

Development of a stretchable and removable electrical interconnection solution for ultra-thin electronic components

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Abstract—

Currently, the solutions for interconnecting electronic components with their active face facing on substrates are based on metallic soldering. The mechanical contacts are therefore rigid. In order to enhance the reliability of the bonding, underfill is usually used to redistribute the thermo-mechanical stress created by the Coefficient of Thermal Expansion (CTE) mismatch between the silicon chip and substrate. Underfill is done with epoxy resins containing silica fillers (SiO₂). As a result, the removal of components is no longer possible. Moreover, this solution is not suitable for devices integrating ultra-thin silicon components (<100 μm) hybridized on flexible substrates that may be subject to deformation. This is the case, for example, of medical "patches" worn on the person and continuously solicited. Indeed, the rigid contact points are likely to break. To address this issue, we are developing a thin anisotropic conductive and stretchable adhesive film inspired by the adhesion of the gecko. Thanks to the microstructuration of its toes involving about 1 million setae, the gecko can develop a large contact surface and thus a large force of attraction by the multiplication of van der Waals interactions. In this work, this "dry adhesion" based on the principle of "contact splitting", was implemented in order to improve the adhesion of a flexible interconnection. For this purpose, the surface of a polydimethylsiloxane (PDMS) film was structured with micrometric mushroom-shaped patterns known to be the most efficient form of contact. To this end, silicon molds with varying mushroom geometries were used to shape the PDMS (with different cap and pillar diameters) and the adhesion force microstructured films were assessed (shear and pull-off experiments). To make these films locally conductive through the thickness, a conductive composite was prepared and locally deposited in the mold. One approach we investigated, was using a screen-printing mask. This approach has been implemented and characterized using electrical tests (I-V measurements) in order to select the most suitable films to make a flexible interconnection.

Keywords— Interconnection, flexible, gecko-like adhesion, ACA

I. INTRODUCTION

This work presents an innovative solution for assembling electronic components for applications where mechanical

deformation such as bending and stretching must be tolerated. For example, thin flexible electronic devices can follow the shape of the human body in motion. In addition, this work shows a removable solution to facilitate the replacement of a defective component. During the two last decades, the packaging of electronic component has to satisfy high interconnection density, high data throughput, miniaturization, and reliability needs while keeping a low cost. Flip-Chip interconnection of chips, i.e. with the active face facing the substrate, offers the best solution to meet these requirements. The main techniques involve soldering micro-bump between the chip and the substrate to create an electrical and mechanical bond. In order to enhance the reliability, underfill is usually used to redistribute the thermo-mechanical stress created by the Coefficient of Thermal Expansion (CTE) mismatch between the silicon chip and the substrate. Underfill materials are usually epoxy bases. Once cross-linked, the material is hard and not tolerate deformation. Moreover, the chip cannot be disassembled. Another interconnection solution is to use conductive adhesives. There are two types of conductive adhesives: anisotropic conductive adhesives (ACA) and isotropic conductive adhesives (ICA). Polymers used for these type of interconnections are thermoset with a high Young's modulus as epoxy [1] or acrylic adhesives [2] mixed with conductive fillers. In ICAs, the proportion of conductive fillers is very high: around 80 % weight or higher [3]. ACAs are electrically conductive in the z-axis due to an adding of conductive fillers in low proportion in the polymer matrix [2]. In case of ACAs, films (or pastes) require high pressure and temperature for processing. Pressure is necessary to connect conductive fillers and obtain an electrical continuity and temperature is necessary for the polymer curing (typically $T > 100^\circ\text{C}$). Nevertheless, this approach is still difficult to bend and impossible to stretch. Moreover, there are some difficulties to remove easily the component.

Dry adhesion based on van der Waals interactions [9] is an alternative to process conductive adhesive at room temperature. In nature, dry adhesion was observed for insects or lizards and rely on the microstructuration of their

feet [4]. Thanks to SEM image technology, the hierarchical microstructuration of the gecko lizard toes was observed as divided in three levels: lamellae, setae and spatulae [5] which enable to establish a very large contact area with surfaces [10]. Dry adhesives inspired from the gecko have been studied for around 20 years. Scientists groups tried to mimic by simplifying the gecko microstructuration because of its complexity for technical processes [6–10]. In 2007, Del Campo *et al.* determined that the mushroom shape was the ideal contact shape to obtain a maximum pull-off adhesion [11]. In the literature, most of the microstructured films are built with polymers [8,9], particularly elastomers such as poly(dimethylsiloxane) (PDMS) which have often been chosen for their flexibility, stretchability and ability to be molded into very small structures [14].

