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PDMS-Based Elastomer Tuned Soft, Stretchable, and Sticky for Epidermal Electronics

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Targeting good user experiences, softness and stretchability, are essential features for epidermal devices in body signal monitoring and body area stimulation. A highly soft, stretchable, and sticky polydimethylsiloxane-based elastomer (S3-PDMS) is achieved by a simple process with a widely used siloxane precursors, the properties of which are tuned by adding small fractions of an amine-based polymer, ethoxylated polyethylenimine (PEIE). This allows formation of a thick unobtrusive patch and may ease implementation of epidermal electronics in wearable healthcare applications.

Being able to conformally attach electronic devices onto the human skin with a very low level of mechanical interfacial stress, which otherwise results in inconvenience, epidermal electronics offers an unsurpassed experience to users and is considered one of the revolutionary electronic technologies for the near future.[1] One of the supporting material systems behind this technology, polydimethylsiloxane(PDMS)-based elastomers,[2] serves as a substrate or carrier, bridging the gap between high modulus components such as metal connectors or silicon-based active electronics and soft, stretchable human skin.[3] PDMS is the most commonly used polymer containing the atomic element silicon (Si). It can be used alone to form PDMS elastomers by crosslinking. Often PDMS is combined with other components to form compositions that we in this publication have given the terminology, "PDMS-based elastomer." Recently their use have proven successful, e.g., for mechanical components such as tubes, wires and physical structures, adopted for soft robots.[4] However, compared to our skin, the most widely used, room temperature-curing two component (silicone base and crosslinker) PDMS-based elastomers, such as Sylgard184

(Dow Corning) and ElastosilRT601 (Wacker Chemie), are not soft enough to satisfy the demands of epidermal electronics.

In clinical and healthcare applications, biomedical adhesives for skin have been used a long time. Recently, more advanced and functional adhesives have shown high potential. For example, adhesives of biological systems such as 3,4-dihydroxy-L-phenylalanine (DOPA) from a mussel,[5] gelatin-, collagen-, or chitosan-based materials[6] and a Gecko adhesive[7] have been developed with intention of better performance in, e.g., biocompatibility, adhesion strength, reusability, compliance (softness), degradation time, antifouling, and swelling. To be able to use a silicone elastomer as an adhesive for skin, however, its modulus, wetting and fluidity need to be improved. Requirements for the soft patch include spontaneous conformal contact upon application onto the skin while at the same time providing for ease of detachment.

Previously, many approaches for tuning the mechanical properties of common PDMS-based elastomers have been reported. An early work used a thinner to tune modulus.[8] This approach did not enhance stretchability (elongation at break) and led to less bondability with plasma treatment. More commonly, a softer PDMS-based elastomer was achieved by reducing the amount of a crosslinker or introducing a (photodefined) inhibitor.[9] Unfortunately, such strategies, especially the former, often result in poor mechanical properties. In short, the modified PDMS-based elastomer from the above-mentioned approaches is not compliant (soft) and stretchy enough for compliant epidermal electronics. To achieve such an aim, one approach is to use siloxanes with higher molecular weight base and crosslinker having lower degree of functionalization, which can be provided by commercial suppliers (e.g., SilasticQ7-4720, 4780, Dow Corning). However, the processing of such highly viscous silicone elastomer precursors requires dedicated mixing equipment and hence handling is significantly more inconvenient that of the lower viscosity Sylgard 184 and Elastosil RT601. An alternative way is to use a lower modulus and highly elastic silicone elastomer such as EcoFlex00-30 (Smooth-On). However, this type of silicone elastomer is not as easy to handle as the traditional PDMS based elastomer: the precursor has high viscosity after mixing, is nontransparent and does not easily bond to itself by plasma surface treatment after curing. Very recently, a soft and nonsticky PDMS elastomer was synthesized with a brush-like molecular architecture limiting entanglements to provide a very low modulus.[10]

In this work, we demonstrate a simple way to make a commonly used, vinyl terminated, poly(dimethylsiloxane) and poly(methylhydrosiloxane)-based silicone elastomer, which is curable at room temperature. (Hereafter, the PDMS-based elastomer is Sylgard 184 unless otherwise specified.) into a soft, stretchable and sticky PDMS based elastomer (S3-PDMS) by

