Conductors for smart textiles by 3D printing on textile substrate: adhesion forces and flexibility

Prof. **Ger Brinks**
Eliza Bottenberg, M.Sc.
Jennifer Hesse, Ing.
Richard Groeneveld, Ing.
Saxion UAS, Academy Creative Technology, Research group Smart Functional Materials
Enschede, The Netherlands
e-maila: g.j.brinks@saxion.nl
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Robust and flexible conductors, integrated into textiles is a prerequisite for wearable electronics. However, connecting conductors to electronic components creates a weak spot that hampers the widespread use of conductors in textiles. Our goal was to create intense contact between textiles and conducting material to create one coherent structure that will avoid the existence of stress points. We studied the performance of flexible materials applied through 3D printing in a way that intense bonding between textile substrate and the conducting system could be achieved, while maintaining flexibility. Different polymers were printed on PET/Co surfaces with the aim to increase the adhesion of the polymer onto the textile surface. This knowledge will bring new possibilities into the clothing and technical textile market. We showed that it is possible to deposit flexible polymers on PET/Co blends with excellent bonding strength, while the resulting textile structure has sufficient flexibility for use in wearable textiles.
***Key words**: 3D printing, bonding strength, flexibility, conductive textile, wearables, electronic textiles.*

1. Introduction

Large part of the research of the textile industry is concentrating on the development, production and application of materials for technical (intelligent, wearable "smart") textile. Focus of this research is on new materials, leading to new functionalities. These new materials put an even heavier demand on the industry since these can only be incorporated in textiles if in depth knowledge of processing and application of functional materials is available [1]. The future of smart or wearable textiles lies in the potential of technology where polymers are integrated into fibres and fabrics. This will enable soft intelligent textile products that have a broad spectrum of functions and capabilities that are found in hard electronic products today.

Research into new production methods is a must and an opportunity for the textile industry. One of the future production possibilities of textiles and clothing is 3D printing, a quickly evolving technology that should be taken into consideration [2]. There are only a few random examples of 3D polymer deposition using print technology on textiles: as yet an unexplored field [3, 4].

Smart functional textiles as wearable electronics represent a huge area of research and development. Literally thousands of prototypes have been developed, yet market introduction on a large scale of products based on this technology has not happened yet.

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To quote the Systex vision paper: "The key market hurdles for development and subsequent commercialization of smart textiles applications are of technical, strategic and economic in nature: lack of standardization and regulations for the new products, a lack of shared product development initiatives among the value chain partners as well as a lack of funds for SMEs in new product development. The technological barriers include reliability, durability and ease of use; slow development in the areas like flexibility of electronics, durability and power; and missing interconnection of components. The business barriers include high development and manufacturing costs; high retail prices leading to less consumer acceptance, and scarcity of human resources to carry out new product development. Besides this also a lack of genuine understanding of customer requirements, immature business contexts and, a limited market potential for smart textiles because of cheaper alternatives, needs to be taken into account" [5].

The use of robust and flexible conductors, integrated into textiles, is a prerequisite for wearable electronics. However, connecting conductors to electronic components creates a weak spot that hampers the widespread use of conductors in textiles, in fact it is the main reason why electronics incorporated into textiles have not found widespread application. One major reason is that current conductive materials suffer from one or more of the following drawbacks: not flexible enough (thus bending stress leading to breakages), mechanical properties of textiles and conductors differ too much creating stress spots leading to weak connections, and missing intense bonding between conductor and textile thus creating two different materials in each other's vicinity that react differently to stress.

However 3D printing or polymer deposition on textiles could create interesting applications, particularly direct printing of electronics, PCB's, OLED's, RFID-chips, solar panels and displays. Direct printing of conductive tracks could be very useful and would open the way to robust wearable textiles [6].

Fig.1 Examples of a 3D printed electronic circuits

However: 3D application of functional properties on textiles is not trivial due to the manifold of interactions and binding/adhesion phenomena that occur at the various interfaces. The fact that textiles are flexible, composed of a wide variety of different polymeric materials, prone to several constructions and cloth architectures, and composed of yarn, fibres and filaments makes proper understanding of the interface phenomena a real challenge. Until the processes that play a dominant role in these interfaces are really understood, processing on an industrial scale is limited to trial and error leading to non-generic, ad hoc solutions that cannot be generalized to standardized production processes. These are required for larger scale industrial processes [2,7].

