# Organic Memristors and Adaptive Networks

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**Abstract.** We describe the organic memristor – an element which varies its conductance according to the history of its previous involvement into the signal transfer processes. After the presentation of basic principles and fundamental properties, we present the architecture of the organization of model networks, capabale to learning. Finally, we discuss the possible ways, alternative to the currently existing lithography-based technologies, that would result in the fabrication of statistically organized fiber networks of such elements.

**Keywords:** Organic memristor, conducting and ionic polymers, adaptive networks, polymer fibers.

## 1 Introduction

A fundamental difference of the brain organization with respect to that of the computer is connected to the fact that memory and processor are not separated. Both functions are executed by the same elements. Such organization allows learning. In fact, it implies not only recording of the information but also modification of the "processor" for the better resolving of similar problems in the future.

Considering the realization of electronic analogs of such systems, it is useful to recall a hypothetical element, "memristor", introduced by Chua in 1970 [1]. It must vary its resistance according to the history of its involvement into the signal propagation. Such elements could be the basis of electronic circuits capable to reproduce to some extent the learning processes occurring in the brain.

There are several models describing learning processes. Here we will consider mainly the synaptic type of learning that can be described by the Hebbian rule [2]:

*"When an axon of cell A is near enough to excite cell B and repeatedly or persistently takes part in firing it, some growth process or metabolic change takes place on one or both cells so that A's efficiency as one of the cells firing B is increased*".

It means that the synapses which provide the signal transfer from one neuron to another will be strengthened every time they are used. In terms of electronic circuits, it means that the resistance of the wire, transferring the signal from one non-linear element to the other, must decrease each time when the signal is transferred. Thus, the

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successful realization of the hypothetical memristor, mentioned above, would provide an essential step towards mimicking learning capabilities.

Recently a claim for the first realization of a material memristor, a doped $TiO_2$ film, has been reported [3]. The attribution of the device to the memristor hypothesised by Chua is somewhat doubtful [4] and cannot be useful for the task, mentioned above. Its characteristics reveal bistable electrical behaviour, very useful for memory applications, but not suitable for mimicking learning. In fact, the last item demands the gradual variation of the element resistance, corresponding to the integral charge transferred by the junction.

In 2005 we have reported a polymeric electrochemical element with properties very similar to the hypothetical memristor [5]. Its working principle is based on the dramatic difference in the conductivity of conducting polymers (in particular – polyaniline (PANI)) in a reduced and oxidized states [6]. This difference can reach 8 orders of the magnitude. The transition can be triggered by the applicaton of the appropriate potential. At a first glance, it seems very similar to the electrochemical field effect transistor, known since the end of 1980 ies [7]. However, there are significant differences in the construction and, especially, properties of the device with respect to electrochemical field effect transistors. The closest analogue of our device, reported in the literature, is a polymeric electrochemical rectifying element [8], even if there are significant differences also with this system.

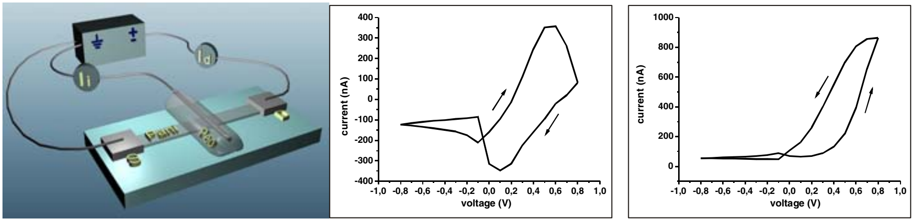
For the aims of learning behaviour mimicking, the most important characteristics of our polymeric electrochemical device are the gradual increase of the conductivity when operating under positive voltage bias, and its gradual decrease when negatively biased. Therefore, the device seems to be a real organic memristor, suitable for the fabrication of adaptive material networks.

In this paper we present the basic ideas underlying the structure, properties and working principles of the single organic memristor. Thus here we do not present details of the technological processes, giving only essential requirements and referring to the published works. Then, we will illustrate the possibility to mimic learning behaviour on simple examples of model circuits. Finally, we will overview approaches that are expected to lead to the realization of complex networks capable of learning, highly parallel information processing, and decision making.

