Adhesion of three-dimensional printing on textile fabrics: Inspiration from and for other research areas

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Abstract
Combining textile fabrics with three-dimensional printed items can be a good approach to save time and money as compared with purely three-dimensional printed large-scale objects, to reach desired position-dependent mechanical properties, for design and technological purposes. The main challenge in such bi-material systems is the adhesion between both partners of the composites. Although some experimental research on this topic has been performed during the last years, only few theoretical investigations exist which may support striving for material combinations with higher adhesion. Here, we give an overview of the recent state of experimental research on adhesion in textile/polymer composites as well as of theoretical investigations on adhesion inside the three-dimensional printed part. Combining both topics, we suggest further research approaches to increase the textile/polymer adhesion.

Keywords
Three-dimensional printing, textile fabric, adhesion, rheology, healing model, wetting, diffusion

Date received: 25 January 2020; accepted: 13 February 2020

Introduction
The term ‘3D printing’ (three-dimensional printing) includes many different technologies, such as stereolithography (SLA), selective laser sintering (SLS), fused deposition modelling (FDM) or polyjet modelling (PJM).¹-⁵ Although these technologies offer large advantages in comparison with previous methods to prepare objects from polymers or metals, such as individualization and fast production without the necessity to prepare an expensive form before, similar to in injection dye casting, there are also disadvantages which have not yet been overcome, such as large anisotropies inside 3D printed objects,⁶-⁸ making mechanical properties hard to control,⁹,¹⁰ a high waviness or roughness, depending on the technology,¹¹-¹⁴ and long printing durations which make the process less interesting for larger numbers of identical objects to be produced.

While the surface structure is immanent to the chosen technology and usually modified by chemical or mechanical after-treatments, both a reduction of time and an optimization of the mechanical properties in spite of the usually low tensile strength of typical 3D printing...
materials can be reached by combining 3D printing with a substrate prepared by another technology, for example, a textile fabric. In such composites, mechanical properties along and normal to the textile plane can be tailored by the mechanical properties of both partners as well as by the distribution and shapes of the 3D printed parts.

Printing on a textile fabric can be quite simple if it is a net fabric with large open areas, allowing for printing through the fabric and embedding it into the 3D printed part, in this way building form-locking connections. In most cases, however, the adhesion between both materials is less easily achieved.

Here, we give an overview of recent research approaches to increase the adhesion of different textile/polymer composites, concentrating on the FDM technology. Although SLS can be used to prepare textile-like structures, similar to FDM, no reports can be found yet in the literature of combining other 3D printing technologies with textile substrates than in FDM.

**Experimental**

Besides the large number of studies concentrating on embedding short fibres into a 3D printing feedstock or an FDM printing filament to improve the mechanical properties of the resulting objects, here we concentrate on embedding fibres and textile fabrics during the printing process since this is a part of the process which can usually be modified without specialized equipment like a screw extruder to prepare FDM filaments.

Printing with polylactic acid (PLA) on open-pore knitted fabrics, the adhesion between both materials is often found sufficient for the respective application. Not all publications provide values of adhesion measurements, which makes comparison sometimes complicated. Other publications report on larger problems with the adhesion inside textile/polymer composites.

Most recently, Eutinoma-Diff et al. found a significant influence of the build platform temperature, on the one hand, and fabric parameters such as orientation and weft density of the woven fabric, on the other hand, on the adhesion between textile substrate and imprinted PLA objects. Generally, they rated the adhesion between both materials as insufficient for the preparation of a composite with good mechanical properties.

Kozior et al. investigated for the first time the possibilities to perform 3D printing on electrospun nanofiber mats prepared from polyacrylonitrile. They found that 3D printing was even possible on the relatively brittle carbonized nanofiber mats and could be used to mechanically stabilize the nanofiber mats for filter applications, which is opposite to most approaches in which the textile fabric provides the mechanical in-plane stability. The combination of 3D printing with electrospinning was reported earlier by Rivera and Hudson, who built a special system combining melt electrospinning and 3D printing of the same material, enabling creating single-material, multidimensional composites.

Oyon Calvo et al. found highest adhesion values for combinations of a porous textile fabric with a flexible filament. Similarly, Meyer et al. reported on the best adhesion of flexible filaments on hairy woven substrates. The advantage of the flexible filaments was attributed to their lower viscosity during printing, enabling stronger penetration into the pores of the substrate and thus building a better form-locking connection. Tadesse et al. also reported sufficient adhesion forces for flexible NinjaFlex filament printed on a polyester fabric. Oppositely, acrylonitrile butadiene styrene (ABS) was found to show a lower adhesion than PLA, possibly due to its higher viscosity during printing.

Besides the viscosity of the filament during the printing process, the distance between nozzle and substrate is also an important factor, as reported several times in the literature for different filament materials and textile fabrics. Other groups investigated other printing parameters and found, for example, a positive correlation with the adhesion for the warp and weft count, fabric thickness and handle.

