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An Artificial Somatic Reflex Arc

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Abstract

The emulation of human sensation, perception and action processes has become a major challenge for bio-inspired intelligent robotics, interactive human-machine interfacing and advanced prosthetics. Reflex actions, enabled through reflex arcs, are important for human and higher animals to respond to stimuli from environment without the brain processing and survive the risks of nature. An artificial reflex arc system that emulates the functions of the reflex arc simplifies the complex circuit design needed for “central-control-only” processes and becomes a basic electronic component in an intelligent soft robotics system. Herein, we report an artificial somatic reflex arc that enables the actuation of electrochemical actuators in response to the stimulation of tactile pressures. Only if the detected pressure by the pressure sensor is above the stimulus threshold, the metal-organic framework-based threshold controlling unit (TCU) can be activated and triggers the electrochemical actuators to complete the motion. Such responding mechanism mimics the all-or-none law in the human nervous system. As a proof of concept, we successfully

integrated the artificial somatic reflex arc into a robot to mimic the infant grasp reflex. This work provides a unique and simplifying strategy for developing intelligent soft robotics, next generation human-machine interfaces and neuroprosthetics.

Organisms possess complex activation tools or behavior cascades that enable the capability for problems solving, which are crucial for the species to survive.^[1,2] Scientists have always been longing for the designing and engineering of robotics and devices that can mimic the behaviors of organisms.^[3-7] Recently, the advancement of technology in soft/flexible electronics has opened up new opportunities for the fabrication of devices that emulate the structures and functions of biological systems.^[3,8-10] Flexible sensors with various sensing mechanisms have emerged to emulate the basic human senses.^[11-22] Both flexible memristors and transistor synaptic devices have been designed to mimic the brain-like fast and power-efficient information processing.^[23-28] Besides, soft actuators based on different actuation mechanisms have been developed for artificial muscles and soft robotics.^[29-36] Multiple flexible devices have been integrated to realize complex functions for emulation of human sensory system, bio-inspired robotics, prosthetics and healthcare.^[3,37-44] For example, tactile perception device was developed to mimic the functions of human afferent nerve by the integration of pressure sensors, organic ring oscillators, and synaptic transistors.^[3] Furthermore, organic optoelectronic sensorimotor synapse device based on organic nanowire synaptic transistor was reported, which realized the light-responsive actuation of artificial muscle actuators emulating the contraction pattern of biological muscles.^[44] However, the increasing complexity results in dramatic computation and transmission burden for integrated systems relying on central-only data processing.^[45,46] In contrast, nature addresses this issue by keeping the necessary data processing local and thereby of minimal complexity.^[2,47] The natural perception-action loops of organisms are in many cases independent of any outer control by the brain.^[47-50] For human and higher animals, the employment of reflex arcs allows the implement of reflexes without central processing by the brain. Reflexes not only enable the rapid reacting in response to a stimulus to outpace harmful elements, but also “liberate” the higher brain from countless low decision

depth operations, such as raising single hairs or adjusting breath and heartbeat to the given needs.^[48,51] Therefore, emulating the responsive behaviors and processes of biological reflex arcs, an autonomous control system that collects physical signals from environment and generates feedback motions in response simplifies the complex circuit design, minimizes requests for central control processing unit and becomes an essential “intelligent” component for bio-inspired robotics.^[3,44] However, how to realize such artificial system by using simple soft electronic components is a major challenge.

In human, the reflex arc enables the cooperation of the muscles and neurons to complete the reflex actions (**Figure 1a**, left).^[48,52] A somatic reflex arc initiates with the sensory receptor, for example, the mechanoreceptor in human skin, which detects the mechanical pressure. Once the stimulus is detected, an action potential passes through the neuron and reaches the integration center, which is the grey matter in the spinal cord. In response to the signal from the integration center, an unconscious action is realized by the contraction of the effector muscle. Once the reflex is triggered, the actions would be completed even the stimulus is removed. Importantly, to trigger the reflex action, an all-or-none law is applied (**Figure 1b**, left).^[53] If the stimulus level reaches the threshold, the action potential can be elicited, and the reflex actions can be triggered. For an individual nerve or muscle fibre, the strength of the response is independent of the strength of the stimulus. Otherwise, there is no response at all. For example, a strike applied on the patellar tendon must be strong enough to trigger the knee jerk reflex.^[51] Although conventional rigid robotic devices and systems based on solid-state electronics have been implemented to mimic biological reflexes, these systems are based on classical control theory, which requires complex hierarchical systems for sensory input, signal processing, motion planning and execution.^[54-57] In addition, the mechanical mismatching compliance of biological materials strictly limits the application of these systems for future health monitoring, soft robotics and

wearable devices (Table S1, Supporting Information).^[58,59] Therefore, mimicking the human somatic reflex arc, an artificial device with response motions responding to tactile stimulations in an all-or-none, threshold dependent pattern simplifies autonomous control loops and becomes a fundamental functional module for bio-inspired robotics.^[60]