To obtain a conductive material with dry adhesive properties, conductive ICA-like gecko films were developed from the last 10 years. The targeted applications were mainly the fabrication of EEG-ECG electrodes applied directly to the skin [15]. To that aim, elastomer composites were prepared including silver powders, carbon powders or carbon nanotubes (CNT) mixed with graphene, and were molded in a mushroom shapes [15–17]. All these conductive microstructured films are fully electrically conductive. Short-circuits come out of a multiple of electrical pads are in contact, the electrical way is blocked. Yet in microelectronic there is a necessity to fabricate a non-continuous conductive films to avoid short-circuit in the system.

In this work, we aimed to create a flexible, stretchable, reversible, anisotropic gecko-like interconnection, adapted for the processing of flexible substrates at room temperature. The concept is presented in Figure 1.

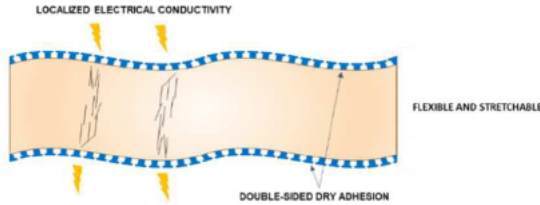


Figure 1 Scheme of the concept of the gecko anisotropic interconnection film

Here, the fabrication process of a conductive and mushroom-shaped microstructured PDMS is described. First, silicon master molds were manufactured. Then, composites based on carbon nanotubes (CNT) and PDMS were prepared and molded. In this work, the electrical conductivities of structured and unstructured composite films are compared. Then, the adhesion of a CNT-PDMS structured film is compared to a pristine PDMS structured film. Finally, a first attempt to localize the conductive composite in the film is presented.

II. EXPERIMENTAL METHODS

A. Conductive and Microstructured PDMS-mushroom

Fabrication of conductive dry adhesives mushroom-shaped molds were fabricated from Silicon On Insulator (SOI) (100 mm - orientation <100>) wafers with a 2 μm -thick BOX (Buried Oxide layer) and a 15 μm thick device layer. To define the location of mushrooms and pillars, a positive

photosensitive resin AZ4562 was spin-coated on the wafer, cured and subsequently exposed to UV (365 nm) through a mask with apertures diameters of 10 μm and spaces of 16 μm in a hexagonal arrangement. After the development of the photosensitive resin, the Si device layer was etched till the BOX in a ICP Oxford tool using SF_6 and CH_2F_2 . The BOX was then etched by HF vapor to define the cap diameter.

Prior to PDMS replica molding, a fluorinated silane was deposited on the SOI master molds by a vapor phase process in a hermetically sealed chamber. Then, liquid precursors of PDMS (Sylgard 184 from Dow) were poured into the molds and spread out by spin-coating. The spin-coating parameters were adjusted to obtain films with a backing layer thickness of approximately 190 μm . The curing of PDMS was carried out at 100°C for 30 min and the microstructured PDMS film were peeled from the SOI mold. To prepare a conductive PDMS, multiwall carbon nanotubes with a mean aspect ratio of 830 (diameter between 6 to 13 nm) were purchased from Sigma-Aldrich. The dispersion protocol of CNTs in the PDMS matrix was inspired from Kim *et al.* [18]. A proportion of 1-wt% (relative to PDMS) of CNTs was added in a beaker with isopropyl(alcohol) (IPA) with a ratio of 1:100 in weight. The mixture was homogenized in an ultra-sonic bath for 10 min to separate the CNT bundles. Then 1-wt % (relative to the CNT and PDMS proportions) of a low viscosity Poly(dimethylsiloxane-co-methylphenylsiloxane) (purchased from Sigma Aldrich), was added to the suspension of CNT/IPA and mixed using a planetary mixer, followed by sonication for 10 min. Finally, IPA was evaporated using a rotary evaporator and the crosslinking agent was added in the mixture. Lastly, the CNT-PDMS mixture was degassed under vacuum and poured into mushroom-shaped molds.

