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Irreversible bonding of polyimide and polydimethylsiloxane (PDMS) based on a thiol-epoxy click reaction

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Abstract

Polyimide is one of the most popular substrate materials for the microfabrication of flexible electronics, while polydimethylsiloxane (PDMS) is the most widely used stretchable substrate/encapsulant material. These two polymers are essential in fabricating devices for microfluidics, bioelectronics, and the internet of things; bonding these materials together is a crucial challenge. In this work, we employ click chemistry at room temperature to irreversibly bond polyimide and PDMS through thiol-epoxy bonds using two different methods. In the first method, we functionalize the surfaces of the PDMS and polyimide substrates with mercaptosilanes and epoxysilanes, respectively, for the formation of a thiol-epoxy bond in the click reaction. In the second method, we functionalize one or both surfaces with mercaptosilane and introduce an epoxy adhesive layer between the two surfaces. When the surfaces are bonded using the epoxy adhesive without any surface functionalization, an extremely small peel strength ($<0.01 \text{ N mm}^{-1}$) is measured with a peel test, and adhesive failure occurs at the PDMS surface. With surface functionalization, however, remarkably higher peel strengths of $\sim 0.2 \text{ N mm}^{-1}$ (method 1) and $>0.3 \text{ N mm}^{-1}$ (method 2) are observed, and failure occurs by tearing of the PDMS layer. We envision that the novel processing route employing click chemistry can be utilized in various cases of stretchable and flexible device fabrication.


Keywords: microfabrication, polymer bonding, PDMS, Kapton, adhesion, peel-testing, thiol-epoxy click chemistry

(Some figures may appear in colour only in the online journal)

Introduction

Polyimides (including DuPont's commercial film, 'Kapton') are popular substrate materials for flexible electronic devices, including antennas [1], organic transistors [2], organic light-emitting diodes [3], and organic solar cells [4]. Polyimides are thermoset polymers with good chemical resistance and dimensional stability over a wide range of temperatures (with demonstrated applications between $-269 \text{ }^\circ\text{C}$ and $400 \text{ }^\circ\text{C}$), and

thus can withstand harsh processing conditions that often occurs in microfabrication [5, 6]. In terms of mechanical deformation, polyimides are capable of withstanding bending and twisting modes, but *are not intrinsically stretchable or compressible*. Strain relief can be introduced into relatively non-stretchable materials (including polyimides [7–9], rigid nanocomposites [10] or zinc oxide [11]) by patterning these substrates or layers with deformable shapes (such as periodic voids, meandering shapes, or wrinkled structures). Various types of polyimide have been shown to be biocompatible, exhibiting low cytotoxicity and limited haemolysis in *in vitro* studies [12], and exhibiting limited changes in properties during incubation under physiological conditions at elevated

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temperatures (60 °C) for up to 20 months [13]. Polyimides are strong candidates for use as substrates in a variety of biomedical devices (both implanted and externally-mounted), such as active and passive microelectrode arrays for recording of neural signals from the surface of the cortex [7, 14, 15], pressure sensors [16], strain sensors [17], wearable electrodes for relaying signals to and from the body [18], and skin-mountable thermoelectric power generators [5].

To insulate and protect these electronic devices, they are often encapsulated with polydimethylsiloxane (PDMS) [18]. PDMS is a biocompatible silicone polymer with high gas permeability that can be in contact with skin for a long time [18], forming a favorable interface between the device and the surface of the body. Another important feature of PDMS is its rubbery mechanical behavior, i.e. *stretchability* [19, 20]. PDMS can be made exceptionally soft and its dimension can adapt to mechanical changes in its surrounding environment in a resilient way, thus it is an ideal substrate material for devices in contact with parts of the body that are soft and under motion. In devices that combine these two materials, the polyimide acts as the substrate for the electronic components, while the PDMS provides a soft flexible interface with the tissue of interest [21]. The stretchability of these devices is an important mechanical feature for various futuristic electronics, including internet of things (IoT) [22].

It can be challenging to bond PDMS to other materials (including polyimides) due to the fact that PDMS is both chemically inert and highly hydrophobic. One strategy that has been successfully demonstrated for PDMS–PDMS bonding is to leverage the adhesiveness of uncured PDMS, which can be applied as a glue between cured layers [23]. In a related approach, uncured PDMS can be dispensed onto a polyimide substrate (on which the electronic components have been patterned) and then cured [9]. Alternatively, cured sheets of oxygen plasma-treated PDMS may be bonded *around* a patterned Kapton device, creating an encapsulated device sandwiched between two PDMS sheets in which the PDMS–PDMS bonding minimizes the tendency of the substrates to delaminate or separate upon deformation [18].

