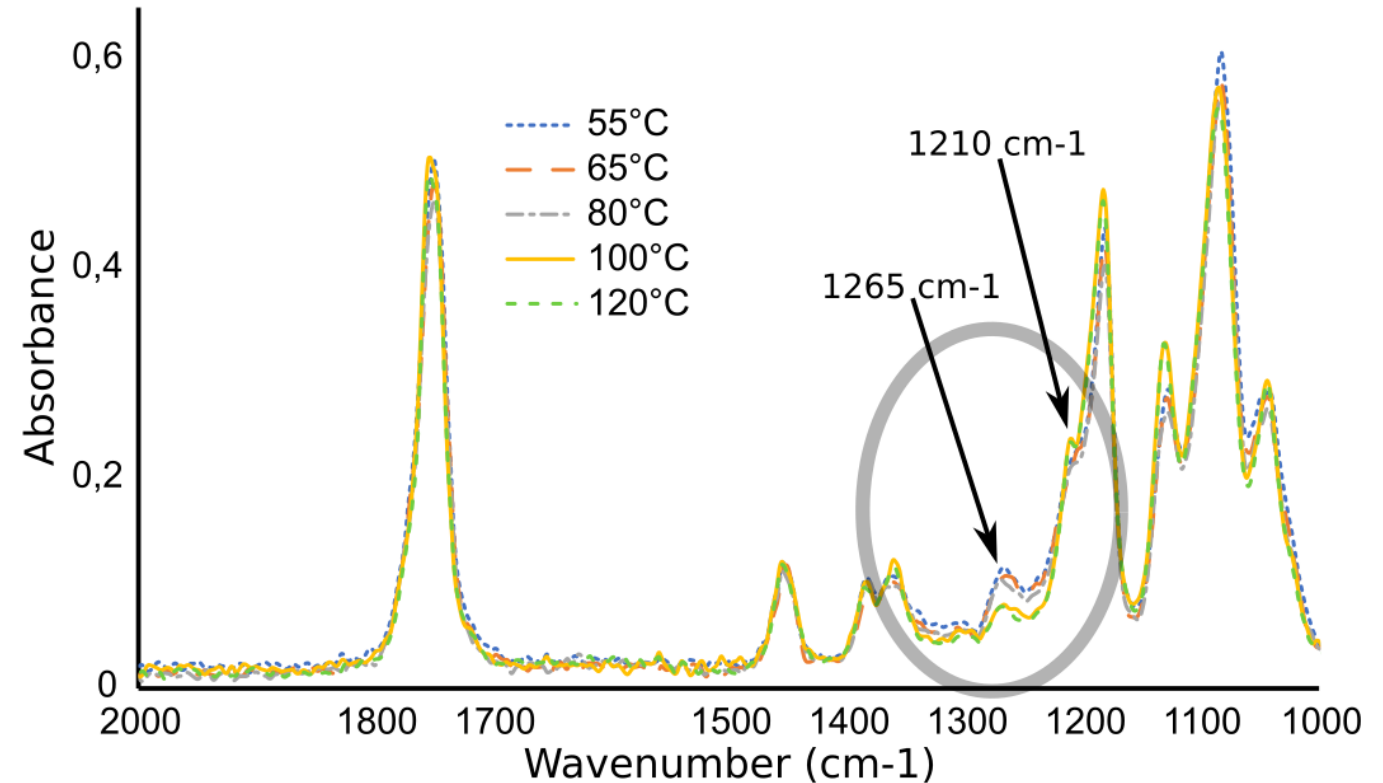
$$\chi_c = \frac{\Delta H_m - \Delta H_c}{\Delta H_m^\infty} \times 100$$

Samples	ΔH_c (J/g)	ΔH_m (J/g)	χ_c (%)
PLA1	12,90	15,09	2,35
PLA2	30,43	35,00	4,91

- PC amorphous: Tg around 145°C
- PLA semi-crystalline: low Tg (around 55-60°C) compared to PC
 - Cold crystallization (115°C)
 - Tm higher for PLA2 compared to PLA1 (170°C vs. 150°C)

