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Illuminating the Lifecycle of Functional Materials Through Multimodal Synchrotron X-Ray Characterization: From Synthesis to Operation and Degradation

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Introduction

Driven by the global demand toward sustainable energy systems, the green transformation of energy materials has become a central focus in modern material science across the global economy [1–5]. This pertains to any change in how a material is synthesized, processed, or used that reduces its overall environmental footprint by lowering energy demand, eliminating or replacing hazardous substances, minimizing waste, and enabling the material to contribute to cleaner applications [6]. Beyond supporting long-term environmental sustainability, these advances can also deliver immediate economic benefits by reducing energy consumption, lowering operational costs, and improving process efficiency. Such improvements not only enhance industrial competitiveness but also provide broader societal benefits through reduced resource use, eliminating toxic waste, and a more sustainable economy. The goal when developing materials in this framework is to enable materials to meet their functional demands without sacrificing essential performance. When studying materials, they can be divided into two key stages: 1) the processing phase, during which raw precursors are turned into functional forms, and 2) the operational phase, during which the material must maintain or enhance its intended behavior under specific conditions. Synchrotron X-ray characterization is central to providing the comprehensive, multiscale insights needed to understand these processes. By revealing the structural, chemical, and morphological evolution that occurs during both the processing and operational phases, synchrotron characterization can help researchers develop technologies that reduce manufacturing energy demands, improve operational efficiency, and utilize resources more effectively while still meeting the rigorous performance requirements of modern applications.

During the processing and creation phase, key parameters such as reaction temperature, solvent choice, and precursor chemistry, directly influence the environmental footprint of a material before it reaches application. For instance, lowering processing temperatures, replacing toxic solvents with water-based alternatives, and minimizing waste generation are all strategies that reduce energy consumption and hazardous byproducts at the point of creation [7]. Once fabricated, a mate-

rial must maintain its functional properties under real-world modes of operation such as mechanical stress, chemical exposure, and repeated thermal cycling. For green materials specifically, performance stability and reproducibility are critical. Degradation during use not only compromises functionality, but additionally erodes the environmental gains achieved during processing. Maintaining little to no degradation during use is the goal, since this preserves the energy savings gained during production, which verifies the material will remain under sustainable operation.

Particularly challenging in the realm of understanding the changes in functional materials during their processes and operation, is their inherent structural and chemical complexity across multiple length scales. Ranging from nanoscale to microscale, and macroscale, the newly designed green materials and their structure will continuously need to be characterized to fully understand their chemistry, structure, and function. Complex systems and heterogeneous mixtures found throughout nature and industry span a wide range of compositions and architectures, which further drives the need for multidisciplinary techniques to characterize them efficiently. Multimodal synchrotron X-ray characterization [8, 9], in which complementary imaging, spectroscopy, scattering, and diffraction techniques are integrated to probe materials across multiple length and time scales, becomes essential, since no single technique can capture the complete structure–property–function relationship in these complex systems. The high brightness, tunability, and penetrating power of synchrotron X-rays enable nondestructive characterization with spatial, temporal, and chemical sensitivity that is often difficult or impossible to achieve using conventional laboratory methods. By combining complementary modalities, researchers can simultaneously access structural, chemical, morphological, and functional information, providing a more complete understanding of material evolution during synthesis, operation, and degradation [10]. Exploration of a material at the nanoscale increases experimental complexity since high quality data resolution is required, but when scaled up, it allows for a wider influence across several scientific fields.

TECHNICAL REPORT

The examples highlighted in this review were primarily performed at the National Synchrotron Light Source II (NSLS-II) and demonstrate how multimodal synchrotron approaches can be applied to address a diverse range of challenges in sustainable materials research (Figure 1). The multimodal synchrotron characterization paradigm has evolved substantially over the past decade. At NSLS-II, a Multimodal Taskforce was established in 2017 to identify scientific opportunities and technical challenges associated with integrating complementary characterization methods. This effort culminated in the 2019 workshop *Multimodal Synchrotron Approach: Research Needs and Scientific Vision* and the

subsequent workshop report published in *Synchrotron Radiation News* in 2020 [8], which articulated the need for coordinated synchrotron imaging, spectroscopy, scattering, diffraction, and microscopy, together with complementary characterizations, modeling, and data-science approaches for complex materials systems. NSLS-II also enabled users to submit proposals requesting access to multiple beamlines within a single experiment, facilitating more integrated multimodal studies. Since then, the user community has demonstrated a growing demand for multimodal characterization, as reflected by the increasing number of proposals requesting access to multiple beamlines and complementary

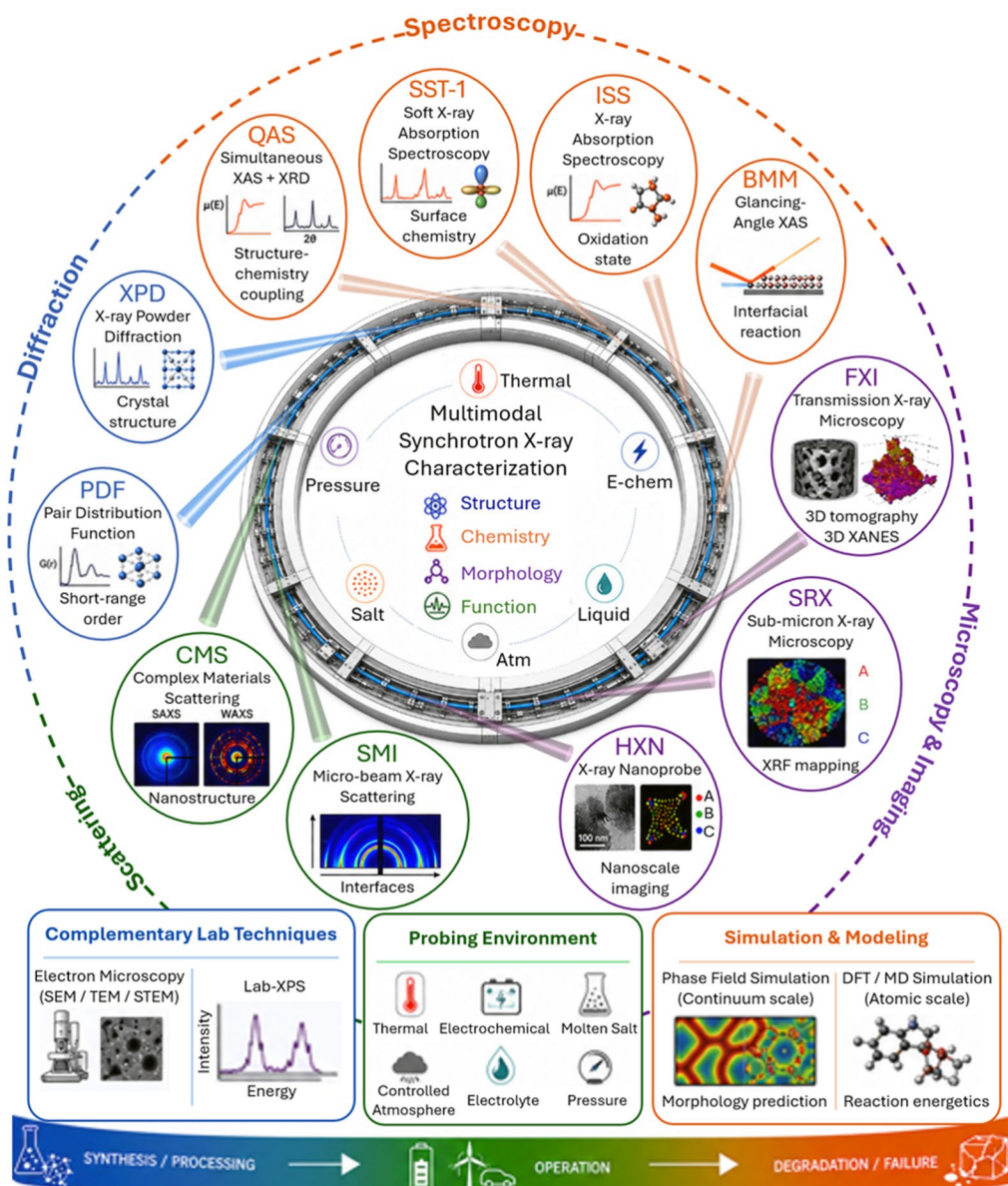


Figure 1: Overview of selected NSLS-II beamlines and complementary multimodal synchrotron X-ray techniques highlighted in this review, spanning diffraction, spectroscopy, imaging, and scattering methods used to investigate material synthesis, operation, and degradation.

techniques, exemplified by the network map of multi-beamline proposal requests [11]. The studies presented in this article demonstrate the power of multimodal characterization across two broad domains: materials in operation, including beyond Li-ion batteries (aqueous zinc ion batteries and sodium metal batteries), thermochemical energy storage, and molten salts; and materials processing, including environmentally friendly surface conversion coatings and nanostructure formation through dealloying.

