

Control of the Neuromorphic Learning Behavior Based on the Aggregation of Thiol-protected Ag/Ag₂S Core–Shell Nanoparticles

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Abstract

The neuromorphic learning switching behavior was investigated in devices which were constructed based on the aggregation of thiol-stabilized Ag/Ag₂S core–shell nanoparticles (NPs). The NPs were synthesized using the two-phase Brust–Schiffrin procedure, and exhibited Ag-S and Ag-S-R bonding states at the surfaces of Ag NPs. The memristive behavior of such device at room temperature under ambient pressure, which can be used to emulate the functions of biological synapses with long and short memories is achieved. The importance of the Ag₂S and the thiol layer at the surface of Ag NPs for the demonstration of learning behavior, such as the potentiation and depression of synapses in the human brain is explained.

Introduction

The performance of the von Neumann computer has improved considerably according to Moore's law that shows that the transistor density of a chip can be increased by downsizing. However, in recent year, the highest allowable number of transistors on a chip has remained constant and has thus failed to solve complex problems. Meanwhile, artificial intelligence (AI) is a intelligence demonstrates by machines that aims, such as artificial neural networks (ANNs) used for document analysis and image recognition.[1] However, the AI today still relies on the von Neumann computer that consumes tremendous electric power for its operation.[2] To overcome these problems, one of the proposed brain-like devices was used for the development of non-von-Neumann computing that is believed to a) be associated with low-power consumption for its operation, and b) possesses the ability to solve more complex problems.[3]

Recently, the memristor (memory resistor) has attracted increased attention because of its potential to replicate some of the functions of the brain, such as simple pattern recognition,[4] learning,[5] and other complex applications.[6,7,8] Various memristors synthesized using several materials have been studied and reported, such as metal oxides,[9,10] sulfides,[11, 12] and other organic molecules.[13] Only a few of them utilized nanomaterials and especially nanoparticles (NPs) that has led to large-scale and low-cost production. Recently, noble metal NPs, such as gold, have been successfully developed for Boolean logic[14] and neuromorphic switching behaviors.[15] However, the disadvantages associated with the use of gold NPs are primarily based on the fact that the NPs have no memory function. Furthermore, in the cases where they are used for switching functions, all the information stored in the device will be erased. By contrast, silver NPs have been studied for use in resistive switching devices.[16] Thus, it is expected that they can maintain information in such highly functional devices if aggregates of silver NPs are used for the switching operations in these devices, even after they are switched off. Herein, we report the study and use of Ag NPs for neuromorphic

switching devices. We first synthesize Ag NPs using the Brust–Schiffrin procedure, which resulted in a thin layer of sulfides and thiols to protect the Ag NPs,[17,18] as depicted in Figure 1(a). Subsequently, the thiol-protected Ag/Ag₂S NPs were placed at the center of the four-electrode device to create random networks among the particles, as shown in Figure 1(b), and brain mimicking properties were studied, such as the learning behavior of the device, based on the application of pulses, as shown Figure 1(c). We found that the states of the Ag₂S-like layers and thiols at the surface of Ag NPs play essential roles in the functionalization of NPs intended for use in neuromorphic devices owing to their ionic conducting materials and subnanometer insulating gaps. Additionally, the neuromorphic behavior was obtained by maintaining the pulses width and interval instead of bias polarity, which practically favorable and useful for future development of non von Neumann architecture.

Experimental procedures

The Ag/Ag₂S core–shell NPs were synthesized with the Brust–Schiffrin procedure according to previous work.[18] Three different samples with Ag/AM molar ratios equal to 0.25/1, 0.5/1, and 1/1 were synthesized based on the control of the amount of AM. The details of the synthetic procedure are described in the supporting information. The structural properties, such as particle size and chemical bonding states at the surface of as-synthesized NPs were then investigated with TEM (JEM 3010) and X-ray photoelectron spectroscopy (AXIS-Nova).