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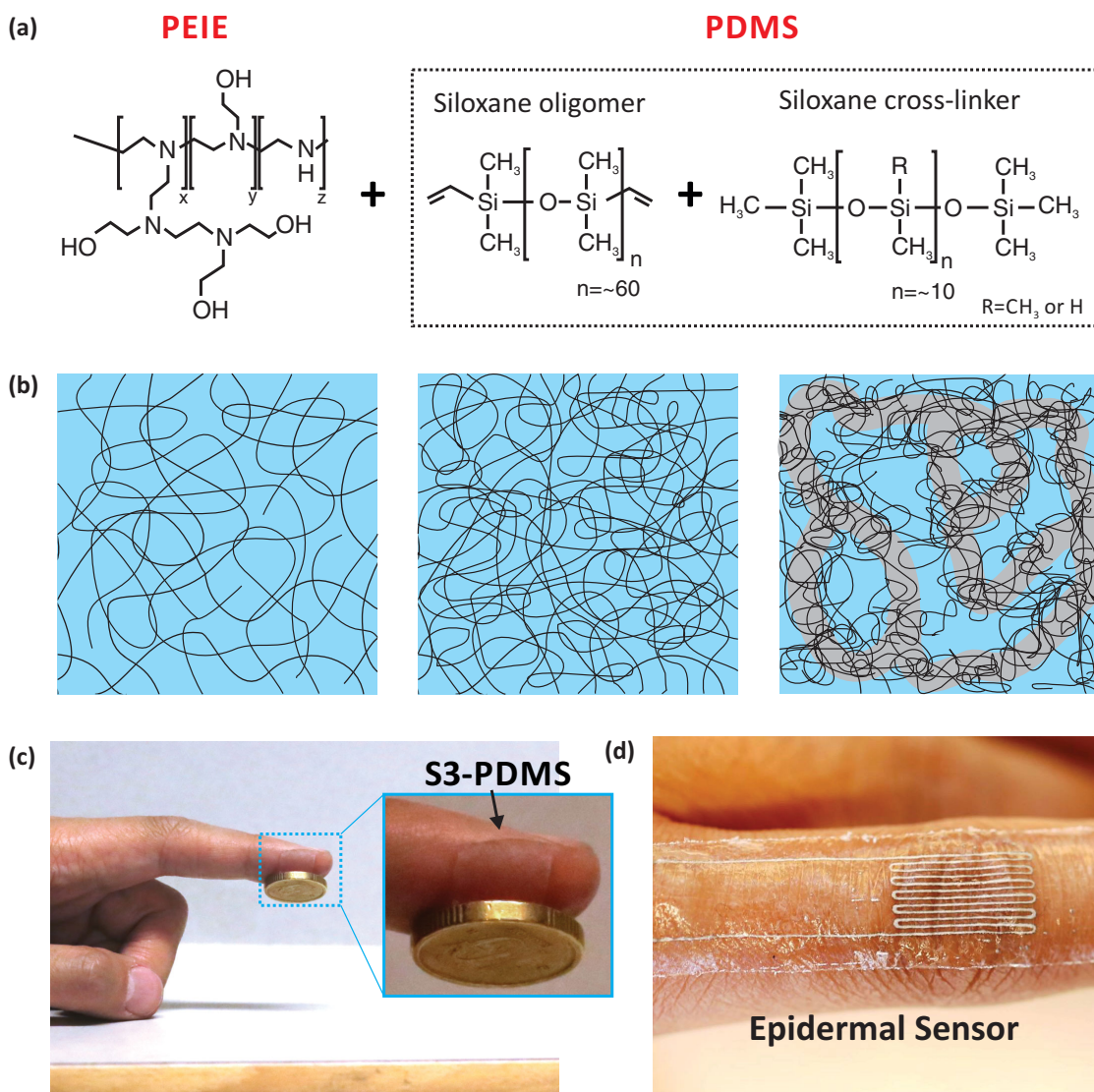Uppsala University
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Figure 1. a) Chemical structures of polydimethylsiloxane and ethoxylated polyethylenimine solution, b) a homogeneous crosslinking network of the PDMS-based elastomer (1:40) with low density of crosslinking network (left), compared to the PDMS based elastomer (1:10) with a high density of crosslinking (middle) and heterogeneous crosslinking network of the S3-PDMS (40 μL PEIE), which includes high-density crosslinking networks (grey-colored areas) and low-density crosslinking networks (right), c) the S3-PDMS conformally attached on the fingertip with a coin above a desk, and d) a stretchable strain sensor adhered on finger skin.

adding a tiny amount of a solution of an amine-based polymer, ethoxylated polyethylenimine (PEIE), into the mixture of the silicone base and crosslinker of the PDMS-based elastomer, **Figure 1**a. More importantly, we show that the mechanical compliance, elongation at break and adhesion force of the PDMS based elastomer to the human skin can be tuned by changing the amount of the PEIE additive. All the time, the S3-PDMS retains good processability, similar to the unmodified one, such as low viscosity after mixing, easy wetting of many surfaces, transparency and strong bonding to itself after plasma treatment. Specifically, it significantly increases the adhesion to human skin, which helps epidermal electronic systems to be conformally attached, allowing large deformation without delamination-induced performance degradation. Figure 1b schematically illustrates the cured PDMS-based elastomer network, which can be used for explaining the mechanism behind our new approach. Figure 1c shows a piece of the S3-PDMS that was conformally attached onto a fingertip with an attached coin to demonstrate functionality. As a demonstration, a self-adhesive and conformal strain sensor, which responded to human motion, was applied to a finger skin without any glue or interface material in Figure 1d.

