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D6.3: Report on in-mold/structural electronics prototyping

17/04/2024 (M35)

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EXECUTIVE SUMMARY

The pre-pilot line for printed electronics at Holst Centre has been tested and optimized to provide functional foils for electronic devices based on printed electronics. The combination of a rotary screen printer for conductive inks and graphical inks with an assembly line for SMT components enables the development of functional foils for electronic parts. Various conductive inks were analysed during the Treasure project to provide high quality printed circuitries from start to finish during a roll-to-roll printing run by employing screens with optimized emulsions and pressures.

Injection molding was applied to flexible printed foils to obtain so-called in-mold (structural) electronic devices (IME). In these IME devices, the electronics are typically protected by the injection molded polymer resin that is applied onto the functional (flexible) substrate, making the device rigid, strong and durable. A significant challenge, however, results from this type of encapsulation for end-of-life treatment of such electronic devices. To remedy this, a design-for-recycling was tested in the Treasure project that incorporated a disassembly layer into the devices that is compatible with roll-to-roll high volume production, without significantly increasing the bill of material and costs of production.

This deliverable introduces in short the Treasure pre-pilot line for flexible / IME electronics and describes the use of the dismantling layer in IME devices. The processing is applied to a mutual demonstrator developed together with WalterPack and Seat. Data for a life cycle intake is provided which as well serves for a comparative life cycle assessment with dedicated EOL metallurgic evaluation as part of D4.8.

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1. In-mold structural electronics

1.1. Introduction

In-Mold Electronics, or IME, is an attractive alternative for conventional electronics based on printed circuitry boards (PCBs) for aerospace, domestic appliances and automotive due its form-factor, light weight, seamless design, high level of integration and recently also its reduced environmental impact(Jääskä and Apilo, n.d.; Jääskä and Muñoz, 2022). IME is a term used for a variant of printed electronics where devices, assemblies or products are realized by combining in-mold decoration or in-mold labelling with functional elements based on printed electronics. In printed electronics, the electronic circuitry is manufactured by printing and curing of conductive inks. The circuitry may consist of conductive tracks for external contacting and internal wiring, but also to create sensors, antennas and other electrical features or functionalities. The circuitry can be combined with elements from traditional electronics, such as semiconducting components that include capacitors, resistors, light-emitting diodes, driving chips, sensors and so forth, but also other type of electronics, such as solar cells, external sensors, external PCB driving boards, and so on. In-mold electronics (IME) is currently being developed for aerospace, domestic appliances and automotive. Unique to IME is the combination of printed electronics on 2D thermoplastic substrates with a glass transition temperature (Tg) well above room temperature and shaping of the substrate a 2½ or 3D shape by a process called thermoforming. Thermoforming occurs at high pressures and a temperature at or just above Tg. Subsequent injection molding of additional plastic onto the printed circuitry provides strength, stability, rigidity and encapsulation of printed electronics from external influences.

The IME market for automotive is growing rapidly and mass manufacturing of IME parts is expected to boom following favourable assessments by OEMs. Due to the novelty of this technology in comparison to conventional electronics based on printed circuitry boards (PCBs), additional measures for disassembly and recycling may be beneficial to increase recycling yields. After a more detailed description of IME, including an overview of a typical composition, this informative section elaborates on disassembly and recycling scenarios.

Example state-of-the-art IME parts for automotive

For visual reference, exemplary state-of-the-art IME and TactoTek's IMSE® prototype parts are provided in Figure 1 and Figure 2.

a) b)

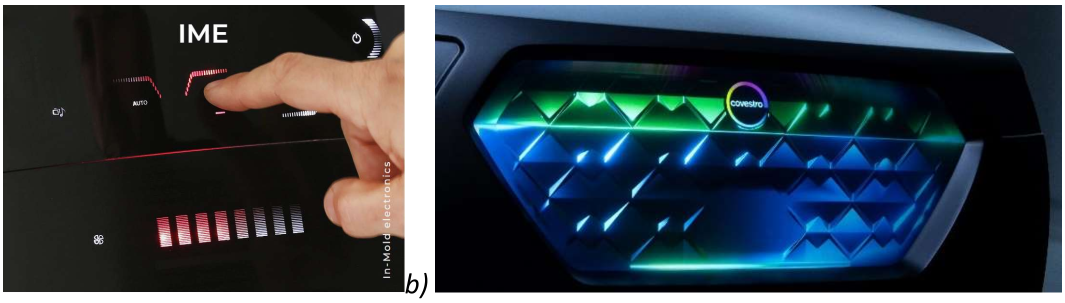

Figure 1a) climate control unit by WalterPack; b) Automotive front grill demonstrator by Covestro: 5D film insert molding (FIM) combines rear 3D structures with color and decoration due to the design freedom enabled by printed polycarbonate films. Integrated metal lines can be used for example for deicing to keep sensor areas clear or provide brand differentiation through integrated LED lighting powered by printed electronics.

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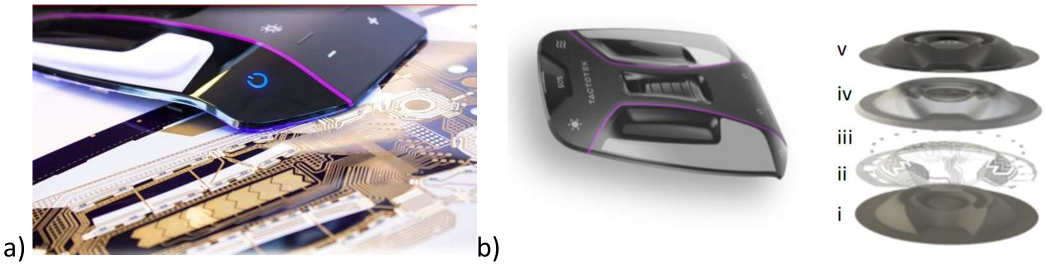

Figure 2: TactoTek's IMSE® automotive panel with example layout in b) showing buildup from bottom to top: i) functional substrate with ii) printed metal circuitry, iii) semiconductor components, iv) injected thermoplastic resin and v) front thermoplastic decorative foil.

