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Two-terminal Resistive-switching Memories based on Liquid $AgNO_3$ as Artificial Synapses

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Abstract

The brain presents impressive characteristics, like memory retention and learning capabilities at low energy consumption, making it the most efficient computational entity known. A promising candidate for the task of mimicking the brain is a new generation of devices showing resistive switching, a process that resembles the synaptic behavior, namely the liquid-based resistive-switching memory (LRSM). Here we present a two-terminal structure that works based on a solution of silver nitrate ($AgNO_3$) and a silver (Ag) electrode. Furthermore, since the goal is to mimic the behavior of synapses, the dynamical properties of resistive switching, endurance and data retention were studied. Low operation power was further achieved (<0.5 V, 0.1 mA). Finally, taking advantage of the liquid medium, in which the synaptic behavior occurs, a flexible device was built using microchannels in Polydimethylsiloxane (PDMS) that allowed for the harvesting of the silver nitrate and the insertion of a silver and copper electrode.

Author Keywords. Resistive Switching, Neuromorphic Computing, Artificial Synapse, Liquid-based Memory.

Type: Research Article

1. Introduction

The current computer technology is moving to the end of Moore's law, which means that, for the average computer, the number of transistors used in microchips capable of processing large data efficiently without reasonable high energy consumption, cost and heat dissipation is reaching its limit. Due to this paradigm, the scientific community turned their attention to other possibilities such as quantum and neuromorphic computing, being that the latter focuses on the most efficient computational entity known, the human brain, and tries to mimic it in a microchip (Wan et al. 2019).

The brain is an impressive human organ, with remarkable characteristics such as memory retention and learning capabilities, with low power consumption (Park, Park, and Lee 2020). Behind these characteristics is a complex network of neurons and synapses, being that the first process information and the second are the vehicles to transmit that information from a pre- to a post-synaptic neuron, so that memory is stored in the synapses in the form of synaptic strength (Park, Kim, and Lee 2020).

A promising candidate for the task of mimicking the human brain is a new generation of devices called resistive random-access memory (RRAM), that are based on a process that resembles the synaptic behavior, the resistive switching, defined by the transition between two or more states of resistivity (Dias et al. 2017). Thorough research has already been performed proving that this can be achieved in both solid and liquid materials (Khan, Hassan, and Bae 2020).

In particular, RRAM devices based on a liquid electrolyte rely on the formation and destruction of a metallic filament connecting the two electrodes across the solution. Liquid as a medium for the artificial synapses is promising due to its high ion mobility. Liquid devices can be classified as either cationic or anionic. The device used in this work is cationic, which means that the formation of the filament is due to the movement and migration of cations (Ag^+).

These cations come from electrochemically active materials, like silver, which are often used as electrodes, part of the active layer of the device or both (Upadhyay et al. 2019).

When no bias is applied to the device, the solution presents disperse silver and nitrate ions. By applying a positive bias, the bottom silver electrode begins to oxidize providing an extra source of silver ions to the solution and the ones in the solution begin to suffer reduction, forming silver atoms. When these atoms form a filament that connects bottom to top electrode, the device is in the On state, a state with low resistivity (see Figure 1a)). When the bias is reversed, the extra source of ions provided by the bottom electrode is ceased and the filament is ruptured leaving the device in the Off state, a state with high resistivity. Thus, by controlling the charge that goes through the device, its conductance can be modified (Jo et al. 2010). Furthermore, since the medium where the artificial synapse occurs is liquid, this device may be fabricated in many shapes, that are able to adapt to the desired application (Kim and Lee 2019).

2. Materials and Methods

Here, a two-terminal device composed of two gold coated POGO tips (for electrical measurements), a thin film composed of a silicon substrate with a silver thin film (100 nm) deposited on top of it, and a silver nitrate solution was implemented. One of the gold tips was directly connected to the bottom silver electrode, while the other was placed in contact with a drop of silver nitrate solution, stacked on top of the Ag layer (Figure 1b)).