We investigated the stress–strain behavior of the S3-PDMS to determine mechanical compliance (Young's modulus, as one of the property of softness) and elongation at break at various concentration of PEIE mixed into the siloxane precursors, **Figure 2**a. Compared to the original siloxanes processed with the recommended mixing ratio from the manufacturer, we found out that a very small amount of PEIE could significantly change the mechanical properties of the PDMS-based elastomer. In contrast to the common way of making the PDMS-based elastomer soft
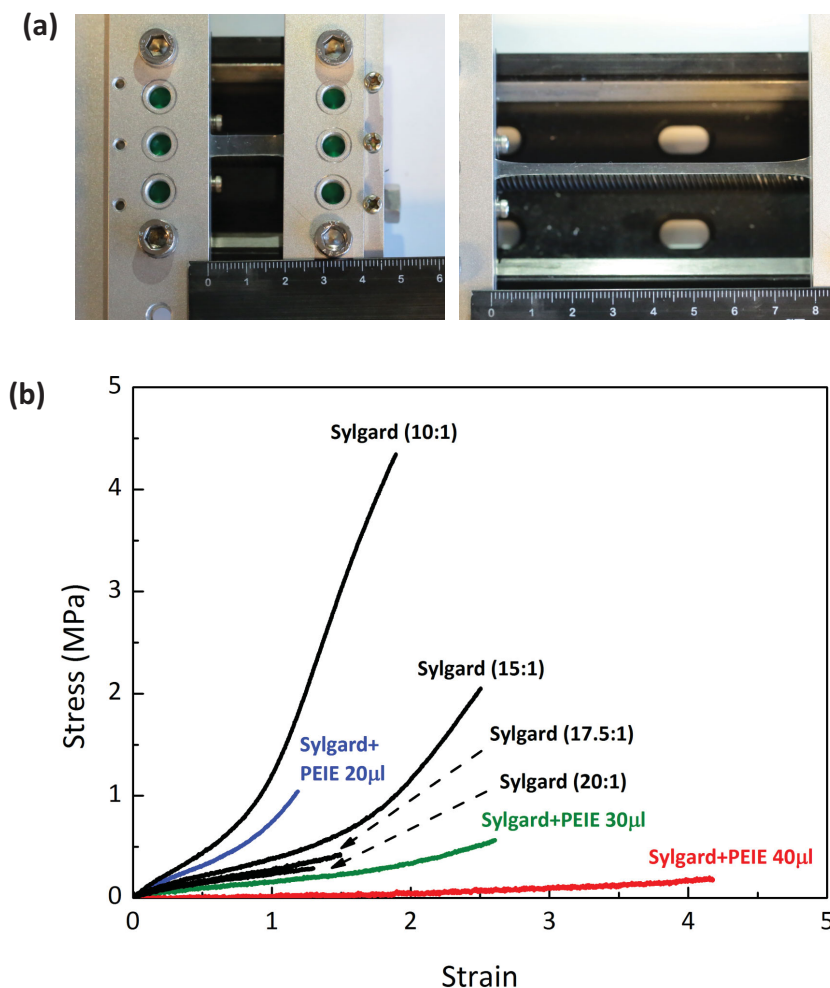
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Figure 2. a) Photographs of an original and stretched sample up to 300% of the initial length, and b) the stress–strain curves of the PDMS-based elastomers.

curing, i.e., hydrosilylation of the vinyl functional base component by the hydrosilyl functional crosslinker, is catalyzed by a platinum complex (Pt). The Pt catalyst is, however, coordinatively unsaturated and hence forms strong complexes with, e.g., amines like those in PEIE.[12] It is therefore likely that the areas around the PEIE containing water droplets are depleted in Pt catalyst yielding reduced or inhibited crosslinking, resulting in a heterogeneously crosslinked system. The curing of the PDMS is partially controlled by the diffusion of Pt and the curing process is thus much slowed down with less Pt.

To better understand the difference in the crosslinked structure of the S3-PDMS, a swelling test with cyclohexane was performed to show four times higher swelling ratio for the S3-PDMS (40 μL PEIE) than the unmodified PDMS-based elastomer with the same feed ratio (e.g., a mixing ratio of the crosslinker to the silicone oligomer, 1:10). The 62% gel fraction of the S3-PDMS is significantly lower than the 95% gel fraction of the corresponding PDMS-based elastomer (Table S1 in the Supporting Information) Therefore, we conclude that the S3-PDMS has a low density of crosslinking or more likely a heterogeneous crosslinking, which makes it more compliant compared to the original PDMS-based elastomer. In addition, to have much higher elongation at break than the original PDMS-based elastomer, the crosslinking network has to be either a homogeneously and loosely crosslinked network, or a heterogeneous crosslinked network with locally low density crosslinking.

To render PDMS-based elastomers softer, one method is to use off-stoichiometry by a lower fraction of the crosslinker to provide a lower degree of crosslinking. When the PDMS-based elastomer is formed at off-stoichiometry, e.g., a mixing ratio of the crosslinker to the silicone base, 1:40, and at the same curing condition as the S3-PDMS (40 μL PEIE), the resulting elastomer has a low crosslinking density and a large fraction of unreacted precursors. These precursors remain as uncrosslinked chains, and are thus compliant. However, to distinguish the network homogeneity, it is not enough to only consider the lower amount of Pt to explain the low elastic modulus of the S3-PDMS.

