Mimosa-inspired design of flexible pressure sensor with touch sensitivity

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([Optional Dedication])

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Keywords: Bio-inspired, mimosa, Flexible, Pressure sensor, Tactile sensor

Abstract

The ability of fabricating active materials with defined microstructures on soft elastomeric substrates is critical in flexible pressure sensors to achieve high sensitivity and quick response time. Previous strategies largely depend on the top-down technique (i.e. by molding from laser-etched silicon patterns), which is known to be a time-consuming, high cost process with limited scalability. Here, we report on a new bio-inspired approach to fabricate flexible, highly sensitive pressure sensors, offering benefits of simplicity, low cost, scalability. The key innovation is that we take advantage of unique surface microstructures of natural mimosa leaves and apply them in a simple yet efficient, environment-friendly and easy scale-up molding process for fabricating flexible pressure sensors. We achieved a high sensitivity (50.17 kPa⁻¹), quick responding time (< 20 ms), and high durability (negligible loading-unloading signal changes over 10,000 cycles). Our bio-inspired strategy opens a new route to fabricate flexible electronic devices at large scale with low cost.
1. Introduction

Substantial recent research interest has been directed to emerging unconventional flexible/stretchable electronics. One device of particular interest is flexible pressure sensors which have broad technological implications for areas ranging from wearable electronics\textsuperscript{[1]}, real-time health monitoring\textsuperscript{[2-4]} to soft robots and prosthetic devices\textsuperscript{[5,6]}. Among various flexible pressure sensors developed recently\textsuperscript{[7-13]}, resistance-type ones are facile to produce at large-scale and in a cost-effective manner due to their relatively simple structures and fabrication process\textsuperscript{[14]}. Typically, flexible pressure sensors can be constructed by embedding conducting fillers into soft elastomeric materials forming 3D percolating networks\textsuperscript{[15-18]}.

In addition, it is often important to introduce microstructures in elastomeric electrode surfaces, such as pyramid\textsuperscript{[19]}, micropillar\textsuperscript{[20]}, interlocking\textsuperscript{[21]} and aligned\textsuperscript{[22]} nanofibers, and microgroove\textsuperscript{[23]}. Such micro-structured surfaces contribute to the enhanced sensitivity with low responding time\textsuperscript{[19]}. Despite this, the fabrication of above-mentioned surface microstructures largely depends on the traditionally lithography technique (i.e. by molding from laser-etched silicon patterns), which is a time-consuming, high-cost process with limited scalability.

Herein, we report on a new bio-inspired strategy to fabricate flexible pressure sensors with touch sensitivity. Inspired by mimosa – a plant can close their leaves under external stimuli, we demonstrate a simple, efficient, low-cost and scalable direct-molding method to flexible pressure sensors which are sensitive enough to mimic mimosa leaves. In the regime of 0 – 70 Pa, a sensitivity of 50.17 kPa\textsuperscript{-1} was obtained, which is higher than that for many flexible pressure sensors reported recently\textsuperscript{[19-22]}. Our bio-inspired sensor could respond to pressure changes within 20 ms at a frequency up to 9 Hz, simultaneously with negligible loading-unloading signal changes over 10,000 cycles. Besides pressing, our bio-inspired pressure sensors could be utilized to detect bending and torsional forces. Furthermore, an artificial touch-sensing “mimosa leaves” has been demonstrated. We believe that our mimosa-inspired
approach opens a new route to fabricate flexible pressure sensors in a cost-effective and scalable manner.

2. Result and discussion

After undergoing billions of years of evolution and natural selection, nature has gained the wisdom on how to get maximal functions at the cost of minimum resources\(^{[24,25]}\). Microstructures widely exist upon the surfaces of natural creatures\(^{[26]}\). Thanks to these unique surface constructions, animals or plants can self-repel the water permeation\(^{[27]}\), obtain highly dry adhesion\(^{[28]}\), and exhibit stimuli-responsive behaviors. *Mimosa*, a plant can close their leaves under external stimuli, is a natural example of flexible pressure sensors. Their leaves are bipinnately compound with one pair of pinnae that consists of ~15 leaflets (Figure 1a). From the scanning electronic microscopy (SEM) observation of a usual leaflet, an irregular pattern of microdomains with an average diameter of 18.4 ± 6.1 µm and height of 16.1 ± 3.7 µm can be found upon the leaflet (Figure 1b-d). Owing to the existence of these protuberant microdomains, a gentle finger touching could be transferred into a biologically electric signal, leading to the release of water from the leaf cell vacuoles\(^{[29]}\). Then, thousands of plant cells would collapse. As a result, the leaflets will close within nearly three seconds (Supplemental Movie S1). Inspired from these “natural tactile sensors”, we demonstrate a new strategy to fabricate flexible pressure sensors in a simple yet efficient matter.