The localization of the conductive composite was performed by screen-printing. The uncured CNT-PDMS composite was deposited through a stencil directly on a SOI master mold patterned with pillars. The stencil was composed of round apertures of 300 μm diameter and a spacing of 500 μm . After the screen printing process, the CNT-PDMS areas were thermally cured and pristine PDMS was deposited by spin coating, cured and demolded following the procedure presented in the experimental section.

B. Measurement Test Methods

Electrical characterizations of the conductive films along the thickness direction were performed by placing the films (microstructured and unstructured) between two flat copper electrodes with a surface of 1 cm^2 . To insure a good contact between the electrodes and the film, a weight (880 g) was added on the top of the box tester electrode.

Current-Voltage curves (I-V) were recorded using an electrical measurement bench developed internally. A forward and backward voltage from 0 to 10 V and from 10V to 0V was applied to the electrodes with a step of 100 mV. Several cycles were performed and the electrical conductivity was extracted from the 4th cycle using the following formula:

$$\sigma = \frac{t}{R \times S}$$

were σ is the electrical conductivity of the composite (S.m^{-1}), R is the resistance (Ω), S the surface (0.0001 m^2) and t is the film thickness (m).

To assess the adhesion performances of the microstructured films, pull-off test were achieved by contacting the mushroom structured PDMS with a flat glass substrate. Before the test, the glass substrates were cleaned with IPA and covered with a protective film to avoid particles contamination. The microstructured film (surface: $5 \times 5 \text{ mm}^2$) was fixed on a piece of glass which was already pasted on aluminum sample holder. Pull-off tests were carried out on a tensile machine Syntax 12 from 3R (Recherches et Réalisations Rémy) equipped with a 100 N load cell. Once positioned between the jaws, the samples were first compressed to a preload of 2.3 N at a velocity of 0.1 mm/min. Then, the sample was pulled-off with a velocity of 0.1 mm/min. The adhesion strength of the microstructured films was determined at the maximum of the pull-off force.

III. RESULTS AND DISCUSSION

A. Non-conductive PDMS Microstructured Films

Scanning Electron Microscopy (SEM) pictures of a mushroom-shaped PDMS film are shown in Figure 2. A dense array of mushrooms arranged in a hexagonal pattern is clearly visible (Figure 2 a.). The PDMS mushrooms are well aligned and defined without collapsing which is favorable in terms of adhesion as all mushrooms can contribute to establish van der Waals interactions. The backing layer which is the part supporting the mushrooms, has a thickness of around $190 \mu\text{m}$. From the Figure 2 b., the dimensions of individual mushroom can be measured precisely: the mushroom pillar height is $15 \mu\text{m}$, the cap diameter is $18 \mu\text{m}$, the pillar diameter is around $10 \mu\text{m}$ and the cap thickness is $2 \mu\text{m}$.

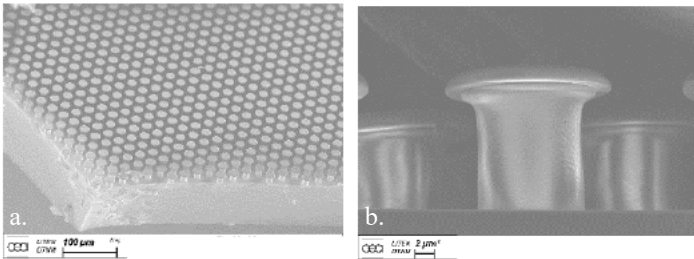


Figure 2 SEM image of a micro-structured PDMS film a. Top view of the film b. Zoomed-in side view of the PDMS mushrooms

A second objective was to perform a molding of a conductive CNT-PDMS. Firstly, a conductive film without structured part was made. Then, once the viscosity parameter adapted for molding, the mushroom-shaped were molded with CNT-PDMS. The next part shows the electrical and mechanical characterization for CNT-PDMS films.

