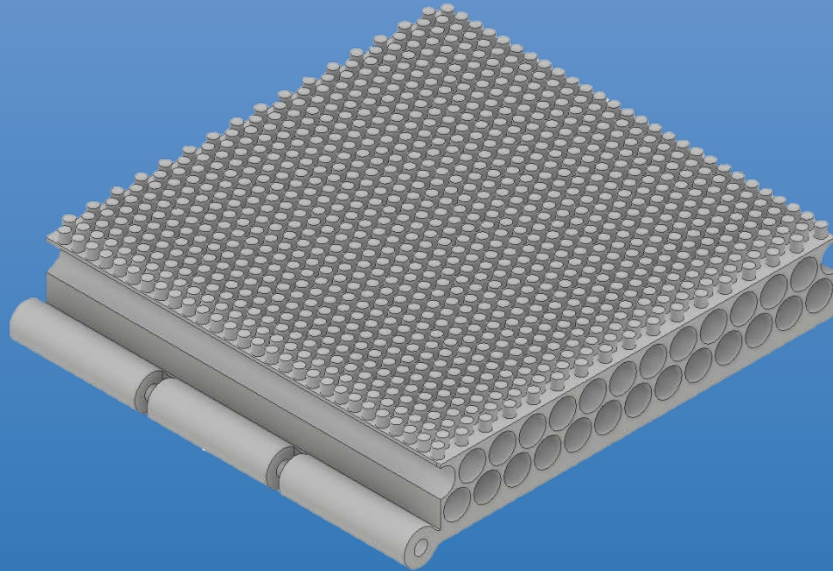


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3D printed bio-inspired adhesives, friction force generation by modulating stiffness.



3D printed bio-inspired adhesives, friction force generation by modulating stiffness.

By

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Preface

After obtaining my bachelor's degree at the Hogeschool Rotterdam of applied sciences, I wanted to obtain more knowledge and decided to apply for a master at the delft university of technology. In the years after, I learned a lot during the bridging program and the master courses, not only theoretically but I also grew personally. It felt that I have learned to learn, analyse and understand instead of just applying the skills and theory. During my master thesis about 3d printed bio-inspired adhesives I had to use all these newly learned skills and push myself. And for me the results have been satisfying.

There are several people that have helped me immensely to achieve this result and finish my thesis project. Firstly, I would like to thank Dimitra Dodou, who has been my supervisor during this project. She has dedicated many hours discussing the progress and direction of the project. Also, I would like to thank Peter van Assenbergh for helping me design the tests and test setup, and for guiding me during the project. Without their help I would not have achieved this result. Furthermore, I thank Tessa Essers with the help and tips she gave on printing the adhesive samples. Finally, I would like to thank Ivan Buijnsters for his interest in the project and for being on my committee.

During my masters I have received a lot of support from my girlfriend and family, for which I am great full. They, and the friends I made during my masters, really supported me in going the extra mile and obtaining the degree. Lastly, I would like to thank Anouk Vinken for proof-reading my thesis.

*F.M. Haring
Delft, Januari 2020*

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Abstract

For over a decade the use of bio inspired adhesives have been explored to achieve high reversible attachment on a wide range of surfaces. Based on the fibrillar adhesive toepads of geckos, many patterned adhesives have been produced that are able to form and preserve contact with rigid and smooth substrates. But on other types of surfaces, such as soft and deformable substrates, the performance of these micro patterned adhesives diminishes. Alternative methods have to be investigated to create adhesives that optimize the ability to form and preserve contact on these types of substrates.

In this study, 3D printing is used to produce a series of adhesives with internal cylindrical porous structures of various geometry. The internal structure modulates the adhesives stiffness in the normal and shearing direction, designed to affect the friction force. Friction forces are measured on glass, soft elastomeric substrates and cloth. On glass, the highest friction forces are generated by the adhesive with the lowest normal stiffness, unaffected by their shear stiffness. On soft substrates, the highest friction forces are achieved with adhesives that combine a low normal stiffness, promoting contact formation, with a high shear stiffness, enhancing contact preservation. The effect of this anisotropic stiffness becomes more pronounced when the stiffness of the substrate decreases, contributing to contact formation. On cloth, generation of friction forces remains challenging, more research is required to create adhesives that excel on these kinds of substrates.

1. Introduction

Non-permanent attachment to substrates is useful in a range of applications, such as performing pick-and-place tasks by robotic devices, climbing robots, and fastening products on tilted or vertical substrates. For several years, researchers have been developing such adhesives, using nature as a base of inspiration. Especially the gecko has been an animal of interest because it is one of the largest animals that has evolved an adhesive system.¹ It has been found that molecular Van der Waals interactions are the main contributor to adhesion of the gecko's system.²⁻⁴

In order to generate enough force with Van der Waals forces, sufficient contact area must be acquired. The gecko achieves this with a fibrillar hierarchical structure. On its toepads there are rows of lamella, or scansorial plates, each lamellae contain rows of millions of setae that further branch in nanoscale spatulae (Figure 1).⁵⁻⁷ While the lamella and setae are made from a stiff β -keratin, the adhesive system itself has a very low effective stiffness due to their high aspect ratio.⁸ Making it able to conform to surface roughness at different length scales, ensuring sufficient contact area between the toepad and the surface (Figure 2).^{5,6}

Furthermore, the stiff β -keratin material in combination with the geometry of the system, gives the adhesive system a high directional in-plane stiffness. The high in-plane stiffness promotes the distribution of friction forces over the established contact area, preventing (shear) stress concentrations and crack initiation. In addition, by splitting up the contact area into a large number of individual contact points, crack propagation is prevented.^{4,9,10}

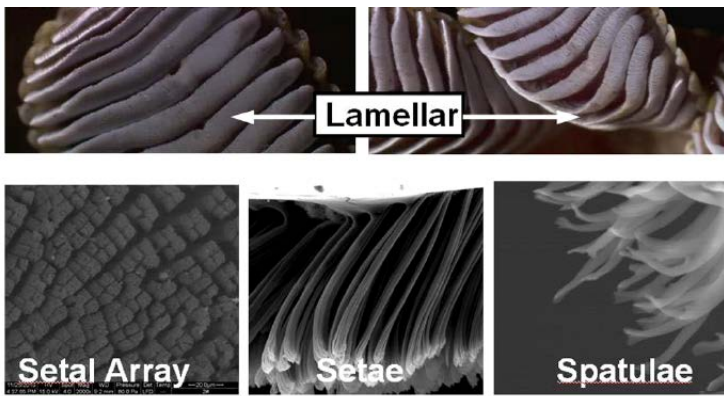


Figure 1: Hierarchical structure of the Gecko (Tian et al. 2013)



Figure 2: Lamella adapting to large scale roughness. (Gillies et al. 2014)

Bartlett 2012 derived a simple framework based on an energy balance to design dry adhesives that incorporate the ability to conform to surface roughness while having a high in-plane stiffness, also called the ability to drape. The framework dictates that there is a maximum sustainable force (F_c) an adhesive bond can withstand before the bond becomes unstable and breaks:

$$F_c \sim \sqrt{G \left(\frac{A}{C} \right)} \quad (1)$$

This force scales with the critical strain energy release rate (G), equal to the work of adhesion, and signifies the release rate at which cracks start to form and propagate without additional energy input. A , is the contact area formed between the adhesive and the substrate. In order to establish a large contact area, thereby establishing strong adhesion, an adhesive system must be able to conform to substrate roughness, which is governed by the systems stiffness in the normal direction.¹¹ In order to achieve a high sustainable force, the established bonds must be maintained and loaded equally. Meaning that, in order to prevent stress concentrations, a low compliance (C) in the direction of loading is required.

When designing an adhesive for a multitude of substrates, the critical energy strain rate becomes an uncontrollable parameter since it is set by the materials of the contact interface. Therefore, the sustainable force is controlled by the A/C ratio. This means that, for adhesives designed to enhance friction, a large contact area must be combined with a low compliance in the shearing direction. Bartlett achieved this set of requirements without making use of a fibril-like structure by producing an adhesive pad out of a fabric combined with a thin elastomeric adhesive layer. The fabric had the ability to drape, conforming at larger length scales while providing stiffness in the direction of loading, and the soft elastomer layer maximizes the contact area at smaller length scales.^{11–14}

While the aforementioned framework does not rely on a specific fibril-like structure, many gecko-inspired synthetic adhesives have been developed with a fibril-like surface micropattern.^{10,15–22} On hard substrates, these micro patterns enhance the synthetic adhesives low effective stiffness and improve the adaptability to the substrates roughness, leading to better contact formation compared to unpatterned adhesives.^{9,20,23–25} The micropatterns also preserve the established contact by splitting up the contact surface, which prevents crack propagation, and improve the stress distribution, achieving a firm and reversible adhesion.^{9,17,20,25,26}

On soft substrates, the improved performance of patterned adhesives over unpatterned ones diminishes. Due to the deformability of a soft substrate, the detachment of the individual fibrils is no longer uncoupled, meaning that crack propagation is no longer prevented.²⁷ Patterned adhesives might even increase the risk of cracks forming on soft substrates, because splitting up the contact leads to a non-uniform stress distribution, with peak forces at the contact edges, at the interface between the adhesive and substrate.²⁸ The adhesive capabilities of patterned dry adhesives on soft substrates are mostly related to the low effective stiffness of the adhesive, which can also be achieved by lowering the elastic modulus of the material itself.^{27,29}

Flat dry adhesives have also been developed by adding a terminal layer on top of the microscale structure. On hard substrates, adhesives with these internal structures have been shown to enhance adhesion by lowering the effective stiffness, but also by preventing crack propagation.³⁰ In addition, adhesives with an internal structure produce higher friction forces compared to micro pillared adhesives.³¹ The increase in friction force might be related to energy losses due to elastic deformations, reattachment of the terminal layer during sliding, and sliding of the internal structure along each other.³¹ However, the increased friction force could be associated to the anisotropic stiffness of the adhesive, and a result of contact preservation due to the high shear stiffness of the internal structure.^{31–33}

In this study, we investigated if anisotropic stiffness, created by a combination of an internal structure and surface structure, could be used to enhance friction forces for single material adhesives. The adhesives were 3D printed and contained a macroscopic internal structure of horizontal cylindrical pores to control the shear and normal stiffness. The surface structure was of similar length scale and was used to improve contact formation. The friction forces were measured on a rigid glass substrate, three poly-dimethoxysiloxane (PDMS) substrates, polyvinyl alcohol (PVA) hydrogel substrates and a plain weave cotton cloth substrate.

2. Methods

2.1. Fabrication of the adhesive structures

20x20 mm² adhesive samples with various inner and surface structures were fabricated. All samples had a solid back layer of 0.4 mm. The minimal wall width between the pores, as well as the minimal thickness of the terminal layer, was 0.15 mm for all structures. To be able to attach the pulling wire for the frictional force measurements, a tube with an inner diameter of 0.7 mm and a wall thickness of 0.5 mm was printed to one side of the back later over the full width of the adhesives. The tube was intersected by two 0.6 mm slits located at one third and two third of the width, where the pulling wire could be attached with the use of a steel rod running through the tube.

