Flexible, Strain Gated Logic Transducer Arrays Enabled by Initializing Surface Instability on Elastic Bilayers

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Developing flexible sensors with a high strain sensing range could enable widespread downstream applications, by allowing intimate, mechanically conformable integration with soft biological tissues. By characterizing interconnected metal electrode arrays on super-flexible substrates, we have established a surface deformation control strategy of an array of strain transducers. The strain gated switches are capable of measuring various compressive strains (up to 60\%) by bringing metal electrodes into self-contact via creasing elastic instability beyond a threshold substrate strain. The designed devices have been developed to explore the geometry design effect on the electrode-elastomer “stiff film on soft elastomer” surface deformation. The enabled transducer array yielded a stepwise strain-electrical resistance switching mechanism which opens up the potential of future interconnected sensor array type of super-compressible devices.

Flexible electronics and transducers where devices are subject to stretching, bending and twisting forces have grown into one of the more interesting technologies for next generation applications such as bio-medical electronics, smart skin, wearable sensors, epidermal electronics, sensors and actuators\textsuperscript{1,2,3,4,5,6,7,8,9,10,12,16}. Recent research on elastic substrates complying with local features such as metal interconnects and integrated transducers has shown great potential to withstand high strain deformation during bending, compressing and stretching\textsuperscript{7,12,13,14}. Such structured elastic surfaces under compressions and stretching can undergo various deformations such as wrinkling, creasing, folding and buckling, which generates interests in

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engineering applications in sensing and actuation\textsuperscript{11,15,16,17,18,19,20,21,22,23,24,25}. Among them, recently developed metal-elastomer strain gated transducers have utilized mechanically gated super-flexible electrical switches to provide sensing mechanisms for wearable electronics\textsuperscript{5, 8, 9, 23}.

Controllably and reversibly generating creasing and other instability patterns on the surfaces of soft materials by electrical\textsuperscript{26}, temperature\textsuperscript{27}, mechanical\textsuperscript{28} and electrochemical\textsuperscript{29} stimulation have attracted considerable interest in developing them into sensing and actuation applications. One of the latest super-compressible (up to 60\%) strain-gated electrical switching sensing mechanisms utilizes surface instabilities such as wrinkles and creases on soft elastomer substrates under compression with stiff film patterns on top (e.g. metal electrodes)\textsuperscript{5}. Such devices with single-pair of metal “finger” electrodes relying on the controllable formation of surface creases to bring initially disconnected regions of the metal electrode into self-contact was previously reported\textsuperscript{5}. When compressed (x-direction in Fig. 1), the stiff Au film (Young’s Modulus 50 - 70 GPa) wrinkles due to its near inextensibility. The much softer (Young’s Modulus 0.4 - 4 MPa) substrate areas in the gap between Au electrodes can have greater deformation due to the local amplification of strain by the stiff films nearby. At higher compression, a crease forms in this gap between electrodes, generating the desired the electrodes self-contact switching mode\textsuperscript{5}. The measured resistance displayed a significant step change from $\sim 10^{13}$ $\Omega$ to $\sim 10^{2}$ $\Omega$ after the self-contact of the electrodes after the substrate compression strain surpass threshold values\textsuperscript{5}. The switching threshold strains can be controlled by geometry design (e.g. Au electrode width $W_\ell$, Length $L_\ell$, and gap width $L_g$) as well as material and structural properties\textsuperscript{5}, and recent research in surface instabilities has shown the deformation mode can also be controlled via such designs\textsuperscript{21}. Moreover, the latest study into surface instabilities on such heterogeneous surfaces with patterned regions of different materials (e.g. stiff metal electrodes and soft elastomers) opens the possibilities for research into advanced surface morphing and more complex application devices\textsuperscript{21}.

For future development of transducer arrays applied to a larger area under compression, it is important to develop electrode interconnect technologies which should ultimately enable row-column addressing. This paper discusses a multi-finger switching mechanism with the controllable competing elastic instability growth on super-flexible surfaces, by design and micro-engineering interconnect more complex Au on Polydimethylsiloxane-Vinylpolysiloxane (PDMS-VPS) bilayer structures. Such structure was fabricated by dual-SAM (self-assembly monolayer) assisted metal transfer process which transfers photolithographically patterned Au electrodes (thickness = 74nm) from silicon to the PDMS-VPS substrates (Fig. 1a) with details in the supplementary document, providing a stepwise resistance-strain response sensing mechanism which could shed a light on the future applications in widespread downstream applications, tunable and stretchable electronics \textsuperscript{24-28}. This work also experimentally studied instabilities (patterned stiff and soft regions) on heterogeneous surfaces under large compression strains (up to 60\%) which could help the development in related theoretical studies, such as initiated in \textsuperscript{21}. 

