Memristors with Carbazole-Functionalized Organic Materials for AI-Driven Development: Unlocking Resistive Memory and Synapse-Mimicking Functionality for Next-Gen Computing

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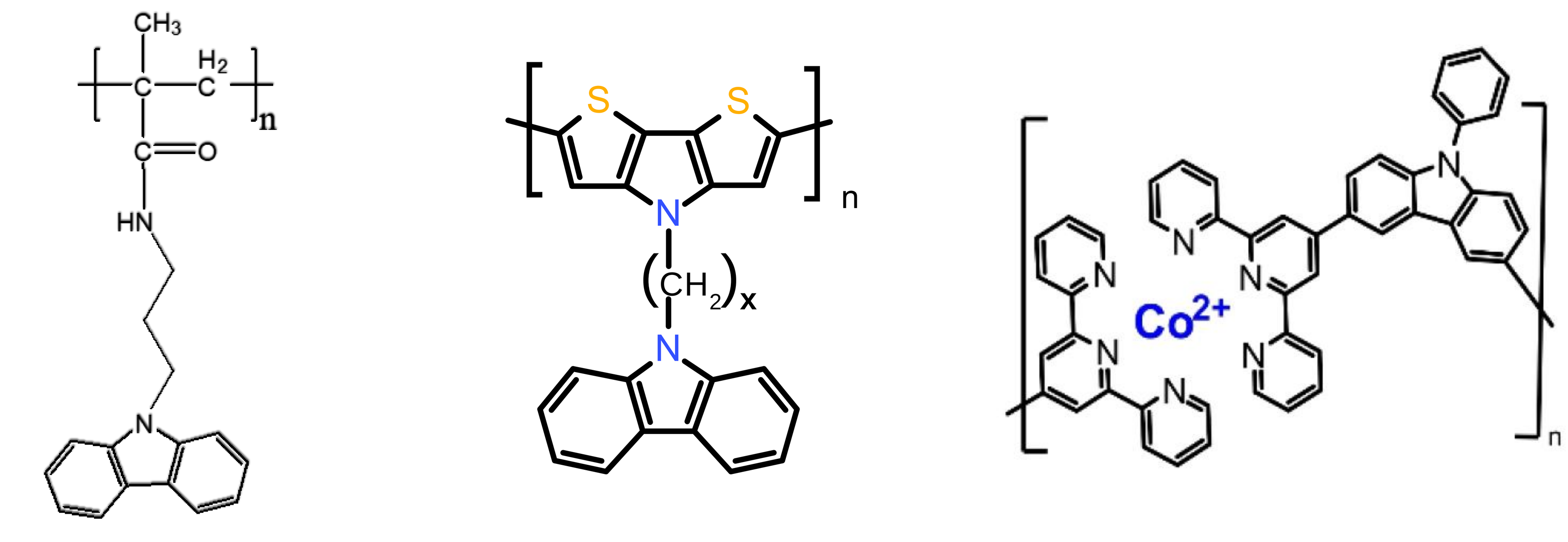
After successful designs and synthesis, the carbazole-functionalized polymers were characterized for memristor devices, along with their applicability in bistable memory and emulation of synaptic plasticity. These polymers integrates carbazole as the functional units into main chain or sidechain demonstrate voltage-induced conductance change when sandwiched between ITO and Al or Au electrodes, thus exhibiting memristive properties.[1, 2, 3] Devices display bistable conductivity and retains data for hours under electric fields above certain threshold. Under moderate fields, characterized by continuous trigger pulses, the devices exhibit conductance modulation akin to neuronal synapses, demonstrating functionalities like short-term and long-term memory, spike-timing-dependent plasticity, and associative learning. The underlying mechanisms, such as voltage-induced conformation changes, charge carrier trapping / detrapping, and redox phenomena that influences above-mentioned properties are thoroughly examined with relevant experimental evidences. Our findings suggest the potential application of these polymers in organic memory devices, artificial intelligence and neuromorphic computing.