Finally, it should be mentioned that some studies report on successful tests to increase the adhesion of the textile fabrics by chemical pre-treatments, mostly following the Korger law that more hydrophilic surfaces allow for a better adhesion, or by polymer coatings.

Generally, these experiments show that properly adjusted low viscosity and high pressure may induce penetration of the polymer into the fabric, resulting in a form-locking connection. This effect may also be supported by good wetting properties which seem to correlate with hydrophilic textiles. For rigid – and thus usually highly viscous – polymers, it may be possible to replace penetration of the 3D printing polymer into the fabric by an intermediate polymer coating layer. There is, however, no theoretical investigation or modelling of these effects yet. This is why the next section gives an overview of theoretical approaches to understand the adhesion within an FDM printed object as the base for a possible transfer to the special situation in textile/polymer 3D printed composites.

**Theoretical considerations**

Why does a 3D printed polymer adhere on the substrate? Transferring this question to the inside of a 3D printed object translates to ‘Why does a printed road adhere to the neighbouring one, and why is a printed layer fixed on the former one?’ This apparently very basic question is nevertheless still discussed in the literature. Possibly, full understanding of these effects necessitates a molecular-level perspective.

For injection or compression moulding, Wool and O’Connor suggested a crack healing theory which is based on five different stages, that is, surface rearrangement, surface approach, wetting, diffusion, and randomization. Here,
we find indeed the wetting mentioned as a possible factor influencing the adhesion as well as the viscosity which correlates with the diffusion, both factors made responsible for most mechanical properties.\textsuperscript{41}

Many authors have transferred this model to the FDM process. Bellehumeur et al. modelled the bond formation between neighbouring polymer filaments and attributed the bond formation between them to the necks formed between them, that is, the wetting, as well as molecular diffusion and randomization at the interface. They suggest modelling the bond formation as sintering process for which wetting is important or as polymer welding in which molecular diffusion is more important. By comparing theoretical and experimental analysis, they come to the conclusion that extrusion temperature is more important for neck formation, that is, bonding, than the envelope temperature and underline that temperature decreases too fast to reach complete bonding between neighbouring filaments so that the mechanical properties of the bonding zone cannot be identical with those inside a filament.\textsuperscript{42} In another study, the group showed that sintering occurred only for a very short time before the filament temperature was reduced below the critical sintering temperature, whereas envelope temperature and convection coefficient variations strongly influenced the cooling temperature profile and thus the bond strength between adjacent filaments.\textsuperscript{43} Seppala et al.\textsuperscript{44} mentioned that the weld time is a crucial factor to increase weld strength, but they could not reach more than 70% of the bulk strength in their experiments, for which different possible reasons were given.

McIlroy and Olmsted developed a non-isothermal welding process model and showed in this way that the mechanical strength should not be limited by the interpenetration depth, but insufficient recovering time during cooling was found for typical printing conditions. Such a disentangled weld structure, resulting from disentanglement inside the nozzle, may increase the entanglement molecular weight and thus reduce the mechanical properties of the weld. They suggest using higher temperatures or less entangled materials to form thicker and more entangled welds, while the printing speed did not influence mechanical integrity in their model.\textsuperscript{45} Geng et al.\textsuperscript{46} even found in their investigation of FDM printing polyether-ether-ketone (PEEK) that higher extrusion speeds could eliminate extrusion defects and was thus preferable to gain a more compact layer surface.

Coogan and Kazmer concentrated on the diffusion across FDM printed layers which was modelled by a one-dimensional transient heat analysis. Rheological data were used to set up a temperature-dependent diffusion model in which time-dependent diffusion coefficients were integrated. The bond strengths modelled in this way were found to correlate with measured values.\textsuperscript{47}

Other groups concentrated on extending the Frenkel sintering model.\textsuperscript{48} Pokluda et al.\textsuperscript{49} based their model on the interplay between surface tension and viscous dissipation, while Gurrala and Regalla\textsuperscript{50} concentrated on viscous sintering of cylindrical filaments as the base to calculate neck growth. Faes et al.\textsuperscript{51} investigated the interlayer cooling time by producing one or more objects at the same time and found an inverse correlation between the cooling time and the ultimate tensile strength and elongation at break as well as a positive correlation between cooling time and variability of these mechanical properties.

Yin et al. concentrated on the inter-molecular diffusion process for which they developed a new model. They found a significant influence of the building stage temperature which was not mentioned as very important parameter in most other studies.\textsuperscript{52} Yan et al.\textsuperscript{53} also found that the forming environment temperature is more important than the nozzle temperature, and that convection in the forming room should be low.

Hart et al. suggested overcoming the low bonding between adjacent layers by thermal annealing above the glass transition temperature inside an aluminium fixture after FDM printing. Using different annealing temperatures from 75°C to 175°C and durations from 2 to 168 h, they found stabilization of crack propagation and an increase in toughness values of up to more than 2000% for relatively high temperatures and durations.\textsuperscript{54} Wu et al.\textsuperscript{55} suggested applying pressure under ultrasonic treatment and found fusing of the pores in the samples, resulting in an increase of bending strength and modulus of ABS printed samples.

