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Increasing Adhesion of 3D Printing on Textile Fabrics by Polymer Coating

Povečanje adhezije 3D potiska tekstilnih tkanin s polimernim premazom

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Abstract

3D printing is a technology which has recently found its way into the field of textile fabrics, from fashion design to technical textiles. By combining both technologies with their advantages, new composites with novel physical properties can be created. Increasing adhesion between both components, however, still remains challenging. This paper suggests a new method to improve the adhesion of a 3D printed object on a textile fabric by previously coating the latter with a polymer layer. In this way, adhesion can be substantially enhanced without significantly changing the bending stiffness and haptic properties of a fabric. In this study, this procedure worked especially well for printing PLA (poly(lactic acid)) on PMMA (poly(methyl methacrylate)) or PLA coatings, while for printing ABS (acrylonitrile butadiene styrene), the best textile coatings were ABS and PLA.
Keywords: 3D printing, composite, adhesion, polymer coating

Izvleček

3D tisk je tehnologija, ki se v zadnjem času uveljavlja tudi na področju tekstilnih materialov, od modnega oblikovanja do tehničnih tekstilij. Z združitvijo prednosti obeh tehnologij lahko ustvarimo nove kompozite z novimi fizikalnimi lastnostmi. Izboljšanje adhezije med obema komponentama pa je še vedno izziv. V članku je predlagana nova metoda za izboljšanje adhezije 3D potiska na ploskovni tekstiliji s predhodnim premazom tekstilije s polimernim slojem. Tako se oprijem lahko bistveno izboljša, ne da bi se pri tem znatno spremenile upogibna togost in haptične lastnosti ploskovne tekstilije. V tej študiji je bil ta postopek posebej primeren za tisk PLA (polimlečne kisline) na premaze iz PMMA (polimetil metakrilata) ali PLA, za tisk ABS (akrilonitril butadien stirena) pa so bili najboljši tekstilni premazi ABS in PLA.
Ključne besede: 3D tisk, kompozit, adhezija, polimerni premaz

1 Introduction

3D printing is one of the new and inspiring technologies which have been developed over the past decades and offer possibilities to establish new production processes, individualisation and creation of objects which can otherwise not be produced in one part [1, 2]. In particular, the Fused Deposition Modeling (FDM) technology is promising, since it is not only relatively inexpensive but also allows for combining 3D printed objects with substrates from different materials, e.g. textile fabrics.

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The printing process in the FDM technology is based on a resistively heated extruder, melting a polymer filament which is transported through this extruder, pressing the melt through a nozzle and depositing it on a printing bed. According to the CAD model, an object is formed layer by layer by printing each layer on the previous one until the object is complete [3]. Typical printing polymers are acrylonitrile butadiene styrene (ABS), poly(lactic acid) (PLA), polyamide ("nylon"), polycarbonate, polypropylene etc. [4]. However, most of these materials and the objects printed from them have insufficient mechanical properties for several applications [5, 6]. In addition, the printing process is still relatively slow, as compared to die casting. Both problems suggest combining 3D printed shapes with the materials which can have higher tensile strength and can be produced at a faster rate, e.g. textile fabrics.

Several attempts have thus been made during the last years to combine 3D printing and textile fabrics. The adhesion between both materials is the most challenging. The easiest way to solve this problem is using net-like fabrics with large open areas between two subsequent printing layers, enabling intermolecular bonding between the polymer layers below and above the textile fabric and thus creating a form-locking connection [7]. The integration of single textile fibres may be useful, but can lead to great problems when untwisted yarns (e.g. carbon fibre yarns) are used [8]. A possible solution for this problem can be an additional heat treatment of the whole composite after the printing process [9]. Ironing thin polymer/textile composites, on the other hand, was found to counteract the adhesion [5]. The extruder and printing bed temperature, in turn, could be used to increase the adhesion [10].

A large impact of the distance between the nozzle and textile fabric has been shown in multiple studies (e.g. [11]). Moreover, the fabric morphology was shown to strongly influence the adhesion – generally, adhesion forces increase at thicker fabrics with larger pores in and between the yarns in which the printing polymer can protrude to build form-locking connections [11].

