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The ZnOrgBAT Project: a new generation of Zinc–based rechargeable batteries

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Abstract

The ZnOrgBAT project aims to develop next-generation zinc-based energy-storage systems by combining highly cyclable Zn anodes with high-capacity organic cathodes. Since the performance and durability of these batteries are critically influenced by the behaviour of the zinc anode, a fundamental understanding of its degradation mechanisms is essential for guiding electrode and cell design. This contribution first provides an overview of the most common challenges associated with zinc anodes, together with the corresponding mitigation strategies typically adopted. It then presents an integrated summary of recently published studies by the co-authors, covering a range of different anode architectures and additive chemistry. Across these works, X-ray microtomography (XµCT) and microradiography (XµR) played a central role in elucidating key morphochemical processes, such as dead-metal formation, redistribution of active phases, gas-induced mechanical degradation, and in correlating these structural dynamics with electrochemical behaviour.

Keywords: Zinc-organic batteries, zinc-anodes, stationary energy storage, 3D and 4D microtomography

1 Introduction

The transition to a carbon-neutral economy relies on energy storage systems that have to be efficient, durable, safe, affordable and sustainable. As renewable energy deployment grows, reliable stationary storage becomes essential to balance supply and demand at both grid and local scales. Currently, most systems rely on lithium-ion batteries (LIBs), originally developed for electric mobility. While LIBs offer high energy and power density, their suitability for stationary use is limited in terms of lifespan, fire safety, cost and dependence on Critical Raw Materials (CRMs), such as lithium and cobalt [1, 2]. These factors raise concerns over price volatility, geopolitics, and environmental impact. To meet stationary storage requirements, alternative battery chemistries based on abundant, non-toxic materials are being explored. Among these, zinc-based rechargeable systems have emerged as a promising route thanks to their intrinsic safety, compatibility with aqueous electrolyte, low environmental impact, high theoretical capacity and potentially lower production costs. Within this context, the ZnOrgBAT project aims at the development of new type of rechargeable zinc batteries, combining highly cyclable Zn anodes [1, 3–6] developed at Polytechnic of Milan, with high-capacity organic cathodes, synthesized at National Institute of Chemistry [7], thereby integrating advances in both electrode materials to enable safe and sustainable energy-storage technologies. However, their performances ultimately depend on the stability of the metal-electrolyte interface and on the electrode's ability to maintain structural integrity during long-term cycling. The scope of this paper is to provide an overview of the published work related to the optimization of the Zn anode performances.

1.1 Challenges of the Zinc Anode

Among the different factors influencing the performance of zinc-based batteries, the behavior of the zinc anode plays a central role, as it can develop instabilities that manifest across different operating regimes. At low to moderate current densities, zinc tends to deposit in mossy and highly porous structures [1]. These deposits are mechanically weak and inclined to fragmentation, eventually leading to the formation of electrically isolated zinc domains and progressive loss of active material. At higher current densities, on the other hand, zinc deposition becomes controlled by mass transport, favoring the formation of dendritic structures. These protrusions can lead to uneven current densities and eventually to loss of capacity and short-circuit [1].

Passivation further complicates zinc anode operation. During discharge, zinc or zincate ions precipitate as ZnO and other insulating phases on the electrode surface. This might lead to high internal resistance and capacity fade. Moreover, the space distribution of passivation has a bearing on anode performance, since uneven passivation, which insulates only some portions of

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the electrode surface, causes a heterogeneous current density, which leads to the formation of pits during discharge and favors the formation of non-compact structures during charge [1, 2, 8].
Additionally, in aqueous electrolyte systems, the Hydrogen Evolution Reaction (HER) introduces a further degradation pathway, as hydrogen gas nucleates within the system, leading to potential structural damages, detachment of active material or decrease of the efficiency of the cell [1, 3].
To mitigate these phenomena, two primary strategies have been explored. The first involves the use of additives (to the electrode chemistry or the electrolyte) able to modify the interface electrode/electrolyte or enhance the electrode stability, however, their effectiveness is highly sensitive to current density, electrolyte composition, and the specific anode architecture [1, 2, 8]. The second strategy focuses on microstructuring the anode, using designs such as nanoparticles or sponge network, to distribute current more uniformly, confine dissolved zinc species, and limit concentration gradients [5, 6, 8, 9].
Understanding the effect of different mitigation strategies requires, alongside electrochemical performance evaluation, direct visualization of the evolving anode structure under realistic operating conditions. In this context, X-ray computed tomography (XμCT) and microradiography (XμR) enable non-destructive access to the internal morphochemical processes governing zinc-anode stability, providing insights that cannot be obtained from electrochemical measurements alone. The following chapter presents an overview of the main recently published results by the co-author, in which the combination of electrochemical measurements and X-ray imaging provided an essential support for elucidating the mechanisms that control Zn-anode behavior.

2 X-ray Imaging Case Studies on Zinc Anodes

Symmetric Zn|Zn configurations provide a controlled environment for assessing the Zn anode response to operating conditions, by removing the complexities caused by the introduction of a cathode with different chemistry. In the work of Rossi et al. [2], the effect of the Quaternary Ammonium Salt (QAS) Tetrabutylammonium bromide (TBAB), an electrolyte organic additive, was tested. The use of XμCT revealed that zinc foils develop a pronounced shape changes during cycling, including the formation of dead-metal domains as a function of current density and additive content [2].