To investigate the nonlinear responses as well as the memristive behaviors of the I–V properties of the NP-based device, a parametric analysis was conducted with an HP 4156B semiconductor analyzer. The memristor is an electrical component that regulates the electrical current and remembers the amount of charge flow through the circuit. It was first proposed Chua,[19] and then was first developed by the Hewlett-Packard (HP) laboratory using a TiO₂

thin film.[20] Herein, a four-electrode device system was used to investigate the nonlinear response of the output current, memristive switching, as well as the learning behavior of the aggregated of Ag/Ag₂S NPs. Electrodes for the device were fabricated with a 200 nm gap using e-beam lithography with a 6 nm thick titanium (Ti) layer followed by a 24 nm thick platinum (Pt) layer.

Given that the memristive behavior is one of the requirements for the development of neuromorphic devices, the learning behavior of the device was then investigated based on the application of the periodic pulse voltage and the maintenance of the frequency, amplitude, duty cycle, and offset of the function generator (Agilent 33120A). The output current was recorded by LabVIEW with a computer which was connected to a data acquisition module (National Instruments, NI USB-9162) and a preamplifier (DLPCA-200) with a 10 k Ω resistor. The control of the formation and annihilation of metallic filaments between NPs played an essential role in the development of the memristive neuromorphic devices.[21,22]

Results and Discussion

Structural properties

The shape and average diameters of the as-synthesized NPs were investigated with transmission electron microscopy (TEM). As shown in Figure 2(a)–(c), NPs have spherical shapes for the Ag/Allylmercaptan (AM) molar ratios of 0.25/1 to 1/1 for various diameters. To obtain the average diameter of the NPs, Gaussian curves were applied to fit the histogram of the particle size distribution, which is depicted in the inset of Figure 2(d). The average diameters obtained from the analysis of the TEM images were 22.7, 35.6, and 45.3 nm for Ag/AM molar ratios of 0.25/1, 0.5/1 and 1/1, respectively, and were relatively smaller compared to polymer-coated Ag NPs.[16] The size of NPs was in close agreement with

extracted data from the X-ray diffraction (XRD) pattern using Sherrer's formula presented in Table S1 (see Supporting Information). The average diameters of NPs were increased as the molar ratio of Ag/AM increased, in agreement with a similar tendency reported previously.[18]

Further investigations of the NP structure were carried out by performing X-ray photoelectron spectroscopy (XPS) measurements to investigate the chemical bonding state at the surfaces of the NPs. The S2p and Ag3d experimental spectra were then analyzed with Lorentzian shaped peaks. As shown in Fig. 3(a), three distinct chemical states for sulfur atoms were obtained around S2p spectra at the binding energies (BE) of 161, 162, and 163 eV, which are respectively associated with the sulfur atom in the Ag₂S-like chemical state (green line),[23,24] bridging of the AM-residue R (R = -CH₂-CH=CH₂) to the Ag ions (Ag-S-R) (blue line),[25,26] and the sulfur atoms of physisorbed AM molecules (orange line). The fraction of S atoms belonging to the third S type was below 20% in all the tested samples (0.25/1 to 1/1). The fraction of S in the Ag₂S-like phase as well as Ag-S-R increased following increases in the Ag/AM molar ratio from ~20% to approximately 40%. Moreover, four peaks were observed after fitting with a separation of 6.0 eV of in the Ag3d XPS spectra, as depicted in Figure 3(b). The first two peaks are located at 368.20 and 368.9 eV, which correspond to Ag3d_{5/2} Ag(0), Ag₂S,[27] and Ag3d_{5/2} Ag-S-R, similar to the Ag bonds within organic structures.[28] For Ag(I) ions in Ag₂S inorganic compound, an Ag3d_{5/2} BE value of 368.00 eV is expected.[26] The small amount detected for the sulfur associated with Ag-S-R (~10%) is consistent with the Ag located at the NP surface and slightly decreases with the increase of the Ag/AM ratio, thus suggesting a smaller surface-to-volume ratio, as expected in the case of larger particles. The existence of Ag-S bonding in all tested samples with gradual increase of NPs as increase of Ag/AM molar ratio indicating the dominance of Ag structure with thin layer of Ag₂S at the outer shell of NPs. The thickness of Ag₂S layer was estimated by calculating the

intensity ratio of Ag and Ag₂S peaks in XRD pattern. The smaller the average size of particle, the higher volume ratios of Ag₂S.

