

Characterization of Ag-PEO₁₀LiCF₃SO₃-PolyPyrrole-Au Neural Switch

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Abstract: - The design, build and characterization of a semiconducting, organic neural switch are carried out. Voltage-Current characteristics for an ionic-electronic interacting neural switch are presented. Neural Switch characteristics analyzed using an electronic equivalent circuit model. The model is proved to describe to a great degree of soundness the interacting mechanism between the used materials. Programmability of the switch is proved to be bi-directional and reversible with hysteresis effect which is due to excess charge storage, with a behavior similar to a biological Synapse.

Key-Words: - Neural, modeling, Synapse, Memory, Information Processing, Polymers, Computing, Intelligence

1 Introduction

Electronic neural network hardware implementations, particularly with parallelism and self-learning abilities, generated a need for miniature variable analog memory elements (synapses). The connection strength (electrical resistance) of such film deposited devices is reversibly modified through an electrochemical process. The electrical resistance of such a device can be modified to, and stabilized at, any desired value over a wide dynamical range.

Such compact devices are used in an artificial neural network arrangement that comprises a collection of simple processors (Neurons), interconnected through a complex network of weighted elements (synapses). Such massively parallel architecture possesses unusual properties applicable to complex problems in concurrent information processing, fault tolerant pattern recognition, and other fields [1, 2, 3, 4, 5].

The development of neural circuits based on correlated activity, relies on two critical mechanisms. The best known of these is activity-dependent synaptic modification along the lines proposed by Hebb. Equally important is a mechanism that forces various synapses to compete with one another so that when some synapses to a given postsynaptic neuron are strengthened, others are weakened.

In an electronic neural network, the weighted synaptic interconnections modulate the communication between the neurons. The key requirement for learning in networks as a result of communication between various neurons is the programmability of synaptic connections to

reversibly alter their magnitude in small increments during learning [13, 14, 15, 16, 17].

In this paper the design of a three terminal double layered, sandwiched (Ag-PEO₁₀LiCF₃SO₃-ppy-Au) device is presented to be used as analogue, reversible, programmable; resistance variable neural switch is introduced. The device behaves in a similar manner to a SYNAPSE through which NEURONS are inter-connected in a sophisticated fashion. The description of network activity at such level, analyzing the switching pattern under different conditions, provides a vast amount of information about neural network behavior. Mathematical modeling is a powerful approach for understanding the complexity of biological systems. Recently, several successful attempts have been made for simulating complex biological processes.

In this paper the fundamental characteristics that describe the main functionality of an artificial synapse are presented, modeled and analyzed. Such a model based analysis provides a good insight into the behavior of the neural switch and any futuristic implementations.

2 Neural Switch Design

Polymer electrolytes, in particular polyethylene oxide - alkali salt complexes, and the extensively studied polyethylene oxide - lithium trifluoromethane sulphonate (PEO₁₀LiCF₃SO₃), have been applied as materials for high energy, long life, flexible re-chargeable batteries with good mechanical and stable electrochemical properties [22, 24, 25, 26]. Moreover the remarkable achievement of producing highly conducting polypyrrole (PPY) with excellent chemical and

thermal stability and its ability to switch between two states, namely conducting (oxidized) and insulating (reduced) has brought about the idea of combining these polymers to produce an ionic-electronic interacting mixed conductor. Such a device should have the ability to store the injected charge, affecting its conduction and switches states due to device resistance change. Such a change would be stable occupying a specified level until another charge is injected or removed. Such advances in polymeric science together with artificial neural networks enabled the design of an analogue neural processor [6, 7, 8, 9].