## 2 Organic Memristor

The scheme of the organic memristor is shown in Fig. 1a. The active channel is formed from PANI, deposited onto a support with two electrodes. The only requirement for the support material is its insulating nature. Initially, we have used glass, but the last generation of the devices and deterministic networks we made on the flexible supports such as polyimide Kapton films – a highly insulating inert material.

An important feature of the active channel is its thickness. It must provide a significant conductivity, but it must be as thin as possible, as processes responsible for the conductivity variation are diffusion controlled (see later). Therefore, Langmuir-Blodgett technique is the most appropriate for the channel formation, allowing to fabricate structures with a resolution at the level of single monomolecular layer [9].




a b c

**Fig. 1.** Scheme of the organic memristor (a) and cyclic voltage-current characteristics of the organic memristor for ionic (b) and electronic (c) currents. Arrows indicate the direction of the voltage scans.

The other important part of the device is a medium for the redox reactions. The central part of the channel is covered with a solid electrolyte. Polyethylene oxide (PEO) was chosen as the material of the electrolyte matrix, as it has demonstrated adequate properties in the case of rechargeable batteries applications [10]. Lithium salts (in particular, lithium perchlorate) were used for the electrolyte formation, as lithium provides the highest mobility, especially in solid electrolytes. The area of the PANI channel in a contact with the electrolyte is the “active zone”, as practically all redox reactions and, therefore, the conductivity variations occur in this area [11].

Finally, we need to have a reference point for the potential. Therefore, silver wire is inserted into the electrolyte stripe and is considered as the reference electrode. This wire, together with one of the electrodes on the support, is connected to the ground potential. The conductivity variations will occur in the active zone according to its actual potential with respect to that of the reference electrode.

A scheme of the electric connections to the memristor is also shown in Fig. 1a. Even if it has only 2 electrodes for connecting to the external circuit, we will consider here 3 electrodes for the better understanding of the working principles. Two currents were measured for the device characterization: $I_i$ – is the ionic current in the circuit of the reference electrode; $I_d$ – total current in the device. For a better understanding of the memristor working principle we report $I_i$ (Fig. 1b) and $I_e$ (Fig. 1c), where $I_e$ is an electronic current, determined as a difference between measured $I_d$ and $I_i$.

Let us consider characteristics reported in Fig. 1. Initially, the active zone is in the insulating state resulting in a low $I_e$ value. At some voltage value we can see the peak in the $I_i$ characteristics (Fig. 1b), corresponding to the significant increase of the electronic current (Fig. 1c). The voltage value corresponds to the oxidizing potential. For the presented configuration, this voltage is about +0.6 V, which is higher than the oxidizing potential of PANI in solution (+0.3 V) [12]. This difference can be easily understood as the potential is applied to the drain electrode and is distributed along the whole length of the channel. Therefore, the actual potential of the active zone is significantly lower with respect to the applied voltage.

After passing the oxidation peak, the active zone is in a conducting state. The voltage cannot be increased to higher values [13]. Reaching this value, the voltage was gradually decreased. The electronic current (Fig. 1c) shows linear Ohmic behavior during the voltage decrease, until the voltage reaches the value of about +0.1 V. For the ionic current (Fig. 1b) we can see the negative peak, corresponding to the

PANI reduction. Then, we see significant decrease of the memristor electronic conductivity. In our first memristors, the conductivity ratio between the conducting and insulating states was about 2 orders of magnitude. However, optimization of the memristor structure and composition have allowed us to increase this ratio to 4 orders of magnitude [14].

In order to mimic the learning behavior it is very important to analyze the kinetics of the conductivity variations at constant applied voltage. This characteristics for positive (upper +0.6 V) and negative (any) applied voltages are shown in Fig. 2.

