Tailoring Ionic Dynamics in Organic Neuromorphic Devices based on PEDOT-Crown

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Neuromorphic hardware translates concepts from biology into electronic circuits to enable more efficient, biologically inspired computing. In biological neuronal systems, the transfer and processing of information fundamentally rely on ionic signals. Materials capable of both ionic and electronic conduction, known as Organic Mixed Ionic Electronic Conductors (OMIECs), have enabled the development of Organic Neuromorphic Devices (ONDs), such as Organic Electrochemical Transistors (OECTs).[1] These devices utilize the ion-driven modulation of the electronic doping state of an OMIEC-based conducting channel in contact with an electrolyte to emulate synaptic plasticity as well as other neuronal functionalities.[2]

A key challenge in OECT-based neuromorphic devices is their fundamental dependence on intrinsic ionic transport dynamics, which govern switching speed, threshold behavior, plasticity timescales, and retention. Recent studies have improved the understanding of the underlying mechanisms and introduced novel materials that enhance OND performance.[3] Here, we investigate a previously developed Crown-ether functionalized OMIEC, PEDOT-Crown, as a channel material in OECTs. PEDOT-Crown has been shown to exhibit cation-responsive electrochemical properties, which makes it particularly interesting for engineering ion-transport-dependent functionalities in OECTs.[4]

For this, PEDOT-Crown transistor channels were electrodeposited onto gold interdigitated electrodes using specifically optimized potentiostatic conditions. PEDOT-Crown was systematically compared with pristine PEDOT across multiple electrolyte compositions using Electrochemical Impedance Spectroscopy and Cyclic Voltammetry, revealing differences in electrochemical properties originating from the crown ether modification. Furthermore, steady-state and transient OECT characterizations demonstrated improvements in device performance, including reduced threshold voltage and altered plasticity dynamics.

This work demonstrates the potential of PEDOT-Crown as a channel material for OECT-based neuromorphic devices, showing that crown-ether functionalization can be used to engineer ionic dynamics and improve device performance. Additionally, the well-known biocompatibility of PEDOT and the low operating voltages of OECTs make this platform a suitable option for future biohybrid interfacing applications.

[1] S. Inal, *et al.*, *Acc. Chem. Res.*, 51, S. 1368-1376, 2018

[2] J. T. Friedlein, *et al.*, *Org. Electron.*, 63, S. 398-414, 2018

[3] P. R. Paudel, *et al.*, *J. Mater. Chem. C*, 9, S. 9761-790, 2021

[4] C. Kousseff, *et al., J. Polym. Sci.,* 60, S. 504-516, 2022