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Stretchable Motion Memory Devices Based on Mechanical Hybrid Materials

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Abstract

Animals possess various functional systems such as sensory, nervous and motor systems, which show effective cooperation in order to realize complicated and intelligent behaviors. This inspires rational designs for the integration of individual electronic devices to exhibit a series of functions, such as sensing, memory and feedback. Inspired by the fact that humans can monitor and memorize various body motions, we developed a motion memory device to mimic this biological process. In this work, we introduced mechanical hybrid substrates, in which rigid memory devices and stretchable strain sensors are integrated into a single module, which enable them to work cooperatively in the wearable state. When attached to the joints of limbs, our motion memory device can detect the deformations caused by limb motions and simultaneously store the corresponding information in the memory device. This work would be valuable in materials design and electronics technology towards the realization of wearable and multifunctional electronic modules.

Bio-inspired electronic devices with excellent stretchability and adaptability have been designed to mimic the architectures and functions of highly evolved bio-systems. This renders

the devices promising not only for highly sensitive and efficient signal detection but also for the integration of functions such as intelligent processing and reflection.^[1-5] For example, smart sensors have been designed to detect signals from environment through the mimesis of human sensing, such as smelling, tasting, touching, visual and auditory sensing.^[6-10] Artificial neuron networks have also been studied to mimic the information processing capabilities of biological brains.^[11-13] There is no doubt that, individually, these functional units of organisms exhibit outstanding performance. However, what is more fascinating is that they are capable of working together to enable complex behaviors. For instance, we human beings are able to perform various motions by controlling our locomotor systems, as well as simultaneously monitor and memorize the corresponding information via our neural systems (Scheme 1a).^[14-16] Thus, the integration of multiple functions, i.e., the combination of individual electronic components such as power supplies, sensors, information storage devices and actuators, is an essential requirement for constructing functional electronic modules and future wearable computers, which would enable potential applications in health monitoring and biological actuation.^[4, 17-19]

One of major challenges for such integration is the rational combination of various device functionalities into a single wearable state. Currently, some basic elements for the construction of electronic circuits, such as electrodes and mechanosensors, have already been engineered for good tolerance towards mechanical deformations through rational structural designs.^[20-24] However, it remains difficult for traditional complementary metal–oxide–semiconductor (CMOS) devices, the properties of which are usually based on brittle materials and sophisticated structures, to be part of such integration due to low flexibility and stretchability.^[25-27] An example would be memory devices, which are essential electronic components in the construction of functional electronic modules. Memory devices exhibit information storage capability through electrically triggered-resistance switching,^[28-30] which usually relies on dynamic atom/ion migrations at the nanoscale that widely exist in various

functional dynamic materials.^[31-33] Due to the delicate sandwiched device configuration and the constituent brittle electronic materials, memory devices show poor ability to accommodate mechanical deformations.^[34] Although some attempts have been made to overcome this issue, the limited choices in elastic active materials and the complexity of the fabrication processes render the realization of stretchable memory devices extremely challenging.^[35-39] In addition, the incorporation of fragile memory devices in a stretchable module with other flexible electronic units would result in other complicate problems and issues.

In this work, a mechanical hybrid substrate was introduced to enable stretchable devices to work together with traditional rigid electronics (Scheme 1). The substrate was spatially separated into patterned domains with different mechanical properties, i.e., high and low moduli, which would exhibit different localized strain under physical forces.^[40-41] As a result, both stretchable and non-stretchable functional devices can be located on the mechanically compatible parts of the hybrid structure, and combined as a complete unit with integrated functions. This method shows potential as a general solution for the construction of wearable electronic modules.^[42-45] Next, inspired by the ability of animals to memorize various gestures by detecting the movements of their body parts and storing the relevant information in their brains (Scheme 1a), we conceived of an integrated device to achieve a human motion memory system. The detection and “memorization” of particular body movements was achieved through a wearable module comprising a rigid resistance switching device based on zeolitic imidazolate framework-8 (ZIF-8),^[46] in combination with a stretchable strain sensor (Scheme 1b). The strain sensor located on the stretchable part of the substrate can detect the deformations on the body or clothes surface. Subsequently, the obtained information on human motion can be directly stored in the resistance switching devices located on the rigid device islands, without the involvement of external equipment (Scheme 1b). In this way, the combination of the two individual devices has realized an integrated process for the transduction of mechanical input into electric signals and the

subsequent conversion to stored informations.^[47-48] This design shows an instructive demonstration in the smart integration of multifunctional devices, and would be a valuable unit in intelligent electronics and robotic technology.^[18, 35, 49]