Materials in operation

Among materials in operation, energy storage systems such as batteries and thermochemical materials present some of the most demanding characterization challenges, where electrochemical or thermal reactions drive structural and chemical changes across multiple phases simultaneously [12]. These systems must perform reliably under repeated electrochemical cycling, reactive environments, or elevated thermal conditions, all while maintaining the efficiency and stability required for clean energy applications. The following multimodal studies demonstrate how synchrotron X-ray characterization at NSLS-II has revealed phase transitions, morphological insights, and the evolution of chemistry in several systems under the operational phase.

Aqueous Zn ion batteries

Aqueous Zn-ion batteries (AZIBs) have gained considerable attention as promising candidates for large-scale energy storage systems owing to their safety, low cost, low redox potential vs. the standard hydrogen electrode, environmental friendliness, and high theoretical specific capacity of Zn metal [13–16]. Among cathode materials for AZIBs, manganese dioxide has emerged as a viable option for AZIBs due to its low cost and high theoretical capacity enabled by a 2-electron[−] redox process [17–19]. However, the Zn/MnO₂ performance is limited by poor cycle life, capacity fade, and uncertainty in their reaction mechanism [20–22]. Their underlying mechanism remains a challenge to characterize [23, 24], however using multimodal synchrotron X-ray techniques, the reaction pathways in Zn/MnO₂ systems can be probed for the structural, chemical, and morphological evolution in real time.

Initially investigated was a Zn/β-MnO₂ system to understand the cathode's evolution during the dissolution-deposition reaction mechanism. The crystalline phase evolution in the electrode was studied via *operando* X-ray diffraction (XRD), revealing the evolution of the pristine β-MnO₂ and formation of a reversible zinc hydroxide sulfate (Zn₄SO₄(OH)₆·xH₂O, ZHS) phase, presented as a waterfall plot in Figure 2A [25].

Complementary to XRD which probes long-range ordered phases, *operando* X-ray absorption spectroscopy (XAS) assisted to probe the local oxidation and coordination environment at Mn and Zn K-edges, providing complementary information to the phase evolution. X-ray absorption near-edge spectroscopy (XANES) analysis uncovered the formation of an amorphous ZnMn₃O₇·2H₂O (layered Zn-birnessite) phase during charging, which contributes significantly to the battery's capacity (Figure 2C), while an irreversible ZnMn₂O₄ (hetaerolite)

phase was simultaneously observed as a major contributor to capacity fade. Figure 2D demonstrates a continuous decrease in Mn-Mn bonds during cycling observed in extended X-ray absorption fine structure (EXAFS) analysis which reflects the formation and evolution of the Zn-Mn complex [25]. Additionally, the morphological evolution of the electrode at various stages of cycling was visualized via transmission X-ray microscopy (TXM) and scanning X-ray microscopy. *Ex-situ* 3D X-ray nanotomography via TXM showed significant changes in the electrode architecture, including the development of a thicker and irregular shell morphology after ~400 hours of cycling (Figure 2E) [26]. To spatially resolve elemental distributions of the Mn and Zn phases in the electrode during cycling, *operando* X-ray fluorescence (XRF) was employed to define the colocalization of Zn and Mn phases depositing near MnO₂ dissolution sites, forming a shell-like structure around MnO₂ (Figure 2F) [26]. Further characterized by XRF nano-imaging using a synchrotron hard X-ray nanoprobe, the electrode reveals at sub-10 nm resolution, the shell-like structure comprises of Zn-Mn complexes surrounding unreacted MnO₂ particles, as well as the reaction heterogeneity leads to incomplete utilization of the active material (Figure 2G).

Building on the insights from the reaction mechanism of β-MnO₂, electrodeposited ε-MnO₂, which has been identified as the dominant phase that forms during charging in long-cycled electrodes irrespective of the starting polymorph [27, 28], was investigated to elucidate the long-term reaction mechanism. *Operando* XRD lattice occupancy analysis revealed proton insertion/extraction, along with partial dissolution of ε-MnO₂ and minor Zn²⁺ insertion (Figure 2B) [28], concluding that proton-coupled electron transfer, proton-mediated dissolution-redeposition, and minor Zn²⁺ intercalation dominate the long-term reaction mechanism of Zn/MnO₂ batteries. Multimodal synchrotron characterization provides a framework for understanding the charge storage mechanism of Zn/MnO₂ batteries and lays the groundwork of optimal AZIBs for grid-scale energy storage.

Sodium metal batteries

Sodium (Na) batteries have also emerged as a promising energy-storage candidate in the context of green transformation and sustainable electrification [29]. Compared with lithium (Li), Na is far more abundant in the Earth's crust and is less constrained by geographically concentrated resources, making it attractive for building a more secure, scalable, and potentially lower-cost battery supply chain [30]. These advantages position Na-based systems as strong candidates for large-scale energy storage, where material availability, sustainability, and affordability are critical [31]. However, significant challenges remain, particularly in understanding interfacial instability and the evolution of the solid-electrolyte interphase (SEI), which strongly govern the reversibility and lifetime of Na metal batteries [32].

Interfacial instability, specifically the formation of dendritic metal deposits and the dynamic evolution of the SEI, a thin interfacial layer usually tens of nanometers thick, continues to impede their practical implementation despite their promising potential [33]. Tools such as