In this work, we report an artificial somatic reflex system that enables the actuation of electrochemical actuators in response to the stimulation of tactile pressures, which mimics the function of the somatic reflex arc in humans and higher animals. The system consists of three main components: a pressure sensor, a threshold controlling unit (TCU) and an electrochemical actuator (Figure 1a, right). A resistive pressure sensor based on microstructured poly(dimethylsiloxane) (PDMS) film coated with carbon nanotubes (CNTs) was constructed to mimic the tactile sensing of the skin. We fabricated a metal-organic framework (MOF)-based TCU to emulate the function of the integration center. The electrical parameters of the MOF-based TCU device can be adjusted for better compatibility with other devices. Finally, a multi-walled carbon nanotube (MWCNT)-based electrochemical actuator with a capacitive double layer configuration was introduced herein to imitate the muscle. Importantly, our system presents a responding mechanism similar to the all-or-none law in biological nervous system owing to the employment of the MOF-based TCU device in the system (Figure 1b, right).

The resistive pressure sensor was constructed using CNT/PDMS film with pyramidal microstructure arrays as the sensing layer (Figure S1, Supporting Information). The microstructured PDMS film was fabricated by curing PDMS precursor on the silicon master. The silicon master with uniform recessed pyramid microstructures was prepared using photolithography and silicon etching methods as previously described.^[11,61] Patterned stretchable Au/PDMS film was prepared by thermal evaporation with a shadow mask.^[37,62,63] By placing the CNT/PDMS film face-to-face on top of patterned stretchable Au/PDMS film

(Figure S2, Supporting Information), a typical pressure sensor device was obtained. The performance of the pressure sensor is displayed in **Figure 2a**. In this work, we aim to match the detection range of the pressure sensor with the range of a gentle fingertip tapping, which ranges from a few hundred Pa to a few thousand Pa (Figure S3, Supporting Information). Therefore, the sensor is designed to exhibit a broad sensing range (Figure 2a, i). Correspondingly, the sensor must show a wide range of resistance change responding to the applied pressures. That is the reason why CNT was employed as the conductive layer. In contrast, the pressure sensor fabricated using silver nanowires (AgNWs) shows comparative limited detection range due to the supreme conductivity of the AgNWs (Figure S4, Supporting Information). Due to the low concentration of CNTs we used, the sensor is non-conductive when no pressure is applied. A series of pressures were applied to the device and the current level remained the same for identical pressures indicating a good reliability of the sensor in a broad range of pressure (Figure 2a, ii). The real-time voltage change of the pressure sensor responding to loading/unloading pressure was measured, indicating little hysteresis of the sensor for pressure loading (< 4 ms) and unloading (< 7 ms) (Figure 2a, iii).

Secondly, we fabricated MOF-based resistive switching device as the TCU, which is the core part of our artificial reflex arc. Such devices are known to be able to switch between high resistance state (HRS) and low resistance state (LRS) upon applying a voltage bias.^[27,37] Such electrical characteristic is utilized to closely mimic the all-or-none response, which requires bistability that can clearly distinguish between non-stimulated state and activated state.^[64] By the further integration of the TCU and the pressure sensor, we realized the pressure threshold-dependent current regulation, which can be used to subsequently control the actuators. However, in order to integrate with other electronic devices and adapt to different circuit designs, the electrical parameters of the device needs to be adjustable to provide better compatibility. Here, the controllable set voltage was implemented for zeolitic

imidazolate framework-8 (ZIF-8)-based device via simply varying the thickness of the ZIF-8 layer. The ZIF-8-based flexible resistive switching device is prepared by the deposition of ZIF-8 film on top of the stretchable Au/PDMS film as previously reported.^[27,37,65] Silver was employed as the top electrode. The resistive switching mechanism is attributed to the formation and rupture of the nanoscale silver conductive filament in the ZIF-8 layer upon applying an electric field.^[27] For a 1-layer ZIF-8 device, the set voltage averagely locates at 1.8 V (Figure S5b, Supporting Information), a characteristic which can be adjusted by further increasing the number of deposition cycles. After the deposition of two layers of ZIF-8, the set voltage is shown to be shifted to 3.5 V and the device is stable with a current compliance of 10 mA (Figure 2b, i). We also tried to deposit a third layer of ZIF-8, but then the obtained device became very insulating, and the device could not be switched to LRS even the applied voltage reached 10 V (Figure S5b, Supporting Information). The change of the electrical property of devices with different numbers of deposition cycles is due to the increasing thickness of the ZIF-8 film (Figure S5a, Supporting Information).^[65,66] We chose the 2-layer ZIF-8 device as our typical TCU device. The device-to-device distribution, cycling test and retention test demonstrated that the HRS and LRS are well-separated and that the TCU device shows excellent reliability (Figure 2b, ii and iii, Figure S6, Supporting Information). Furthermore, the resistive switching characteristic of the ZIF-8-based TCU device was able to perform well at bending states, indicating a good flexibility of the device (Figure S7, Supporting Information).