Ethylene oxide (CH₂ CH₂ O) is highly reactive with particular importance as a chemical intermediate due to the tendency of the ring to open in the presence of ionic catalyst. Co-dissolving an alkali metal salt with PEO [HO(CH₂CH₂O)_nH] and removal of the solvent will result in a mixed phase system which retains, in part, a complex of the polymer and the salt. It is shown that the paired ether oxygen electrons, which give the polymer strong hydrogen - bonding affinity, can also be involved in association reactions with a variety of monomeric and polymeric electron acceptors with polymer-alkali salts recognised as fast ionic conductors with wide charge storage using them as solid electrolytes in batteries, chemical sensors and other applications e applications .For pure PEO the crystalline phase occupies approximately 80% of the available volume, with the remainder being taken up by a dispersed amorphous phase. On addition of LiCF₃SO₃, Li⁺ ions form cross-links between ethylene oxide segments of neighbouring chains as the cation (Li⁺) forms ionic bonds with the negatively polarised oxygen atoms forming a closed helical structure with the (Li⁺) ions inside. The CF₃SO₃⁻ anion forms another ionic bond with the methylene group outside the helical structure when the ring structured monomers. Ionic conduction in PEOxLiCF₃SO₃ occurs by the cation (Li⁺) hopping between sites in either the single or double helix structures. However, this hopping model was discredited other by studies, which showed the ionic conductivity occurred mainly in the amorphous phase rather than in the crystalline phase. Furthermore experiments with dopant ions that form immobile ion pairs indicated that transport was not occurring down a helical path. A variety of experiments showed that ion transport in the majority of the polymer-salt complexes occurs through segmental motion of the polymer.

Despite some apparent similarities between electrically conducting polymers and conventional, doped inorganic semiconductors, their physics is

very different. A fundamental reason for this difference is that the polymers are molecular materials. Therefore, charge - transfer (electron-excitation) processes cause appreciable local modifications (relaxation) of the polymer chain geometry. The equilibrium geometry of organic materials in the ionized or excited state is different from that in the ground state because electron-phonon coupling occurs in addition to the local geometrical modifications of the chain, markedly affecting the electronic structure by inducing localized electronic states in the band gap. These are related to the charge-transfer-induced changes of the π -system of the polymer that are intrinsic to the parent material (organic polymer). In contrast inorganic semiconductors have states in the gap related to their dopant levels. The evidence that ionization or electronic excitation results in a local relaxation of the geometric and electronic structures of organic materials forms the basis of analysis of conducting polymers. Conducting polymeric films can easily be obtained by electrochemical oxidation/polymerization of the pyrrole monomer C₄H₅N. This electro synthesis has two advantages:

- (1) The polymer is formed in the conducting state.
- (2) The films possess good mechanical properties.

Poly(pyrrole) is considered to be one of the most interesting materials because of its accessibility, substantial conductivity and reasonable stability with considerable efforts made to establish the relationship between molecular structure, morphology, texture and electrical properties have been made. It is suggested that transport occurs as a three-dimensional variable-range-hopping process (VRH) where charge carriers not only hop between sites of closest contact of adjacent chains of poly(pyrrole) at low temperatures, but also to more remote sites in order to minimise the consumed energy. At higher temperatures the conduction dependency changes to cover exclusively the nearest sites.

Figure1 illustrates a classical structure of a neuron with synapses indicated, while figure 2 shows a simplified view of a synapse as the primary information-processing element based on its ability to control the current in or out of an electrical node by the potential on the other node. Hence, the synapse will control the conductance of the membrane between the inside of the postsynaptic cell and the extracellular fluid, which is controlled by the potential across the presynaptic membrane.

Inside this membrane is a high accumulation of specific neurotransmitting molecules where depolarized calcium channels permit its ions (Ca^{+2}) to proceed to the presynaptic cell from the synaptic gap activating the subcellular mechanism that releases the neurotransmitter molecules into the synaptic gap from which they diffuse to the postsynaptic membrane. This initiates a sequence of events resulting in the opening of ion-specific channels with three patterns of behavior [10, 11, 12]:

(1) Excitatory: depolarization of the postsynaptic membrane as a result of presynaptic depolarization due to the channels opened by neurotransmitters being specific to other ions like sodium (Na^{+}).

(2) Inhibiting: hyperpolarization of the post synaptic membrane as the potassium-specific channels are opened (Ca^{+2}).

(3) Shunting: when chloride channels open and increase the membrane conductance without a significant change in potential.