The chemical properties of textile fabrics and the printing polymers were also found to significantly influence the adhesion between both materials [12]. This resulted in the idea to increase the adhesion by washing or plasma treatment of diverse materials, both of which showed positive or negative effects on different textile fabrics. Generally, more hydrophilic textile fabrics showed better adhesive properties [13]. A more detailed study of the influence of diverse chemical pre-treatments was performed recently, verifying the idea of hydrophilic properties being supportive of increased adhesion for several treatments, while a few outliers indicated that additional effects have to be taken into account [5].

A completely different approach which has not yet been reported in the scientific literature is coating textile fabrics with a polymer solution prior to the 3D printing process. Such polymer solutions can be used to create thin layers on the top of single fibres or yarns, leaving the existing pores open and thus not completely blocking water vapour or air transition [14].

In this paper, we report on the first tests of printing PLA and ABS on a thin cotton fabric with different polymer coatings and show that in some combinations, significant improvements could be achieved compared to uncoated fabrics.

2 Materials and methods

The textile fabric used for printing was a thin cotton woven fabric (thickness 0.21 mm), which showed very low adhesion in the former investigation [11]. This fabric was used in its original state and with different polymer coatings:

- PLA (5% dissolved in hot 1,2-dichlorobenzene)
- ABS (5% dissolved in acetone)
- PMMA (poly(methyl methacrylate), 5% dissolved in acetone)
- PA (soluble amorphous copolyamide from ε-caprolactam, hexane-1,6-diamine, hexanedioic acid and 4,4-diaminodicyclohexylmethane, 5% dissolved in 80% ethanol and 20% water).

After dissolving polymers by stirring them for 2 h using a magnetic stirrer (in the case of PLA, the hot plate was set to 120 °C), they were applied on the cotton fabric with a metering wire coating (wet film thickness 100 μm), which was used to homogenise the thickness of the fluid film, and dried in an exhaust hood at room temperature for one day.

As printing materials, PLA and ABS (both purchased from Filamentworld, Neu-Ulm/Germany) were used, which are often applied in the FDM printing and resulted in the best and worst adhesion in former test series [11].

For 3D printing, an Orcabot XXL (Prodim, The Netherlands) with the nozzle diameter of 0.4 mm, layer thickness of 0.2 mm, nozzle temperature of 200 °C (PLA)/220 °C (ABS) and printing bed temperature of 60 °C was used.
For the adhesion tests, rectangles of 250 mm × 25 mm in size with the thickness of 0.4 mm (corresponding to two layers) were printed on uncoated and coated fabrics. The tests were performed using a universal testing machine according to DIN 53530 and evaluated according to ISO 6133, applying the method for more than 20 peaks. Using this standard method, one end of the printed rectangle was separated manually from the textile fabric. The printed rectangle and the textile fabric could thus be fixed in one of the clamps of the universal test machine. Afterwards, the clamps were moved apart with the speed of 100 mm/min and the force-displacement curve was measured. 3 samples per coating material were tested.
The z-distance between the printing bed and nozzle was fixed at (0.28±0.02) mm, i.e. 0.07 mm above the textile surface. This value was found to be small enough to reach certain adhesion by pressing the filament into the textile fabric and large enough to avoid the clogging of the nozzle and the undesired roughness increase of 3D printed layers. Both effects typically occur when the distance between the nozzle and textile surface is too small and the filament is blocked too much when pressed through the nozzle.
An optical evaluation was performed using a digital microscope VHX 600 / VH-Z20R (Keyence, Neu-Isenburg, Germany) and a confocal laser scanning microscope (CLSM) VK-9000 (Keyence). The contact angles were measured 10 times on each material, using the aforementioned digital microscope with the option to orient it in parallel to the ground, enabling taking images from the side. Droplets of 18 µl distilled water were used. The contact angles were calculated by image evaluation, fitting the droplets by ellipses and the surface by a straight line.

