A new opportunity for the emerging tellurium semiconductor: resistive switching device implementation

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Abstract

The electrochemical (EC) resistive switching (RS) cross-point arrays, composed of nonvolatile RS (NV-RS) memories and volatile RS (V-RS) selectors, hold promise for high-density data storage, in-memory computing and neuromorphic computing. However, the conventional EC-RS devices based on metallic filaments suffer from the notorious current-volatility dilemma that the low and high current requirements for NV-RS memories and V-RS selectors, respectively, cannot be satisfied simultaneously, due to the dominant EC nature of the RS. In this work, we demonstrate electrochemically active, low thermal-conductivity and low melting-temperature semiconducting tellurium filament-based RS devices that solve this dilemma, enabling NV-RS memories to operate under lower currents than do V-RS selectors. This novel phenomenon arises as the consequence of the adversarial EC and Joule heating (JH) effects. The devices also show unusual stimulus frequency dependent long-term plasticity (LTP)-to-short-term plasticity (STP) transition. Devices with this property can be generically utilized as spatial-temporal filters in spiking neural networks (SNNs) for high-performance event-based visual recognition tasks, as illustrated in our noise filtering simulations. By regulating the EC-JH relationship using dielectric materials with decreasing thermal conductivities, full functional-range tunable Te filament-based devices, from always-NV RS, to NV-to-V transitionable RS, and to always-V RS, are also demonstrated. The tellurium filament-based RS devices are promising enablers for functional cross-point arrays.
Introduction

The resistive switching (RS) cross-point arrays are emerging technologies for high-density data storage and nonconventional information processing, such as neuromorphic computing, in-memory computing and machine learning.[1-19] For large-scale arrays, each cross-point consists of a nonvolatile RS (NV-RS) memory device, and in series, a three-terminal transistor or a two-terminal volatile RS (V-RS) selector device to suppress the undesired sneak-path currents. So far, although transistors enable the most reliable array operations, V-RS selectors are of great promise for maximizing the density of integration.[20-24] There are various types of NV-RS memories based on a broad category of materials and physical mechanisms,[25] including phase change memories,[26-30] ferroelectric memories,[31-33] magnetoresistive memories,[34-35] valance change memories,[36-38] electrochemical (EC)-RS memories,[39] and so on. For V-RS selectors, nonlinear resistors such as Schottky diodes[40] and RS devices such as ovonic threshold switches (OTS),[41-42] Mott selectors,[43-44] EC-RS selectors,[45] and the mixed-ionic-electronic conduction type selectors[46] and field assisted superlinear threshold selectors based on undisclosed materials,[47] have gained considerable research interests. Among these device technologies, EC-RS devices are particularly attractive in terms of the simplicity of the working principles and the diversity of functions, including NV-RS memories[87-89], V-RS selectors[90] and neuronal emulators[55].

In addition to the different switching behaviors, NV-RS memories and V-RS selectors also have distinct operating requirements: the operating currents of NV-RS memories need to be as low as possible to minimize power consumption, while the V-RS selectors should be able to operate under high ON-state currents to ensure successful writing to the memories and provide sufficient read margins. Considerable progress has been made recently to satisfy these requirements that NV-RS memories with operating currents as low as 10 pA[48-51], and nonlinear resistor-type of V-RS selectors[40] and chalcogenide-based OTS V-RS selectors with ON-state current densities exceeding 10^7 A/cm^2 and 20 MA/cm^2,[41-42,52] respectively, have been demonstrated.

Despite the performance boosts for NV-RS memories and V-RS selectors by exploiting materials satisfying the respective requirements, a long-standing challenge, namely, the current-volatility dilemma, has impeded the use of the same materials and therefore the same mechanisms for both types of devices, which significantly increases the design complexity of the RS cross-point arrays. To be specific, for the same RS device, V-RS normally occurs under low operating current, and a transition from V-RS to NV-RS can take place if the current is sufficiently high. These characteristics are in contrast to the device requirements aforementioned. This dilemma has been confronted with by almost all existing RS devices.

In an attempt to solve the current-volatility dilemma, Zhao et al.[56] introduced graphene with controlled structure defects to the Ag/SiO_2-based EC-RS devices and realized low and high operating currents for NV-RS memories and V-RS selectors, respectively, based on the same materials. However, this is achieved at the expense of fabrication complexity of using CMOS-incompatible graphene material and engineering graphene
properties for different devices. In other EC-RS devices, either V-RS phenomena under high currents\cite{57} or NV-RS under low currents\cite{51} have been demonstrated. However, the current-volatility dilemma has never been solved in the same devices.