Analogy can immediately and easily be drawn between the behaviour of a synapse and the organic neural switch shown in figure3, as in the organic neural switch the gate bias potential affects the dc resistance between the source and drain. When programmed, the bias applied between the gate and

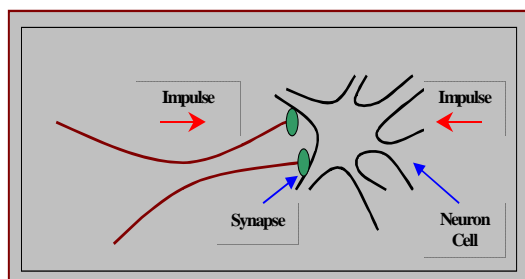


Fig.1: Biological Neuron

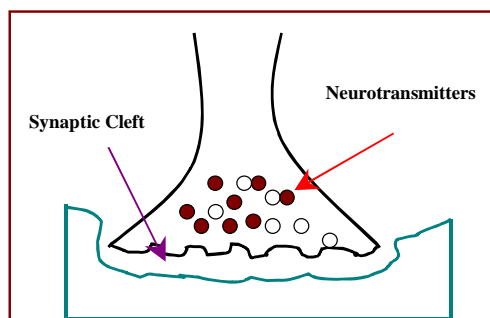


Fig.2: Biological Synapse

the shorted source/drain terminal will set up a field that will oxidize or reduce the polymer, and hence when the programming voltage is removed and a constant current is applied between the source and drain, the measured conductance will be related to

the previously applied bias and will be fixed until another latching potential is introduced. So, the switch may be used to control and modulate the communication between different artificial neurons in applications such as:

- Pattern recognition as in speech, image and odor discrimination,
- Complex optimization problems,
- Adaptive equalization in communication links

In the process of planning the overall switch pattern, efforts were made to allow the following:

- Uncomplicated and practical electrochemical growth and deposition of polymeric material (e.g. poly(pyrrole)), by making the size of the contact pads of the device large enough compared to the working electrode connections used to apply the potential to them.
- Elimination of the edge effect, in the z-direction, on the conduction behavior of the device, by permitting sufficient electrode length at either side of the polymer window.
- Simplification of the bonding task and formation of relatively strong bonds by deposition of NiCr on the contact pads.

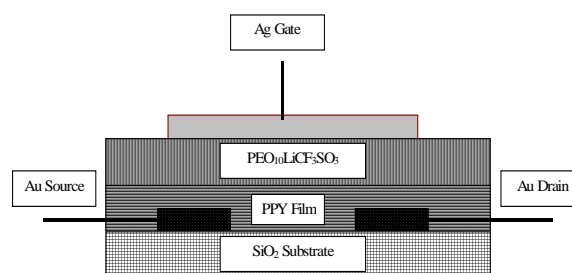


Fig. 3: The Designed Neural Switch

3 Experimental Arrangements

3.1 Processing of the device structure

In order to make the final device, barriers or insulations are necessary between the conductive polymeric materials (poly(pyrrole), $\text{PEO}_{10}\text{LiCF}_3\text{SO}_3$) and the silicon substrate [23, 27, 28, 29]. Holes or windows are etched through one barrier (photoresist) wherever contact is desired. The windows shapes are transferred to the surface of the oxidized silicon wafer using photolithography as a refined type of the photoengraving method. The process involves transferring the patterns from the masks to a light-sensitive material (photoresist), after which chemical etching is used to transfer the

windows geometry from the photoresist to the insulating material (SiO₂) on the surface of the silicon wafer. The photolithographical process comprises all the steps required to transfer a prototype from the mask to the surface of the silicon wafer. During the lithography process clean conditions are maintained as any dust particles on the original substrate, or particles dropping on the surface of the wafer during production, can result in defects in the final resist coating. So, prior to use, silicon wafers were cleaned to remove any particles on the surface as well as any remains of organic, ionic, and metallic impurities.

3.2 Polymers Preparation

3.2.1 Poly(pyrrole)

Poly(pyrrole) films were grown electrochemically using a three electrode cell consisting of:

- (1) Working electrode
- (2) Reference electrode
- (3) Counter electrode

The working electrode is the finite gold electrodes structure. The reference electrode used is a calomel electrode while platinum gauze was used as the counter electrode.