Thirdly, to complete the artificial reflex arc, we employed MWCNT-based electrochemical actuators with a capacitive configuration (Figure S8, S9 and S10, Supporting Information). The electrochemical actuator, which belongs to the category of ionic polymer-metal composite (IPMC) actuator, is a promising candidate for artificial muscles due to their large deformation and operation stability in air.^[32,67] Besides, one important reason we chose

such type of actuator is that they can be operated at a voltage level as low as 2-3 V, which is easy to integrate with other devices. The as-prepared actuator strips show large bending deformation and can bend to both directions by changing the polarity of the applied voltage bias (Figure 2c, i and Figure S11, Supporting Information). Due to the capacitive double layer configuration, the actuator presents electrochemical characteristics of a double layer supercapacitor. When a constant voltage bias is applied across the two sides of the actuator strip, an exponential decay of the current is expected, which resembles the curve of charging a supercapacitor (Figure S12, Supporting Information). At the same time, the strip bends to the direction of the anode. The actuator device then was subjected to a periodical voltage switching between 3 V and -3 V. The instant current change demonstrated the reversible charging and discharging of the device (Figure 2c, ii). The maximum displacement value for 10 cycles suggested a good reversibility and reliability (Figure 2c, iii).

Next, in order to mimic the all-or-none response, the pressure sensor and the TCU were first integrated to establish the pressure-responsive resistive switching, which was further used for current regulation and the control of the actuators. To maintain the flexibility, the system was constructed on PDMS elastic film, on top of which, there is the stretchable gold layer serving as the conductive layer.^[62,63] The TCU device was prepared by the deposition of 2 layers of ZIF-8 film on top of the desired area of the circuit (**Figure 3a**, left). The microstructured CNT/PDMS film was attached to the designated area to complete the pressure sensor. Such configuration realizes the series connection of the two devices and thus the pressure-dependent programming of the TCU resistance state. As mentioned before, the biological sensory system responding to a certain stimulus follows the all-or-none law. Only if the level of the stimulus, e.g. a pressure, reaches the threshold, a response action can be elicited (Figure 3a, right). Now we show that our pressure sensor and TCU integrated system possesses a similar responding pattern. A voltage bias of 4 V was applied to the

system, which was placed on a curved surface (Figure S13, Supporting Information). A series of pressures were applied onto the pressure sensor. We found that the level of the current stayed very low ($<10^{-9}$ A) until the loading pressure reached 1 kPa. The abrupt increasing in the current suggested that the TCU had switched to LRS (Figure 3b). When the applied pressure is below 1 kPa, the resistance of the pressure sensor ranges from $10^6 \Omega$ to $10^8 \Omega$, which strictly limits the current flow in the system to be lower than 10^{-6} A. Such low level of current is not adequate to switch the 2-layer ZIF-8 device from HRS to LRS.^[68-70] To obtain more evidence, we further monitor the resistance state of TCU after different pressures were applied by reading the resistance of the TCU separately (Figure 3c). We found that the TCU stayed at HRS ($\sim 10^{10} \Omega$) before 1 kPa was applied. When the applied pressure was above 1 kPa, the TCU switched to LRS with a typical resistance lower than $10^3 \Omega$. The results demonstrate that 1 kPa is the threshold pressure for the TCU to switch to LRS. This threshold value can be used to distinguish between a very gentle finger tapping (~ 500 Pa) and a firm finger tapping (~ 2000 Pa). We further demonstrated that, the threshold pressure is tunable over a broad range by simply changing the concentration of the CNTs used to fabricate the pressure sensor (Figure 3d). Importantly, the response time of the integrated system is estimated to be *ca.* 0.8 ms, which is compatible with the response time in biological systems (Figure S14, Supporting Information).^[71]

Lastly, to complete our artificial reflex arc, the integrated pressure sensor and TCU were connect to the electrochemical actuator to accomplish the tactile pressure-responsive actuation (Figure 3e). The electrochemical actuator has a configuration of a supercapacitor, which generates mechanical strain due to the ions' intercalation and deintercalation in the electrodes. A photograph of the integrated circuit on polyethylene terephthalate (PET) film is displayed in Figure 3f to show the flexibility. The designed circuit consisting of a controlling circuit (C_{Ctrl}) and an actuation circuit (C_{Act}) is illustrated (Figure S15, Supporting

Information). Before sensing any pressure, the current flow in the controlling circuit is very low ($<10^{-9}$ A) due to the HRS of the TCU. Correspondingly, there is no current flow in the actuation circuit (Figure S15, Supporting Information). Once the pressure sensor detects a pressure beyond the threshold, the TCU is activated and switched to LRS. Subsequently, the current in the C_{Ctrl} is adequate to activate the actuation circuit. We monitor the real-time current change upon applying a swift finger tapping with a pressure above the threshold (1 kPa). Responding to the tapping, an abrupt current increasing was observed in both C_{Ctrl} and C_{Act} (Figure 3g). In C_{Ctrl} , the current remained steady. At the same time, an exponential current decay was observed in C_{Act} , which indicates that the charging of the actuator device was taking place. These results demonstrate that the circuit design is successful and the system can realize the actuation of the electrochemical actuators in response to a tactile pressure above a threshold.