Like other electronics and electronic devices, IME combines plastics with metals, semiconductor technology and carbon-based coatings that improve aesthetics or provide a function within the layer stacking as protective, adhesion or otherwise supporting layer. The coatings and bulk plastics are chosen and tuned to provide a highly reliable part that can last for years. Metals and components are largely fully embedded within the plastics. While this is highly favourable for protecting the electronic functionalities, this is less than favourable for recycling at end-of-life.

1.2. Treasure solution to improve recyclability of IME devices

In end-of-life vehicles (ELV), IME parts are likely to be readily accessible for removal from the car due to their usage as part of the human machine interface. If not removed, the panel may be shredded as part of the car and end up in different recycling fractions, depending on the sorting processes applied in the plant, such as density separation and eddy-current separation, and the properties (composition) of the IME, including, if not foremost, the ratio of metal and non-metal. IME may contain only a few % of printed metal and may not be recognized as metallized plastic by automated systems that are tuned for printed circuitry boards (PCBs). If removed from the ELV, the IME parts may instead be sold and re-used, or collected as a separate waste stream if these are not deemed valuable or re-usable. The latter gives the possibility to properly repair or recycle the IME part. We believe that this is accomplished by introducing design-for-recycling principles and product-centric end-of-life processing as advocated by MARAS b.v. In EU Treasure, a design-for-recycling was pursued for IME that uses a dismantling layer within the stack design.

For further information regarding why design-for-recycling (DfR) adds to the recovery rates of plastics and metals, see Harkema *et al.* (Harkema et al., 2024) and references therein. Also further described in this article is the implementation of the DfR in IME. See also public Treasure deliverable 5.7.

2. Pre-pilot line description

2.1. Introduction

A roll-to-roll line for printed electronics has been developed at Holst Centre for prototyping purposes and research on roll-to-roll printing, assembly and optimization of processing. In the EU Treasure project, we looked more into the details of the printing process, the power usage and its applicability to a specific rigid form of printed electronics: in-mold structural electronics (IME). This deliverable will describe the roll-to-roll pre-pilot line and describe our activities.

A further description of the pre-pilot line and involved experiments can be found in D5.6 and D5.7 (public).

2.2. Pre-pilot line lay-out

The pre-pilot line consists of two sections, each with winding and unwinding stations for rolls of plastic foil. These sections were not developed to provide a continuous process from printing of graphics to assembly of SMT components. This pre-pilot line was designed for research & development of printed electronics and is suitable for low volume prototyping. The power usage per station is provided in the corresponding figures for the graphics line and the SMT assembly line, being Figure 3 and Figure 4. For the development of roll-to-roll printing, we opted for a single rotary screen for printing in the first section of the line, as each layer needs to be optimized for printing and drying. Later sections of this deliverables will further elaborate. Industrial production using roll-to-roll methodologies would / could focus on multiple rotary screens and short bursts of drying, followed by a stage of more extensive drying. This line runs at a speed of at least 5 meters per minute.

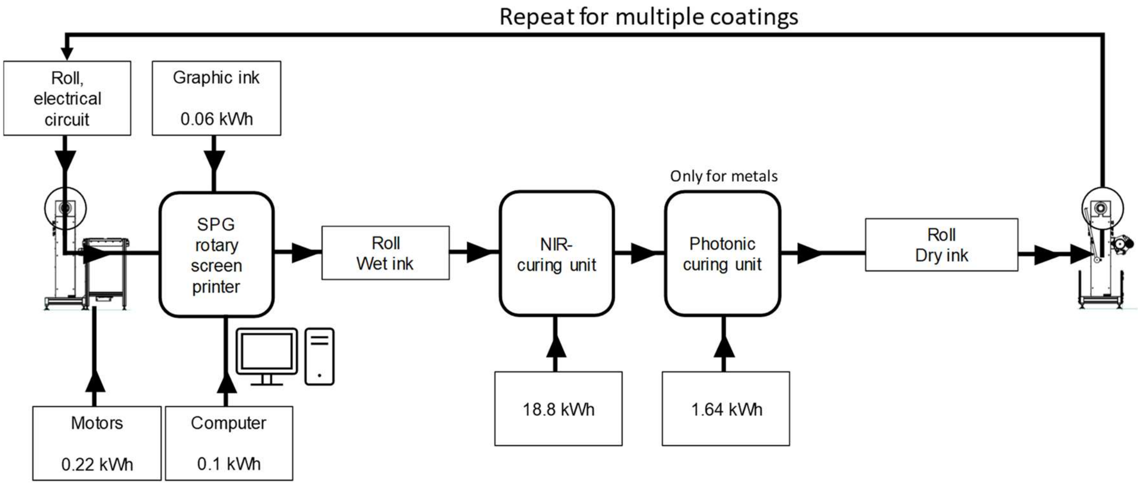

Figure 3: pre-pilot line lay-out for rotary screen printing of conductive and non-conductive inks

