High-Throughput Solution-Processed Graphene Transistors: Enabling sub-Femtomolar Point-of-Care Immunosensing and Ultra-Low Power Synaptic Neuromorphics

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Abstract

Utility of graphene in nanoelectronics is proclaimed due to its unparalleled electron mobility, atomically thin structure, and bandgap tunability which enables ultra-fast, low-power devices at the nanoscale. In context of graphene-based transistors be it field-effect (FET) or electric double layer (EDLT), chemical vapor deposition (CVD)-derived graphene has been favoured for its superior quality, reliability, and reproducibility. However, its specialized instrumentation requirements like along with high-temperature and vacuum necessities renders it costly and impractical for nanoelectronics sensors. Despite advancements, protocols like liquid-phase exfoliation (LPE) are known to induce polydispersity in graphene nanosheets leading to huge device-to-device variability, originating from randomized junction resistances and erratic baseline response that hinders demonstration of reliable graphene-based transistors. Herein, we demonstrate two distinct works wherein graphene obtained from high-throughput production methods like shear-mixing is utilised and in order to keep the fabrication cost to a minimum spray printing is employed. Firstly, a multi-parametric machine learning (ML) approach is demonstrated on a multiplexed few-layer graphene (FLG) transistor array, wherein a unique feature amalgamation encompassing steady-state and transient features have been employed as a quantitative descriptor of Traumatic Brain Injury (TBI) biomarkers (GFAP and S100B) and utilised for model training. The drain current change (M), settling time (S), noise density (N), and frequency roll-off-rate (R) are identified as the optimal amalgamate of parameters, yielding the highest classification accuracy of ~93% and quantification accuracy of 90% at 0.5 fM. Secondly, we demonstrate industrially integrable high-shear mixing of bulk graphite to yield few-layer graphene inks, further facilitated by integration with industry-standard spray printing for channel fabrication. By electrolytic gating of the fabricated transistors, we utilise the dynamic spatio-temporal iontronics for graphene doping modulation to exhibit robust emulation of synaptic responses including spike-rate-, duration-, number-, and frequency-dependency in both excitatory and inhibitory regimes. Furthermore, the devices demonstrate exceptional cyclic endurance with <5% deviation even after 10,000 cycles and ultra-low power consumption of ~1 pJ which is reported for the first time for solution processed graphene. Employment of phosphate-buffer-saline (PBS) as the electrolytic medium emphasizes viability for bioelectronic immunosensing, with electrochemical impedance measurements (EIS) unravelling ion-specific interaction-driven conductance modulation. Finally, a cyberphysical system is demonstrated which integrates these devices for on-site human face recognition, achieving high-fidelity neuromorphic processing at ~1 pJ energies per event. This high-throughput ink formulation, coupled with spray printing, enables adoption of 2D material-based neuromorphic and PoC sensors, mitigating fabrication bottlenecks and fostering scalable, low-power hardware for energy-efficient AI and bioelectronics.