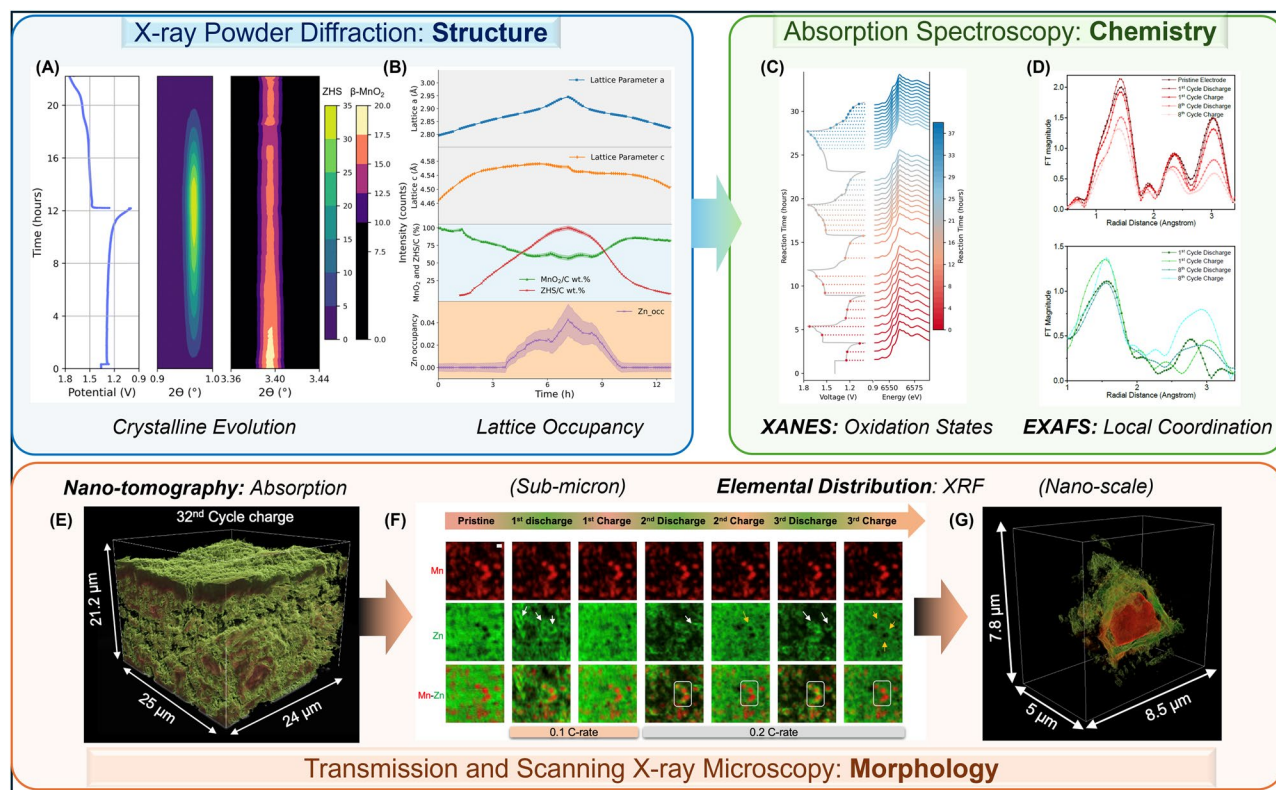


Figure 2: Multimodal synchrotron characterization of reaction mechanisms in MnO_2 cathodes for AZIBs. Operando XRD shows (A) the phase evolution of the $\beta\text{-MnO}_2$ electrode [25], and (B) lattice occupancy analysis of the $e\text{-MnO}_2$ electrode, highlighting structural site evolution during cycling [28]. Operando XAS shows (C) Mn K-edge XAS results indicating the gradual conversion of the $\beta\text{-MnO}_2$ structure during cycling [25], and (D) EXAFS R-space spectra comparing local coordination environments between the first and eighth cycles [25]. Transmission and scanning X-ray microscopy show (E) 3D X-ray nanotomography visualization of the charged MnO_2 electrode after the 32nd cycle (~400 h) [26]; (F) operando XRF imaging revealing the evolution of Mn (red) and Zn (green) phases during cycling; and (G) high-resolution XRF nano-imaging highlighting spatially resolved Zn-Mn distributions and the formation of shell-like structures in long-cycled Zn/ MnO_2 cathodes [26]. Adapted with permission from [25] and [26] (©2023 and 2025 Royal Society of Chemistry) and [28] (©2026 American Chemical Society).

scanning electron microscopy (SEM) and X-ray photoelectron spectroscopy (XPS) have been widely used to characterize metal deposit morphology and SEI chemistry, respectively [34]. However, further mechanistic understanding requires techniques capable of probing both morphological and chemical changes to capture the full spatiotemporal evolution of these highly reactive interfaces. Characterizing these systems using multimodal synchrotron X-ray thus can provide key insights beyond the scope of laboratory methods. For example, TXM enables visualization of internal structural features, while grazing-incidence wide-angle X-ray scattering (GIWAXS) is well suited for tracking thin-film and interfacial phase evolution. By tuning the incidence angle, GIWAXS provides access to different X-ray probing depths, ranging from a few to tens of micrometers, thereby enabling bulk-sensitive characterization of the SEI. In contrast, soft X-ray absorption spectroscopy (sXAS) offers nanometer-scale surface sensitivity through different electron detection modes: partial electron yield (PEY) and total electron yield (TEY), with typical probing depths of approximately 5 and 10 nm, respectively. Although the SEI is generally described as a thin

film covering the Na metal surface, and due to the porous nature of the Na deposit, SEI components may extend into the interior of the metal rather than being confined solely to the outer surface [35]. The complementary nature of hard and soft X-ray techniques in terms of penetration depth and light-element sensitivity, together with the complementary capabilities of transmission and grazing-incidence geometries for 3D tomography and surface-sensitive characterization, are illustrated in Figure 3.

In terms of Na internal morphological investigation, Figure 3A, [35] shows the nano-tomography analysis of electrodeposited Na on Cu current collectors. Despite a fixed area capacity corresponding to a theoretical $54\text{ }\mu\text{m}$, the actual film thickness progressively increases with current density. It may be observed that electrodeposits on the Cu collector are not fully dense and that the electrodeposit is morphologically heterogeneous. With increasing current density, porosity becomes more apparently evident.

Operando GIWAXS was employed to track SEI evolution during Na electrodeposition and stripping in a Na-Cu half-cell. Figure 3B