To mimic the stimuli-responsive performance of mimosa leaves, a square piece of pre-cleaned mimosa leaflet (4×1 cm\(^2\)) was used as the template for fabricating micropatterned polydimethylsiloxane (PDMS) thin film, as illustrated in Figure 2a. PDMS was chosen as the device substrate in this study due to its good elastic properties, its biomedical compliance with human tissue, and its high performance as a dielectric material in flexible electronics\(^{[30]}\). After two-step negative/positive molding from the mimosa leaflet, PDMS films can be endowed with precise surface microstructures as the plant owns. The thickness of the PDMS
thin film was controlled in the range of 300 ± 40 µm. The morphology and microstructure of structured PDMS film were observed by SEM with different view angles (Figure 2c,d), which show similar microdomain patterns to that of the mimosa leaflet (Figure 1b,c). After generating rough and flexible PDMS substrates, a thin layer of metal coating (~5 nm thick titanium plus ~30 nm thick gold) has been deposited upon the microstructure to effectively transport the electrons (the resistance is 110 ± 14 Ω sq⁻¹). Thinner gold layers would yield unstable/high resistance (Supplemental Figure S1) while thicker ones might waste electrical energy. Hence, an optimum ~30 nm thick gold layer was used in this study. Consequently, two layers of coated PDMS films with the microstructured surfaces have been placed face-to-face, and conductive wires were anchored on the edge of both films as the source-drain electrodes (Figure 2b and device details can be found in Supplemental Figure S2). Therefore, a mimosa-inspired flexible pressure sensor has been fabricated. The reason for utilizing this rough-to-rough type device construction is due to its high impedance change (Supplemental Figure S3). Under 156 Pa applied pressure, rough-to-rough type device has generated more than 0.6 mA current increase while the flat-to-flat counterpart showed a tiny current change (~0) and rough-to-flat one exhibited smaller change (~0.4 mA).

The sensing mechanism of the device is attributed to pressing-force-dependent contact between top and bottom rough surfaces. Unlike flat thin films, a small compressive deformation of microstructured PDMS surfaces would happen when applied an external pressure, yielding increased conductive pathways and corresponding current enhancement (Figure 2e). On unloading, these microstructured PDMS films would recover to their original shapes, leading to the decrease of the current to the primary value.

The pressure sensitivity (S) of the bio-inspired polymer sensor has been tested by utilizing a computer-controlled stepping motor and a force sensor, shown in Figure 3a. The sensitivity S can be defined as:
\[
S = \frac{I - I_{\text{off}}}{\Delta P} \tag{1}
\]

Where \(I\) is the current when applied pressure on the devices, and \(I_{\text{off}}\) is the current of device with only base pressure; \(\Delta P\) is the change of applied pressure.

The points in Figure 3a can be divided into two regions based on difference in sensitivity. In the low pressure region (purple region, 0-70 Pa), the sensitivity of the device is 50.17 kPa\(^{-1}\), which is much higher than that of the relatively large pressure region (1.38 kPa\(^{-1}\) in yellow region, 200-1500 Pa). The separated fittings of these points in both regions showed good linear behaviors (\(R_{\text{purple}}^2 = 0.943\), and \(R_{\text{yellow}}^2 = 0.961\), see Supplemental Figure S4a,b), indicating the device can serve as a reliable pressure sensor. The reason for different sensitivities can be explained on the basis of the previous reports\(^{[19-23]}\). In low-applied-force purple region, the pressure increase resulted in decreased gap between top/bottom microdomains, yielding close contact between the microstructures. In contrast, it is quite possible that the gap distance has already reached zero from the turning point in yellow region, and further increasing pressure only works for enhancing the microdomain deformation. In other words, the contact area tended to saturate in yellow region, and the applied pressure has little effect on the current variation. This trend became more obvious in larger pressure ranged from 2 kPa to 10 kPa (see Supplemental Figure S4c). Briefly, high device sensitivity of 50.17 kPa\(^{-1}\) (0-70 Pa) can be generated by this bio-inspired pressure sensor, which is larger than most reported records in flexible pressure sensors\(^{[19-22]}\). So far as we know, only Bao’s group reported a higher sensitivity value \~56.0-133.1 kPa\(^{-1}\) with pressure range from 0 to 30 Pa\(^{[23]}\). Furthermore, the key resource of surface microstructures upon polymers in this study were fabricated by directly molding of natural mimosa leaves, rather than laser-etched silicon patterns\(^{[19-23]}\). Thus, it indicates a simple, environment-friendly and easy scale-up fabrication process of these flexible pressure sensors.
The responses of our bio-inspired sensors to both static and dynamic mechanical pressures were characterized. The sensor exhibited a steady response to static pressure and the resistance under each pressure applied (Supplementary Figure. S5). The resistance became saturated when the applied pressure \( > \sim 1 \text{ kPa} \), this result fits the sensitivity measurement well.