Electrical properties

The NPs with an Ag/AM molar ratio of 0.25/1, which had the smallest average diameters and highest volume ratios among the tested Ag₂S samples were then drop-casted on the electrodes using heating, which was provided by a hot plate at 50°C to evaporate the solvent. Similar to other filament-based switches, such as metal-oxide thin films[29] and Ag NPs,[16] a forming step was usually required to activate the NP assemblies and memristors. To perform the activation, repeated sweeping of forward and reversed bias only in the positive region were applied to the device, as shown in Figure 4(a). A nonlinear response and a gradual increase of the output current with a history of successive sweeps were obtained. At the initial sweep, some fluctuation of current was observed in the range of 0–4.5 V, thus indicating that the forming process randomly occurred at various gap points among the NPs. Subsequently, the current started to increase and reached 10⁻⁵ A with a dropping resistance magnitude in the range of 3 to 100 kΩ which allowed the formation of some of the metallic bridges among the NPs. During the reverse bias of the first sweep, the output current exhibited hysteresis with the forwarding current. At the second sweep, the output current started to increase at an applied bias voltage of approximately 1 V, which is lower than that of the first sweep. This followed the path of the reverse sweep after the first sweep, and is indicative of typical memory of the device. Additional bias sweeps resulted in the gradual increase of the output current up to 10⁻⁴ A and to a resistance of 10 kΩ at 5 V with hysteresis, which justified the activation of the device. The activated network of NPs exhibited robust, strong memristive behavior, manifested by hard switching, as shown in Figure 4(b).

The formation and annihilation of metallic bridging between the particles resulted from adequate electric fields applied across the NPs. Based on these fields, the number of tunneling electrons were injected into the device and led to the reduction and oxidation (redox) process at the surface of the Ag₂S layer of the NPs.[20] In this NP-based device, however, gaps were created between the NPs in justification of the existence of the Ag₂S layer and thiols that led to the multigap system. Once the bias voltage was applied to the system, Ag NPs were attached to the counter electrode and started to oxidize it. Ag⁺ ions migrated through the Ag₂S layer at the outer shell of the NPs in all directions. Additional energy was applied and led to the breakage of organic layer. Accordingly, Ag⁺ ions were reduced by the tunnelling electrons injected from one of the electrodes. The entire redox process as well as filament formation among the NPs are indicated by the stepwise increase of the conductance following the application of a constant direct current (DC) voltage, as shown in Figure 4(c). The redox reaction led to the migration of Ag from the surface of Ag₂S and to the formation of a metallic filament between the NPs. This explains the hysteretic I–V characteristic of the memristor.

Another essential characteristic of the biological brain is neuroplasticity,[30] manifested (among others) by memorization and learning processes, whereby the organization of the brain changed as a result of experience or training. The memorization process in the human brain starts from the instant external input signals are received. Signals are then transmitted through axons via the interconnecting synapses, and the synaptic potential thus increases. If the frequency of the firing increases, this will lead to a long-term memorization state. This process is manifested by increased synaptic responses, which are followed by the potentiating pulse of electrical stimuli, commonly referred to as long-term potentiation (LTP).[31] The pulse bias was applied to the device to perform such measurements. The desired pulse input, including the pulse ratio (pulse width (t_w)/pulse interval (t_i)) and the pulse

amplitude were controlled by the frequency, amplitude, duty cycle, and offset of the function generator.

At first, amplitudes of 1 V with respective a pulse width and an interval of 8 s and 2 s (pulse ratio of four), were set and applied to the device. The output current of the device with thiol-protected Ag NPs exhibited a gradual increase up to 80 s. At that time, the current decreased slightly and its value was maintained over the next 10 s period. As depicted in Figure 5a, the minor current drop occurred several times, while the general tendency was increasing. This increasing tendency was attributed to the breakage of the metallic filaments owing to Joule heating at some of the connection points among particles. As additional pulses were applied, the output current tended to increase gradually until a maximum current intensity of 1.0 μA was reached. This suggests that the metallic filaments which connected the particles were rearranged after the breakage. Herein, the maximum conductance was lower compared to the single atomic contact of 77.5 μS [32] owing to the thiols at the outer shell that increased the contact resistance between particles. The process of metallic bridging formation, including the rearrangement of the filament based on the application of the input pulses, emulated the strengthening of synapse in the biological brain.