3 Results and discussion

Firstly, the coated samples were investigated with CLSM. The results are depicted in Figure 1.
Despite all coatings being prepared with 5% solid content and identical coating procedures, differences were visible between different materials. Especially PLA showed a thicker layer on fibres, most probably resulting from the special solvent. Since PLA was dissolved at a higher temperature to obtain sufficient solubility, it can be assumed that it solidified during the coating process due to the cooling down and could thus not penetrate between single fibres, but remained on the yarn surface, building large closed areas.
The other three coatings showed fine layers on the top of single fibres, while the pores between the yarns were in most cases still open.
Next, the hydrophobic properties of coated fabrics were tested and compared with the properties of the original cotton fabric. While the latter was strongly hydrophilic, all coatings significantly increased the hydrophobicity. The PA coating still resulted in a slightly hydrophilic surface, while all other polymer coatings led to hydrophobic surfaces. It should be mentioned, however, that in all cases the droplets used for the contact angle measurements penetrated into the fabric,

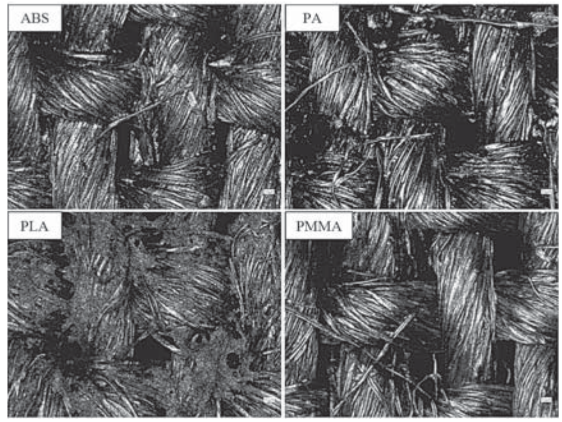

Figure 1: CLSM images of polymer-coated samples (nominal magnification 400×)

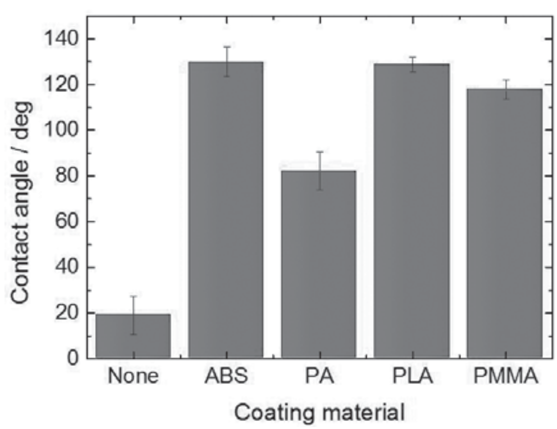

Figure 2: Contact angles of uncoated cotton and polymer-coated fabrics

most probably as the cotton fibres were not completely surrounded by the respective polymers and the water passed through them by capillary forces.

With respect to the findings mentioned before that hydrophilic textile fabrics often result in higher adhesion forces to an imprinted polymer than hydrophobic ones, polymer coatings do not appear to be an ideal solution to increase adhesion between both materials. Thus it must be mentioned that these previous results were based on mainly physical (form-locking) connections between both partners, while in our case molecular interactions got (more) relevant.

The results of the adhesion tests are depicted in Figure 3. For the uncoated cotton fabric, both PLA and ABS resulted in very low adhesion forces, meaning that both materials could already be separated by simply bending the composite. Increasing the physical adhesion could be done by reducing the z-distance between both materials, resulting in an uneven surface and the danger of a clogged nozzle. For the fabric under examination, the smallest z-distance was –0.02 mm, meaning that the nozzle would scratch the glass printing bed if no textile fabric separated the two. In this way, significantly increased adhesion forces could be reached for PLA and ABS ("min z"). Comparing these values with those gained on the polymer-coated fabric showed an inconsistent result. On the one hand, printing PLA on PMMA or PLA coatings resulted in significantly increased adhesion forces, as compared to the printing at minimised z-distance. On the other hand,