- PLA1 less crystalline than PLA2
- Low enough crystallinity to consider these two PLA more amorphous than crystalline

- Can we see the variation of PLA crystallinity depending on the temperature of heat treatment?
- PLA1 film specimens at five temperatures for 20 min
- Small peak variations
- No effect of crystallinity expected during IME process

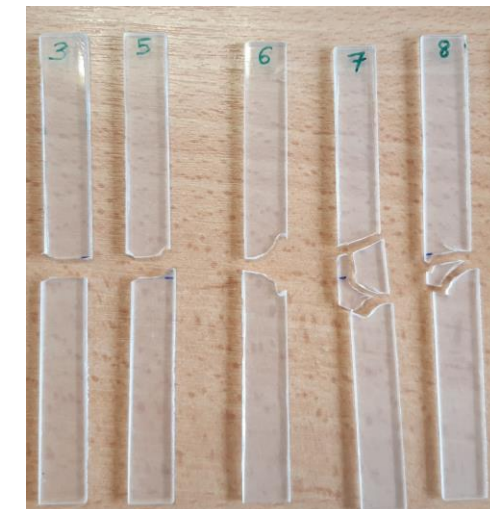
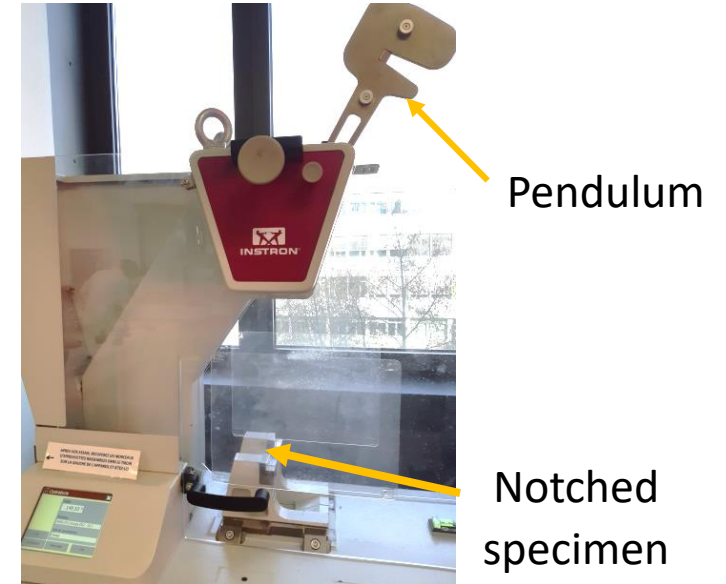


FTIR curves of PLA films heat treated at different temperatures for 20 min

- Shock resistance standard tests

Charpy tests PC2 15J pendulum	Impact resistance (kJ/m ²)
2 mm notch	8,9 ± 0,2
2,5 mm notch	8,4 ± 0,2

Charpy tests 1J pendulum		Impact resistance (kJ/m ²)
PLA1	Unnotched	15,6 ± 0,5
	Notched (2mm)	1,7 ± 0,1
PLA2	Unnotched	13,7 ± 0,6
	Notched (2mm)	1,7 ± 0,3