To bond or laminate *cured* sheets of PDMS to a polyimide substrate, some form of chemical modification is usually required. Adhesion promoters can be introduced directly into the PDMS during the curing process [24], or new chemical functional groups may be introduced on the surface of fully cured PDMS in combination with an O₂ plasma or a corona treatment [25]. A thin Cr/SiO₂ bilayer (3 nm/33 nm) has been evaporated onto Kapton as an adhesion layer prior to joining the material to plasma-treated PDMS in a transfer-printing process [26]. However, this process requires a vacuum deposition system, which significantly increases production cost and time.

A number of functional groups have been deposited from solution or vapor phases in the effort to create irreversible bonds between cured PDMS sheets and various types of plastics. Vlachopoulou *et al* modified PMMA with

(3-aminopropyl)trimethoxysilane (APTES), then plasma treated both the PDMS and the modified PMMA and pressed the substrates together at 80 °C to form a strong bond (they hypothesized that the second plasma treatment removed the amine groups from the silane, and that the bond formed was Si–O–Si) [27]. A variety of plasma-treated thermoplastics have been bonded to plasma-treated PDMS by applying APTES to one of the two plasma-treated surfaces as a linker and then bringing the two surfaces into conformal contact at room temperature [28]. Amine-PDMS linkers have also been applied to the surfaces of corona-treated plastics (including polyimides) to form the basis of strong bonds with corona-treated PDMS; urethane bonds form at the plastic substrate, and Si–O–Si bonds at the surface of the PDMS [25]. Tang *et al* demonstrated a chemical gluing strategy to bond PDMS to a variety of non-silicone polymers based on the reaction between an epoxy-silane applied to one substrate and the amine-silane applied to another; this process did not include an oxygen plasma step [29]. This mechanism has also been applied using a silicone adhesive tape as an intermediate layer to bond PDMS to PMMA (with oxygen plasma), where the PMMA was functionalized with (3-aminopropyl)trimethoxysilane and the silicone epoxy was functionalized with (3-glycidoxypropyl)trimethoxysilane [30].

An unutilized, but a promising route to promote chemical bonding to PDMS surface is epoxy-thiol click chemistry [31]. The thiol-epoxy reaction has a high reaction efficiency even at or below room temperature, and can thus be used to achieve bonding between two surfaces in mild processing conditions. In addition, the reaction generates hydroxyl groups as by-products [31], opening the possibility for subsequent functionalization of the modified surface.

In this work, we describe a simple and effective technique to form an irreversible bond between cured PDMS sheets and Kapton films by utilizing the thiol-epoxy bonding as an adhesion promoter. Kapton—specifically poly-oxydiphenylene-pyromellitimide (Dupont, Wilmington DE)—is a well-known and widely used commercial polyimide. Two main bonding procedures are explored. In the first method, each surface is exposed to UV ozone and then functionalized with a particular chemical group; the surfaces are then pressed together overnight at room temperature. Within this method, either aminosilanes or mercaptosilanes are applied to Kapton for bonding with epoxysilane-functionalized PDMS; this results in the formation of either amino-epoxy bonds or thiol-epoxy bonds. In the second method, each substrate (Kapton and PDMS) is exposed to UV ozone and functionalized with mercaptosilane; these substrates are sandwiched on either side of a thin layer of epoxy (applied to one of the substrates). In each method, the parameters by which the substrates are functionalized are varied, for example both liquid and vapor deposition of the mercaptosilane solution is tested to see which deposition method offered better adhesion. The effect of treating one of the substrates only is also explored. In all instances, bonding strength is characterized using a 90° peel test.

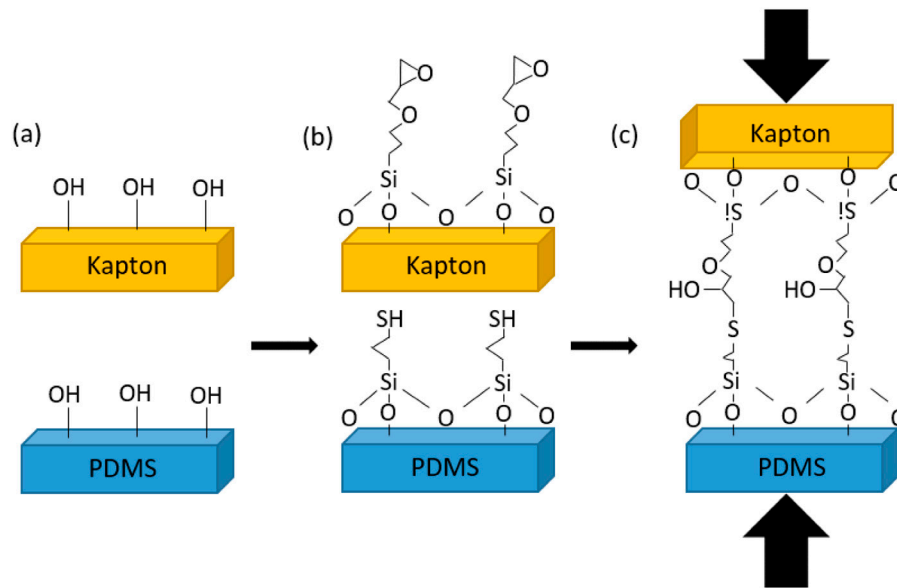


Figure 1. Method 1: schematic of surface treatment of Kapton and PDMS for the MPTMS/GPTMS bonding procedure: (a) substrate hydroxylation by UV ozone treatment for 10 min, (b) attachment of mercaptosilane and epoxysilane by liquid deposition for 1 h, (c) contact of the two substrates overnight under 30 kPa at room temperature. The resulting bonds illustrated are based on [31]. In a modified version of this procedure (unsuccessful), the PDMS substrate is functionalized with aminosilane rather than mercaptosilane.

