Frequency Driven Organic Memristive Devices for Neuromorphic Short Term and Long term Plasticity.

S. Battistoni[1], V. Erokhin[1] and S. Iannotta[1]*

[1] CNR- IMEM, Institute of Materials for Electronics and Magnetism, Parco area delle Scienze 37/A, Parma, 43124 Italy

E-mail: silvia.battistoni@imem.cnr.it

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Abstract

The concomitant and concurrent presence of cellular mechanisms, such as the activity-dependent Long Term Potentiation (LTP) and Depression (LTD), and the short-term plasticity (STP) is believed to be at the basis of encoding memories in brains. Thus, the best possible emulation of these fundamental brain activities is essential for developing bioinspired circuits and novel adaptive technologies, and, looking in perspective, an efficient brain-machine interface. In this framework, memristive devices are increasingly considered as key elements in the emerging fields of neuromorphic engineering and computing because of their synaptic-like plasticity properties. Here we demonstrate the application of organic memristive devices (OMDs), based on polyaniline (PANI), mimicking controlled neuromorphic functions. In particular our OMDs exhibit LTD or LTP by varying the polarity or, more interestingly, by varying the frequency of the incoming stimuli, according to the typical biological patterns. In both cases, OMDs show also an effect analogous to the transition between short term to long term memory, as a function of the total number of received pulses. We validate that organic memristors represent an important step toward an "intelligent" neuroprosthetics demonstrating through the hardware implementation of OMDs neuromorphic functionalities, the possibility of pairing the frequency dependence of synaptic signals with the non-volatile evolution of the internal memory states of the device.

1. Introduction

Interfacing nervous systems with electronic elements for the realization of neuroprosthetic devices and/or brain machine interfaces (BMIs) is still a major challenge since the direct coupling of live neurons with (or through) functional electronics devices has to pass several bottlenecks. One of this is the need of devices able to "speak the same language" and to establish a bi-directionally communication with biological neurons for enabling a "near-physiological" tuning of neuronal activities within an "intelligent" adaptive closed-loop [1]. Adaptation processes increasing neuron synaptic efficiency in response to stimuli from surrounding cells, are the expression of the fundamental biological property called synaptic plasticity [2-5]. An efficient emulation of this feature has been a critical task for the neuromorphic computing community since, in standard Von Neumann architectures, both computing complexity and energy dissipation increase rapidly with the system sophistication, progressively compromising the computing speed [6]. Biological architectures are much more efficient in handling adaptation processes in complex systems because of both their high parallel processing power due to the large network connectivity [7] and their ability of tuning synaptic strength in response to external stimuli and the whole history of their involvement into the formation of the signal transfer chains.

Memristive devices are among the best systems mimicking synapse activity since, exactly as their biological counterpart, they change their memconductance (weight function) in response to a sequence of received stimuli [6, 8-10].

PANI based OMDs (represented in Figure1) are characterized by a nonlinear response of the current showing a gradual transition between the low and high resistivity states due to electrochemical reactions [11-16].

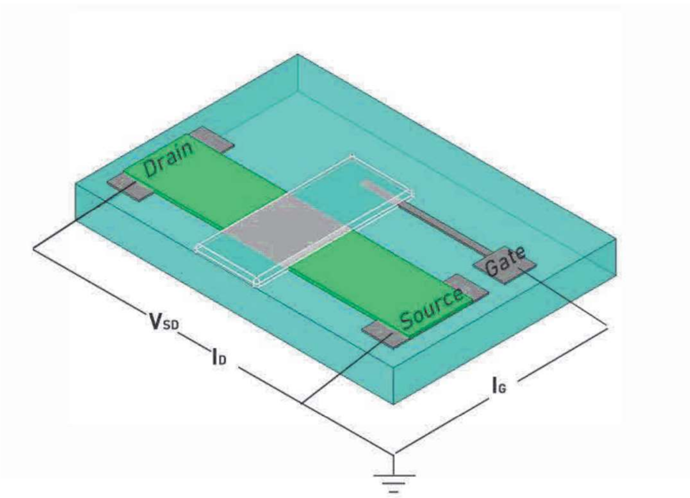

Figure 1: Scheme of the organic memristive device (OMD) and characterization circuit: in green the polyaniline conductive channel deposited on two chromium electrodes identified as Source and Drain. The third electrode, Gate, is a silver wire directly in contact with the polyelectrolyte, represented in light grey. Gate electrode is always grounded and the current that flow in this circuit is referred as Gate current (I_G); to the circuit formed between source and drain is applied a voltage potential, producing the current (Is_D).