As a proof-of-concept, we constructed a robot whose fingers can respond to the touch on the chest to mimic the grasp reflex, which is a well-known primitive reflex for human babies below 6 months old (**Figure 4a**).^[72] The 3D-printed robot was equipped with our artificial somatic reflex arc (Figure 4b). The pressure sensor and the TCU integrated circuit was fabricated using the same approach but with a different layout on PDMS (Figure 4c). The original left arm was rebuilt into the actuation system (Figure 4d and Figure S16, Supporting Information). Three actuator strips were employed as the fingers, which can grip objects when bent (Figure 4d). The actions of making a grasp posture can be accomplished by applying a stimulus (finger tapping) on the sensor. When a gentle tapping was applied, the fingers did not give any response due to the pressure sensed was below the threshold (Figure 4e, g and Movie SI, Supporting Information). In contrast, a stronger firm tapping with a pressure value above the threshold triggers the actions of the actuators (Figure 4f, h and Movie SII, Supporting Information). The fingers of the robot started to move

immediately and completed the action of making the grasp posture. Similar to a biological reflex, once the threshold-dependent artificial reflex is triggered, the actuators complete the actions without the necessity of the stimulus being present throughout the response period. A multiple tapping test further demonstrated the threshold-dependent response (Figure S17, Supporting Information). The release of the posture can be achieved simply by reversing the voltage polarity of the actuation circuit (Figure S18, Supporting Information).

In summary, here, we designed and constructed an artificial somatic reflex arc system, which mimics the function of the biological reflex arc. The pressure sensor detects the physical pressure signal and transduces it into electrical signal and passes it to the threshold controlling unit (TCU), which subsequently activates the electrochemical actuators to accomplish the motions. To mimic the all-or-none response in biological nervous system, we employed a ZIF-8-based TCU device. As a result, only a pressure above the threshold can activate the TCU and triggers the response of the actuators. The electrical parameters of the TCU can be easily adjusted by controlling the thickness of the ZIF-8 film. This is the very first time that a complete artificial reflex arc is constructed by multiple flexible electronic components. This work provides new insights for a nanomaterials-based simplification of autonomous control and is promising for the applications in intelligent soft robotic systems, interactive wearable electronics and neuroprosthetics.

Experimental Section

The detail experimental process is available in the Supporting Information.

Supporting Information

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

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Figure 1. Illustration of the concept of the artificial somatic reflex arc. a) Schematic comparison of the somatic reflex arc in human and our artificial somatic reflex arc. The somatic reflex arc consists of three main parts: sensory receptor (skin), integrating center (spinal cord) and effector (muscles). Correspondingly, our artificial electronic somatic reflex arc is composed of three flexible devices as the counterparts: a pressure sensor, a threshold controlling unit (TCU) and an electrochemical actuator. b) Illustration of the artificial reflex arc mimicking the all-or-none response in biological system. Knee jerk reflex is illustrated as an example. The level of stimulus must reach the threshold to trigger an action potential and thus the completion of the reflex. Similarly, in our artificial reflex arc system, the action of the actuators can be triggered only if the pressure sensed by the pressure sensor is above the threshold.

Figure 2. Overview of the characteristics of the pressure sensor, the threshold controlling unit (TCU) and the electrochemical actuator. a) Electrical characterization of the microstructured CNT/PDMS-based pressure sensor. i) The resistance response of the pressure sensor to the applied pressures. Inset shows the typical I - V curves of the sensor at various pressures. ii) Instant current change in response to different applied pressures. iii) Voltage change in accordance to loading/unloading a constant pressure (10 kPa) for multiple times. The inset displays an enlarged view of the voltage change for an individual loading/unloading process showing the response of the sensor. The device was in series with a constant resistor (500 k Ω) and a constant voltage of 1 V was applied. Pressures were applied by a motorized vertical test stand. b) Electrical characterization of the TCU device. i) Typical I - V curve of the TCU with a 2-layer ZIF-8 structure. The inset displays the configuration of the device. ii) Device-to-device distribution of the HRS and LRS of the TCU device. iii) Retention performance of both HRS and LRS at a constant readout voltage of 50 mV. c) Electrical

characterization of the electrochemical actuator. i) Digital photographs showing the configuration of testing a typical MWCNT-based capacitive actuator (25×5 mm) and the bending motion of the actuator strip at a constant voltage of ± 3.0 V. ii) Input voltage signal and current response at an applied voltage of ± 3.0 V with a frequency of 0.1 Hz. iii) The maximum displacement of the actuator strip at an applied voltage of ± 3.0 V with a frequency of 0.1 Hz for 10 cycles.