2
The designed and fabricated transducer arrays and interconnects are shown in Figure 1. Each of the paired Au finger electrodes based on previous design with key dimensions (e.g. finger electrode width $W_f$, length $L_f$, and gap width $L_g$) were interconnected by two different configurations. Fig. 1(b) shows SEM image (Tescan® Mira3) of the bulk interconnect configuration linking 4 pairs of finger electrode transducers (labelled F1 to F4) with 2 contact electrodes (labelled E). Fig. 1(c) shows the serpentine interconnect configuration which helped to protect the Au electrodes from damage caused by perpendicular direction stretching due to Poisson effect (details in supplementary document). This research focuses on the relationship between geometry design parameters such as $W_f$, $L_f$, and $L_g$ and 2D (top-view) deformation for the Au electrodes and gaps in-between at different substrate compression strains $\varepsilon_{sub}$. Other parameters such as contact electrode length of $L_e$, and the adjacent serpentine interconnect spacing $W_e$ (Fig. 1c) were also considered.

The uniaxial (x-direction) substrate compression was provided by releasing the pre-stretched PDMS-VPS mounting layer from $L_0$ to $L$. Wrinkles on Au finger electrodes started to develop at low strain, and eventually both wrinkles on Au and creases on PDMS after substrate strain $\varepsilon_{sub} = (L_0-L)/L_0$ went beyond threshold as illustrated in Fig. 2a. Figure 2a and 2b shows that the reversible wrinkling process on Au has reduced finger length from $L_{g0}$ to $L_f$, resulting a local strain change $\varepsilon_{Au-x} = (L_{g0}-L_f)/L_{g0}$. Meanwhile as discussed, the gap area in-between (softer PDMS surface) will have its local strain amplified due to surrounding stiffer film patterns with $\varepsilon_{Gap-x} = (L_{g0}-L_g)/L_{g0}$.

Fig. 2c shows the Au wrinkling 3D profiles obtained by Atomic Force Microscopy (AFM Bruker™ 3100) scan with which progressively grew under different substrate strains. Based on the critical wrinkling strain of $\varepsilon_w=0.25\left(\frac{3E_s}{E_f}\right)^{2/3}$, the expected theoretical $\varepsilon_w = 6.7 \times 10^{-4}$ calculated based on the elastic plane-strain moduli of $E_f = 7 \times 10^{10}$ Pa and 74nm gold film of $E_s = 4 \times 10^5$ Pa. In reality, the wrinkles started to form at a substrate strain $\varepsilon_{sub} = 9 \times 10^{-3}$. This indicates and confirms that the local strains on stiff Au electrodes significantly lagged behind those of the substrate and soft PDMS gap areas, which is welcomed as a helpful technique to protect metal films under large compression strains. The wrinkles on Au continuously grew under further x-direction compression, covering the majority of the Au electrodes at the substrate strain of 0.12. The wrinkle wavelength at substrate strain $\varepsilon_{sub} = 0.12$ was calculated to be 17µm according to $\lambda_0 = (2\pi h_f(\frac{E_f}{3E_s})^{1/3}$, which closely agreed with the actual measured value of 18µm (shown in supplementary document).

To further understand this local strain difference between Au and PDMS areas against the substrate strain, characterization experiments were designed and conducted using Nikon® LV-100 optical microscope. We focused on local strains of each Au finger electrodes $\varepsilon_{Au-x} = (L_{g0}-L_g)/L_{g0}$, as well as PDMS gap area $\varepsilon_{Gap-x} = (L_{g0}-L_g)/L_{g0}$ between the paired finger electrodes. The
lengths data $L_{g0}$, $L_f$ and $L_{g0}$, $L_g$ were all measured by the Nikon system from top view. The relationships between $\varepsilon_{Au-x}$, $\varepsilon_{Gap-x}$ and $\varepsilon_{sub}$ were comparatively studied against other key geometry design parameters of the Au electrodes such as $L_{g0}$, $L_g$, finger width $W_f$ and locations in the transducer array (E, F1 to F4).

