Electrospraying and ultraviolet light curing of nanometer-thin polydimethylsiloxane membranes for low-voltage dielectric elastomer transducers

Bekim Osmani*[a], Tino Töpper[a], Matej Siketanc[a], Gabor Kovacs[b], and Bert Müller[a]
[a]Biomaterials Science Center, Department of Biomedical Engineering, University of Basel, 4123 Allschwil, Switzerland; [b]Swiss Federal Laboratories for Materials Science and Technology, Überlandstrasse 129, 8600 Dübendorf, Switzerland.

ABSTRACT

Dielectric elastomer transducers (DETs) have attracted interest as actuators, sensors, and even as self-sensing actuators for applications in medicine, soft robotics, and microfluidics. To reach strains of more than 10 %, they currently require operating voltages of several hundred volts. In medical applications for artificial muscles, however, their operation is limited to a very few tens of volts, which implies high permittivity materials and thin-film structures. Such micro- or nanostructures can be prepared using electro-spraying, a cost-effective technique that allows upscaling using multiple nozzles for the fabrication of silicone films down to nanometer thickness. Deposition rates of several micrometers per hour have already been reached. It has been recently demonstrated that such membranes can be fabricated by electro-spraying and subsequent ultraviolet light irradiation. Herein, we introduce a relatively fast deposition of a dimethyl silicone copolymer fluid that contains mercaptopropyl side chains in addition to the methyl groups. Its elastic modulus was tuned with the irradiation dose of the 200 W Hg-Xe lamp. We also investigated the formation of elastomer films, using polymer concentrations in ethyl acetate of 1, 2, 5 and 10 vol%. After curing, the surface roughness was measured by means of atomic force microscopy. This instrument also enabled us to determine the average elastic modulus out of, for example, 400 nanoindentation measurements, using a spherical tip with a radius of 500 nm. The elastomer films were cured for a period of less than one minute, a speed that makes it feasible to combine electro-spraying and *in situ* curing in a single process step for fabricating low-voltage, multilayer DETs.

Keywords: dielectric elastomer actuators, electro-spraying, thiol-functionalized polymers, ultraviolet light curing, nanoindentation, nanometer-thin films, atomic force microscopy.

1. INTRODUCTION

Soft dielectric elastomer transducers (DETs) are used for a growing number of applications, including tunable optics, microfluidics, soft robotics, and haptic devices [1-5]. Their main drawback is still the driving voltage of several hundred volts. In medical applications such as artificial muscles, however, their operation is limited to a very few tens of volts. Many research groups report approaches on how to reduce the operating voltage, either by increasing the dielectric constant, using fillers such as TiO_2 or $BaTiO_3$, or by reducing the thickness of the elastomeric layer [6-9]. A recent work describes the combination of a curable liquid acrylic composition and multilayer fabrication method that allows actuation without pre-stretching, though the elastomer layers were more than 35 µm thick and required several kilovolts for operation [10]. As the compressive strain s_z is inversely proportional to the square of the film thickness h_p [11]:

$$s_z = -\frac{\varepsilon_0 \varepsilon}{E}\left(\frac{U}{h_p}\right)^2, \tag{1}$$

with its relative permittivity ε, elastic modulus E, and operating voltage U, we target the fabrication of soft, micrometer-thin elastomer layers, which can be operated at a few tens of volts.

Thin polydimethylsiloxane (PDMS) films can be prepared by spin-coating, electro-spraying, or molecular beam deposition (MBD) techniques. Although the homogeneity of thermal evaporation of PDMS film is superior with respect to spin-coating or electro-spraying, its applicability for DETs is restricted. The evaporation of liquid PDMS pre-polymers is limited to oligomers at low deposition rates, due to the thermal degradation of the used polymer [12]. As shown recently, electro-spraying of vinyl-terminated PDMS with an average molecular weight of 6,000 g/mol leads initially to a partially coalescent drop-like film with an average thickness of about 150 nm [13]. The confluent layers shown here were covered only by 75 %. Even a subsequent deposition for 588 s resulted in 500 nm films and did not fully cover the substrate. In addition, long-lasting ultraviolet (UV) irradiation for around 15 minutes was necessary to cure the PDMS film at a distance as low as 2 cm. The crosslinking mechanism was based on the scissions of Si-CH_3 and –CH_2-H bonds using a deuterium broad-band UV lamp with its maximum intensity at a wavelength of 210 nm.

Another challenge for thin-film DETs is the low adhesion of the Au electrode to the PDMS membrane. It can be increased by the introduction of an adhesive layer of Cr or Ti, but this implementation increases the overall stiffness by orders of magnitude [14]. Another approach is based on oxygen plasma treatment and the functionalization of the PDMS surface using the molecular glue 3-mercaptopropyltrimethoxysilane (MPTMS) [15]. This bifunctional molecule interfaces the relatively stiff Au electrode and the relatively soft elastomer layer. Furthermore, its thiol or mercaptan head (-SH) can form covalent bonds to Au, whereas the three methoxy (-O-CH3) functional groups bind to silanized PDMS (-OH) surfaces, which are introduced via oxygen plasma or UV/ozone treatments [16].

Here, we show that by using an SH-functionalized PDMS with a low molecular weight of 3’600 g/mol, one can fabricate smooth dielectric films at one-micrometer in thickness. An adhesive layer between the electrode and the elastomer is not required, as the Au clusters form strong covalent bonds to the existing SH groups within the elastomer network. We also show than PDMS films can be cured in air, using a high-power Xe-Hg lamp, and tune the elastic modulus between 165 and 1’300 kPa in less than a minute, a speed that allows for combining electro-spraying and *in situ* UV curing in a single process step for fabricating low-voltage, multilayer DETs.