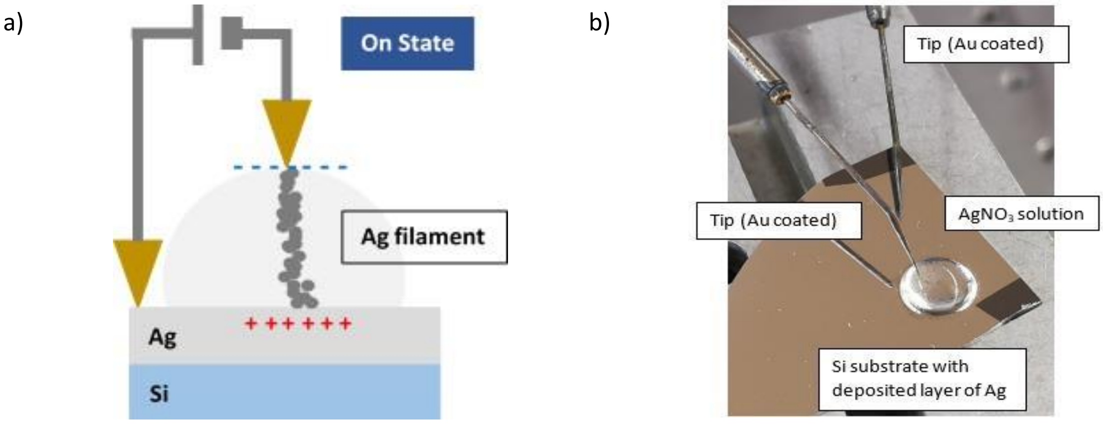

Figure 1: a) Scheme of operation of the device and b) experimental setup showing the silver bottom electrode in a silicon substrate, the $AgNO_3$ aqueous solution and the gold top electrode in a microtip

Three types of concentrations of silver nitrate were studied, namely 0.05 mol, 0.1 mol and 0.5 mol to investigate the most desirable characteristics, such as highest On/Off resistivity ratio, fastest switching and smaller operation voltage. For each of the concentrations, three kinds of measurements were made: voltage sweeping, endurance, and data retention. For the voltage sweeping measurements, the height of the top tip emerged in the liquid was varied in three positions (top, intermediate, bottom) to evaluate the effect of filament size in the operation parameters. The other measurements were performed in the "intermediate" position.

To perform measurements of voltage sweeping, endurance and data retention, bias was applied that changed the state of the device between On and Off. Voltages ranging from -300 mV to 0 V and from 0 V to 500 mV were applied, with steps of 10 mV. For the endurance measurements close to 100 consecutive cycles of voltage sweeping were performed and in the data retention measurements close to 15 min of reading the behavior of the solution when in a fixed state (On and Off) was performed, using a small reading voltage of 0.01 V.

A flexible device was also fabricated, with the help of a Polydimethylsiloxane (PDMS) mold. A microchannel of 850 µm in diameter was built inside a PDMS cylinder with 3 cm height and a diameter of 0.5 cm. The microchannel was used to insert the liquid silver nitrate (0.5 mol $AgNO_3$ concentration) and two electrodes, one made of silver (ground), and another made of copper, that were connected to a power source. A picture of the PDMS prototype with the visible microchannel is presented in Figure 2. The voltage cycles applied ranged from -500 mV to +500 mV, with a current compliance of 1 mA.

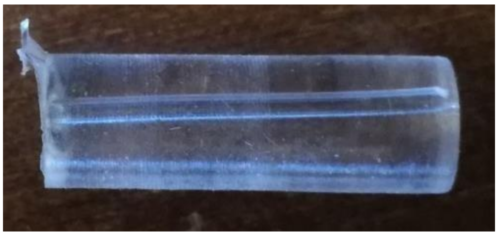

Figure 2: Flexible transparent PDMS device showing the interior microchannel

3. Discussion

In the present work low operation power was achieved (<0.5 V, 0.1 mA) with resistive switching being observed under electrical stimulation at different electrode spacing (top electrode depth), as shown in Figure 3. The On state (set process) was achieved between the voltages of 0 to -300 mV. The Off state (reset process) was reached for voltages of 0 to 500 mV. These low operation voltages are on the same order of magnitude of the ones reported by Kim et al. in a similar system (Kim and Lee 2019), which are both significantly lower than the ones reported by Khan, Hassan, and Bae (2020) on another liquid-based device, ranging between -1.5 and 1.5 V. The resistive switching is bipolar with set at negative voltage and reset at positive, when the silver electrode is grounded. This behavior may be explained by the diffusion of silver atoms inside the solution when an electric field is being applied (Rajan et al. 2016). The bottom silver electrode represents the pre-synaptic neuron, and the top gold coated tip (top electrode) represents the post-synaptic neuron. The artificial synapse is then reproduced in the liquid medium, the silver nitrate.

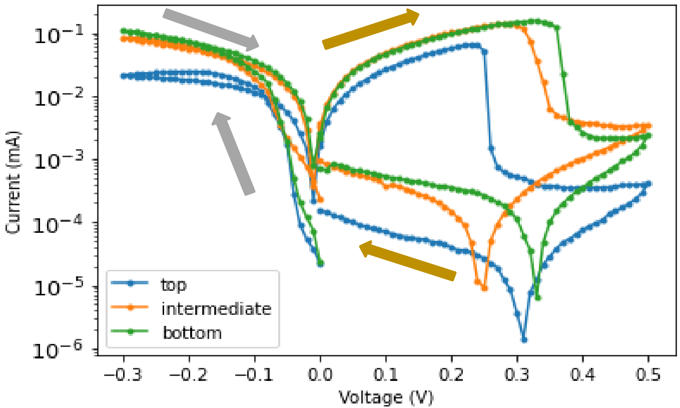

Figure 3: Resistive switching behavior in a solution of silver nitrate at 0.05 mol. The label "top", "intermediate", and "bottom" refer to the depth of the top electrode in the $AgNO_3$ solution, being that "top" is the least submerged and "bottom" the most profound. The arrows show the loop direction for set (grey) and reset (golden) processes

We observed that better performance was achieved with lower concentrations of the silver nitrate solution, due to increasingly rapid overheating with larger concentration. It was also noted that, for the three configurations of the tip emerged in the liquid, the more emerged the tip was, the larger the achieved current operation. This may be explained by a smaller filament forming in a thinner gap, as it would be expected according to the existing literature (Dias et al. 2017; Kim and Lee 2019; Khan, Hassan, and Bae 2020; Rajan et al. 2016).