Figure 3a shows the relationship between the Au local strains against the substrate strains. The $\varepsilon_{Au-x}$ always lagged behind the $\varepsilon_{sub}$ which is indicated by the dash line. The strain gap was observed to be constantly ~0.03, until $\varepsilon_{sub} = 0.45$ where this gap increased to around 0.05 when large creases started to appear on PDMS surfaces which absorbed more strain energy. This has confirmed previous assumption that the local strains on stiff Au electrodes significantly lagged behind those of the substrate. Together with the reversible wrinkling mechanism, most Au electrodes remain intact after > 10 cycles of repeated compression. Essential to determine the switching strain values that brings Au finger pair electrodes into self-contact, Fig. 3b to 3d show detailed comparative study results of the PDMS gap strain $\varepsilon_{Gap-x}$ versus the substrate strain $\varepsilon_{sub}$. When $\varepsilon_{Gap-x} = 1$ at $L_g = 0$, it was an indication that the PDMS area in the gap was completely folded into the crease from the top view, as illustrated in Fig. 2a. The corresponding value of $\varepsilon_{sub}$ at this point is close to the switching strain.

Fig. 3b shows the results comparison between different finger electrodes (F1 to F4) on the same transducer array where $W_f = 35 \mu m$, initial $L_{g0}$ ranges from 225 \mu m to 250 \mu m, and $L_{g0}$ from 10 \mu m to 55 \mu m respectively. All $\varepsilon_{Gap-x}$ are far greater than the $\varepsilon_{sub}$ (indicated by dash line) due to the expected strain amplification effect. It can be observed for pair electrode F1 with smallest $L_{g0}$, the self-contact strain is around $\varepsilon_{sub} = 0.08$, significantly lower than typical creasing strains of around 50% on plain PDMS surfaces\cite{5,20}. As $L_g$ gradually increased, it required much higher substrate strains to bring F2 and F3 pair electrodes into self-contact, at $\varepsilon_{sub} = 0.2$ and $\varepsilon_{sub} = 0.5$ respectively. For F4 electrodes, it is understandable that the large $L_{g0}$ made it impossible for the pair of electrodes to reach each other before the 0.6 substrate strain limit (Fig. 3b), due to insufficient self-contact depth (estimated to around 19 \mu m) before reaching the limit. Since the natural wavelength of crease is ~ 3.5 times of substrate thickness $H$ ($H=110 \mu m$ in this case), multiple creases may occur at larger distances, which prevents the electrodes from achieving contacts even at higher strains.

We also compared finger electrode width $W_f$ (25 \mu m, 35 \mu m and 50 \mu m) effect on gap strain $\varepsilon_{Gap-x}$ as shown in Figure 3c and 3d. The initial gap distance $L_{g0}$ for F2 electrodes (Fig. 3c) was kept at 15 \mu m with $L_{g0}$ for F3 (Fig. 3d) kept at 25\mu m. The electrode lengths $L_{g0}$ are 250 \mu m and 245 \mu m respectively. It was clearly observed in both cases that electrodes with narrow $W_f$ (25 \mu m) had much lower $\varepsilon_{sub}$ of 0.1 and 0.4 respectively, when electrode pair made self-contact. This suggests a wider Au
electrode may require more energy to be pulled into the creasing created in the PDMS gap area, since the electrode prefers small wrinkling.

Based on the above deformation study, we demonstrated multi-step or “stepwise” electrical resistance change corresponding to different levels of $\varepsilon_{\text{sub}}$. The Everbeing EB8 manual probe station (with EB-05 probes) which connected to a Keithley® 4200 analyzer (I-V mode, -1V to +1V sweep, with 0.2V/step) was used to characterize the resistance value change. As shown in Figure 4, the step-wise strain-resistance sensing has been achieved by surface elastic instability induced multi-finger electrodes with different distances between the electrodes. For the designed $L_g$ values ($L_f = 510 \ \mu m$ and $520 \ \mu m$ respectively), then the estimated finger electrode resistance would be in the region of $50 \ \Omega$. When the first pair electrodes were in contact, the calculated resistance of would be expected to be $\sim 100 \ \Omega$, assuming the contact resistance is zero. This will be reduced to $50 \ \Omega$ when the second switching being achieved. Fig 4b shows when the substrate strain ranged greater details of the two-step switching achieved. At between 0.45 and 0.52, the first step switching stage was achieved, with a resistance of $\sim 120 \ \Omega$. Under further compression, the second step switching stage has happened at the substrate strain range of $0.54 < \varepsilon_{\text{sub}} < 0.58$ by measuring the resistance to be $\sim 50 \ \Omega$. Note the error bars indicate multiple measurements at different current levels that in most cases indicate that Joule heating is not influencing the measurement. The probe-Au electrode contact resistance was characterized to be $\sim 7.9 \ \Omega$ with a standard deviation of 1.17. The variation of the experimental resistance values may mainly be caused by the contact resistance between the touching Au finger electrodes. The corresponding substrate strains at switching was also in good agreement with the electrode deformation observations.