2. EXPERIMENTAL

2.1 Preparation of GP367 silicone films

The equipment for the fabrication of the PDMS films is shown in Figure 1. A dimethyl silicone copolymer fluid, containing mercaptopropyl (-SH) side groups with an average molecular weight of 3’600 g/mol (GP-367, Genesee Polymers Corporation, USA), was dissolved in ethyl acetate (laboratory reagent grade, Fisher Scientific, Loughborough, UK) to obtain 1 %, 2 %, 5 %, and 10 vol% spray solutions. A 3 mL glass syringe with a metallic Luer-lock (Eternal-Matic, Sanitex, HUBERLAB, Aesch, Switzerland) was used to introduce the spray solutions into the electro-spray chamber through a custom-built PEEK flange and a stainless steel needle (25 ga, 20 mm, pst 3, Hamilton, Bonaduz, Switzerland) with an inner diameter of 260 µm. The syringe pump (Aladdin AL1600-220Z, World Precision Instruments Germany GmbH, Berlin, Germany) was set to a constant flow rate of 10 µl/min. A rectangular alternating voltage signal at a frequency of 1 Hz (Tektronix AFG3021C, Linktronix AG, Thalwil, Switzerland) was amplified to ±1.5 kV using a high-voltage power amplifier (TREK 10/40A, TREK INC., Lockport, NY, USA). The optimal gap between the nozzle and the 2-inch Si substrate (2P0/1-10/ 279 ± 25, Siegert Wafer, Aachen, Germany) was found to be 50 mm. Post UV curing was performed with a 200 W Hg-Xe UV lamp (super-quiet mercury-xenon lamp L2423, Hamamatsu Photonics K.K., Hamamatsu, Japan) at a distance of 50 cm.

The built-in lens and mirror adjustments allowed for broadening the beam and irradiating the entire 2-inch specimen in one shot. To improve the homogeneity of the UV-cured films, the samples were irradiated on a rotating stage at a speed of 60 rpm. All experiments were carried out in ambient conditions.

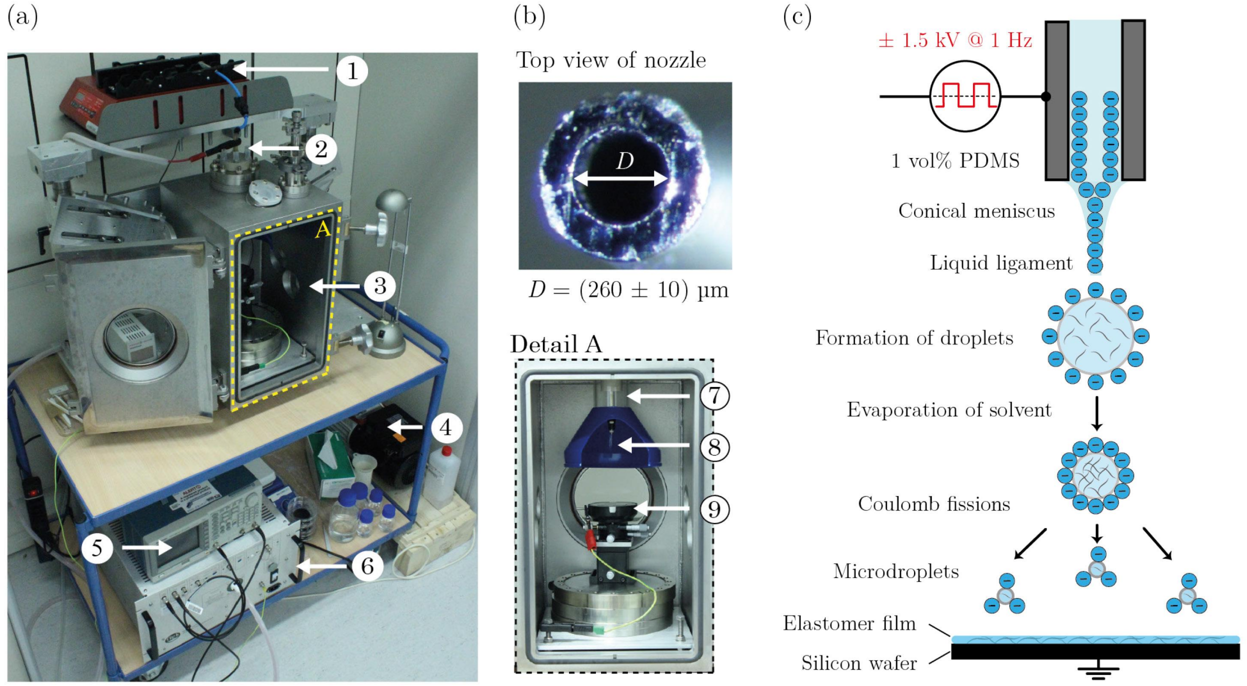


Figure 1. Electro-spraying of PDMS films. (**a**) Photograph of the experimental setup consisting of: (1) Syringe pump, (2) DN63 PEEK flange for the metallic nozzle feed-through, (3) Floatable vacuum chamber to guarantee well-defined conditions, (4) Rotary vane pump, (5) Function generator, (6) High-voltage power amplifier, (7) Plexiglas tube, (8) Stainless steel nozzle, and (9) Si wafer; (**b**) Optical micrograph of the used nozzle; (**c**) Scheme representing the formation of the conical meniscus and the droplet breakup into finer droplets. The repulsion forces caused the fine droplets to self-disperse.

2.2 Morphology of the PDMS films

Non-contact measurements on crosslinked GP-367 films were performed with a Keyence confocal 3D laser microscope (Keyence VK-X200, Keyence International, Belgium), using the supplier's VK Viewer acquisition software. Scans were taking using a 20× objective at a pixel resolution of 690 nm in the two lateral directions and a pitch of 100 nm in the third direction.