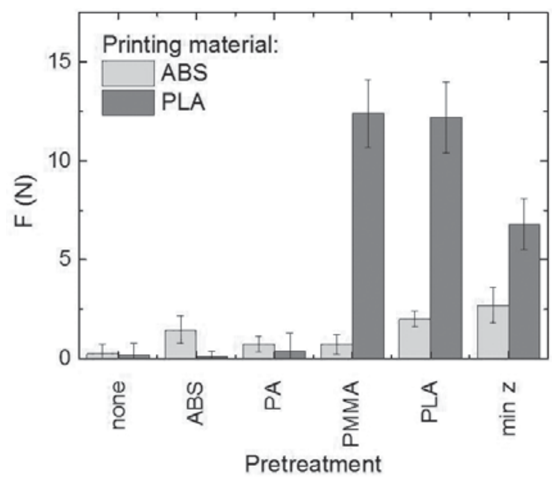

Figure 3: Adhesion forces of ABS and PLA when printed on uncoated cotton, differently polymer-coated cotton and additionally uncoated cotton with minimised z-distance between nozzle and printing bed

ABS showed the best results for the minimised z-distance, although the differences to the printing on coated fabrics were in most cases not significant.

Before these results were interpreted, microscopic images were taken to investigate the printed surfaces as well as the interfaces between the textile fabrics and imprinted polymers.

Figure 4 shows a comparison between the surfaces of two layers of PLA, printed at a reasonable z-distance (z = 0.28 mm) and at the minimised value (z = –0.02 mm). The results for ABS look identical. Obviously, by pressing the polymer with too much force into the textile fabric, the typical linear surface structure got destroyed, and the roughness was increased. This finding was supported by the tactile examination of fabrics – printing at very low z-distances resulted in uncomfortably rough surfaces for PLA as well as ABS. This method is thus more suitable for thicker objects in which this roughness can level out during the following layers.

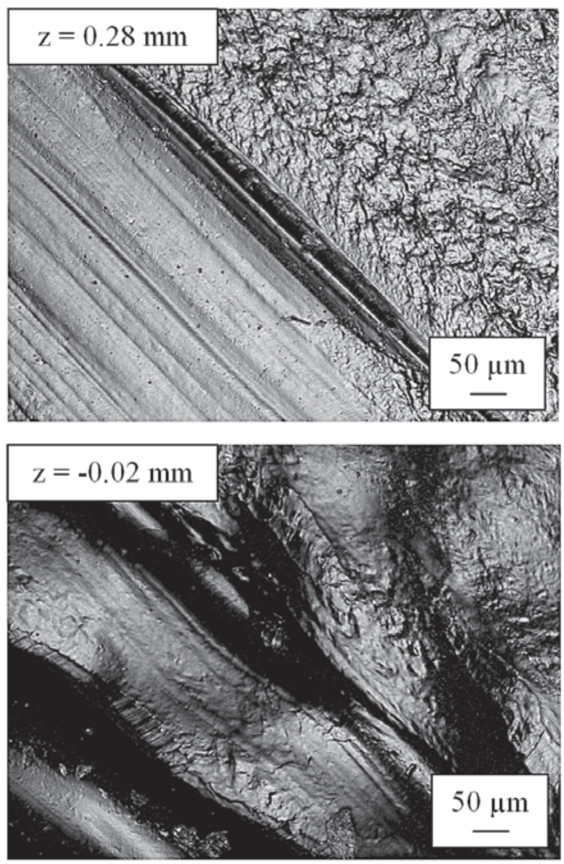

Figure 4: CLSM images of surface of printed PLA rectangles with two layers height, printed in two different z-distances (nominal magnification 400×)

Figure 5 depicts microscopic images of ABS and PLA, respectively, printed on pure cotton. Both layer heights are similar. Both polymers did not penetrate into the textile fabrics. In the case of PLA, even the air layer seems visible along the interface. The lack of interpenetration of both materials clearly explains the very low adhesion forces for 3D printing on pure fabrics.

Next, Figure 6 shows the interfaces between printed ABS and polymer-coated fabrics. The air layers are visible for PA- and PMMA-coated cotton fabrics on which the adhesion was not significantly increased by the coating. For the ABS and PLA coatings, the textile fabric and polymer seem to be in direct contact.

Figure 7 shows the interfaces between printed PLA and polymer-coated cotton fabrics. While on the ABS-coated fabric, an air layer is clearly visible, the print on PMMA seems to be very well-connected, with the polymer completely following the morphology of the textile surface.