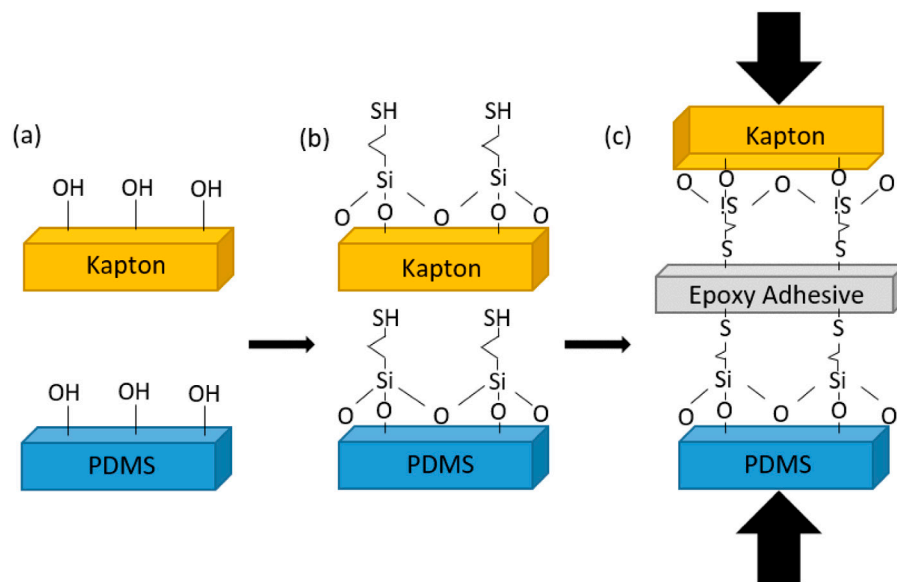


Figure 2. Method 2: schematic of surface treatment of Kapton and PDMS for epoxy adhesive bonding procedure: (a) substrate hydroxylation by UV ozone treatment for 10 min, (b) attachment of mercaptosilane group by liquid or vapor deposition for 1 h, (c) application of epoxy adhesive and contact of the two substrates overnight under 30 kPa at room temperature. In a modified version of this procedure, only the PDMS substrate is chemically modified before bonding via the epoxy adhesive.

Experimental details

Chemicals and materials

PDMS (10:1 Sylgard 184 Silicone Elastomer Kit) was purchased from Ellsworth Adhesives. To form sheets, the PDMS was mixed in a standard 10:1 ratio of base to cross-linker, and cured at 60 °C for 2 h. The typical thickness of the resulting sheets was ~2 mm. 1 mil Kapton (25 μm) was purchased from American Durafilm. 3M Kapton tape (with silicone adhesive) was purchased from Digi-Key Electronics (5413 AMBER 1/2IN X 36YD (Manufacturer: 3M)). The aminosilane, epoxysilane, and

mercaptosilane, (3-aminopropyl)triethoxysilane (APTES, 99%, PRODUCT #440140), (3-glycidyloxypropyl)trimethoxysilane (GPTMS, 98%, PRODUCT #440167) and (3-mercaptopropyl) trimethoxysilane (MPTMS, 95%, PRODUCT #175617), respectively, were purchased from Sigma-Aldrich. LePage Gel Epoxy adhesive was purchased from a local hardware store.

UV ozone treatment

All UV ozone treatment was performed in a UVO Cleaner Chamber (UVO 342, Jelight Company Inc.) The distance