2.3 Thickness measurements and surface scans using AFM

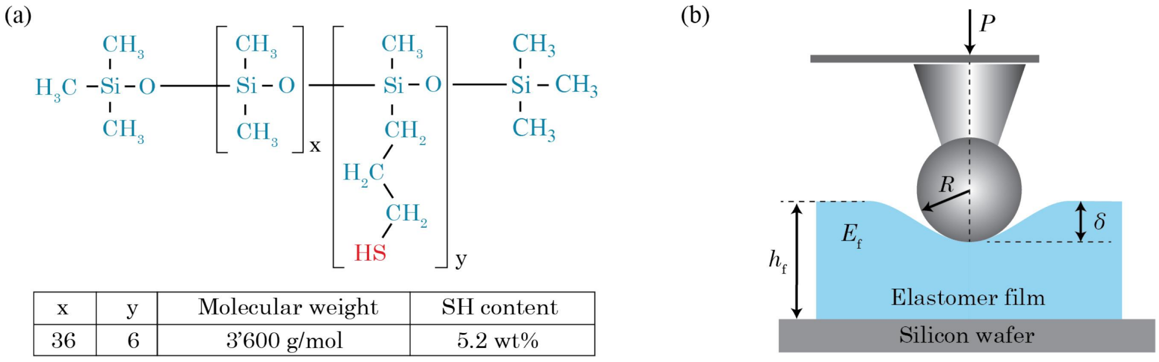
AFM scans in tapping mode were performed on manually induced scratches (Budget Sensors Tap190-G, Nano and More GmbH, Wetzler, Germany) to determine the film thickness. The vibration amplitude was set to 4 V, using a set point of 60 % on a FlexAFM system (Nanosurf AG, Liestal, Switzerland). For the mean surface roughness on 2 × 2 μm^2 spots, the vibration amplitude was set to 900 mV and the set point to 90 %.

2.4 Nano-indentations on PDMS films

AFM nanoindentations (FlexAFM C3000, Nanosurf AG, Switzerland) were applied on 10 × 10 μm^2 spots using an AFM cantilever with a spherical tip ($R = (490 \pm 5)$ nm, B500 FMR, Nanotools GmbH, Germany), as shown in Figure 2. The spring constant $k = (2.6 \pm 0.1)$ Nm of the AFM cantilever was determined using the Sader method [17]. Each spot was partitioned into 400 subdomains, serving as a nanoindentation site. The elastic modulus for each subdomain was calculated from the force distance curves. It has been reported that the nanoindentation measurements using large spheres on soft polymers are affected significantly by adhesion forces F_{adh} [18]. The Johnson-Kendall-Roberts (JKR) contact model takes into account F_{adh} and can be described by the following equation [19]:

$$E_f = \sqrt{\frac{S^3(1-\upsilon_f^2)^2}{6R}\left(\frac{1}{P + 2F_{adh} + 2F_{adh}\sqrt{\left(\frac{P}{F_{adh}} + 1\right)}}\right)}\,, \tag{2}$$

where P is the applied load, R is the radius of the spherical tip, υ_f is the Poisson ratio of the silicone film, and F_{adh} is the maximum adhesive pull-off force between the tip and the sample during unloading. Stiffness S represents the slope of the unloading curve. The JKR model is implemented in FLEXANA® software (FLEXANA, Nanosurf AG, Switzerland), offering a fully automated post-processing and analysis of the data.



x	y	Molecular weight	SH content
36	6	3'600 g/mol	5.2 wt%

Figure 2. (**a**) Structure of dimethyl silicone copolymer fluid GP367 containing mercaptopropyl SH-side chains in addition to the conventional CH_3-methyl groups. (**b**) Scheme of the AFM nanoindentation using a spherical tip with a radius $R = (490 \pm 5)$ nm. The elastic modulus E_f of the silicone film was calculated from the force distance curves and the applied load using the Johnson-Kendall-Roberts contact model.

2.5 Evaporation of the Au electrode

The 15 nm-thin Au electrodes were thermally evaporated in ultra-high-vacuum conditions at a residual pressure of 10^{-7} mbar. High-temperature effusion cells (HTEZ, Dr. Eberl MBE Komponenten GmbH, Weil der Stadt, Germany) with 10 cm^3 PBN crucibles served as molecular beam sources. The evaporation temperature was set to 1'440 °C, corresponding to a deposition rate of 0.5 nm per minute. The substrate was mounted at a distance of 40 cm away from the crucibles and rotated at a speed of 10 rpm to obtain a homogenous film and to minimize the thermal impact of the source on the underlying PDMS film.

2.6 Contact angle measurements

The specimens were placed on the stage of a contact angle goniometry device (OCA 15 EC, DataPhysics Instr. GmbH, Germany), supplied with LED illumination and a USB camera for video-recording. High-purity water droplets at a volume of 5 μL (Elga, Purelab UHQ II, UK) were dispensed by a glass syringe and taken up on the surface of the polymer by raising the stage upwards. The SCA 20 software (DataPhysics Instr. GmbH) was used to record and analyze the data.

3. RESULTS AND DISCUSSION

3.1 From islanding to confluent elastomer films

First, we tuned the electro-spray's deposition parameters, namely the applied alternating high voltage, its frequency, the flow rate through the nozzle, and the distance between the nozzle and the substrate, to create a stable and reproducible mono-dispersed, single-cone jet.

In a next step, we varied polymer concentration from 1 to 10 vol% and investigated film formation with respect to deposition time, as displayed in Figure 3. After depositing for a duration of 10 s, we observed islands but no confluent film for the polymer concentrations used herein. The island heights corresponded to about 30 nm for the 1 vol% and increased to 750 nm for the 10 vol% solution. Nevertheless, the islands were rather flat and exhibited a diameter-to-height ratio of about 500.