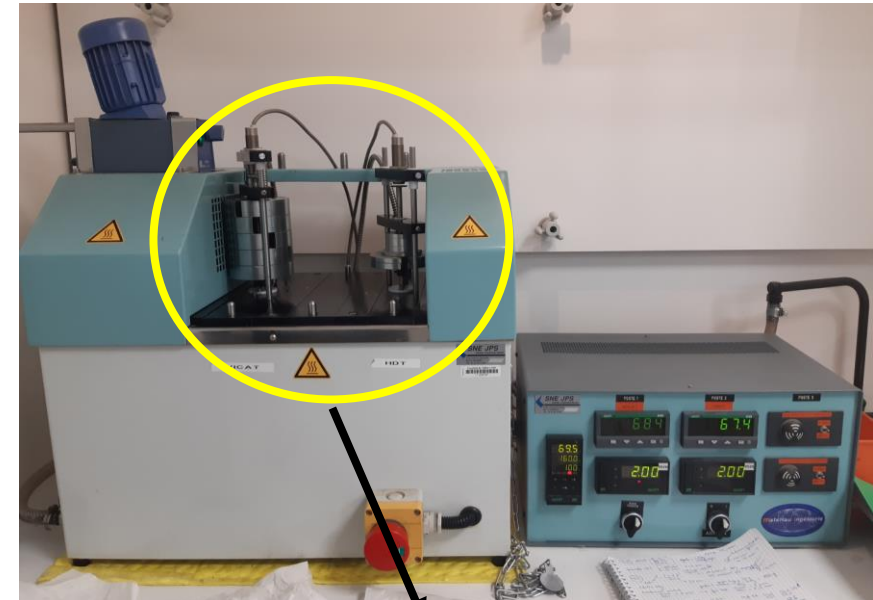
PLA specimen after test

- Confirmation PLA brittleness and PC great ductility
- But PLA objects in hands are still great!

Vicat and HDT tests

	Vicat (°C) (B method)	HDT (°C) 1,8 MPa (A)
	PC2	147
PLA2	60,9	58,6
PLA1	59,9	57,6

- Excellent thermomechanical properties of PC2
 - No plastic deformation before 140°C
 - PLA: poor thermomechanical properties
- Is PLA good enough for thermomechanical holding of electronic IME device?

