Pore-confined ToF-SIMS electrochemistry

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Electrode-electrolyte interfaces govern charge transfer, mass transport, solvation, adsorption, and structural evolution in electrochemical energy systems. However, the dynamic molecular processes remain difficult to probe under operando conditions. Pore confined time-of-flight secondary ion electrochemical mass spectrometry integrates ToF-SIMS with a vacuum-compatible electrochemical microfluidic cell, enabling direct interrogation of liquid electrochemical interfaces under operando conditions. By stabilizing a nanoscale liquid-vacuum interface within a confined aperture, this strategy enables real-time acquisition of mass-resolved molecular information from the electrode surface during electrochemical reactions. With high surface sensitivity, mass resolution, and spatiotemporal resolution, this technique enables the identification of transient intermediates, reaction products, solvated species, spectator ions, and electric-double-layer evolution. Representative studies demonstrate its capability to probe molecular transformations, proton-coupled electron transfer, ion-solvent interactions, electrocatalytic pathways, and solid-electrolyte interphase formation in battery systems. This presentation will highlight the working principle, analytical capability, and representative applications of pore confined ToF-SIEMS, while outlining current challenges in device design, vacuum stability, interfacial sampling, and quantitative interpretation, as well as future opportunities in electrochemistry,

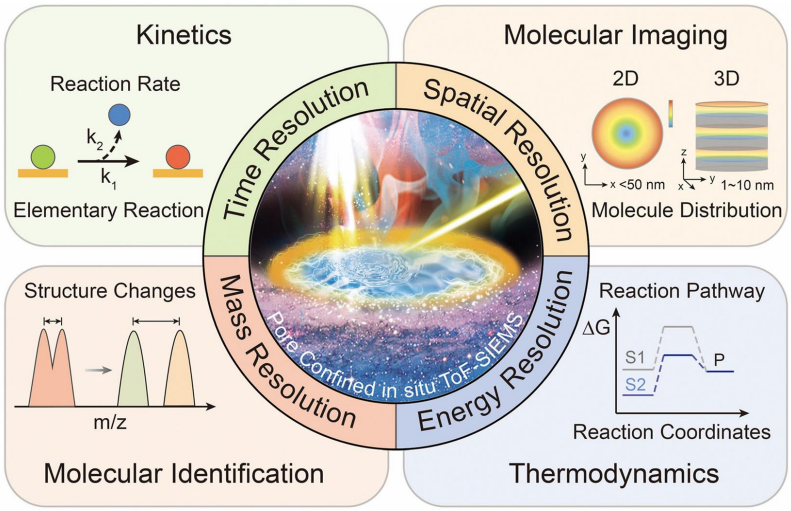

Figure 1. Concept of ToF-SIEMS probing the EEIs through the pore confined configuration.

energy conversion, bioanalysis, and chemical sensing.

[1] X. Hua, H.-L. Xia and Y.-T. Long, Chem. Sci., 2019, 10, 6215–6219.
[2] J.-G. Wang, R.-J. Yu, X. Hua and Y.-T. Long, Chem. Soc. Rev., 2023, 52, 2596–2616.