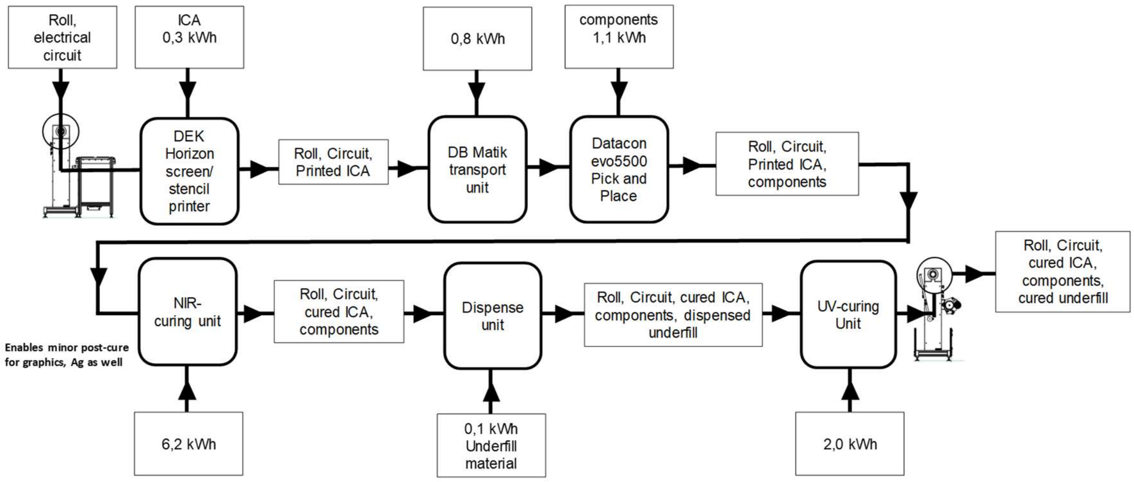

Figure 4: pre-pilot line lay-out for stencil printing of conductive adhesives, SMT component pick & place, underfilling with structural adhesive and curing

The assembly line consists of a stencil printing station for deposition of conductive adhesives, pick & place station for SMT components, NIR curing, optionally in combination with photonic sintering / curing, underfilling and finally UV curing for the structural adhesive. In our pre-pilot line, the pick & place station is time-determining step, which translates in a relatively high-power usage per device in comparison to a faster industrial process.

3. Development of Treasure demonstrator

3.1. Demonstrator design

Activities using the pre-pilot line were carried out in WP5 and 6 in the EU Treasure project. WP5 tasks concerned simulation of IME processing using the pre-pilot line and optimization of the printing. WP6 tasks concerned prototyping automotive parts in collaboration with WalterPack. These developments are close to concluded and will be completed before the end of the project.

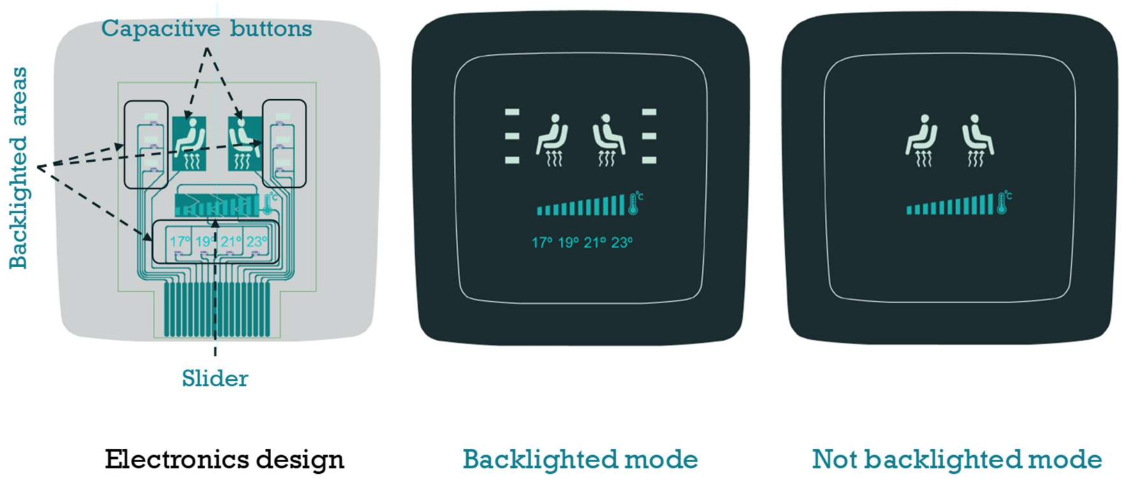

Figure 5: design of decorative foil A

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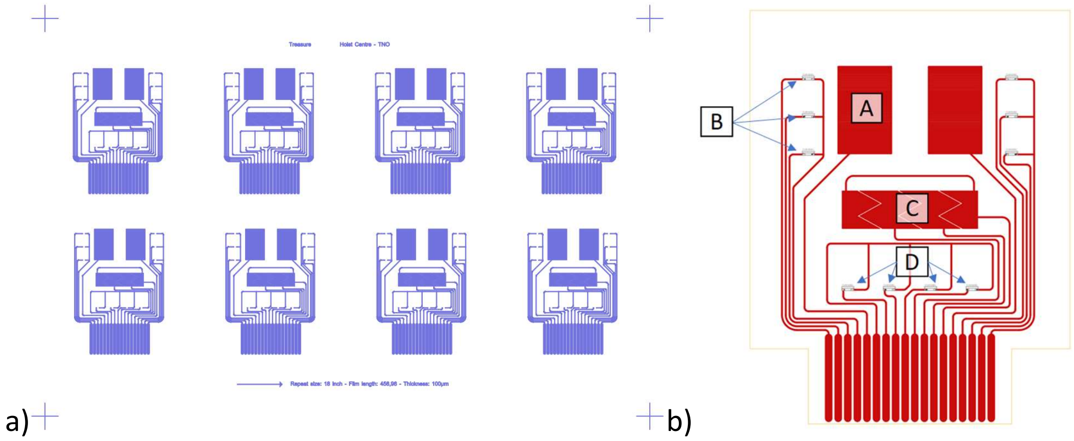

Figure 6: a) design for rotary screen of Ag circuitry on functional foil B; b) design of a single device with labels A-D for functionalities: A: capacitive touch area, icon for seat heating B: 3 LEDs indicating seat heating level C: capacitive touch slider for aircon temp. D: 4 LEDs for indicating aircon temp. LEDs are Side firing; type OSRAM CUW Y3SH.B2.

The designs of the decorative foil A and functional foil B are provided in Figure 5 and Figure 6. Figure 7**Error! Reference source not found.** shows the process flow for foils A and B with a multitude of printing and curing steps to deposit the functional and decorative coatings onto each foil. These two foils, of which foil A is thermoformed to provide a curve shape, are then combined into a single IME device using injection molding.