Figure 3. The design and characteristic of the artificial somatic reflex arc system. a) Schematic illustration of the pressure sensor and the TCU integrated on a flexible PDMS substrate and the all-or-none law in a biological sensory system. b) The instant current response of the integrated pressure sensor and the TCU to different pressures applied. A voltage bias of 4 V was applied to the integrated system, which was placed on a curved surface. c) The resistance change of the TCU as a function of the pressure applied with a reading voltage of 50 mV. The threshold level of pressure to shift the TCU from HRS to LRS is *ca.* 1 kPa. d) The evolution of the threshold pressure as a function of the CNT concentration used for the pressure sensor. e) Illustration of the complete artificial somatic reflex arc consisting of a pressure sensor, a TCU and an electrochemical actuator. The

MWCNT-based electrochemical actuator has a configuration of a supercapacitor and the electromechanical strain is generated due to the ions transferring under the voltage applied.

f) Digital photograph of the integrated pressure sensor and TCU attached on to a PET surface.

g) The instant current change (top row) of the controlling circuit (I_{ctrl}) and the actuation circuit (I_{Act}) in response to a pressure above the threshold (1 kPa) applied by a finger tapping (bottom row). The inset displays an enlarged view of the instant pressure change.

Figure 4. Demonstration of a 3D-printed robot equipped with the artificial somatic reflex arc to mimic the grasp reflex. a) Photograph of a baby presenting the grasp reflex. b) Photograph of the 3D-printed robot with the artificial somatic reflex arc. c) Enlarged view of the integrated pressure sensor and the TCU. d) Enlarged view of the actuation system (artificial fingers for the robot). e) and f) The photo images showing the different responses of the robot hand to a gentle finger tapping and a firm tapping. For a gentle tapping, the artificial fingers did not accomplish the grasping action due to the low level of pressure applied. In contrast, the artificial fingers bent towards the middle and formed a grasping posture in response to a firm tapping with a pressure level above the threshold. g) and h) The instant current change of the actuators responding to the pressure applied on the pressure sensor. The insets display the enlarged view of the pressures applied.

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An artificial somatic reflex arc is presented, which enables the actuation of electrochemical actuators in response to tactile stimulations. The pressure sensor perceives the pressure and only if the sensed pressure is above the threshold, the threshold controlling unit can be activated and triggers the actuators to complete the reflex actions, which mimics the all-or-none response.

Keywords: artificial reflex arc, pressure sensors, resistive switching devices, electrochemical actuators, all-or-none response

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An Artificial Somatic Reflex Arc

Supporting Information

An Artificial Somatic Reflex Arc

*Ke He, Yaqing Liu, Ming Wang, Geng Chen, Ying Jiang, Jiancan Yu, Changjin Wan, Dianpeng Qi, Meng Xiao, Wan Ru Leow, Hui Yang, Markus Antonietti and Xiaodong Chen**

Experimental section

Materials. Zinc nitrate hexahydrate (98%), 2-methylimidazole (99%), 1-ethyl-3-methylimidazolium tetrafluoroborate (EMIMBF₄, ≥99.0%), poly(vinylidene fluoride-co-hexafluoropropylene) (Pvdf (HFP), M_w 400,000), N,N-dimethylacetamide (DMAC, ≥99.9%), 4-methyl-2-pentanone (≥99.5%), propylene carbonate (99.7%) and silver nanowires (20-50 μm) were purchased from Sigma-Aldrich. Single-walled carbon nanotubes (SWCNT, Bu-202) were purchased from BuckyUSA. Carboxylated multi-walled carbon nanotubes (MWCNT, TNMC2) were purchased from Time Nano. Monomer and curing agent for polydimethylsiloxane (PDMS, Sylgard 184) were purchased from Dow Corning and used as received.

Preparation of patterned stretchable Au/PDMS film.

The stretchable Au/PDMS film was prepared as previously described.^[1] The PDMS precursor with a cross linker to monomer weight ratio of 1:10 was stirred vigorously and centrifuged at 8000 rpm for 20 min to remove the bubbles. The mixture was then spin-coated onto silicon wafers at 800 rpm for 30 s and allowed to cure at a temperature of 60 °C for 4 h. The completely cured PDMS film was then transferred to the thermal evaporation chamber with a shadow mask fixed on top. After the vacuum reached 1.5×10⁻⁶ Torr, a layer of chromium with a thickness of 2 nm was deposited on to the film, followed by a layer of gold with a thickness of 40 nm.

Preparation of pressure sensor.

Silicon master with the pyramidal microstructure was prepared using typical photolithography and wet etching process.^[2,3] The PDMS precursor (10:1, w/w) was cast on the microstructured silicon master by spin-coating (500 rpm, 30 s) and allowed to be cured in an oven at 60°C for 4 h. The obtained PDMS film was first treated by oxygen plasma (80% of power, 1 min). Then an aqueous suspension of SWCNT (2×10^{-3} mg/mL, 20 μ L) was cast on the PDMS film. The CNT-coated PDMS film was dried at room temperature for 4 h. To fabricate the pressure sensor device, the CNT/PDMS film with the pyramidal microstructure was placed face-to-face on a patterned Au/PDMS film which works as the bottom electrode. Silver paste and copper wires were used for connection.

Preparation of threshold controlling unit (TCU).