Four types of internal structures were printed: The first structure consisted of a single row of 2.7 mm diameter cylindrical pores, positioned perpendicular to the pulling direction. The second structure consisted of the same 2.7 mm diameter cylindrical pores, but now positioned parallel to the pulling direction. The third structure contained two rows of 1.275 mm diameter cylindrical pores, positioned parallel to the pulling direction. A fourth structure was a reference sample without any pores. Each of the 4 internal structures were fabricated either without surface structure, or with a pattern of 0.5 mm diameter circular pillars, or with a pattern of conical 45° tilted pillars with a diameter of 0.5 mm at the base and 0.2 mm at the top. The height of both structures was 0.5 mm. In total we fabricated 12 structures (4 internal structures x 3 surface structures). Each of these 12 structures were printed twice, resulting in a total of 24 samples. 3D files of the adhesive samples can be found in appendix E.

The samples were printed on a Form2 printer (Formlabs, Somerville, MA, USA) using a flexible rubber like resin (Flexible V2 (FLFLGR02), Formlabs, Somerville, MA, USA) with the suppliers recommended print settings and a layer thickness of 0.05 mm. The uncured resin was removed from the samples with the use of isopropyl alcohol (IPA) and compressed air (EW5601, Ewent, Geleen, Netherlands). After cleaning, the samples underwent a full post-curing in a Form cure (Formlabs, Somerville, MA, USA) using the suppliers recommended settings.

2.2. Mechanical characterization

To mechanically characterize the adhesive samples, we performed two tests. An indentation test was performed to determine the effective stiffness in the normal direction, and a single lap shear test was performed to determine the effective stiffness in the shearing direction. In addition, to gain insight in the shear stress distribution of the internal structures, a Finite Element Analysis (FEA) was performed.

2.2.1. Indentation tests

Indentation tests were performed with an Aerotech ACT115 linear motion stage, controlled with an Aerotech Soloist CP controller and a custom-built GUI in MATLAB R2016a. A FUTEK S-Beam load cell with a 10 lb. capacity was mounted on the motion stage to record the forces. The load cell signal was amplified with a CPJ2S signal conditioner. Adhesives were compressed using a spherical glass lens indenter (radius of curvature = 34.5 mm) (Thorlabs, Newton, NJ, USA). Adhesives were indented over a depth of 1.5 mm (0.5 mm for the adhesives with an internal reference structure), and forces and displacement were recorded at a frequency of 200 Hz. For the indentation tests, the samples were fixed to a Perspex sample holder with double-sided tape. Recorded data of the indentation tests and Matlab script used to analyze the data can be found in appendix E.

2.2.2. Single-lap shear tests

The effective shear stiffness of the adhesives was characterized with a single-lap shear test, performed with a LLOYD testing machine (LR5K, AMETEC Inc., Berwyn, PA, USA), with a 5 KN S-Beam force sensor. The adhesives were attached to a custom-made sample holder with the use of two metal connection strips glued (Loctite universal 60sec, Henkel AG & Co, Dusseldorf, Germany) to the bottom and top sides of the adhesives (Figure 3). The adhesives were pulled apart with a speed of 1 mm s^{-1} until they were completely torn apart (20 mm). Recorded data of the single-lap shear tests and Matlab script used to analyze the data can be found in appendix E

2.2.3. FEA of adhesives

FEA was performed to gain insight in the deformation and stress distribution of the internal structures. A nonlinear static analysis was performed with Inventor Nastran2020 (Autodesk Inc., San Rafael, CA, USA). The material model was built using the mechanical properties provided by Formlabs (appendix F). The elastic modulus was derived from the shore hardness using the relations from Mix and Giacomini.³⁴ A mesh was generated using the default linear elements, with an element size of 0.15 mm. In addition, the central axis of the tube was fixed for any vertical displacement, ensuring a pure horizontal deformation under the applied load. A bearing-load with a magnitude of 6 N was applied at the inside of the tube in the direction of pulling.

2.3. Frictional force measurements

Friction experiments were performed using a custom-made positioning stage (ACT115, Aerotech inc, Pittsburg, PA, USA) with a 9 N load cell (LSB200, FUTEK, Irvine, CA, USA), controlled with a GUI build in MATLAB R2016a (Mathworks, Natick, MA, USA). The signal from the load cell was amplified and converted with a CPJ2S signal conditioner. Samples were connected to the load cell via a HPPE Dyneema braided wire (BBVFS25, Berkley, Columbia, SC, USA). Before testing, samples were placed and aligned by applying a small pre-tension in the direction of pulling. This was done with a small weight hooked to the back of the sample (Figure 4).

Samples underwent a 2.45 N (250 gr) normal preload for 10 seconds, and a 0.49 N (50 gr) normal load during sliding. The sliding velocity was 1 mm s^{-1} , for a duration of 5 seconds. The friction force measurements were performed on six substrates: glass, plain cotton cloth, PVA hydrogel and three PDMS substrates of various stiffness degrees. For each substrate, the 24 samples were measured 5 times each, in a randomized order, resulting in 10 measurements for each sample type. The substrates were cleaned with ethanol after each 10 measurements. The PDMS were replaced after each 20 measurements. The cloth and PVA hydrogel substrates were not cleaned with ethanol but replaced every 10 measurements instead. The substrates were fixed to a substrate holder that was placed on a LabJack and aligned with the pulley (appendix D). The glass substrate was placed on the holder with the use of double-sided tape. The cloth was fixed to the glass using eight paper clamps. The PDMS and PVA substrates were fixed to the substrate holder using eight m3 bolts and a fixating plate (Figure 4, appendix E). Recorded data and Matlab script used to analyze the data can be found in appendix E.



Figure 3: Single lap shear test setup used to measure the shear stiffness of the printed adhesive structures.

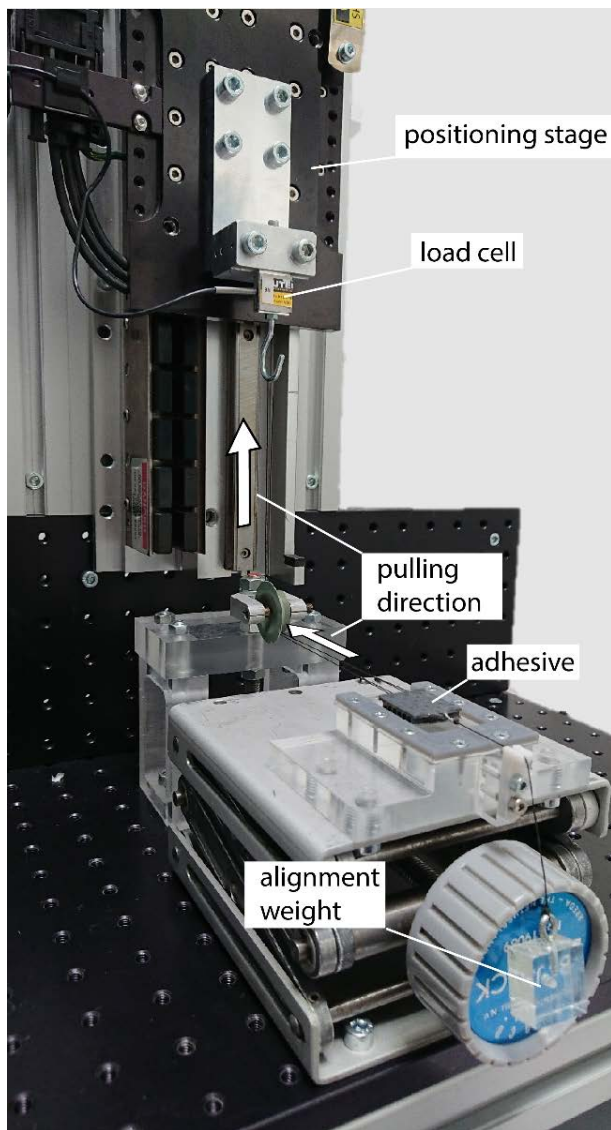


Figure 4 Upper right: the test setup used to test the adhesives on PVA substrates; the sample is aligned with the use of a small weight. Upper left: the test setup equipped with the glass slide and a piece of cloth clamped to the glass with the use of paper-clips; the alignment weight is removed from the sample and the normal preload is applied. Bottom: friction test on PVA.

2.4. Substrate preparation

The glass substrate consisted of a glass slide, which was used as received.

The cloth substrates were cut out of a plain weave sheath, with the weave of the samples being kept at 0° and 90° degrees (parallel and orthogonal to the direction of pulling).

Poly-dimethoxysiloxane (PDMS, Sylgard 184, Dow Corning, Midland, MI, USA) substrates were prepared by mixing pre-polymer and curing agent mixed in 10:1, 20:1 and 30:1 weight-based ratios, obtaining substrates of 2200 kPa, 700 kPa and 300 kPa, respectively. The pre-polymer - curing agent mixtures were degassed, in measured quantities poured in 140 mm petri dishes, degassed again and finally cured for at least 2h at 70°C to obtain substrates of 2 mm in thickness.

The polyvinyl alcohol (PVA) hydrogel was prepared by dissolving PVA in deionized water at 90°C in 1:10 weight-based ratios. The solution was poured in 140 mm petri dishes and cured in three cycles of freezing for at least 6 h at -20°C and thawing for 6 h at 7°C. Before testing, the PVA substrates were taken out of the freezer to warm to room temperature for a minimum of 30 minutes.

2.5. Statistical analysis

2.5.1. Indentation tests

The effective elastic modulus of the adhesive samples was determined by fitting the Hertz model to the force indentation data. The force and position data were processed with MATLAB 2018a. Prior to the analysis, the data were filtered with a zero-phase weighted moving average filter function with a window of 21 samples to reduce noise. Then, the point of contact between the indenter and sample was determined and indentation was calculated. In order to take into account, the initial phase of contact, a fitting parameter δ_{off} was included to the indentation δ , such that the fit is done on the loading portion of the indentation curve.³⁵

$$F = \frac{4}{3} E_{eff} * R^{0.5} * (\delta - \delta_{off})^{\frac{3}{2}} \quad (2)$$

2.5.2. Single lap shear tests

Extension and force data were processed with MATLAB 2018a. The stiffness was calculated between each sample point and the maximum stiffness increment was determined and used to compare the samples.

2.5.3. Friction force measurements

All the recorded data was processed with MATLAB 2018b. Prior to analysis, the recorded force and position data was filtered with a zero-phase unweighted moving average filter with a window of 21 samples (41 for the PVA hydrogel data), to reduce the noise in the signal. The data from the first 0.5 seconds was used to determine the initial position and to remove any static offset (weight of the cord) from the force measurements. From the recorded force-time data, the peak force corresponding to the moment when the sample started sliding was used in the analysis.

Several analysis of variance (ANOVA) with a post-hoc Turkey-Kramer test were conducted to test the effect of the variable and their interaction on friction. One analysis compared the effect of the internal structures with respect to substrates with decreasing stiffness (glass, PDMS 1:10, PDMS 1:20, PDMS 1:30). A second analysis compared the effect of the surface structures with substrates with decreasing stiffness. Three separate analysis were performed comparing the internal and surface structures on glass, PVA and cloth.

