

# Development of ‘Artificial’ Memristive Synapses using Various $sp^2$ C (graphene-like) - $sp^3$ C (diamond) Heterojunctions as Neuromorphic Devices

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The integration of allotropic  $sp^2$ -/ $sp^3$ -bonded carbon ( $sp^2$ C/ $sp^3$ C) interfaces and devices has evoked increasing attention since they offer versatile and rich playground for carbon-based electronics, electrochemical sensing platforms, and optoelectronic neuromorphic computing attaining enhanced performance [1-5]. Moreover, inspired by human brain functionality and its low power consumption of only 10W, memristors for neuromorphic computing have gained significance for implementing solid-state neurons and ‘artificial’ synapses due to their nanoscale footprint and reduced complexity. In this talk, we present the the fabrication of various carbon-based heterojunctions comprising graphene-like ( $sp^2$ C)–diamond ( $sp^3$ C) architectures using microwave plasma-assisted chemical vapor deposition on nanodiamond seeded  $p$ -Si (100),  $SiO_2/p$ -Si (100) and  $Fz$ -Si (001) substrates. These are the key elements emulating the characteristics of biological synapses and memory functions which are game-changing energy saving computing devices. The resulting heterojunctions behave as memristors (*i.e.*, resistors with tunable memory) having multiple resistance states and nonvolatile memory functions, a phenomenon that refers to the ability of synapses (neuronal links) to adapt in response to an increased or decreased activity, essential to human memory and learning. The resulting heterojunctions behave as memristors (*i.e.*, resistors with tunable memory) having multiple resistance states and nonvolatile memory functions, a phenomenon that refers to the ability of synapses (neuronal links) to adapt in response to an increased or decreased activity, essential to human memory and learning. We performed  $I$ - $V$  characteristics with temperature (up to 250 °C) and in response to photoirradiation at 365 nm, 532 nm, and 633 nm in addition to comprehensive microstructural properties. Interestingly, high or low resistance states (equivalently, short-term and long-term potentiation) can be controlled by combined applied bias and light irradiation, giving a resistive switching ratio of  $\sim 10^6$ , observed in sparse materials and/or heterostructures. They exhibit quasi-linearity and symmetry when subjected to identical input pulses, essential for their role in online training of neural networks. The linearity holds for a range of pulse width, amplitude and applied pulse number. We ascribe the observed behavior to redox reaction (or reorganization of carbon orbitals) at the  $sp^2$ - $sp^3$  interfaces and the role of hydrogen and oxygen movement by applied bias. Finally, heterostructure arrays could be designed for better memristive devices and memory functions and photo sensing (image sensors) applications.

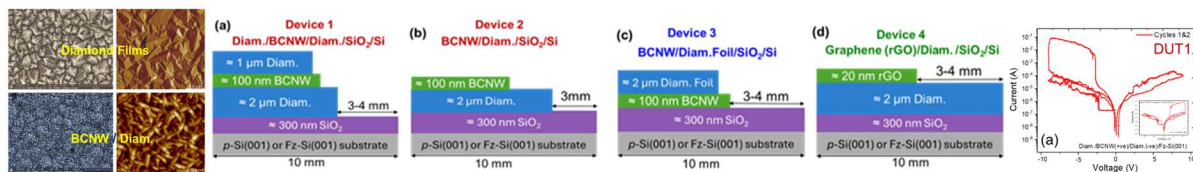


Figure 1. Schematic of Diamond films, diamond foil, and BCNW synthesis using MWCVD method [7]. (a-d) Heterojunction device architectures showing various diamond-graphene-like interfaces. Also shown is  $I$ - $V$  characteristics measured upon irradiation at  $\pm 8.5$  V for 15s with UV light showing the junction change between HRS and LRS state combined with applied bias.

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### Supplementary information:

We have also characterized these interfaces using advanced electrochemistry at solid-liquid interfaces. The hierarchical features constituted by complex morphology defined with microcrystallite diamond grains intertwined with vertically aligned BCNW as thin layer revealed using electron microscopy complemented with structural, electrical, and electrochemical properties such as activation energy ( $E_A$ ), electron transfer rate ( $k_{\text{eff}}$ ) and redox potential shifts ( $\Delta E_p$ ). The hydrogen plasma during deposition plays an effective role in the transformation of  $\text{sp}^2\text{C} \leftrightarrow \text{sp}^3\text{C}$ , eventually leading to various complex morphologies. While the flatband potential and hole-acceptor carrier concentration were estimated using Mott-Schottky relationship, the fabricated hybrid carbon interfaces exhibited electroactivity toward ferro/ferricyanide redox couple. The redox peak separation value ranged between 112–149 mV (and 67–71 mV) for all the samples studied with both redox probes and the electron transfer rate was determined using different analytical procedures. The experimental findings are ascribed to graphitic  $\text{sp}^2\text{C}$  pathway paired with surface conductive channel of H-terminated diamond films surface for electrons transportation and their robust nature. This work promotes the development of high-performance electrocatalytical, photoelectrochemical platforms based on most known hybrid allotropic carbon interfaces and the method proposed here also provide an effective strategy to construct diamond - graphene junctions for diverse applications.