Synthesized Set of Polymers under Study

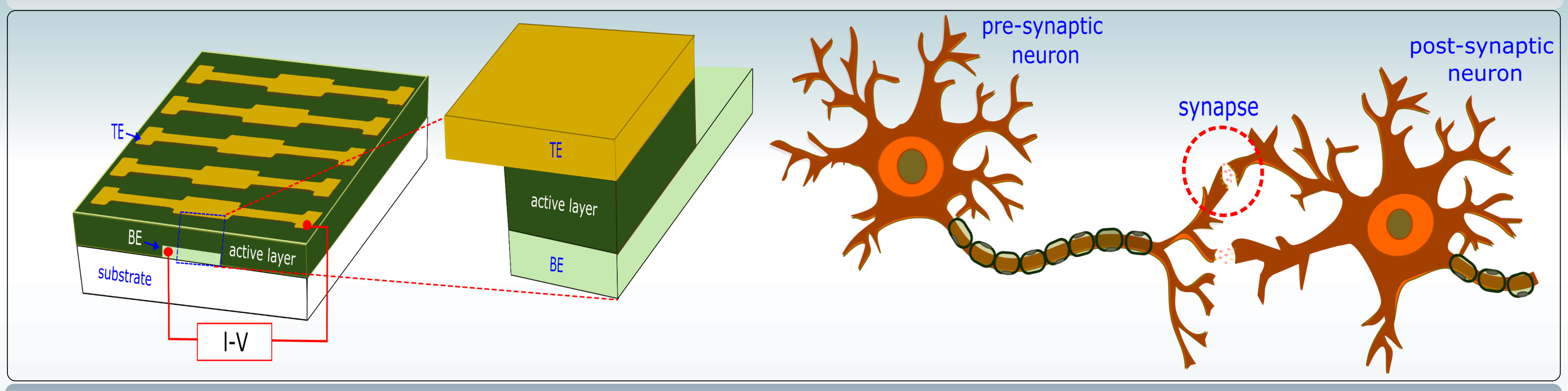


Poly-N-(3-(9*H*-carbazol-9-yl) propyl)methacryl amide **(PCaPMA)**

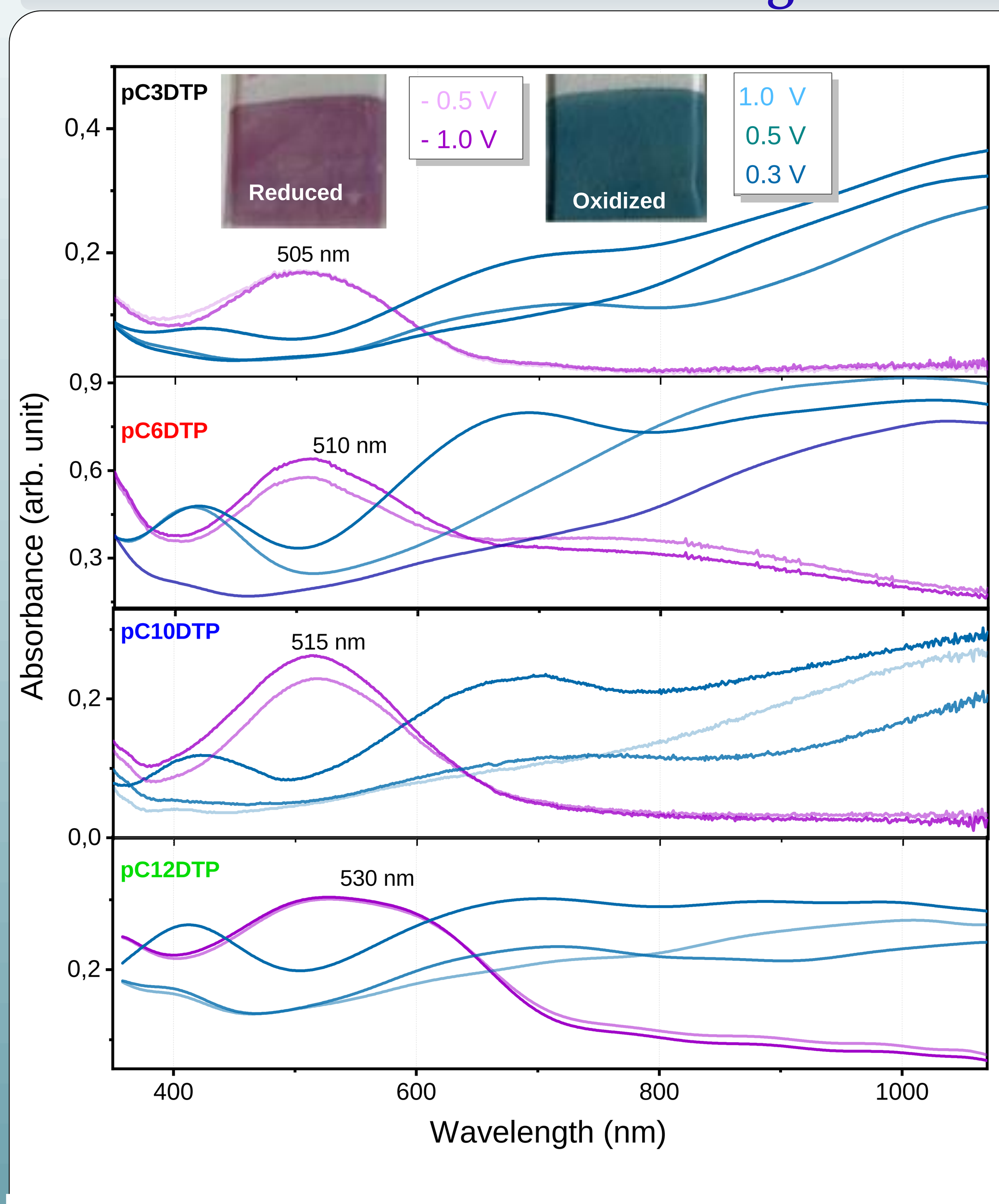