Before growth of the poly(pyrrole) was initiated, nitrogen was bubbled for 20 minutes, through the working solution of 50 mmol dm⁻³ pyrrole dissolved into 0.1 mol dm⁻³ TSA (sodium salt) as the supporting electrolyte. This degassing process was carried out in order to remove any oxygen and nitrogen was continuously passed over the surface of the solution during the growth operation to prevent re-oxygenation.

The films were grown by applying a potential step of 800 mV across the reference electrode and the working electrode, while submerging the finite electrode structure in the growth solution. The current flowing between the working electrode and the counter electrode was measured over a specified period of time. The film thickness is known to be proportional to the length of time for which the potential is applied. In this case films are grown for 60 seconds. This growth technique has the advantage of being able to produce thin films of poly(pyrrole) as the devices produced for this research have average film thickness of 3.5 μm.

3.2.2 PEO₁₀LiCF₃SO₃

Polymer films were prepared using poly(ethylene oxide) with average molecular weight of 5 x 10⁶ Da. and density of 1.21 TM at 65 °C, and lithium trifluoromethane sulfonate 97% pure. The required amounts of lithium salt and PEO, to give an O:Li

ratio of 10:1, were dissolved in acetonitrile 99% pure under nitrogen atmosphere. The solution was then stirred at room temperature for 48 hours after which the solution was filtered to remove any undissolved materials, and stored in 14, 2 ml plastic containers and kept well isolated from the atmosphere.

The polymeric films were deposited using a drop coating technique and then dried by heating for 30 minutes in an oven at 50 °C.

4. Neural Switch Programming

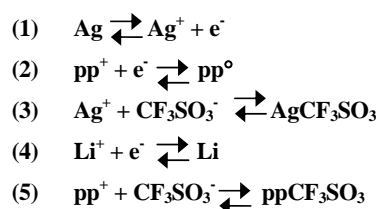
The Neural Switch is programmed by means of charge storage process, which proceeds as follows:

- (1) The source and drain are shorted together.
- (2) A biasing potential is applied between the shorted terminals and the gate.
- (3) After a specific time (5 minutes) the bias voltage is removed and the source and drain disconnected from each other.
- (4) The change in the device resistance is measured as the gate is left floating.

The above process was carried out over a range of positive and negative biasing potentials with the temperature kept constant at 25 °C using heating element under normal room atmosphere. The heating element was used not only to stabilize temperature at a certain value, but also to help testing the device at various temperatures. The typical programming cycle consisted of applying 0 volts up to +65 mV, and then back from +65 mV down to 0, then immediately after that a negative cycle started by applying 0 mV to -65 mV and back from -65 mV to 0.

4.1 Gate Programming

When the gate is programmed, a biasing potential is applied between the gate and the shorted source-drain terminals, then a constant current is injected to measure the change in the device resistance between the source and drain while the gate is to float. The following interactions are thought to occur:



(1), (2) and (4) accounts for the polarity of the gate bias which contributes to ionization of silver and oxidation/reduction of poly(pyrrole), while (3) and

(4) hypothesize cation/anion migration and the effective role that anions plays in the overall conduction process.

Programming of the neural switch was carried out over two gate polarities comprising different levels of biasing as follows:

4.2 Positive Programming Cycle

As positive bias is applied between the gate and source-drain, the following set of interactions would occur:

- (a) $Ag \rightarrow Ag^+ + e^-$
- (b) $CF_3SO_3^-(\alpha) \rightarrow CF_3SO_3^-(\beta)$
- (d) $Ag^+ + CF_3SO_3^-(\beta) \rightarrow AgCF_3SO_3$
- (e) $pp^+ + e^- \rightarrow pp^0$

From these reactions it is apparent that ionization, neutralization and reduction processes have taken place, resulting in an ionic-electronic exchange between Ag and CF₃SO₃, and reduction of poly(pyrrole). During the process CF₃SO₃ ions travel toward the gate due to the positive field set up by the bias while Li⁺ moves toward the shorted drain-source electrode.