The ZIF-8-based TCU device was fabricated according to our previously reported method.^[4-6] Patterned Au/PDMS film on silicon wafer was prepared as mentioned before. After oxygen plasma treatment (80% of power, 1 min), two layers of ZIF-8 were allowed to grow on top of the desired Au/PDMS area by immersing the Au/PDMS film into a mixed methanol solution of $\text{Zn}(\text{NO}_3)_2$ (25 mM) and 2-methylimidazole (50 mM) for 2 growth cycles (30 min for each cycle). The unwanted area was masked by covering a thin layer of PDMS. The film was then rinsed thoroughly with methanol and dried by nitrogen flow. A copper wire was embedded in PDMS and the two ends were exposed. One end was dipped into silver paste and dried in air. This silver paste painted tip was used as the top electrode to connect the ZIF-8 area.

Preparation of electrochemical actuator devices.

The MWCNT-based actuators were fabricated according to the literature with minor modification.^[7,8] Typically, 100 mg of MWCNTs, 100 mg of MnO₂, 240 mg of EMIMBF₄ and 160 mg of PvdF (HFP) were mixed and dispersed in 20 mL of DMAC using ultrasonication. MnO₂ powder was prepared according to the reported literature.^[9] The electrode film was prepared by casting 2.5 mL of the resultant suspension on a glass slide (75×25 mm) and evaporating the solvent at 80°C in an oven. To enhance the conductivity, a layer of gold (400-600 nm) was deposited on the electrode film using thermal evaporation. To fabricate the electrolyte film, 1 g of EMIMBF₄, 1 g of PvdF(HFP) and 2.5 g of propylene carbonate were dispersed into 20 mL of 4-methyl-2-pentanone. The suspension was refluxed at 80°C for 2 h. 1 mL of the solution was cast on a glass slide and dried at 80°C. The final actuator devices was fabricated by laminating one piece of electrolyte film in between two pieces of electrode film using hot-pressing.

Methods and characterization. For morphology study, field emission scanning electron microscopy (FESEM) was performed using a JEOL/JSM-7600F equipment. Electrical characterization was performed by Keithley 4200 semiconductor characterization system. For loading pressure, a motorized force test stand with PC control (Mark-10) was employed. Alternatively, standard weights were also used. The responding time of the pressure sensor was measured by using an oscilloscope (Tektronix DPO5054B). Force/pressure testing was carried out by using the force gauge (Mark-10, M5-2) or by using an analytical balance. 3D-printing was performed by Eden 260 and Ultimaker 2.

Figure S1. Morphology characterization of the CNT/PDMS pressure sensor. a,b,c,d) Scanning electron microscope (SEM) images of the CNT/PDMS films with the pyramidal microstructure. The length of an individual pattern is 30 μm. The CNTs deposited can be identified in the high magnification images (c,d).

Figure S2. Configuration of the patterned Au/PDMS film as the bottom electrode. a) Digital photograph of the patterned Au/PDMS film. The dashed rectangles denote the area designed for further device integration. b) Illustration of stretching the Au/PDMS film.

Figure S3. Measurements of the pressure of human fingertip tapping. The subject (male, 30 yr old) was instructed to perform fast tapping on the force gauge for many times. The subject was requested to perform very gentle tapping first. Then the subject was requested to perform firm and stronger tapping. As shown in the figures, the gentle tapping ranges from 200 Pa to 1200 Pa and the average value is about 500 Pa. The firm tapping ranges from 1100 Pa to 8200 Pa and average value is about 2200 Pa.

Figure S4. Performance of the pressure sensor based on microstructured AgNW/PDMS. a) Typical I - V curves of the pressure sensor at various pressures. b) Instant current change in response to different applied pressures. c) Plotting of the resistance response to the applied pressures. Due to the excellent conductivity of the AgNWs, the resistance of pressure sensor only ranges from several tens of ohms to a few hundred ohms even the applied pressure is in low range (<1 kPa). As a result, the pressure detection range is limited. d) High magnification SEM image of the AgNW/PDMS film showing the AgNWs deposited on top of the PDMS.

Figure S5. Morphology and performance of the ZIF-8-based devices with different numbers of deposition cycles. a) Cross-sectional SEM images of different layers of ZIF-8 film deposited on silicon substrates. The SEM images suggest that the thickness of the ZIF-8 film increases with increasing the number of deposition cycles, which is consistent with the reported results.^[6] b) Typical I - V curves of TCU devices with 1 layer and 3 layers of ZIF-8 deposited.

Figure S6. Supplementary electrical characterization of the TCU device. a) The HRS and LRS resistance of a typical TCU device for 15 cycles. b) Device-to-device distribution of set and reset voltages of a typical TCU device.

Figure S7. Flexibility characterization of the ZIF-8-based TCU device. a) The HRS and LRS resistance of a typical TCU device at different bending states. b) The set and reset voltages of a typical TCU device at different bending states. The device was placed on a polyethylene terephthalate (PET) substrate and the testing configuration is shown as the inset.

Figure S8. Morphology characterization of the actuator electrode films. a) Digital photographs of the MWCNT/MnO₂ electrode films without (left) and with (right) gold-coating. b) SEM image of the film without gold-coating. c,d) High magnification SEM images of the films without (c) and with (d) gold-coating. As shown in (c), the entangled MWCNTs form a network with the MnO₂ particles dispersed uniformly, which enhances the capacitance. To increase the conductivity, a thick layer of gold (400-600 nm) was deposited on top of the film (d).