The temporal evolution of the memristor output current, shown in Figure 2, is driven by the concurrent presence of a proper voltage bias and an appropriate application time, thus, by varying one of these two parameters, the conductivity of the device can be precisely tuned in multiple resistive states.

This intrinsic feature of memristors makes them ideally suitable for neuromorphic applications [14, 17, 18] and for implementing models of human memorization [3, 19], that has been demonstrated in organic [20] and inorganic devices [3, 21, 22] varying intensity or duration of the training routine. Potentiation/depression synaptic operations have been demonstrated in inorganic devices [23-26] inducing the increase/decrease of the conductivity through the application of a positive/negative bias to the device, respectively. However, even if this approach is rather common and well established in the memristive community (inorganic in particular), biological systems are governed by the common effect of the frequency and the number of received stimuli rather than by the different voltage amplitudes[2, 27-29]. In a recent paper, Wang et al [10] propose a memristor for implementing this functionality with “volatile diffusive properties” that, after the activation in the ON state, naturally relaxes back to the OFF state when the bias is removed, excluding the possibility of operation on stable conductivity levels.

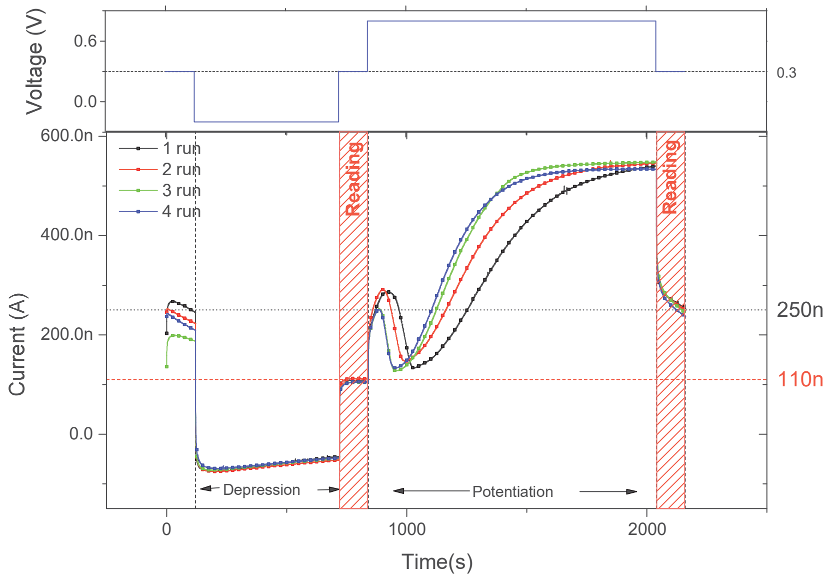

Figure 2: Temporal evolution of the output current of the organic memristor in function of the applied potential: top panel) voltage profile applied between source and drain during the measurement (V_{SD}); bottom panel) memristive output current (I_D). Reading phases are characterized by the application of +0.3V, depression by -0.2V and potentiation by +0.8V. The induced reactions (oxidation and reduction respectively) lead to a relevant difference (about a factor 2.3) in device conductivity that changes from about 110nA to 250nA and remains constant for all the different cycles of measurements.

In our approach, since the properties of the OMD conductive channel were precisely and gradually tuned, we use them for a better implementation of common neuromorphic functions in two distinct operational modes. We achieve long term functions by varying the polarity of the incoming stimuli (voltage dependence) or by tuning the frequency of a fixed stimulus (frequency dependence). Moreover, in both cases the number of the stimuli determines the occurring of the STP-to-LTP transition.