For EC-RS devices, the difficulty to solve the current-volatility dilemma originates from the fact that the stabilities of the filaments are monotonically dependent on the strengths of the EC effects, and therefore on the operating currents\cite{54}, that govern the filament growth during RS. The Joule heating (JH) effect is normally considered to be secondary and synergetic that it assists the lateral diffusion of atoms or ions, resulting in the increase of the thickness of the filament and therefore its stability.\cite{58} In light of this, a clue to the solution of this dilemma is using effects that counteract the EC effect in filament growth.

Here, we consider the possibility of using the intrinsic JH effect as the counteractive effect. To this end, filaments composed of materials with low melting temperatures are useful because they tend to be ruptured when heats are accumulated. Furthermore, filamentary materials with low thermal conductivities are able to confine heats and facilitate heat accumulations. In addition, of course, new materials should also be electrochemically active to enable filament growth.

Currently, main reported filamentary materials for EC-RS devices are all metals with relatively high thermal conductivities and high melting temperatures\cite{25}. Recently, tellurium (Te), as an emerging semiconductor material for the next-generation transistors\cite{65-68}, has been found to be electrochemically active\cite{59-64}. Associated with its semiconductivity, Te also has low thermal conductivity (1.6 W m^{-1} K^{-1})\cite{69} compared to those of the metallic filamentary materials (e.g. Cu: 401 W m^{-1} K^{-1}; Ag: 429 W m^{-1} K^{-1})\cite{83}. This has been one of the main reason behind its attractive thermoelectric performance\cite{69}. In addition, Te has the second lowest melting point (452 °C)\cite{82} among all elemental semiconductors. These properties endow Te with the aptitude to be the new filamentary material.

In this work, we demonstrated RS devices based on semiconducting Te filaments. Unlike conventional EC-RS devices based on metallic filaments, our devices showed novel behaviors that the NV-RS occurred under lower operating current than did the V-RS, solving the notorious current-volatility dilemma. The adversarial EC-JH relationship also led to unusual stimulus frequency dependent long-term plasticity (LTP) to short-term plasticity (STP) transition under electrical pulse train measurements, in contrast to the commonly observed STP-to-LTP transition in EC-RS devices. This device character could be utilized in spatial-temporal filter layers in spiking neural networks (SNNs) for high-performance event-based visual recognition tasks, as demonstrated in our noise filtering simulations. We further demonstrated full functional-range tunable Te filament-based devices, from always-NV RS, to NV-to-V transitionable RS, and to always-V RS, using dielectric materials (Bi_{2}Te_{3}, Sb_{2}Te_{3} and TiTe_{2}) with decreasing thermal conductivities, which could also be understood on the basis of the regulated EC-JH relationships.
Results and discussion

The transmission electron microscopy (TEM) image of the cross-section of the Te/Sb$_2$Te$_3$/Te RS device is shown in figure 1(a). The energy-dispersive X-ray spectroscopy (EDS) mapping confirmed that the deposited Sb$_2$Te$_3$ film was close to its stoichiometry, with 39.37% of Sb and 60.63% of Te, as shown in figure 1(b).

![Figure 1](image)

**Figure 1** (a) TEM image of the cross-section of the Te/Sb$_2$Te$_3$/Te RS device. (b) EDS elemental mapping image of the Te/Sb$_2$Te$_3$/Te device. (c) I-V curves of the Te/Sb$_2$Te$_3$/Te device under the CC of 300 μA, 500 μA, 700 μA and 900 μA. (d) Endurance test of the device under 500 μA CC. (e) I-V curves of the device under the CC of 1.5 mA, 2 mA, and 2.5 mA. (f) Endurance test of the device under 1.5 mA CC.
The I-V curves for the RS processes of a 2×2 μm² device under four compliance currents (CCs) are shown in figure 1(c). When the CC was 300 μA or above, the measured current under the positive voltage sweep from 0 V was sharply increased to the CC limit at ~1 V and the device reached its state of low resistance (LR). This is defined as the SET process (arrow 1). The device kept LR under the backward sweep from the positive voltage region (arrow 2) to the negative voltage region until the negative voltage reached ~ -0.7 V at which the current rapidly dropped to a low value, switching the device back to the state of high resistance (HR). This is defined as the RESET process (arrow 3). The device then kept HR under the sweep from the negative voltage region (arrow 4) to the positive voltage region before the next SET switching occurred. These were typical NV-RS behaviors because the resistance states of the device were maintained even if the applied voltages approached 0 V. The RS windows (hysteresis loops) were enlarged for devices under increased CCs. The NV-RS behavior was still significant under more than 10² cyclic sweeps, as shown in figure 1(d).