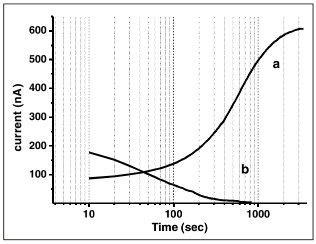



**Fig. 2.** Kinetics of the conductivity (current) variations of the organic memristor for the when biased with +0.6 V (a) and –0.2 V (b)

For both cases we can see a gradual variation of the conductivity. However, kinetics of these variations are different. This difference was explained by the fact that in the case of the positive bias to the memristor in the insulating state, only part of the active zone is under the oxidizing potential. Thus, we have gradual transformation of the active zone into the conducting state. Instead, for the negative bias the whole active zone is under the reduction potential and the conductivity transformation occurs in the whole zone, determining the faster kinetics. A more detailed explanation of the observed behavior and the model, describing it, can be found in [15]. It is interesting to note that our last results [14] have demonstrated the possibility to vary the kinetics for the positive bias just by increasing the conductivity of the PANI layer, in good agreement with the developed model.

The characteristics presented in Fig. 2 demonstrate explicitly that the device can be really considered as a memristor. In reaity, our last results, based on real time comparison of the conductivity and grazing angle X-ray fluorescence, have demonstrated that the conductivity variation is directly connected to the transferred ionic charge. These characteristics can be the basis for adaptive behavior. In fact, the increase of the conductivity at positive bias represents the fundamental property necessary for unsupervised learning. In agreement with the Hebbian rule, which claims the strengthening of the synapses, the behavior according the Fig. 2a will result in the formation of preferential signal pathways in a network composed of a large number of memristors. Thus, the solution of a problem will modify the whole network, yielding a processor which can solve more effectively similar problems in the future.

The decrease of the conductivity at negative bias will be useful for two main reasons. First, if we consider the network composed of a large number of memristors, providing multiple signal pathways between input-output pairs, we can expect that for the positive bias the system, after working for a long time, will arrive to the situation when all signal pathways will become preferential ones (all memristors will be in a high conducting state). This fact, by the way, was the reason of the beginning of the research activity decrease on the hardware realization of the neuron networks in 1980ies [16]. In biological beings the prevention of the saturation is performed by so-called inhibitory neurons. In the case of the artificial networks, we can use the characteristic, shown in Fig. 2b, as the analog of the inhibitory activity. Periodic application of the negative bias between all input-output pairs will result in the partial inhibition of the signal pathways, created during unsupervised learning according to the Fig. 2a. Thus, the system will never come to the equilibrium, allowing further strengthening of the synapses analogs. Second, the curve shown in Fig. 2b is a basis for the supervised learning. In fact, *a priori* wrong preferential signal pathways, formed during unsupervised learning, can be easily inhibited by the external training action, just applying negative bias between the corresponding input-output pairs.

Summarizing, the presented characteristics allow us to claim that we have realized an element, which behaves similarly to synapses, the most complex node of the nervous system. The next step is to demonstrate its applicability to the formation of adaptive networks, imitating the possibility of supervised and unsupervised learning.

## 3 Adaptive Circuits

The possibility of unsupervised learning has been demonstrated on a circuit containing only one organic memristor. The circuit was constructed for mimicking the learning behavior of pond snail during feeding. The learning in this case means the association of the touching stimulus with the presence of the food. After touching the snail lips with sugar, it begins to open the mouth after touching even without sugar. The constructed circuit contained two inputs for signals, corresponding to the mentioned stimuli (touch and test). The value of the output current corresponds to the signal for the task execution (mouth opening). This action can be performed only if the value of this signal is higher than some threshold value. The memristor is initially in the insulating state. Both applied voltages separately are not enough for its transferring into the oxidized conducting state (+0.3 V each). However, when the voltages are applied together, the memristor will be transferred into the conducting state and further application of one signal only will result in the significant increase of the output signal that will overcome the threshold level necessary for the task execution. Initially, this possibility has been demonstrated in dc mode [17]. However, more adequate mimicking of the biological beings demands the work in the pulse mode, and in particular imitating spike signal propagation in the real snail [18]. Therefore, investigation of the organic memristor learning capabilities have been also performed in the pulse mode [19]. The response to the test input was increased from 45 nA to 107 nA after strengthening training. We then proceeded with 15 pairs of inhibition training cycles, after which the output of the device was reduced to 59 nA. Then the system was subjected to the strengthening training again, resulting in the output value increase up to 150 nA.

It is interesting to note that the relative increase of the output signal, obtained in the artificial system is comparable with that experimentally measured in the real snail [20].