3. Results

3.1. Fabrication of the adhesive structures

Figure 5 shows the four internal structures and three surface structures that were fabricated. From now on we will refer to the internal structure of the adhesive samples by a reference code, from I1 to I4. I1 has an internal structure containing 2.7 mm pores perpendicular to the pulling direction. I2 has pores of the same diameter but the pores are aligned with the pulling direction. I3 has two rows of pores with a diameter of 1.275 mm perpendicular to the direction of pulling and I4 was a reference sample without any structure. Three different surface structures were printed upon the internal structures: a pillared structure of cylindrical pillars 0.5 mm in diameter and height, a hooked structure containing conical 0.5 mm high pillars tilted 45°, with a base diameter of 0.5 mm and top diameter of 0.2 mm and a flat reference structure.

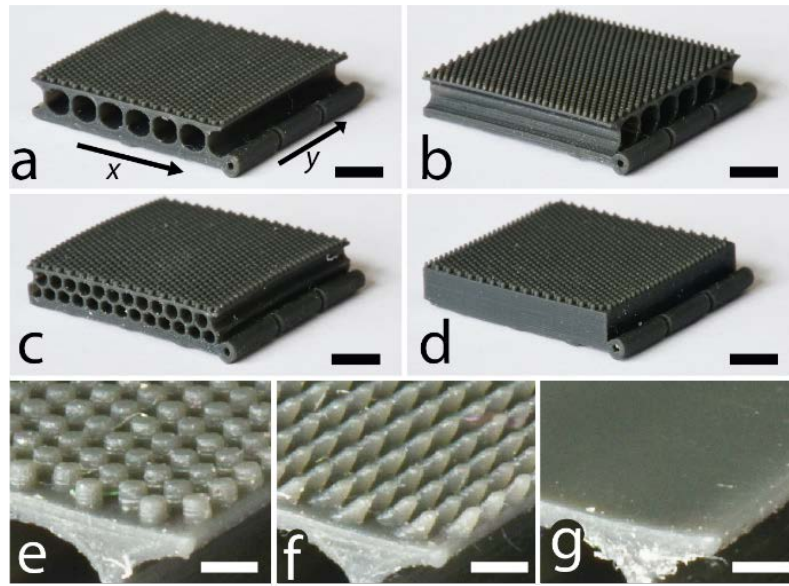


Figure 5 Photographs of the 3D-printed adhesives. During the force measurements, the samples were pulled in the x-direction. a) Internal structure I1, with 2.7 mm pores perpendicular to the pulling direction (y-direction). b) Internal structure I2, with 2.7 mm pores aligned in the x-direction. c) Internal structure I3, with two rows of 1.275 mm pores in the x-direction. d) Reference sample without internal structure (I4). e) A surface structure of cylindrical pillars with a diameter of 0.5 mm and height of 0.5 mm. f) Surface structure of conical, 0.5 mm high, pillars tilted 45° with a diameter of 0.5 mm at the base and 0.2 mm at the top. g) No surface structures. Scale bars in a-d are 5 mm and 1 mm in e-g.

3.2. Mechanical characterization.

3.2.1. Indentation tests

For the indentation tests, we considered the normal stiffness of I1 and I2 to be the same since these samples have identical internal structures. From the Hertz fit, we derived the effective stiffness (Figure 6). We used a one-way ANOVA to compare the stiffness between the internal structures and found a significant main effect, $F(2,10) = 733.86$, $p < 0.001$. Post-hoc analysis showed that effective stiffness was not significantly different between I1 (and I2) and I3 ($p = 0.182$) and that I3 had the lowest effective stiffness 0.25 ± 0.03 n/mm² followed by I1 (and I2) with an effective stiffness of 0.72 ± 0.16 n/mm². The solid reference sample I4 was significantly stiffer (9.99 ± 0.80 n/mm², $p < 0.001$), compared to the adhesives with a porous internal structure.

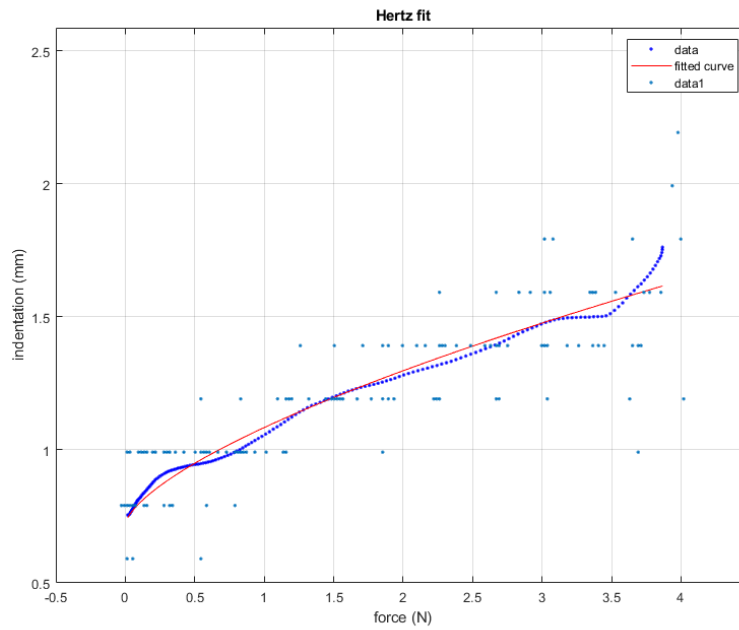


Figure 6: A fit (red) of the Hertz model on the filtered data (dark blue) of the force indentation curve.

3.2.2. Single-lap shear tests

For each shear test we calculated the incremental stiffness between each sample point and used the maximum value to compare the internal structures (Figure 7). A one-way ANOVA showed a significant difference in shear stiffness for the adhesives with different internal structures, $F(2,12) = 12.23$, $p = 0.001$. Post-hoc analysis showed that I1 had the lowest average maximum stiffness of $46.14 \pm 3.43 \text{ n/mm}^2$, followed by I3 with an average maximum stiffness of $51.95 \pm 6.02 \text{ n/mm}^2$. I2 had the highest measured stiffness, $69.32 \pm 8.97 \text{ n/mm}^2$. We were unable to record a reliable effective shear stiffness for the reference samples I4, since these samples were so stiff that tearing occurred on the layer of glue between the sample and the connection strips. The post-hoc analysis showed that the stiffness of I2 was significantly higher compared to I1 and I3 ($p < 0.001$ and $p = 0.008$, respectively). The stiffness of I1 and I3 did not differ significantly ($p = 0.322$).

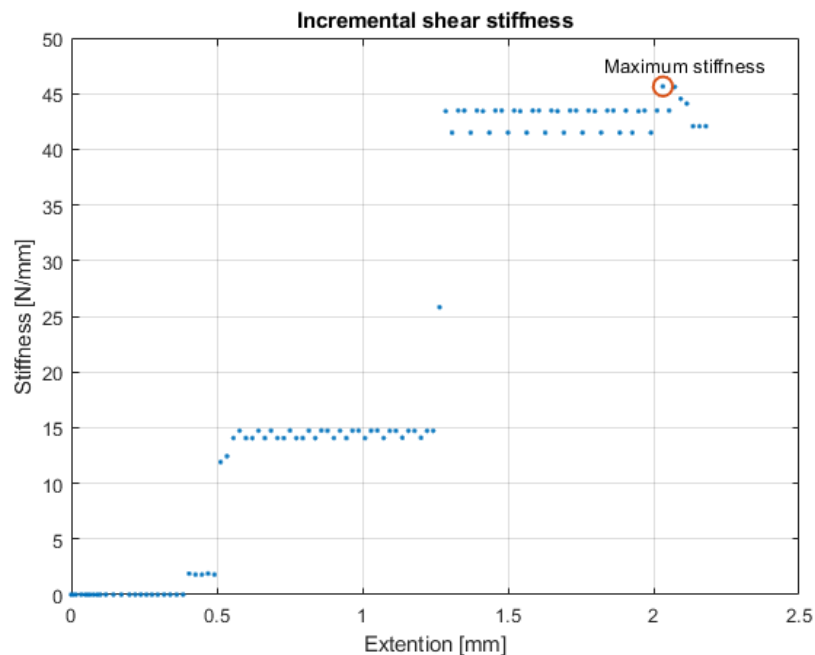


Figure 7: Calculated incremental shear stiffness. Only the increments up to the maximum force are displayed. As expected, the stiffness increased up to a maximum during loading.

3.2.3. FEA and empirical data

Figure 8 shows the results of the FEA for the four internal structures. The photographs show, from top to bottom, the adhesives without any load, under a normal load of 6N and a shear load of 6N. In addition, the structural stress under an applied shear load of 6N are shown, predicted by the FEA.

I4 (Figure 8a): High stresses only occurred at the point where the shear load was applied. The FEA predicted a deformation of 0.075 mm. The empirical test also did not detect any deformation under either shear or normal load.

I1 (Figure 8b): FEA showed high stresses at the first rows of internal walls, whereas stresses in the terminal layer were a third of the stresses in the internal walls. FEA further showed that, under a shear load, this structure exhibited the largest shear deformation (0.332 mm). This is in line with the empirical data, where I1 exhibited the largest shear deformations under a similar shear load. Derived from Figure 8, with the use of a pixel count, the lateral deformation was around 1.5 mm. The indentation depth was around 1.5 mm.

I2 (Figure 8c): The FEA shows similar stress concentrations in the internal walls, compared to I1, at the point where shear forces are introduced to the adhesive sample. However, since the pores are aligned with the direction of loading, the stresses are distributed to all the pore walls equally, instead of a few as is the case for the I1 structure. This leads to a better stress distribution at the terminal layer. The FEA predicted a limited shear deformation of 0.171 mm. The corresponding empirical data confirmed the prediction and showed a lateral deformation of less than 0.1 mm. The indentation depth of this structure was the same as I1, around 1.5 mm.

I3 (Figure 8d): Stresses in the walls of the internal structure were high in the first few rows but decreased gradually, with the rows at the end of the sample showing no stress in the walls of the structure. The FEA predicted a shear deformation of 0.166 mm. The empirical test showed a deformation in the shear direction of about 0.8 mm and an indentation depth of 0.45 mm.