For the 1 vol% solution, the films became percolated after a deposition period of 50 s. The average film thickness reached (220 ± 70) nm after a deposition period of 75 s and resulted in a clearly present irregular thickness, even for deposition periods exceeding 100 s. These thickness modulations reflect the still micrometer-sized droplets landing on the substrate. Note that the islands were high with respect to film thickness. The images represented in Figure 3 further indicate that an increase in concentration to 10 vol% resulted in an earlier transition to the percolation threshold and to confluent films.

Confluent films are required in the preparation of functional elastomer films in DETs. Therefore, the minimal deposition time is 75 s (with an exception at 10 vol%, which equates to 50 s). Related average film thickness, however, depends linearly on polymer concentration, as indicated by the numbers above the images. These numbers are red-colored for the films that only partially cover the substrate. Numbers in black mark the layers that cover the entire substrate and are suitable for building DET structures. Our experiments clearly demonstrate that using a polymer concentration of 5 or 10 vol% only allows for fabricating micrometer-thick films, whereas lower concentrations enable us to build nanometer-thin confluent elastomer films.

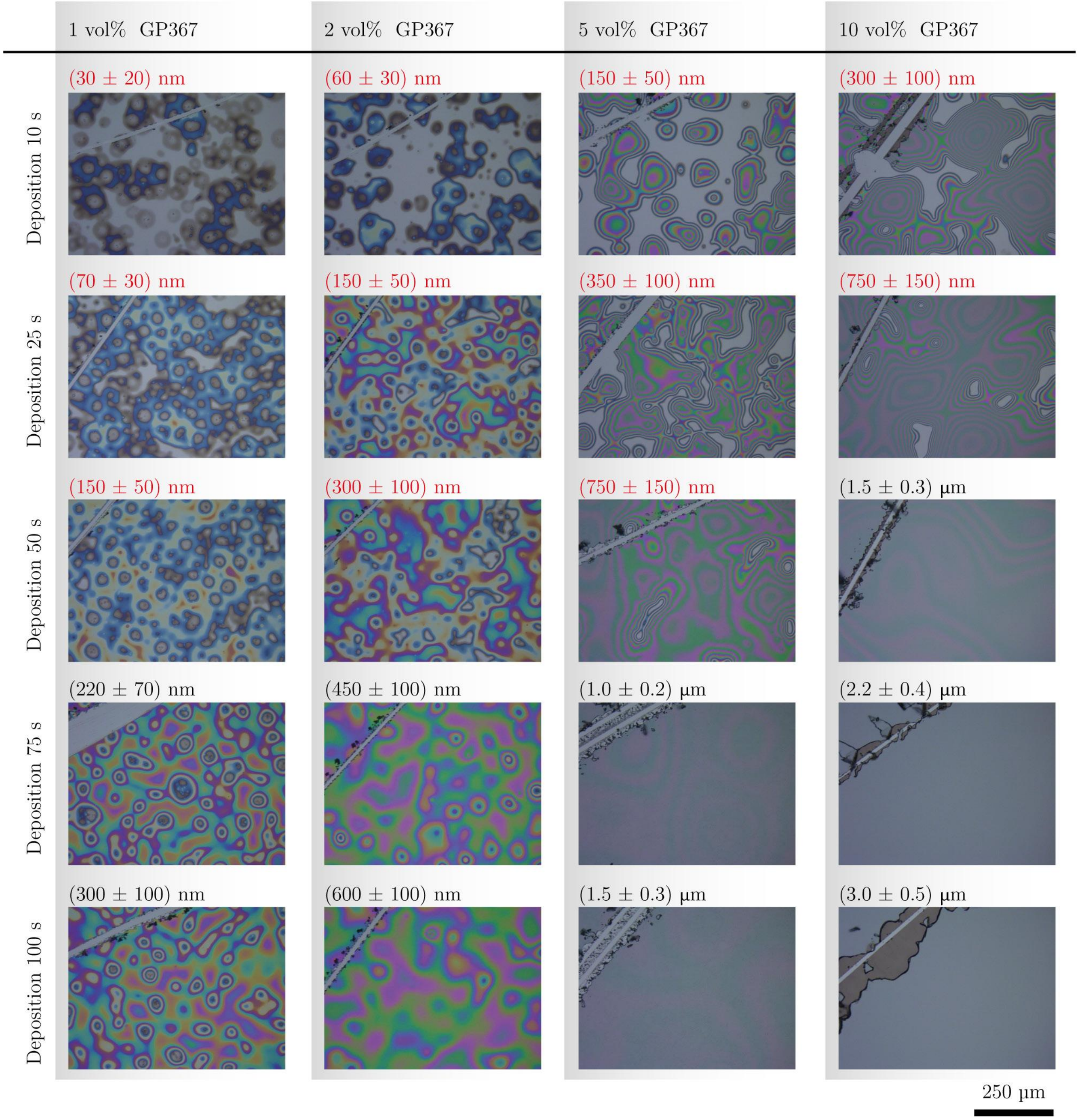


Figure 3. Optical micrographs of AC electro-sprayed and UV-cured PDMS films on Si wafers. PDMS was dissolved in ethyl acetate to realize concentrations between 1 and 10 vol%. The PDMS ethyl acetate solution was electro-sprayed for deposition times ranging from 10 to 100 s. More or less interconnected islands, which do not fully cover the substrate, are formed during the early stages of deposition and marked in red color. They will result in a short-circuit fault in the DET structures. For 1 and 2 vol% concentrations, confluent films were found even at nanometer thicknesses. These films, however, were rough. Smoother PDMS films were found for a thickness above 1 µm. Note: The scratches were built on purpose.