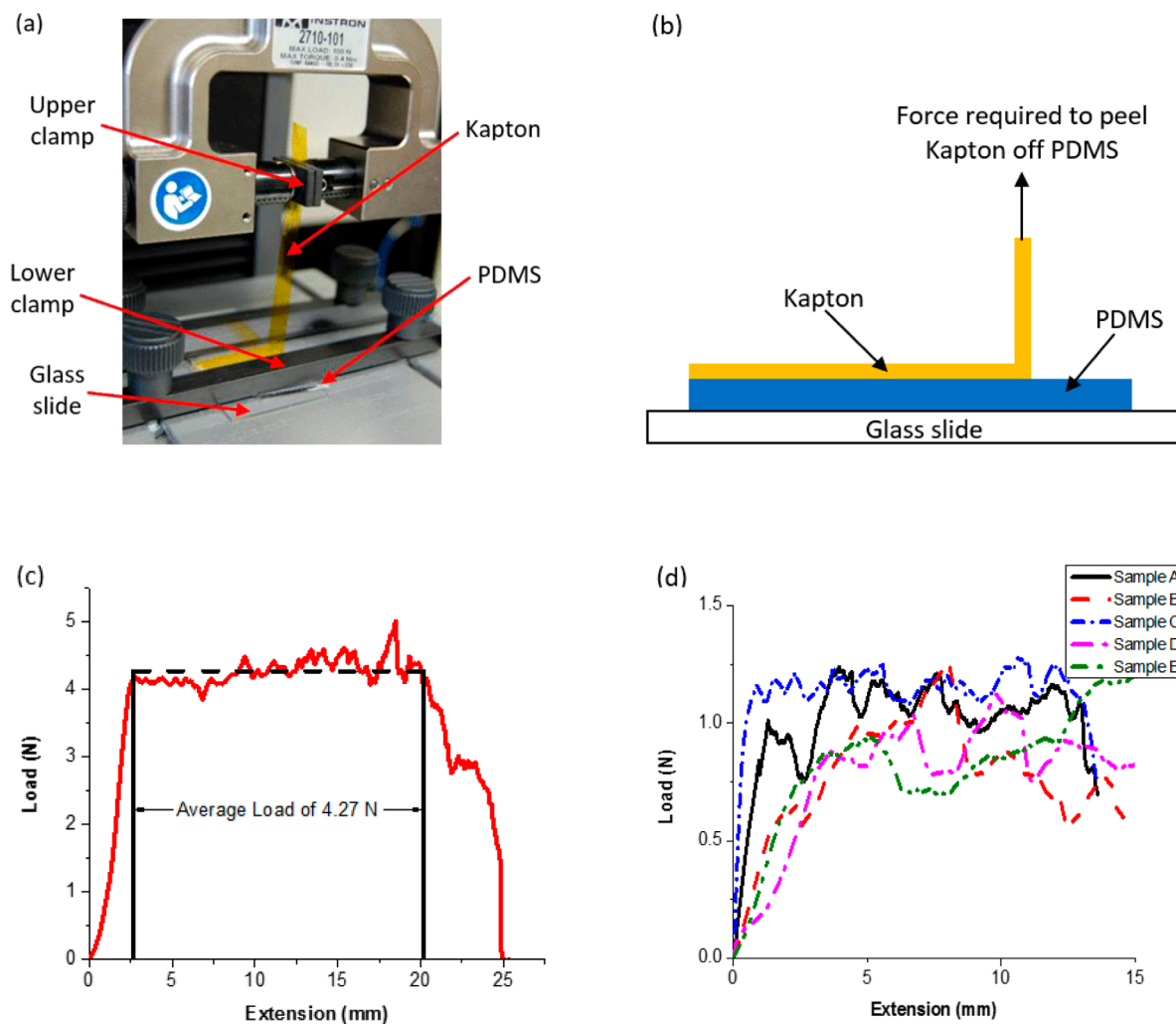


Figure 3. (a) Set up of 90° peel test. The sample is glued onto a glass slide to prevent the PDMS from slipping out of the lower clamp. (b) A schematic of a sample during the peel test. (c) Representative graph of a peel test. (d) Overlapping peel tests of five samples made by bonding Kapton tape to UV ozone-treated PDMS.

between the UV lamp and the surface of samples was consistently maintained at 3 millimeters. The typical treatment time was 10 min.

Kapton-PDMS bonding

Two methods were investigated to bond Kapton to PDMS. In the first method, the surfaces of these materials were treated with oxygen plasma, chemically functionalized, and mechanically pressed together (figure 1). In the second method, to further promote adhesion, a thin layer of epoxy adhesive was applied as an intermediary layer between the functionalized surfaces prior to pressing the surfaces together (figure 2). Two additional types of samples were prepared for comparison: (1) adhesive Kapton tape was bonded directly to PDMS substrates by simply applying the tape to UV ozone-treated PDMS sheets (treated for 10 min); (2) uncured PDMS was coated onto UV ozone-treated Kapton and polymerized at 60 °C for 2 h.

Method 1: bonding of substrates functionalized with either MPTMS or APTES, and GPTMS

In the first bonding method, both the PDMS and Kapton were treated with UV ozone for 10 min to generate hydroxyl groups at the surface (figure 1(a)). In one procedure, PDMS and Kapton were immersed in a 1% (v/v) solution of MPTMS in methanol and GPTMS in methanol for 1 h, respectively (figure 1(b)). In a slightly modified procedure, the PDMS substrate was instead soaked in a 1% (v/v) solution of APTES. After the surface treated substrates were washed with deionized water and dried, they were bonded together and left overnight at room temperature under 30 kPa (figure 1(c)). This pressure was selected (based on our experimental observations) to be large enough to bring the substrates into intimate contact, without being so large as to deform the 2 mm thick sheet of PDMS at the interface with the Kapton. If too large a pressure is applied, a pre-strain mismatch may form between the substrates, which can lead to delamination upon release of the pressure.