This means that investigating polymer deposition onto textiles of polymers like PLA, doped with carbon could be a versatile route to achieving economic and sustainable conducting textiles. If the mechanism underlying the bonding of polymers with textiles can be controlled for processing then a new route to achieving conductive grids would be opened.

One critically important requirement for textiles is flexibility and related to this: drape. Drape is defined in terms of shape characteristic or the way a fabric hangs down in folds. Bending and shear stiffness influence the drape [20]. For fashion and interior applications obviously a critical parameter, but there are also many technical applications where drape ability is important such as in transport (tarpaulins). Depositing functional polymers may thus influence the drape of a fabric in a negative way and consequently we need to know how critical this may be, so this is one of the subjects in this research project.

Our goal was to create intense contact between textiles and conducting material so as to avoid the aforementioned differences and create one coherent structure that will avoid the existence of stress points. For that reason we studied the performance of flexible materials applied through 3D printing, or better, through polymer deposition, in such a way that intense bonding between textiles substrate and the conducting system could be achieved. This also meant that a suitable new functional polymer printing process had to be developed. We used the basic fused deposition modelling technique that is currently applied in 3D printing. Preliminary examples are shown in Fig.1. In addition the aim is to expand the knowledge and knowhow of 3D printing and make this technology applicable for deposition of functional polymers onto textiles in such a way that process parameters are clearly understood, and pre-defined final product specifications can be met [8]. In this research the main focus was on how to optimize the adhesion of 3D printed polymers to textile surfaces, while maintaining a flexible textile. In previous

research it was discovered that it is possible to print polymers onto textiles but the adhesion of the polymer onto the textile material was not sufficient. Therefore different polymers were printed on PET/Co surfaces to elaborate this problem with the aim to increase the adhesion of the polymer onto the textile surface. This knowledge will bring new possibilities into the clothing and technical textile market. In his extensive review, Baldan described a number of bonding mechanisms. In our case we assume the mechanical interlocking model to be the predominant mechanism for the bonding between the deposited polymer and the textile substrate. This assumption is based on earlier analyses where we found that the viscosity of the melt in combination with the porosity of the substrate and the surface energies of the substrate and the polymer, the viscosity is the predominant factor and thus penetration followed by rapid solidification of the melt, leading to anchoring of the polymer into the textile porous mass is the mechanism that leads to bonding of the polymer [2, 9, 10].

The results, showing the bonding strength of various polymers to polyester/cotton blends and the resulting flexibility of these constructions will be discussed. The main conclusion is that is it possible to deposit flexible polymers on PET/Co blends with excellent bonding strength, while the resulting textile structure has sufficient flexibility for use in wearable textiles. In order to understand the binding phenomena of 3D printing on textiles, we started with a number of commercially available non-conductive polymers. Application in a model electronic conductive system will be shown.

2. Materials and methods

The research was conducted on a 3D FDM (fused deposition modelling) printer Cartesio from Mauk CC, which has a 400×200 mm heated built-in printbed and an E3D V5 extruder, which is able to print flexible polymers with 1.75 mm cross section. With this system it is possible to apply a slight pressure between the melt and the substrate. Tab.1 summarizes the materials used and some characteristic properties for the printing experiments. It also shows the print parameters.

The system parameters used for all printing experiments were: Nozzle diameter n = 0.6, layer height = 0.2 mm, fill density = 80%, infill pattern = rectilinear.

Tab.1 Materials used and their characteristics

Nr.	Composistion	Elasticity (supplier data)	Textruder, °C	Tprintbed, °C	Extrusion speed, mm/s
1	Poly(lactic acid)	Non flexible	250	60	50
2	Thermoplastic Co-Polymer (TPC Flex 65)	Medium	240	50	50
3	Thermoplastic Polyurethane (Ninjaflex®)	Flexible (1000% elongation)	235	80	30
4	Thermoplastic Polyurethane (Filaflex®)	Flexible (700% elongation)	250	60	30
5	Thermoplastic elastomer (rapeseed oil based TPU)	Medium	220	50	30
6	Modified Styrene Butadiene (Bendlay®)	Medium	235	55	50

The substrate used to print on was polyester/cotton 65/35 (170 g/m^2) woven. Porosity was measured using the autoporosimeter from TRI Princeton USA, wicking and dynamic contact angels were measured by the Dataphysics 200.The optical porosity was measured using Perkin Elmer Lambda 900 UV/VIS/NIR spectrophotometer.