3.2. Demonstrator process flow

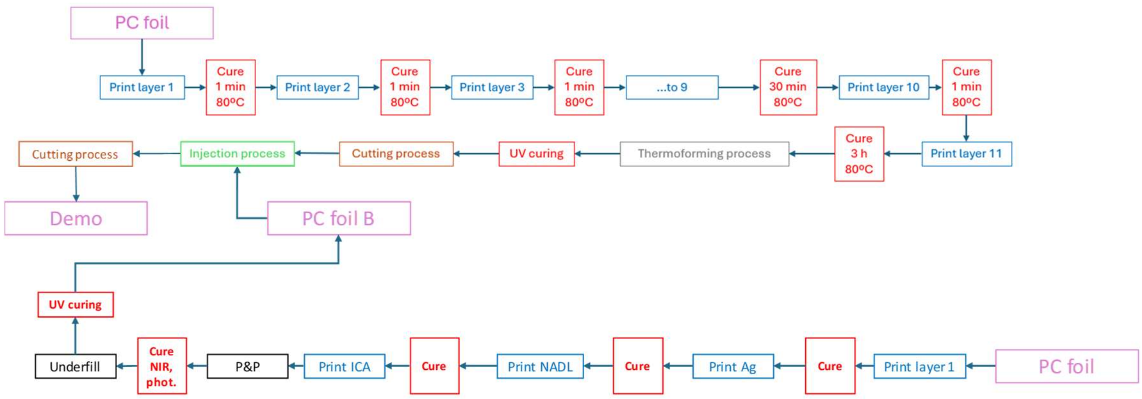

Figure 7: process flow for the Treasure IME demonstrator

Process flow in Figure 7**Error! Reference source not found.** shows the process flow for the decorative foil, starting in the top left, including the merging of foils A and B by injection molding, and in the bottom, the flow for the functional foil. The process for foil A consists of the application of 9 coatings, but is more complicated than shown, as the final part contains haptics by surface textures, a unique feature added to the part by WalterPack. This allows the user to feel where the slider is located and provides an intuitive interaction to adapt the air temperature using the slider. The materials used for the decorative foil are also not as easily summarized as for Foil B, as some layers may be achieved by a combination of materials. A legend is therefore omitted. Foil B includes the application of semiconductor components (LEDs) by pick & place technology. LED bonding by pick & place is followed by application of structural adhesive and

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curing thereof. In the final stage of assembly, foils A and B are brought together in an injection molding tool and bonded together by the solidification of the injected hot polycarbonate.

3.3. Demo processing

Processing of the foils proceeded as indicated in the process flow. Figure 8a) below show the rotary screen by which the printing of the conductive tracks with silver ink proceeds (shown in b). Figure 8c) shows drying by NIR, followed by rolling up the web in d). After bonding of the LEDs, and cutting of the devices to size, samples as shown in Figure 10**Error! Reference source not found.** were obtained and provided to WalterPack for further processing (combining with foil A).

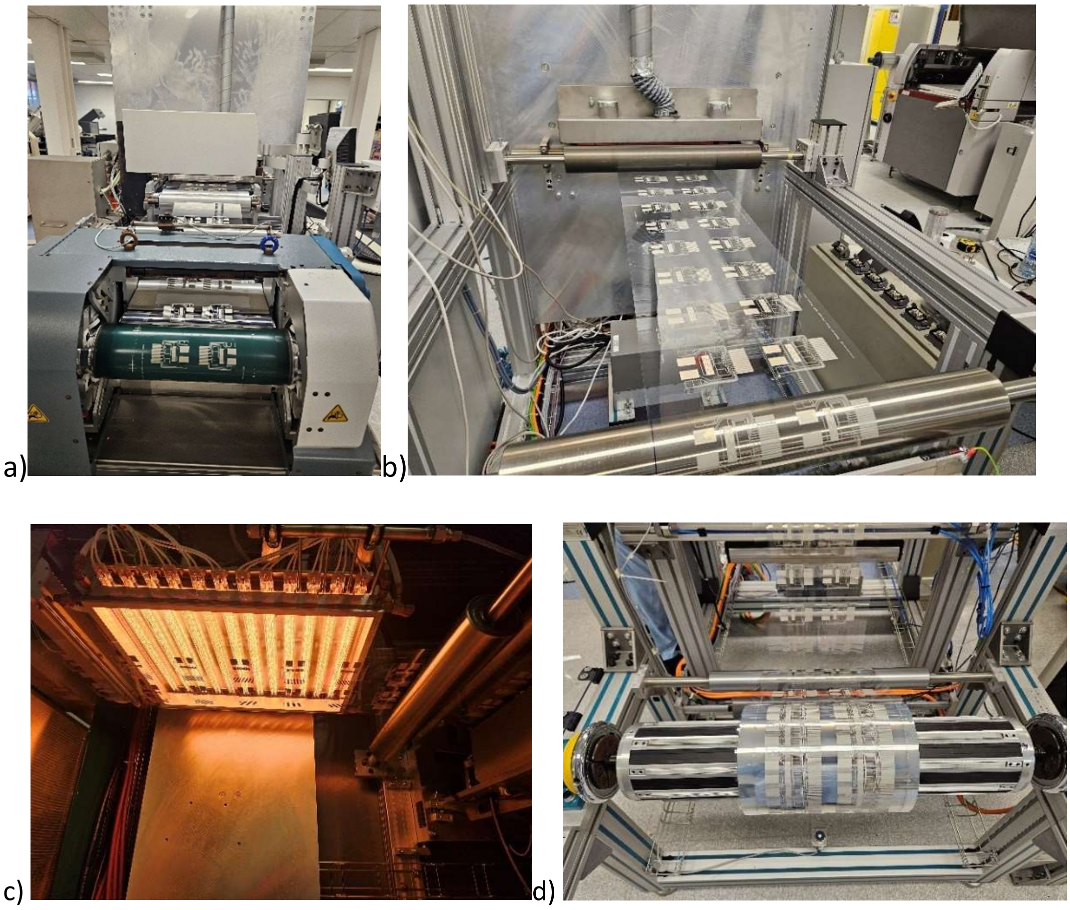

Figure 8: roll-to-roll deposition of the conductive silver tracks for the demonstrator in a) and curing underneath the NIR heater in b).