poly(4-(6-(9H-carbazol-9-yl)hexyl)-4H-dithieno[3,2-b:2′,3′-d]pyrrole) **(pC6DTP)**

Co^{2+} Coordinated polymers with bis(tpy)carbazole ditopic ligand **(coU)**

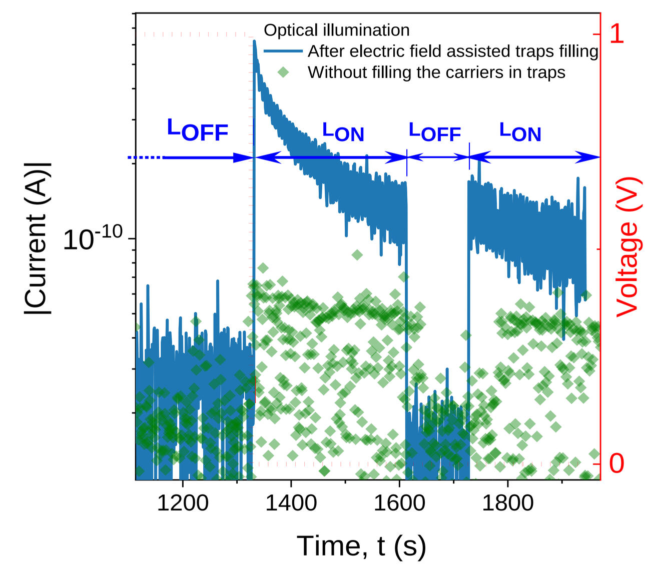
Schematic of the Memory Device and Neuronal Synapse