The possibility of the supervised learning has been demonstrated on a network composed of 8 discrete elements, connecting two inputs and two outputs [21]. The possibility to strengthen or inhibit signal pathways between any desirable pairs of input-output electrodes was demonstrated by the application of an adequate training voltage between them. Currently, such circuits have been fabricated in the integrated mode on the flexible support (Fig. 3).

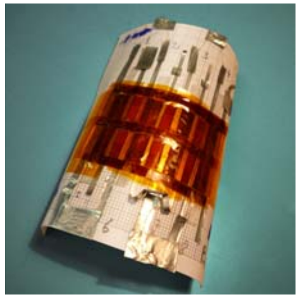


**Fig. 3.** Adaptive network with 8 organic memristors fabricated on flexible support

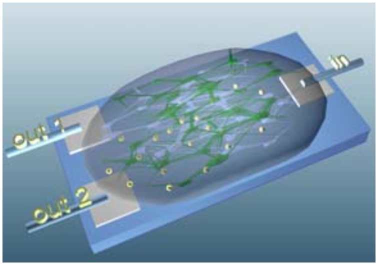
This structure has demonstrated the adaptation capabilities similarly to the network composed from discrete elements. In particular, strengthening of the pathways between selected input-output pairs of electrodes by training with positive bias has resulted in the four times increase of the output current. Instead, inhibition of the pathways between selected input-output pairs of electrodes by training with negative bias has resulted in the three times decrease of the output current.

The training of the network can be repeated, bringing the system to the initial state or even inverting the strength of signal pathways between input-output pairs. We want to underline that this rather simple network allows adaptations just under the action of external stimuli and does not require any variation of the circuit architecture.

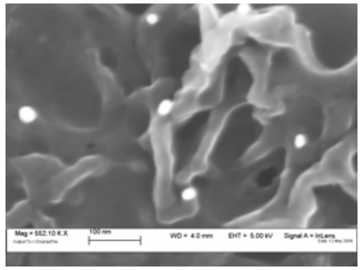
## 4 Composite Organic-Inorganic Structures

The abovementioned memristor itself and the model networks have demonstrated to possess the necessary fundamental properties to permit Hebbian learning and to represent to some extent the properties of synapses. Further mimicking of the nervous system requires the realization of the neuron body analogs. This element must allow the income of all signals into it, but to provide output only when the integral of all incoming signals will overcome some threshold value. As it was shown at the macroscopic scale, the Shottky effect can be used for these purposes [21]. It demands the organization of the contact of materials with significant difference in the work functions. In particular, the structure where PANI is in the contact with gold has shown rectifying characteristics.

Complete reaching of the aim of the mimicking the neuron body behavior requires the use of gold particles distributed in the PANI matrix. Schematic representation of the organization of such network is shown in Fig. 4a.




a



b

**Fig. 4.** Adaptive network composed from conducting/ionic polymers – gold nanoparticles composite structure (a). SEM image of Au nanoparticles – PANI composite layer (b).

Considering the currents as the signal (its polarity must provide no barrier for the signal income but a barrier for the out-going signal), it will be necessary to accumulate enough charge on the particle, to provide the potential higher than the Shottky barrier. It is interesting to note that it is possible to vary the threshold value of the accumulated current by simple variation of the gold particle diameter (of course, if we have the possibility to work with arrays of monodisperse particles; in the case of polydisperse particle arrays, we will have statistical distribution of the threshold values from one particle to the other, what can be also useful for the fabrication of statistical adaptive networks). In fact, the potential on the particle will be determined by the accumulated charge (integral of the input currents) and the capacity of the particle, directly connected to its diameter.

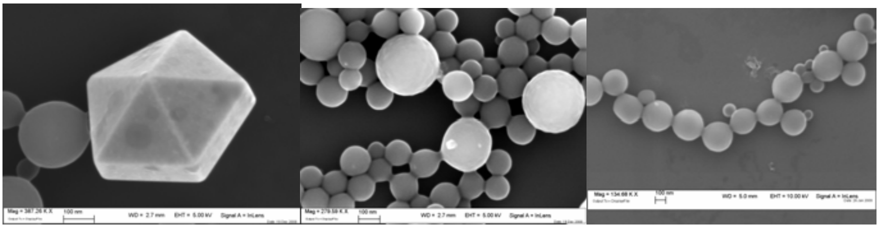
The actual realization of a network shown in Fig. 4a demands solving several preliminary tasks, such as growth of Au particles down to the nm size and their effective binding to PANI matrix.