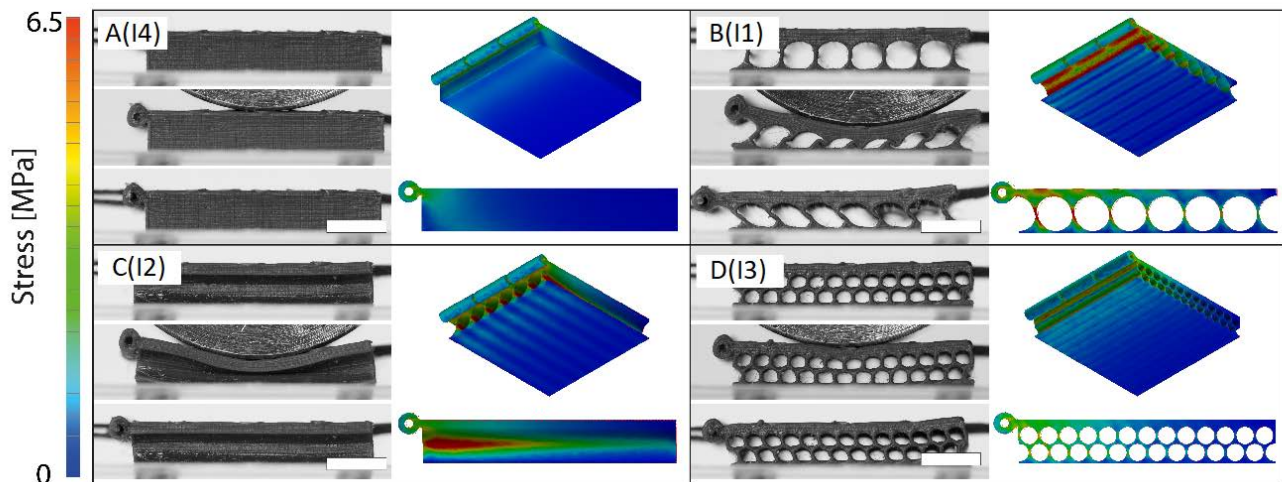


Figure 8: The deformation of the four internal structures under no load, normal load and shear load (from top to bottom). The stresses determined by the finite element analysis are shown in bottom and side view representations. Scale bars are 5 mm.

Table 1: Results of the mechanical characterization. The effective shear stiffness of I4 could not be determined since the samples were too stiff and tearing occurred in the glue layer.

Internal structure	Indentation depth empirical. mm	Effective normal stiffness n/mm ²	Effective shear stiffness n/mm ²	Lateral deformation predicted by FEA: mm	Lateral deformation empirical. mm
I1	1.5	0.72±0.16	46.14±3.43	0.33	1.5
I2	1.5	0.72±0.16	69.32±6.02	0.17	<0.1
I3	0.45	0.25±0.03	51.95±8.97	0.17	0.8
I4	<0.1	9.99±0.80	*	0.08	<0.1

3.3. Frictional force measurements

3.3.1. Effect of the internal structures on substrates with decreasing stiffness

A two-way ANOVA comparing the performance of adhesives without surface structures for substrates with varying stiffness (glass, PDMS 1:10, PDMS 1:20 and PDMS 1:30) and internal structures (I1 to I4) showed significant main effects for both the substrate, $F(3,144) = 352.69$, $p < 0.001$, and the internal geometry, $F(3,144) = 35.17$, $p < 0.001$, as well as for the interaction between substrate and internal geometry, $F(9,144) = 7.71$, $p < 0.001$.

Internal structures on glass

Post-hoc analysis showed that within the internal structure group on glass, the adhesives with a single-row pore structure, I1 and I2, had the highest friction force ($F_{I1} = 2.83 \pm 0.47$ N, $F_{I2} = 2.84 \pm 0.26$ N) and performed significantly better than the double-row pore structure I3 ($F_{I3} = 1.95 \pm 0.17$ N, $p < 0.001$) and the reference structure I4 ($F_{I4} = 1.61 \pm 0.22$ N, $p < 0.001$) (Figure 9).

Internal structures on PDMS 1:10

Post-hoc analysis showed that, on PDMS 1:10 substrates, there was no significant difference in friction force within the internal structure group. On average, adhesives with the reference structure I4 generated the highest forces ($F_{I4} = 2.55 \pm 0.26$ N), followed by I2 ($F_{I2} = 2.43 \pm 0.59$ N), I1 ($F_{I1} = 2.01 \pm 0.48$ N) and adhesives with a I3 structure generated the lowest forces ($F_{I3} = 1.90 \pm 0.29$ N).

Internal structures on PDMS 1:20

On PDMS 1:20, post-hoc analysis showed that, the highest friction forces were generated by I1 and I2 ($F_{I1} = 4.02 \pm 1.39$ N, $F_{I2} = 4.72 \pm 1.13$ N), followed by the adhesives with I3 ($F_{I3} = 3.49 \pm 1.14$ N). The reference structure I4 exhibited the lowest friction force ($F_{I4} = 3.16 \pm 0.70$ N). On this substrate, only the difference between I2 compared to I3 and I4 reached statistical significance ($p = 0.008$, $p < 0.001$, respectively).

Internal structures on PDMS 1:30

On the softest substrate tested in this series, the PDMS 1:30, post-hoc analysis showed that, I1 and I2 generated the highest friction forces ($F_{I1} = 7.31 \pm 0.44$ N, $F_{I2} = 8.16 \pm 0.34$ N). Adhesives with the double-row structure, I3, generated significantly lower friction than the single-row structures, I1 and I2 ($F_{I3} = 6.20 \pm 0.74$ N, $p = 0.032$, $p < 0.001$ respectively). The reference I4 generated the lowest friction forces among the four internal structures ($F_{I4} = 4.99 \pm 0.91$ N, $p \leq 0.001$).

Effect of the substrate stiffness

Post-hoc analysis showed no significant difference in friction force between the two stiffest substrates, glass and PDMS 1:10. Adhesives with a porous internal structure (I1, I2 and I3) showed a significant increase in friction force with a decrease in PDMS substrate stiffness

($p < 0.001$ in all cases). Adhesives with the reference structure (I4) showed an increase in friction force on the PDMS 1:30 substrates only, compared to PDMS 1:10 and PDMS 1:20 ($p < 0.001$ for both), no statistical difference was found between PDMS 1:10 and PDMS 1:20.

I1 and I2 showed the largest increase in friction force as a function of substrate stiffness, followed by I3. The friction force of the reference sample I4 was the least affected by the substrate stiffness (Figure 9).

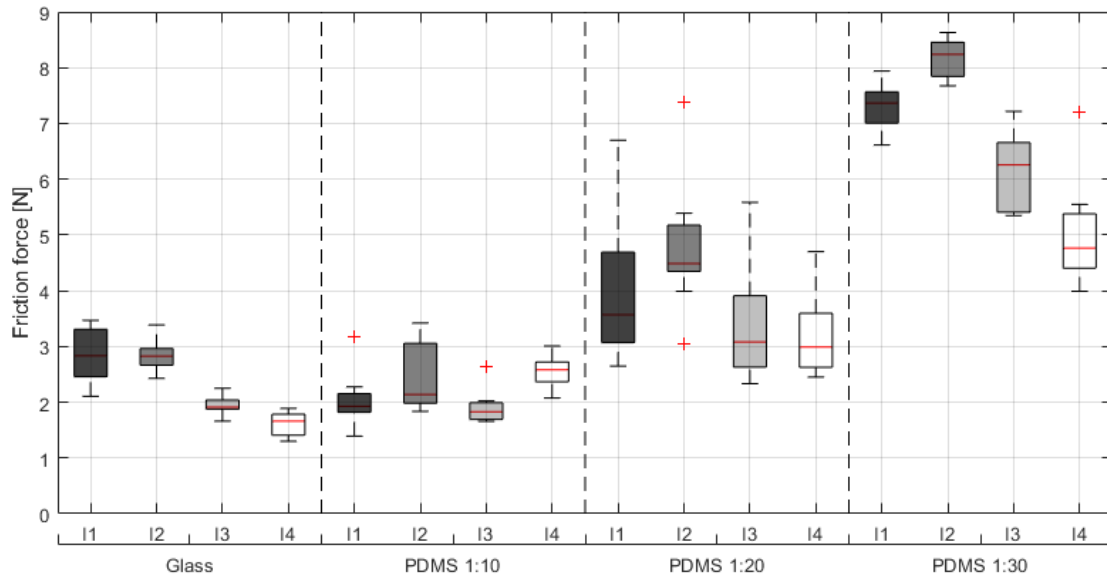


Figure 9: Friction force of samples with various internal structures and without surface structure on substrates with decreasing stiffness. Figures of the friction force measurements of adhesives with surface structures can be found in appendix A.

3.3.2. Effect of the surface structures on substrates with decreasing stiffness

A two-way ANOVA comparing the effect of the surface structures (flat, pillared and hooked) with respect to the different substrates (glass, PDMS 1:10, PDMS 1:20 and PDMS 1:30) showed significant main effects for the surface structures, $F(2,108) = 302.09$, $p < 0.001$ and substrates, $F(3,108) = 110.66$, $p < 0.001$, as well as a significant effect for the interaction, $F(6,108) = 25.9$, $p < 0.001$.

A post-hoc analysis showed that, the effect of surface structures on friction was similar for all the tested substrates. Specifically, adhesives without surface structure generated significantly higher force compared to the pillared and hooked surface structures (on glass $p = 0.03$, $p = 0.002$ respectively, on all other substrates $p < 0.001$ for all cases), followed by the adhesives with pillared surface structures. Adhesives with a hooked surface structure generated the least force. The friction force between the pillared and hooked surface structures was significantly different only on PDMS 1:30 ($p < 0.001$).

The adhesives with a flat surface structure showed an increase in friction force with a decrease in substrate stiffness, on all substrates (between PDMS 1:10 and PDMS 1:20 $p = 0.029$, $p < 0.001$ for all other cases). For the pillared adhesives, the friction force was significantly higher only on PDMS 1:30 substrates ($P < 0.001$ in all cases). No difference in friction force was found for the hooked adhesives on different substrates (Figure 10). Friction forces measured for patterned adhesives are presented in appendix B.

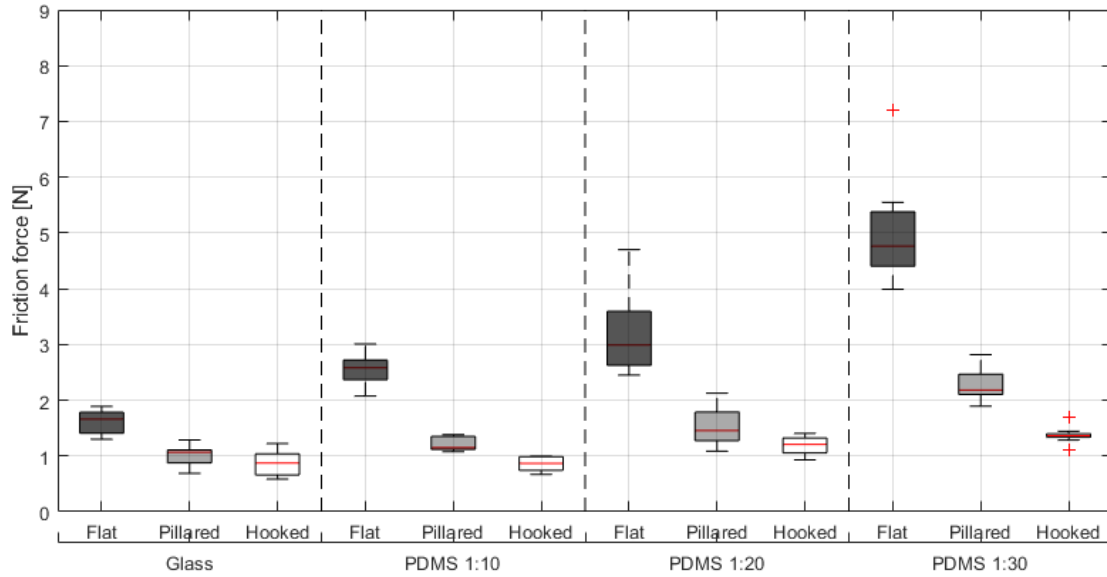


Figure 10: Friction force of adhesives as a function of surface structure on substrates with decreasing stiffness. Only the results of the samples without internal structure are shown.