Moreover, the annihilation of metallic bridging that indicated the depression of synapses was also obtained by the maintenance of the input pulse based on the widening of the pulse interval to 1 s and the narrowing of the pulse width to 0.25 s (pulse ratio of 0.25) at a constant amplitude of 1 V, as shown in Figure 5(b). At the end of the first five pulses, the conductance was still at its maximum value, and indicated the existence of metallic filaments between the particles. Subsequently, as more pulses were applied, the current decreased gradually to approximately 0.1 nA at 25 s. The decrease of the conductance indicates the annihilation of the metallic bridging that mimics the depression of the synaptic strength.

The key mechanism behind potentiation and depression of the NP-based neuromorphic device lies on maintaining the interfacial energy between Ag nanoparticles and surrounding materials. Applying bias voltage into the system facilitate redox reaction and increase the interfacial energy between Ag and Ag₂S so that the conduction channel formation process is induced, as demonstrated before by applying relatively high DC constant bias to the device. On the contrary, removing the bias causing the spontaneous relaxation dynamics that owing to minimizing the interfacial energy between Ag and Ag₂S. Although the spontaneous relaxation of Ag nanocluster in oxide material is about a few tens of milisecond,[33] different material involved such as Ag₂S and organics may affect the relaxation time of Ag nanoclusters. Here, during the depression process, shorter time of electrons injected to the device cannot maintain the conduction channel formation, leading to rupturing owing to dominant of relaxation time. The longer the bias is applied/removed, the more active/relax of Ag nanocluster. Thus, combining constant high voltage and low voltage with controlling the time of electron injection as well as interval may lead to the competition between formation and annihilation of conduction channel between NPs. Previously, it was reported that the potentiation of Ag-Ag₂S based-neuromorphic device was controlled by maintaining the polarity of pulses,[11] which is inconvenient for practical in industry. Here, by utilizing the aggregation of Ag-Ag₂S nanoparticles, the state of interfacial energy was successfully controlled by maintaining the ratio of pulse width and interval of input bias without changing the polarity. This was justified by the fact that the potentiation occurred following the application of a pulse with a width which was four times longer than the pulse interval to the device. Accordingly, the bridging formation was dominant compared to the relaxation, and led to the gradual increase of the number of metallic connections between NPs, and to the increase of the conductance. These findings justify the potentiation effect. By contrast, the shorter pulse width with the longer

interval facilitated the dominance of the relaxation time and caused a gradual drop of conductance that mimicked the depression of synapses in the brain.

Using a simple two-phase Brust-Schiffrin synthesis procedure, we have created functional neuromorphic devices based on aggregation of Ag/Ag₂S NPs system. This is the first report that demonstrates control of the potentiation and depression of synaptic memory using the aggregation of the NP device. Different polarity of input voltage was used in single solid-state atomic switch as reported previously,[11] however such neuromorphic behavior in our device is obtained by maintaining the pulse width and interval, which is favorable for practical application in real device. However, one should note that compared to the polymer-coated Ag NPs[16] that exhibited typical resistive switching, the thiol-coated Ag/Ag₂S NPs-based device exhibited synaptic-like behavior that was indicative of the essential role of the thiol and Ag₂S thin layer at the surface of Ag NPs as a mixed ionics and electronics conducting materials [21]. The successful demonstration of synaptic-like behavior with the utilization of the aggregation of thiol-protected Ag/Ag₂S NPs, provides a good opportunity to overcome the limitation of Moore's law.

Conclusions

We successfully synthesized thiol protected Ag/Ag₂S NPs with various diameters based on the control of the molar ratio of Ag/AM. The diameter of the particle increased as the molar ratio increased. The I–V characteristics of the NPs exhibited nonlinearity and memristive behavior with an On/Off ratio of approximately 10⁴. The input pulse was then applied to the system to investigate their learning behavior. By maintaining the pulse width and interval at the same pulse amplitude, the formation and annihilation of metallic bridging between particles, which emulated the potentiation and depression of synapses in the brain, was successfully controlled. This successfully demonstrated the potentiation and depression effects following the utilization of the aggregation of NPs owing to the Ag₂S and thiol layer at the outer shell of Ag NP.