To evaluate possible heterogeneity the swollen samples of the S3-PDMS were compared to the off-stoichiometric soft PDMS-based elastomer (1:40) at the same gel fraction (Table S1 in Supporting Information). First, the equilibrium swelling ratio of the S3-PDMS is 17% higher than that of the off-stoichiometric PDMS-based elastomer (1:40) at a similar gel fraction, implying different crosslinking network structures. Second, the mechanical integrity of the S3-PDMS was lower than that of the off-stoichiometric PDMS-based elastomer (1:40) at the swollen state when they were handled in the swelling test. This

by changing the mixing ratio between the crosslinker and the silicone base, the compliance and elongation at break of the S3-PDMS were both improved by increasing the PEIE amount (Figure 2b). This shows that the mechanical characteristics of the PDMS-based elastomer can be uniquely tuned by varying the PEIE concentration. Most importantly, without introducing large changes in the processing protocol of the PDMS-based elastomer, the S3-PDMS could be made softer than human skin (Young's modulus of the human skin is in the order of 1 MPa or less[11]). The elongation at break of the S3-PDMS was increased to over 300% and the Young modulus of the S3-PDMS was decreased to 24 kPa from the 1 MPa of the original PDMS-based elastomer (with the recommended mixing ratio from the supplier). Unfortunately, the elongation at break was often reduced by edge damage in our experiments, introducing a larger variation at high strains. At high PEIE concentrations, the curing time of the S3-PDMS was prolonged compared to that of the original PDMS-based elastomer. Still, we obtained a useful and controllable processing window for the S3-PDMS.

To improve compliance and elongation at break of the S3-PDMS, we added a small amount of PEIE to change the crosslinking network structure. In our system, the PDMS

strongly supports the idea that the S3-PDMS is heterogeneously crosslinked as we shown in Figure 1b. In addition, the loosely crosslinked regions can be more easily ruptured than the homogeneously crosslinked off-stoichiometric PDMS-based elastomer (1:40) (Figure S2 in Supporting Information). But the S3-PDMS does not lose structural integrity compared to the off-stoichiometric PDMS-based elastomer (1:40) because the elongation at break over 300% of the S3-PDMS has a similar level as that of the off-stoichiometric PDMS based elastomer (1:40), which indicates that the S3-PDMS has regions with various crosslinking density.

The increased compliance and elongation at break in a heterogeneously crosslinked network are likely to originate from the local sol inclusions of uncrosslinked or loosely crosslinked domains around PEIE.[13] Rheology tests were made to investigate the viscoelastic properties of the S3-PDMS by measuring the storage modulus, G′, and a loss modulus, G′′, as a function of frequency (Figure S3 in the Supporting Information). These tests support the swelling and sol–gel characterization results by showing the stronger frequency dependence of G′ and G′′ for S3-PDMS indicative of higher dependence on entanglement and viscous flow.

To analyze if any water inclusion was entrapped in the S3-PDMS, drying at 90 °C for a month was performed. The weight loss of the S3-PDMS was only 0.6%. In addition, thermal gravimetric analysis (TGA) on a freshly prepared S3-PDMS showed a 0.5% weight loss at 100–120 °C, which corresponds well to the drying test. In fact, water vapor permeability through the PDMS-based elastomer is known to be high. PEIE dissolved in water is suspended at 0.1% concentration into the PDMS-based elastomer resulting in a slightly hazy material. Scattering,

or transparency reduction, in the S3-PDMS compared to that of the PDMS-based elastomer is likely due to the dispersed PEIE (Figure S1 in the Supporting Information).

Apart from the tunability of the mechanical property, the S3-PDMS became stickier when more PEIE was added. As shown in **Figure 3**a, the adhesion force of the S3-PDMS was increased by adding more PEIE. The S3-PDMS with 40 µL PEIE gave the highest adhesion in our experimental conditions, which were fulfilled below 100 °C for curing. The adhesion force of the S3-PDMS with 40 µL PEIE to human skin could be more than 10 times higher (1.2 N) than that of the PDMS-based elastomer of 1:10 ratio (0.1 N) in our adhesion force measurement (for more details, refer to the experimental section). In the image of the field-emission scanning electron microscope (FE-SEM, Leo 1550, Zeiss), micrometer-sized wrinkles were observed on the S3-PDMS surface (Figure 3b). According to the previous study,[1a] such an enhanced surface contact with a thin and large area system can increase adhesion strength due to the geometry and improved development of van der Waals forces with better wetting and spreading. These forces are regarded to contribute to the main adhesion mechanisms affecting the human skin in epidermal electronics. The adhesion force in shear direction was also tested with the same method (Figure S4 in the Supporting Information).