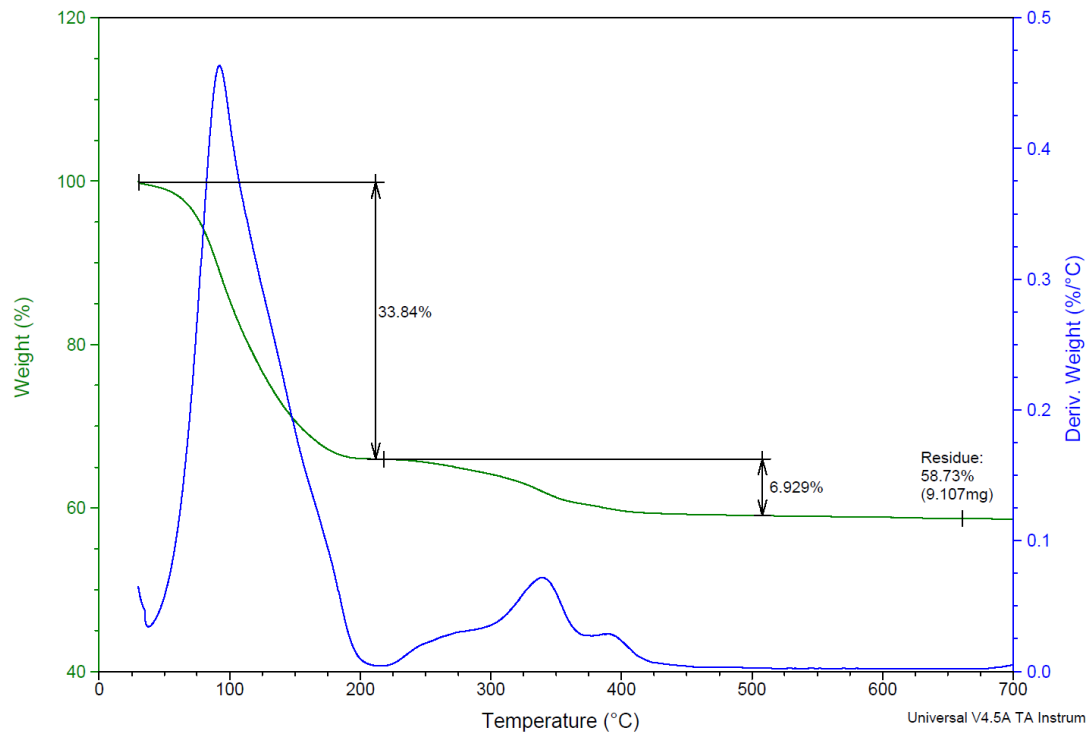


Vicat

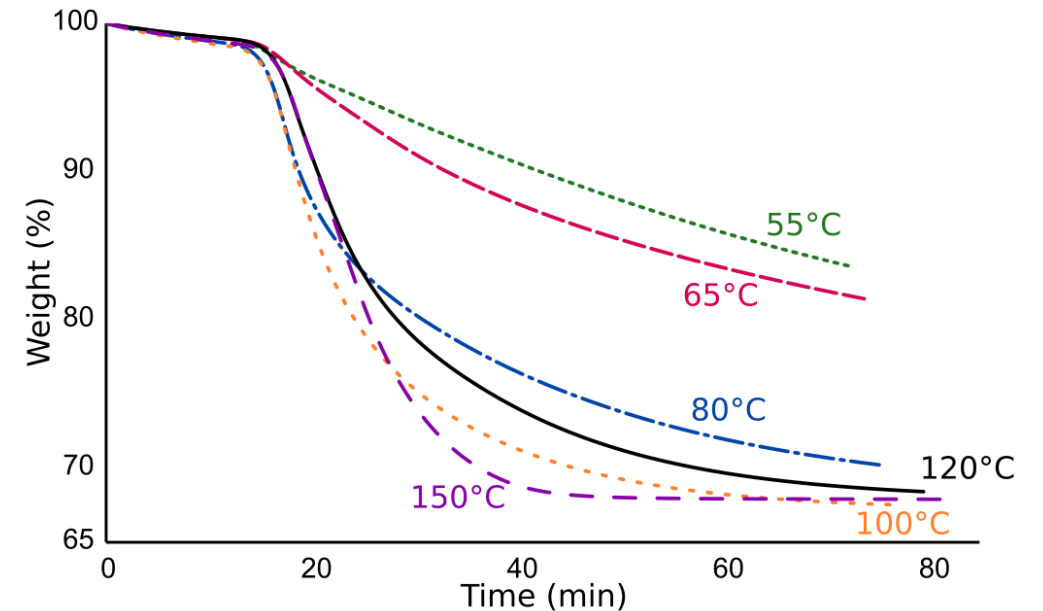


HDT

- ME603 silver conductive ink from DuPont:
 - 3 or 4 chemical compounds (blue curve)
 - 58%wt residue = silver particles (green curve)



Thermal study of ME603 conductive ink



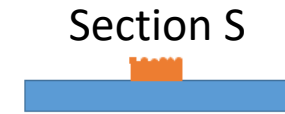
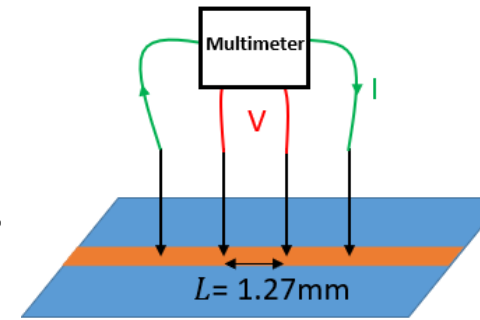
Isothermal study of ME603 drying to adapt on PLA substrate

- On PLA substrate: drying at lower temperature but for longer time
 - Choice: 55°C or 65°C during 2h30

IME process

Experimental work

- Resistance measurements
 - 4-point probe method
 - S cross-sectional area with confocal measurements
- ME603 electrical properties



$$\rho = \frac{S \cdot R}{L}$$

	Bulk silver (in tables)	ME603 supplier data on PC (120°C – 20 min)	ME603 on PLA (55°C – 20 min)
Resistivity ($\mu\Omega \cdot \text{cm}$)	1,59	89	150-200

- Resistivity of ME603 dried at 55°C on PLA much higher than when dried at 120°C on PC
 - Ink drying temperature lower on PLA
- Photonic curing to reach similar results as PC with PLA?





