Highly Stretchable Metal Films on Polymer Substrates: Mechanics and Mechanisms

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ABSTRACT

Stretchable interconnects are required for flexible electronic devices where a typical system comprising metallic interconnect and elastomer substrate needs to undergo large deformation while maintaining its mechanical and electrical integrity. This paper reports on a highly stretchable interconnect system consisting of indium metal periodically bonded to a compliant polydimethylsiloxane (PDMS) substrate via a thin, cracked chromium interlayer. It is shown that the system can sustain large tensile strain of ~100% without mechanical breakage and about 30% strain without significant electrical and mechanical damage. It is shown that the In-layer overlying the narrow gaps between adjacent Cr islands are strained much more than that possible in bulk In. The increased failure strain is demonstrated to be associated with micro-scale effective gauge lengths between Cr islands using digital image correlation (DIC). It is further demonstrated via experiments on freestanding thin In foils that the ductility of the foil increases with decreasing sample length, particularly when the sample length is less than 100 µm. This suggests that the root cause for obtaining such large strain-to-failure in periodically bonded In films on PDMS is the delay in the film plastic instability as a result of the reduction in the sample gauge length.

INTRODUCTION

Flexible electronic devices are used in several new areas such as smart clothing[1], sportswear[2] and medical diagnostics[3]. Such devices are predicted to lead the revolution of Internet of Things in the upcoming years. A typical flexible electronic device requires that its major components (e.g. power source, logic, sensors, memory and communication devices) be in connection with each other on a non-rigid substrate using thin or thick metallic interconnects. The non-rigid substrate can ideally be made of cross-linked polymers with inherent ability to stretch to large strains without undergoing permanent deformation. One important aspect of such devices that has been a constant challenge to many research groups is that the interconnect films need to undergo numerous, large cyclic tensile and flexural strain without mechanical failure or reduction of electrical performance. To date, the principal strategy to produce metallic lines on flexible substrates has comprised creating serpentine structures of metal films[4-12] or out-of-plane buckling structures on elastomer substrates[13-16]. These geometrically modified features on flexible substrates have proven to be space-inefficient, and thus prohibits high density interconnect system. Moreover, thin Cu films on polyimide substrates have shown severe cracking at or above 20% strain and resulted in delamination[17]. Use of Au as an interconnect material have produced higher strain levels but would be expensive for real world applications[18].

In this paper, the authors propose a novel approach where the ductility of metallic film is significantly enhanced by confining the deformation into numerous periodic narrow regions that delay the plastic instability and postpone overall rupture of the metal-polymer system. Through systemically carried out experiments, it is shown that Indium interconnect films, a relatively low melting point metal can show high ductility while being periodically bonded to polymer substrates with low rise in resistivity with strain. Here, it shown that such unique architecture can be utilized to get overall film strain more than 70% without accumulating significant mechanical and electrical damage. Although the demonstration of higher stretchability of these metal-polymer systems proves the viability of flexible electronic devices, the underlying science behind this phenomenon is still not fully known. This paper aims at identifying the mechanics and mechanisms behind this high stretchability of such systems by experiments in macro and microscale and Digital Image Correlation(DIC).

EXPERIMENTAL

Since the interconnect system consists of a set of heterogeneous materials (metal and polymer), their choice is based on mutual compatibility, mechanical properties, cost and manufacturability. For the choice of metal, Indium was chosen because of its low yielding stress (2.5-3MPa) and has a melting point of 157^{o} C that enables the material to undergo creep relaxation and dynamic recrystallization at room temperature (homologous temperature T/Tm=0.7) and a relatively low electrical resistivity (8×10^{-8} Ohm-m). Polydimethylsiloxane(PDMS) was chosen as the non-