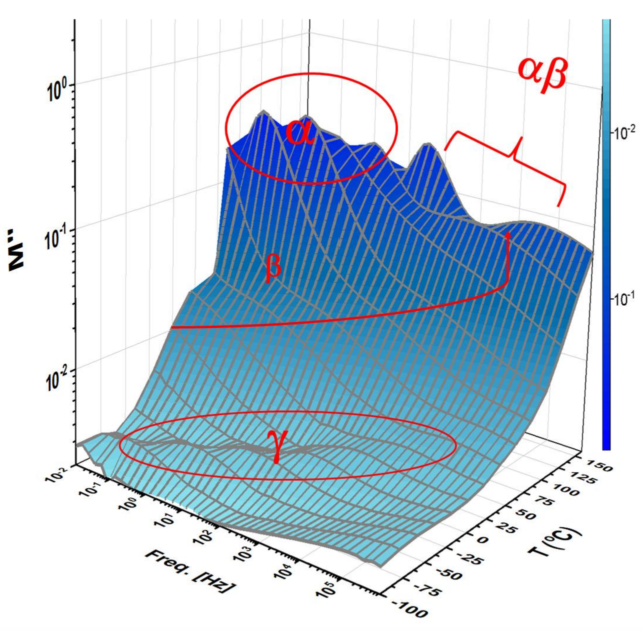
Working Mechanisms



Spectro-electrochemical (SEC) Measurement of pCxDTP samples



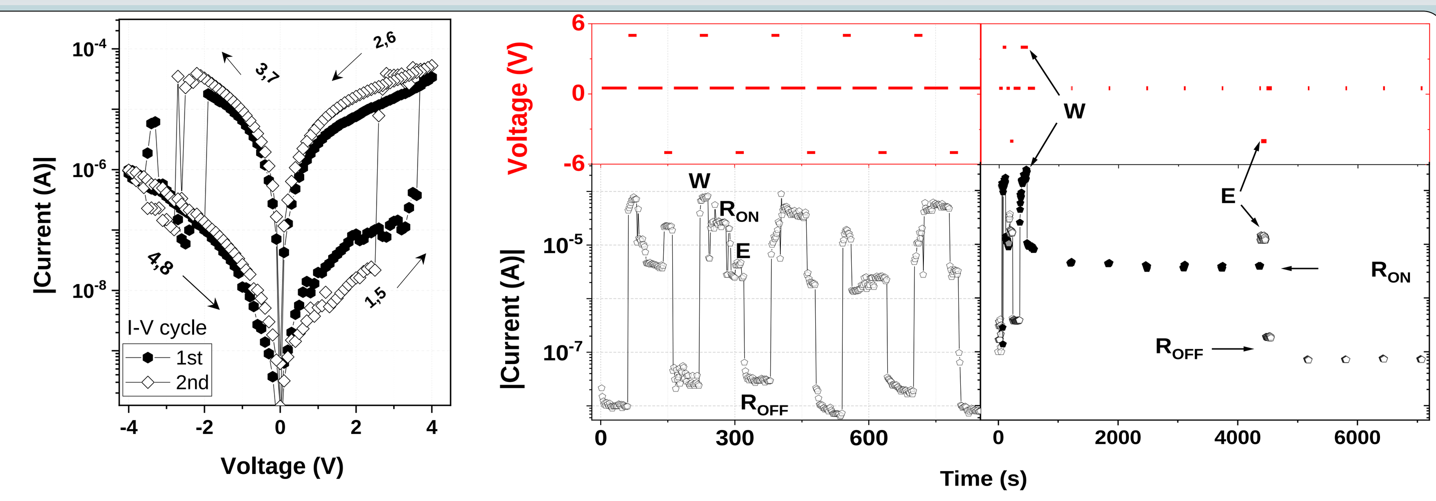
Optical Detrapping of Trapped Carriers



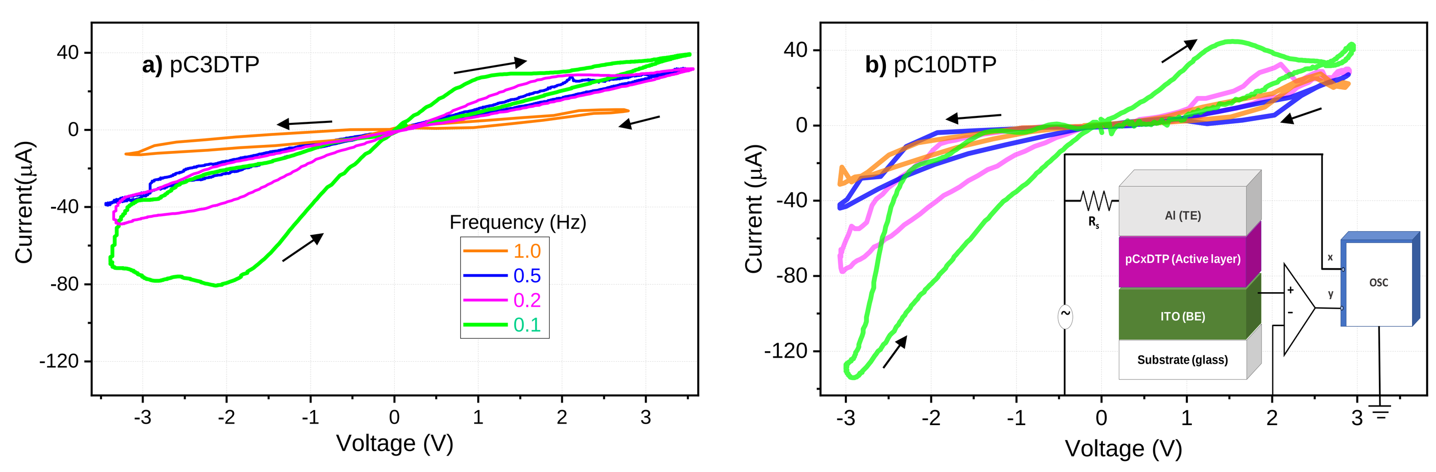
Relaxations in Loss Modulus

Conductance Switching, Binary Conduction and Non-volatile Memory

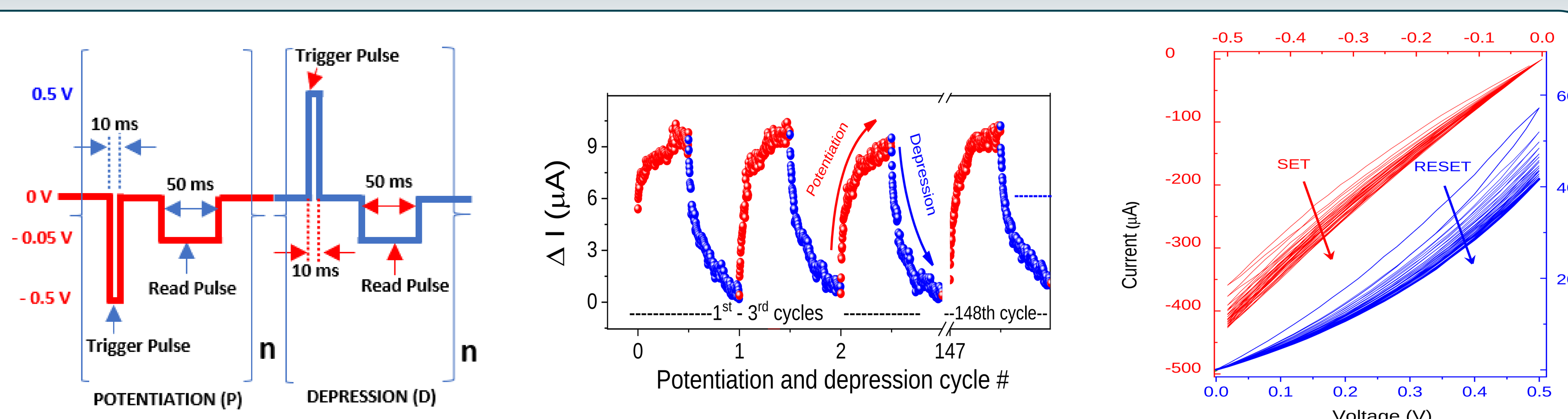
DC response

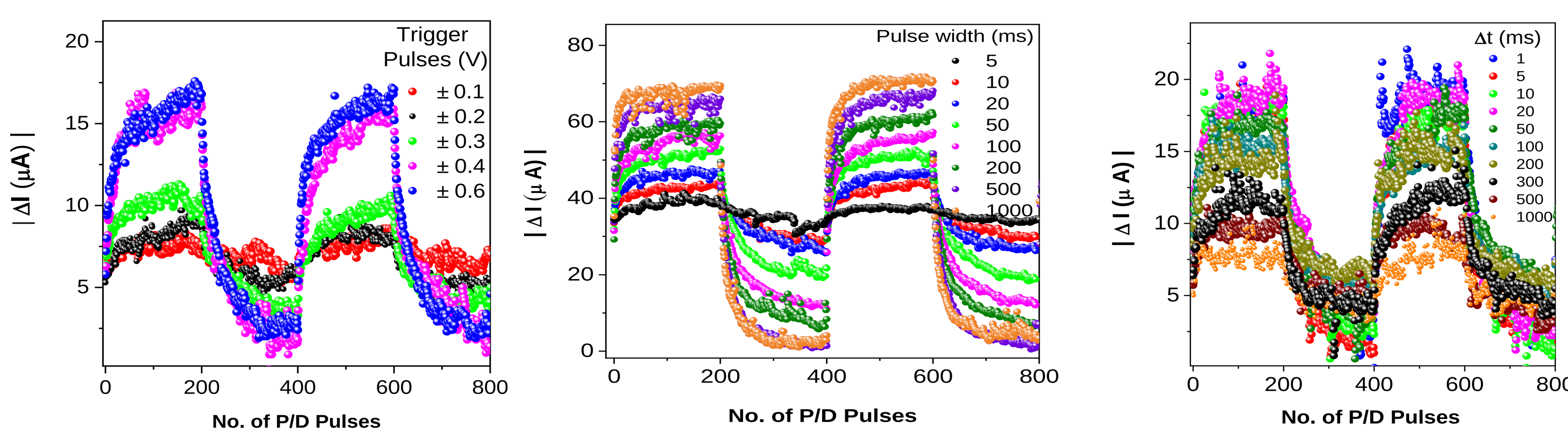


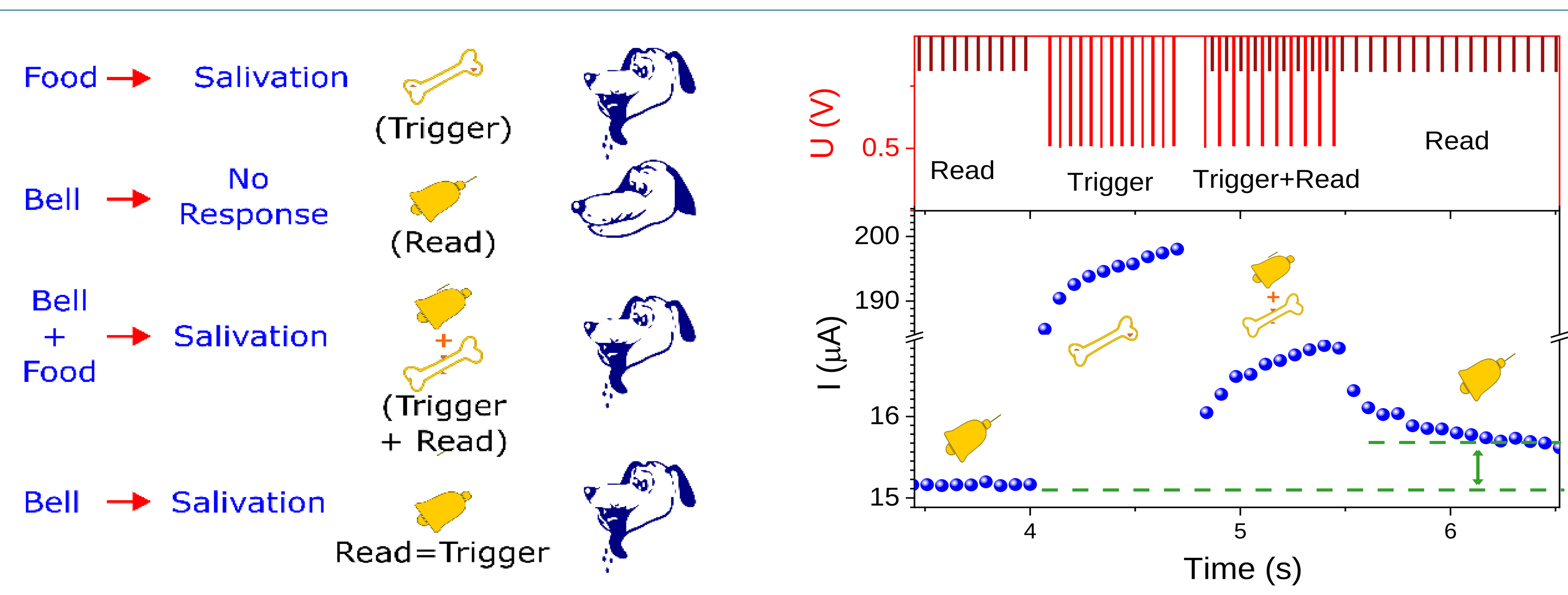
AC response



Analog Properties: Mimicking Synaptic Plasticity







Associative learning

Summary

- All studied polymers exhibited good solubility in many organic solvents and high thermal, ambient and photo stability.