The nanometer-thin films, however, are comparably rough and only qualify partially for the preparation of DETs. The film roughness reflects the micrometer-sized droplets. For a 5 vol% concentration, the average radius of the micro-droplets was (7 ± 2) µm, which could be reduced to (3 ± 1) µm for the 1 vol% solution. For the micrometer-thick films, however, the roughness is comparably small and the impact of droplets less obvious.

As previously reported [20], voltages of ±5 kV usually lead to unstable, multi-cone jets and therefore to local variances in deposition rate and film thickness. Flow rate and PDMS concentration determine the mean droplet size, whereas the other operation parameters, including applied voltage, nozzle diameter, and electrical conductivity of the solvent, seem to have a modest effect [21].

3.2 Thermal treatment of electro-sprayed elastomer films

The molecular weight of GP367 corresponds to 3’600 g/mol and is therefore relatively low. Thus, we investigated the influence of thermal processing on film morphology. For this purpose, a solution with an oligomer concentration of 5 vol% was sprayed over a period of 25 s to obtain a PDMS film that would partially cover the substrate. In the next step, these liquid-like films, with an average thickness in the sub-micrometer range, were processed at temperatures between 25 and 150 °C for a period of ten minutes.

The resulting morphology is best represented using the optical micrographs in Figure 4. Up to an annealing temperature of 100 °C, part of the Si substrate is visible. Annealing to 150 °C results in a film that covers the entire substrate. This film, at a thickness of (500 ± 150) nm, however, still exhibits height modulations of more than 100 nm, as identified by the interference pattern on the optical micrograph.

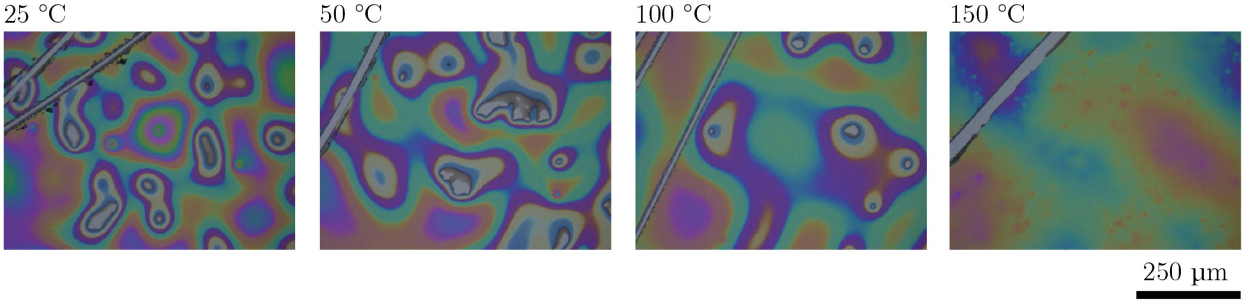


Figure 4. Optical micrographs of thermally treated liquid-like PDMS films at a mean thickness of (550 ± 150) nm. The holes were filled after thermal treatment at a temperature of 150 °C for ten minutes. Subsequent to the thermal treatment and cooling down to room temperature, the PDMS layers were UV-cured. Note: The scratches were built on purpose.

3.3 Tuning the elastic modulus via UV irradiation

Figure 5(a) shows the increase in the elastic modulus E of the electro-sprayed films as a function of irradiation time. The experimentally derived values are also listed in Table 1. The prolongation of irradiation time from 30 to 120 s results in elastic moduli increasing from 165 to 1’300 kPa.