- PLA films: initial relaxation of internal stress necessary
 - Heat chamber between metal plates: 75°C – 15 min

- Adhesion ISO 2409:2007

➤ Good adhesion: 100% success

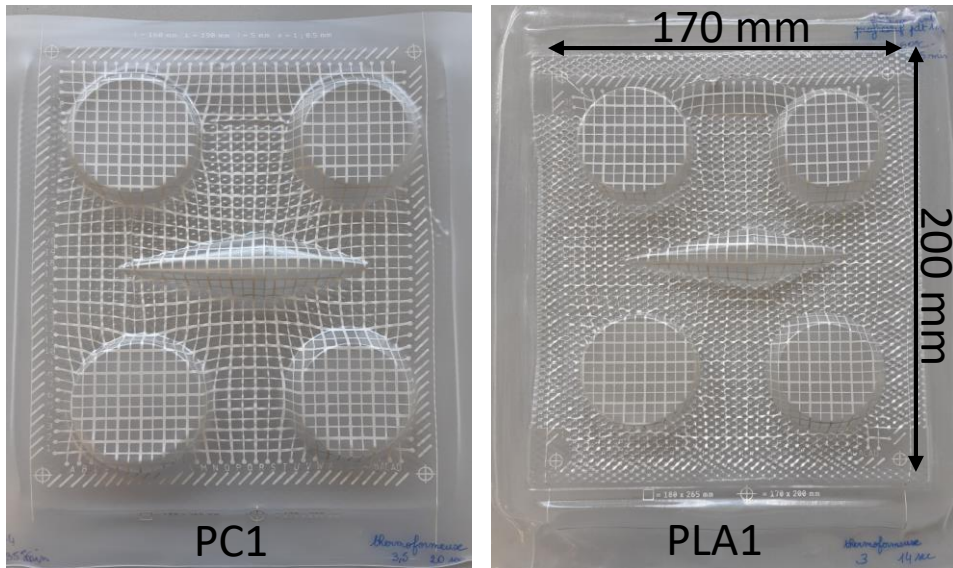
➤ Quantitative measurements

ongoing

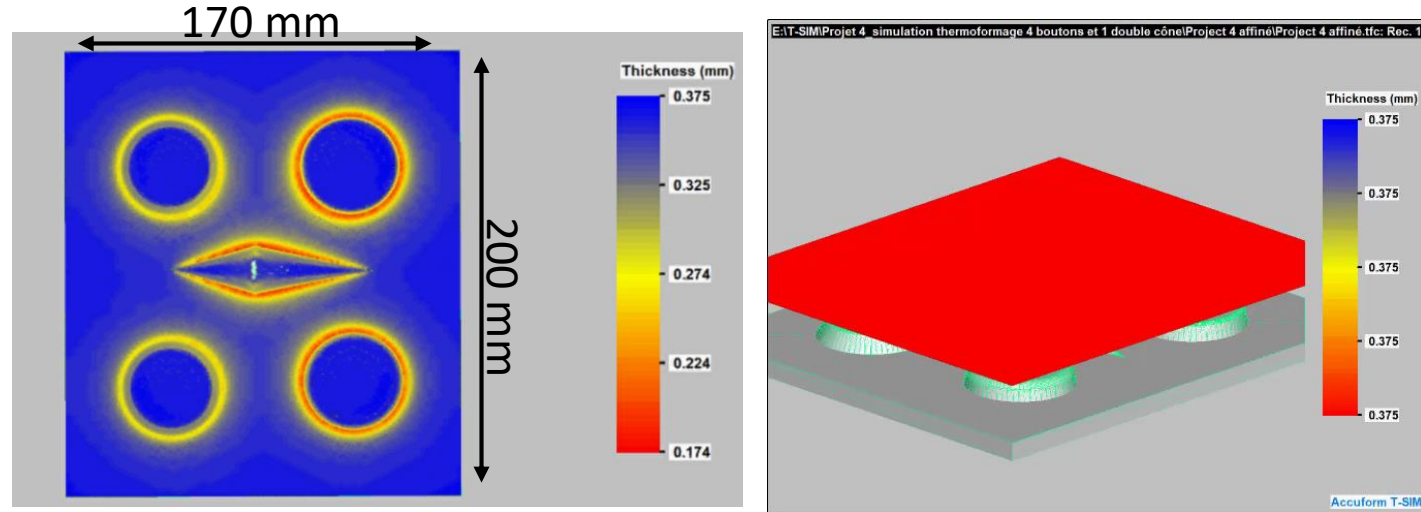
Standardized cross-cut test (Scotch test)	Before	After
PC (120°C – 20 min)		
PLA (65°C – 30 min)		