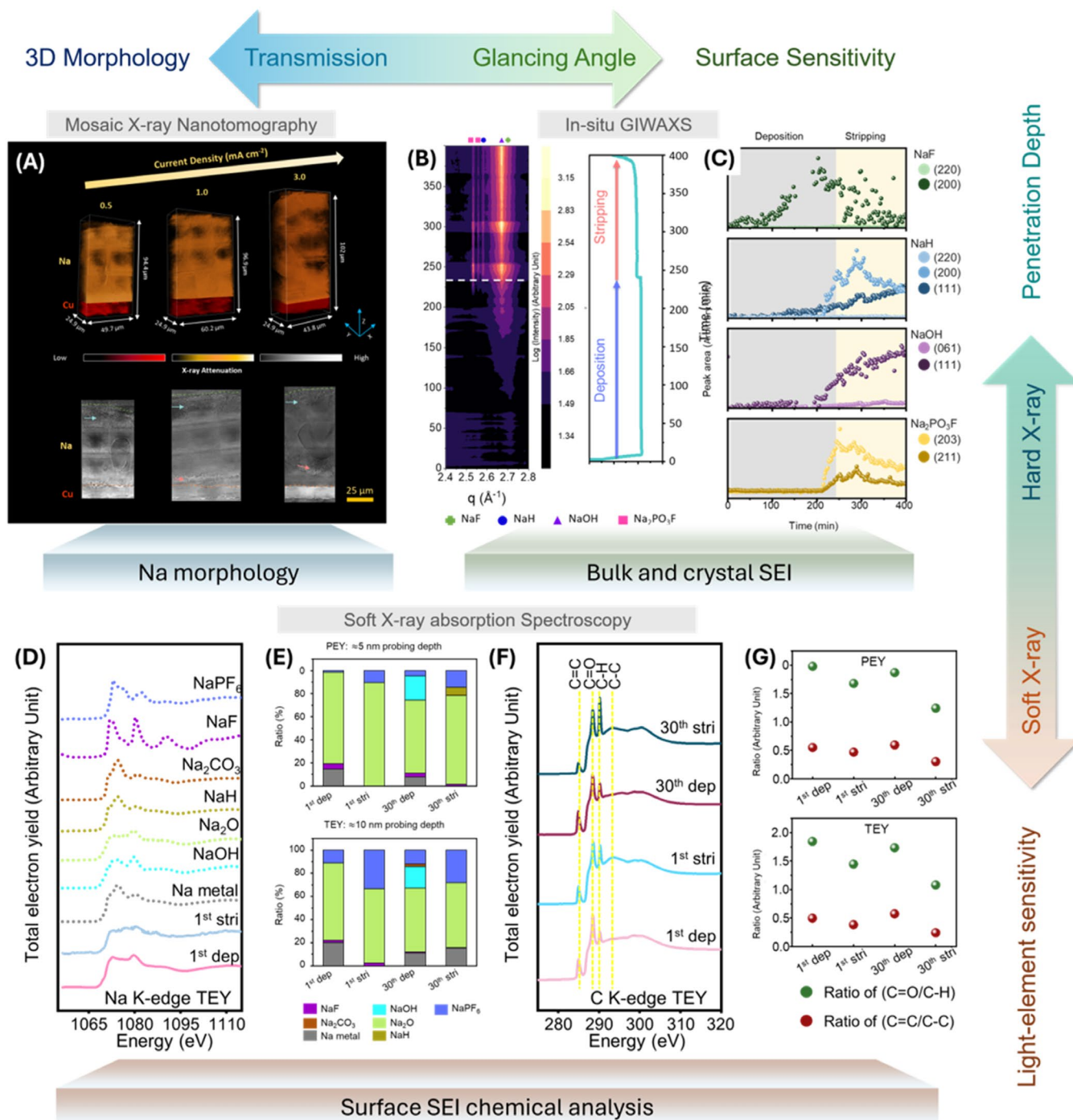


Figure 3: Multimodal characterization of Na deposits and SEI evolution with complementary soft and hard X-ray techniques, in different geometries. (A) Synchrotron X-ray nano-tomography by TXM revealing the three-dimensional morphology and spatial distribution of deposited Na [35]. (B–C) Operando grazing-incidence WAXS tracking the formation and evolution of crystalline SEI phases during cycling. (D–E) Soft X-ray absorption spectroscopy (sXAS) providing depth-sensitive information on the inorganic chemical composition of the SEI. (F–G) Carbon K-edge sXAS probing the evolution of the organic SEI chemistry. Together, these complementary techniques provide correlated morphological, structural, and chemical insights into Na deposition and interphase evolution [36]. Adapted with permission from [35] (©2025 John Wiley and Sons) and [36] (©2026 American Chemical Society).

shows the time resolved heatmaps and corresponding voltage profiles reveal the dynamic formation and dissolution of multiple SEI phases. During stripping, the identified SEI species show distinct evolution behaviors (Figure 3C, [36]). In particular, the significant change in NaF intensity highlights its transient nature, indicating that it undergoes continuous formation and loss rather than remaining static in the SEI. Fluorine-derived species such as NaF and $\text{Na}_2\text{PO}_3\text{F}$ are generally considered important for passivation because of their mechanical robustness [37]. Therefore, the decrease in $\text{Na}_2\text{PO}_3\text{F}$ intensity during stripping suggests partial dissolution or degradation of the passivation layer, which may expose fresh Na to the electrolyte and promote further side reactions. This process is consistent with the continued increase in NaH and NaOH during stripping.

Additionally, sXAS offers supplementary surface-sensitive information on SEI chemistry, especially for the light elements. Figure 3D shows Na K-edge TEY spectra collected during the first cycle, representing the inorganic components of the SEI, while Figure 3E further summarizes the depth-resolved SEI composition under different cycling conditions, including deposition and stripping in the first and 30th cycles. The disappearance of NaF after stripping indicates the instability of the SEI in 1 M NaPF₆ diglyme, consistent with the *operando* GI-WAXS results. With continued cycling, additional inorganic species such as NaOH, NaH, and Na_2CO_3 emerge, suggesting increasing chemical complexity of the SEI.

To further probe organic SEI evolution, C K-edge sXAS was analyzed using characteristic peaks assigned to C=C, C=O, C-H, and C-C species (Figure 3F) [38, 39]. The derived C=C/C-C and C=O/C-H ratios serve as qualitative indicators of the SEI's electronic structure and oxidation state [40]. Figure 3G shows both the outer and inner SEI exhibit similar trends: the C=C/C-C ratio remains nearly unchanged after the first cycle but decreases with extended cycling, suggesting the SEI becomes more insulating, while the C=O/C-H ratio is consistently higher during deposition than stripping and gradually declines over time, implying a less oxidized, less compact, and potentially less protective SEI with prolonged cycling [41]. Monitoring and understanding cyclic activity for Na battery components through multimodal approaches further expands the realm for such materials to be pushed toward sustainability and long-term energy storage in the future.

Thermochemical energy storage

Thermochemical energy storage materials (TCMs) represent a promising strategy for enabling high-temperature energy storage in concentrated solar power and other renewable thermal systems [42, 43]. Among the candidate materials, multivalent metal oxides have attracted particular interest due to their favorable redox thermodynamics, fast reaction kinetics, ability to operate with air as the working fluid [44, 45], and well-defined phase transitions at elevated temperatures [46, 47]. Among these oxides, Co_3O_4 serves as an ideal model system for mechanistic studies, despite its relatively high material cost, because its well-characterized redox behavior enables clear evaluation of reaction kinetics and pathways across nano- and meso-scales, provid-

ing insights that can guide the development of more cost-effective thermochemical materials [48–51]. However, practical application is hindered by performance degradation over repeated redox thermal cycles, which is closely linked to coupled morphological and chemical evolution across multiple length scales [52–54]. Key degradation mechanisms include particle sintering, cracking, and especially the dynamic evolution of pore structures, all of which directly impact oxygen transport and reaction reversibility [48, 55–57]. Key challenges are thus to provide insight into internal and isolated pore evolution and capture the spatially resolved chemical changes occurring during cycling [58]. To address these challenges, our recent work employed a multimodal synchrotron-based framework combining *in-situ* TXM and *in-situ* 2D XANES imaging, and post-analysis 3D XANES tomography, to probe the redox-driven transformation of Co_3O_4 across nano to microscale regimes through thermal cycling (Figure 4) [59].

This approach allows simultaneous visualization of 3D morphological evolution and spatially resolved oxidation-state changes during thermal cycling. The results reveal that micro- and nanoscale particles follow fundamentally different evolution pathways during thermal cycling. Nanoscale particles exhibit faster and more uniform redox conversion without the formation of significant porous structures, whereas microscale particles develop heterogeneous pore networks, including internal and isolated pores that persist and evolve over cycling. Importantly, these internal pores can trap evolved oxygen, creating localized high-concentration environments that impede subsequent reduction and contribute to kinetic limitations. The dynamic evolution of pore structure, including the transformation between open and closed porosity and its coupling with coarsening processes, plays a critical role in governing reaction pathways and long-term stability. By directly correlating 3D structural evolution with spatially resolved chemical state changes, this multimodal study provides mechanistic insights into degradation processes in TCMs, offering guidance for the rational design of more robust and efficient thermochemical systems for sustainable energy applications.

Molten salts

Molten chloride salts have emerged as critical materials for next-generation energy technologies, including molten salt reactors (MSRs), concentrated solar power systems, thermal energy storage, electrorefining, and high-temperature electrochemical processes, due to their high thermal stability, excellent heat-transfer capability, low vapor pressure, and wide operational temperature window [60, 61]. However, the extreme chemical and thermal environment at molten salt–metal interfaces lead to complex corrosion, restructuring, and mass-transport phenomena that remain poorly understood and continue to limit the long-term stability of structural materials [62, 63]. To address these challenges, we have developed a synchrotron-based multimodal characterization framework to directly visualize and quantify the morphological, structural, and chemical evolution of metals and alloys in molten salt environments across multiple length and time scales [64–66]. A major advance from these studies has been the application of *in-situ*