The procedures for the fabrication of the mechanical hybrid substrate and the ZIF-8 based memory device are shown in Figure S1 (Supporting Information). In the hybrid film, rigid SU-8 flakes with high modulus (4 GPa) are embedded in a soft PDMS substrate to help maintain the surface structure during the stretching process and serve as device islands, while the PDMS substrate with low modulus (1 MPa) works as elastic interconnection between the device islands and enables stretchability (Scheme 1c). The finite element modeling (FEM) analysis of the hybrid substrate revealed that the maximum principal strain on the surface of device islands is less than 0.1% when the substrate was subjected to 50% uniaxial strain, which indicated that mechanical forces would not damage the devices on top of the SU-8 flakes (Scheme 1, Figure S2a,b, Supporting Information). This was further proven by a control experiment which showed that ZIF-8 based memory devices on the device islands did not sustain mechanical damage, whereas devices fabricated on the soft part of the hybrid substrate were destroyed (Figure S3, Supporting Information). In addition, this strategy can accommodate different shape designs of SU-8 flakes in order to extend the adaptability of the substrate for different purposes (Figure S2, Supporting Information).

To prepare the ZIF-8 based stretchable memory device, gold film with microcrack morphology was thermally evaporated onto the mechanically hybrid substrate as the bottom electrode, which can maintain good conductivity under stretching.^[20, 50-51] Next, patterned ZIF-8 films containing uniform ZIF-8 nanocrystals (Figure S4, Supporting Information) were fabricated on the device islands in an array structure with patterned Ag electrodes on top (Figure 1a, Figure S2c,d, Supporting Information). The I-V curves of the Ag/ZIF-8/Au device showed typical resistance switching characteristics (Figure 1b) when a voltage bias was applied to the top Ag electrode while the Au electrode was grounded. At first, the device

exhibited the high resistance state (HRS). Upon continuously increasing the voltage, the device current gradually increased from 5.09×10^{-14} A to 1.04×10^{-10} A. As the voltage reached the setting threshold of 2.20 V, the current showed a sudden increase to the compliance limit (1×10^{-3} A), which implied that the memory device switched from HRS to the low resistance state (LRS). After the resistance switching, the device could maintain LRS during the subsequent sweep from zero to positive voltage, which indicated its non-volatile nature. Finally, the device could be made to switch back to HRS by a negative voltage sweep approaching the reset voltage of -0.85 V. Device-to-device distribution and endurance tests showed that the LRS and HRS were well-separated and the resistance OFF/ON ratio was as large as 10^7 , which demonstrated the reliability of the memory device in the data storage operation (Figure S5, Supporting Information) and was consistent with our previous work.^[46] In addition, multi-level storage could be realized through the application of different compliance currents (Figure S5e, Supporting Information). Based on Ag/ZIF-8/Au device structure, the resistance switching behavior relied on the electron hopping effect between the Ag nanoparticles formed in the insulating ZIF-8 layer, which occurred due to electro-migration of the electrode material under an electric field, and would be fractured during a reverse voltage sweep.^[46, 52-53]

Moreover, the mechanical hybrid substrate allowed the Ag/ZIF-8/Au device to perform stable information storage even under bending or stretching (Figure 1). The memory devices could be bent to a radius of approximately 1.1 mm or stretched to 50% strain with no obvious effect on the resistance switching performance (Figure 1c,d, and Figure S6a,b, Supporting Information). The HRS resistance remained unchanged under different bending or stretching states, whereas the LRS resistance of the memory device showed slight increase with stretching, which could be attributed to the decrease in conductivity of the microcrack-based gold film due to the strain (Figure 1d). Moreover, the retention tests showed that the HRS and LRS remained stable for more than thousands of seconds at a continuous read

voltage of 50 mV under the bending or stretching states (Figure 1e, and Figure S6c, Supporting Information).

Besides the exhibiting memory performance in static states, stretchable memory devices are also required to show stable information storage, including writing, reading, and erasing processes in dynamic states, under which mechanical forces would be continuously applied on the devices. Continuous reading tests showed the HRS and LRS of the device remained stable even after 1000 cycles of 50% strain stretching (Figure 2a,b, Supporting video SI), which indicated that the information stored in stretchable memory device can be properly maintained under consecutive stretching and releasing cycles. It should be noted that the resistance of LRS would vary with the applied strain on the substrate, which was consistent with the result under static states (Figure 2a). This phenomenon could have resulted from the change in the resistance of the microcrack-based gold film on the PDMS interconnection at different strains. This renders the microcrack-based gold film capable of functioning as a strain sensor, which could be used to construct the motion memory system.