Combining the results of these microscopic evaluations with simple chemical rules now allows for explaining the results of the adhesion tests. Firstly, equal materials should form intermolecular bonds, explaining the relatively good adhesion of ABS on the ABS coating and of PLA on the PLA coating. Secondly, the microscopic images suggest that ABS on the PLA coating and PLA on the PMMA coating should also result in increased adhesion values, which corresponds to the results of the adhesion tests depicted in Figure 3. Neither ABS nor PLA adhere more strongly on PA coatings, indicating that the hydrophobicity does not play a role here.

Since thermoplastic polymers are usually very poor heat conductors, it can be assumed that the coating does not melt significantly due to the contact with the printing material. According to Vojuckii's diffusion theory of adhesion, the adhesion forces are determined *inter alia* by the partial diffusion of polymers into the

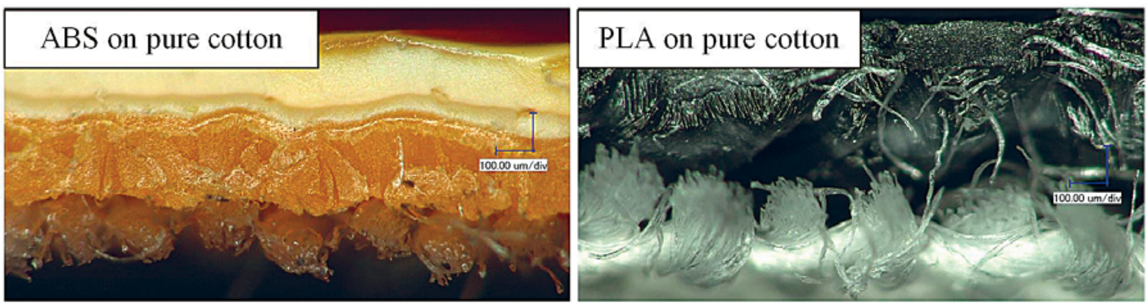

Figure 5: Microscopic images of interfaces between ABS (orange) and PLA (black) rectangles with two layers height, printed on pure cotton (nominal magnification 200×)

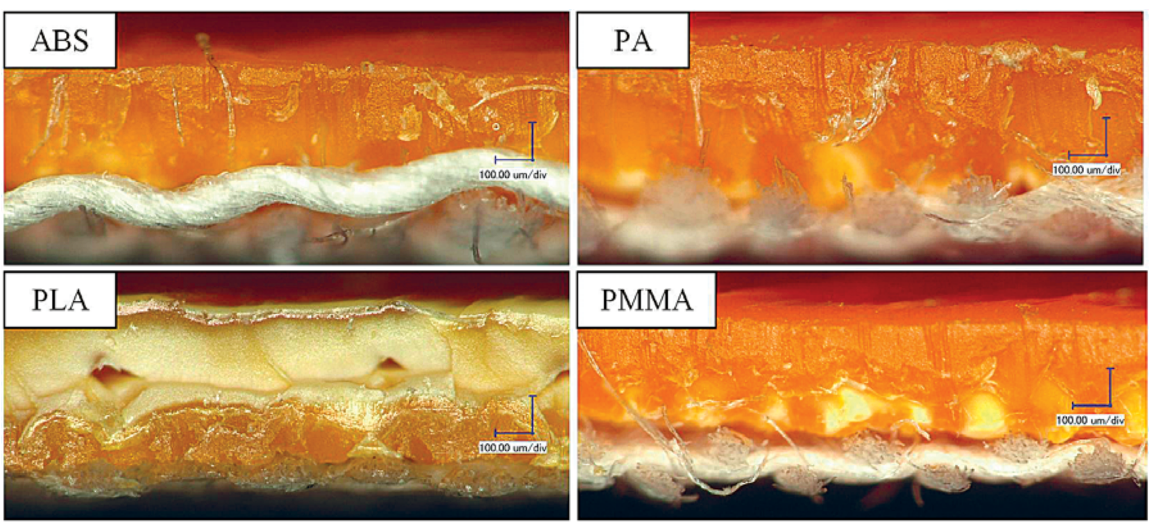

Figure 6: Microscopic images of interfaces between printed ABS and cotton fabrics coated with different polymers (nominal magnification 200×)