1) CNT-PDMS based film

In order to prepare a conductive microstructured film, a study was first performed involving the preparation of unstructured conductive PDMS films by introducing MWCNTs with different wt% in the mixture and determining the electrical

percolation threshold. To be able to mold the CNT/PDMS mixture, a minimum CNT content (low viscosity) must be aimed for and the quality of the dispersion of CNT in the elastomer matrix is therefore a critical aspect. The conductivity of the composites with different filler contents was assessed using I-V measurements. Figure 3 shows an example of I-V curves for an unstructured and a structured sample (composite PDMS – CNT (1 wt%)).

Whatever the CNT loading (in the range investigated), the I-V curves are nonlinear. The electrical resistivity varies with the voltage, the resistance decreases with the voltage increases. This behavior is typical of inhomogeneous materials containing conductive fillers in a polymer matrix and was already observed on composites of PDMS/CNT [19]. Wang et al. [25] have determined that the nonlinear behavior was mainly due to electron hopping effect and tunneling effect for relatively low voltages. In this study the resistance was determined by fitting the I-V curves to a straight line with an imposed start at 0.

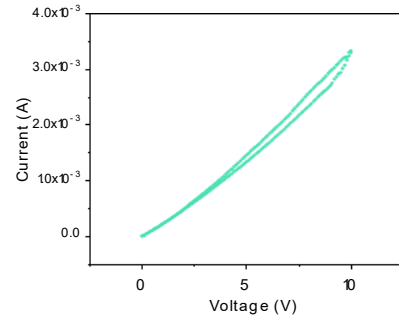


Figure 3 I/V curves measurement for a CNT-PDMS structured film

Figure 4 presents the variation of the electrical conductivity as a function of the filler content (mass fraction) for unstructured composites (with CNT mean aspect ratio of 830). For a MWCNTs mass fraction of less than 0.4 wt% the composite has a conductivity of around 10^{-12} S/m . At the opposite, for a higher mass fraction of MWCNTs of 1 wt% and above, the conductivity rises sharply up to 0.1 S/m . The region in between corresponds to the percolation transition. In a conductive composite, the percolation threshold is the lower content of fillers to obtain an electrical path through the composite.

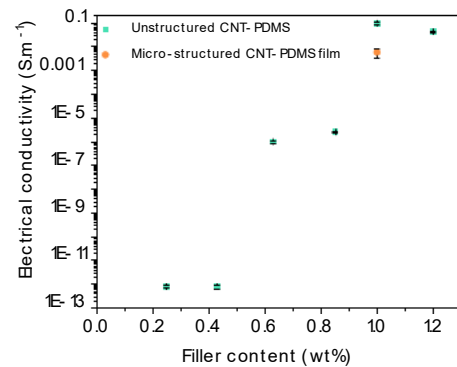


Figure 4 Variation of the electrical conductivity as a function of the filler content for CNT-PDMS composites

The electrical conductivity of a composite depends on volume or mass fraction of the conductive fillers and can be described by the following equation [20] :

$$\sigma \propto \sigma_0(p - p_c)^t$$

with σ electrical conductivity of the composite, σ_0 electrical conductivity of the fillers, p volume fraction of CNT, p_c percolation threshold and t a critical exponent which depends on the connectivity and aspect ratio of the fillers

The percolation threshold can be estimated from Figure 4 and is between 0.4 and 0.6 wt% of MWCNTs in the PDMS. The percolation threshold depends strongly on the filler aspect ratio but also on their dispersion in the matrix. The presence of MWCNTs aggregates observed on SEM image (Figure 5) increase the percolation threshold. Indeed, the estimation of the percolation threshold as a function of the aspect ratio (833), is around 0.17 wt% of CNT in a matrix. Here, the aspect ratio corresponding to this percolation threshold is between 240 and 350. A similar result was observed for Ag nanowires (NWs with a mean aspect ratio of 220) in a polymer matrix where a discrepancy of a factor 2 was evaluated between the experimental results and the theoretical one [21]. To prepare a microstructured conductive film we have chosen a MWCNT mass fraction well above the percolation threshold i.e. 1 wt%.

For that proportion, the mixture of uncured PDMS and MWCNTs prepared with the protocol described in the experimental method section, is sufficiently fluid to be molded. Figure 5 shows a cross section of a CNT-PDMS microstructured film. The mushrooms can be distinguished on the upper side of the film and do not show any defects or collapse. In the backing layer, lighter zones appear corresponding to aggregates of MWCNTs suggesting that the mixing process can still be optimized. Some mushrooms on the film surface appear shinier confirming the presence of MWCNTs. Finally, one can notice an irregular thickness of the backing-layer resulting from the difficulty to spin-coat the mixture of PDMS/MWCNTs.