Interestingly, when the applied CC was increased to ~1.5 mA, a transition from NV-RS to V-RS occurred. As illustrated in figure 1(e), the device was first switched to the LR state under forward sweep in the positive voltage region (arrow 1), similar to the SET behavior of devices under lower CCs. Under the backward sweep to 0 V, a dramatic drop of the current occurred at 0.7 V (arrow 2), indicating the transition from LR to HR. When the backward sweep continued that the sign of the applied voltage reversed, no RESET behavior was observed. Instead, the device underwent SET switching again (arrow 3). This confirmed the volatile nature of the LR state obtained after the first SET switching. After the second SET switching, the applied voltage was swept from the negative voltage region to 0 V. Again, a dramatic drop of the current at ~0.7 V occurred (arrow 4), indicated by a box-shape RS window. This also reflected the volatile nature of the LR state obtained after the second SET switching. The V-RS behavior under 1.5 mA CC was still significant under more than fifty cyclic sweeps, as depicted in figure 1(f). When the CC was further increased to 2.5 mA, the volatile nature of the LR state obtained after the SET switching under both the positive and negative voltages was still evident.

Devices with such V-RS behavior are always used as the selectors in series with the NV-RS memory devices in the crossbar arrays to suppress the sneak-path problem. One of the main requirements for selector devices is high ON current to provide sufficient read margins and ensure successful writing to the memory devices. However, this is very challenging due to the aforementioned current-volatility dilemma. Our devices not only solved this dilemma that they showed NV-RS behavior under low CCs but V-RS behavior under much higher CCs but also achieved the 2.5 mA ON-current, equivalent to 6×10⁴ A/cm², which was among the highest current density values achieved by the state-of-the-art EC-type selector devices.

To understand the mechanism of RS, we first noted that Sb₂Te₃ is a well-known chalcogenide phase change material (PCM) whose solid phase transition between the amorphous state and the crystalline state results in NV-RS. However, our devices were not likely to have such phase change-type switching because of the much higher required current, namely, 19 A for 2×2 μm² devices (extrapolated from the reported
RESET current density value for a device with much smaller size),\textsuperscript{[70]} than the maximum CC under test. Actually, PCMs have often been utilized as the dielectric layer in EC-RS devices but most of these devices used electrochemically active Ag or Cu electrodes.\textsuperscript{[71-74]} Recently, the use of Te electrodes was also found to enable RS \textsuperscript{[59-64]} in which the formation of local conducting filaments led to HR-to-LR transition while the rupture of filaments led to the reverse transition. In order to confirm whether the RS phenomenon for our Te/Sb\textsubscript{2}Te\textsubscript{3}/Te devices was based on such process, we measured the electrode area dependent HRs and LRs of the device in the NV-RS (under 500 μA CC) and V-RS (under 1.5 mA CC) modes, respectively. As shown in figure 2(a), the HR was decreased with increasing area, whereas the LR was almost independent of the electrode area. These strongly indicated that the RS was originated from the formation and rupture of the localized conducting filaments. Considering the use of Te as the electrode material, the filaments formed in our devices were most likely to be Te.