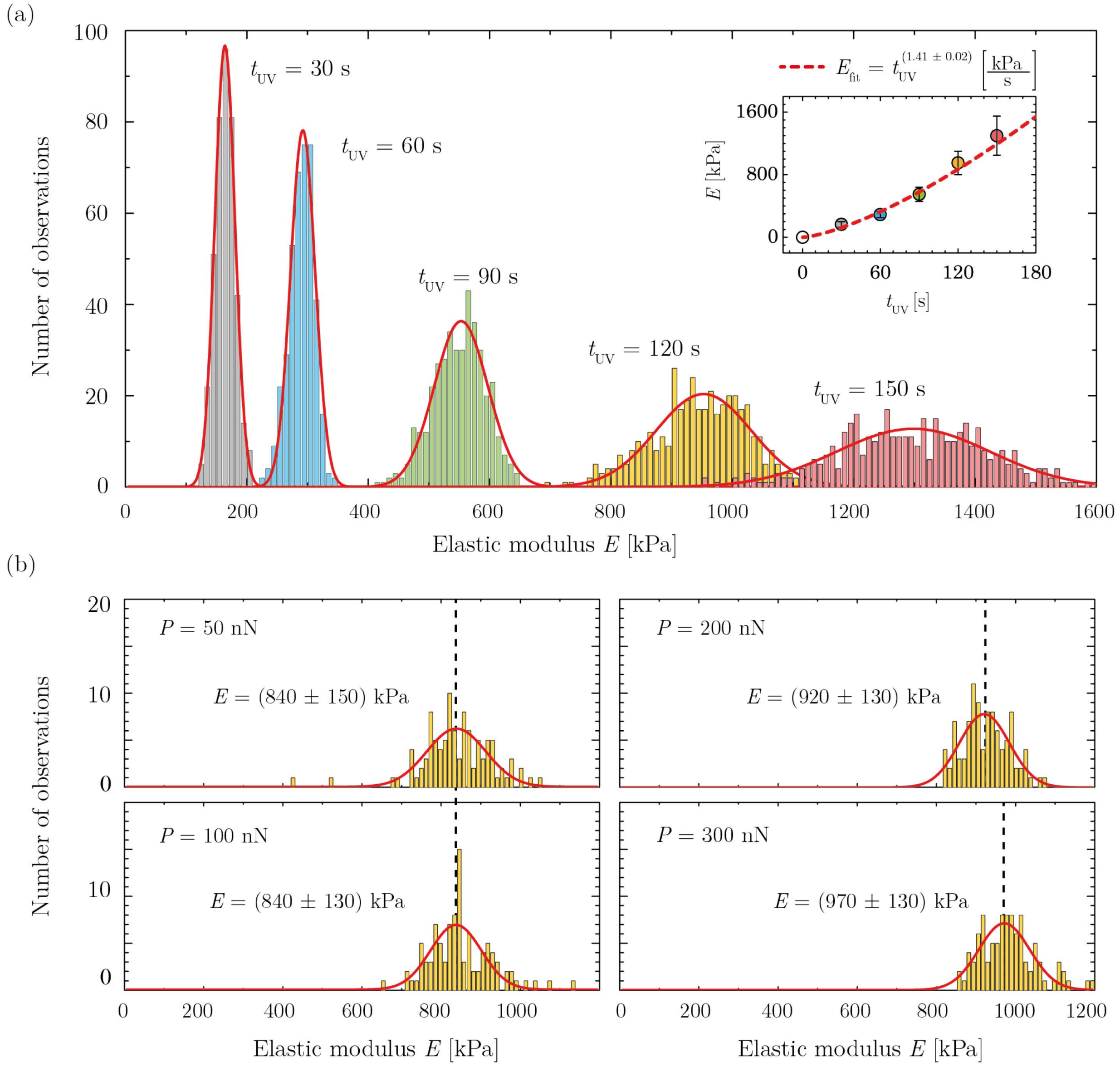


Figure 5. (**a**) UV-curing of GP367 silicone films. The elastic modulus E can be tuned with the irradiation dose using a 200 W Hg-Xe lamp. (**b**) Elastic modulus of a UV-cured silicone film, calculated for selected loads P and related nanoindentation depths $\boldsymbol{\delta}$. A six-fold higher load $P = 300$ nN with an average indentation depth of (420 ± 50) nm reveals that the silicone film is uniformly cured over the whole thickness. The thickness of the investigated sample was (2.1 ± 0.2) µm.

Table 1. Elastic moduli and water contact angles of the UV-cured PDMS films for selected irradiation times.

UV irradiation time [s]	**Elastic modulus *E*** [kPa]	**Shear modulus *G*** [kPa]	**Water contact angle** [deg]
30	165 ± 35	55 ± 12	111 ± 1
60	290 ± 40	100 ± 14	107 ± 1
90	550 ± 90	180 ± 30	106 ± 1
120	950 ± 150	950 ± 50	105 ± 1
150	1300 ± 250	430 ± 84	103 ± 1

The crosslinking mechanism responsible for the stiffness increase, however, was not completely identified [22]. It is assumed that thiol radicals, which are formed by UV irradiation, play a crucial role in the photo-polymerization of thiol-ene polymers. These radical species are also probably responsible for the crosslinking of the polymer chains into a three-dimensional network. It has been reported that under ambient conditions, oxygen is incorporated into the polymer network [23] and reacts with carbon radicals to form peroxyl radicals. Indeed, we observed smaller water contact angles for the longer irradiation times shown in Table 1. The longer the sample has been irradiated, the more peroxyl radicals may form and the polymer membrane becomes more and more hydrophilic.

The data shown in Figure 5(b) indicate that the PDMS films are uniformly cured. The calculated elastic modulus does not change significantly for the selected loads and resulting indentation depths, though one recognizes a slight increase in the highest selected load. Presumably, this marginal increase relates to substrate effects. Here, the average indentation depths were already (420 ± 50) nm. A possible gradient in crosslinking, with a stiff, skin-like layer on top and a soft/liquid bulk, as reported for a UV-cured, vinyl-terminated PDMS using a deuterium UV lamp, can be excluded [24].

3.4 Increased interaction of the Au electrode with the PDMS membrane

The limited adhesion of Au to commercially available Sylgard® 184-PDMS is well known [25]. A peel-off test, as displayed in Figure 6, clearly demonstrates that the Au electrode interacts only with the SH-functionalized PDMS network, whereas the Au electrode on Sylgard® 184 peeled off easily. The increased adhesion of Au results from the thiol groups and formed disulfide bonds. Thiols and disulfide bonds interact strongly with Au [22].