Compared to the Sylgard 184 and EcoFlex 00-30, the adhesion force between the S3-PDMS and the human skin was much stronger, as shown in Figure 3c. The relevant properties in comparison to the Sylgard 184 or EcoFlex 00-30 are summarized and listed in **Table 1**. The reusability of S3-PDMS (40 µL PEIE) was examined on forearm skin over 100 times, and the adhesion force was evaluated in the same manner with
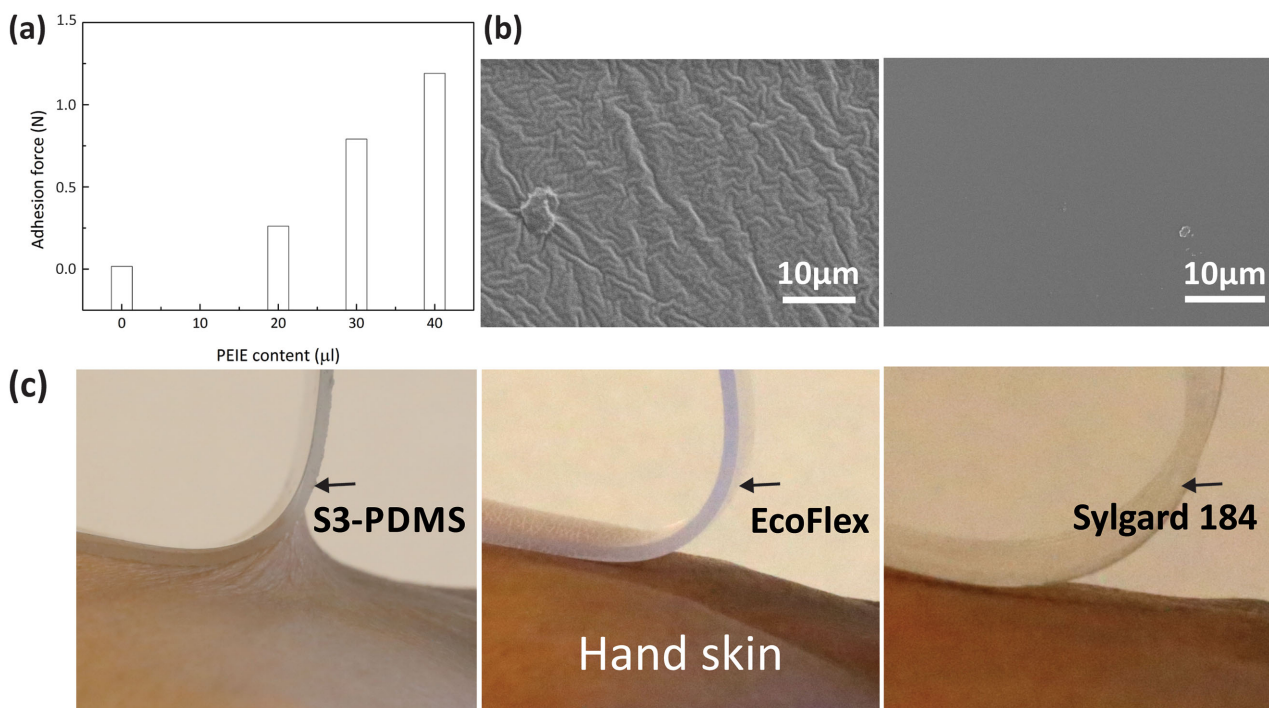

Figure 3. a) Comparison of the adhesion forces of the S3-PDMS on human forearm skin, b) SEM images of the S3-PDMS with 40 µL PEIE, and c) photographs of the S3-PDMS adhesion on the skin of a hand with references.

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Table 1. Comparison of properties of two silicone elastomers with the S3-PDMS.

	S3-PDMS[b]	00EcoFlex 00-30	Sylgard 184
Adhesion force[a]	1.2 N	<0.1 N	<0.1 N
Young's modulus	24 kPa[c]	35–69 kPa[14]	1 MPa[c]
Elongation at break	>300%	900%[d]	>150%

All the data were from our measurements unless otherwise specified. [a]The adhesion area in the adhesion test was 17.5 cm². The sample was peeled off from one side; [b]The S3-PDMS with 40 µL PEIE was tested; [c]The moduli were fitted in the region from 0% to 100% strain; and d) the data are from the datasheet provided by the manufacturer.

a spring gauge (Figure S5 in the Supporting Information). To check the stability of the S3-PDMS, the change of adhesion force and weight was measured between before and after washing out in solvents such as DI water, isopropyl alcohol (IPA), or acetone for 1 d. The adhesion force after washing did not change much, varying less than 20% of the initial adhesion force (Table S2, Supporting Information). Further, the S3-PDMS-made self-adhesive patches were intimately attached on the forearm skin of three volunteers, and stayed for 12 d when scratch/friction was prevented. There were no observed negative effects on the skin, e.g., red color spots or itching during the days. This simple test showed that such a PDMS-based elastomer can be used on human skin but a proper biological evaluation regarding to biomedical devices is required to further progress. In addition, to check if water inclusion could affect the adhesion, it was examined during the time that the sample was kept in a vacuum oven at 25 °C under 100 mbar for 5 d. The results showed no influence from the vacuum drying (Figure S6, Supporting Information).

The S3-PDMS can be reversibly attached and detached on human skin by finger force. Hence, the increased adhesive force is mainly be due to the very high compliance (very low elastic modulus) and viscous surface adaptation of the S3-PDMS, allowing development of van der Waals interactions with a underlying substrate. The high compliance permits relaxation to provide for a low stress interfacial region of the adhesive S3-PDMS, to contribute to the overall adhesion. In addition to the intimate contact between the S3-PDMS and human skin to maximize the van der Waals force, dangling ends and free oil contribute to the good adhesion (high tack).[15]

Apart from the Sylgard 184, such an approach could also be extended to the Elastosil RT601. By adding the PEIE into the Elastosil RT601, the stretchability (Figure S7 in the Supporting Information) could be enhanced as well, but the modified Elastosil was not as sticky as that based on the Sylgard 184 (S3-PDMS) when the same amount was added. The 40 µL PEIE modified Elastosil RT601 was smoother than that of the S3-PDMS, as seen in the micrograph of Figure S8 in the Supporting Information. Similar to the S3-PDMS, the modified Elastosil became stickier when more PEIE was added.