3.3.3. Performance of adhesives combining the internal and surface structures

On glass, a two-way ANOVA comparing surface structures (flat, pillared, hooked) with internal structures (I1 to I4) showed significant main effects, $F(2,108) = 209.09$, $p < 0.001$ and, $F(3,108) = 62.01$, $p < 0.001$, as well as a significant interaction effect, $F(6,108) = 11.09$, $p < 0.001$.

A post-hoc analysis showed that there was no statistical difference in friction force among the adhesives with the four internal structures and a hooked surface structure. Among the adhesives with pillared surface structures, all three porous internal structures (I1, I2 and I3) generated higher friction forces than the reference structure (I4) ($p < 0.001$ in all cases). Furthermore, I1 generated significantly higher friction forces than I3 ($p = 0.009$). The results of the comparison of the internal structures of the adhesives with a flat surface structure were the same as shown in section 3.3.1.

All flat surface structured adhesives generated higher friction forces compared adhesives with hooked surface structures and corresponding internal structures ($p < 0.001$ in all cases). Only the adhesives with a pillared surface and porous internal structure (I1, I2 and I3) generated higher friction forces compared to their counterparts with hooked surface structures ($p < 0.001$, $p < 0.001$ and $p = 0.002$ respectively). Adhesives with I1, I2 and I4 internal structures and a flat surface structure, outperformed corresponding adhesives with a pillared surface structure ($p < 0.001$ in all cases).

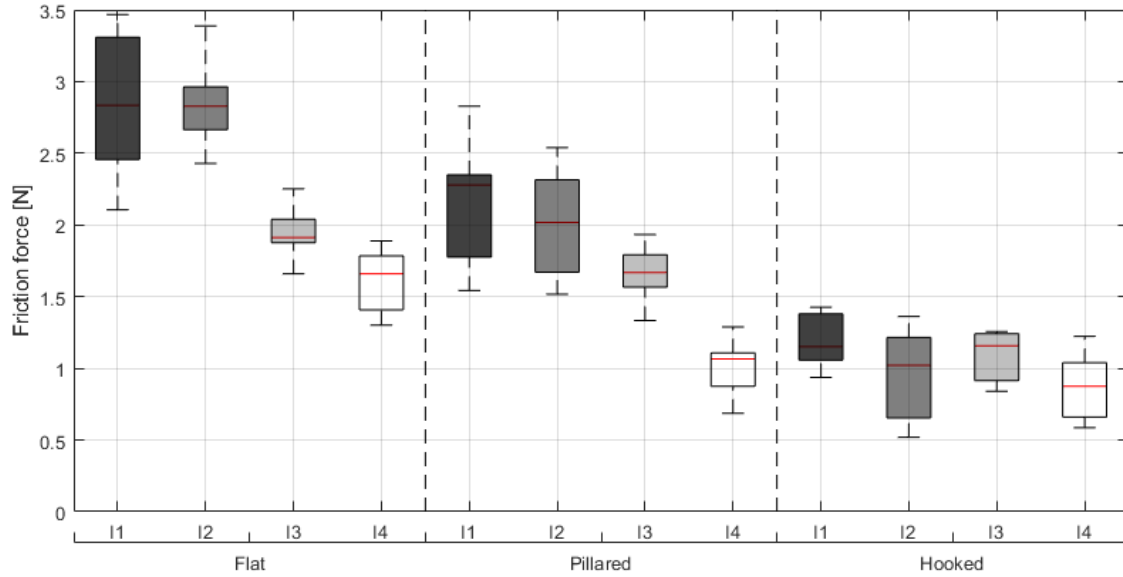


Figure 11: Friction forces of the adhesives on glass.

3.3.4. Performance of structured adhesives on PVA

A two-way ANOVA for surface structures (flat, pillared, hooked) and internal structures (I1 to I4) on PVA showed a significant main effect for the surface structures only, $F(2,108) = 70.97$, $p < 0.001$. The presence of an internal structure did not significantly contribute to the friction force on PVA substrate. Moreover, post-hoc analysis on the significant main effect showed that the presence of a surface structure had a negative impact on the generated friction force ($p < 0.001$). The forces generated on PVA were considerably lower than the lowest forces generated on the softest PDMS substrate tested (Figure 12).

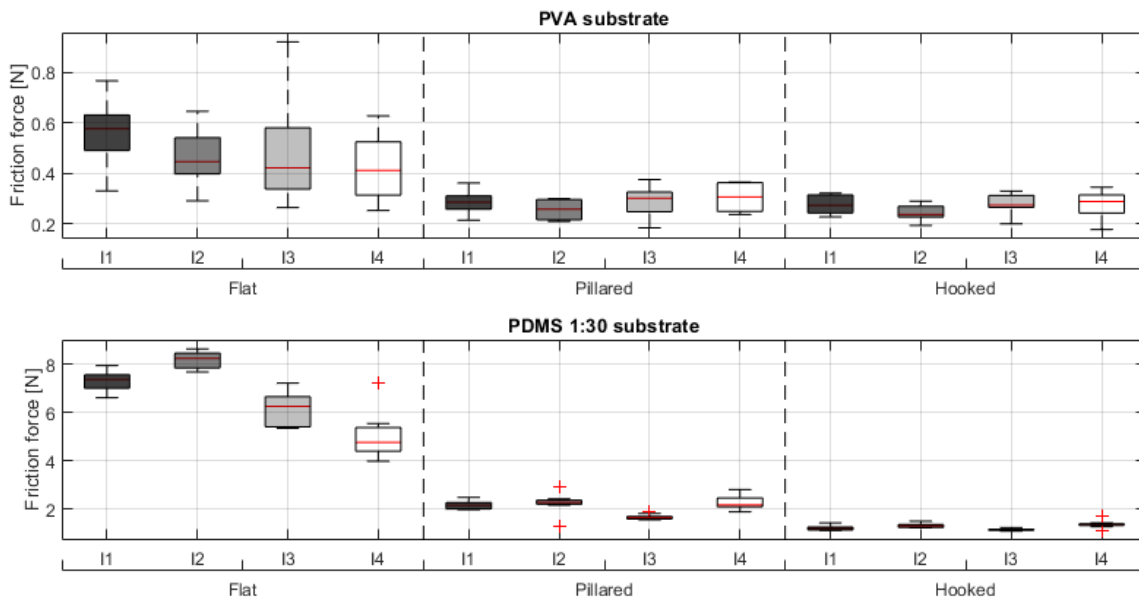


Figure 12: Friction force of the samples on PVA substrate and on PDMS 1:30 substrate.

3.3.5. Performance of structured adhesives on cloth

A two-way ANOVA for surface structures (flat, pillared, hooked) and internal structures (I1 to I4) on cloth showed significant main effects for both the surface structures, $F(2,108) = 3.41$, $p < 0.034$ and the internal structures, $F(3,108) = 35.04$, $p < 0.001$. An interaction effect was also found, $F(6,108) = 2.33$, $p < 0.037$.

Post-hoc analysis showed that only the difference between adhesives with a flat and pillared surface structure and I2 internal geometry was statistically significant ($p = 0.013$). In all other cases a different surface structure did not affect the friction on cloth.

Comparing the internal structure group per external surface structure, showed that for flat surface structures, adhesives with internal structure I2 generated higher friction forces compared to I1 and I3 ($p < 0.001$) and I4 structures performed better than I3 ($p < 0.001$). Adhesives with a pillared surface structure and I3 internal structure generated higher friction forces compared to pillared adhesives with a I2 or I4 internal structure ($p = 0.005$, $p < 0.001$ respectively). Adhesives with a hooked surface structure and I1 or I3 internal structure performed better than adhesives with a I2 or I4 internal structure (I1($p = 0.031$, $p = 0.005$), I3($p = 0.012$, $p = 0.002$), respectively) (Figure 13).

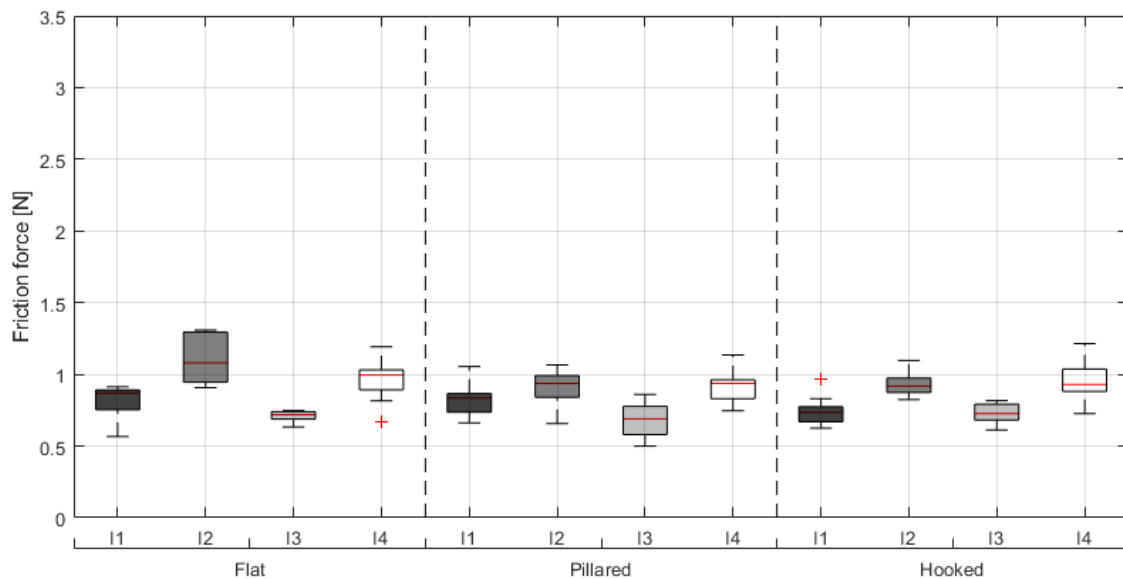


Figure 13: Performance of the adhesive samples on cloth.

4. Conclusion & Discussion

4.1. Fabrication of the adhesive structures

We successfully fabricated adhesives with anisotropic properties out of a single material by changing their internal and external architectures. By printing porous internal structures, using a stereolithographic 3d printing method, we created adhesives with a larger deformability than expected based on the reported resin stiffness. With this method designs are printed in successive layers of 0.05 mm, where each cured layer is peeled from the printing bed. Thus, it is key to reduce peeling forces to prevent printing errors during peeling of each successive layer. Commonly this is done by minimizing the surface area of each layer. However, we encountered several problems by printing our samples this way. To overcome these problems, we maximized the mechanical stability of each layer by adjusting the print orientation and minimizing the height of the print. In appendix C we elaborate on the printing process of the adhesives.