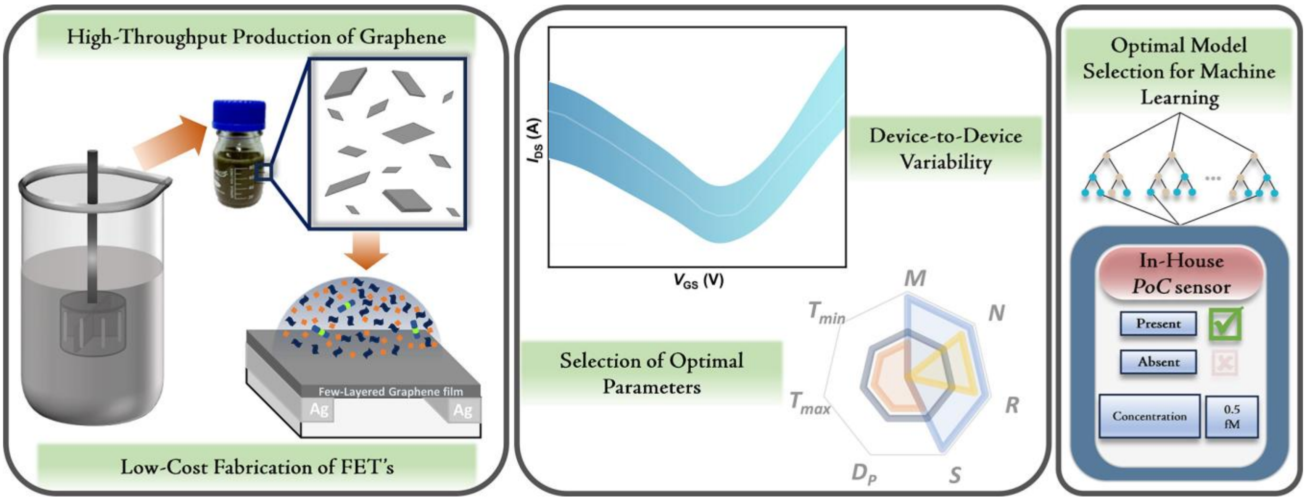

Figure 1: A multi-parametric machine learning (ML) approach on few-layer graphene transistor array, wherein a unique feature amalgamation encompassing steady-state and transient features have been employed as a quantitative descriptor of Traumatic Brain Injury (TBI) biomarkers (GFAP and S100B).

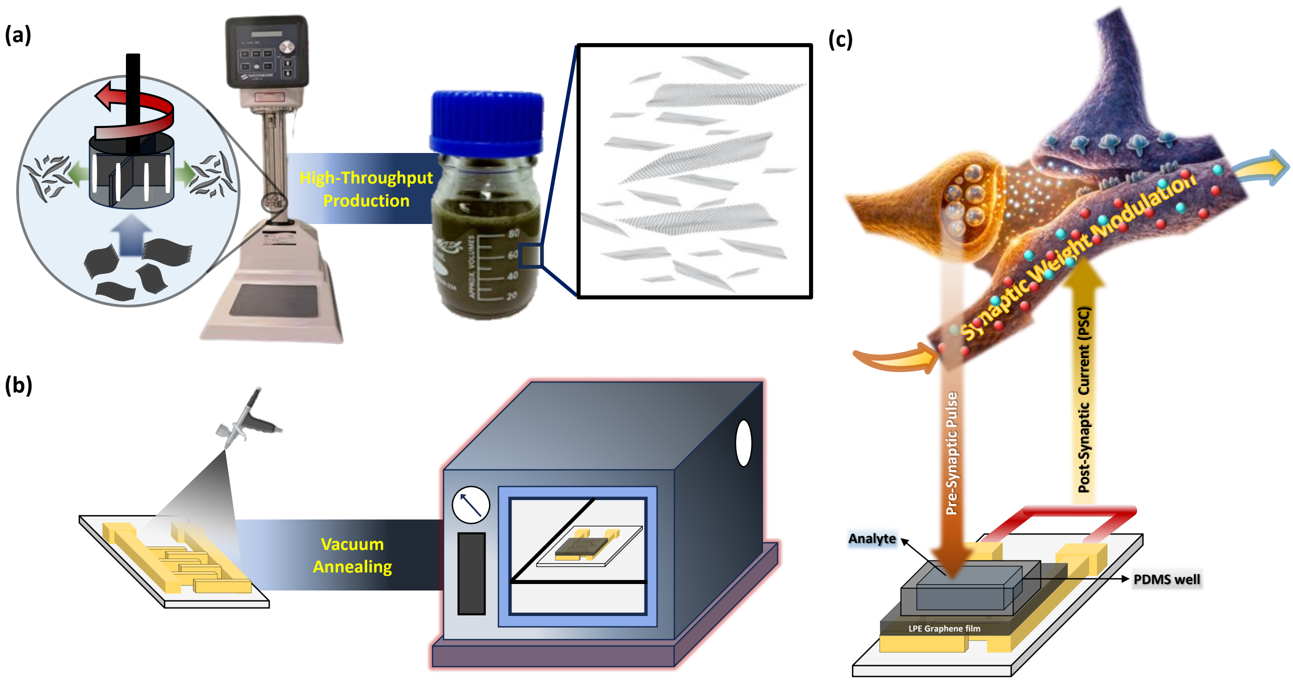

Figure 2: (a) High-throughput graphene ink production, (b) Spray coated graphene transistors, and (c) Is a schematic representation of the emulation of biological synapse by the fabricated transistor.