Thermoforming of PLA

- 10 mm-high molds:
 - Cylinder-shaped
 - Fillet radius: 0,5 mm or 1 mm
 - Draft angle: 10° or 30°
- 15 mm-high double cone-shaped mold



- Simulation:



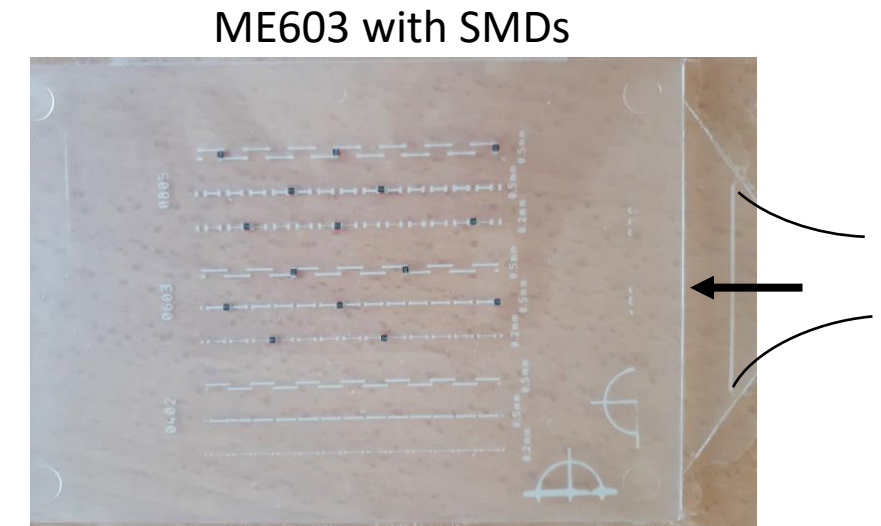
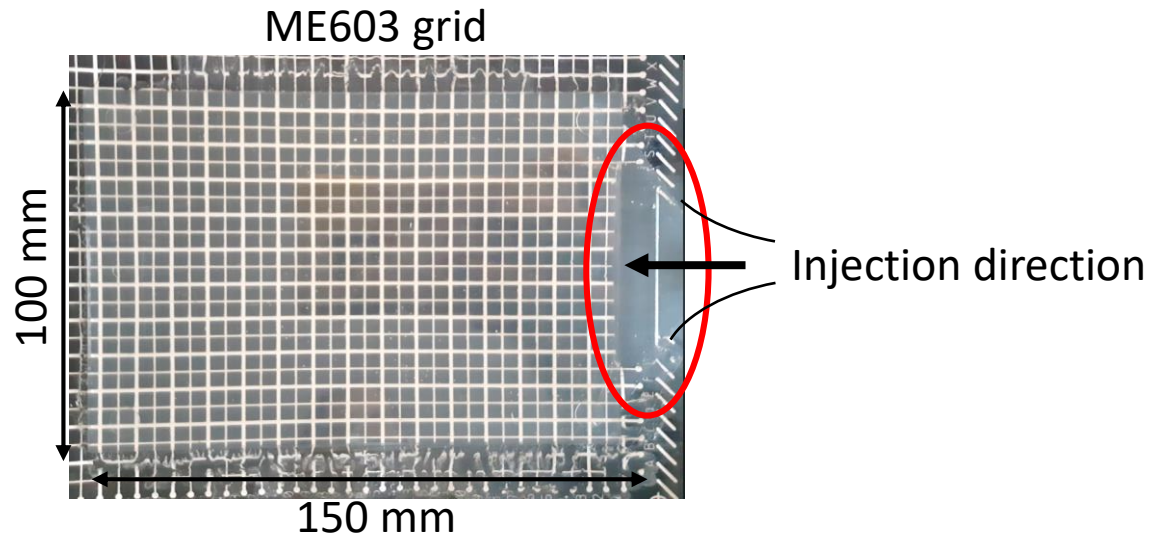
Thermoforming simulation with 375 μm thick PC film (T-SIM software)