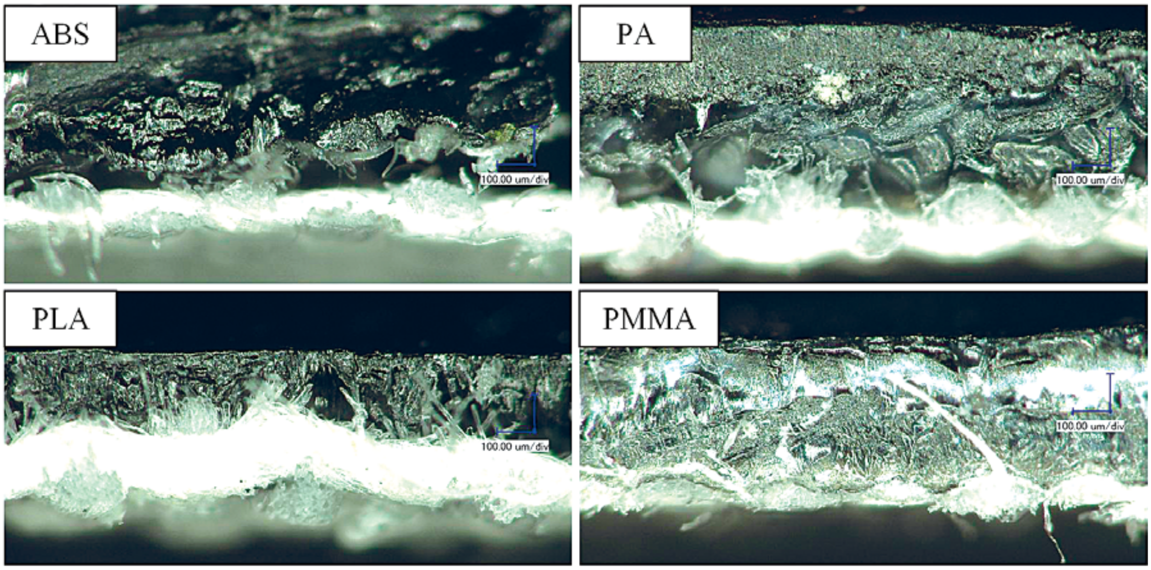

Figure 7: Microscopic images of interfaces between printed PLA and cotton fabrics coated with different polymers (nominal magnification 200×)

surface of the coating material [15]. Since in this case the printing material has melted, the diffusion of the coating material into the printing material is likely to play a subordinate role. Apparently, the adhesion between a 3D printing polymer and a textile fabric can be increased by previously coating the fabric with an appropriate polymer. A significant improvement, however, could only be achieved for PLA, while the absolute values of adhesion of ABS on all coated fabrics were still low. In comparison with the best coatings for the printing of PLA, PMMA is advantageous due to the simpler coating method and the more homogeneous coating. In the next step, these experiments should be extended to combining physical and chemical adhesion by using thicker fabrics with larger pores and evaluating whether in this case, an additional coating can also increase adhesion between both materials.

4 Conclusion and outlook

Cotton fabrics were coated with different polymers before 3D printing with ABS or PLA on them. Some combinations resulted in a significant increase of adhesion between the polymer and textile fabric. Besides the expected result that ABS on the ABS coating and PLA on the PLA coating was going to lead to improved adhesion, ABS on the PLA coating and PLA on the PMMA coating were also found to be supportive which could be explained by the diffusion of the PLA coating through the ABS printing and the reduction of the air layer at the interface for PLA on the PMMA coating.

Future research will concentrate on testing more coating and printing polymers and combining different adhesion mechanisms by using thicker textile fabrics with more pores.

