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Advanced architectures of electrochemical interfaces



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Communications Chemistry is delighted to introduce a Collection of articles on the intricate subject of electrochemical interfaces. Here, the Guest Editors introduce the topic, outline some of the themes covered by the Collection content, and look to the future of the field.

"The interface is the device" was coined by Nobel laureate Herbert Kroemer. Electrochemistry is the science of interfaces, and interface issues are prevalent in various energy storage devices. These include physical, chemical, and electrochemical interface problems commonly encountered, such as electrode fracture and pulverization during cycling; loss and failure of physical contact surfaces; decomposition of the solid–electrolyte interface (SEI); charge accumulation and mutual diffusion of elements at the solid-state electrolyte (SSE) interface; dendrite growth; lattice mismatch, among others. These challenges pose significant obstacles to the performance enhancement and reliable operation of energy storage/catalytic devices. Due to the lack of comprehensive understanding of the dynamic changes at interfaces and the complex mechanisms of electrochemical reactions, the foremost factor towards achieving sustainable and efficient electrochemical devices lies in constructing stable interfaces.

Collection themes

The concept of wettability was first introduced by Thomas Young in 1805. The wettability at the interface between the electrode and electrolyte determines the degree and efficiency of their interaction, which significantly influences the electrochemical storage and conversion efficiency of materials. Developing a profound understanding and effective regulation of electrode wettability have become pivotal research directions for enhancing the performance of energy storage devices. For example, chemical treatment of substrate interface wettability and conductivity can enhance the capacitance (<https://doi.org/10.1038/s42004-022-00719-y>)¹. In metal-based batteries, wettability affects nucleation overpotential, nucleation sites, dendrite formation and growth,

as well as the composition and constitution of the interfacial layer (<https://doi.org/10.1038/s42004-024-01350-9>)².

Throughout their lifecycle, the degradation of electrochemical devices is often based on the interactions and interlocking dependencies among various mechanical, physicochemical, and electrochemical degradation mechanisms. The relevant significance of these influencing parameters evolves, making the detection of real aging mechanisms and the establishment of predictive models particularly challenging. The growth, rupture, and repair of the SEI are primary mechanisms contributing to battery aging. In practical applications, commonly selected parameters such as different solvents, salts, electrolyte concentrations, and water content can be utilized to evaluate the formation and long-term evolution of SEIs (<https://doi.org/10.1038/s42004-024-01381-2>)³.

The materials and interfaces involved in the field of electrochemistry possess various structures, including crystalline, amorphous, and glassy states, and often contain a significant number of light elements. These materials exhibit high chemical reactivity and are sensitive to impurities, air, and electron irradiation, making it challenging to obtain authentic and accurate structural information about the samples. The SEI film is highly complex, thin, and characterized by a small gradient in electron/ion concentration. Achieving accurate electrochemical reactions often necessitates specialized designs for structure, composition and space charges. For example, the conductive properties of hydrogenated diamond-based electric double-layer transistors have been utilized to evaluate the electric double-layer effect and the suppression at the interface between solid electrolytes and electronic materials (<https://doi.org/10.1038/s42004-021-00554-7>)⁴.

The development of the emerging anode-less lithium metal solid-state battery (ALMSSB) technology requires an understanding of the reactivity between current collectors and solid electrolytes. Therefore, it is necessary to optimize the interface that forms during charge and discharge processes. For example, study of the reactivity of various current collectors with a solid electrolyte enables identification of metals that

lead to good stability versus metals that lead to more pronounced degradation phenomena, enabling optimized design (<https://doi.org/10.1038/s42004-025-01609-9>)⁵. The emergence and innovation of more sophisticated characterization techniques have provided powerful tools for exploring the physicochemical changes at interfaces. Cryo-electron microscopy (cryo-EM) offers the potential to resolve the composition and spatial arrangements of SEI components at the atomic level (<https://doi.org/10.1038/s42004-021-00521-2>)⁶. However, the interface not only involves solid–solid interactions; the formation and participation of gases (such as O₂, CO₂, and H₂) during electrochemical reactions contribute to the evolution of solid–gas and liquid–gas interfaces, which remains poorly understood. Time-of-flight secondary ion mass spectrometry (TOF-SIMS) can determine the chemical composition and morphology of SEIs through the control of the type of sputtering ions, allowing for depth profiling and compositional analysis (<https://doi.org/10.1038/s42004-025-01426-0>)⁷. Solid-state nuclear magnetic resonance (ss-NMR) is useful for probing the chemical environments of target atomic nuclei, providing rich information on ionic diffusion dynamics and complex electrochemical reaction mechanisms, with higher spatial resolution. This technique can also assess the impact of interfacial changes on ionic transport within lithium aluminum titanium phosphate (LATP) electrolytes (<https://doi.org/10.1038/s42004-025-01505-2>)⁸.

Electrochemical analytical methods hold advantages such as their non-destructive nature, ease of operation, and high efficiency, and are widely used for assessing interfacial behavior. Techniques such as electrochemical impedance spectroscopy (EIS), incremental capacity analysis (ICA), and differential voltage analysis (DVA) help obtain information on the local-scale transfer of electrons and ions at the interface, quantifying the contributions of electrode and interfacial processes. Mechanical instability at the interface and byproducts can also be monitored by developing new characterization techniques. For example, spectroscopic ellipsometry (SE) can characterize the accumulation and depletion layers within SSE; combined with theoretical models, it can elucidate the physical properties of

the space charge layer (<https://doi.org/10.1038/s42004-023-00923-4>)⁹.

Outlook

Significant progress has been made in advanced characterization techniques, simulation/modeling capabilities, machine learning, and high-throughput screening in the complex field of electrochemical interfaces. However, independent analyses are insufficient to provide a comprehensive understanding of the electrochemical, morphological, microstructural, and chemical changes that occur during device operation. The development of a series of cross-scale and multimodal in situ characterization methods and theories is crucial for real-time and accurate observation of the dynamic evolution of electrochemical interfaces. This collaborative effort will help reveal the intricate mechanisms underlying interface evolution, which is essential for deepening our understanding and optimizing device performances.

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Competing interests

The authors declare no competing interests.

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