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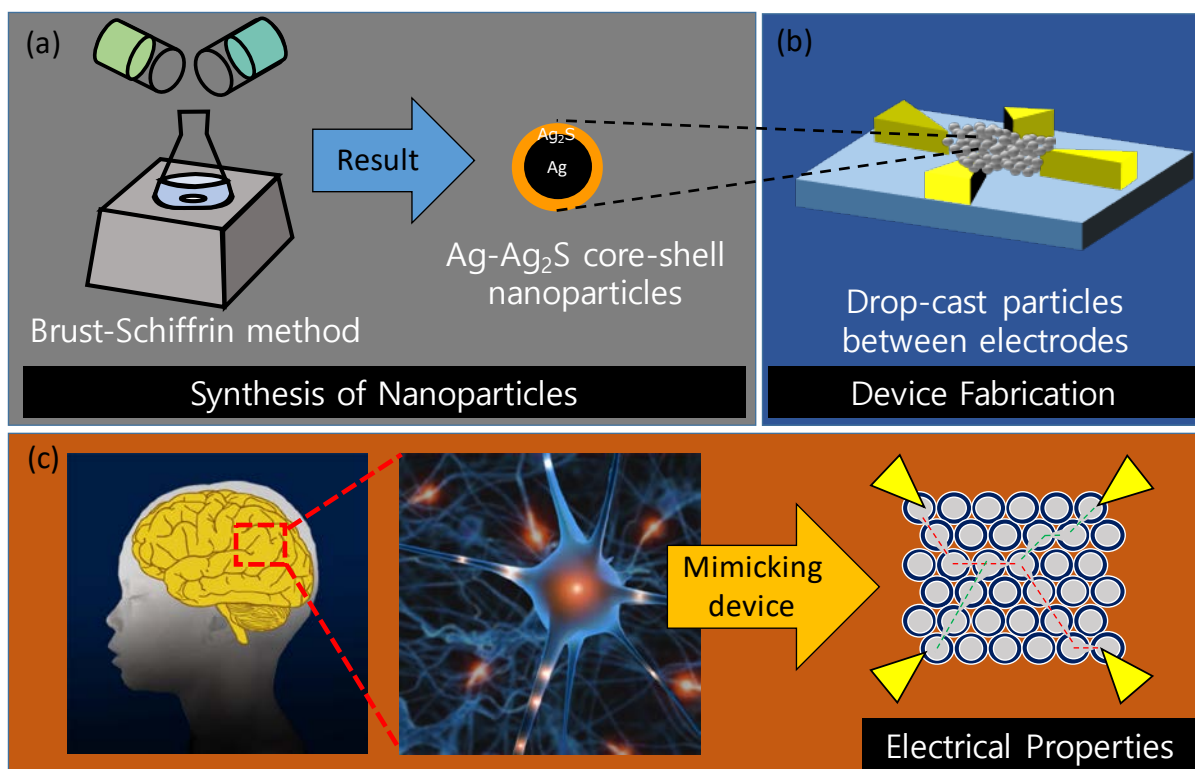


Figure 1. Summary of procedure adopted in this study. (a) To implement a brain-like device, thiol-protected Ag/Ag₂S NPs were synthesized using the Brust–Schiffrin procedure. (b) NPs were then placed at the centers of the four-electrode system devices and (c) investigated the electrical and brain-mimicking properties, such as the memristive and learning behaviors.