4.2. Mechanical characterization

Under an applied normal load of 6N, we observed that the amount of deformation is related to the porosity of the adhesives. Under normal loading, the pore walls of the single row porous structures showed an S shaped deformation when buckling. Resulting in an additional lateral deformation under normal loading. This observed lateral deformation could explain the unexpectedly high effective stiffness determined by the indentation tests. During these tests lateral deformation of the adhesive was not possible, meaning that the single-row porous adhesives were forced to deform in a different buckling mode, resulting in a higher effective stiffness.

Under shear loading, the adhesives with internal structures I1 and I3 showed the largest deformations. Bending of the pore walls was the main contributor to deformation under shear load. When the walls of the pores were aligned with the direction of loading, which was the case for adhesives with a I2 internal structure, bending of the walls was not possible, resulting in less deformation and thus a higher effective shear stiffness. Compared to I1, I3 has a higher shear stiffness, due to the increase in number of pores, and thus pore walls, as well as a decrease in aspect ratio of pore walls. Adhesives without internal structure, I4, did not exhibit observable deformations under loading. The stiffness of adhesives with this structure are best described by the isotropic tensile strength of the resin determined by the manufacturer, which is 7.7 – 8.5 MPa after post-curing.

4.3. Frictional force measurements

4.3.1. Effect of the internal structures

For adhesives without a surface structure it was found that, on a glass substrate, adhesives with the lowest normal stiffness, which are the adhesives with a single pore row I1 and I2, produced the highest friction forces. The difference in shear stiffness between these two internal structures did not lead to a significant difference in pull off force. Although the performance of adhesives with a higher shear stiffness (I2) was more consistent, making it more reliable compared adhesives with the I1 internal structure.

The increase in friction force for the adhesives without surface structure was mostly related to the amount of contact area formed between the adhesive and the glass substrate. The adhesives with a porous internal structure, and lower normal stiffness, deformed better to the substrate and created more contact area. The contact area that was created consisted out of multiple patches and form a projection of the internal structure. For the single row adhesives, these patches had a larger width, compared to the double row adhesives, indicating a more intimate contact was formed in these cases. In addition, the adhesives with a single-row internal structure preserved more contact area when the preload was removed (a 50gr normal load remained), compared to

adhesives with a double-row internal structure, which showed a large decrease in contact area (Figure 14).

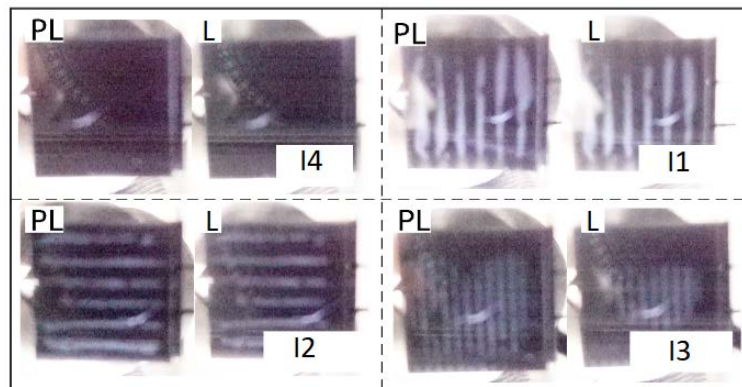


Figure 14: Bottom view of the adhesives without surface structures. The formed contact area can be seen as white patches, and form a projection of the internal structures. The two frames of each adhesive show the contact area before and after removing the pre-load.

On the PDMS substrates, the adhesives with different internal structures showed similar contact formation compared to the glass substrate. Adhesives with the lowest normal stiffness maintained the largest contact area on all PDMS substrates.

We found no difference in performance between the glass and PDMS1:10 substrate. Although an increase in friction force was expected due to a larger contact area related to the deformability of the PDMS substrate. We observed that contact area maintained on PDMS 1:10 was slightly higher compared to that of glass (Figure 15). It is likely, that the critical strain energy release rate might be lower on PDMS compared to that off glass, nullifying any benefit that was gained from the increased contact area.

For PDMS substrates, we found that the deformability of the substrate had a positive contribution to the amount of contact area that was formed and maintained. On PDMS 1:10 no effect of the internal structures was observed, for the PDMS 1:20 and PDMS 1:30 substrates, a lower normal stiffness of the adhesive was increasingly beneficial for the established contact area, and thus generated friction force. The highest forces were generated by the adhesives that combined a low normal stiffness with a high shear stiffness. Indicating that a difference in shear stiffness can be beneficial for the amount of friction force that is generated, but only when sufficient contact area is established.

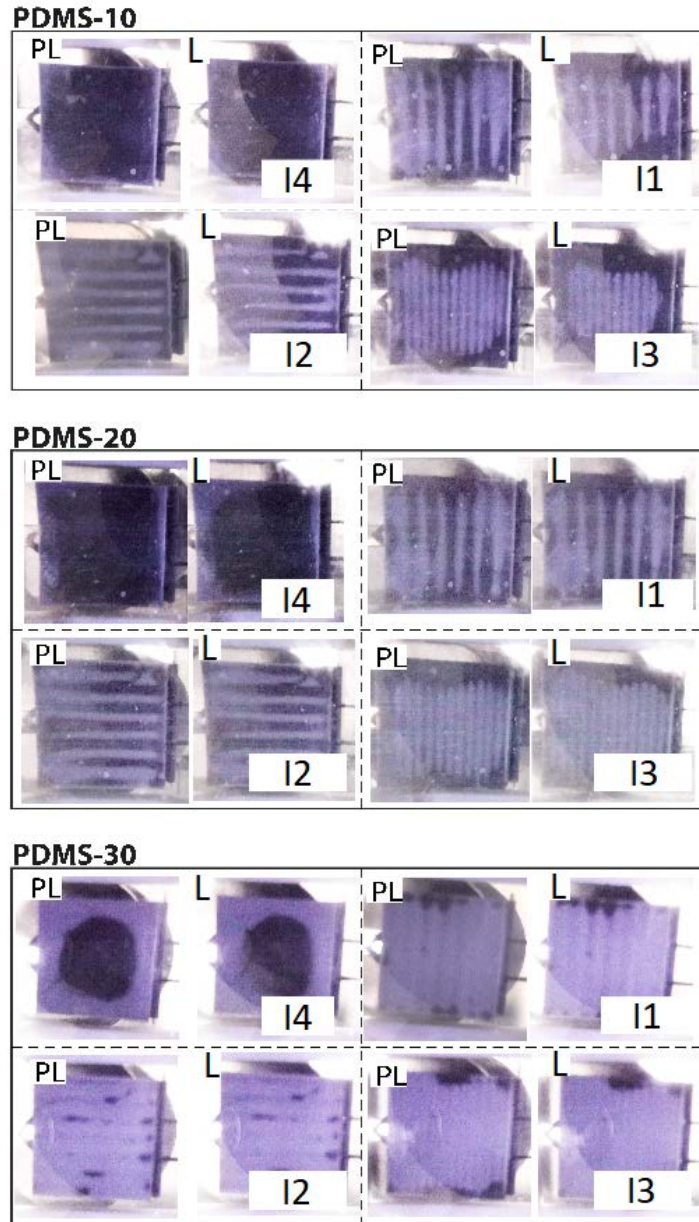


Figure 15: Bottom view of the unpatterned adhesives on the three PDMS substrates. As substrate stiffness decreases, the formed contact area increases.

4.3.2. Effect of the surface structures

Patterning the surface of the adhesives lowered the friction force on glass substrates compared to adhesives without surface structure. Probably because of the limitation with the 3D printing method, used surface structures were not fine enough and the packing density of the pattern was too low to benefit from any contact splitting effects. In addition, the contact area that could be achieved was reduced dramatically compared to adhesives without a surface structure, leading to a low friction force. For the adhesives with a hooked surface structure on glass, the reduction in contact area was so severe that the effect of internal structures was nullified.

On the PDMS substrates, adding a patterned surface structure resulted in lower friction forces compared to adhesives with a flat surface structure, probably due to the reduced contact area. As a result, the effect of the internal structures were also reduced on these substrates.

Although the effect did not lead to an improvement in friction force, we did observe that adhesives with a patterned surface structure had a better distribution of contact over the interface, and presumably less stress concentrations (Figure 16).

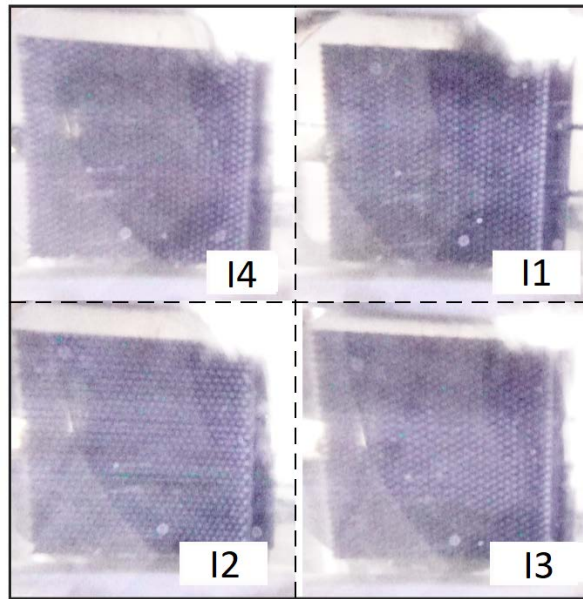


Figure 16: Contact area formed on PDMS 1:20 by adhesives with a pillared surface structure and varying internal structures. Adhesives with the reference internal geometry (I4) show a large contact area at the edge of the adhesive. Adhesives with I1 and I2 structures show longitudinal contact area's aligned with the pore direction. Adhesives with the I3 internal structure show a centered uniformly distributed contact area.

4.3.3. Performance of structured adhesives on PVA

On PVA, we only found an effect for the surface structure. Adhesives with a flat contact surface generated slightly higher friction forces compared to the patterned structures, likely because the latter two structures had a lower contact area due to a low stacking density. Compared to the softest PDMS substrate, the friction forces on PVA were much lower. It is likely that this is caused by water present in the PVA. When the adhesive was pressed on the PVA substrate the water is pushed out of the PVA, making the interface slippery. Since the adhesives with a porous internal geometry almost achieved full contact on PDMS 1:30, one might argue that the decrease in friction force might be caused by the decrease in shear stiffness of the system. Because when full contact is realized, lowering the stiffness further does only increase the systems compliance thereby decreasing the friction force that is generated. However, if this was the case, one would see an improved performance of the adhesives with a higher shear stiffness. Since no effect of the internal structures is present the decrease in friction force is likely related to the difficulty of making contact and forming adhesive bonds.

4.3.4. Performance of structured adhesives on Cloth

On cloth, we observed no relation between the surface structure of the adhesives and friction force generated. The patterned surface structures were probably too coarse to be able to conform to the roughness of the cloth substrate. Also, due to the roughness of the cloth, the flat adhesives probably did not form a larger contact area. A slight effect of the internal structures was found, the adhesives with a higher shear stiffness (I1 and I4) performed slightly better compared to adhesives with a low shear stiffness. Meaning, these adhesives were able to distribute the stress better over the formed contact.