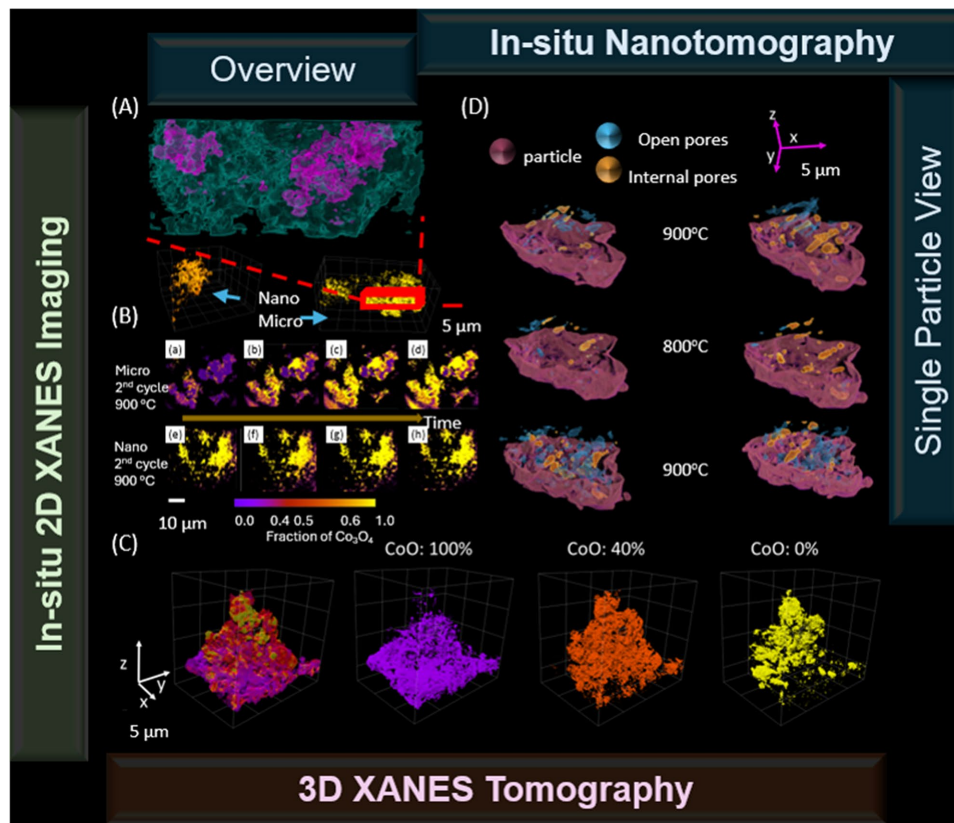


Figure 4: Multimodal characterization of chemical and morphological evolution in micro- and nanoparticle Co_3O_4 during high-temperature thermal cycling. (A) 3D morphology of Co_3O_4 micro- and nanoparticles in air during thermal redox cycles at 900°C , obtained by in situ synchrotron X-ray nanotomography; sub-figure shows internal and isolated pores in microparticles. (B) Chemical-state evolution for different particle sizes, shown by fitted 2D XANES images, indicating the Co_3O_4 phase fraction at selected time points in the second cycle at 900°C . (C) 3D XANES tomography of microparticles quenched after the second reduction cycle (900°C , 30 min), showing surface-localized CoO and Co_3O_4 retained in the interior. (D) Morphological evolution of a single microparticle during cycling between 800 and 900°C , showing formation of internal and open pores during isothermal holds. [59].

synchrotron X-ray nano-tomography (TXM), combined with *in-situ* bulk XANES and XANES imaging, as well as, *in-situ* diffraction, and post-analysis using STEM/EDS. In addition, atomistic simulation and phase-field modeling was integrated with *in-situ* tomography to provide mechanistic understand the corrosion and coarsening behavior of post-corrosion microstructures in molten salts (Figure 5).

Prior work on pure Ni and Ni–20Cr alloys in molten KCl-MgCl_2 demonstrated that molten salt corrosion is fundamentally different from conventional aqueous corrosion and proceeds through complex interfacial diffusion and selective dissolution pathways. Real-time synchrotron X-ray nano-tomography revealed the formation of bicontinuous porous structures during molten salt dealloying of Ni–20Cr, directly linking Cr dissolution, pore formation, and long-range diffusion processes, as shown in Figure 5A. In addition to capturing the 3D morphological evolution, 2D XANES chemical imaging provided complementary information on the local chemical state during the reaction. The 2D XANES spectra collected from selected regions of interest closely matched the Ni foil reference, indicating that Ni remained largely metallic during

molten salt exposure. This confirms that the observed porous structure formation was primarily associated with selective Cr dissolution rather than significant oxidation of the Ni-rich framework [67]. By further comparing the results between coarsening stage of the X-ray nano-tomography with phase-field modeling, we directly linked the experimentally observed interfacial morphology with the simulated evolution of post-corrosion coarsening in molten salts (Figure 5) [68].

Subsequent studies expanded the observations by systematically investigating how temperature, impurities, and dissolved metal ions influence interfacial evolution. Temperature-dependent synchrotron TXM revealed a transition between different corrosion pathways, including intergranular corrosion vs. bicontinuous morphology formation, likely depending on the balance between interfacial reaction kinetics, surface diffusion and long-range diffusion. Such findings highlight that corrosion morphology in molten salts is governed not only by thermodynamics but also by competing kinetic processes at the salt–metal interface [70]. Further work explored the effects of metal-ion additives such as Ni^{2+} , Ce^{3+} , and Eu^{3+} in molten KCl-MgCl_2 . By combining synchrotron nanoto-