References

1. BEN-NER, Avner, SIEMSEN, Enno. Decentralization and localization of production: the organizational and economic consequences of additive manufacturing (3D printing). *California Management Review*, 2017, **59**(2), 5–23, doi: 10.1177/0008125617695284.
2. DUARTE, Lucas C., CHAGAS, Cyro, RIBEIRO, Luiz Eduardo B., COLTRO TOMAZELLI, Wendell K. 3D printing of microfluidic devices with embedded sensing electrodes for generating and measuring the size of microdroplets based on contactless conductivity detection. *Sensors and Actuators B: Chemical*, 2017, **251**, 427–432, doi: 10.1016/j.snb.2017.05.011.
3. NOVAKOVA-MARCINCINOVA, Ludmila. Application of fused deposition modeling technology in 3D printing rapid prototyping area. *Manufacturing and Industrial Engineering*, 2012, **11**(4), 35–37.
4. NOORANI, Rafiq. *Rapid prototyping: principles and applications*. New Jersey : John Wiley & Sons, 2005.
5. KOZIOR, Tomasz, DÖPKE, Christoph, GRIMMELSMANN, Nils, JUHÁSZ JUNGER, Irén, EHRMANN, Andrea. Influence of fabric pretreatment on adhesion of 3D printed material on textile substrates. *Advances in Mechanical Engineering*, 2018, **10**(8), 1–8, doi: 10.1177/1687814018792316.
6. FAFENROT, Susanna, GRIMMELSMANN, Nils, WORTMANN, Martin, EHRMANN, Andrea. 3D printing of polymer-metal hybrid materials by fused deposition modeling. *Materials*, 2017, **10**(10), 1199, doi: 10.3390/ma10101199.
7. SABANTINA, Lilia, KINZEL, Franziska, EHRMANN, Andrea, FINSTERBUSCH, Karin. Combining 3D printed forms with textile structures – mechanical and geometrical properties of multi-material systems. *IOP Conference Series: Materials Science and Engineering*, 2015, **87**, doi: 10.1088/1757-899X/87/1/012005.
8. RICHTER, Christoph, SCHMÜLLING, Stefan, EHRMANN, Andrea, FINSTERBUSCH, Karin. FDM printing of 3D forms with embedded fibrous materials. *Design, Manufacturing and Mechatronics*, 2015, 961–969, doi: 10.1142/9789814730518_0112.
9. MORI, Ken-ichiro, MAENO, Tomoyoshi, NAKAGAWA, Yuki. Dieless forming of carbon fibre reinforced plastic parts using 3D printer. *Procedia Engineering*, 2014, **81**, 1595–1600, doi: 10.1016/j.proeng.2014.10.196.
10. SANATGAR, Razieh Hashemi, CAMPAGNE, Christine, NIERSTRAZ, Vincent. Investigation of the adhesion properties of direct 3D printing of polymers and nanocomposites on textiles: Effect of FDM printing process parameters. *Applied Surface Science*, 2017, **403**, 551–563, doi: 10.1016/j.apsusc.2017.01.112.
11. GRIMMELSMANN, Nils, LUTZ, Mirja, KORGER, Michael, MEISSNER, Hubert, EHRMANN, Andrea. Adhesion of 3D printed material on textile substrates. *Rapid Prototyping Journal*, 2018, **24**, 166–170, doi: 10.1108/RPJ-05-2016-0086.
12. PEI, Eujin, SHEN, Jinsong, WATLING, Jennifer. Direct 3D printing of polymers onto textiles: experimental studies and applications. *Rapid Prototyping Journal*, 2015, **21**(5), 556–571, doi: 10.1108/RPJ-09-2014-0126.
13. KORGER, Michael, BERGSCHNEIDER, Julia, LUTZ, Mirja, MAHLTIG, Boris, FINSTERBUSCH, Karin, RABE, Maike. Possible applications of 3D printing technology on textile substrates. *IOP Conference Series: Materials Science and Engineering*, 2016, **141**(1), doi: 10.1088/1757-899X/141/1/012011.
14. WORTMANN, Martin, FRESE, Natalie, HES, Lubos, GÖLZHÄUSER, Armin, MORITZER, Elmar, EHRMANN, Andrea. Improved abrasion resistance of textile fabrics due to polymer coatings. *Journal of Industrial Textiles*, 2018, online first, doi: 10.1177%2F1528083718792655.
15. VOYUTSKII, S. S., VAKULA, V. L. The role of diffusion phenomena in polymer-to-polymer adhesion. *Journal of Applied Polymer Science*, 1963, **7**(2), 475–491, doi: 10.1002/app.1963.070070207.