To investigate the pressure range of our bio-inspired sensors towards dynamic forces, a piezoelectric stepping positioner with minimum displacement of only 1 mm was applied to the sensors. As shown in Figure 3b, a pressure of 10.4 Pa could be detected, which indicates the weight of a water droplet (\( \sim 10.4 \mu \text{l} \)) on a surface of 10 mm\(^2\). At the higher pressure range (0.5-1 kPa), the noise-free, stable continuous responses could be observed. The cycling stability of our prepared bio-inspired pressure sensor was tested under a pressure of 156 Pa at a frequency of 1.6 Hz (Figure 3c,d). The consistent resistance change with pressure applied on the surface of the pressure sensor can be maintained after 10,000 loading–unloading cycles, implying long working life and reliability of this bio-inspired pressure sensor.

To investigate the hysteresis/response time of our sensors to applied forces, the output current signals and the dynamic pressure inputs have been compared at a frequency of 0.11-9 Hz (Supplementary Figure. S6). The output electrical signals remained stable without obvious change in amplitude at typical frequencies of 0.34, 4.17 and 9.00 Hz. It should be pointed out that the hysteresis time decreased from \( \sim 1.1 \text{ s} \) to nearly 0 s following the increase of pressure frequency (0.11 Hz to 9 Hz). This hysteresis may be contributed by elastic deformation and viscoelastic effects of microstructured PDMS films during loading-unloading process\(^{[31]}\). Lower frequency indicated slower deformation as well as contact of microstructured polymers, leading to the appearance of the hysteresis time. Furthermore, the response time (\( \sim 20 \text{ ms} \)) of the device has also been observed (Supplementary Figure. S7).

In addition to pressing forces by the piezoelectric stepping machine (Figure 3) or finger touching (Figure 4a), this bio-inspired pressure sensor could simultaneously be utilized to detect the bending and torsional forces (Figure 4b,c). Notably, these response curves could
be distinguished for diverse mechanical forces. For example, the full width at half maximum (FWHM) of responding peaks by torsional forces (~0.5 s) is larger than those by pressure (~0.2 s) or bending forces (~0.3 s). The reason for increased FWHM of curves for diverse forces might attribute to applied-force-dependent contact and recovery between top and bottom microstructured surfaces. For the pressure force, the contact area would focus on the center of finger tip. Thus, the microstructures would recover easily, yielding a small FWHM. Differently, for the bending case, the top/bottom contact model would change from a point to a linear region, indicating increased time to return. In contrast, torsional forces led nearly full contact between these two microstructured layers. Therefore, it would cost the most time to return to its primary condition. Briefly, our bio-inspired sensor can be used to detect different forces and high signal-to-noise ratios existed in all the three types of force measurements, further demonstrating the high sensitivity of this pressure sensor. Besides bending by fingers, this device also exhibited stable responses to various bending angles by employing a stepping machine (Figure 4d).

We further used our bio-inspired pressure sensors to build an “artificial mimosa leaves” with an integrated system of “touching-responding-feedback”, shown in Figure 5a. Three pairs of “bio-inspired pressure sensors” have been cut into natural leaflet shapes, and placed onto a 3D-printed bracing frame (Figure 5b). These flexible pressure sensors were connected with two control parts: the electrical part served as the function of “touching-responding” while the mechanical system worked for the behavior of “responding-feedback”. Furthermore, we tailored the lengths of physically connecting lines to mimic different closing levels of the leaflets. Once a pressure-sensor leaflet has been touched (Figure 5c), the external force could be transferred into a changed electrical signal, and triggered the motor to rotate. This process is similar to the performance of natural mimosa leaves. Owing to the unidirectional rotating of the motor, the lines connected with the leaflet pairs would be tightened up, yielding the closing of “artificial mimosa leaflets” (Figure 5d). This mechanism is to repeat the cell-
collapse-induced closing of natural leaves by utilizing a simple mechanical process. Finally, the whole performance of this “artificial mimosa leaves” has been fulfilled within one second (Supplemental Movie S2), which is quicker than the natural counterpart (Figure 1).