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rigid substrate material because of its high deformability, chemical stability and biocompatibility. Lastly, Cr was chosen as the thin intermittent layer between the metal and polymer because of its proven adhesive capabilities with PDMS[19]. Commercially available Sylgard 184 Silicone Elastomer Kit and a curing agent were mixed at 10 parts to 1 ratio by weight and were thoroughly mixed together using a stirrer. Then debubbling of the mixture was done by keeping it in an ultrasonic bath (Manufacturer: Cole Palmer) followed curing at 80° C for 3 hours. Surface treatment was done by an atmospheric oxygen plasma system (Surfx Atomflo 400) with helium as secondary gas for a minute at 100W power. 0.5mm thick PDMS blocks were prepared and cut into dog-bone shaped samples. Two types of deposition methods were used to manufacture the metal-polymer system. First, a 5-10nm thin layer of Cr was deposited on top of PDMS which was followed by deposition of 1μm thick Indium layer using a magnetron sputtering chamber (BOC Edwards Auto 306 and Denton DV 502-A) at room temperature. Note that no external heating or cooling of the substrate was done inside the chamber. The base pressure was maintained at 10^{-7} torr. The sputtered 1μm In film was used as a seed-layer to provide electrical continuity for the subsequent electroplating of metal since the previously deposited thin interlayer of Cr was found to be discontinuous. Finally, a 5μm metal layer was deposited using a commercially available acidic electroplating bath (Indium Sulfamate, Indium Corporation, US). The metal film was at the center of the dog-bone shaped PDMS samples. During stretching of the samples while recording the change in resistance, a constant displacement rate of 0.035mm/s was maintained which translated to an overall strain rate of 1.3×10^{-3}/s using a custom-made tensile testing machine with LVDT recorder. Fiducial marks on the film was placed on top the films to track the actual strain on the In film during stretching of the metal-polymer system (Fig.1).

For the DIC experiments, high resolution Scanning Electron Microscope (SEM) images were taken at different strain levels of the metal-polymer system. For this set of experiments, 2μm of In was deposited on top of 5nm thin Cr interlayer and PDMS. Thickness reduction of the primary metal layer was done to better observe the effect of the underlying layer and effect on the top layer. To identify the effect of Cr interlayer, in-situ stretching of thin Cr film on PDMS was done systematically and recorded film strain to correlate with the results from DIC. A MATLAB based package NCORR was used to carry out the simulation.

For finding the effect of gauge length in delaying the necking instability of the freestanding In films, foils of In of 47-50μm thickness was rolled from a 99.9% pure bulk In. High vacuum annealing at 120°C was done to reduce the dislocation density induced because of cold working. A custom-made micro-tensile stage was used to perform tensile testing on these samples. To simulate the adhesion between the film and Cr, Loctite Ultra gel glue was used to bond the freestanding In films with the stainless steel plates for stretching from one end while the other being fixed. A servomotor enabled linear stage (Zaber Technologies, Canada) was integrated with a load cell (Futek, USA) and a capacitance gauge (Capacitec, USA) to perform tensile testing. A LabVIEW code was used to perform computer-assisted automated testing while recording the load and displacement data.

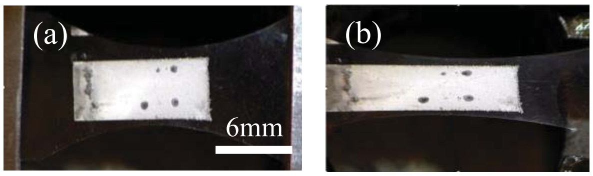

Fig.1 Electroplated In films on PDMS a) unstretched and b) stretched condition. The black dots (fiducials) are tracked during tension test to confirm actual strain induced in the metal film[20].

RESULTS

In the authors' previous works[20, 21], it was shown that a stretchable interconnect system consisting of 5-10μm thick electroplated film on In on PDMS substrate with a discontinuous adhesion layer of Cr can be stretched to a very high linear strain (more than 70%) without film failure.

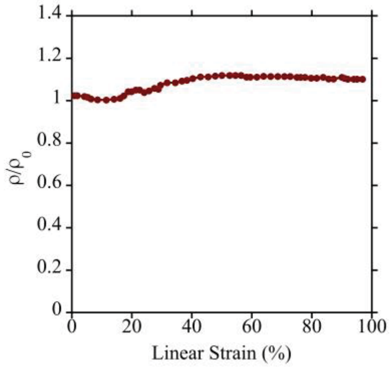

Fig.2 Resistivity change of the In interconnect film stretched to more than 90% strain while bonded to the PDMS substrate.

