Closed-Loop Control of Microgel-Based Biohybrid Synapses

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Over the last decades, there has been a growing research interest in organic neuromorphic devices. By mimicking parts of the human nervous system, these devices enable a better understanding of the highly efficient information processing techniques that have naturally arisen through evolution - efficiencies that remain unmatched by technology to date [1]. Organic electrochemical transistors (OECTs), typically based on organic mixed ionic electronic conductors (OMIECs) as their active channel material, such as PEDOT:PSS, have proven to be ideal candidates, as their architecture and operating principles efficiently and inherently mirror the biological chemical synapse [2]. Specifically, the presynaptic transistor gate drives an ionic flux through the electrolyte - acting as the synaptic cleft - which ultimately modulates the transconductance of the postsynaptic transistor channel [1].

Although current neuromorphic platforms already can biomimetically emulate synaptic short and long-term plasticity (STP and LTP) *via* the slow kinetics of ion movement and the oxidation of dopamine (DA) [3-4], mechanisms as neurotransmitter homeostasis and local learning still remain little investigated. Therefore, this project aims to emulate the exocytosis-based neurotransmitter release from the presynaptic terminal by engineering PEDOT:PSS-based microfabricated OECTs.

Here, synaptic vesicles are implemented by synthetic stimuli-responsive microgels loaded with DA and located on the gate terminal. The microgels based on vinylcaprolactam (VCL) can release previously loaded small molecules upon stimulation (temperature, pH, light) *via* the reversible disruption of hydrogen bonds [5]. This mechanism enables them to be used as both reservoirs and dispensers for the neurotransmitter. Analysis methods comprising NMR, FTIR, Raman spectroscopy and DLS were used to confirm the successful sythesis of the functional microgels. Further, the spin-coated microgel-PEDOT:PSS interface was morphologically and electrochemically investigated using AFM and EIS, revealing successful particle deposition and an altered impedance of the modified interface.

By implementing a supervised or unsupervised local learning strategy (e.g., Hebbian learning or spike-timing dependent plasticity (STDP)) - an on-chip control algorithm later will promote DA homeostasis and dynamically tune the synaptic weight through the regulated release and oxidation of DA, as well as the antagonistic redoping of the PEDOT:PSS channel. Functioning as an artificial analogue to the dopamine transporter (DAT), this redoping mechanism mimicks the reuptake of DA and can be carried out by applying gate-source voltage pulses or by utilizing a microfluidic setup to wash the channel with hydrogen peroxide H_2O_2 [4].

Ultimately, this work aligns with broader research initiatives whose overarching goal is to technically implement fundamental concepts of highly efficient biological systems, thereby paving the way for the native integration of such technology into advanced biohybrid applications and sophisticated neuroelectric interfaces.

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