TEM observations of the device after NV-SET operation and V-RS operation were also carried out. Local RS region was identified from a wide range of cross-section area. For the NV-SET sample, a cone-like Te filament (5 nm thick) connecting the top and bottom electrodes was observed with EDS signal showing predominant Te composition (~82.32%), as seen in figure 2(b). For the V-RS sample, instead of connected filaments, isolated Te nanoclusters dispersed in the Sb\textsubscript{2}Te\textsubscript{3} matrix were observed, as seen in figure 2(c). The Te nanoclusters were expected to be the remains of the ruptured filament.
Figure 2 (a) Dependence of the resistance on the electrode area for both the high- and low-resistance states. Cross-sectional TEM images of the Te/Sb$_2$Te$_3$/Te device after (b) NV-SET and (c) V-RS. Simulations of the temperature distributions in (d) Ag filament-based device and (e) Te filament-based device.
As previously introduced, the current-volatility dilemma arises from the monotonic relationship between current and the stability of the filament which is a natural result of the dominant EC effect and the synergetic effect from JH in the filamentary process. Therefore, breaking this monotonic current-filament stability relationship by inducing counteraction between these two effects may overcome the dilemma. In fact, the JH effect does not always assist the growth of the filaments. For example, JH has been believed to rupture the filaments in devices showing unipolar-RS or unusual V-RS behaviors [80,81,91-96]. In those cases, the grown filaments after the SET processes were conducting enough that even low RESET voltages led to large currents and therefore accumulated significant JH to fuse the filaments. It should be noted that in our cases, JH accumulation might be more pronounced because of the low thermal conductivity of Te and Te was more vulnerable to overheat because of the lower melting point.

COMSOL simulations were carried out to study the different temperature fields evolved in a Te filament-based device and a Ag filament-based device under the same CC of 1 mA. According to our TEM analysis, truncated-cone-shaped filaments were adopted in the simulations. Both systems under simulations adopted the same geometry and size. To simulate the generation of JH, the experimental electrical conductivities of the filament materials at room temperature were used as input parameters and their temperature-dependent evolutions were explicitly simulated. To simulate heat transfer, we used the same room-temperature thermal conductivities of the filament materials throughout the simulations because they are relatively temperature independent. The results of the simulations are depicted in figure 2(d,e), from which we found that JH was more localized at the thinner end of the Te filament than was in the Ag filament because Te has poorer thermal conductivity. In addition to the stronger thermal confinement for the Te filament observed from the simulations, it was also clear that under the same simulated CC of 1 mA the highest local temperature in the Te filament reached its melting temperature, while the melting temperature of Ag (961°C) was not reached in the Ag filament-based device.

Based on the experimental and simulation results, a comparison between the operation mechanism of the metallic filament-based device and that of the Te filament-based device is schematically depicted in figure 3. The abnormal NV-RS-to-V-RS transition in Te/Sb:Te/Te device could be attributed to the JH effect that counteracted the EC effect and became dominant under high CCs.
The unique NV-to-V transitional RS in the Te/Sb2Te3/Te device was also observed in the pulse train measurements, as illustrated in figure 4(a,b). Two write pulse (±1 V) widths were tested, 1 μs and 1 ms. Read pulses of 0.1 V immediately prior to and after the write pulses were used to read out the resistance states of the device before and after pulse programming. After being stimulated by a positive pulse of the width of 10 μs, the measured after-write current was obviously larger than the before-write current by ~100 μA, verifying the occurrence of the NV HR-to-LR RS, namely, the SET process. Successive stimulation by the -1 V pulse of the same width resulted in the reversed NV LR-to-HR RS, namely, the RESET process, that the device completely recover to its initial HR state. In stark contrast to the phenomena under short pulse stimulations, 1 ms pulse stimulations led to V-RS. After being simulated by the pulse, the after-write current was almost identical to the before-write current, which was a typical feature of the V-RS. This NV-to-V transitional RS can be understood as the result of JH accumulation over time. Under longer pulses, the JH effect became too prominent to counteract the EC effect, resulting in the spontaneous rupture of the just-formed filament. The HR/LR ratios for the device under both RS modes remained as large as $10^3$ after repeated pulse stimulations with alternating voltage polarities, as showed in figure 4(c,d).

The competition between the EC and JH effects had led to an unusual stimulus frequency dependent LTP-to-STP transition. The width and the amplitude of the pulse were fixed at 10 μs and 0.6 V, respectively, to avoid the over-accumulation of JH in a single pulse. The stimulus frequencies were adjusted by changing the pulse intervals.
When the interval was set to 10 μs, an analog RS behavior was observed that the resistance of the device gradually decreased, as shown in figure 4(e). Interestingly, when the interval was reduced to 1 μs, the resistance of the device first underwent a more rapid decrease and then a sudden increase back to an early HR, mimicking a transition from LTP to STP. According to the mechanism we proposed above, this could also be understood as the result of the accumulated JH that gradually surpassed the EC effect with decreasing stimulus interval to allow JH dissipation.