Figure S9. Electrochemical characterization of the actuator. a) Cyclic voltammetry at a scanning rate of 1mV/s and b) galvanostatic charge-discharge at a current density of 1 A/g of a typical actuator strip. The specific capacitance is calculated to be 58 F/g according to the CV curve. And the equivalent series resistance (ESR) is calculated to be 70 Ω (25×5 mm sized) according to the charge-discharge curve.

Figure S10. Electrochemical characterization of the actuators prepared by electrodes with different MWCNT/MnO₂ weight ratios. a) Cyclic voltammetry at a scanning rate of 10 mV/s of actuator strips with electrode films prepared using different MWCNT/MnO₂ weight ratios. b) Photograph of the electrode film prepared with a ratio of 1:2.0. The CV curve of ratio 1:2.0 possesses the largest specific capacitance due to the presence of larger amount of MnO₂. However, the electrode film prepared with ratio of 1:2.0 easily cracks.

Figure S11. Bending performance of the actuator. Digital photographs of the actuator strip bending towards both directions upon applying a constant voltage of ± 3 V.

Figure S12. Electrical characterization of the actuators prepared using electrode films with and without gold-coating. Typical I - T curves of the actuator strips without and with gold-coating at an applied constant voltage of 3 V. The inset shows the testing configuration. As shown in the figure, the current flow increases significantly as the conductivity is enhanced greatly after gold-coating. As a result, the bending of the actuator strip becomes faster.

Figure S13. Testing configuration of the pressure sensor and TCU integrated system. a,b) Digital photographs showing the top view (a) and side view (b) of the integrated system placed on a curved surface with a bending radius of 0.7 cm. c) Liquid metal and copper wires were used for connection.

Figure S14. Response time of the pressure sensor and TCU integrated system. a) Schematic illustration of the pressure sensor and the TCU integrated system. b) Illustration of the testing configuration. c) The response time of the pressure sensor and the TCU integrated system (CH 2) and the pressure sensor (CH 1) alone. The response time of the integrated system was measured by an oscilloscope using pressure sensor (CH 1) alone as a reference. Pressure was applied on to both pressure sensors (CH 1 and CH 2) simultaneously by applying a standard weight. The devices were in series with a constant resistor (R_0 , 1 M Ω) and a constant voltage of 3 V was applied. The voltage changes on R_0 were monitored. The response time of the integrated system is estimated to be *ca.* 0.8 ms, which is compatible with the response time range in biological systems. The abrupt current transition between point a and b (CH 2) is due to the resistance state change of the TCU device. Because a typical MOF-based memristor displays a much faster response (~ 70 μ s),^[5] the total response time (point a-c) of the integrated system depends on the pressure sensor, which is slower.

Figure S15. Assembly of the reed relay and the designed circuit. a) Photograph of a commercial reed switch. b) Photograph of a coil recovered from an alarm clock. c) Photograph of the reed relay composed by the reed switch and the coil. d,e) Illustration of connecting an LED bulb using the relay with a relay switching current as low as 0.58 mA. f) Design of the circuit for the artificial reflex arc system (C_{Ctrl} : controlling circuit, C_{Press} : resistive switching circuit, C_{Act} : actuation circuit). A constant voltage is applied for all the circuits (V_1 : 2.5 V, V_2 : 4.0 V, V_3 : 3.0 V). A current compliance of 1 mA was applied to C_{Ctrl} and a compliance of 100 mA was applied to C_{Act} .

To realize higher current supply and hence fast actuation, the integrated system and the actuators were not electrically connected but coupled using a reed relay. The designed circuit

is comprised of a controlling circuit (C_{Ctrl}) and an actuation circuit (C_{Act}). Besides, a resistance switching circuit (C_{Press}) is set up to change the resistance state of the TCU. Before sensing any pressure, the TCU is in HRS and the corresponding current flow in the controlling circuit is very low ($<10^{-9}$ A), which is not sufficient to switch on the reed switch and thus the actuation system. Once the pressure sensor detects a pressure beyond the threshold, the TCU is activated and switched to LRS. Subsequently, the current flow in the controlling circuit would be adequate to switch on the reed switch and the actuation circuit.

Figure S16. Circuit components and design of the actuation system for the robot demo. Digital photographs showing a,b) the circuit components and c,d) the actuation system designed for the robot demo. The robot was printed using an Eden 260. The original left arm was removed and reconstructed. The scaffold for the left arm was printed using Ultimaker 2. The L-shaped Au-coated PET film was prepared by typical thermal evaporation.

Figure S17. Multiple tapping test. The instant current change of the actuators responding to multiple finger tapping applied on the pressure sensor of the artificial somatic reflex arc of the robot. To demonstrate the threshold-dependent response, a multiple tapping test was performed. As shown in the Figure, the actuators only responded to a pressure above the threshold indicated by the charging current in the actuators.

Figure S18. Reversing actuation of the actuators. Digital photographs showing the hand releasing actions. To achieve this opposite actuation, we only need to reverse the polarity of the input voltage for the actuation circuit (C_{Act}).