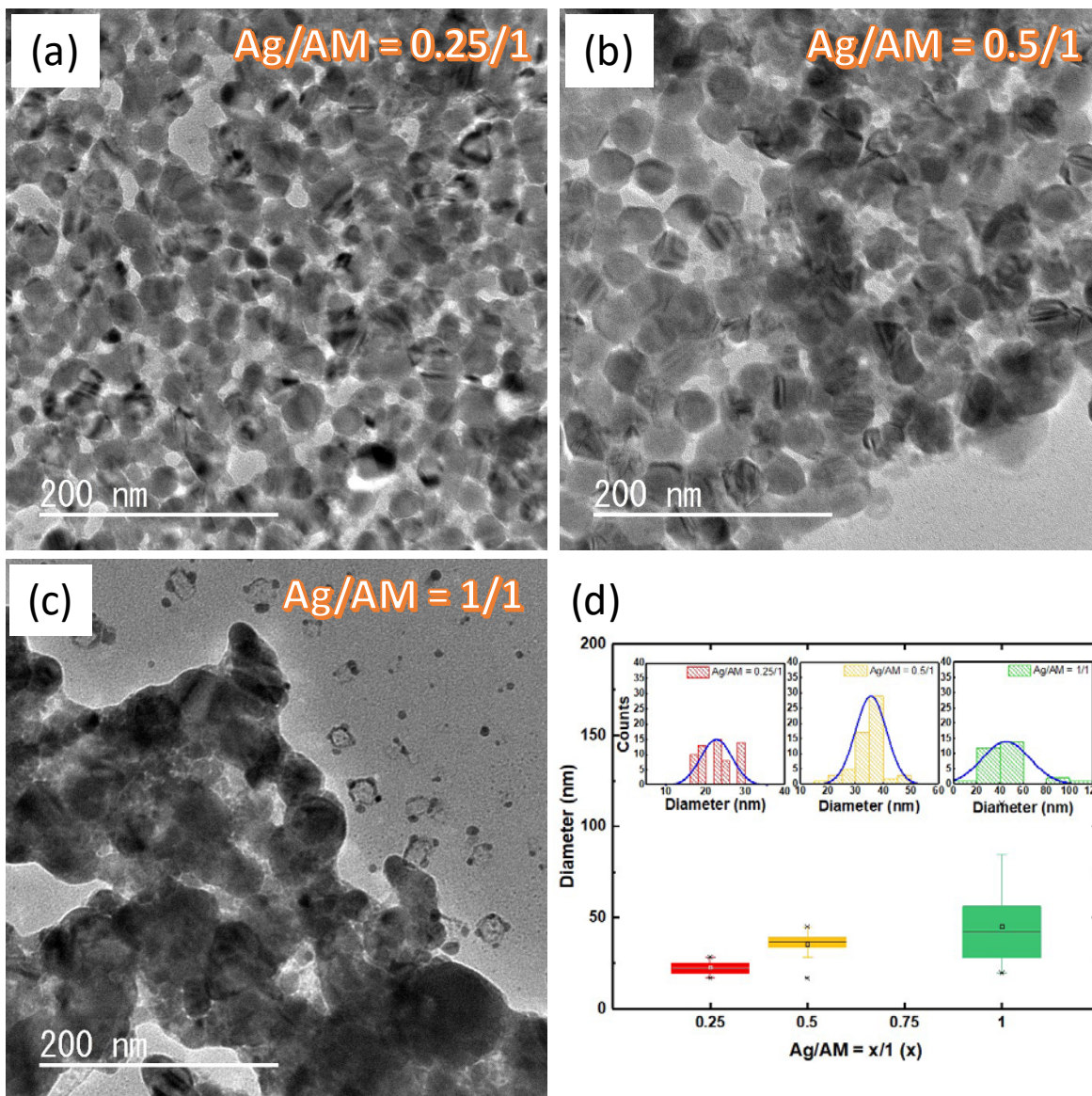


Figure 2. (a)-(c) Transverse electron microscopy (TEM) images of NPs with the Ag/Allylmercaptan (AM) molar ratios of 0.25/1, 0.5/1, and 1/1. (d) The distribution of the diameters of particles extracted from TEM images for Ag/AM molar ratios of 0.25/1, 0.5/1, and 1/1. Inset: average diameters of tested samples were calculated by applying normal distribution. The average diameter increases as the molar ratio increases.