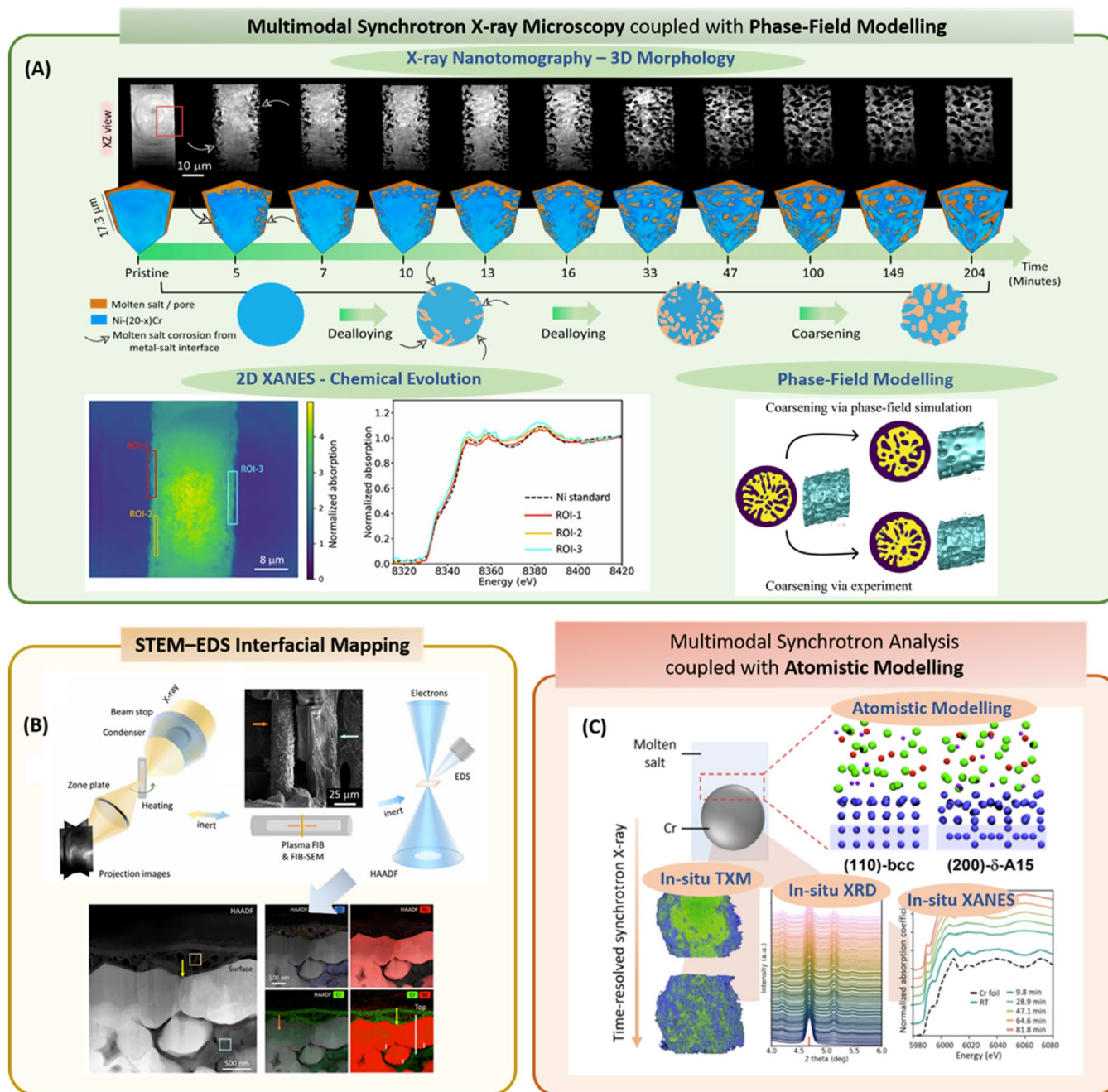


Figure 5: Multimodal synchrotron characterization of molten salt-metal interfacial evolution. (A) In-situ synchrotron X-ray nano-tomography showing time-resolved 3D morphological evolution of a Ni-Cr alloy during molten salt exposure. The reconstructed volumes reveal corrosion initiation at the molten salt-metal interface, followed by dealloying, pore formation, and coarsening over time. Blue represents the Ni-Cr alloy phase, while orange represents molten salt/pore regions, 2D XANES analysis showing representative spectra from selected regions of interest [67] and phase-field simulation of post-corrosion coarsening, showing qualitative agreement with the in-situ tomography-derived microstructural evolution in molten salt [68], (B) correlative post analysis using STEM-EDX can be conducted on the in-situ corroded sample from TXM, with the surface condition preserved through FIB sample preparation under inert atmosphere, showing the corroded molten salt-metal interface and elemental redistribution after exposure [64], and (C) In-situ multimodal synchrotron analysis of Cr microparticles evolution, where X-ray nano-tomography, XRD and XAS all collected during molten salt exposure, showing phase evolution as a function of time [69]. Adapted with permission from [64] (©2025 American Chemical Society) and [69] (©2024 Royal Society of Chemistry).

mography, spectroscopic imaging, and STEM analysis, our studies unveil the interplay between interfacial diffusivity and chemical reactivity determine whether alloys undergo dissolution, dealloying, coarsening, or interfacial stabilization. This provides a mechanistic framework linking molten salt chemistry with structural evolution pathways in engineering alloys. Post-exposure STEM/EDS analysis on the *in-situ* TXM samples provided nanoscale evidence of interfacial corrosion and elemental redistribution (Figure 5B) [65]. More recently, synchrotron X-ray studies of Cr microparticles in chloride salts further highlighted the importance of interfacial chemistry and impurity effects. *In-situ* diffraction (Figure 5C) revealed that Cr can undergo dramatic morphological transformation accompanied by phase evolution and changes in coordination environment during interaction with molten KCl–MgCl₂ [69]. In a different study of Ni–20Cr in eutectic LiCl–KCl demonstrated that impurity levels strongly influence alloy stability: under controlled conditions, Cr₂O₃ passivation suppresses large-scale morphological evolution, whereas water- and oxygen-containing environments accelerate corrosion and promote Cr dissolution and oxidation [71]. Time-resolved synchrotron imaging directly captured these impurity-dependent pathways in real time.

Collectively, these studies demonstrate how synchrotron-based multimodal imaging has transformed the understanding of molten salt–metal interactions by enabling direct visualization of interfacial processes that were previously inaccessible. The ability to correlate 3D morphological evolution, local chemistry, phase transformations, and diffusion behavior *in situ*, provides a powerful scientific framework for designing corrosion-resistant alloys and advancing molten salt technologies for operational materials regarding nuclear, solar and thermal energy storage.

Materials processing

From synthesis and processing to fabrication, materials are developed within a broad parameter space in which composition, precursor chemistry, temperature, additives, and reaction environments collectively determine the resulting structure and functionality. Understanding how these processing variables influence material evolution is essential for designing sustainable manufacturing pathways that reduce energy consumption, minimize hazardous chemicals, and improve resource efficiency. Multimodal synchrotron characterization provides unique insight into these processing–structure relationships by enabling the simultaneous investigation of chemical transformations, phase evolution, morphology development, and nanoscale structural changes during material formation. By correlating processing conditions with the resulting material architecture and properties, these approaches provide a foundation for rational process optimization and accelerated materials development. The following studies illustrate how multimodal synchrotron characterization has provided mechanistic understanding of environmentally friendly conversion coatings and enabled the controlled synthesis of functional architected nanomaterials through laser-induced solid-state dealloying. Highlights include industrial collaborations for sustainable coating development in the automobile

industry, International Space Station exposure studies, and the integration of machine learning and autonomous experimentation to accelerate materials discovery.

Industrial surface conversion coatings

Chemical conversion coatings are widely used surface treatments for improving corrosion resistance and paint adhesion on metallic substrates, especially in automotive, aerospace, and industrial applications [72–75]. Traditional chromate- and phosphate-based coatings have been effective, but their toxicity, carcinogenicity, and environmental concerns have increased the need for safer and more sustainable alternatives. Zirconium-based conversion coatings formed from hexafluorozirconic acid solutions have emerged as promising chromate-free systems because they can form protective ZrO₂-rich layers on steel surfaces through simple immersion-based processing. Our prior work on Zr-based CCCs has progressively established a processing–structure–chemistry–performance understanding through multimodal characterization (Figure 6).

In the first study, the fundamental formation mechanism of Cu-containing Zr-based conversion nanocoatings was investigated using a combination of *in-situ* synchrotron XRF microscopy, XANES spectroscopy, laboratory XPS, and electron microscopy. As shown in Figure 6A, the SEM image of the sample and the Fe XRF map before the experiment provide the initial substrate morphology and elemental distribution, while the Fe and Zr XRF maps after the experiment reveal changes in the substrate signal and the formation of the Zr-containing coating layer. Figure 6B further shows the fluorescence intensity of different elements during the treatment process, allowing the time-dependent evolution of Fe, and Zr to be tracked during immersion, solution exposure, and rinsing. SEM-based morphological analysis further quantified the size, density, and distribution of Cu-rich clusters as a function of processing time and temperature, showing that coating growth follows a reaction-controlled mechanism. Complementary XANES analysis, represented in Figure 6C, confirmed the formation of ZrO₂-rich coating chemistry. Together, these X-ray and electron-based modalities connected coating growth kinetics, elemental evolution, surface morphology, and depth-dependent chemistry in a way that could not be achieved by a single technique alone [76].

Building on this mechanistic foundation, we then introduced a hybrid organic–inorganic coating strategy by combining Cu²⁺ ions with poly(amidoamine) (PAMAM) dendrimers. This work further expanded the multimodal approach by using FIB-SEM to reveal the cross-sectional coating structure and interfacial void formation, STEM-EDS to map the nanoscale distribution of Zr and Cu within surface clusters, and XPS with Ar sputtering to resolve chemical changes through the coating depth. As shown in Figure 6D, the fitted Cu 2p XPS spectra of F-P0, F-P50, and F-P100 at the surface and after 20 min of sputtering reveal how PAMAM concentration changes the Cu chemical state and depth-dependent composition within the coating. These results show that PAMAM modifies Cu deposition and promotes the formation of Cu–PAMAM-containing species, particularly at higher PAMAM concen-