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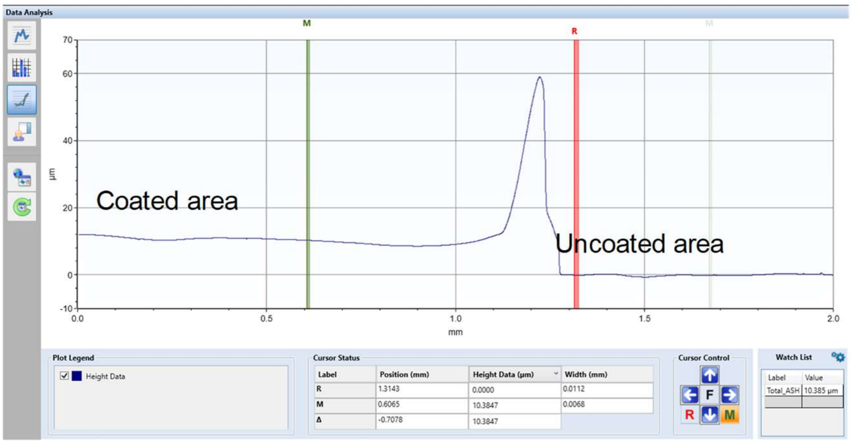

Figure 9: thickness measurement of the dismantling layer

To include the dismantling layer in the device, the coating material was applied to the functional substrates. A thickness measurement of the transparent dismantling layer indicated a coating thickness of 8 to 12 µm. The artifact at the edge of 60 microns high is caused by a piece of tape that was coated along with the dismantling layer for the purpose of having a clean surface for the thickness measurement after removal of the tape.

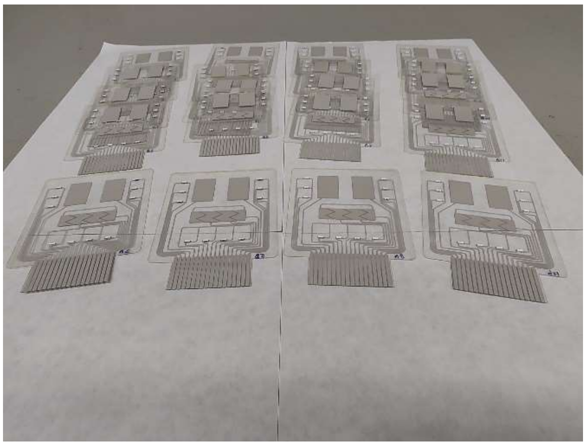

Figure 10: cut and assembled functional foils before combining these with the decorative foils at WalterPack facilities by injection molding.

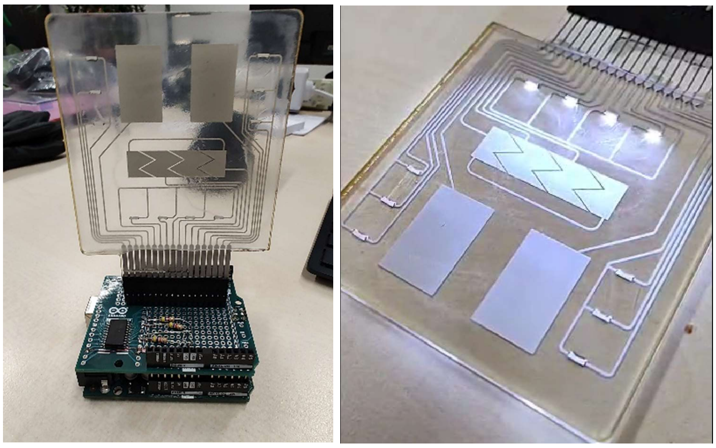

Figure 11: a,b) functional foil hooked up with an Arduino Uno PCB driver.

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The foils were tested with a simple Python code using an Arduino Uno. Figure 11a) shows the connection for testing and in b) with all LEDs of the capacitive touch slider on. Including PCB driver, the functional foil consumes 2 Wh with all LEDs on. A more typical value is 1.5 Wh: not all LEDs are continuously on. The PCB driver is responsible for a significant part of the power consumption: 1 Wh.

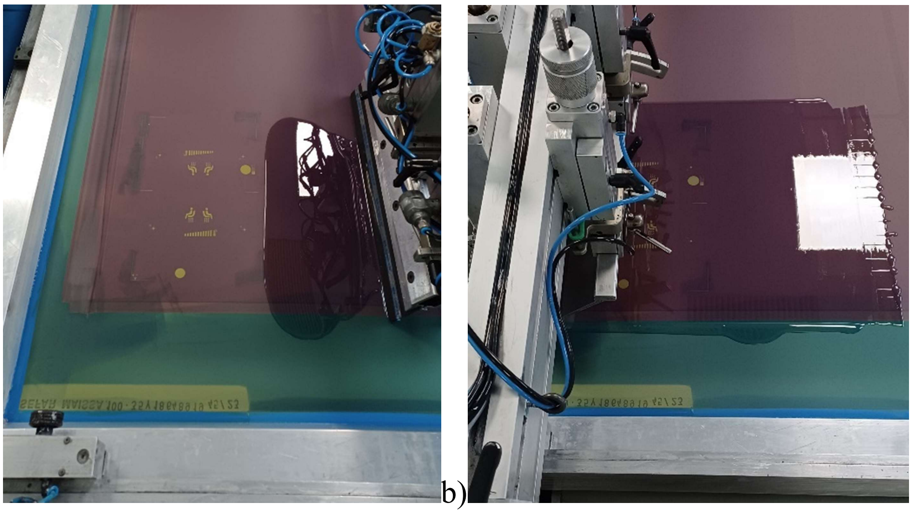
a)  b)

Figure 12: a) ink deposition for screen printing the decorative foil at WalterPack; b) wet coating formed by screen printing

The decorative foil was separately processed and thermoformed by WalterPack, as shown in Figure 12 and further described in the process flow in Figure 7.