4.3 Negative Programming Cycle

When negative bias is applied between the gate and the source-drain, the following set of reactions believed to have occurred:

- (a) $pp^0 \rightarrow pp^+$
- (b) $CF_3SO_3^-(\beta) \rightarrow CF_3SO_3^-(\alpha)$
- (c) $Ag^+ \rightarrow Ag$

4 Characterization of the Neural Switch

Figures 4-8 illustrate the characterization curves obtained for the neural switch. These curves cover biasing voltage from 0 mV up to 65 mV.

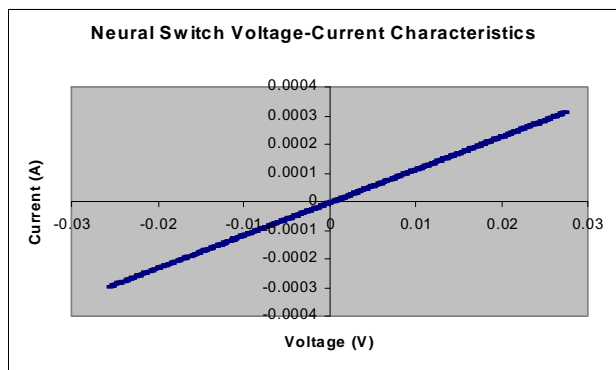


Fig.4: Voltage-Current characteristics of the Neural Switch at 0 mV gate bias.

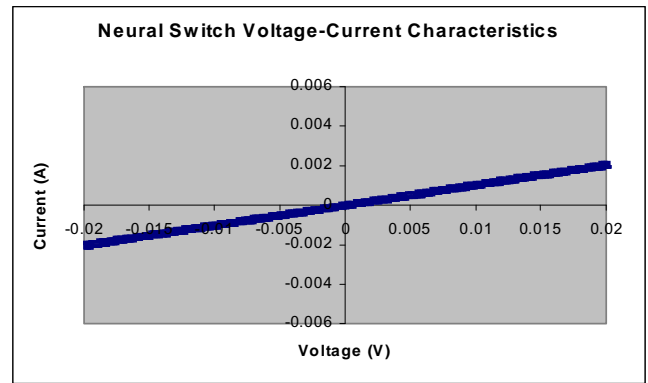


Fig.5: Voltage-Current characteristics of the Neural Switch at 20 mV gate bias.

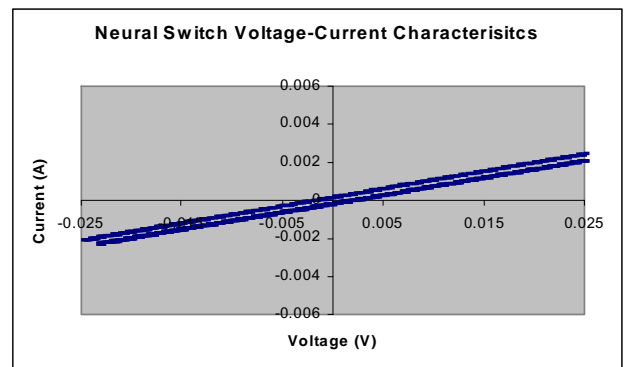


Fig.6: Voltage-Current characteristics of the Neural Switch at 40 mV gate bias.

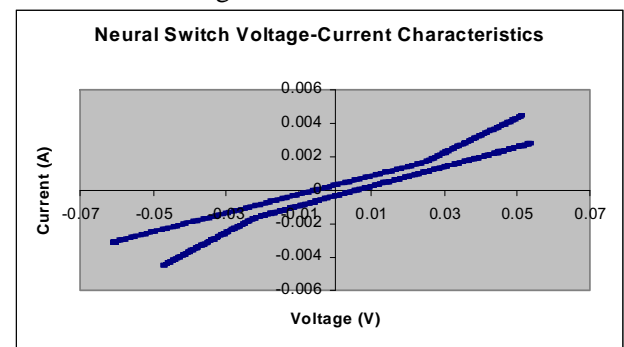


Fig.7: Voltage-Current characteristics of the Neural Switch at 50 mV gate bias.