Table S1. Comparison of recent works relating to artificial reflex arc for robotics

Typical relative work	Biomimicking concept /Functionality	System architecture	System size	Major components and flexibility		System response time	Technical performance	Reference
				Components	Flexibility (Elastic modulus)			
Works based on solid-state electronics	Artificial reflex arc Built a hierarchical controlling system with the low level controlling system reproducing human spinal reflex		~ m	Sensing Force sensor Position sensor Angular sensor	Not flexible Rigid materials >100 GPa ^[10]	Sensing + Processing	1) Conventional rigid-bodied robots can be programmed to perform complex tasks precisely and efficiently 2) Require complex hierarchical device configuration for sensory input, processing and motion control 3) Large in size and heavy in weight	[11]
				Processing PC	Not flexible Silicon-based 130 GPa ^[12]	Not given		
				Actuation McKibben artificial muscles	Flexible Rubber (bladder) 40-50 MPa Nylon (shell) 1-3 GPa	Sensing + Processing + Actuation		
						Not given		
	Artificial reflex arc Realize the locomotion of a limbless robot with local leg reflexes mimicking the biological reflex arcs		0.45 m	Sensing Touch switch (Omron D2MQ) Position sensor (Futaba s3003)	Not flexible Rigid materials >100 GPa	Sensing + Processing	4) Mechanical mismatching with biological materials and organisms Not able to be integrated into soft robotics.	[14]
				Processing PC	Not flexible Silicon-based 130 GPa	Not given		
				Actuation RC servo motor (Futaba s3003)	Not flexible Rigid materials >100 GPa	Sensing + Processing + Actuation		
						< 0.5 s		
	Artificial reflex arc Realized hand artificial reflex for automatic readjustment during hand transport		> 30 cm	Sensing Dry EMG electrodes with integrated amplifiers Creative Senz3D camera	Not flexible Rigid materials >100 GPa	Sensing + Processing	4) Mechanical mismatching with biological materials and organisms Not able to be integrated into soft robotics.	[14]
				Processing Standard PC with 16GB RAM and four-core i7@2.9 GHz CPU	Not flexible Silicon-based 130 GPa	Sensing + Processing + Actuation		
				Actuation Michelangelo left-hand prosthesis with a wrist rotator	Not flexible Rigid materials >100 GPa	< 0.75 s		
						> 0.85 s		
	Artificial reflex arc Realized a local closed-loop tactile feedback control system to mimic the natural reflex for adjustments during grasping		> 30 cm	Sensing Tactile force sensors (Stretchy Piezo LTT-SLPA-150k) EMG electrodes (Myobock, Ottobock, Austin, USA)	Flexible Nylon /Spandex /conductive coating > 1 GPa	Sensing + Processing	Not given	[15]
				Processing Prosthesis controller (Infinite Biomedical)	Not flexible Rigid materials >100 GPa	Sensing + Processing + Actuation		
					Not flexible Silicon-based 130 GPa	~ 1 s		

				Technologies, USA) PC				
				Actuation Motors	Not flexible Rigid materials >100 GPa			
Works based on flexible devices	Hybrid bioelectronic reflex Realized pressure signal detection, digital signal conversion and signal processing by synaptic transistor.		~ 4 cm	Sensing Resistive pressure sensor Processing Organic ring oscillators Synaptic transistors Actuation A discoid cockroach leg	Flexible CNT/elastomers ~ 3 GPa ^[16] (polyimide substrate) Flexible Pseudo-complementary CMOS inverters ~ 3 GPa (polyimide substrate) Flexible Polymer semiconductor and ion gel ~ 3 GPa (polyimide substrate) Flexible Musculo-skeletal < 1 GPa (muscles) ~ 13 GPa ^[18] (apodeme)	Sensing + Processing Not given. Sensing + Processing + Actuation > 0.1 s	1) Realized sensory input and signal processing using simple device configuration. 2) Smaller in size and lighter in weight. 3) All the major devices are flexible and suitable to be integrated into soft robotics.	[17]
	Artificial somatic reflex arc Realized an “all-or-none” actuation of electrochemical actuators in response to tactile stimulation above the stimulus threshold emulating a biological somatic reflex action. Employed pressure sensor, threshold controlling unit (TCU) and electrochemical actuators to mimic the three major components of a biological reflex arc (sensory receptor, integrating center, and effector).		~ 3 cm	Sensing Resistive pressure sensor Processing Metal-organic framework (MOF)-based threshold controlling unit (TCU) Actuation Electro-chemical actuator	Flexible CNT/PDMS ~ 2 MPa (PDMS substrate) Flexible ZIF-8/PDMS ~ 2 MPa (PDMS substrate) Flexible MWCNT/Pvdf/MnO ₂ 80-120 MPa ^[8]	Sensing + Processing ~0.8 ms Sensing + Processing + Actuation < 1.0 s	1) Realized sensory input signal processing and response motion using simplified autonomous control loop. 2) The motion responds to the stimulus in an all-or-none pattern. 3) Smaller in size and lighter in weight. 4) All the major devices are flexible and suitable to be integrated into soft robotics.	This work

Supporting References

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