Generally, despite the large amount of theoretical studies and comparison with experiments, it does not seem to be fully clear yet whether wetting or diffusion or other factors are most important. Only recently, the contact area which is related to the pressure of the molten polymer onto previous layers was modelled, based on a model used for composites,\textsuperscript{56} by Coogan and Kazmer\textsuperscript{57} who showed that melt pressure at the nozzle exit correlated with viscosity, flow rate, road width and layer height. They suggested small layer heights as most important to increase the contact with the previous layer and underlined the importance of contact area while wetting could be nearly neglected.

While the basic parameters for the adhesion between neighbouring and adjacent filaments inside 3D printed objects are more or less known, although their respective influence is still discussed and not consistently modelled nor measured, transferring this knowledge to the original problem of 3D printing on textile fabrics is not easy. Most importantly, the materials of textile fabric and 3D printed object usually differ, with the first not even necessarily being a thermoplastic polymer. Simple experiments show directly that, for example, printing with a flexible polymer
like NinjaFlex or FilaFlex on relatively rigid PLA results in a much better adhesion than the opposite stacking order.

In addition, the mechanical and morphological properties of a textile fabric differ from those of a 3D printed layer in terms of flexibility, bendability, compressibility, structural orientation and so on. Since the overall process of material deposition takes place in several stages, the final characteristics and the time-dependent intermediate ones can be classified using image-processing tools. In this way, the quantitative parameters describing technological ones can be classified using image-processing tools. In this way, the quantitative parameters describing technological mechanisms can be obtained using adequate statistical or fractal-related analysis.58

The next section will thus discuss which of these findings may be used as an inspiration for further research on 3D printing on textile fabrics – and oppositely, which open questions could be addressed in the theoretical and experimental research on interlayer adhesion.

Discussion

The following parameters were found important for the adhesion inside an FDM printed object and may possibly also be related to the adhesion of an FDM imprinted layer on a textile fabric:

- **Wetting.** This process can also be expected to be crucial for 3D printing on textile fabrics, as Korger et al.37 showed for the first time by investigating the dependence of adhesion on the hydrophobicity of a textile fabric.
- **Diffusion.** This process could only be taken into account for FDM printing on textile fabrics if the latter consisted of or were coated with a similar or identical polymer. Coating was indeed found to increase adhesion for several coating/printing polymer combinations.28,39
- **Pressure.** This parameter correlates partly with the z-distance which was found crucial in 3D printing on textile fabrics.32 It also explains the experimental findings that with higher printing temperature, the maximum adhesion was not increased, but the dependence on the z-distance was shifted towards larger distances.53

On the other hand, some of the experimental and theoretical investigations in interlayer adhesion do not have a counterpart yet in the research on 3D printing on textile fabrics. The following points should thus be evaluated:

- **Printing speed.** Here, different findings are available in the literature for interlayer adhesion, although there seem to be no clear trends for 3D printing on textile fabrics yet.
- **After-treatment.** Although annealing or ultrasonic strengthening is not unusual for fully 3D printed fabrics, the necessity to perform this after-treatment step inside a form to avoid undesired shape deviations may make this step complicated for 3D printed objects on textile fabrics; however, basic research on this topic should be performed.

Other parameters, especially the building stage temperature, cannot be easily transferred onto the idea of 3D printing on textile fabrics. Oppositely, the often observed ‘air buffers’ between textile fabrics and 3D imprinted polymers, especially for printing with ABS,28,32 have not yet been observed in interlayer adhesion investigations and necessitate a theoretical explanation.

This and other challenges found in 3D printing on textile fabrics can thus also be used as an inspiration for the investigation of pure 3D printed objects. As mentioned in the last section, important differences are from the physical point of view, mechanical and morphological properties, and from the chemical point of view, the differences in the materials and thus surface energies.

Our suggestion is thus to start modelling and experimentally investigating bi-material FDM printed objects, taking into account the usual materials typically used for 3D printing, as well as an extended spectrum including flexible and other special filaments which can nowadays be printed with several inexpensive FDM printers. On the other hand, variations of infill patterns in such theoretical and experimental investigations may approximate diverse textile structures with their mechanical and morphological parameters.

Conclusion

We gave an overview of the scientific studies available on 3D printing on textile fabrics as well as on theoretical and experimental investigations of the adhesion between neighbouring and adjacent layers in fully 3D printed objects. Comparing the findings from both research areas, we discuss possible theoretical explanations for experimental findings in the adhesion of FDM printed polymers on textile fabrics. We also give suggestions for further research on this topic, derived from previous findings on interlayer adhesion, and vice versa.

In this way, both research areas can inspire and learn from each other.

Declaration of conflicting interests

The author(s) declared no potential conflicts of interest with respect to the research, authorship and/or publication of this article.

Funding

The author(s) received no financial support for the research, authorship and/or publication of this article.

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