The changes in mechanical and electrical properties were recorded before and after stretching the metal-interconnect systems for multiple samples. A geometric effect was observed which resulted in out-of-plane wrinkling of the overlaying In film upon release from the strain. Change in resistance was recorded and converted to change in resistivity using the following equation where volume was assumed to be constant because of plastic deformation of the metal film

$$\frac{\rho}{\rho_o} = \frac{R}{R_o}\left(\frac{L_o}{L}\right)^2 \tag{1}$$

Here, ρ_o, R_o, L_o denote resistivity, resistance, and length before stretching respectively and ρ, R, L denote resistivity, resistance, and length after stretching respectively. The increase in resistance in the film is due to the increase in film length which results in a decrease in the cross sectional area. From the Fig.2, it is observed that the resistivity increases because of the rise in dislocation density and other defects in the film with the increase in tension of the metal-polymer system. In other words, deviation from the unity in the change in resistivity value suggests the effects of plastic deformation and defects in the form of micro-cracks formation. The change of slope at around 35% suggests of existence of one or multiple dynamic recovery mechanism during stretching beyond that critical strain level. Since In film has a high homologous temperature, this can be explained either through dynamic recovery/recrystallization mechanism that actively limits the growth of dislocation density or that further plastic deformation is concentrated near the expanding defects (e.g. micro-cracks), which in turn, constraints the increase of plastic strain in the film. When compared with other metals such as Au or Cu where the rise in resistivity occurs due to the increase in plasticity throughout the entire stretching period, In film here showed rise in resistivity in up to a critical strain level and then plateaued, suggesting now subsequent rise in electrical resistivity with the increase in further strain in the film. To investigate the effect of Cr interlayer on the overall stretchability of the In film was carried out by correlating separation of Cr islands (Fig.4) with the results obtained from Digital Image Correlation where strain localization at multiple locations is observed rather in a single location. It is hypothesized that the small length scale of the cracked Cr islands may have helped stretch the overlaying In layer in between two adjacent Cr islands far beyond they would had the length scale (effect gauge length) been larger. The cracked Cr islands were observed to be in close

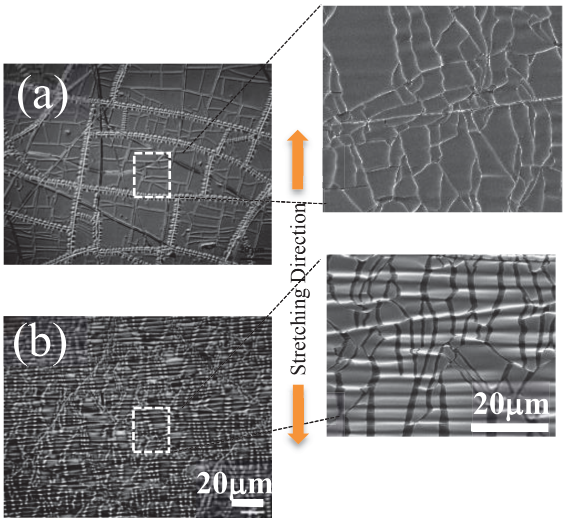

Fig.3 Discontinuous layer of Cr interlayer on PDMS deformed by island separation when PDMS is a) not stretched and b) stretched to 35% strain.

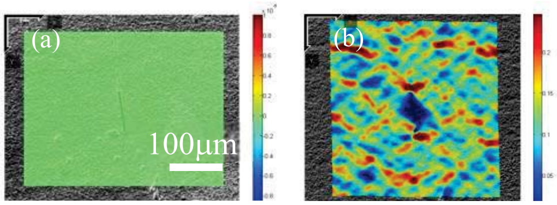

Fig.4 DIC result of the In film[(a)0% strain, (b)25% strain] on Cr/PDMS for the same strain as (a,b) showing strain localization at multiple numerous periodically spaced bands[21].

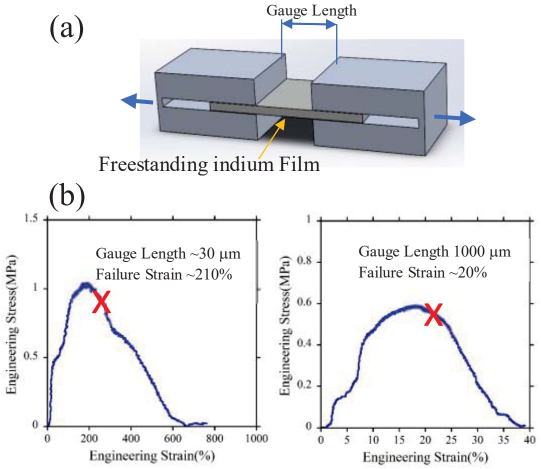

Fig.5 (a) Schematic for investigating effect of gauge length on the stretchability of freestanding In films. (b) Stress-strain curves for freestanding In films stretched to failure. The strain to failure increases from 20% to 210% when the effective gauge length is decreased from 1000µm to 30µm. The sample thickness was 47µm and width 6.12mm.

proximity to each other while stretched to 35-40% strain where the gaps between the islands were 20nm-40μm.