Figure 4 (a) Pulse train measurement with write pulse width of 10 μs and amplitude of ±1 V. (b) Pulse train measurement with write pulse width of 1 ms and amplitude of ±1 V. (c) 50 cycles of 10 μs-pulse measurements with alternating voltage polarities. (d) 50 cycles of 1 ms-pulse measurements with alternating voltage polarities. (e) Pulse train measurement with write pulse width of 10 μs, amplitude of ±0.6 V and 10 μs interval. (f) Pulse train measurement with write pulse width of 10 μs, amplitude of ±0.6 V and 1 μs interval.
As well been recognized, synaptic plasticity over different timescales have a wealth of computational functions\(^1\). For example, LTP is the neuronal foundation of learning and development, and STP can act as a temporal filter. Here, we made use of our Te/Sb\(_2\)Te\(_3\)/Te device that showed LTP-to-STP transition under high stimulus frequency in a unique event-based vision preprocessing layer that functioned to reduce the high-frequency noise and thus enhanced the overall recognition accuracy of the SNN. Event-based camera, based on dynamic vision sensor (DVS), is a kind of bio-inspired camera that senses continuous flows of asynchronous spatial events, and responds as they occur or stay silent otherwise, as shown in figure 5(a). We used the event-based Neuromorphic MNIST (N-MNIST) dataset as our training and test set\(^1\). Compared to traditional frame-based MNIST datasets, N-MNIST contains richer temporal features and sparser information representations. As shown in figure 5(b), each original image frame from MNIST dataset can be transformed into dynamic events by the saccade of the event-based camera. Figure 5(c) represents the recording results for a sample image of digit 5 along different saccade paths. Two detection channels that respond to different brightness change directions of the pixels, brighter and darker, are shown in red and green, respectively.

Despite its bio-plausibility and low power consumption, the event-based camera usually suffers from noise disturbance\(^1\). To simulate the inherent noise during visual information acquisition, we used uniform distribution functions to produce certain noise patterns (pixel coverage, PC) and added the noise to a certain percentage (noise frequency) of the N-MNIST test data. The event-based SNN visual recognition framework is depicted in figure 5(d). The SNN had already been trained using noise-free training data from the same dataset (see Implementation details of network simulation). During test, a LSL before the SNN was added to preprocess the visual inputs. The LSL was parameterized according to the measured behavior of the real devices (see Implementation details of network simulation). In the LSL, event noises with high frequencies (noises being added to a high percentage of the test data) were filtered out as the LSL shifted to the STP operation mode. Genuine event signals remained almost intact due to their low-frequency nature (the few activated DVS pixels changed constantly). After preprocessing, visual recognition continued as usual by the following SNN processing.

The recognition accuracy is shown in figure 5(e). It was seen that with increasing noise frequency the performance of the SNN without LSL degraded monotonically. In particular, the relative recognition accuracy for highly noisy (100%) data with respect to that for noise-free data decreased below 30%. With the LSL included, the problem of accuracy degradation was mitigated. Interestingly, with increasing noise frequency the recovery of the accuracy was more complete. This was understandable from the frequency dependent STP strength. For highly noisy (100%) data, the accuracy could even be recovered to the same level as noise-free data recognition. The simulation results provided a glimpse of the potential computational advantages of the Te/Sb\(_2\)Te\(_3\)/Te device.
In order to satisfy the device requirements in different application domains, the ability to regulate the RS behaviors is important. Previous studies have shown that for the same filamentary materials the dielectric matrixes might have significant influence on the RS behaviors [74,97-99]. The rate of the EC redox process, ion mobility and dielectric conductivity have been considered as the main regulators. As discussed above, Te filament has low thermal conductivity compared to conventional metallic filaments, which is likely to be associated with the unusual current-volatility relationship in Te/Sb$_2$Te$_3$/Te device. Here, we further investigated whether the thermal conductivity of the dielectric matrix had an effect on the RS behavior of the Te filament-based device. To this end, two control devices, Te/Bi$_2$Te$_3$/Te and Te/TiTe$_2$/Te, whose binary telluride dielectrics have thermal conductivities higher and lower, respectively, than that of Sb$_2$Te$_3$ (~1.2 W m$^{-1}$K$^{-1}$[84] for Bi$_2$Te$_3$, ~0.78 W m$^{-1}$K$^{-1}$[86] for Sb$_2$Te$_3$ and ~0.12 W m$^{-1}$K$^{-1}$[86] for TiTe$_2$) were investigated.