2. Results and Discussion

We tune OMD's current response by varying the number and the polarity of stimuli with a fixed frequency. In fact the application of incoming signals reported in Figure 3 a) and b), leads to the instauration of potentiation and depression processes in the device (respectively) that can be quantified in terms of the observed relative change ($\Delta I/I_0$) as a function of the number of stimuli "experienced" by the memristor (Figure 3 c) and d)).
As it is shown in Figure 3 c), a total relatively small number of positive pulses (< 5k) does not induce a significant variation in the conductive state of the device so that a small, practically zero, change is observed in the current before and after the training pulses (see the bottom right inset of Figure 3 c)) . This effect, already reported in [3, 20], can be considered as a short term potentiation that is a temporal enhancement of synaptic connections, which then quickly decays to its initial states[2, 3, 19]. By increasing the total number of pulses, the difference between the two different conduction states becomes gradually wider up to the limiting value of 400 % relative change after about 15k stimuli, achieving a prominent potentiation effect (LTP).

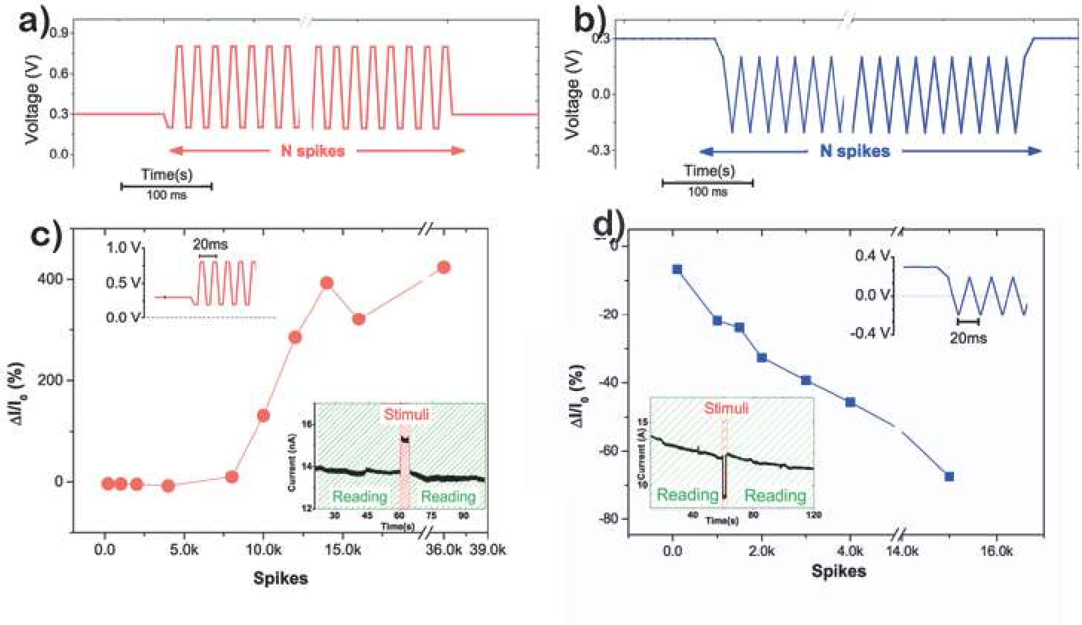

Figure 3: LTP and LTD induction with different voltage pulses: **panel a)** voltage pulses used for the potentiation and, **panel b),** for the depression of the memristor **panel c)** percentage variation of the current ($\Delta I/I$) versus the number of spikes in the potentiation process: in the top inset the pulse profile used, in the bottom inset example of the output profile of the current obtained for a low number of applied stimuli. The application of a limited number of positive spikes to the organic memristor doesn't affect the internal conductivity of the device (bottom inset) that after the stimulus exhibits a current level comparable to the initial one. Increasing the number of stimuli, the variation between the initial and the final values of the current becomes wider, demonstrating the so called LTP function; **panel d)** percentage variation of the current ($\Delta I/I$) versus the number of spikes in the depression process: in the inset the pulse profile used; the application of negative spikes to the memristive device leads to a gradual decrement of the final current in analogy with the LTD neuromorphic function.