Besides *mimosa*, the leaves of other plants, such as *cymbopogon citratus* or *populus lasiocarpa*, have also been employed to generate flexible pressure sensors in similar processes (Supplementary Figure. S8). Unfortunately, the height of microstructures upon *cymbopogon citratus* was too large (≈78 µm) to deposit continuous electricity active coating, leading to the block of electron transport. *Cymbopogon citratus*, on the other hand, owned smaller microstructure height and enabled successful generation of pressure sensor. However, the current change was smaller than that of *mimosa* at the same applied pressure. In this control experiment, it is obvious that the height of surface microstructures played an important role in deciding the sensor sensitivity. Carefully choosing the plant template type, such as the *mimosa*, would yield great performance of bio-inspired pressure sensors.

3. Conclusion

In summary, we demonstrate naturally microstructured plant leaves can serve as the low-cost and environment-friendly templates for fabricating super-sensitive, rapid-responding and reliable pressure sensors at low cost. These bio-inspired contact-resistance-type pressure sensors, with the advantages of high sensitivity (50.17 kPa⁻¹), quick responding time (< 20 ms), and durable stability (negligible loading-unloading signal changes over 10,000 cycles). Importantly, the whole device fabrication process is facile without the need of complex or expensive equipment. We believe that our methodology opens a new route to low-cost flexible pressure sensors with a wide range of applications in future wearable electronics.

4. Experimental Section
Preparation of Flexible Microstructured PDMS Conducting Films: First, the leaves of plants, such as *mimosa*, *cymbopogon citratus* or *populus lasiocarpa*, have been cut into $4 \times 1$ cm$^2$ (might need several leaves due to their limited surface areas) and washed with deionized water for 10 min, followed by sonication in deionized water for 20 min. Then, they were blown dry with a nitrogen gun to get pieces of pre-cleaned plant templates. Secondly, the fabrication of microstructured PDMS film is performed. Polydimethylsiloxane (Sylgard 184) precursor was mixed with a curing agent in the proportion of 10:1 by weight. The PDMS mixture was put into a centrifuge to remove air bubbles (2000 rpm, 5 min). Then the mixture was poured onto the plant templates. To make great replicating of the leave surfaces, the remaining air bubbles between the PDMS and leaves were removed in a vacuum chamber. After heating at 60 °C for 4 h, the leaves were carefully peeled. Then, a PDMS negative template was modified using fluoride molecules (heptadecafluoroctyltrimethoxysilane) in a decompressed environment at room temperature for 24 h and then heated at 80 °C for 3 h. The fluoride molecules could decrease the surface energy of PDMS negative templates and facilitate the peeling of positive microstructures. Toluene-diluted PDMS (PDMS:toluene = 5:1 w/w) was poured on the modified negative template and then cured at 80 °C for 3 h. The artificial PDMS leaves with similar plant surface microstructures were also carefully peeled off the negative template. Thirdly, to introduce a thin layer of electricity active coating onto the PDMS microstructures, Ti/Au interdigitated coating (Thickness at 5 nm/30 nm) were gradually deposited using an electric beam evaporator (Intlvac Nanochrome II, 10 kV). Finally, two layers of coated PDMS films with the microstructured surfaces have been placed face-to-face, and conductive wires was anchored on the edge of both films as the source-drain electrodes, yielding the generation of bio-inspired flexible pressure sensors. 

Fabrication of “artificial mimosa leaves”: A rigid polymeric bracing frame with three holes has been generated by the 3D-printer (Objet Eden 260V). Then, bio-inspired flexible pressure sensors have been cut into natural leaflet shapes and placed onto the bracing frame.
flexible pressure sensors were connected with two control parts: the electrical part served as the function of “touching-responding” while the mechanical system worked for the behavior of “responding-feedback”. The motor for receiving the electrical signal and giving mechanical feedback was detached from a fan toy purchased from ebay online.