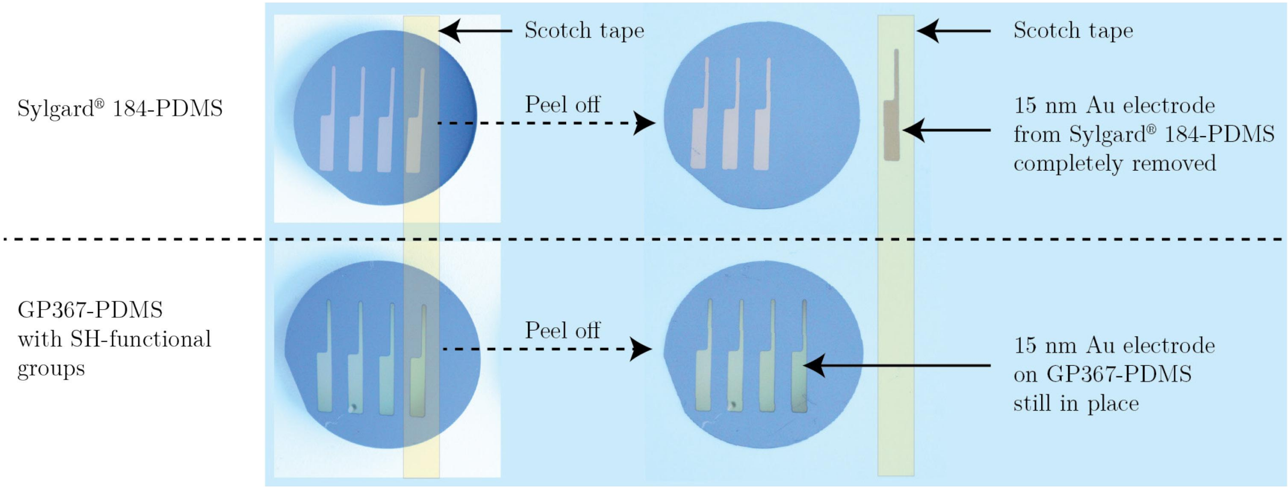


Figure 6. Adhesion of an Au electrode to thermally-cured Sylgard® 184-PDMS and UV-cured GP367-PDMS. A peel-off test with Scotch tape reveals the limited adhesion of Au on PDMS to be fixed using SH-functional groups.

Au films on pure PDMS most likely follow the Volmer-Weber growth mode, leading to a cluster-like Au electrode [25]. Au clusters with a nominal size of (40 ± 20) nm were observed on both PDMS films, as shown in Figure 7. However, the root-mean-square roughness (RMS) of Au on GP367 was significantly smaller than for Au on Sylgard® 184, showing values of (3.8 ± 0.5) and (5.3 ± 0.5) nm, respectively. The initial RMS value of the sprayed and UV-cured PDMS film was determined at only (0.3 ± 0.1) nm.

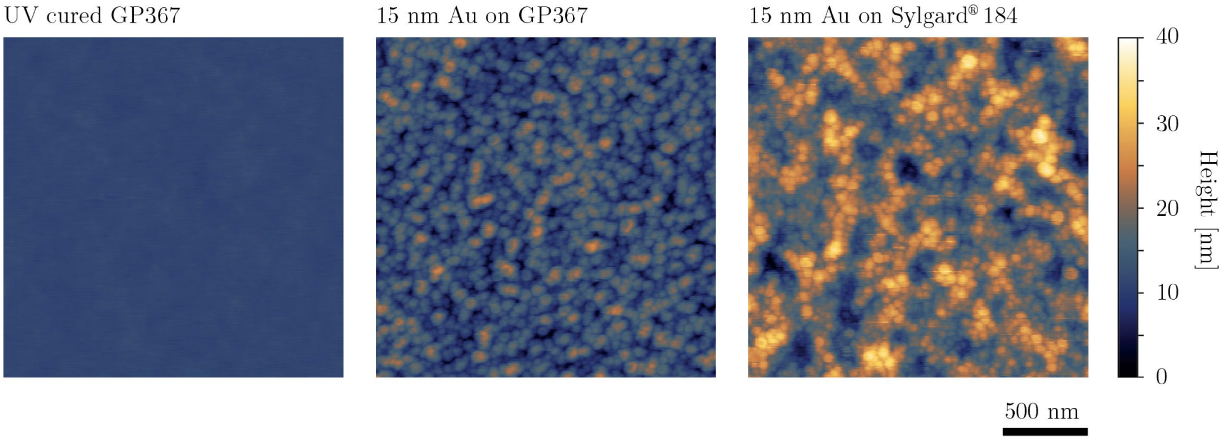


Figure 7. The three AFM scans show 2 × 2 µm² areas of a (left) UV-cured GP367 PDMS film, (middle) a 15 nm-thin Au electrode on GP367, and (right) a 15 nm Au electrode on Sylgard® 184 PDMS film. The initial root-mean-roughness (RMS) value of (0.3 ± 0.1) nm for the UV-cured GP367 increased to (3.8 ± 0.5) nm after the evaporation of 15 nm Au at a distance of 40 cm. The RMS value for Au on Sylgard® 184, at (5.3 ± 0.5) nm, was even higher.

4. CONCLUSIONS

Using SH-functionalized PDMS, soft and smooth elastomer films with thicknesses of only 1 µm can be fabricated. The elastic modulus of these layers can be tuned between 165 and 1’300 kPa. Compared to previous studies, the stable single-cone jet enabled us to produce a film that fully covered the substrate in less than one minute [13]. Subsequent UV-polymerization, without applying any photoinitiators, allowed us to form a homogenously crosslinked thin film. In addition, the adhesive layer between the electrode and elastomer was not required, as the Au nanoclusters form covalent bonds to the SH groups on the elastomeric network. Confluent PDMS layers were obtained even at a thickness as thin as (220 ± 70) nm using 1 vol% solutions. Films thicker than 1 µm are required to obtain smooth films suitable for the fabrication of low-voltage DETs. Selecting the experimentally proven values for the dielectric layer with $E = 290$ kPa, $\boldsymbol{\varepsilon} = 2.7$, $h_{\mathrm{p}} = 1$ µm and a voltage of $U = 42$ V, the calculated compressive strain s_z corresponded to about 15 %. In the next step, a multiple nozzle arrangement and a rolling endless sheet substrate should be employed to increase film homogeneity further. With thermal treatment and *in situ* UV-curing, this process will enable the fabrication of low-voltage, multilayer DETs.

ACKNOWLEDGMENTS

The financial support of the nano-tera.ch initiative, project SmartSphincter, as well as the Swiss Nanoscience Institute (SNI) for the financial contribution to the AFM are gratefully acknowledged. The authors wish to thank Irene Piskoti, from Genesee Polymers Corporation, Burton MI, USA, for kindly providing one pint of GP-367 at no charge. The authors also wish to thank to Monica Schönenberger, from the Nanotech Service Lab at the University of Basel, for her assistance with the 3D laser microscope.