- DC current-voltage (I-V) characteristics of sandwich device showed an abrupt switching and erasing above threshold voltages, with high ON/OFF current ratio and good nonvolatile behaviors. AC I-V characteristics showed strong frequency dependence and the response was dependent on structure of the materials used in the device.
- In the moderate field, the device exhibited analog conductance changes dependent on trigger pulse amplitude and frequency, mimicking biological synapse behavior. Trigger pulses of very low amplitude, ± 500 mV and short pulse-width, 10 ms, induced conductance changes, capable of being monitored by reading pulses of -50 mV. The device proved high reproducibility >150 cycles, mimicking synaptic potentiation and depression
- The device mimics both short-term and long-term memory/forgetting, and associative learning properties akin to **Pavlovian Conditioning** of dog.
- Device working mechanisms were elucidated with several experimental methods and theoretical analysis. Photodetrapping of trapped carriers in polymer thin film was carried out to find out the role of traps in conductance change. Space-charge-limited-current – voltage characteristics observed confirmed the role of traps in conductance switching.
- Broadband dielectric spectroscopic study showed three relaxations properties, γ- β- and α- relaxation in loss modulus (M′′), with γ-relaxation signifying the rotation of carbazole group responsible for conductance switching.
- The spectro-electrochamical study explores redox reactions in a polymer under varying electric field, revealing significant optical absorption changes linked to carbazole unit oxidation and reduction, influencing molecular conformation and device conductance.

References:

[1] Y. R. Panthi, J. Pfleger, D. Výprachtický, A. Pandey, M. A. Thottappali, I. Šeděnková, M. Konefał and S. H. Foulger, *J. Mater. Chem. C*, 2023, **11**, 17093–17105

[2] Y. R. Panthi, A. Pandey, D. Výprachtický, A. Šturcová, S. H. Foulger, J. Pfleger, under review in *J. Mater. Adv., RSC*, 2024.

[3] A. Pandey, A. Chernyshev, Y. R. Panthi, J. Zedník, A. Šturcová, M. Konefał, O. Kočková, S.H. Foulger, J. Vohlídal, J. Pfleger, Polymers 2024, 16, 542.

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