Currently, different technological approaches are used to produce nanoparticles of different materials, including gold. For instance, we have produced Au nanoparticles by sodium citrate reduction of $HAuCl_4$. A SEM image of the composite layer of these particles with PANI fibers is shown in Fig. 4b.

Very interesting results were obtained when the Au particles were grown under the preformed Langmuir monolayer of PANI at the subphase, containing $10^{-4}$ M $HAuCl_4$. PANI monolayer was compressed till the surface pressure of 10 mN/m. The described system was left at ambient conditions for a week for the Au particles growth. It is interesting to note that nanoparticles even with pentagonal shape were observed (Fig. 5a).

However, most typical SEM images of the realized structures are shown in Fig. 5b.

As it is clear from the image, the average sizes of the particles were increased. This is probably due to their covering by the PANI layer during the growth and transferring onto solid supports. In some zones these composite particles form linear chains (Fig. 5c). Controlled growth of such chains would be an important step in the field of nanoarchitecture.



a b c

**Fig. 5.** SEM image of Au structure formed under PANI LB monolayer (a). Typical SEM images of Au nanoparticles formed at PANI monolayer (b, c).

## 5 Statistical Networks of Polymer Fibers

Effective mimicking of the brain structure and function require the organization of the networks, composed of millions of memristors. Application of the current fabrication methods, based mainly on high resolution lithography, can solve the problem only partially. It will allow the miniaturization of the elements sizes to sub-micron level. It will also allow to increase significantly the integration degree, arriving to the circuit with millions of the elements. However, the realized structures will be planar in nature, while we have 3D organization of nervous systems in biological beings.

Therefore, the search for the new approaches, based on bottom-up self-assembling techniques, widely discussed now [22-24], seems more promising.

In this work we will limit ourselves to the discussion of the statistical networks of polymeric fibers. The capacity of polymers (PEO, in particular) to form thin fibers is well-known. For example, they can be fabricated by the thinning of the PEO gel in the rather high electric field [25]. In our study we have used another method of fiber formation [26], i.e. vacuum treatment. A PEO gel was cast onto the surface of the solid support and rapidly pumped. As a result, a fibrillar structure of the PEO was realized. In the second step, this fibrillar matrix was used as a scaffold for fabricating PANI fibers on it. The initial hypothesis of the possibility to form memristor-like junctions in such structure if the number of formed fibers is rather large and statistical probability of the crossing of conducting and ionic polymers and reference voltage source is rather high, has been verified demonstrating rectifying behavior of the junction with the fibrillar active zone instead of deterministic layer of PANI and PEO stripe with reference electrode [27].

As the next step, it is necessary to demonstrate that this approach allows to reproduce properties of not only a single memristor, but that it is possible to fabricate the adaptive network.

We started by reproducing the properties of the previously discussed network with 8 memristors. It must contain one input and two output electrodes and allows to have higher signal at any desirable output when applying an appropriate training procedure. The network was organized by fibrillar structures formation. Its schematic representation is similar to that shown in Fig. 4a. Solid support with one input and two output electrodes was covered by PEO gel (silver wire, that will act later as a reference electrode, was placed into this gel), placed into the essiccator and exposed to vacuum, created by rotary pump. As the next step, the structure was covered by PANI solution and again exposed to vacuum. The only difference with respect to Fig. 4a is the absence of Au nanoparticles in the structure. Input and reference electrodes were connected to the zero potential level, while the voltage was applied to output electrodes. Two current values were considered as output signals. For testing, the voltage of +0.4 V was applied and the current value in circuit of each output electrode was analyzed. Training was performed by the application of different voltages to two electrodes. In particular, we applied +1.2 V to the first output electrode and -0.6 V to the second one. The results, demonstrating the training capabilities of this network, are presented in the Table 1.