The electrical measurement appears dependent of the surface finish. Indeed, the electrical conductivity decreases from 10^{-1} S.m^{-1} for the unstructured flat conductive film to 10^{-2} S.m^{-1} for the microstructured rough CNT-PDMS film (Figure 5). It should be noticed this electrical conductivity is estimated using the real surface in contact with the electrode on the mushroom face (i.e. 0.13 cm^2). The lower conductivity measured for microstructured CNT-PDMS composites could be due to an imperfect contact with the copper electrode, the backing layer flatness being not optimized. Kim *et al.* have obtained a slightly higher conductivity of 0.6 S.m^{-1} for similar composite (with 1 wt% CNT with AR of 1000 in PDMS) and even 1 S.m^{-1} at 1 wt% of CNT and graphene fillers [16]. It

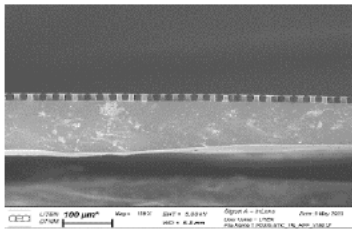


Figure 5 SEM image of a cross-section of a structured CNT-PDMS composite

should be mentioned that higher loadings cannot be used to perform structured conductive composites. Indeed, PDMS mixed with a high CNT loading is too viscous to be used for the filling of micrometer structures.

To confirm the dry adhesion properties of these microstructured films, adhesion tests in pull-off configuration were performed. Pull-off adhesion tests performed normal to a given surface, are a classical tests used to characterized gecko inspired dry adhesives [10], [22], [23] and usually imply a compression step followed by a retraction step. During the compression step the mushrooms are pressed against the glass substrate enabling them to maximize the contact area and as a result improve the adhesion of the film. In this study, the preload value of 2.3 N was is the limiting force value to prevent mushroom buckling. The Figure 6 shows the pull off force - displacement curve obtained for a microstructured CNT-PDMS film with compressive forces shown as positives and tensile forces shown as negatives.

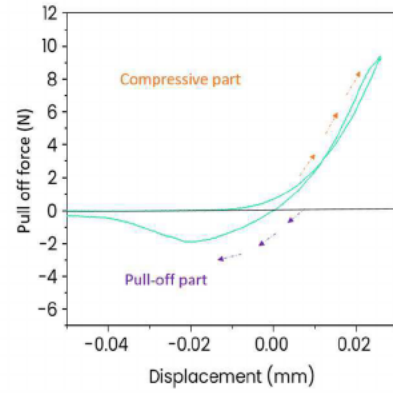


Figure 6 Graph of a pull-off test for CNT-PDMS (1wt%) micro-structured film

The adhesion strength of a microstructured film is determined as the maximum force of the pull-off part. On this example, the adhesion force is 1.9 N.cm^{-2} .

To assess the impact of the presence of fillers in the film on adhesion strength, the micro-structured CNT-PDMS film was compared to the same micro-structured film made of pure PDMS (Figure 7). Several successive measurements were performed on each sample under identical conditions to assess the repeatability of the measurement six times. To facilitate the visualization of the results, the six strength-displacement curves in the pull-off part are shown as positive.

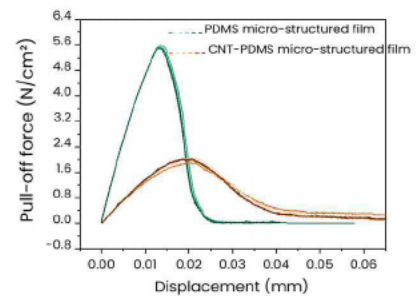


Figure 7 Graph of the comparative adhesion force in pull-off for PDMS micro-structured film and CNT-PDMS structured film

For each sample, the obtained curves overlap and the maximum pull-off forces are repeatable. The pristine microstructured PDMS shows a superior adhesion strength with a maximum adhesion force of 6 N.cm^{-2} compared to 2 N.cm^{-2} when 1% CNT fillers were added in the PDMS-matrix. In addition, CNT-PDMS curves appear more spread with a maximum pull-off shifted to higher values of displacement than for the microstructured PDMS film.

