Enhanced Catalysis in Wrinkled Graphene Revealed by Multimodal *in-situ* Microscopy

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Introducing curvature into graphene's typically inert lattice can fundamentally reconfigure its electronic structure [1], unlocking new catalytic behaviors without the need for metal doping [2]. Despite theoretical predictions of strain-driven reactivity, direct spatial correlations between surface topography and catalytic performance have remained largely unresolved [3]. In this work, we present a systematic investigation of wrinkle-engineered graphene, where pre-stretched elastomer substrates guide the formation of periodic, well-defined wrinkle arrays upon relaxation of CVD-grown films.

We employ a multimodal *in-situ* characterization approach to unravel the interplay between curvature, local structure, and electrochemical activity. High-resolution scanning electrochemical cell microscopy (SECCM) reveals linear zones of elevated oxygen reduction current, precisely aligned with the wrinkle crests. Concurrent electrochemical atomic force microscopy (EC-AFM) captures nanoscale variations in both topography and interfacial impedance, indicating that catalytic enhancement correlates more strongly with geometric curvature than with defect density.

Kelvin probe force microscopy (KPFM) shows a localized decrease in work function along the wrinkled ridges, consistent with curvature-induced shifts in Fermi energy favoring O_2 adsorption. Raman spectroscopy confirms the presence of uniaxial tensile strain, reaching up to 2%, while tip-enhanced Raman spectroscopy (TERS) provides sub-30 nm resolution of sp^2 to sp^3 hybridization transitions at highly curved sites. Across thousands of data points, we establish a quantitative relationship between curvature and catalytic response, with wrinkled graphene exhibiting up to 10× higher limiting currents than flat controls and sustaining this activity over extended periods of time.

Our study delivers the first spatially resolved strain–activity map for graphene electrocatalysts, highlighting the potential of curvature as a design parameter. These insights provide comprehensive guidance for the development of next-generation 2D materials in sustainable energy applications.

References

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