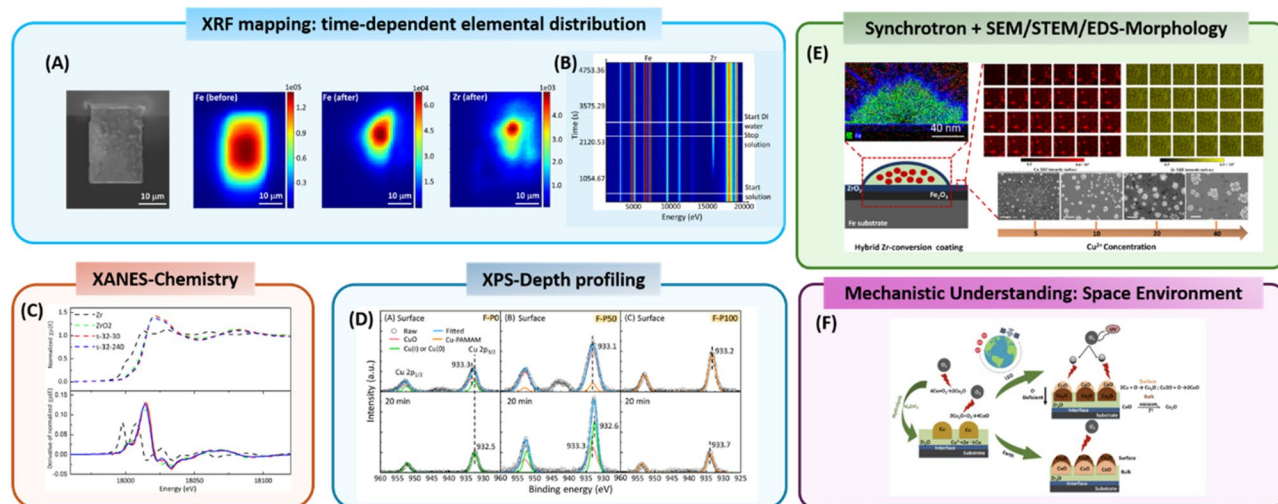


Figure 6: Multimodal characterization of Zr-based chemical conversion coatings. (A–B) SEM and Synchrotron XRF provides elemental mapping and in-situ monitoring of coating formation. (C) XANES reveals the chemical state and coating chemistry. [76]. (D) XPS provides depth-resolved surface chemical information. [77]. (E) In-situ synchrotron XRF microscopy coupled with SEM and STEM-EDX to characterize coating morphology and structure [78]. (F) Mechanistic understanding on the coating evolution for samples returned from the international space station, where the coating samples were exposed to the low-earth orbit environment. Together, these complementary techniques correlate coating growth, chemistry, morphology, and composition across multiple length scales. Adapted with permission from [76] and [77] (©2025 American Chemical Society), [78] (©2025 Elsevier), and [79] (©2024 Royal Society of Chemistry).

trations. Together, these complementary measurements showed that PAMAM strongly affects Cu deposition, coating thickness, interfacial void formation, and Cu distribution within the Zr-based matrix [77].

More recently, the effect of Cu^{2+} concentration was systematically studied in hybrid Zr-based coatings under chloride-rich conditions relevant to winter road-salt exposure using 3.5 wt% NaCl (Figure 6E). SEM and FIB-SEM revealed that increasing the Cu^{2+} concentration from 20 to 40 ppm changed the Cu-rich cluster size, density, and agglomeration behavior. STEM-EDS elemental mapping of the coating before and after NaCl exposure, highlighting how the coated film responded to chloride treatment and how the elemental distribution changed between untreated and NaCl-treated samples. Analysis quantifies the elemental distribution in cluster and non-cluster regions, showing preferential Cu enrichment in the cluster regions while the surrounding coating remains more Zr-rich. Together, these results demonstrate that Cu concentration strongly controls both the nanoscale coating structure and its chemical stability under chloride-rich exposure. Electrochemical potentiodynamic polarization further linked these chemical and morphological changes to corrosion performance. Overall, Cu20 provided a balance between adhesion and corrosion protection, whereas Cu40 showed stronger corrosion resistance but greater Cu agglomeration, highlighting the need to optimize Cu content for both surface uniformity and durability [78].

Finally, the environmental stability of these coatings was evaluated under low-Earth orbit (LEO) conditions using the MISSE-13 platform outside the International Space Station (Figure 6F). This work extended the CCC study from terrestrial corrosion environments to extreme aerospace exposure, where samples experienced atomic oxygen, ultraviolet

radiation, high vacuum, and repeated thermal cycling for approximately 500 days. A multimodal post-exposure characterization strategy was used. Comparison of pristine, Earth-stored twin, and ISS-exposed samples showed that Cu–Zr coatings retained a Zr-rich matrix with embedded Cu-rich clusters after LEO exposure. XANES further revealed $\text{CuO}/\text{Cu}_2\text{O}$ formation, depth-dependent redox evolution, and partial Zr chemical changes near Cu-rich regions, indicating that the coatings remained chemically resilient while adapting to the oxidative space environment. These results demonstrate the potential of Cu–Zr hybrid CCCs as environmentally friendly, non-chromate protective coatings for both terrestrial and aerospace applications [79].

Together, these studies show that these mechanistic understanding of Zr–Cu hybrid conversion coatings was driven by a truly multimodal characterization framework. *In-situ* synchrotron XRF provided real-time elemental evolution during coating growth and chloride exposure; XANES and micro-XANES identified oxidation-state changes and chemical speciation; XPS depth profiling revealed surface-to-interface chemical gradients; SEM, FIB-SEM, and STEM-EDS connected morphology, thickness, interfacial voids, and nanoscale elemental distribution; and electrochemical testing linked these structural and chemical features to corrosion performance. By integrating these complementary techniques, the studies established how processing parameters, organic and inorganic additives, environmental exposure, coating chemistry, and protective behavior are connected. This work highlights the central role of synchrotron and multimodal characterization in guiding the design of scalable, environmentally friendly CCCs for corrosion protection across automotive, industrial, and aerospace environments.

Nanostructure formation via dealloying

Dealloying provides a kinetic pathway at the nanoscale, where selective dissolution is designed to provide functional nanoarchitectures for a wide range of applications including catalysis, energy storage, sensing and actuation [80]. This spontaneous process is facilitated through the selective dissolution of one element in an alloy drives self-organization of the remaining components into bicontinuous structures composed of interconnected nanoporous-metallic networks [81]. Through porous architectures, these nanostructures directly contribute to applications in catalysis, while maximizing active surface area, which lowers the energy barriers for chemical reactions and reduces the need for precious metals. Achieving these benefits, however, depends on the ability to precisely control nanostructure morphology, feature size, and chemical composition during the dealloying process.

Prior work on thin-film solid-state metal dealloying (SSMD) has gradually built a comprehensive framework utilizing multimodal synchrotron characterization techniques to understand nanostructure formation. In this context, each dealloying system follows an A-B/C ternary configuration. A-B represents the parent alloy, and C is the dealloying agent, yielding a resulting bi-continuous structure composed of A and B-C components post-dealloying.

Initially, XRF and STEM tomography as a multimodal approach permitting resolution of features 5–15 nm small, which further enable the high surface area to volume ratios required for catalytic performance (Figure 7A) [82]. Figure 7B shows the evolution of this approach combining EXAFS, XRD, and STEM-EDS, revealing the chemical evolution, phase transitions, and morphological changes of dealloying in the solid-state [83]. This work demonstrated that the morphological transitions from globular to lamellar