In addition to reading, the writing and erasing behaviors of the stretchable memory devices also showed good stability during stretching. No significant disturbance on the writing and erasing process was observed at various stretching speeds (Figure 2c). These results demonstrated that the memory device on the mechanical hybrid substrate was able to function normally under dynamic states, rendering it suitable for application in wearable electronics. This was further demonstrated by attaching a stretchable memory device directly onto a knuckle; stable memory performances were observed during the finger bending process (Figure 2d, Supporting video SII).

In the locomotor system of the human body, the stretching and contraction of muscles play an important role for human to realize different motions, which are attributed to the complex cooperation of muscular and skeletal systems.^[54-55] We demonstrated that information on human motions could be stored in a wearable memory device integrated with

stretchable strain sensors, which would detect stretching on the body or clothes surface caused by the elongation of muscles. The strain sensor used was the aforementioned microcrack-based gold film, which also functioned as the bottom electrode in the stretchable memory devices. The stretchability of the gold film was mainly based on the composite structure of cracks and transverse wrinkles (Figure 3a), which could effectively release strain energy.^[56-57] Moreover, the gold film strip exhibited good performance when employed as a stretchable strain sensor; the resistance was able to increase and decrease according to the stretching and retraction, respectively (Figure 3b, and Figure S7a, Supporting Information). The resistance of the gold film could also be tuned by simply adjusting the width of the strip, in order to meet specific requirements for practical applications (Figure S7b, Supporting Information).

The construction of the integrated module was based on a shunt circuit consisting of a gold film strain sensor and a stretchable memory device with a resistor connected in series (Figure S8a, Supporting Information). The resistance of the resistor was adjusted to match the sensing range of the strain sensor in order to keep the divided voltage on the memory device (V_{memory}) lower than V_{set} . This meant that the memory device would show HRS prior to the stretching of the strain sensor. Upon the application of stress, the elongation of the strain sensor would result in the increase in resistance, which would increase the voltage on the shunt circuit until it reached V_{set} , thus setting the memory device to the “on” state (a typical test was shown in Figure S8b, Supporting Information). Due to the fast operation speed of the ZIF-8 memory device ($\sim 70 \mu\text{s}$) and the microcrack gold strain sensor ($\sim 31 \text{ ms}$), the stretching rate would have a dominate influence on the response time of the whole integrated system (Figure S9, Supporting Information). In addition, a cycling test was conducted by a cyclic stretching the strain sensor followed by the electrical reset of the ZIF-8 memory device (Figure S10, Supporting Information). This design demonstrated that the writing process of memory could be triggered by a strain sensor and showed the possibility of applications in monitoring and storing of information on stretching processes.

Furthermore, through simple modification of the bottom electrode, we can integrate a strain sensor and a memory device in a single circuit on a mechanical hybrid substrate (Figure 4a). To achieve a visual demonstration of the LRS and HRS, we connected a LED lamp in series with memory device in the shunt circuit. As proof of concept, we directly attached the chip onto the upper arm near the elbow to monitor and store the information on the elbow flexion, by detecting the stretching of the skin caused by the elongation of the triceps brachii muscle (Figure 4b). In the elbow extension state, the stretchable memory device showed HRS. In response to the flexing of an elbow, the elongation of the strain sensor would switch the memory device to the “on” state, causing the LED lamp to light up and indicate that information on this motion had been stored in the memory device. Despite the subsequent extension of the elbow, the LED lamp remained lighted, indicating that the memory device remained under LRS (Supporting video SIII). The memory device can be then switched back to HRS by an electrical reset process. The information of complex motions can also be stored through the cooperation of multiple motion memory systems located on different joints. By setting up a one-to-one match between each motion memory device and muscular relaxation (or contraction), we would be able to collect information on all of the performed motions by reading the resistance level in each motion memory device. For example, by monitoring deformations on the human body or clothes caused by the stretching of the teres major muscle and the triceps brachii muscle, we can store information of the composite motions of shoulder abduction and elbow flexion (Figure 4c).

In summary, we fabricated stretchable memory devices located on the mechanical hybrid substrate, which exhibited information storage property even under dynamic processes. Furthermore, through circuit design and device engineering, we achieved the human motion memory system by integrating strain sensors with the stretchable memory devices, which showed potential applications for wearable sensory memory in health monitoring and medical treatment. This work would be instructive and valuable in material design and electronic

technology to realize the wearable electronic modules with integrated functions, which would be essential for constructing smart devices and future intelligent systems.