Table 1. Average peel strength of several methods of bonding Kapton to PDMS.

PDMS bonded to	Bonding method	Substrate and parameters	Average peel strength (N mm ⁻¹)	Failure mode
Kapton tape (with silicone adhesive)	UV ozone treatment	PDMS only	0.072 ± 0.009	Adhesive
	Uncured PDMS	UV ozone-treated Kapton	0.010 ± 0.005	Adhesive
	APTES/GPTMS (1% v/v solution) (method 1)	PDMS/Kapton in water	0.0013 ± 0.0006	Adhesive
PDMS/Kapton in methanol		0.0027 ± 0.0006	Adhesive	
Kapton (25 μm)	MPTMS/GPTMS (1% v/v solution) (method 1)	PDMS/Kapton in methanol	0.20 ± 0.04	Cohesive (PDMS)
	Epoxy adhesive	PDMS and Kapton	0.0017 ± 0.0009	Adhesive
	MPTMS by liquid deposition and epoxy adhesive (method 2)	PDMS only	0.41 ± 0.07	Cohesive (PDMS)
		PDMS and Kapton	0.46 ± 0.04	Cohesive (PDMS)
	MPTMS by vapor deposition and epoxy adhesive (method 2)	PDMS only	0.31 ± 0.07	Cohesive (PDMS)
PDMS and Kapton		0.33 ± 0.04	Cohesive (PDMS)	

Method 2: bonding of MPTMS-functionalized substrates using an epoxy adhesive

Liquid deposition. Either both the PDMS and Kapton or only the PDMS were treated with UV ozone for 10 min (figure 2(a)). The desired substrate(s) was/were then soaked in a 1% (v/v) solution of MPTMS in methanol for 1 h (figure 2(b)). After washing and drying, a thin layer of epoxy adhesive (LePage Gel Epoxy) was applied between the two surface-treated substrates and left overnight at room temperature under 30 kPa (figure 2(c)). For samples for which only the PDMS was silane treated, Kapton was treated with UV ozone immediately before the application of the epoxy adhesive.

Vapor deposition. In a slightly modified procedure, the functional molecules were deposited onto one or both substrates from the vapor phase. The desired substrates (PDMS and Kapton or PDMS only) were treated with UV ozone (figure 2(a)), as described above. Each substrate was then placed into a separate plastic Petri dish and 2–3 drops of MPTMS were added to each dish beside the substrate. The dishes were closed and left at room temperature for 1 h under rough vacuum (figure 2(b)). A thin layer of epoxy adhesive was applied between the two substrates and left overnight at room temperature under 30 kPa (figure 2(c)). For samples for which only the PDMS was silane treated, Kapton was treated with UV ozone immediately before the application of the epoxy adhesive.

Peel strength analysis

A 90° peel test was conducted using the Instron 5943 with a 1 kN load cell (figure 3(a)). The PDMS sample was secured to a platform, and the Kapton was secured in the upper clamp. As the upper clamp rose, it pulled the Kapton up at a 90° angle. At the same time, the lower platform shifted to the right to maintain 90°. As the upper clamp rose at a steady rate, the load cell measured the force required to peel the Kapton off of the PDMS. The Kapton was pulled off of

the PDMS at a speed of 10 mm min⁻¹. Samples were glued onto glass slides using a silicone adhesive (GE Silicone I All Purpose Sealant) to prevent the PDMS from lifting up during the peel test (figure 3(b)); the glass slides were clamped to the platform during testing. The average peel strength was calculated by measuring the average load of the peel test and dividing it by the width of the bonded area (i.e. the width of the Kapton tape, figure 3(c)). To calculate the average peel strength, the average load was taken over the part of the graph with the most constant load. Ideally, the peel strength should be consistent throughout the test. Each bonding method was tested with at least five different samples to ensure reproducibility (figure 3(d)). During this testing, some samples underwent *tearing* of the PDMS layer (i.e. cohesive failure mode) instead of *peeling* of the two layers (i.e. adhesive failure mode).

Results and discussion

Peel strength analysis of substrates functionalized with either MPTMS or APTES, and GPTMS (method 1)

At least five sets of each sample types were prepared and their average peel strengths were measured using the Instron 5943 (table 1). As a reference, Kapton tape was bonded onto UV ozone-treated PDMS. The tape could be peeled with a force of 0.072 ± 0.009 N mm⁻¹ (figure 4). This adhesive failure (characterized by delamination of the Kapton from the PDMS without any tearing) is shown in figures 5(a) and (b), and is indicative of reversible bonding. Practically speaking, the low peeling energy indicates that there is no chemical bonding that binds Kapton or Kapton/Epoxy to PDMS. Adhesive failure is also seen for the second reference sample, consisting of UV ozone-treated Kapton onto which PDMS was cured. For this sample, the Kapton delaminated from the PDMS during testing, and the average force was 0.010 ± 0.005 N mm⁻¹ (figure 4).