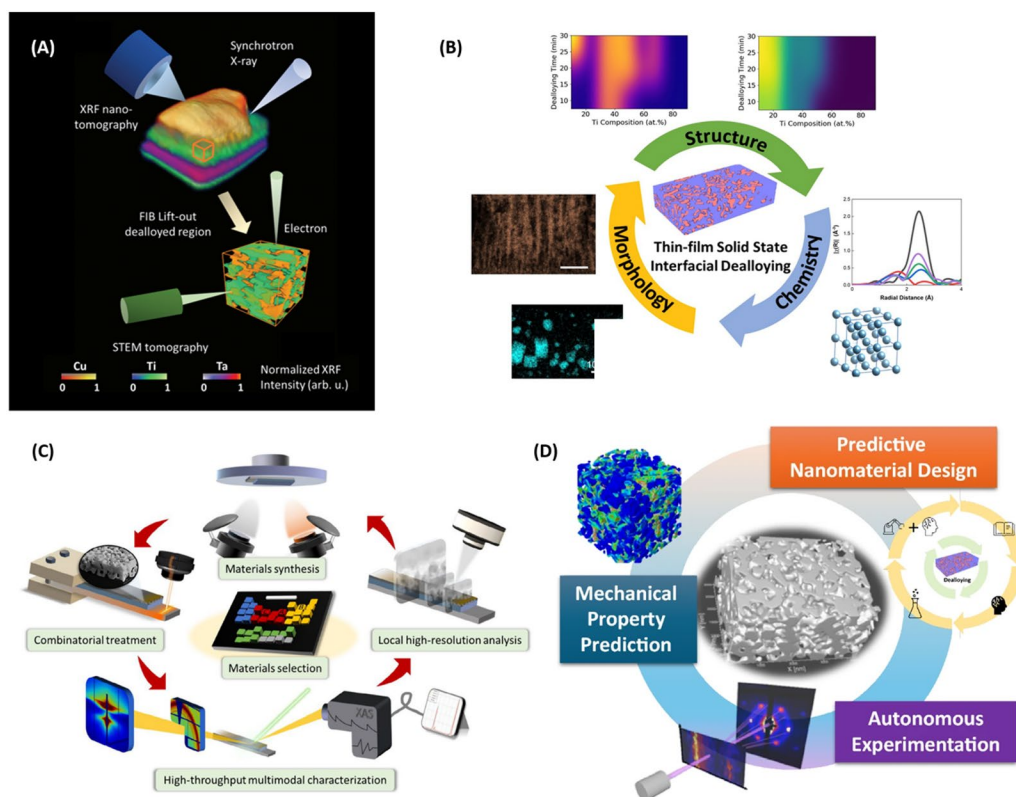


Figure 7: Multimodal characterization of thin films solid-state dealloying, SSMD. (A) Synchrotron XRF nanotomography combined with STEM-EDX tomography resolves 3D elemental distributions of Cu, Ti, and Ta across the dealloyed region [82]. (B) TEM imaging captures the morphology of the dealloyed nanostructure, XRD probes phase transitions and crystallographic structure, and EXAFS characterizes the local chemical environment. With combined techniques, a complete chemical and structural description can be seen in thin film solid-state dealloying systems [83]. (C) ML-guided workflow for accelerating nanostructured materials discovery in SSMD. Candidate systems are selected by an ML model, synthesized under combinatorial temperature-gradient conditions and characterized using high-throughput synchrotron techniques, coordinating between two beamlines (GIWAXS/GISAXS and XAS). Further verification of nanostructure formations is provided via STEM imaging. All results obtained aid in the feed back into the model to guide the next selected materials for SSMD [85]. (D) Vision for autonomous materials discovery enabled by multimodal characterization, integrating predictive nanomaterial design, machine-learning-driven property prediction, autonomous experimentation, and closed-loop optimization to accelerate the intentional engineering of functional materials [86]. Adapted with permission from [82] (© 2021 Royal Society of Chemistry), [83] (© 2023 Elsevier), [85] (© 2025 Wiley), and [86] (© 2025 Springer Nature).

to bi-continuous structures are strongly dependent on parent alloy composition. The kinetics of dealloying were measured across various length scales, which would remain unachievable without the versatility of techniques spanning from diffraction, imaging, and spectroscopy. Further investigation into Nb-Al/Sc systems identified oxidation as an unexpected but critical driving mechanism in thin-film SSMD, leading to the formation of bi-continuous metal-metal oxide nanocomposites while also highlighting the importance of incorporating oxidation as a key parameter in the thin-film dealloying system design and ML model refinement [84].

In the most recent study, laser-based thermal gradients were implemented in addition to a streamlined multimodal characterization (including GIWAXS, GISAXS, XANES, and STEM), to more efficiently explore the dealloying parameter space across ML predicted systems (Figure 7C) [85]. This type of approach allows for identification of critical phase transitions, nanostructure formation, and chemical evolution that demonstrates the power of integrating ML and multimodal synchrotron characterization for accelerating functional sustainable materials discovery [86].

Future directions

Multimodal synchrotron X-ray characterization has proven to be an indispensable tool used for understanding chemical and morphological complexity within various materials systems that are instrumental for green transformation. The studies presented across both materials in operation and materials processing, open several directions for future investigation, spanning from energy storage, thermochemical systems, molten salt corrosion, surface coatings and nanostructure formation. While each system presents unique challenges, commonalities emerge across both domains in terms of advancing *operando* capabilities, integrating ML, and expanding established frameworks to new material systems.

A recurring limitation across many case studies is temporal and spatial resolution during *operando* measurements. Among materials in operation, aqueous and sodium batteries such as SEI evolution and reaction heterogeneity at the electrode occur on timescales and length scales that current *operando* setups only partially resolve. Capturing short-lived intermediates more completely would strengthen the overall mechanistic conclusions. Similarly, in thermochemical energy storage, the relationship between porous network evolution and chemical state changes in thermal cycling, involve transient structural transformations that higher resolution and faster data acquisition could more comprehensively characterize. For molten salt systems, the competing kinetics of selective dissolution, coarsening, and long-range diffusion at the salt-metal interface, can be highly heterogeneous across the spatiotemporal domains. Coupling *operando* and *in situ* 2D/3D imaging with other modality to provide chemical, elemental and structural contrast, would allow these processes to be distinguished clearer for real-time experiments. As beamline capabilities evolve, complex *in situ* experimental setups will increasingly be utilized and developed. Representative of operating conditions observed in real time, *in situ operando*

studies will provide insight for design of materials relevant to applications in clean energy.

For synchrotron characterization, materials processing had similarly revealed the critical needs to continue advancing *in-situ* and *operando* capabilities. For instance, current analysis revealed the formation and degradation mechanism of the surface conversion coating across several conditions, yet the dynamic evolution of coating chemistry during deposition remains difficult to resolve at time scales at which nucleation and growth occur. Additionally, the formation of nanostructures via dealloying, using high-throughput multimodal characterization, mapped out the phase transitions and morphological evolution across temperature-time gradients with faster acquisition rates that contributes to understanding the early-stage kinetics of these bicontinuous structures. As *in situ* sample environments for thin-film and coating systems continue to develop, using *operando* synchrotron techniques under realistic processing conditions will provide the detail needed to design sustainable, scalable processes for energy storage and catalysis.

Another important future direction is the development of advanced *in-situ* imaging approaches for radiation-sensitive materials. As synchrotron sources continue to increase in brightness and coherence, beam-induced effects may increasingly influence the materials systems under investigation, particularly in electrochemical, catalytic, and soft-matter environments. Future efforts will benefit from integrating multimodal imaging techniques, including *operando* TXM, ptychography, coherent diffractive imaging (CDI), and tomography, to quantitatively distinguish intrinsic material evolution from radiation-induced artifacts. Recent developments in dynamic CDI have demonstrated the ability to reconstruct time-dependent structural evolution from diffraction data with minimal prior constraints [87], opening new opportunities for dose-efficient imaging of radiation-sensitive systems. Combined with advances in high-speed tomography and ptychographic imaging, these approaches will enable simultaneous characterization of morphology, chemistry, and kinetics across multiple length scales while minimizing radiation damage. Such capabilities are expected to provide unique insight into nucleation, growth, degradation, and interfacial phenomena in complex energy materials under realistic operating conditions.

Overall, integration of machine learning with these characterization frameworks also represents a promising direction. ML approaches could help map or predict the relationship between phase transitions, chemistry of a system, and morphological formations across a wider range of processing parameters. For dealloying, the combination of ML-guided system selection and high throughput characterization techniques, have already shown how the compositional and processing parameter space can be explored more efficiently. Employing multimodal studies, simultaneously, and autonomously, can be a pivotal direction for sustainable, efficient materials discovery. Extending this approach to other material systems, and establishing real-time feedback between beamline measurements, could shift outlook toward active guidance of material design. This is particularly valuable for accelerating the discovery of more high-performing and cost-effective functional materials

across batteries, thermochemical systems, molten salts and catalytic nanostructures.

Across many disciplines, multimodal synchrotron characterization provides a well-established and flexible framework for connecting processing conditions, material structure, and functionality in ways that are difficult to achieve through conventional techniques alone. Advances in multimodal instrumentation, data fusion analysis, and cross-beamline workflows are critically needed to further accelerate the adoption of multimodal experiments, enabling the characterization of complex materials. Ultimately, the continued development and application of both established and emerging materials systems will remain central to advancing the broader aspects of green transformation.

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