As illustrated in figure 6(a), the Te/Bi$_2$Te$_3$/Te device always underwent NV-RS over the range of CCs from 100 μA to 1.6 mA, a comparable CC range to that tested for the Te/Sb$_2$Te$_3$/Te device. The upper limit of the CC was not further increased to avoid the occurrence of hard breakdown. On the other hand, the Te/TiTe$_2$/Te device always underwent V-RS over the range of CCs from 200 μA to 1.5 mA, as shown in figure 6(b). Therefore, full functional-range tunable Te filament-based devices, from always-NV RS, to NV-to-V transitionable RS, and to always-V RS, are achieved by using Bi$_2$Te$_3$, Sb$_2$Te$_3$ and TiTe$_2$ dielectric materials, respectively. The rich dielectric dependent RS phenomena in Te filament-based devices can also be understood from the EC-JH competitive mechanism that high (low) thermal-conductivity dielectric

**Figure 5** (a) Schematic diagram of neuromorphic data acquisition via event-based camera. (b) Three saccade trajectories of the camera during data acquisition process. (c) Event-based data acquired by the camera along the three saccade trajectories. (d) Schematic diagram of SNN processing of the N-MNIST data with LSL. (e) Performance comparison between SNNs with LSL and without LSL.
facilitates (suppresses) JH dissipation and therefore makes the JH effect less (more) pronounced to counteract the EC effect.

Thermodynamics simulations of these three devices under the CCs ranging from 0.7 mA to 1.5 mA (in the vicinity of the NV-to-V transition point of the Te/Sb$_2$Te$_3$/Te device) were performed to further clarify the temperature distribution in the devices. For simplicity, cylinder-shaped filaments of the same size were taken as models. As shown in figure 6(c), the simulated highest local temperature in the Te/Bi$_2$Te$_3$/Te (Te/TiTe$_2$/Te) device was always lower (higher) than the melting temperature of Te. For Te/Sb$_2$Te$_3$/Te device with intermediate dielectric thermal conductivity, a crossover was observed. Although not in fully quantitative agreement with the experiments due to the use of simplified models, the simulated temperature trends well reproduced the experimental RS behaviors of the three devices. Figure 6(d) shows the temperature fields in these three devices under the CC of 1.3 mA. Due to the lowest dielectric thermal conductivity, the Te/TiTe$_2$/Te device showed the most confined temperature distribution around the filament and the highest peak local temperature of 688 °C, sufficient to fuse the Te filament. In contrast, the Te/Bi$_2$Te$_3$/Te device with the highest dielectric thermal conductivity showed the most expanded temperature distribution and the lowest peak local temperature of 401 °C, lower than the melting temperature of Te. The Te/Sb$_2$Te$_3$/Te device was intermediate.

Figure 6 I–V curves of the (a) Te/Bi$_2$Te$_3$/Te device and (b) the Te/ TiTe$_2$/Te device under different CCs. (c) Simulated CC dependent local peak temperature in the Te/Bi$_2$Te$_3$/Te, Te/Sb$_2$Te$_3$/Te and Te/TiTe$_2$/Te devices. (d) Simulated temperature distributions in these three devices under 1.3 mA CC.
Conclusion

To conclude, we have demonstrated a new application opportunity for Te in RS devices. Te-based RS devices solved the long-standing current-volatility dilemma, enabling NV-RS memories to operate under lower currents than do V-RS selectors as required by cross-point array applications. This novel phenomenon can be attributed to several indispensable materials properties that are integrated in Te, namely, sufficient electrochemical activity, low thermal conductivity and low melting point, which can lead to adversarial EC and JH effects and therefore reversing the current-volatility relationship. The adversarial EC-JH relationship also led to unusual stimulus frequency dependent long-term memory (LTM) to short-term memory (STM) transition under electrical pulse train measurements in our devices, in contrast to the commonly observed STP-to-LTP transition in conventional EC-RS devices[99]. Our SNN simulations indicated that devices with this property could be generically utilized as spatial-temporal filters to improve the accuracy of the recognition of the event-based visual information with high noise levels. By modulating the EC-JH relationship using dielectric materials with different thermal conductivities, full functional-range tunable Te filament-based devices, from always-V RS, to V-to-NV transitionable RS, and to always-NV RS, were also demonstrated. This work demonstrated that the tellurium filament-based RS devices are promising enablers for functional cross-point arrays.