These results differ from those obtained by Seong *et al.* who demonstrated that the pull-off adhesion force was almost unchanged by the addition of low mass fraction of CNT in the PDMS matrix (0.5-2 wt%) for a given mushroom geometry [24]. We assume that in our case, this reduction of pull-off force could be the result of a misalignment of the sample with the substrate result of the thickness variation of the backing layer (shown in Figure 5) and the lack of flatness of the sample. Booth *et al.* observed that for misalignment of 0.1° the adhesion measurement type assigned is a peel-like test [25]. Thus, to maintain an adhesion force around 6 N/cm^2 , a smooth backing layer is mandatory. Indeed, a film with a rough backing layer pasted on the glass is still irregular (composed with bumps and hollows). Hence, the mushroom contact is not homogeneous on all the sample surface. Finally, the adhesion is characterized only on the contact surface which is possible only on bumps areas.

2) Localization of Composite in Pillar Structured Film

The fabrication of a removable anisotropic interconnect requires to localize conductive areas at certain points in the microstructured film.

Figure 8 shows a picture of the obtained film with localized CNT-PDMS areas (filler loading of 1 wt%).

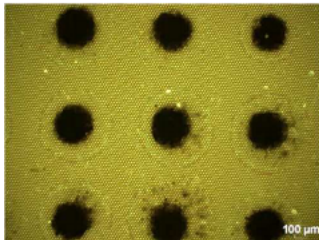


Figure 8 Optical microscope image of pillar-shaped microstructured film with conductive CNT-PDMS inserts.

In the picture, one can distinguish 9 large black zones corresponding to CNT-PDMS composite whereas lighter zones correspond to pristine PDMS. The small dots on the picture are the micrometrics pillars. A circle around the pillar of CNT-PDMS is observed, which could be a disturbance zone for the pillar PDMS.

For a better understanding of the microstructured film architecture, SEM observations were carried out on the cross section of the film (Figure 9).

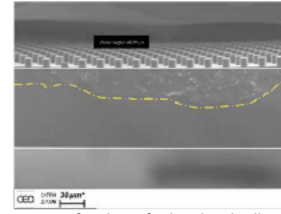


Figure 9 SEM image of a slice of a localized pillar structured film

In this picture, the conductive composite of CNT-PDMS is clearly visible in the upper part of the backing layer (just below the pillars). To guide the eye, a dashed yellow line was added and delimits this area. The lower part of the backing layer is a pure PDMS layer indicating that the conductive areas do not pass through the entire thickness of the film. As a result, the electrical conductivity of this microstructured film in z-axis is very high (10^{-10} S/m). These preliminary results are not surprising since the non-conductive PDMS was poured over the conductive areas and completely covers them. To avoid this situation, strategies are currently implemented to reduce the excess PDMS over the conductive areas and chemically or physically etch the remaining PDMS.

IV. CONCLUSIONS AND PERSPECTIVES

In this work we prepared and molded a fully conductive composite based on high aspect ratio MWCNTs and PDMS. To this end, composite formulation and mixing protocols were optimized to obtain a homogeneous and fluid mixture with a low mass fraction of MWCNTs (1 wt%). The electrical conductivity obtained for such composites is 10^{-1} S.m^{-1} but decreased by a decade for microstructured films (10^{-2} S.m^{-1}). The adhesion performances of the mushroom-shaped microstructured conductive films were characterized by pull-off tests and compared to a non-conductive equivalent. An adhesion force of 1.9 N.cm^{-2} was obtained for the microstructured conductive composite (CNT 1 wt% - PDMS) whereas for a similar mushroom geometry, the adhesion force on structured PDMS is 6 N.cm^{-2} . This deterioration in adhesion and electrical conductivity could be explained by the lack of planarity of the composite films inducing misalignment during the pull-off test and a decreased contact area with electrode during the electrical characterization. This limitation will be the subject of further works especially to improve the smoothness of the backing layer. Finally, firsts encouraging results for the localization of the MWCNT-PDMS composite in microstructured films were obtained. Inserts of conductive MWCNTs-PDMS were successfully screen printed on a silicon mold. Further work will be performed to obtain the z-axis conductivity in the microstructured films.

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