In addition to the above experiments, effect of gauge length on the stretchability of In film over the narrow gaps between neighboring Cr islands was found to be much pronounced in the films with shorter effective gauge length. Fig.5 shows engineering stress-strain curve where evidence of this effect is shown by comparing two different cases with different gauge lengths. For the shorter gauge length, the failure strain was found to delayed which resulted in higher deformability of the freestanding In film.

DISCUSSION

Based on previous findings [22], where it was observed that a shorter gauge length resulted in a higher work hardening exponent(n), it is hypothesized that since the true strain to failure will be equivalent to n. This ultimately resulted in a delay in plastic instability and therefore enhanced overall ductility of the In film. One of the reasons that was mentioned in the finding was a shorter gauge length might have led to pile-ups of dislocations along the lines where the slip plane intersected the boundaries between the grip sections and the gauge.

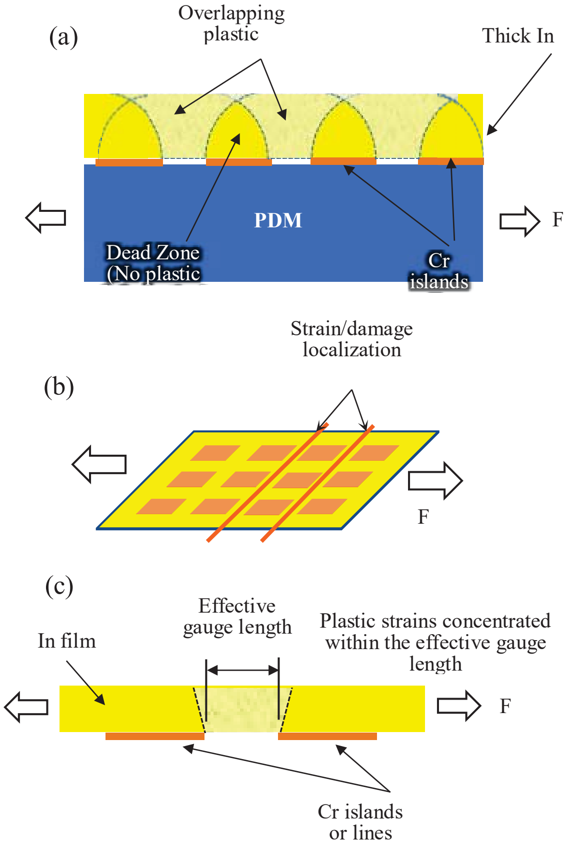

Fig.6 (a,b) Schematic of metal interconnect periodically bonded to PDMS and stretched to large strain showing overlapping plastic strain fields and (c) effective gauge lengths.

As shown in Fig.4, large strains accumulate in the In just above the neighboring Cr gaps where it was not bonded to the PDMS substrate. If it is assumed that most of the strains are being concentrated in between these gaps only at the bottom surface of the In film and spread out beyond the gaps throughout the entire films as the distance from the bottom increases, specifically as the externally applied strain increases (Fig.6a), the strain induced in the film is still several hundred percent. However, plastic instability in bulk In within a few tens of percentage of strain that results in reduced fracture strain. This suggests that a length scale effect on the onset in plastic instability exists because of the small gaps between the Cr islands which ultimately may have helped achieve higher overall strain in the overlaying In films.

CONCLUSION

In the paper, we demonstrated successful stretching for inherently ductile metallic interconnect films with periodic adhesion layer showing an unique approach towards engineering metal-polymer system that can be potentially used for flexible electronic devices. We hypothesized the effect of effect gauge length in shorter scale. We used digital image correlation and carried out systematic experimentation to gain more insight into the mechanics and mechanism of the unusually large strain in the In film while bonded to the polymer substrate. However, further experimentations at different length scales will be carried out in future and will be part of another paper.

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REFERENCES

[1] Gartner Report at http://www.gartner.com/, 2014.
[2] J.-H. Ahn, J.H. Je, Stretchable electronics: materials, architectures and integrations, Journal of Physics D: Applied Physics 45(10) (2012) 103001.
[3] T. Adrega, S.P. Lacour, Stretchable gold conductors embedded in PDMS and patterned by photolithography: fabrication and electromechanical

characterization, Journal of Micromechanics and Microengineering 20(5) (2010) 055025.

[4] H. Yung-Yu, K. Lucas, D. Davis, B. Elolampi, R. Ghaffari, C. Rafferty, K. Dowling, Novel Strain Relief Design for Multilayer Thin Film Stretchable Interconnects, Electron Devices, IEEE Transactions on 60(7) (2013) 2338-2345.