**Characterization.** SEM images were characterized using a FEI NovaNanoSEM 430 operated at 5 kV beam voltage. The sheet resistances of coated PDMS substrates were carried out on a Jandel four point conductivity probe by using a linear arrayed four point head. The current differences and the I-V characteristics for the pressure sensor were recorded by the Parstat 2273 electrochemical system (Princeton Applied Research). For the dynamic low pressure measurement, a piezoelectric stepping positioner (SLC-1730) was used by a custom LabView programme and the force data was measured by an electrical balance (Mettler Toledo NewClassic MF, MS105DU). For the bending investigation, the sensor has been fixed and bent by a stretchable machine (Thorlebs, LTS150/M).

**Supporting Information**
Supporting Information is available from the Wiley Online Library or from the author.

**Acknowledgements**
This research was financially supported under Australian Research Council's Discovery Early Career Researcher Award (DECRA) funding scheme (DE140100541) and Discovery projects funding scheme (DP120100170 and DP140100052). We also acknowledge Romiza Mazid (Monash University, Australia) and Xiaonan Kan (Institute of Chemistry, Chinese Academy of Science, China) for helpful experimental assistances. This work was performed at the Melbourne Centre for Nanofabrication (MCN) in the Victorian Node of the Australian National Fabrication Facility (ANFF).

Received: ((will be filled in by the editorial staff))
Revised: ((will be filled in by the editorial staff))
Published online: ((will be filled in by the editorial staff))
References


Figure 1. “Naturally” flexible pressure sensors of *mimosa* leaves.  a) Digital photographs of mimosa leaves. They are bipinnately compound with one pair of pinnae that consists of ~15 leaflets. A gentle touch by one human finger can yield the leaflets closing within less than three seconds. b) Top and c) side view scanning electronic microscopy (SEM) observations of a usual leaflet, showing the existence of an irregular microdomain pattern. The height of microdomains is ca. 16 µm. d) Statistical histograms of the microdomain diameter (top) and height (bottom) obtained from 50 regions.
Figure 2. Polymer-based flexible pressure sensors learned from the mimosa leaves. a) Schematic illustration of the fabrication process of flexible microstructured PDMS films and corresponding pressure sensors. After two-step negative/positive molding from the mimosa leaflet, PDMS films can be endowed with precise surface microstructures as the plant owns. Then, a thin layer of electricity active coating has been deposited upon the microstructure to effectively transport the electrons. Finally, two layers of coated PDMS films with the microstructured surfaces have been placed face-to-face, yielding a bio-inspired pressure sensor. b) Digital photograph showing the bending ability of the sensor. c) Top and d) side view SEM observations of a molded PDMS film with the similar microstructure to that of mimosa leaves. e) Current changes in responses to pressure and reversibility. The compressive deformation of microstructure could yield the current increase and this change would return to the primary state when removing the external force.
Figure 3. High-sensitivity, stable and reliable flexible pressure sensors. a) Pressure-response plots for the bio-inspired pressure sensor. The structured PDMS films exhibit a high pressure sensitivity of 50.17 kPa$^{-1}$ during 0 and 70 Pa and relatively low pressure sensitivity of 1.38 kPa$^{-1}$ during 200 and 1500 Pa. The inset image is schematic of the experimental set-up. A force sensor and a stepping motor connected with a computer were utilized to control the applied pressure and frequency. b) Plots of current response of the sensor as a function of time (pressure input frequency: 1.6 Hz) for the applied pressures in the range of 10.4 – 780.3 Pa. c) The life-time test under a pressure of 156 Pa at a frequency at 1.6 Hz. The current change curves were recorded after each 2,000 cycles and 200 cycles of data were presented in each recording. d) Magnified view of the part of the ΔI–t curve in c) after 10,000 loading–unloading cycles.
Figure 4. Wide adaption of this bio-inspired sensor to detect diverse types of mechanical forces. The dependence of current on time of the pressure sensor by a) finger touching, b) hand torsion and c) hand bending. The devices can responses to dynamic loading and unloading cycles. Inset images are digital photographs showing the test statements of the sensor. d) Plot of current response of the sensor as a function of time for the applied bending angle change in the range from ~2° to ~20°. The sensor has been regularly bent by a stretchable machine.
Figure 5. An “artificial mimosa leaves” with an integrated system of “touching-responding-feedback”. a) Schematic illustration of the generation of “artificial mimosa leaves”. A rigid polymeric bracing frame with three holes has been generated by the 3D-printer. Then, bio-inspired flexible pressure sensors have been cut into natural leaflet shapes and placed onto the bracing frame. These flexible pressure sensors were connected with two control parts: the electrical part served as the function of “touching-responding” while the mechanical system worked for the behavior of “responding-feedback”. The lengths of physically connecting line have been tailored to mimic different closing levels of the leaflets. b-d) Digital photographs of the performance of “artificial mimosa leaves”. Once a pressure-sensor leaflet has been touched, the external force could be transferred by the sensor into a changed electrical signal, and triggered the motor to rotate. Owing to the unidirectional rotating of the motor, the lines connected with the leaflet pairs would be tightened up, yielding the closing of “artificial mimosa leaflets” within one second.