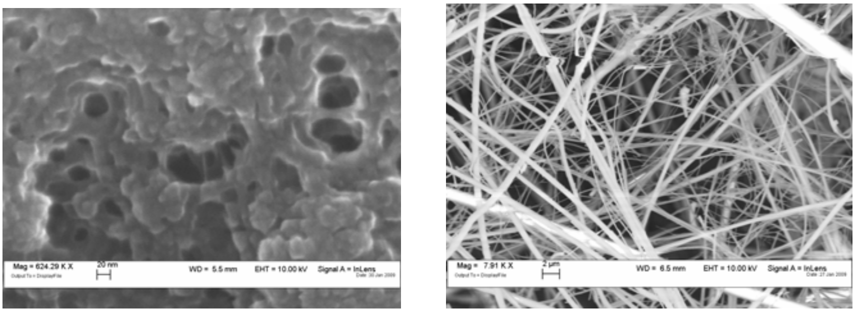
**Table 1.** Learning capabilities of the statistically formed network of polymer fibers

| | **In-Out1** | **In-Out2** |
|---|---|---|
| Before training | 20 nA | 20 nA |
| After training | 200 nA | 20 nA |

As it is clear from the Table 1, the network has demonstrated the possibility to adapt (strengthening and inhibition of signal pathways), reproducing the supervised learning, similar to that observed in the deterministic network composed of 8 discrete memristors [21]. We would like to underline here the importance of the presented results: the reported network has not been realized by the fabrication of separated elements and their interconnections. The system is a statistically distributed array of conducting and ionic polymer fibers with the source of the reference potential somewhere in this structure. However, as the system is complicated enough, it provides statistical formation of at least several areas where suitable mutual arrangement of the necessary elements have been reached. Once fabricated, the network can be externally trained to yield the desired behavior.

Even if the statistical fiber network is a very important step towards the realization of complex adaptive systems, it is still very far from the practical applications. The main critical point is the stability of the structure and properties of such networks. In fact, for the described structure we have observed the variation in the conductivity practically immediately after the beginning of the measurements. After about 40 minutes, the conductivity decreased of about one order of magnitude, and after 2 hours the network was practically destroyed. Such behavior is not strange considering that we deal with free standing polymer fibers. These structures are not stable even in unbiased conditions. In the case of the current passing through induced preferential pathways, we must have thermal gradients in the network resulting in the fiber deformations and melting. Therefore, we need to stabilize the structure for making it more appropriate for complex information processing. In other words, we need to fabricate a rigid frame, maintaining the fibrillar network, as it occurs in nature (f.i. skeletons for the bodies stabilization).

We are considering two different rigid frames in order to solve the stability problem. The first one is based on the use of porous materials as a support for the fiber network fabrication. A SEM image of the polymer fibers formed on a porous stone by the vacuum treatment is shown in Fig. 6a.



a b

**Fig. 6.** SEM image of the polymer fibers formed on porous support (a). SEM image of the glass fiber matrix covered by PANI layer (b).

As it is clear from the image, the method allows to form fibers with very small diameter (10 nm), what will serve for the increase of the integration degree, providing more functional units within the same volume. The image demonstrates also that the fibers are very stable, maintaining their structure even in the high vacuum of the electron microscope and under the electron beam irradiation.

The second approach is based on the use of glass fibrillar structures (glass filters) as solid supports. In this case, it is planned to cover the glass fibers by PANI as the first step and then to fabricate PEO fiber matrix between them. An image of the glass fiber matrix after its covering with PANI is shown in Fig. 6b.

The image demonstrates clearly the effectiveness of the glass fibers coating with conductive PANI. In fact, it was very difficult to acquire images from the original matrix due to its insulating nature. After the covering, instead, the images have revealed the significantly improved contrast.

## 6 Conclusions

In the present paper we have summarized the basic principles of the organic memristor organization and properties. Its conductivity variations and the memory effects are connected to the redox processes occurring in the active zone of the memristor (contact area of the PANI channel and PEO solid electrolyte). The organic memristors were shown to be absolutely adequate for the realization of adaptive networks capable for supervised and unsupervised learning. We have not discussed here such important aspects as the stability of properties [27] and possibility to produce current auto-oscillations at fixed bias conditions [28]. Finally, we have discussed alternative approaches allowing to reach the same goals (learning, adaptations) using statistically formed networks of polymer fibers.

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