Experimental

Fabrication of mechanically hybrid substrate: A thin layer (~20 μm) of PDMS (10:1) was first fabricated on fluoride-treated silicon wafer by spin-coating. SU-8 flakes of various shapes were then fabricated on top of the PDMS film via photolithography with designed shadow masks. Followed by the spin-coating and curing of another PDMS film (~100 μm) on top, this mechanical hybrid film was peeled off from the silicon wafer and served as the stretchable and flexible substrate for the fabrication of memory devices. It should be noted that, to enhance the connection between SU-8 and PDMS, strong plasma treatment (100 W, 3 mins) was applied to the samples between each steps.

Fabrication of ZIF-8 based memory device on hybrid substrate: The chemicals, $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and 2-methylimidazole, were used as received from commercial suppliers without further purification. Upon the thermal evaporation of microcrack Au films (30 nm) with Cr film (3 nm) as the adhesion layer on top of the hybrid substrates, patterned ZIF-8 thin films were grown on the substrates with a PDMS mask according to the pattern of device islands through immersion in a fresh mixture of 5 ml $\text{Zn}(\text{NO}_3)_2$ solution (25 mM) and 5 ml 2-methylimidazole solution (50 mM) for 40 minutes at room temperature. The substrates were then washed with methanol and dried under nitrogen flow. Lastly, patterned Ag electrodes (150 nm) were thermally evaporated on the ZIF-8 films as top electrodes with a shadow mask.

Material characterization: The powder X-Ray diffraction (XRD) patterns were obtained through a Bruker 6000 X-ray diffractometer using a $\text{Cu}/\text{K}\alpha$ source ($\lambda=1.5406 \text{ \AA}$). AFM images were taken with a Veeco Multimode SPM system with a Nano-Scope V control

station under the tapping mode. FT-IR spectra were obtained with a PerkinElmer Frontier FT-IR spectrometer.

Device characterization: The memory performances were tested by a closed cycle cryogenic wafer probe station (Lakeshore CRX-4K, Westerville, OH, USA). The I-V characteristics and response time of the memory devices and strain sensors were obtained by a semiconductor parameter analyzer (Keithley 4200-SCS, Cleveland, OH, USA) and an oscilloscope (Tektronix DPO5054B). In the wearable state, the circuit of integrated devices, including motion memory modules, commercial LED lamps and button cells, were assembled through patterned gold electrodes on the elastic chip, thin copper wires, silver paste, and liquid metal (Gallium–indium eutectic, Aldrich) (Figure 4a). The measurements were typically conducted at atmospheric pressure and room temperature.

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Scheme 1. Motion memory devices based on the mechanical hybrid substrate. a) Humans can perform, sense and memorize various motions with cooperation of different organs. b) The design of integrated device modules comprising a stretchable memory device and a strain sensor, which combines strain detection and information storage properties. c) Schematic figure of the devices on the mechanical hybrid substrate containing rigid and soft domains, with a FEM analysis of a single island under stretching.

Figure 1. Stretchable memory arrays and the corresponding data storage performances under static conditions. a) Stretchable memory arrays fabricated on a hybrid substrate with patterned device islands. b) I-V curves of the Ag/ZIF-8/Au device in voltage sweeping mode at room temperature (inset: structure of the device). The compliance current was set to 1×10^{-3} A. c) Resistance states of stretchable memory devices at different bending states. The bending test machine used is shown in the inset figure. d) Resistance states of stretchable memory devices versus strain. e) Retention characteristics of HRS and LRS under a continuous 50 mV readout voltage at different stretching states.

Figure 2. Memory behaviors of the stretchable memory devices at dynamic conditions. a) Reading process of a stretchable memory device in a stretching and releasing cycle. b) Retention characteristics of two resistance states under a continuous 50 mV readout voltage for 1000 cycles of stretch and release. c) Writing and erasing processes of stretchable memory devices at different stretching rates. d) Reading process of a stretchable memory device on a knuckle during finger bending.

Figure 3. Morphology and performance of stretchable strain sensor based on microcrack gold films. a) SEM image of gold surface under 50% strain. b) Strain sensing by a gold film strip (width = 650 μm) for 1000 cycles of stretch and release.

Figure 4. Motion memory system with detection and storage functions for monitoring human motion. a) Integration of a strain sensor and a stretchable memory device on a single chip by modification of the bottom Au electrode. b) A motion memory device detecting and storing information on elbow flexion. c) Multiple motion memory systems monitoring the composite motions of shoulder abduction and elbow flexion.

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Motion memory devices based on mechanical hybrid substrate are designed with integration of memory devices and strain sensors, in order to store information on human motion by monitoring the deformations of the body surface caused by the elongation of muscles.

Keyword: motion memory device, strain sensor, mechanically hybrid substrate, wearable electronics, human motion

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Stretchable motion memory devices based on mechanical hybrid materials

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