A bio-inspired flexible pressure sensor has been generated with high sensitivity (50.17 kPa$^{-1}$), quick responding time (< 20 ms), and durable stability (negligible loading-unloading signal changes over 10,000 cycles). Notably, the key resource of surface microstructures upon sensor substrates came from the directly molding of natural mimosa leaves, indicating a simple, environment-friendly and easy scale-up fabrication process of these flexible pressure sensors.
**Keyword:** Bio-inspired, Flexible, Pressure sensor, Touch sensor,

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Supporting Information

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Figure S1. The dependence of surface resistance on the thickness of deposited gold layer. Thinner gold layers would yield larger resistance while thicker ones might waste the recourse. Thus, a 30 nm thick gold layer has been chosen in this study.
**Figure S2.** Schematic illustration of the setup of bio-inspired pressure sensors. Two layers of coated PDMS films with the microstructured surfaces have been placed face-to-face, yielding a bio-inspired pressure sensor. Conductive wires were anchored on the edge of both films as the source-drain electrodes.
Figure S3. Rough-to-rough type device construction can yield the highest impedance change. Plots of current response of diverse sensor as a function of time (pressure input frequency was 1.6 Hz and the applied pressures were 156 Pa). Rough-to-rough type device has generated more than 0.6 mA current increase while the flat-to-flat counterpart showed a tiny current change and rough-to-flat one exhibited smaller change.
Figure S4. Magnified images of pressure-response plots for the bio-inspired pressure sensor ranging from a) 0-70 Pa, b) 200-1,500 Pa and c) 2,000-10,000 Pa, respectively. The structured PDMS films exhibit a high pressure sensitivity of 50.17 kPa⁻¹ during 0 and 70 Pa, a relatively low pressure sensitivity of 1.38 kPa⁻¹ during 200 and 1500 Pa and a tiny pressure sensitivity of 0.04 kPa⁻¹ during 2,000 and 10,000 Pa.
Figure S5. The sensor exhibited a steady response to static pressure and the resistance under each pressure was constant. The detailed I-V curve for mechanical loads at various pressures.
Figure S6. Time-resolved measurements of the sensor response. Time-resolved plots of the output signal for an applied pressure frequency of a) 0.34 Hz, b) 4.17 Hz and c) 9.00 Hz. The applied pressures were 156 Pa. d) The dependence of sensor hysteresis time on the frequency. The hysteresis time decreased from ~1.1 s to nearly 0 s following the increase of pressure frequency due to the elastic deformation and viscoelastic effects of microstructured PDMS films during the loading-unloading process.
Figure S7. Instant response of the bio-inspired pressure sensor, which exhibits a response time of 20 ms. The applied pressures were 156 Pa and the frequency is 1.6 Hz.
Figure S8. The height of surface microstructures played an important role in deciding the sensor sensitivity. Digital photographs of a) *mimosa*, d) *cymbopogon citratus* and g) *populus lasiocarpa* leaves. b,e,h) and c,f,i) are top and side view SEM observations of a,d,g), respectively. Irregular microdomain patterns existed upon natural plant leaves. j) The comparison of current changes of sensors inspired by these three kinds of leaves. The applied pressures were 156 Pa and the frequency is 1.6 Hz. The height of microstructures upon *cymbopogon citratus* was too large to deposit continuous electricity active coating, leading to the block of electron transport. *Cymbopogon citratus* owned smaller microstructure height, however, produced smaller current value than that of *mimosa*. Briefly, carefully choosing the plant template type would yield great performance of bio-inspired pressure sensors.