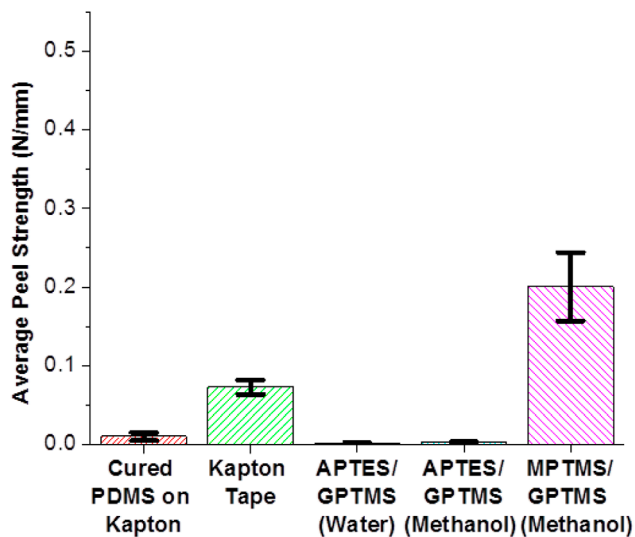


Figure 4. Average peel strength of Kapton bonded to PDMS by functionalization of PDMS with APTES or MPTMS, and of Kapton with GPTMS (method 1). Each sample was repeated five times, and the error bars depict the standard deviation.

A method involving the surface treatment of PDMS with an aminosilane (APTES) solution and Kapton with an epoxy silane (GPTMS) solution was tested. The solutions were prepared with both water and methanol to compare the efficiency of the solvent. Methanol was chosen over other alcohols as the PDMS experienced minimal swelling while submerged in methanol. The adhesion of both samples failed by delamination, indicating that only a reversible bond was formed (figure 5(a)). Changing the solvent from water to methanol improved the peel strength from $0.0013 \pm 0.0006 \text{ N mm}^{-1}$ to $0.0027 \pm 0.0006 \text{ N mm}^{-1}$ (figure 4), but adhesive failure at the interface still occurred, indicating that only reversible bonding had occurred.

In the next bonding method, the aminosilane groups were replaced with mercaptosilane groups [32]. Based on the results of the amine-epoxy method, samples were prepared using a methanol solution. Switching the chemical used to treat PDMS from APTES to MPTMS remarkably improved the average peel strength from $0.0027 \pm 0.0006 \text{ N mm}^{-1}$ to $0.20 \pm 0.04 \text{ N mm}^{-1}$ (figure 4). Samples failed by tearing of the PDMS (figure 5(c); cohesive failure), indicating that a *permanent* bond was formed between PDMS and Kapton.

Peel strength analysis of MPTMS-functionalized substrates bonded using an epoxy adhesive (method 2)

To simplify the bonding procedure and further explore the mechanism of adhesion, we attempted to eliminate the surface treatment step and bond Kapton directly to PDMS using a thin layer of epoxy. Each substrate was treated with UV ozone, a thin layer of epoxy adhesive was applied, and the substrates were brought into contact under pressure. Adhesive failure occurred at the interface between the epoxy and the PDMS (figure 5(b)) at a low average peel strength of $0.0017 \pm 0.0009 \text{ N mm}^{-1}$.

To improve the adhesion, the PDMS sample was functionalized with a mercaptosilane (MPTMS from either the