Furthermore, data retention and endurance were tested. It was observed that both are achieved with low current drop as the time or number of cycles in the device increased, as shown in Figure 4a) and Figure 4b). However, a drop is still evident, being that, for data retention, thisis probably due to the fact that the device loses capacity of withholding the information for larger periods of time and, for endurance, this is most likely due to the formation of colloidalsilver in each resistive switching cycle that would not return to the bottom electrode, remaining in the solution and interfering with filament formation, maybe even interrupting it.This is further proven by the fact that the solution was clear in the beginning of the experiment but had turned grey by the end. These results are in accordance with what would be expectedfrom the material properties and previous works (Upadhyay et al. 2019). Nevertheless, we alreadyobtained an endurance of up to 70 cycles, which is comparable with the order of magnitude of 10^2 previously reported in other works (Kim and Lee 2019; Khan, Hassan, and Bae 2020). Since the goal is to mimic synapses, some dynamical properties like potentiation and depression, properties that define the device in terms of its memory capabilities will be also studied.

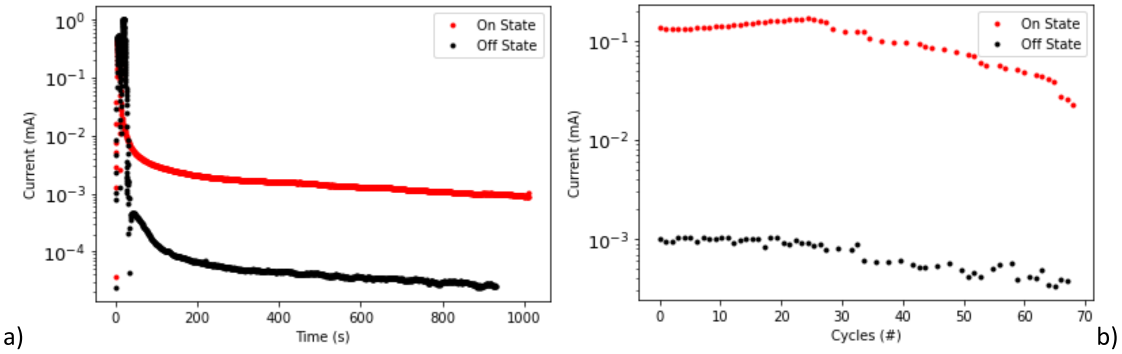

Figure 4: a) Data retention and b) endurance in a solution of silver nitrate at 0.05 mol, measured at 10 mV

To prevent the evaporation of the silver nitrate solution under cycling due to overheating, especially when it comes to higher concentrations of silver nitrate, a flexible device was built that allows for the liquid to be encapsulated inside of it.

Regarding the flexible device, resistive switching was observed in an operation range of -500 mV to 500 mV, with the same mechanism as the previous device, metallic filament formation (Figure 5). Further measurements in the device need to be made to see if it presents other synaptic characteristic and to see if it retains its characteristics with an applied deformation, such as bending.

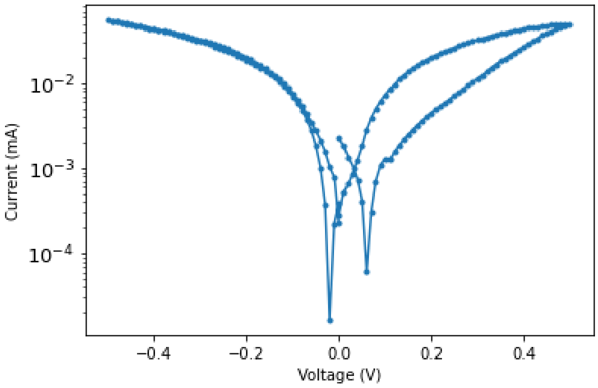

Figure 5: Resistive switching behavior in a flexible device with liquid $AgNO_3$ at 0.5 mol concentration

4. Conclusions

In conclusion, a device was built with resemblance of the synaptic behavior described by the nervous system. This device has great promising qualities such as low power operation, bipolar resistive switching behavior, endurance, and retention. By taking advantage of the useof a liquid medium for the artificial synapses, a flexible device made from PDMS was also built,that can sustain the liquid inside and presents resistive switching, showing promising capabilities to be used as a device for soft electronics.

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