When instead the device is exposed to multiple trains of negative voltage pulses (Figure 3 d)), the system analogously shows a depression of the conductivity (LTD) as a function of the number of incoming signals while the STD, the counterpart of the STP for the depression, is not observed. In fact, even for a very low number of stimuli (100 pulses in 2 s) the devices show a 10% relative change that is significant when compared to that obtained with a larger number of stimuli.

In order words, the device stores and integrates nonlinearly the incoming received signals, and, when the number of collected input pulses exceeds a threshold, it varies its internal conductivity as a function of the intensity and the polarity of the training routine, achieving the LTP or the LTD neuromorphic functions.

This behavior well mimics the biological synaptic phenomena called temporal integration: in biological network each neuron, due to the large connectivity, summarizes and integrates nonlinearly various presynaptic inputs and generates an action potential only if a threshold level of summation is reached [3, 20, 30-34].

Results, shown Figure 3, confirm the adaptive response of the memristive devices and extend, for the first time to the OMDs, the ability to control the efficiency of neuromorphic performance by varying the polarity of the applied pulses.

In natural synapses, repeated stimuli called neuronal action potentials (Figure 4a in red) on a specific natural synaptic site potentiate or depress the synapse's strength depending on the frequency of the pulses that the cells received. A few seconds of a tetanic stimulation (high-frequency sequence of individual pulses) enhance the synaptic strength while long periods of low frequency stimulation induce the depression of the connection[2]. Typically for inducing LTP a 2-5 ms stimuli delivered at 100 Hz are required, while for inducing the depression, the required frequency is reduced to 1Hz[28, 29, 35, 36].
For this purpose, we have tested our memristive devices applying the pulses with the same shape (in terms of amplitude and duration) for both potentiation and depression but varying the frequency of the spikes, in analogy with the typical shape of the biological action potential (Figure 4 a)). We induced LTP and LTD respecting the natural ratio between the pulse duration (5 ms) and time interval between them (from 10 to 1000 ms, respectively) that is about ½ in the first case and 1/20 in the latter one [28]. The typical voltage profile used in the experiment is shown in Figure 4 b): the training routine is reported in blue (and in Figure 4 c)) while the initial and the final intervals (colored in red) are the reading phases.