[5] Y.-Y. Hsu, M. Gonzalez, F. Bossuyt, F. Axisa, J. Vanfleteren, I. De Wolf, The effect of pitch on deformation behavior and the stretching-induced failure of a polymer-encapsulated stretchable circuit, Journal of Micromechanics and Microengineering 20(7) (2010) 075036.

[6] R. Taylor, C. Boyce, M. Boyce, B. Pruitt, Planar patterned stretchable electrode arrays based on flexible printed circuits, Journal of Micromechanics and Microengineering 23(10) (2013) 105004.

[7] Y. Zhang, H. Fu, Y. Su, S. Xu, H. Cheng, J.A. Fan, K.-C. Hwang, J.A. Rogers, Y. Huang, Mechanics of ultra-stretchable self-similar serpentine interconnects, Acta Materialia 61(20) (2013) 7816-7827.

[8] Y. Zhang, S. Wang, X. Li, J.A. Fan, S. Xu, Y.M. Song, K.J. Choi, W.H. Yeo, W. Lee, S.N. Nazaar, Experimental and theoretical studies of serpentine microstructures bonded to prestrained elastomers for stretchable electronics, Advanced Functional Materials 24(14) (2014) 2028-2037.

[9] Y.-Y. Hsu, M. Gonzalez, F. Bossuyt, F. Axisa, J. Vanfleteren, I. De Wolf, The effects of encapsulation on deformation behavior and failure mechanisms of stretchable interconnects, Thin Solid Films 519(7) (2011) 2225-2234.

[10] M. Jablonski, F. Bossuyt, J. Vanfleteren, T. Vervust, H. de Vries, Reliability of a stretchable interconnect utilizing terminated, in-plane meandered copper conductor, Microelectronics Reliability 53(7) (2013) 956-963.

[11] M. Gonzalez, B. Vandevelde, W. Christiaens, Y.-Y. Hsu, F. Iker, F. Bossuyt, J. Vanfleteren, O. Van der Sluis, P. Timmermans, Design and implementation of flexible and stretchable systems, Microelectronics Reliability 51(6) (2011) 1069-1076.

[12] C. Lv, H. Yu, H. Jiang, Archimedean spiral design for extremely stretchable interconnects, Extreme Mechanics Letters (2014).

[13] S. Béfahy, S. Yunus, T. Pardoen, P. Bertrand, M. Troosters, Stretchable helical gold conductor on silicone rubber microwire, Applied Physics Letters 91(14) (2007) 141911.

[14] J.A. Rogers, T. Someya, Y. Huang, Materials and mechanics for stretchable electronics, Science 327(5973) (2010) 1603-1607.

[15] E. Kim, H. Tu, C. Lv, H. Jiang, H. Yu, Y. Xu, A robust polymer microcable structure for flexible devices, Applied Physics Letters 102(3) (2013) 033506.

[16] D.-Y. Khang, H. Jiang, Y. Huang, J.A. Rogers, A stretchable form of single-crystal silicon for high-performance electronics on rubber substrates, Science 311(5758) (2006) 208-212.

[17] N. Lu, X. Wang, Z. Suo, J. Vlassak, Metal films on polymer substrates stretched beyond 50%, Applied Physics Letters 91(22) (2007) 221909.

[18] Y. Xiang, T. Li, Z. Suo, J.J. Vlassak, High ductility of a metal film adherent on a polymer substrate, Applied Physics Letters 87(16) (2005) 161910.

[19] A. Vohra, R.S. Carmichael, T.B. Carmichael, Developing the Surface Chemistry of Transparent Butyl Rubber for Impermeable Stretchable Electronics, Langmuir 32(40) (2016) 10206-10212.

[20] Y. Arafat, I. Dutta, R. Panat, Super-stretchable Metallic Interconnects on Polymer with a Linear Strain of up to 100%, Applied Physics Letters 107 (2015) 081906.

[21] Y. Arafat, I. Dutta, R. Panat, On the Deformation Mechanisms and Electrical Behavior of Highly Stretchable Metallic Interconnects on Elastomer Substrates, Journal of Applied Physics 120 (2016) 115103.

[22] D. Kiener, W. Grosinger, G. Dehm, R. Pippan, A further step towards an understanding of size-dependent crystal plasticity: In situ tension experiments of miniaturized single-crystal copper samples, Acta Materialia 56(3) (2008) 580-592.