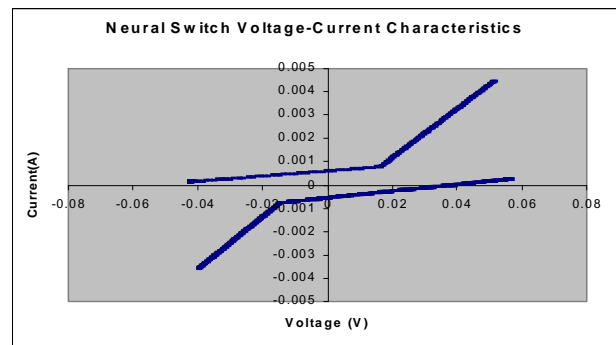


Fig.8: Voltage-Current characteristics of the Neural Switch at 65 mV gate bias.

5 Analysis, Discussion and Conclusion

Analysis of the designed and tested Neural Switch is achieved through an electrical circuit model employing a diode as the semiconducting device as shown in figure 9, while figure 10 represents an electronics circuit equivalent model for the device.

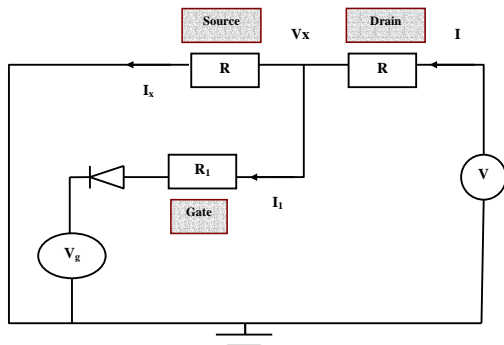


Fig. 9: Electrical circuit model for the Neural Switch

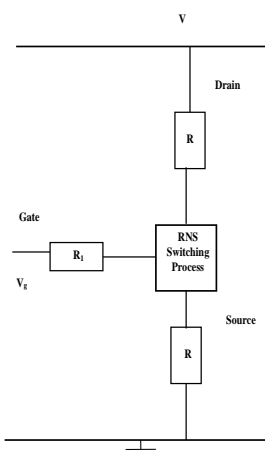


Fig.10: Electronic circuit model for the Neural Switch

In applying the model and on the assumption that both gate-drain and gate-source resistances are equally matched, two cases can be considered:

(1) $V_x \leq V_g$: The potential across the diode is either negative or zero, hence, it is not in its conducting state (reversed biased), which reduces the circuit to two resistances in parallel with each other giving voltage-current expression of the form:

$$(1) \dots I = \frac{V}{2R}$$

Equation 1 expresses an overall linearity of the I-V characteristic ($V_g \leq 20$ mV).

(2) $V_x > V_g$: The diode is in its conducting state (forward biased) and the circuit can be analyzed assuming that $(V - V_x) > 0$ and $(V_x - V_g) >$ diode barrier potential of ~ 0.6 volts.

Thus

$$(2) \dots I = \frac{V - V_x}{R}$$

But

$$(3) \dots I = I_x + I_1$$

So;

$$(4) \dots V - V_x = I_x R + I_1 R$$

From

$$(5) \dots I_1 = \frac{V}{R} - 2I_x$$

We obtain

$$(6) \dots V_x = I_x R = I_1 R_1 + V_d + V_g$$

Now substituting for I1

$$(7) \dots I_x R = \left(\frac{V}{R} - 2I_x \right) R_1 + V_d + V_g$$

So,

$$(8) \dots I_x (R + 2R_1) = \frac{VR_1}{R} + V_d + V_g$$

Then,

$$(9) \dots I_x = \left(\frac{VR_1}{R(2R_1 + R)} + \frac{V_g + V_d}{(2R_1 + R)} \right)$$

But

$$(10) \dots I = I_x + I_1 = \frac{V - V_x}{R} = \frac{V}{R} - I_x$$

Hence,

$$(11) \dots I = \frac{V}{R} - \left(\frac{VR}{R(2R_1 + R)} \right) - \left(\frac{V_d + V_g}{(2R_1 + R)} \right)$$

So;

$$(12) \dots I = \left[V \left(\frac{R + R_1}{R(2R_1 + R)} \right) - \left(\frac{V_d + V_g}{(2R_1 + R)} \right) \right]$$

If R is of unit value (R=1), then the value of I becomes:

$$(13) \dots I = V \left(\frac{R_1 + 1}{2R_1 + 1} \right) - (V_d + V_g) \left(\frac{1}{2R_1 + 1} \right)$$

As the diode in the equivalent circuit becomes saturated or unable to learn, the value of R1 will show a gradual increase proportional to the amount of exposure time to programming charge. Thus a power-like curve is produced as shown in figure 11.