The bending tests were executed with the Pure bending tester KES-FB2, Katotech co LDT, No. 8512 – 84. The print was in fact a strip printed on textile, both in weft and in warp direction (55 × 3 mm). This strip was excised resulting in 5 samples of width 0.5 cm. Subsequently the bending curves were measured. In all cases typical bending curves were found. Using these graphs and data slopes and hysteris were calculated. The bending parameter B is the average of the slopes as can be seen in Figure 2 [11]. The same figure also shows the calculation parameters for the hysteresis, 2HB. Some samples showed such a rigidness that real measurements could not be done.

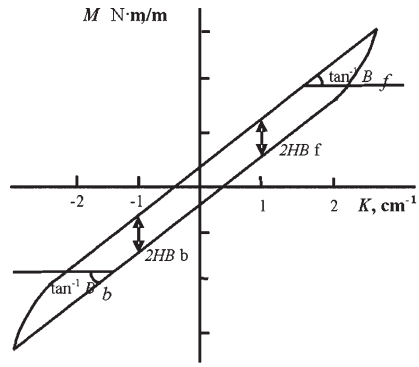


Fig.2 Typical KES bending hysteresis curve [11].

Tensile/peel off tests were performd with the Testometric™ M350-20 CT tester. In all tests the clamp speed was 500 mm/min, load cell type used: ‘6/1000. Sample length 50mm. The test were performed in both warp and weft direction.

Interaction parameters are calculated in accordance with the methodes described by Van Krevelen [12] and extensively described in earlier re-

ports [13]. The overall solubility coefficient δt was calculated using Eq. (1) [12]:

$$\delta t = \sqrt{Ecoh/V} \quad (1)$$

For this analysis we calculate the molar attraction constant F ($(J/cm^3)^{1/2}.mol^{-1}$) based on group contribution and use this to calculate the polar, disperse and hydrogen bonding parameters using the following set of equations (2), (3), (4) and (5):

$$\delta p = \frac{\sqrt{\Sigma Fp^2}}{Va} \quad (2)$$

$$\delta d = \frac{\Sigma Fd}{Va} \quad (3)$$

$$\delta h = \sqrt{\frac{\Sigma Eh}{Va}} \quad (4)$$

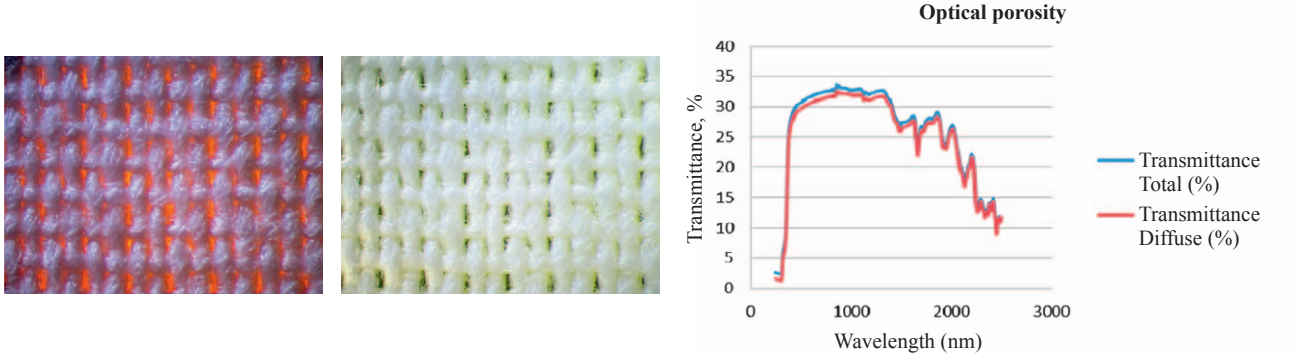
In addition the following equation holds: $\delta t^2 = \delta d^2 + \delta p^2 + \delta h^2 \ldots$ (5)
In these equations V is the molar volume of the constituting monomer, δd, δp, δh, and δt are respectively the dispersion, polar, hydrogen bonding and total solubility/interaction parameters and are a measure for the type and size of attraction between polymers, and are directly related to cohesive energy E_{coh}, the polar and dispersive attraction constant respectively Fp and Fd, and Eh the cohesive energy contribute related to hydrogen bonding.
The surface tension is calculated by the Sugden Parachor equation (6):