To demonstrate the advantages of this new technique, a liquid alloy strain sensor in the S3-PDMS packaging was fabricated. It had a resistance of 12 Ω and was 150 µm thick in the total device. The strain sensor was conformally adhered on the finger skin after being transferred from a water-soluble adhesive paper substrate, **Figure 4**, Figure S9 and videos in the Supporting

Information. During the evaluation of the strain sensor over a period of 2 h, no delamination from the skin of the finger was observed. The sticky surface and the softness of the S3-PDMS packaging provided conformal implementation of the strain sensor on the rough finger skin surface. The self-adhesive strain sensor was elongated by bending the finger at different angles (90° and full bending, 0.03 Hz), Figure 4a. The liquid alloy resistor response of the strain sensor due to the finger bending motion was measured with good repeatability. High-frequency bending and expanding of the finger were well monitored with 1 Hz, Figure 4b and sensor performances at different frequencies and different bending angles were obtained with good resolution, Figure 4c. A further calibration of the strain sensor for the real motion with a motion detection system can help the sensor to be useful in real applications on the human body.[16]

In summary, we demonstrated a simple and easy way of tuning mechanical stretchability, compliance and adhesion force to the human skin of the PDMS-based elastomer. Depending on the amount of the PEIE content, mechanical compliance, elongation at break and adhesion force can be tuned according to a curing condition. We obtained the multifunctional S3-PDMS, having at the same time higher compliance, elongation at break and adhesion force than the PDMS-based elastomer Sylgard 184. The resulting S3-PDMS, was crosslinked with a solvent-free, one-step simple process by adding a tiny amount of PEIE to the commonly used, vinyl-terminated PDMS-based elastomer prepolymer. The S3-PDMS gave tremendous improvements in stretchability, softness, and adhesion. The compliance and elongation at break of the S3-PDMS were obtained from the heterogeneity of the crosslinking density, resulting in a liquid inclusion effect. A compliant surface and dangling ends of the S3-PDMS enhanced the adhesion of the S3-PDMS. An epidermal strain sensor with a liquid alloy resistor that spontaneously monitored finger movements was conformally attached to the finger skin. In conclusion, by combining the enhanced softness and its sticky surface, the S3-PDMS significantly enhances the conformability when it is used as a packaging material or an interface layer in epidermal electronic systems. We anticipate that the S3-PDMS can facilitate new devices and applications of epidermal electronics when conformal attachment is demanded to very soft or/and complex surfaces or dynamic biointeraction systems.[17] Moreover, the S3-PDMS can be further studied for biomedical adhesive applications by tuning the adhesion force and by practical testing on human skin.[18]

Experimental Section

PDMS-Based Elastomer Tuning: The crosslinking of the PDMS-based elastomer (Sylgard184, Dow corning) mixed with the PEIE (80% solution of ethoxylated polyethylenimine, 35–40 wt% in H_2O, average M_W = 70 000, Sigma-Aldrich) was done by hand mixing with a glass rod in a plastic cup for 2 min. A pipette (10 µL, Eppendorf) was used for PEIE sampling. The silicone base, curing agent of the PDMS-based elastomer and the PEIE solution were mixed together at the same time. The S3-PDMS was prepared with the PEIE variation of 20, 30, and 40 µL in 10 g of the silicone base of the PDMS-based elastomer. The curing agent was applied as a standard ratio 10% to the base, as usually. The mixture was cured at 90 °C for 2–3 h in an oven depending on the amount of the PEIE content. When Elastosil RT601 was used, the curing
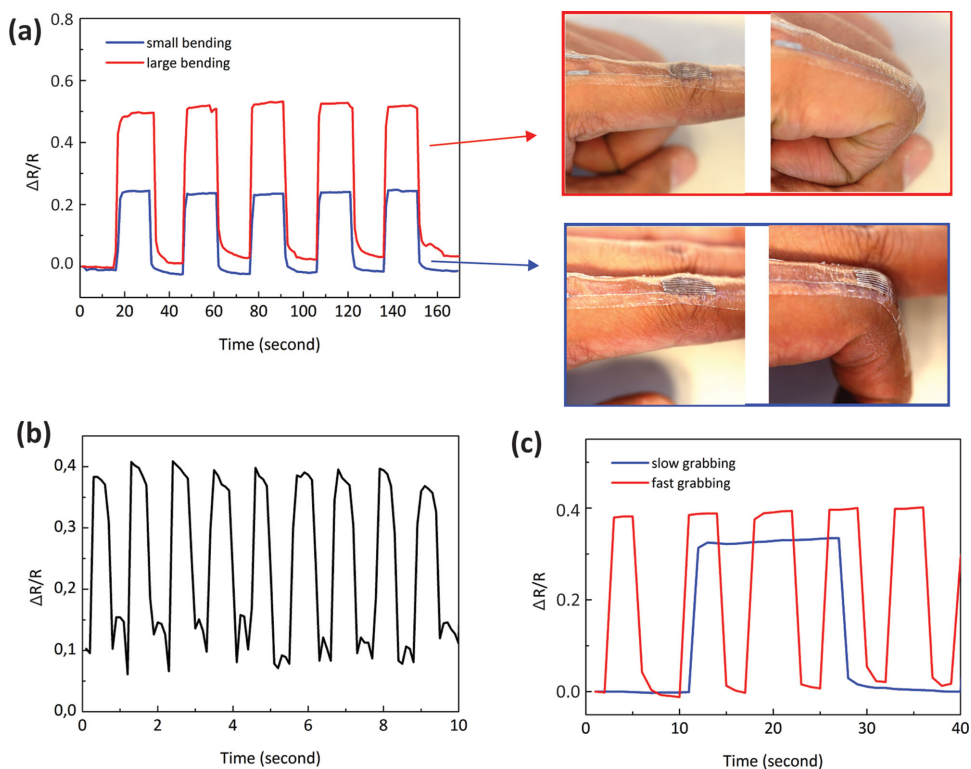
Figure 4. Performance of the self-adhered strain sensor on a finger. a) $\Delta R/R$ vs strain with different angle movements, b) higher frequency folding and unfolding motion of the finger by grabbing a 50 mm diameter pipe, c) sensing of grabbing motions of a finger on the pipe with different frequencies.