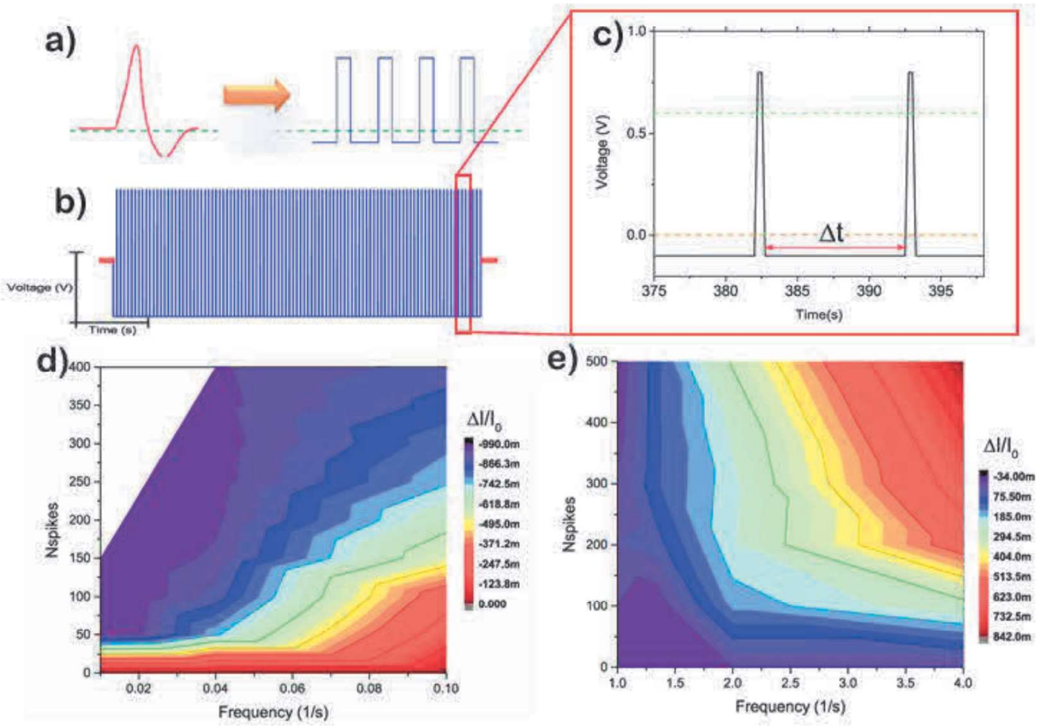

Figure 4: LTP and LTD induction with different frequency pulses: panel a) in red a biological Action potential, in blue the pulse used for the experiment: dotted green line highlights the "resting" potential; **panel b)** a typical voltage profile used in the experiment: the conductivity was acquired through a reading phase (red) before and after the training routine (blue); **panel c)** detail of the pulse used in the experiment as training routine: dotted lines highlighting the oxidation (green) and reduction (yellow) threshold voltages ;Δt is the time interval between the end of the first and the begin of the second pulse; **panel d)** color map of the variation of the current (ΔI/I) in function of the number and of the frequency of spikes applied to the memristor n the LTD regime; exploring low frequencies pulses, the memristive device exhibits a negative variation of current, indication of the performing of a depression process; **panel e)** color map of the variation of the current (ΔI/I) in function of the number and of the frequency of spikes in the LTP regime. In the case of higher frequencies, the increment of the device conductivity, and so the successfully performed LTP, is demonstrated by a positive variation on the current between the initial and the final current values.

We report in Figure 4 d) and e) the color coded maps of the relative current changes in the cases of lower and higher frequencies stimulations respectively as a function of the number of applied spikes and frequencies explored.
For the low frequency stimuli, all the tested combinations between number and frequency of the input signals leads to the decreasing of the device output current (Figure 4 d)) that results in a negative variation of the output current value ($\Delta I/I0 < 0$). In other words, totally in agreement with what reported in Figure 3 d), even a relatively short training routing can induce an effective depression of the device conductivity. This behavior is well documented in biological synapses[37] where the effect of a prolonged, low fixed frequency, stimulation is comparable to multiple trains of shorter periods of stimulation. An individual short stimulation in the latter protocol induces LTD of small entity that, however, can be cumulated with successive trains of inputs.
Moreover, as it is shown in Figure 4 d), the lower is the frequency the smaller is the number of spikes, necessary to switch off the device conductivity while, increasing the frequency, a more intense training routine should be applied to achieve the same result.
When a single OMD experiences a high frequency training routines (Figure 4 e)) it increases its internal conductivity proportionally to the number and the frequency of the incoming input signals. To obtain the highest current change, the concomitant presence of both high frequency stimuli and high number of applied pulses is necessary. Again, this behavior is in a very good agreement with and perfectly emulates the already mentioned biological temporal integration as well as the biological LTP dependence of the protocols on the number of the stimulations (for a fixed frequency)[29]. Moreover, in Figure 4 e) a region where the training routine is not effective can be highlighted (colored in purple): under a certain stimulating frequency the OMDs does not produces any increase of the conductivity even with the application of an intense training routine. In addition, in all the frequencies explored under a threshold number of input stimulations, the organic memristor does not show any significant variations in its internal conductivity, in a total agreement with the temporary behavior (STP) already discussed in Figure 3 d). Results reported in Figure 4 e) also demonstrate the already mentioned non-linear synaptic temporal integration. In fact, above a certain number of given incoming inputs, all the responses to the higher frequencies tested show a gradual variation of the current that increases non linearly with the number of stimuli in a very good agreement with the LTP biological behavior.

3. Conclusion

Summarizing, the overall trends reported in the two color coded maps of Figure 4 and in Figure 3 d) and e) finely reproduce in several aspects the typical LTP and LTD [28, 29, 35] functions of biological synapses. In analogy with other inorganic or organic devices (memristive or not), OMDs present a transition between LTD and LTP as a function of the polarity of the incoming stimuli. A feature that strongly distinguishes them from other kinds of devices is, however, the possibility of inducing the transition between depression and potentiation by means of a frequency variation using a fixed shape pulses instead of a polarity change. A second very relevant neuromorphic result is that our OMDs exhibit the natural transition from the temporary (STP) to the permanent (LTP) memorization levels triggered by the number of the applied stimuli, perfectly emulating the typical biological synaptic behaviors. From this point of view it is worth emphasizing that, since the working principle of the device is based on PANI redox reactions, any permanent increase (as well as decrease) of conductivity, is characterized by a stable behavior leading to multiple and stable memorization levels.