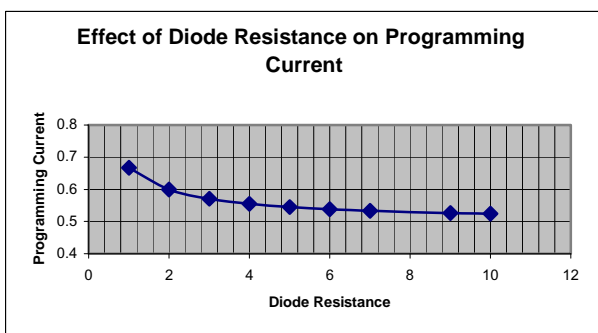


Fig. 11: Effect of gate resistance on programming current.

Two extreme cases are realized:

(1) $R_1 \rightarrow 0$

The programming current expression is reduced to:

$$(14) \dots I = \frac{V}{R}$$

(2) $R_1 \rightarrow \infty$

$$(15) \dots I = \left(\frac{V}{2R} \right)$$

These extreme cases are represented in table 1, and figure 12.

R	I(R1→0)	I(R1→∞)
1	1*V	0.5*V
2	0.5*V	0.25*V
3	0.33*V	0.167*V
4	0.25*V	0.125*V
5	0.2*V	0.1*V
6	0.167*V	0.083*V
7	0.143*V	0.0714*V
8	0.125*V	0.0625*V
9	0.111*V	0.0556*V
10	0.1*V	0.05*V

Table 1: Voltage-Current Relationship

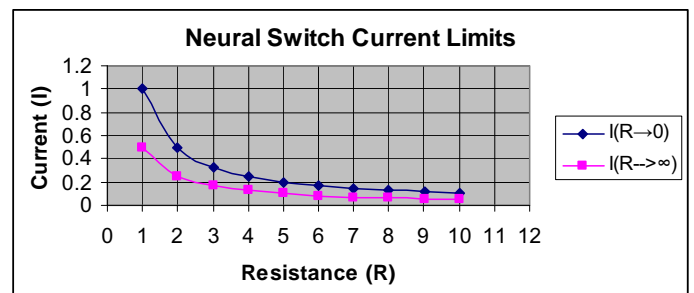


Fig.12: Resistance-Current Limits.

Based on the previous, the storage and switching ability of the designed Neural Switch is established. The Switch stores charges at a low or high resistance slope until the biasing value of the gate is changed, then it switches at the appropriate point to a different resistance slope to allow reprogramming of the switch to take effect.

In the process of testing our designed Neural Switch, other factors governing its performance and functionality were tested [33]. Such factors are:

- (1) The effect of electrode separation on the uniformity of the electrostatic field between the electrodes.
- (2) The importance of the central gap variation on the stability of the field lines and consequently on the conductance to film thickness relationships is studied.
- (3) Temperature effect on $\text{PEO}_{10}\text{LiCF}_3\text{SO}_3$ film as it moves from crystalline to amorphous phase is analyzed.
- (4) The observed behaviour of the Neural Switch, which was studied through complex impedance analysis, is thought to be due to the effect of $\text{PEO}_{10}\text{LiCF}_3\text{SO}_3$ capacitive component and Polypyrrole inductive component. This further supports the assumption of ionic-electronic interaction occurring between ppy and $\text{PEO}_{10}\text{LiCF}_3\text{SO}_3$.

The implementation of a system that includes such designed Neural Switch involves replacing a portion of the nervous system with an artificial device [30, 31, 32]. Aside from developing hardware and providing for its physical and physiological integration with the nervous system, this requires logical development which comprises the following:

- (1) Abstract model that shows what computations need to be done, independently of implementation.
- (2) Theory of how these computations are implemented in real neurons.
- (3) Technology for implementing them in an artificial computer.
- (4) Plan for logical integration of the neural and the electronic computations so that they jointly have the desired effect.

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