agent was added with the recommended weight ratio of 1:9 (curing agent:silicone base) from the supplier.

Mechanical Property Testing: The tensile test (engineering stress and engineering strain) was done in the universal tensile test instrument (AGS-X, Shimadzu) and a specimen shaped like a dog bone was prepared by cutting with a scalpel after curing in a plastic petri dish. The stroke speed of the tensile test was 100 mm min^{-1}.

Swelling Test and Sol–Gel Characterization: The PDMS-based elastomer (1:10 and 1:40 ratio of the crosslinker and silicone base) and the S3-PDMS (20, 30, and 40 µL PEIE fraction in 1:10 ratio of the PDMS-based elastomer) were prepared as described in the experimental section. Cyclohexane (Sigma-Aldrich, CHROMASOLV, for HPLC, ≥99.7%) was used for swelling and sol extraction. The samples (2.8 mm diameter, 1.25 mm thickness) were kept in cyclohexane (50 mL) in a glass bottle for 3 d to reach equilibrium swelling and measured the weight with a balance. The gel weight was measured with a balance after sol extraction for 7 d by the same process as the swelling test and drying for 3 d in air at room temperature with covering the sample to prevent quick drying causing cracks on the sample.

Adhesion Force Testing: The adhesion test of the S3-PDMS was done with a dynamometer (no. 22, 200 g, MWT) and a linear guide stage (A-LSQ, Zaber). The dynamometer (a spring force gauge) was fixed to the linear guide and the test sample was attached on the forearm skin that was clamped with one end of the sample by a plastic clip, Figure S10 in the Supporting Information. The sample size was 25 mm wide and 70 mm long for the measurements in Figure 3a and Table 1 while the rest acquired data were from sample sized 15 mm × 70 mm, which were specified in the Supporting Information. For the adhesion force test sample preparation, the mixture of the S3-PDMS was laminated on a plastic film (Transparency film, Canon) with a film applicator (PA2041, BYK). The thickness of the laminated layers was 50 µm. The peeling speed was controlled by the linear guide and the peeling speed was 20 mm s^{-1} with 100 mm stroke. The maximum adhesion force was read during peeling off the test sample from the skin of the forearm.

Fabrication and Characterization of the Strain Sensor: A liquid alloy (Galinstan, Geratherm medical) was patterned on the semi-cured S3-PDMS surface by a spraying technique via a metal stencil mask (the experimental details can be found in Figure S9, Supporting Information).[18] The strain sensor was transferred to the finger skin via a water-soluble layer (Toner transfer paper, Pulsa professional fx). The resistance change was monitored with a multimeter (34405A, Agilent Technologies) at the different strain conditions. The detailed schematics of the processing are given in Figure S9 in the Supporting Information.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

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[1] a) D.-H. Kim, N. Lu, R. Ma, Y.-S. Kim, R.-H. Kim, S. Wang, J. Wu, S. M. Won, H. Tao, A. Islam, K. J. Yu, T.-I. Kim, R. Chowdhury, M. Ying, L. Xu, M. Li, H.-J. Chung, H. Keum, M. McCormick, P. Liu, Y.-W. Zhang, F. G. Omenetto, Y. Huang, T. Coleman, J. A. Rogers, *Science* **2011**, *333*, 838; b) C. Dagdeviren, Y. Shi, P. Joe, R. Ghaffari, G. Balooch, K. Usgaonkar, O. Gur, P. L. Tran, J. R. Crosby, M. Meyer, Y. Su, R. C. Webb, A. S. Tedesco, M. J. Slepian, Y. Huang, J. A. Rogers, *Nat. Mater.* **2015**, *14*, 728.

[2] a) Y. Xia, G. M. Whitesides, *Annu. Rev. Mater. Sci.* **1998**. *28*, 153; b) N. Bowden, S. Brittain, A. G. Evans, J. W. Hutchinson, G. M. Whitesides, *Nature* **1998**, *393*, 146.

[3] a) A. Carlson, A. M. Bowen, Y. Huang, R. G. Nuzzo, J. A. Rogers, *Adv. Mater.* **2012**, *24*, 5284; b) T. Sekitani, T. Someya, *Adv. Mater.* **2010**, *22*, 2228; c) M. L. Hammock, A. Chortos, B. C.-K. Tee, J. B.-H. Tok, Z. Bao, *Adv. Mater.* **2013**, *25*, 5997; d) X. Wang, L. Dong, H. Zhang, R. Yu, C. Pan, Z. L. Wang, *Adv. Sci.* **2015**, *2*, 1500169.