In conclusion, a new strategy has been established to detect the multiple strain values on a soft elastic substrate to study the metal-elastomer deformation behaviour of a super-compressible strain transducer array. We have characterised the relationship between electrode geometries and compression strain ratios, together with the growth and co-existence of wrinkles and creases on multi-switching electrodes. Multi-steps of electrical resistances were generated at different switching strains on an individual device. This has been demonstrated by using the multiple finger electrode soft electronics with different distances between the electrodes, which has potential for future stretchable/epidermal electronics, flexible sensors, health monitoring, and wearable device applications.
FIG. 1. (a) Schematic (middle) and microscopic image (left) of patterned multi-switching Au strain transducer array, with Au patterns transferred from silicon to PDMS-VPS substrate by dual-SAM (self-assembly monolayer) assisted metal transfer process; and SEM top-view images showing two different interconnect configurations: (b) 4 pairs of finger transducers “F” interconnected with two bulk contact electrodes “E”; (c) 4 pairs of finger transducers interconnected with contact electrodes by serpentine structures. (Scale bars = 200 µm)
FIG. 2. (a) 3D Schematics showing the Au wrinkling - PDMS creasing 3D deformation process under uniaxial substrate compression – pair finger electrodes into self-contact due to PDMS creasing, and wrinkling on Au finger electrodes; (b) Top view optical microscopic image showing local strain changes on Au due to wrinkling, and PDMS gap area due to creasing; (c) AFM scan 3D surface profile view of the wrinkle development on Au electrode at various substrate strains for a device with $W_f = 25 \, \mu m$, $L_g = 50 \, \mu m$ and $L_f = 225 \, \mu m$. 
FIG. 3. Local strain change comparisons: (a) Au electrodes strain vs. substrate strain comparing F1 to F4, and contact pads E1; (b) The PDMS gap strain vs. substrate strain comparing between electrodes with varied $L_{g0}$ and $L_{g1}$; (c-d) Finger electrode widths $W_f$ effect on the PDMS gap strain vs. substrate strain for F2 and F3 electrodes; all dashed lines in Figures indicate substrate strain value as a reference.
FIG. 4. (a) Resistance of the test structure as a function of strain. The resistance change during the two-stage switching are highlighted in a red ring and detailed in (b) Two-stage resistance switching strain sensing: Resistance of the transducer array as a function of strain during the two-stage switching period ($0.45<\varepsilon<0.52$, and $0.54<\varepsilon<0.6$).