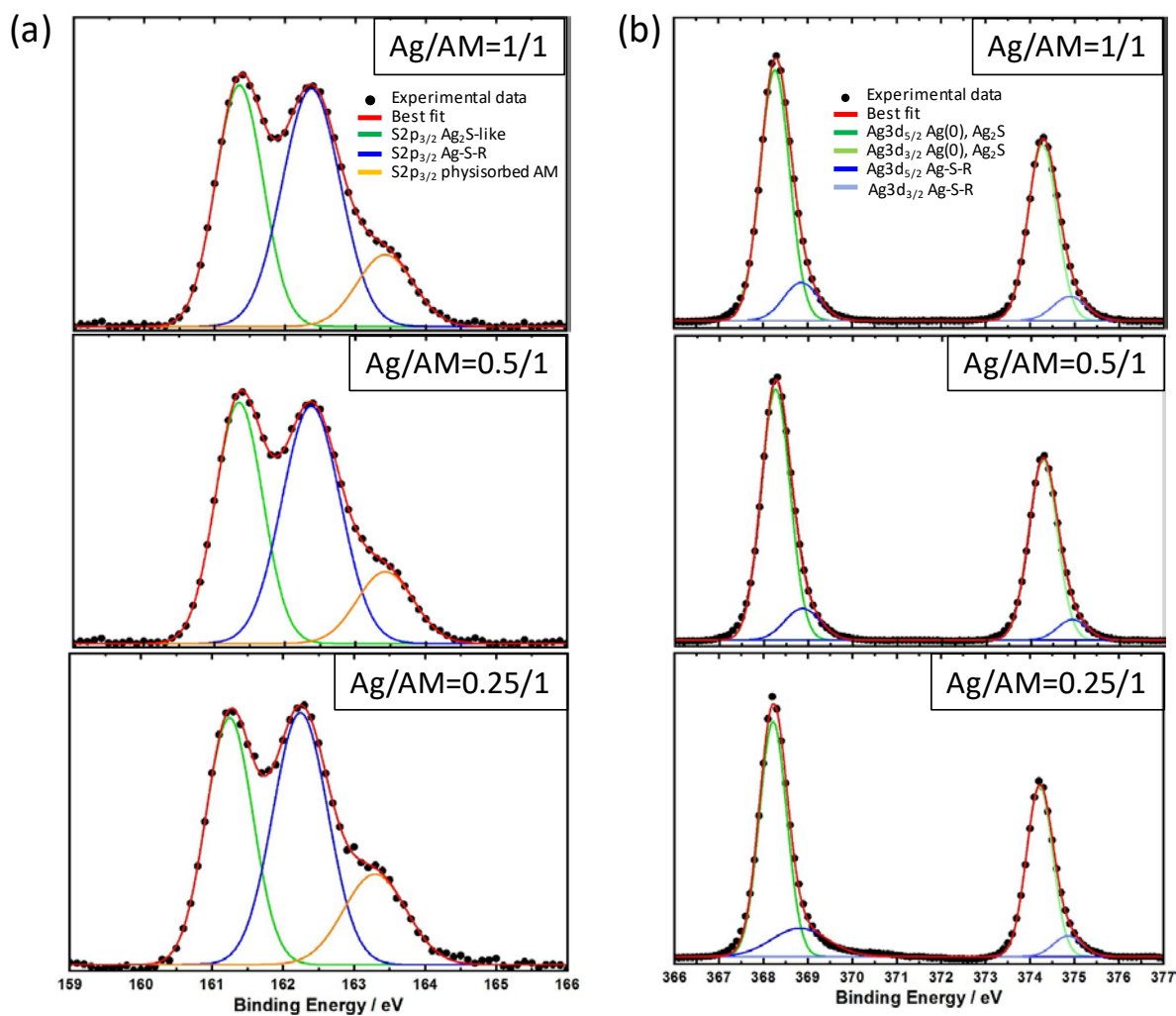


Figure 3. (a) S2p X-ray photoelectron spectroscopy (XPS) data of all tested samples. After the fitting process, three distinct peaks are obtained at 161, 162, and 163 eV, which are respectively attributed to S2p_{3/2} Ag₂S-like, S2p_{3/2} Ag-S-R, and S2p_{3/2} physisorbed AM. (b) Ag3d XPS data of all the tested samples showed four peaks that fitted well to the experimental data. The peaks located at 368.20 and 368.9 eV are attributed to Ag3d_{5/2} Ag(0), Ag₂S, and Ag3d_{5/2} Ag-S-R, similar to the Ag bonds within organic structures. The other two peaks, which are separated by 6.0 eV are respectively attributed to another spin-orbit pair, namely to Ag3d_{3/2} Ag(0), Ag₂S, and Ag3d_{3/2} Ag-S-R.