Finally, the separate foils were placed in the injection molding tool at the WalterPack facilities and were combined to create the part we refer to as the Treasure IME demonstrator (Figure 14). At the time of writing this report, injection molding had been completed to an extent, but testing and dismantling could not yet be performed.

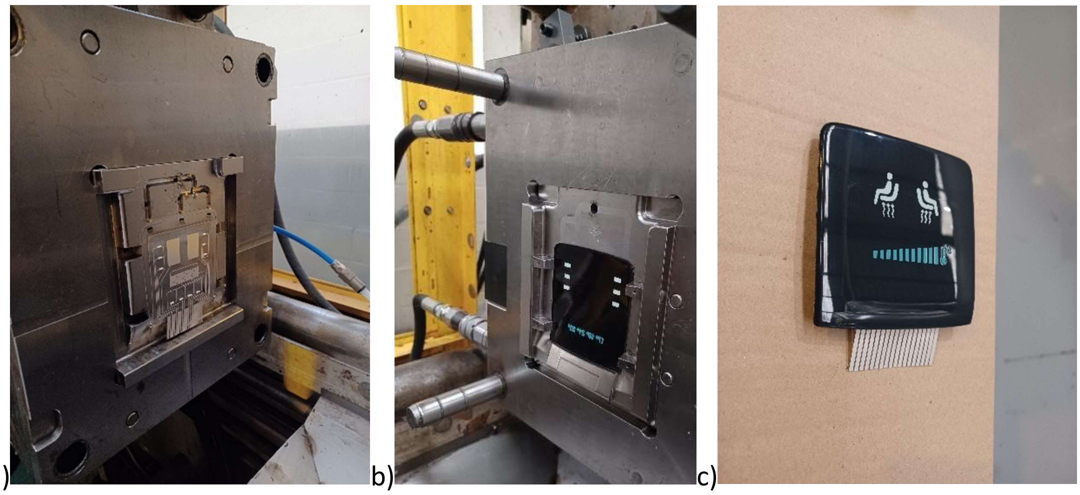
a)  b) c)

Figure 13: injection molding at WalterPack: a) functional foil inserted into the injection mold, b) decorative foil inserted into the injection mold, c) assembled part

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4. Life cycle intake of Treasure demonstrator

4.1. Composition

The Treasure IME part, as shown in Figure 13b), is generally comprised of:

- Thermoplastic bulk material as substrates and resins: 96.8% by weight, depending on the design of the part, and concerns pure polycarbonate purchased from Covestro.
- Thermoplastic coatings: 2.9% by weight and concern graphic coatings on foils A and B and an anti-scratch coating on foil A. Inks for graphical coatings were purchased from Proell. MSDS data is not clear on what polymer system the ink is based on. We assume that these are based on polyurethane.
- Thermoset adhesives: << 1% by weight and concern e.g. structural adhesives e.g. based on epoxy-resins. Commercial adhesives from Henkel were used.
- Printed conductive tracks, 0.2% by weight as pastes, containing 20-40% solvent. The solid weight of printed conductive material is predominantly metallic, but may be contained in a polymer matrix in the concentration range of 50-80%. Celanese's MicroMax™ ME604 was used.
- Semiconductor components may be present in IME automotive electronics in 0-1 weight-% as heterogenous elements that have been separately encapsulated in black epoxy molding compound before being embedded in IME parts. The composition of these components varies heavily on the provided functionality and generally may consist of various elements, including silicon, copper, gold, silver, germanium, phosphorus, boron, indium and gallium, to name a few. Strategic/critical metals are present in semiconducting components in low to very low amounts (in the order of 10^1-10^3 g/ton for e.g. Ga, In, Ag, Pd, Au). Side-firing LEDs (OSRAM) CUW Y3SH.B2 were used. LED composition is now known. No further data could be found.

Figure 14 provides a pie chart diagram for the IME demonstrator (see Figure 13c)). Close to 97 weight-% of the part consists of polycarbonate (substrate and resin) with nearly 3 weight-% of graphical inks. Silver, here calculated as the metallic part of the ink, is present as a minor 0.2 weight-% fraction. This is an exceptionally low amount which may not be detectable in industrial recycling machines that typically sort printed circuitry boards (PCBs). These have 20-30 weight-% Cu. Therefore, relevant is also the distribution of weight after dismantling. Figure 15 provides a pie chart of the functional substrate. The weight-% of Ag is nearly a factor 10 higher with 90% of the bulk plastics removed from the composition. Univaq has provided a recycling strategy and economic viability analysis of IME recycling. For a lower Ag concentration of 0.93 weight-%, a yield of 7.7 kg of Ag from a ton of (dismantled) IME could be recuperated. Revenues for a hydro-metallurgic process using the Univaq pre-pilot line was estimated between 4,000 and 5100 Euro, depending on the depreciation of Ag from an average price of 740 Euro per kg of Ag. A higher concentration of 2.1 w-% would result in the recovery of 17.7 kg of Ag and lead to revenues of that process between 9 and 11.5 kEur.