SUPPLEMENTARY MATERIAL

This supplementary materials is available online, and contains the experimental and fabrication method, as well as the additional results to support the results and claims in the main context.
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1 D.-H. Kim, N. Lu, R. Ma, Y.-S. Kim, R.-H. Kim, S. Wang, J. Wu, S. M. Won, H. Tao, A. Islam, K. J. Yu, T.-I. Kim, R. Chowdhury, M. Ying, L. Xu, M. Li, H.-J. Chung, H. Keum, M. McCormick, P. Liu, Y.-W. Zhang, F. G. Omenetto, Y. Huang, T. Coleman and J. A. Rogers, Science. 333, 838-843 (2011).
2 W. Wu, X. Wen and Z. L. Wang, Science. 340, 952 (2013).
3 W. Gao, S. Emaminejad, H. Y. Nyein, S. Challa, K. Chen, A. Peck, H. M. Fahad, H. Ota, H. Shiraki, D. Kiriya, D.-H. Lien, G. A. Brooks, R. W. Davis and A. Javey, Nature. 529, 509 (2016).
4 S. R. Madhvapathy, Y. Ma, M. Patel, S. Krishnan, C. Wei, Y. Li, S. Xu, X. Feng, Y. Huang and J. A. Rogers, Adv. Funct. Mater. 28, 1802083 (2018).
5 B. Xu, D. Chen and R. C. Hayward, Adv. Mater. 26, 4381-4385 (2014).
6 D.-Y. Khang, J. A. Rogers and H. H. Lee, Adv. Funct. Mater. 18, 1-11 (2008).
7 T. Pan, M. Pharr, Y. Ma, R. Ning, Z. Yan, R. Xu, X. Feng, Y. Huang and J. A. Rogers, Adv. Funct. Mater. 27, 1702589 (2017).
8 C. Wang, B. B. Xu, J.G. Terry, S. Smith, A. J. Walton and Y. Li, Proceedings of the 30th International Conference on Microelectronic Test Structures, Grenoble, France, 28 March-30 March 2017, pp. 150-155.
9 C. Wang, B. B. Xu, J.G. Terry, S. Smith, A. J. Walton and Y. Li, 1st IEEE International Flexible Electronics Technology Conference, Ottawa, Canada, 7-9 August 2018.
10 Y. Chu, X. Wu, J. Lu, D. Liu, J. Du, G. Zhang and J. Huang, Adv. Sci. 3, 1500435 (2016).
11 D. Chen, L. Jin, Z. Suo and R. C. Hayward, Mater. Horiz. 1, 207 (2014).
12 C. Wang, C. Wang, Z. Huang and S. Xu, Adv. Mater. 1801368 (2018).
13 X. Xue, S. Wang, C. Zeng, L. Li and C. Li, Surf. Interface Anal. 50, 180-187 (2018).
14 K. E. Crawford, Y. Ma, S. Krishnan, C. Wei, D. Capua, Y. Xue, S. Xu, Z. Xie, S. M. Won, L. Tian, C. Webb, Y. Li, X. Feng, Y. Huang and J. A. Rogers, Extreme Mechanics Letters. 22, 27-35 (2018).
15 Y.-C. Chen and A. J. Crosby, Adv. Mater. 26, 5626-5631 (2014).
16 C. F. Guo, Q. Liu, G. Wang, Y. Wang, Z. Shi, Z. Suo, C.-W. Chu and Z. Ren, PNAS. 112, 12332 (2015).
17 X. Guo, X. Wang, D. Ou, J. Ye, W. Pang, Y. Hung, J. A. Rogers and Y. Zhang, npj Flexible Electronics. 14, (2018).
18 A. Auguste, L. Jin, Z. Suo and R. C. Hayward, Extreme Mechanics Letters. 11, 30-36 (2017).
19 D. Wang, N. Cheewaruangroj, Y.Li, G. Mchale, Y. Jiang, D. Wood, J. S. Biggins and B. B. Xu, Adv. Funct. Mater. 28, 1704228 (2018).
20 D. Chen, S. Cai, Z. Suo and R. C. Hayward, PRL. 109, 038001 (2012).
21 T. Ouchi, J. Yang, Z. Suo and R. C. Hayward, ACS Appl. Mater. Interfaces. 10, 23406-23413 (2018).
22 K. Nan, H. Luan, Z. Yan, X. Ning, Y. Wang, A. Wang, J. Wang, M. Han, M. Chang, K. Li, Y. Zhang, W. Huang, Y. Xue, Y. Huang, Y. Zhang and J. A. Rogers, Adv. Funct. Mater. 27, 1604281 (2017).
23 G. Lee, T. Lee, Y. W. Choi, P. V. Pikhitsa, S. J. Park, S. M. Kim, D. Kang, and M. Choi, J. Mat. Chem. C. 5, 10920 (2017).
24 D. Rhee, W-K. Lee, and T. Odom, Angew. Chem. Int. Ed., 56, 6523(2017).
25 H. Lv, Y. Liu, B. Xu, D. Hui and Y. Fu, Composites Part: B, 122, 9 (2017).
26 B. Xu and R. Hayward, Advanced Materials, 25, 5555 (2013).
27 J. Kim, J. Yoon, and R. Hayward, Nature Materials, 9, 159 (2010).
28 S. Cai, D. Chen, Z. Suo, and R. Hayward, Soft Matter, 8, 1301 (2012).
29 B. Xu, Q. Liu, Z. Suo and R. Hayward, Adv. Funct. Mater., 26, 3218 (2016).