In conclusion, we believe that the results achieved here on a single OMD device constitute a novel accurate and detailed synaptic model that makes it ideally suitable for artificial neural and hybrid networks as well as for neuromorphic computing. In perspective, our results could support a variety of bioelectronic and neuroprosthetic applications, paving the way for a true revolution toward "intelligent" neuroprosthetics and augmentation of brain function.

4. Materials and Methods

4.1. Device's preparation

For the realization of the device we used the method already reported in several papers[11-13]. Briefly, a solution (0.1 mg mL^{-1}) of emeraldine base form of Polyaniline (PANI) (Sigma Aldrich, Mw≈100 000) in 1-methyl-2-pyrrolidinone (Sigma Aldrich ACS reagent ⩾99.0%) with the addition of 10% of Toluene (AnalaR NORMAPUR® ACS) is deposited by means of the Langmuir-Schaefer technique onto a masked glass substrate with two evaporated separated chrome electrodes (Source and Drain) using purified water (MilliQ) as a subphase.
60 layers of PANI form the channel of the device (of about 5x15mm) that has to undergo to two doping processes performed by dipping the sample into HCl 1M for 30 s and, after a resting time of 30-40 minutes, for 15s.
A water solutions (20 mg ml^{-1}) of polyethylene oxide (Mw = $8 \cdot 10^6$ Da (PEO)) doped with 0.1M $LiClO_4$ (Sigma) is used as polyelectrolyte solution and deposited as stripes of about 2 mm width on the PANI active channel in a crossed configuration. . A reference electrode made of a silver wire of 0.05 mm diameter is inserted into the PEO matrix. Since the pH of the polyelectrolyte gel is neutral, it was necessary to reset the pH of the conductive polymer exposing the device to HCl vapors for few seconds.

4.2. Device's characterization

For qualifying our devices, we considered the differential current, Idiff= I_D-I_G where I_G is the gate current and I_D is the drain current (Supplementary information). The application of the voltage and the measurements of the total current were performed with two Source Measure Units (NI PXle 4138/9) and drove by an ad hoc labview code.
All our training routines consist of 4 different steps: 1) potentiation or depression of the conductive properties of the organic memristive device with the application of 0.8V or -0.2 V respectively for the reset of the system from the previous operation; 2) application for 1 minute of a "reading" voltage of 0.3 V; 3) application of N number of spikes working as stimuli and 4) again the application of the reading voltage.
The ΔI/I parameter is obtained calculating the difference of the output currents between the first and the second reading intervals divided by the initial current value.
In the case of the demonstration of the voltage dependent transition between LTP and LTD, the applied spikes (shown in Figure 3 a) and b)) present different voltage profiles: for the potentiation, stimuli had an amplitude of 0.8 V and a duration of 10 ms and two subsequent pulses were separated by 10 ms in which the bias potential of 0.2 V was applied; in the case of the depression, the pulses had an amplitude of -0.2 V .
For demonstrating the possibility of driving this transition using a frequency dependent incoming stimuli, we used spikes with the duration of 500 ms and 0.8 V of the amplitude superimposed to the bias voltage of -0.1 V. The frequency was changed by varying the time

between two consequent spikes ($f_i = {}^{1}/_{\Delta t_i}$). Our system was tested using 1, 1.33, 2 and 4 Sampling/s and 0.1, 0.04, 0.02, 0.01 Sampling/s as selected frequencies. Furthermore, after the application of the i-th train composed by n_i number of spikes with the proper Δt spacing time (blue lines in Figure 4 b), we applied the already mentioned reading voltage (0.3 V) to test the conductivity level and to reset the system in analogy with the biological resting potential (red lines in Figure 4 b).

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Contributions

S.B. manufactured the devices, performed the measurements and the analysis of data; S.B, V.E., S.I. supervised the work and wrote the manuscript.

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