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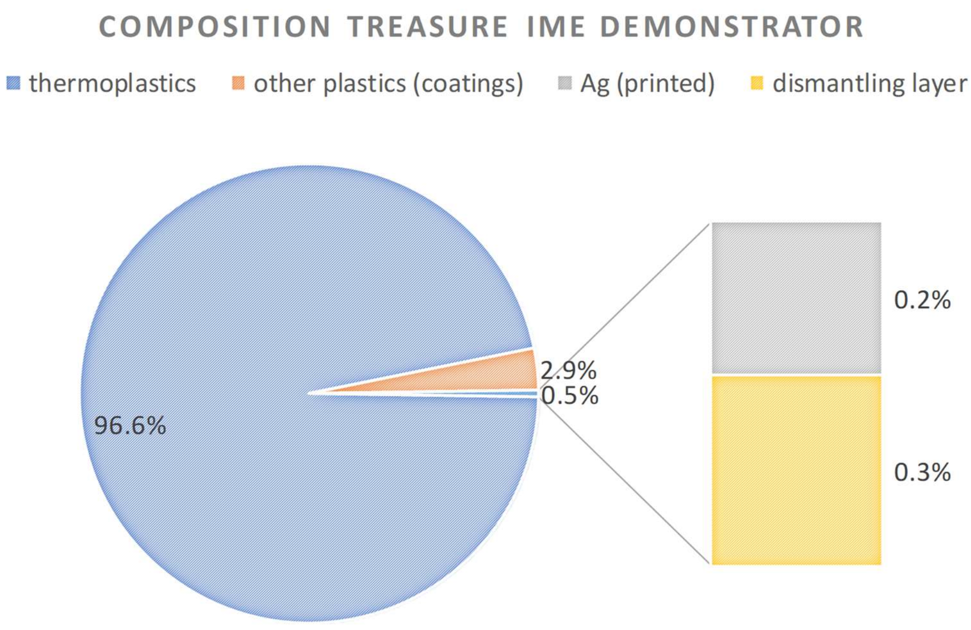

Figure 14: example composition for the polycarbonate-based IME demonstrator, as developed in the EU Treasure project. Semiconductor components and external PCB driver are excluded.

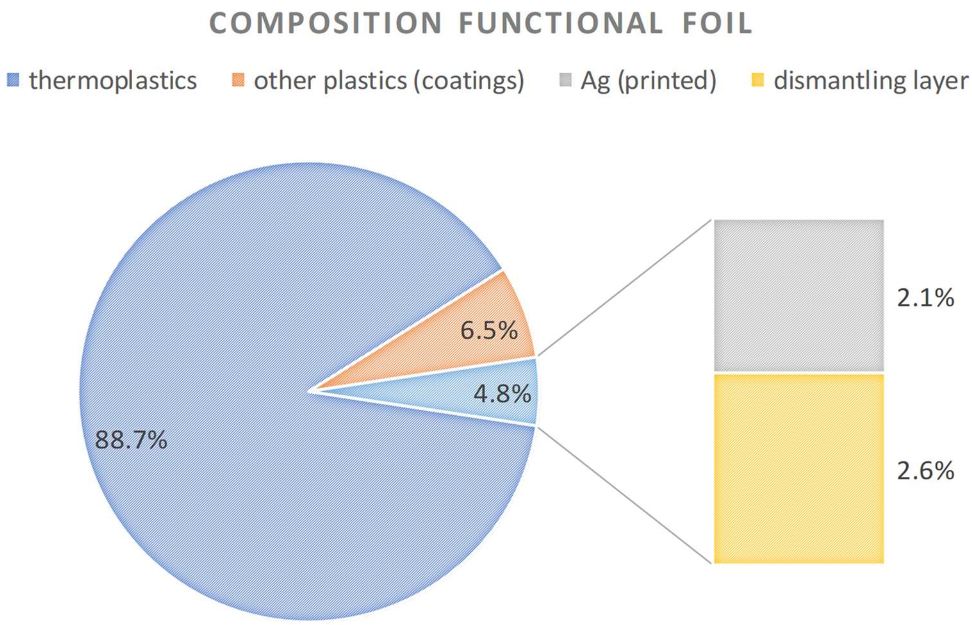

Figure 15: composition of the functional substrate. Semiconductor components are excluded.

A more detailed overview of materials used in each IME device and during production is shown in Figure 16. It should be noted that it was not possible to get exact names for materials from available MSDS files from suppliers. Concrete materials appear to be exchanged with more general descriptions that are useful from LCA point of view, but are too general for anything else. Another note concerns the polymer matrix of coatings and silver ink: this polymer is assumed to be thermoplastic polyurethane, while in practice this cannot be confirmed without breaking purchasing conditions. The dismantling layer is assumed to be polyvinyl alcohol based, based on its behaviour, as further elaborated in D5.7.

Row Labels	Sum of amounts
(2-methoxymethylethoxy)propanol	0.046
1-methoxypropan-2-ol	0.091
2-butoxyethyl acetate	0.055
2-methoxy-1-methylethyl acetate	0.064
2-Propenoic acid, 2-methyl-, 2-(dimethylamino)ethyl ester, polymer with butyl 2-propenoate, comps. with polyethylene glycol hydrogen maleate C9-11-alkyl ethers	0.003
4,4'-Isopropylidenediphenol, oligomeric reaction products with 1-chloro-2,3-epoxypropane	0.000
4,4'-Isopropylidenediphenol, oligomeric reaction products with 1-chloro-2,3-epoxypropane, reaction products with 2-methylimidazole	0.000
Aromatic hydrocarbons, C9	0.540
Calcium bis(2-ethylhexanoate)	0.001
Cyclohexanol, 4,4'-(1-methylethylidene)bis-, polymer with 1,6-diisocyanatohexane, 5-isocyanato-1-(isocyanatomethyl)-1,3,3-trimethylcyclohexane and 2-Propenoic acid, reaction products with pentaerythritol, 2-hydroxyethyl acrylate-blocked	0.047
EPOXY / PHENOLIC RESIN	0.000
ethyl 3-ethoxyproprionate	0.027
Formaldehyde, oligomeric reaction products with 1-Chloro-2,3-epoxypropane and Phenol	0.000
Hexanoic acid, 2-ethyl, zinc salt, basic	0.002
Hydrocarbons, C10, aromates, < 1% naphta	0.353
Hydrocarbons, C10-C13, aromates, < 1% naphta	0.118
Maleic anhydride	0.001
Methanol	0.007
Naphtalene	0.005
Polyurethane (assumed)	0.884
Polyvinyl Alcohol (Ethenol, homopolymer)	0.083
Propylidynetrimethanol	0.000
p-tert-butylphenyl 1-(2,3-epoxy)propyl ether	0.000
Silver powder	0.040
Solvent naphtha (petroleum), light arom.	0.091
Tris(nonylphenyl) phosphite	0.000
Water	0.158
Grand Total	**2.617**
Row Labels	**Sum of amounts**
polycarbonate	36.437
Grand Total	**36.437**

Figure 16: Material balance of the Treasure demonstrator. Includes production waste, ink solvents and cleaning solvents for the screens. Material names do not correspond to structural formulae (and are not exact), but correspond to data reported by material suppliers in the Material Safety Data Sheets.