4.4. Conclusion and outlook

By 3D printing porous geometries, we were able to fabricate single-material adhesives with anisotropic stiffness. We showed that by modulating the stiffness in the normal and shear direction, by using various internal structures, adhesives that generate friction forces on a multitude of substrates, can be created. On stiff glass substrate, we observed that a decrease in normal stiffness leads to an increase in contact area and that adhesives with the lowest normal stiffness performed best. On flexible PDMS substrates we still observed an improved contact area

for adhesives with a lower normal stiffness, but the substrate also contributed to the formed contact. In addition, on these substrates the adhesives that combine a low normal stiffness with a high shear stiffness performed best.

With the flexibility of 3D printing, this approach can easily be used to create adhesives with unique stiffness profiles with specific direction dependent properties. The internal structures could be improved to generate a geometry-controlled stiffness gradient in the material to enhance the stress distribution under loads.

Alternative 3D printing methods should be explored to obtain higher resolutions, making it possible to create internal geometries with smaller features. This would improve the functionality of the internal geometries. Furthermore, printing at a higher resolution, could benefit the packing density and feature size of a patterned surface structure, making it possible to investigate whether the adhesives can benefit from contact splitting. Furthermore, surface structures that contain smaller features might be able to conform to the surface roughness of substrates like cloth enhancing the performance on rough substrates.

In addition, investigating the use of different printing materials should be considered to improve the adhesive abilities under different conditions. For example, a material could be used that improves the adhesive properties under wet conditions.¹⁹ Combining a specific adhesive material with a substrate might improve the critical strain energy release rate and improve the generated forces.

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Appendix A

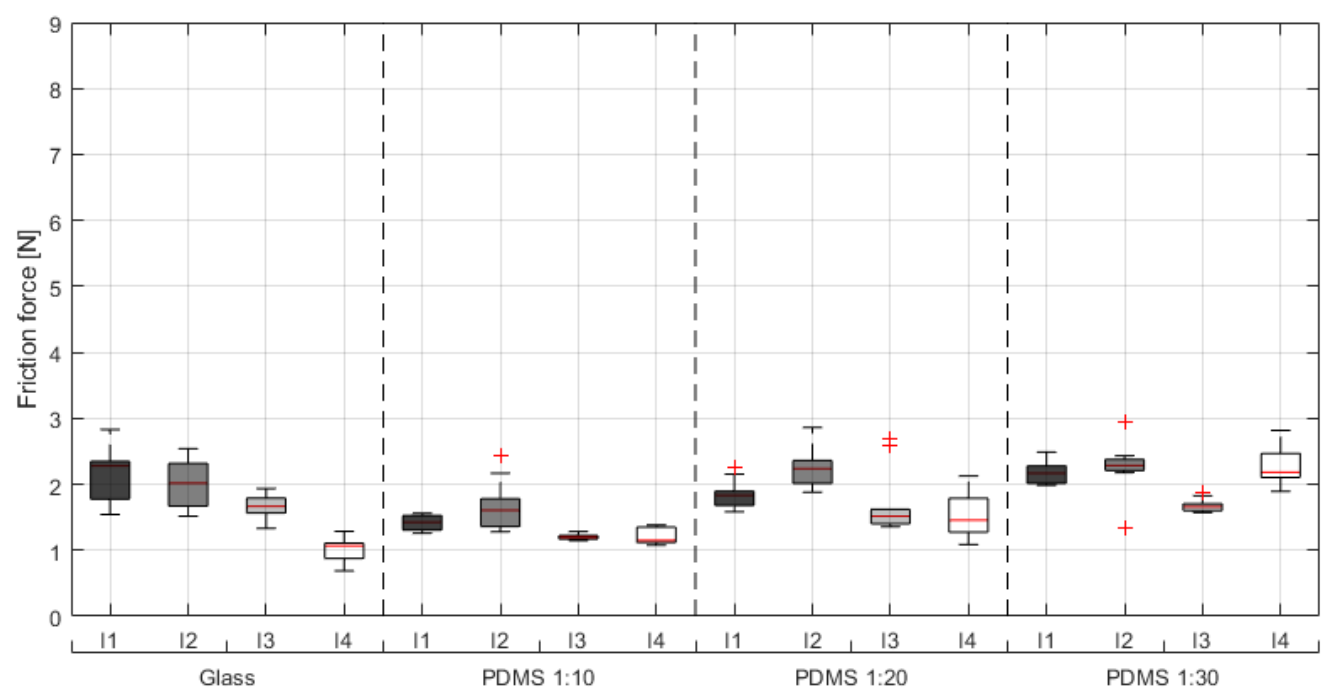


Figure 17: Friction force measurements of adhesive samples with a pillared surface structure.

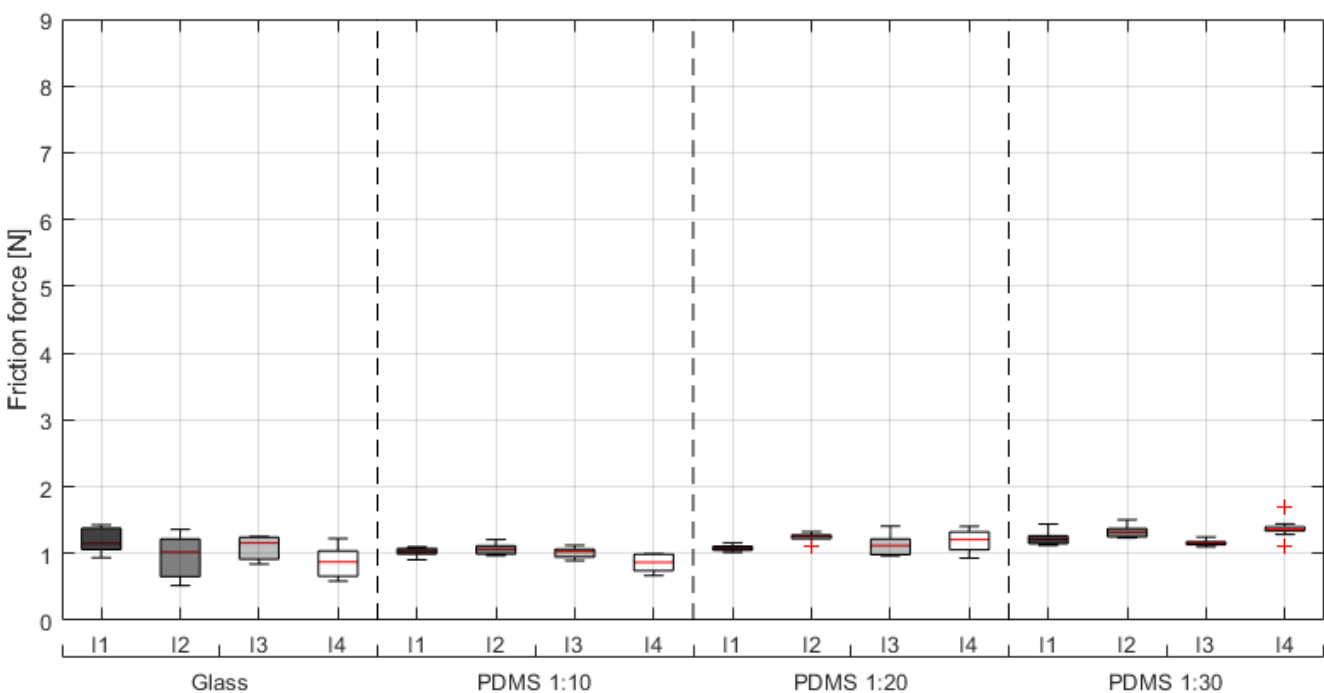


Figure 18: Friction force measurements of adhesive samples with a hooked surface structure.

Appendix B

Table 2: Means and standard deviations of the friction force measurements derived from ten repetitions for each condition

	Internal structure	Flat	Pillared	Hooked
		Mean $\pm$ std	Mean $\pm$ std	Mean $\pm$ std
Glass	I1	2.83 $\pm$ 0.47	2.14 $\pm$ 0.40	1.19 $\pm$ 0.18
	I2	2.83 $\pm$ 0.26	2.00 $\pm$ 0.38	0.97 $\pm$ 0.31
	I3	1.96 $\pm$ 0.17	1.65 $\pm$ 0.18	1.10 $\pm$ 0.16
	I4	1.61 $\pm$ 0.22	1.02 $\pm$ 0.19	0.86 $\pm$ 0.23
PDMS 1:10	I1	2.01 $\pm$ 0.50	1.41 $\pm$ 0.11	1.02 $\pm$ 0.06
	I2	2.43 $\pm$ 0.59	1.68 $\pm$ 0.37	1.06 $\pm$ 0.08
	I3	1.90 $\pm$ 0.30	1.20 $\pm$ 0.05	1.01 $\pm$ 0.08
	I4	2.55 $\pm$ 0.26	1.22 $\pm$ 0.13	0.86 $\pm$ 0.13
PDMS 1:20	I1	4.02 $\pm$ 1.40	1.85 $\pm$ 0.22	1.08 $\pm$ 0.05
	I2	4.72 $\pm$ 1.13	2.24 $\pm$ 0.30	1.24 $\pm$ 0.07
	I3	3.49 $\pm$ 1.14	1.71 $\pm$ 0.50	1.13 $\pm$ 0.16
	I4	3.16 $\pm$ 0.70	1.51 $\pm$ 0.32	1.19 $\pm$ 0.16
PDMS 1:30	I1	7.31 $\pm$ 0.44	2.18 $\pm$ 0.17	1.23 $\pm$ 0.10
	I2	8.16 $\pm$ 0.34	2.26 $\pm$ 0.39	1.32 $\pm$ 0.08
	I3	6.20 $\pm$ 0.74	1.68 $\pm$ 0.10	1.16 $\pm$ 0.05
	I4	5.00 $\pm$ 0.91	2.28 $\pm$ 0.27	1.38 $\pm$ 0.15
Cloth	I1	0.82 $\pm$ 0.11	0.83 $\pm$ 0.11	0.74 $\pm$ 0.10
	I2	1.10 $\pm$ 0.16	0.90 $\pm$ 0.14	0.93 $\pm$ 0.08
	I3	0.71 $\pm$ 0.04	0.69 $\pm$ 0.13	0.73 $\pm$ 0.07
	I4	0.96 $\pm$ 0.14	0.93 $\pm$ 0.12	0.96 $\pm$ 0.15
PVA	I1	0.57 $\pm$ 0.13	0.29 $\pm$ 0.04	0.28 $\pm$ 0.04
	I2	0.46 $\pm$ 0.11	0.26 $\pm$ 0.04	0.24 $\pm$ 0.03
	I3	0.48 $\pm$ 0.19	0.29 $\pm$ 0.06	0.28 $\pm$ 0.04
	I4	0.42 $\pm$ 0.13	0.30 $\pm$ 0.05	0.27 $\pm$ 0.05

Appendix C

Sample Printing

The samples were printed on a Form2 printer (Formlabs, Somerville, MA, USA) using a flexible rubber like resin (Flexible V2 (FLFLGR02), Formlabs, Somerville, MA, USA) with the suppliers recommended print settings and a layer thickness of 0.05 mm. This printer is a bottom-up printer that peels the cured resin from the bottom of the tank after each print layer. The peeling motion consists of a small sliding motion before the bed is lifted from the resin tank. In order to reduce peel forces that cause delamination and print failure it is common practice to minimize cross-sectional area of each layer and maximize the stiffness in the sliding direction. This meant that the adhesives should be printed under an angle. (Figure 19a) However due to the flexibility of the material and thin features, printing under an angle or in a vertical position resulted in a wrinkled surface of the adhesive sample. (Figure 19b) Therefore we printed the parts in an upright position, using a custom support structure, with the holes of the adhesive structures aligned with the peeling direction of the printer. This gave the structures the highest stiffness against the peeling forces, and a smooth surface. Printing right of the print bed was not possible since the Form 2 compresses the first 3 millimeters to ensure bonding with the print bed.