Effect of geometrical parameters on film stretching

- Reduction in thickness to 174 μm
- Experimental:
 - PLA film well thermoformed: no cracks or holes
 - Ink slightly cracked in areas of greater deformations (\rightarrow smaller draft angle)

Injection molding of PLA on PLA

- Laboratory-scale injection molding of PLA
 - Injection mold: 2 mm-thick plate, side injection point
 - Polymer melt temperature: 210°C
- PLA1 screen printed films overmolded with PLA2
 - Injection mold at room temperature
 - Screw speed: 60 mm/s



➤ Conductive ink only washed out close to nozzle where melt is the hottest

➤ No wash-out, SMDs perfectly in place in plate mold area

Comparison of PLA with PC

PLA		PC	
Advantages	Drawbacks	Advantages	Drawbacks
Biosourced and biodegradable	Low impact resistance	Great impact resistance	During injection molding, possible wash-out of conductive paths and SMDs on substrate
Very high rigidity	Brittle	Excellent thermal stability up to 140°C	
Great chemical reactivity (-OH end groups)	Poor temperature performances of sheets (quick falling down)		
Good enough thermomechanically	Wash-out being investigated*	Screen printing inks specifically designed for IME on PC	

*Possibly less problematic than PC because of lower injection temperature for PLA...

- SMDs glued on circuit
 - Conductive paste
 - Choice of dielectric structural paste
 - Quantitative adhesion strength tests
- 3D shaping: ongoing development of an injection mold
- Integration of the electronic functions
 - Improving the ink performances with photonic curing?
 - Replacing the conductive paste with solder paste by photonic soldering?

- Improving mechanical/thermal properties of PLA
 - D. Notta-Cuvier *et al.*, “Tailoring polylactide (PLA) properties for automotive applications: Effect of addition of designed additives on main mechanical properties,” *Polymer Testing*, Jun. 2014
 - V. Nagarajan *et al.*, “Perspective on Polylactic Acid (PLA) based Sustainable Materials for Durable Applications: Focus on Toughness and Heat Resistance,” *ACS Sustainable Chem. Eng.*, Jun. 2016
 - PLA recycling or degradation
 - Industrial compost: norm ISO 14855 (60°C with 60% humidity)
 - Chemical recycling: chemical or enzymatic depolymerization
 - Tsuji *et al.*, *A New Strategy for Recycling and Preparation of Poly(L-lactic acid): Hydrolysis in the Melt*, 2003
 - Patent WO2010118954A1 : *Chemical recycling of PLA by hydrolysis* (2010), Galactic SA Belgium
 - Grewell *et al.*, *Depolymerization of Post-Consumer Polylactic Acid Products*, 2014
 - Hajighasemi *et al.*, *Biochemical and Structural Insights into Enzymatic Depolymerization of Polylactic Acid and Other Polyesters by Microbial Carboxylesterases*, 2016
 - Domestic compost:
 - Evanesto® from CARBIOLICE
 - Chemical degradation:
 - Rheinberger *et al.*, *RNA-Inspired and accelerated degradation of polylactide in seawater*, 2021
- Towards dismantling of IME object

Thank you for your attention!

Do you have questions?