$$\gamma = \left(\frac{Ps}{Va}\right)^4 \quad (6)$$

ϒ is the surface energy and Ps is the so called Sugden molar parachor.

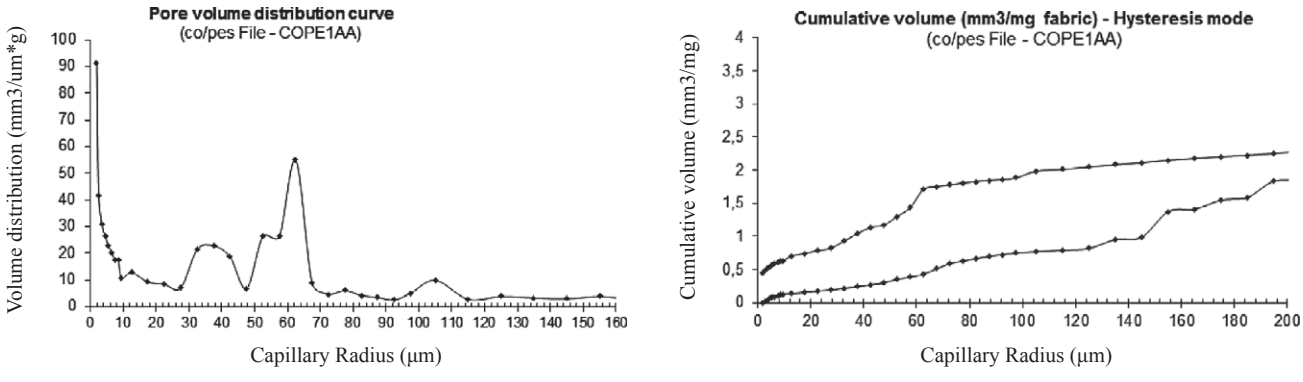
3. Results and discussion

Fig.3 shows the results of the optical porosity tests. The openness factor is a measure for the amount of space available for penetration of molten polymer into the cloth. Usually it is quantified at 280 nm. In the test samples in these experiments the openness factor is found to be 1.2% with a total transmission of 2.65%.
Wicking time analysis showed that the average wicking time was 533 ms, with an initial dynamic surface contact angle (θ) of 75°. Fig.4 shows the pore size distribution measurement revealing inter-yarn pores from 30-70 µm and intra yarn pores in the range of less than 5 µm. There are no pores above 110 µm.
Based on these results it is obvious that for 3D printed layers on textiles, e.g. for wearable applications, PLA is too rigid. We also found the modified Styrene Butadiene (Bendlay®) less suitable. From a flexibility point of view the polyurethane and the Thermoplastic Co-Polymer (TPC Flex 65) polymers are the most suitable polymers for wearable applications.
The results of the peel off tests are summarized in Tab.3.



a) b) c)

Fig.3 Optical porosity of used standard substrate: a) top exposure, b) downside exposure and c) transmision measurements



a) b)

Fig.4 Porosity of used standard substrate: a) pore volume distribution and b) cumulative pore volume