Experimental section

Device fabrication: Electrode/dielectric/electrode-structured cross-point devices with various junction areas (2×2 μm², 4×4 μm², 8×8 μm², 16×16 μm²) were fabricated on a thermally oxidized Si substrate. A 50 nm-thick Te bottom electrode was deposited via radio frequency (RF) sputtering with a 50 nm-thick Pt adhesion layer (via direct current (DC) sputtering) beneath it. Photo-lithographically patterned Sb₂Te₃/Te/Pt stacked films were then deposited, forming cross-point with the bottom electrode. The Sb₂Te₃ film was prepared by RF sputtering from a stoichiometric target. After Sb₂Te₃ deposition, a 50 nm-thick Te top electrode and a 50 nm-thick Pt protecting layer were prepared.

Two other dielectrics, Bi₂Te₃ and GeTe, were used for control devices. The Te/Bi₂Te₃/Te and Te/GeTe/Te devices were fabricated by the same process as that for the Te/Sb₂Te₃/Te devices.

Characterization: The cross-section TEM specimens were fabricated by focused ion beam (FIB), in Field Emission-Environment Scanning Electron Microscope (QUANTA 200 FEG), 2 kev Ga ion beam was used to cut the cell. The HRTEM images and EDS composition analysis were accomplished with a Field Emission Gun/TEM (JEM-2100F) operated under 200 kV voltage.

Electrical measurement: Cyclic quasi-DC voltage sweep measurements were performed by the Keysight B1500A semiconductor analysis system. The Keysight B1530A waveform generator/fast measurement unit was used to perform the pulse
measurements. Using a two-probe (W tips) configuration, DC and pulsed voltages were applied to one electrode with another electrode grounded.

**SNN simulation:** To parameterize the LSL, pulse trains each consisting of 10 pulses with identical amplitude (+0.6 V) and width (10 μs) but different intervals (0.1 μs, 1 μs, 2 μs, 3 μs, 4 μs, 5 μs, 6 μs, 7 μs, 8 μs, 9 μs and 10 μs) were applied to the Te/Sb₂Te₃/Te devices. The devices showed gradual switching behavior. We recorded the conductances of the devices before and after the pulse train stimulations. The differences of the conductances between the initial and final states were then normalized as values of weight changes (Δw), as seen in table 1. Finally, the Δw-interval relationship was extracted by exponential fitting. The fitted curve was:

\[
\Delta w = 0.205 \cdot e^{5.549 \cdot i} - 0.192
\]

where the interval was denoted as i.

**Table 1:** Δw versus pulse interval

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The leaky integrate-and-fire (LIF) model as the basic neuron unit was adopted and spatial-temporal backpropagation (STBP) algorithm\(^{[103,104]}\) for training was used. K3S1P1C128- K3S1P1C256- K3S1P1C256- FC128-FC10 (K: convolution kernel size, S: stride, P: padding, C: outputting channel, FC: full-connected dimension) network structure was used. Adam optimizer\(^{[105]}\) with an initial learning rate of 0.0005 and dropout technique were used. The dropping proportion was set to 0.25 for first layer and 0.4 for the other layers. The network was pretrained using 50000 training samples with a batch size of 100 for 50 epochs. 10000 noisy test samples were used in the test procedure. The time window was set to 8, the threshold to 0.4, and the decay factor to 0.5. Gradient substitution method with rectangular length of 0.5\(^{[103]}\) was used. All simulations were performed using PyTorch on one RTX 2080Ti GPU.

**Acknowledgements**

The authors acknowledge funding from National Natural Science Foundation (grant no. 61974082, 61704096 and 61836004), National Key R&D Program of China (2018YFE0200200), Youth Elite Scientist Sponsorship (YESS) Program of China Association for Science and Technology (CAST) (no. 2019QNRC001), Tsinghua-IDG/McGovern Brain-X program, Brain-Science Special Program of Beijing under grants Z181100001518006 and Z191100007519009, the Suzhou-Tsinghua innovation leading program 2016SZ0102, and CETC Haikang Group-Brain Inspired Computing Joint Research Center.
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