To prevent swelling and tearing of the adhesive structures during cleaning, we minimized the use of isopropyl alcohol (IPA) to remove uncured resin from the structures. First, we let most of the uncured resin drain from the structures and removed remaining uncured resin inside the pores with compressed air (EW5601, Ewent, Geleen, Netherlands). Finally, to ensure that no uncured resin remained sticking to the walls of the pores, the structures were immersed in IPA for a few seconds and dried directly using the compressed air. The external structures cleaned by brushing them off with a brush soaked in IPA. This made the uncured resin that was trapped between the features of the external structure more fluid such that it could be removed with compressed air. In most cases the cleaning of the external structure had to be repeated a few times until all the resin was removed.

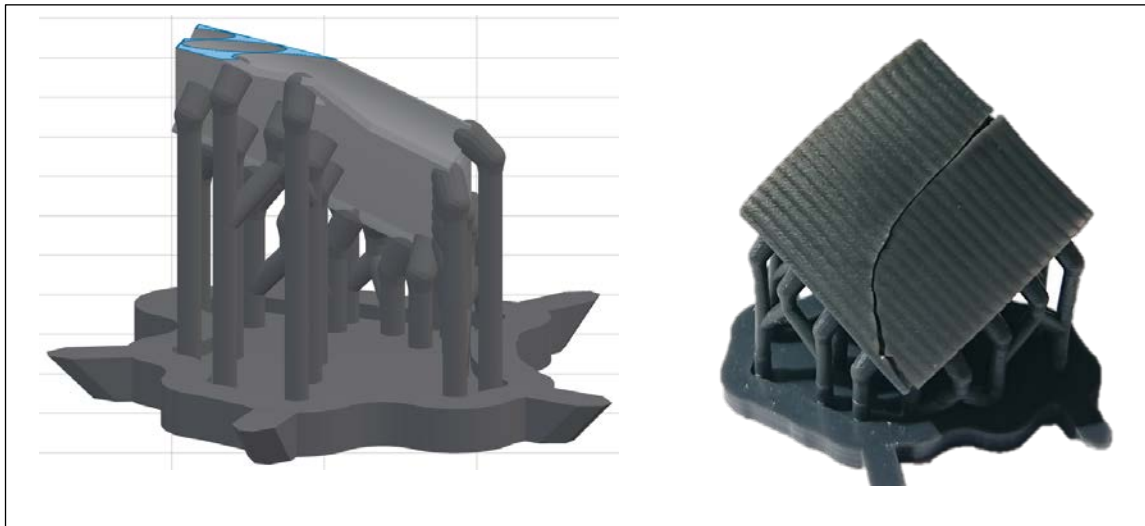


Figure 19 A) A adhesive structure that is orientated and supported using the recommended settings and support structure. The layer cross section (blue) is minimized to lower peel forces. The touchpoints of the support enclose large parts of the internal structure. B) Printed with the recommended settings resulted in a deformed and wrinkled surface. The part is torn due to stresses caused by swelling.

Elastic resin

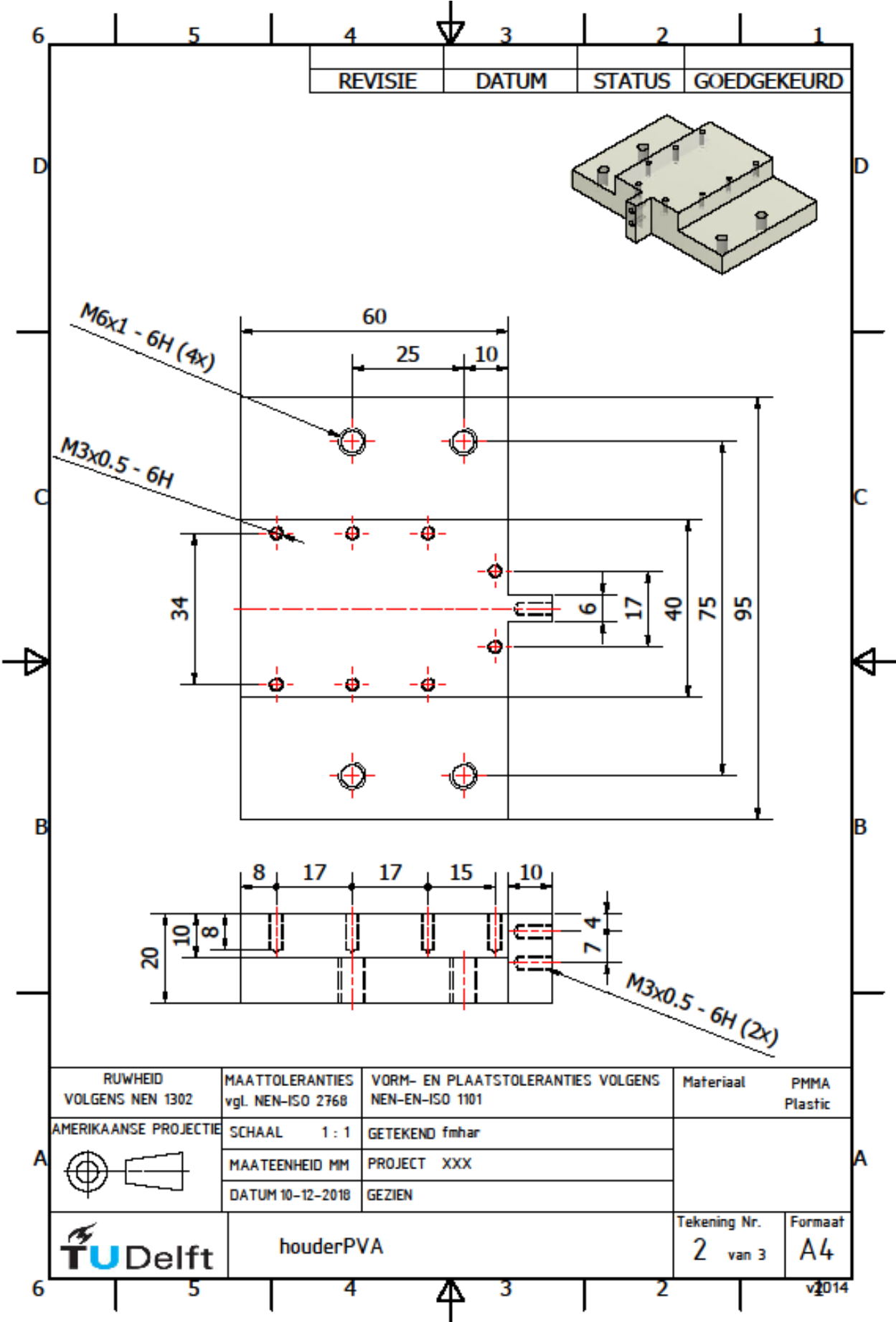
Besides printing with the flexible resin, we also printed adhesive samples using an elastic resin (Elastic (FLELCL01), Formlabs, Somerville, MA, USA), with the recommended layer thickness of

0.1 mm. This resin had a lower stiffness compared to the flexible resin (shore hardness 50A compared to 80A).

However, printing with this resin proved to be difficult. Since this resin is clear the UV light would bleed through the printed layers and the uncured resin inside the pores would also cure, clogging the internal structures. Therefore, we investigated if adding a light blocker (Fine Tuner LB1 Bio, 3Dresyns, Barcelona, Spain) to the resin would minimize the amount of light bleeding through the printed layers. To find the required concentration to eliminate the bleeding, we printed several test samples and increased the concentration of light blocker with 50ML (steps of 0.2%) each time. We found that when 2.8% light blocker was added to the resin the bleeding of light was almost eliminated and we were able to produce adhesives with the desired internal structures.

However, we were not able to successfully print any surface pattern, the resin between the pillars or hooks would still cure, even with the light blocker that was added. In addition, it turned out that adhesive samples produced with this material did not have the desired adhesive properties, presumably due to a low critical strain energy release rate. Therefore, we did not perform any tests with samples made with this type of material.

Appendix D



Appendix E

Mechanical charactersation tests		Friction force measurements	3D print files
Shear test data  Shear Data.zip	Indentation test data  Indentation Data.zip	Friction force data  AllData.rar	Friction force test setup part(s).  PVAPlate.stl
Shear test matlab script  Analyze_Shear.m	Indentation test matlab script  Analyze_Indentation.m	Friction force matlab script  Analyze_2.m	Adhesive samples  adhesive sample files.zip

Appendix F

FLEXIBLE FLFLGR02					
	METRIC ¹		IMPERIAL ¹		METHOD
	Green	Postcured ²	Green	Postcured ²	
Mechanical Properties					
Ultimate Tensile Strength ³	3.3 – 3.4 MPa	77 – 8.5 MPa	483 – 494 psi	1110 – 1230 psi	ASTM D 412-06 (A)
Elongation at Failure ³	60 %	75 – 85 %	60 %	75 – 85 %	ASTM D 412-06 (A)
Compression Set ⁴	0.40 %	0.40 %	0.40 %	0.40 %	ASTM D 395-03 (B)
Tear Strength ⁵	9.5 – 9.6 kN/m	13.3 – 14.1 kN/m	54 – 55 lbf/in	76 – 80 lbf/in	ASTM D 624-00
Shore Hardness	70 – 75 A	80 – 85 A	70 – 75 A	80 – 85 A	ASTM 2240
Thermal Properties					
Vicat Softening Point ⁶	231 °C	230 °C	448 °F	446 °F	ASTM D 1525-09

NOTES:

¹Material properties can vary with part geometry, print orientation, print settings and temperature.

²Data was obtained from parts printed using Form 2, 100 µm, Flexible settings and post-cured with 80.5 mW/cm² of 365 nm fluorescent light for 60 minutes.

³Tensile testing was performed after 3+ hours at 23 °C, using a Die C dumbbell and 20 in/min cross head speed.

⁴Compression testing was performed at 23 °C after aging at 23 °C for 22 hours.

⁵Tear testing was performed after 3+ hours at 23 °C, using a Die C tear specimen and a 20 in/min cross head speed.

⁶Thermal testing was performed after 40+ hours with a 10 N loading at 50 °C/hour. Cracks formed in samples during testing.