[4] D. Rus, M. T. Tolley, *Nature* **2015**, *521*, 467.

[5] H. Chung, R. H. Grubbs, *Macromolecules* **2012**, *45*, 9666.

[6] a) K. Su. C. Wang, *Biotechnol. Lett.* **2015**, *37*, 2139; b) Y. Lee, J. W. Bae, D. H. Oh, K. M. Park, Y. W. Chun, H.-J. Sung, K. D. Park, *J. Mater. Chem. B* **2013**, *1*, 2407; c) J. H. Ryu, S. Hong, H. Lee, *Acta Biomater.* **2015**, *27*, 101.

[7] A. Mahdavi, L. Ferreira, C. Sundback, J. W. Nichol, E. P. Chan, D. J. D. Carter, C. J. Bettinger, S. Patanavanich, L. Chignozha, E. Ben-Joseph, A. Galakatos, H. Pryor, I. Pomerantseva, P. T. Masiakos, W. Faquin, A. Zumbuehl, S. Hong, J. Borenstein, J. Vacanti, R. Langer, J. M. Karp, *Proc. Natl. Acad. Sci. USA* **2008**, *105*, 2307.

[8] a) F. Schneider, T. Fellner, J. Wilde, U. Wallrabe, *J. Micromech. Microeng.* **2008**, *18*, 065008; b) I. D. Johnston, D. K. McCluskey, C. K. L. Tan, M. C. Tracey. *J. Micromech. Microeng.* **2014**, *24*, 035017.

[9] a) A. Mata, A. J. Fleischman, S. Roy, *Biomed. Microdevices* **2005**, *7*, 281; b) M. A. Unger, H.-P. Chou, T. Thorsen T, A. Scherer, S. R. Quake, *Science* **2000**, *288*, 113; c) A. Y. Fu, H.-P. Chou, C. Spence, F. H. Arnold, S. R. Quake, *Anal. Chem.* **2002**, *74*, 2451; d) D. P. J. Cotton, A. Popel, I. M. Graz, S. P. Lacour, *J. Appl. Phys.* **2011**, *109*, 054905.

[10] L.-H. Cai, T. E. Kodger, R. E. Guerra, A. F. Pegoraro, M. Rubinstein, D. A. Weitz, *Adv. Mater.* **2015**, *27*, 5132.

[11] a) C. Li, G. Guan, R. Reif, Z. Huang, R. K. Wang, *J. R. Soc., Interface* **2012**, *9*, 831; b) C. Dagdeviren, Y. Shi, P. Joe, R. Ghaffari, G. Balooch, K. Usgaonkar, O. Gur, P. L. Tran, J. R. Crosby, M. Meyer, Y. Su, R. Chad Webb, A. S. Tedesco, M. J. Slepian, Y. Huang, J. A. Rogers, *Nat. Mater.* **2015**, *14*, 728; c) X. Liang, S. A. Boppart, *IEEE Trans. Biomed. Eng.* **2010**, *57*, 953.

[12] C. F. Carlborg, T. Haraldsson, M. Cornaglia, G. Stemme, W. van der Wijngaart, *J. Microelectromech. Syst.* **2010**, *19*, 1050.

[13] a) R. W. Style, R. Boltyanskiy, B. Allen, K. E. Jensen, H. P. Foote, J. S. Wettlaufer, E. R. Dufresne, *Nat. Phys.* **2015**, *11*, 82; b) F. Di Lorenzo, S. Seiffert, *Polym. Chem.* **2015**, *6*, 5515.

[14] Y. Menguc, Y.-L. Park, H. Pei, D. Vogt, P. M. Aubin, E. Winchell, L. Fluke, L. Stirling, R. J. Wood, C. J. Walsh, *Int. J. Robot. Res.* **2014**, *33*, 1748.

[15] M. Mikrut, *PhD Thesis*, University of Twente **2007**.

[16] a) W.-H. Yeo, Y.-S. Kim, J. Lee, A. Ameen, L. Shi, M. Li, S. Wang, R. Ma, S. H. Jin, Z. Kang, Y. Huang, J. A. Rogers, *Adv. Mater.* **2013**, *25*, 2773; b) F. Tramacere, A. Kovalev, T. Kleinteich, S. N. Gorb, B. Mazzolai, *J. R. Soc., Interface* **2014**, *11*, 20130816.

[17] B. C.-K. Tee, A. Chortos, A. Berndt, A. K. Nguyen, A. Tom, A. McGuire, Z. C. Lin, K. Tien, W.-G. Bae, H. Wang, P. Mei, H.-H. Chou, B. Cui, K. Deisseroth, T. N. Ng, Z. Bao, *Science* **2015**, *350*, 313.

[18] B. Laulicht, R. Langer, J. M. Karpa, *Proc. Natl. Acad. Sci. USA* **2012**, *109*, 18803.

[19] S. H. Jeong, K. Hjort, Z. G. Wu, *Sci. Rep.* **2015**, *5*, 8419.