4D-STEM and EELS Analysis of Complex C-based Sensor Architectures

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Background
The transformation of precursors with high carbon content into open-porous carbon foams and coatings via thermal processes is gaining traction as a promising and eco-friendly method for a range of applications including catalysis, energy utilization, and sensing [1]. In this investigation, we employ a one-step laser patterning technique to locally pyrolyze doctor-bladed ink coatings on flexible PET substrates. This process yields highly porous and intricate CO2 sensor architectures with a thickness of around 50 µm. The ink composition includes glucose as a pore-forming agent and adenine as a nitrogen source, contributing to the sensing capabilities. Laser treatment in an oxygen-rich environment results in flexible and highly porous sensor structures with distinct nitrogen and oxygen functionalities within different regions. The laser intensity varies with depth, leading to the creation of a well-defined highly porous graphitic surface layer (acting as the electric transducer layer) and a less porous nitrogen-rich lower sensor layer, interconnected by a thin transition zone [2].

Methods
To understand the formation of these structures and their relationship with functionality, we conducted a thorough TEM investigation on cross-sections (0.25 - 0.5 t/λ) of the entire device prepared by microtomic cross-sectioning.

Results & Conclusion
STEM-EELS elemental distribution maps reveal a clear distinction in chemical composition between the upper and lower layers of the sensor. Principal component analyses were utilized to unravel the complex sensor structures, comprising various crystalline and amorphous phases containing carbon and nitrogen. 4D-STEM analysis unveils the distribution of the crystalline graphitic phase and the alignment of graphite basal planes relative to the pore walls of the open-porous sensor. Understanding these parameters is crucial for deciphering the electrical performance of the sensor (refer to Figure 1) [3].

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Graphic:

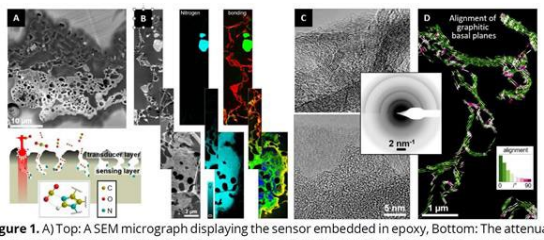

Figure 1. A) Top: A SEM micrograph displaying the sensor embedded in epoxy, Bottom: The attenuation of thermal laser intensity varying with depth. B) Left: A HAADF-STEM micrograph revealing distinct sensor layers, Middle: A STEM-EELS nitrogen distribution map, Right: PCA bond analysis. C) HRTEM images of the sensor from the transducer (top) and sensor (bottom) layers with corresponding SAED image of respective ROIs (inset). D) 4D-STEM analysis illustrating the misalignment angle between the normal of the graphitic carbon basal plane and the pore surface normal.

Keywords:

4D-STEM, open-porous carbon, laser pyrolysis

Reference:

[1] M. Hepp, et al. npj Flexible Electronics 6, 1-9 (2022)
[2] H. Wang, et al. Advanced Functional Materials 32, 2207406 (2022)
[3] C. Ophus, et al. Microscopy and Microanalysis 28, 390-403 (2022)