Tab.2 Bending tests

Nr.	Composition	Layer thickness (mm)	Bending rigidity, B 10^{-2}, ($gf.cm^2/cm$)		Bending hysteresis, 2HB 10-2 (gf.cm/cm)	
			warp	weft	warp	weft
Ref.	Untreated textile substrate	0	1.0 +/- 0.2	0.4 +/- 0.2	1.3 +/-0.3	Too low
1	Poly(lactic acid)	Too rigid				
2	Thermoplastic Co-Polymer (TPC Flex 65)	0.75 +/-0.14	5.3+/-8.0	45.3+/-18.7	4.9+/-6.8	11.8+/-6.8
3	Thermoplastic Polyurethane (Ninjaflex®)	0.62 +/-0.03	9.6 +/- 2.0	7.8 +/- 1.0	4.6+/-0.8	3.2+/-0.6
4	Thermoplastic Polyurethane (Filaflex®)	0.66 +/-0.06	14.5+/-4.4	10.3+/-1.6	5.3+/-1.0	4.0+/-0.5
5	Thermoplastic elastomer (rapeseed oil based TPU)	0.62 +/-0.03	8.9 +/-0.6	20.6 +/-3.5	10.4+/-1.6	7.9+/-1.9
6	Modified Styrene Butadiene (Bendlay®)	0.82 +/-0.03	144 +/-36.5	135+/-35.6	Too rigid, printed strip loosened	

In all tests: n=5

Tab.3 Peel off tests

Nr.	Composition	Max. Peel-off strength (N)		Max peel-off distance (%)	
		Warp	Weft	Warp	Weft
1	Poly(lactic acid)	75+/-14	63+/-4	23+/-3	33 +/-6
2	Thermoplastic Co-Polymer (TPC Flex 65)	24+/-9	11+/-2	27+/-2	30+/-2
3	Thermoplastic Polyurethane (Ninjaflex®)	43+/-7	58+/-9	42+/-6	33+/-3
4	Thermoplastic Polyurethane (Filaflex®)	363+/-31	49+/-15	16+/-2	16+/-3
5	Thermoplastic elastomer (rapeseed oil based TPU)	28+/-19	18+/-5	26+/-4	28+/-1
6	Modified styrene Butadiene (Bendlay®)	30+/-4	4+/-2	24+/-3	22+/-3

In all tests: n=5

Tab.4 Interaction parameters

	δt $(J/cm3)^{1/2}$ Van Krevelen	δd $(J/cm3)^{1/2}$	δp $(J/cm3)^{1/2}$	δh $(J/cm3)^{1/2}$	δt $(J/cm3)^{1/2}$ Hoftijzer/Van Krevelen	Υ mN/m
Poly(urethane)	27.7	17.1	6.9	7.9	20.1	51
Styrene Butadiene	17.6	17.2	0.8	0	17.2	38
Poly(lactic acid)	17.2	16.1	15.7	9.5	24.4	75
PET/Cellulose, 65/35	18.3	17.5	10.1	15.1	26.0	57.8

As can be seen in Tab.3, PLA shows good bonding on the chosen PET/cotton. The three polyurethane variants also show reasonable bonding strengths. With the exception of the Ninjaflex variant all show higher bonding in warp direction. A possible explanation for this is that printing in warp direction creates more accessible intra yarn pores since the filaments in the yarn are oriented in the yarn direction and the molten polymer may have the tendency to penetrate the interstitial space in the yarn because of the slight pressure asserted while printing. The filaments act so to say as minor barriers forcing the molten material into the yarn.

Tab.4 gives a summary of the interaction parameters per type of polymer. The substrate is not very open and has limited pore size as shown above (Fig. 3 and 4). We are aware of the fact that the major mechanism for bonding of the molten polymer onto the cotton is mainly through penetration of the molten polymer into the pores followed by solidification in the pores of the cotton woven (locking mechanism). However, some effect of the surface characteristics and physicochemical properties may be expected since in such a relative thin 3D construction as the used woven. Based on the relative low surface energy of Styrene Butadiene it may be expected that the bonding strength between cotton and Styrene Butadiene is less than for the other polymers, as indeed is the case. Next to the locking mechanism and based on the polar en hydrogenbridge forming capacity the more charged polymers PLA and PUR should show higher bonding-strengths, as is indeed the case.

4. Conclusions

In conclusion: the polyurethane polymer variants are more suitable for

printing flexible conductive tracks on textiles for wearable applications. The results are based on one type of polyester/cotton. Further research will be conducted including several cotton and polyester/cotton wovens.

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