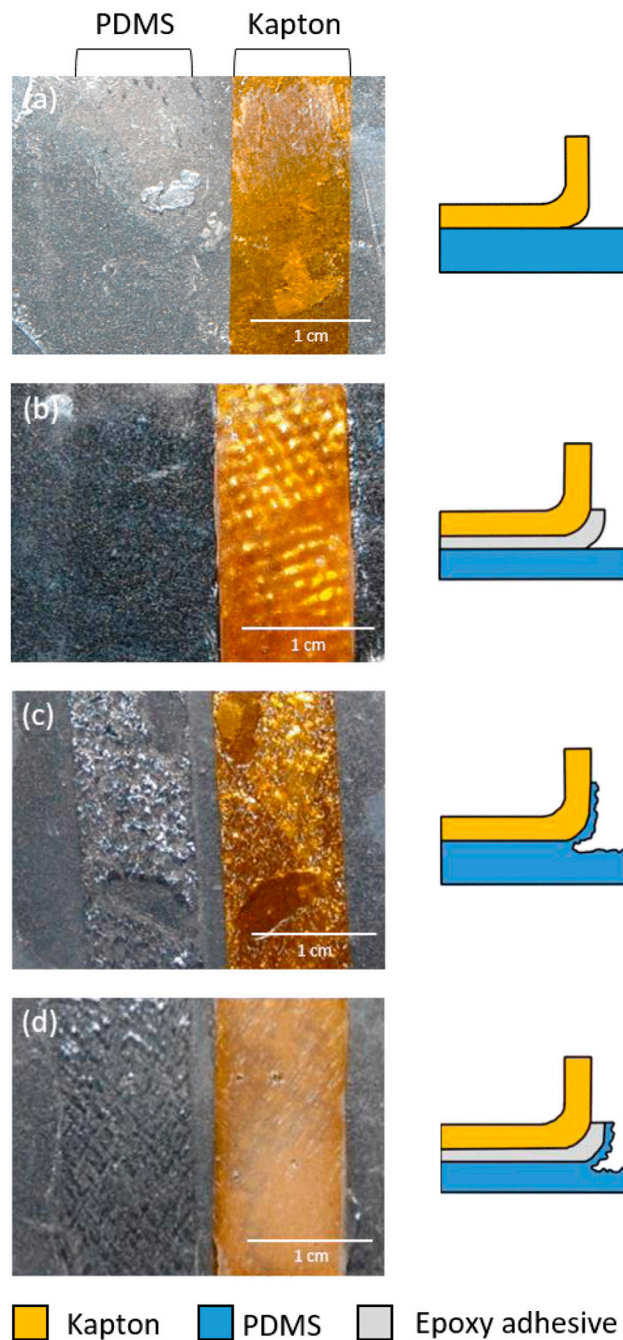


Figure 5. Different types of failure: adhesive failure (i.e. *peeling*) occurs by delamination of the bonded layer ((a) Kapton, or (b) Kapton via an epoxy adhesive) from the PDMS; cohesive failure (i.e. *tearing*) of the PDMS, shown here for bonding either (c) directly to Kapton or (d) to Kapton via an epoxy adhesive. The exemplar samples were prepared as follows: (a) method 1: the PDMS was functionalized with APTES, while the Kapton was functionalized with GPTMS via liquid deposition, (b) method 2: an epoxy adhesive was applied between UV ozone-treated PDMS and Kapton (no further surface treatment), (c) method 1: the PDMS was functionalized with MPTMS, while the Kapton was functionalized with GPTMS via liquid deposition and (d) method 2: both the PDMS and the Kapton were surface treated with MPTMS via liquid deposition and an epoxy adhesive was then applied between the two substrates.

liquid or vapor phase). Substantial improvements to the adhesion were seen regardless of whether the PDMS was treated

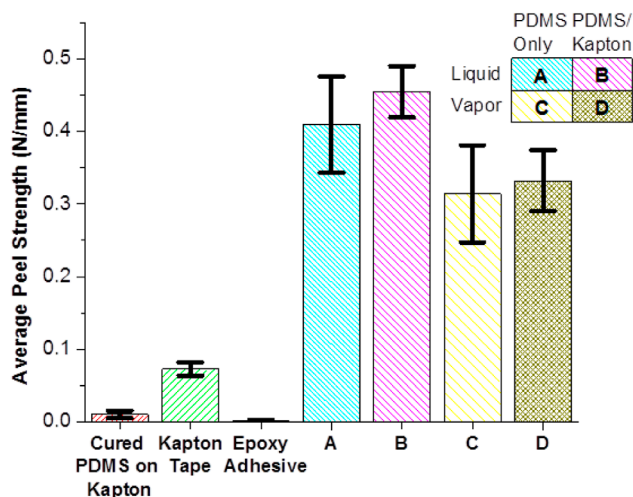


Figure 6. Comparison of different methods of bonding Kapton to PDMS by applying epoxy adhesive between MPTMS treated PDMS/Kapton, where the MPTMS was applied from either the liquid or vapor phase (method 2). Each sample was repeated five times, and the error bars depict the standard deviation. A and B correspond to samples for which either the PDMS only or PDMS and Kapton, respectively, were modified by MPTMS from the liquid phase prior to bonding via the epoxy adhesive. C and D correspond to samples for which either PDMS only or PDMS and Kapton, respectively, were modified with MPTMS from the vapor phase prior to bonding via the epoxy adhesive.

alone or if both the PDMS and Kapton were treated; likewise, applying the MPTMS from the liquid or vapor phase yielded similar results (figure 6). In all four cases, average adhesion energy exceeded 0.3 N mm^{-1} , which falls into the category of *irreversible* bonding.