REFERENCES

[1] Anderson, I. A., Gisby, T. A., McKay, T. G. et al., “Multi-functional dielectric elastomer artificial muscles for soft and smart machines,” *J. Appl. Phys.* **112**(4), 041101 (2012).

[2] Brochu, P., and Pei, Q., “Advances in dielectric elastomers for actuators and artificial muscles,” *Macromol. Rapid. Commun.* **31**(1), 10-36 (2010).

[3] O’Halloran, A., O’Malley, F., and McHugh, P., “A review on dielectric elastomer actuators, technology, applications, and challenges,” *J. Appl. Phys.* **104**(7), 071101 (2008).

[4] Rosset, S., and Shea, H. R., “Small, fast, and tough: Shrinking down integrated elastomer transducers,” *Appl. Phys. Rev.* **3**(3), 031105 (2016).

[5] Sia, S. K., and Whitesides, G. M., “Microfluidic devices fabricated in poly(dimethylsiloxane) for biological studies,” *Electrophoresis* **24**(21), 3563-3576 (2003).

[6] Molberg, M., Crespy, D., Rupper, P. et al., “High breakdown field dielectric elastomer actuators using encapsulated polyaniline as high dielectric constant filler,” *Adv. Funct. Mater.* **20**(19), 3280-3291 (2010).

[7] Poulin, A., Rosset, S., and Shea, H. R., “Printing low-voltage dielectric elastomer actuators,” *Appl. Phys. Lett.* **107**(24), 244104 (2015).

[8] Töpper, T., Weiss, F. M., Osmani, B. et al., “Siloxane-based thin films for biomimetic low-voltage dielectric actuators,” *Sens. Actuators A* **233**, 32-41 (2015).

[9] Weiss, F. M., Madsen, F. B., Töpper, T. et al., “Molecular beam deposition of high-permittivity polydimethylsiloxane for nanometer-thin elastomer films in dielectric actuators,” *Mater. Des.* **105**, 106-113 (2016).

[10] Duduta, M., Wood, R. J., and Clarke, D. R., “Multilayer dielectric elastomers for fast, programmable actuation without prestretch,” *Adv. Mater.* **28**(36), 8058-8063 (2016).

[11] Pelrine, R., Kornbluh, R., Joseph, J. et al., “High-field deformation of elastomeric dielectrics for actuators,” *Mater. Sci. Eng. C* **11**(2), 89-100 (2000).

[12] Töpper, T., Lörcher, S., Weiss, F. et al., “Tailoring the mass distribution and functional group density of dimethylsiloxane-based films by thermal evaporation,” *APL Mater.* **4**(5), 56101 (2016).

[13] Weiss, F. M., Töpper, T., Osmani, B. et al., “Thin film formation and morphology of electrosprayed polydimethylsiloxane,” *Langmuir* **32**, 3276-3283 (2016).

[14] Loo, Y. L., Willett, R. L., Baldwin, K. W. et al., “Additive, nanoscale patterning of metal films with a stamp and a surface chemistry mediated transfer process: Applications in plastic electronics,” *Appl. Phys. Lett.* **81**(3), 562-564 (2002).

[15] Osmani, B., Deyhle, H., Weiss, F. M. et al., “Morphology and conductivity of Au electrodes on polydimethylsiloxane using (3-mercaptopropyl) trimethoxysilane (MPTMS) as an adhesion promoter,” *Proc. SPIE* **9798**, 979822 (2016).

[16] Byun, I., Coleman, A. W., and Kim, B., “Transfer of thin Au films to polydimethylsiloxane (PDMS) with reliable bonding using (3-mercaptopropyl) trimethoxysilane (MPTMS) as a molecular adhesive,” *J. Micromech. Microeng.* **23**(8), 085016 (2013).

[17] John, E. S., James, W. M. C., and Paul, M., “Calibration of rectangular atomic force microscope cantilevers,” *Rev. Sci. Instrum.* **70**(10), 3967-3969 (1999).

[18] Gupta, S., Carrillo, F., Li, C. et al., “Adhesive forces significantly affect elastic modulus determination of soft polymeric materials in nanoindentation,” *Mater. Lett.* **61**(2), 448-451 (2007).

[19] Carrillo, F., Gupta, S., Balooch, M. et al., “Nanoindentation of olydimethylsiloxane elastomers: Effect of crosslinking, work of adhesion, and fluid environment on elastic modulus,” *J. Mater. Res.* **20**(10), 2820-2830 (2005).

[20] Weiss, F. M., Töpper, T., Osmani, B. et al., “Electrospraying nanometer-thin elastomer films for low-voltage dielectric actuators,” *Adv. Electron. Mater.* **1500476**, (2016).

[21] Tang, K., and Gomez, A., “Monodisperse electrosprays of low electric conductivity liquids in the cone-jet mode,” *J. Colloid Interface Sci.* **184**(2), 500-511 (1996).

[22] Grönbeck, H., Curioni, A., and Andreoni, W., “Thiols and disulfides on the Au (111) surface: the headgroup−gold interaction,” *J. Am. Chem. Soc.* **122**(16), 3839-3842 (2000).

[23] Cramer, N. B., Scott, J. P., and Bowman, C. N., “Photopolymerizations of thiol−ene polymers without photoinitiators,” *Macromolecules* **35**(14), 5361-5365 (2002).

[24] Töpper, T., Wohlfender, F., Weiss, F. M. et al., “Characterization of ultraviolet light cured polydimethylsiloxane films for low-voltage, dielectric elastomer actuators,” *Proc. SPIE* **9798**, 979821 (2016).

[25] Graudejus, O., Görrn, P., and Wagner, S., “Controlling the morphology of gold films on poly(dimethylsiloxane),” *ACS Appl. Mater. Interfaces* **2**(7), 1927-1933 (2010).

---

*Further author information: (send correspondence to B.O.)