4.2. Power consumption

EU TREASURE IME POWER CONSUMPTION / TREASURE IME PRE-PILOT LINE								
PER ROLL	PRINTING		ASSEMBLY		THERMOFORM		INJECTION MOLD	TOTAL PROCESS
200 m	FOIL A	FOIL B	FOIL A	FOIL B	FOIL A	FOIL B	FOIL A+B+RESIN	
	total energy	total energy	total energy	total energy	total energy	total energy	total energy	total energy
	kW	*kW*	*kW*	*kW*	*kW*	*kW*	*kW*	*kW*
	163.6	98.8	0	616.9	409	0	22.8	**1311 kW per roll**
devices	average power per device							
3492	PRINTING		ASSEMBLY		THERMOFORM		Injection mold	TOTAL PROCESS
	FOIL A	FOIL B	FOIL A	FOIL B	FOIL A	FOIL B	FOIL A+B+RESIN	
	total energy	total energy	total energy	total energy	total energy	total energy	total energy	total energy
	kW	*kW*	*kW*	*kW*	*kW*	*kW*	*kW*	*kW*
	4.7E-02	2.8E-02	0	1.8E-01	1.2E-01	0	6.5E-03	**0.38 kW per device**

Figure 17: power consumption per roll and per device

A roll of 200 m could fit a total of 3492 devices in its current design. Processing is limited by especially by the assembly of components onto the printed circuitry. In the roll-to-roll line used in EU Treasure, pick & place is relatively slow. The design (Figure 6a) contains 2 devices that are

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simultaneously printed and bonded, meaning 20 LEDs. With a time per component of 3 sec, the set of two devices require 60 s for bonding. As a result, bonding LEDs on a full roll of devices would require 58 hours. During this time, heaters and UV curing are on, resulting in a relatively high-power contribution of the assembly process. Industrial P&P machines are considerably faster and as a result, the power contribution in such a process will be lower. In rotary printing, the curing time is limited by the ink composition. The slowest possible setting is taken for the rotary printing process, which is 5 m/min. The graphic inks of the decorative foil were assumed to have been processed with the roll-to-roll tool to provide a LCA for R2R prototyping and manufacturing of an IME part. Post-curing of the graphic inks was included. An industrial system would have to be tuned more dedicatedly for the materials. Finally, thermoforming requires a considerable amount of energy, as the infrared heaters are continuously on during transport of the sample and the actual forming process. The thermoforming step is considered to take 31 s per sample. Due to the design, we can assume that in a further development, a set of 2 devices may be simultaneously thermoformed, leading to a reduction of the power contribution by a factor 2 (0.12 kW per device to 0.06 kW per device). This was not yet included here. Injection molding is a relatively power efficient step in comparison. TNO’s injection molder was taken as example and consumes 0.47 kW/kg PC. With the injected weight being so low, only 0.01 kW can be allocated for this process. Devices processed efficiently in an industrial setting will require less energy to produce. Especially the power used for assembly can be reduced considerably. P&P in 0.5 sec would cut the power usage by a 83% and the overall power used to process 1 device by 0.15 kW (-39%). Reduction of tact time of the other steps, e.g. thermoforming, will also contribute drastically.

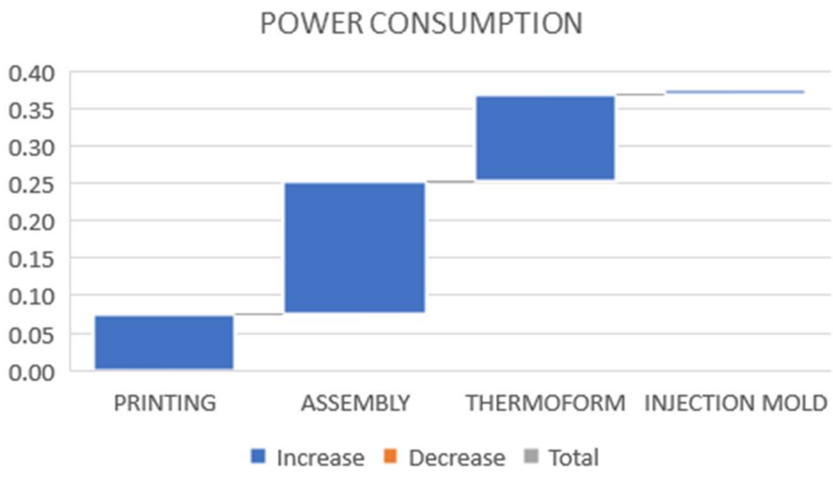

Figure 18: power consumption per processing step

4.3. Power consumption use phase

Including PCB driver, the functional foil consumes 2 Wh with all LEDs on. A more typical value is 1.5 Wh: not all LEDs are continuously on. The PCB driver is responsible for a significant part of the power consumption: 1 Wh.

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5. Conclusions

IME prototyping was accomplished in collaboration with WalterPack and Seat using the Flexible Electronics pre-pilot line at TNO. An IME device with similar functionalities as a climate module intended for use in the rear section of the car was created by a combination of printing of graphic and conductive inks, assembly of semiconductor components, thermoforming and injection molding. A life cycle intake was prepared for evaluation by Treasure partners.

6. Abbreviations

IME	In-mold Structural Electronics
Ag	Silver
NADL	Non-adhering dismantling layer
PCB	Printed circuitry board (conventional electronics)
R2R	Roll-to-roll
S2S	Sheet-to-sheet
UV	Ultra-violet
NIR	Near-infrared
SMT	Surface-mounted technology
SMD	Surface-mounted device
IJM	Injection molding
LCA	Life cycle assessment
LCI	Life cycle intake

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