The PDMS and Kapton were treated with MPTMS by liquid and vapor deposition to compare the efficiency of the different types of deposition. To determine whether it was necessary to surface treat both PDMS and Kapton to improve the adhesion strength significantly, samples were made by surface treating both substrates and compared to samples made by surface treating only the PDMS. The PDMS and Kapton that were surface treated using liquid deposition showed the highest peel strength of $0.46 \pm 0.04 \text{ N mm}^{-1}$ (irreversible bonding). This sample failed cohesively (figure 5(d)): the PDMS tore, leaving a thick layer on the Kapton. When only the PDMS was surface treated, a lower peel strength of $0.41 \pm 0.07 \text{ N mm}^{-1}$ was observed, but the sample also failed cohesively (indicating that the substrates were irreversibly bonded) (figure 5(d)). Both of the samples made using vapor deposition also failed cohesively (figure 5(d)), with average peel strengths from $0.31 \pm 0.07 \text{ N mm}^{-1}$ to $0.33 \pm 0.04 \text{ N mm}^{-1}$ (table 1). It made little difference whether only the PDMS was treated or whether both surfaces were treated; the difference between the average peel strengths measured was less than the standard deviation of the measurement. In all cases, permanent bonding was observed.

Samples that underwent cohesive failure are considered to have formed an irreversible/permanent bond between Kapton and PDMS. This mechanism of failure occurred for five of the methods described above, including bonding

of epoxy-functionalized Kapton to mercapto-functionalized PDMS, and bonding utilizing a thin coating of epoxy where either the PDMS or PDMS and Kapton substrates were chemically-modified with a mercaptosilane (deposited either from the vapor phase or from solution). For these samples, the peel test actually measured the tear resistance of PDMS since the mechanism of failure was by tearing of the PDMS. This peel strength was higher for all of the samples prepared using a thin film of epoxy adhesive ($0.31 \pm 0.07 \text{ N mm}^{-1}$ to $0.46 \pm 0.04 \text{ N mm}^{-1}$) than for samples prepared using surface functionalization only ($0.2 \pm 0.04 \text{ N mm}^{-1}$). This may be explained by two possible factors: (1) a lower effective density of thiol-epoxy bonds may result when functionalized substrates are brought into contact than when a liquid epoxy layer is applied, due to challenge associated of forming intimate, conformal contact of solid substrates (dust and other particles may also disrupt this contact) (2) the epoxy adhesive layer—whose bending stiffness lies between that of Kapton and PDMS—acts as a stress damping layer during the peel test, leading to a larger apparent fracture toughness of PDMS. These arguments, however, are speculative; a rigorous study is necessary to elucidate the mechanism.

Conclusion

In this paper, the bonding of Kapton and PDMS sheets was explored. Two reference samples were included in the study: adhesive-coated Kapton tape applied to UV ozone-treated PDMS, and UV ozone-treated Kapton on which PDMS was cured directly. Both of these samples underwent adhesive failure (delamination at the interface) at relatively low forces (0.072 ± 0.009 and $0.010 \pm 0.005 \text{ N mm}^{-1}$, respectively), indicating that the samples were only *reversibly* bonded. Two basic procedures for the *irreversible* bonding of Kapton to PDMS were presented. The permanent nature of this bonding was evident by the fact that the cohesive failure was observed: during peel testing the PDMS would tear. In the first method, chemical bonding was formed between mercaptosilane-functionalized PDMS and epoxysilane-functionalized Kapton. In the second method, mercaptosilane-functionalized substrates were bonded to a thin layer of epoxy adhesive.

In the first method, mercapto- and aminosilane coupling agents, MPTMS and APTES, were each functionalized on separate PDMS substrates for chemical bonding to epoxysilane-functionalized Kapton. A much stronger, irreversible bond was achieved using MPTMS (peel strength of $0.20 \pm 0.04 \text{ N mm}^{-1}$ when methanol was used as the solvent for the MPTMS functionalization) than APTES, which underwent adhesive failure (peel strength of $0.0027 \pm 0.0006 \text{ N mm}^{-1}$ when methanol was used as the solvent for the MPTMPS functionalization).

In the second method, an epoxy adhesive was sandwiched between the modified substrates and cured. The use of the epoxy adhesive alone was not sufficient to create a permanent bond between the Kapton and the PDMS; while the epoxy adhered well to the Kapton, it consistently delaminated from the PDMS (peel strength $0.0017 \pm 0.0009 \text{ N mm}^{-1}$). However,

when either the PDMS alone or the PDMS and Kapton were functionalized with a mercaptosilane, cohesive failure was observed during peel testing (peel strength $0.31 \pm 0.07 \text{ N mm}^{-1}$ – $0.46 \pm 0.04 \text{ N mm}^{-1}$). Similar results were achieved regardless of whether the MPTMS was deposited from either the liquid or vapor phase onto the UV ozone-treated substrates. For all cases involving one or more functionalized substrate and an epoxy layer, irreversible bonding was achieved.

We have demonstrated an effective method for irreversibly bonding Kapton to cured PDMS substrates. These two materials are important materials in the fabrication of flexible and stretchable electronic devices; these results can be leveraged in a variety of application areas where a strong bond between these materials is required. Such applications include the fabrication of biosensors (either mounted on the surface of the skin or utilized *in vivo*), and devices for the internet of things (IoT) with various form factors.

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