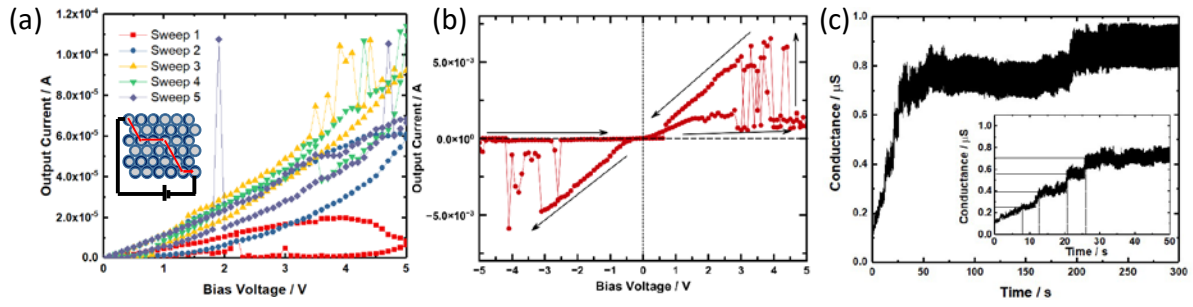


Figure 4. (a) Gradual increase of output current after multiple sweeps of input voltage that is indicative of the activation of metallic bridging formation. (b) Hysteresis is observed in the I–V characteristics of the NP-based devices indicative of the memristive behavior as the most essential property of the neuromorphic device. (c) Time traces of conductance as a result of the application of a constant direct current (DC) voltage of 1 V to the system. Inset: stepwise conductance increases in the first 50 s, thus indicating the metallic filament formation process responsible for the linking among the NPs. The fluctuation of the conductance observed at a certain period indicates the rearrangements of the filaments.

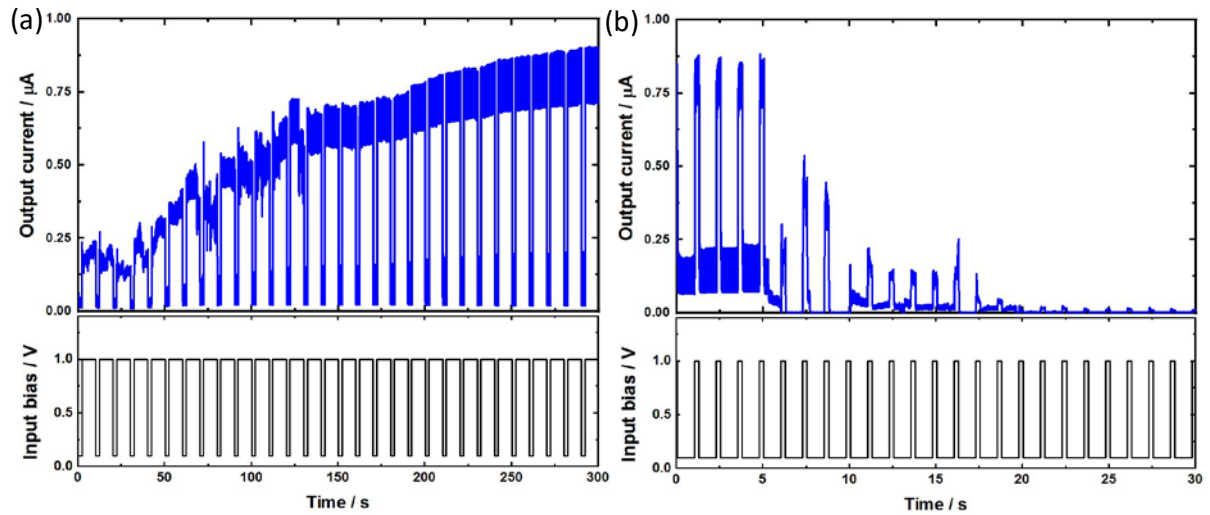


Figure 5. (a) Periodic input pulses with a pulse ratio of four were applied to the device. Gradual increases of the output current are evoked until a conductance value of 0.8 mS is attained. This conductance value indicates the occurrence of the potentiation of synaptic activity in the human brain. (b) The emulation of the depression of synaptic activity was also achieved following the application of periodic input pulses with a pulse ratio of 0.25.