The role of organic additives to the electrolyte solution was further investigated in mildly acidic aqueous electrolytes containing different QAS, including TBAB [1]. In this condition, complementary electrochemical measurements and in situ XμCT provided additional insight on the morphological impact of TBAB, revealing a counterintuitive dependence on current density. Indeed, the presence of the additive in the electrolyte induced a smoothing deposition at lower values (outer electrode, 10 mAcm^{-2}) but favouring unstable growth at higher rates (inner electrode, 25 mAcm^{-2}) (Fig. 1), thereby emphasising the complex interplay between electrolyte composition, additive chemistry, operating conditions and interfacial deposition mechanisms.

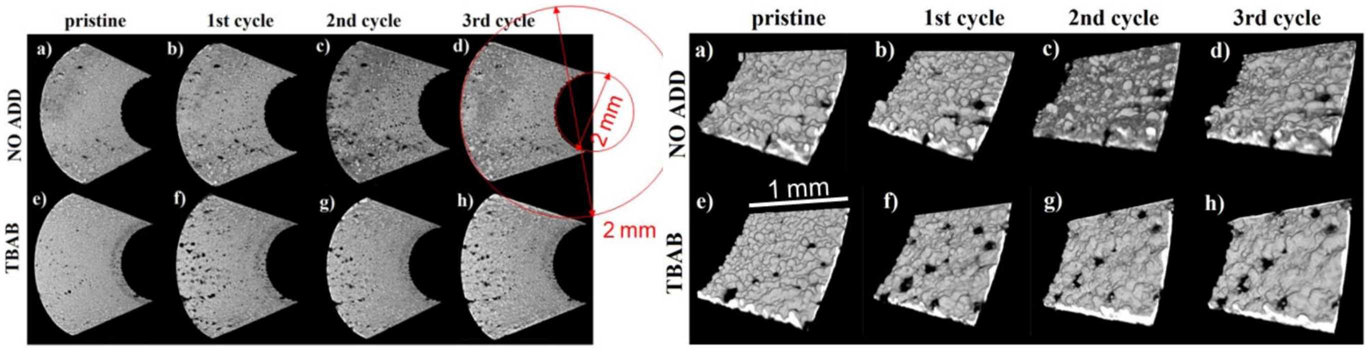

Fig. 1: *left*) Results of in situ XμCT measurements: 3D rendering of a sub-VOI corresponding to a section of the outer Zn electrode (10 mA cm−2) after subsequent cycling intervals; *right*) Isosurfaces renderings showing representative regions of the electrolyte side of the outer Zn electrodes. Modified after [1]

Microstructured zinc architectures were also explored as a route to mitigate dendritic growth and improve cycling stability. Among these, porous Zn sponge electrodes were investigated for their ability to distribute current more uniformly and confine dissolved zinc species. In this perspective, a pioneering work applied laboratory based *in-situ* XμCT to this class of electrode, providing the possibility to capture changes in porosity, ZnO fraction, and the connectivity of the percolating Zn network during cycling (Fig. 2) [9]. This approach was further developed by applying the first synchrotron-based *in-operando* microradiography (XμR) and *in-situ* XμCT to provide further insight on the formation and operating conditions of Zn sponge anodes [3]. In detail, *in-operando* XμR, under different potentostatic conditions, allowed direct visualization of the impact of hydrogen evolution on the mechanical stability of Zn sponge anodes during the formation, ultimately leading to the irreversible structural damage of the electrode (Fig 3). Complementarily, *in-situ* XμCT allowed to track the evolution of porosity, and Zn and ZnO distribution during formation and first discharge (Fig. 4) [3].

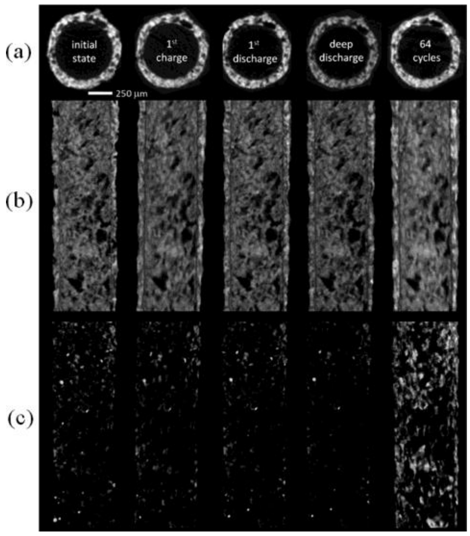

Fig. 2: *In-situ* XμCT: (a) Reconstructed axial slices, (b,c) volume renderings of graphite-supported Zn-sponge anodes under different electrochemical conditions: (i) initial state: as-fabricated, in the discharged state, at OCP; (ii) after first charge; (iii) after first discharge; (iv) after deep discharge; and (v) after 64 charge–discharge cycles. Voxel size: 5 μm^3. The volume renderings of Panel (c) represent the Zn backbone connectivity for the same electrochemical conditions. Modified after [9]

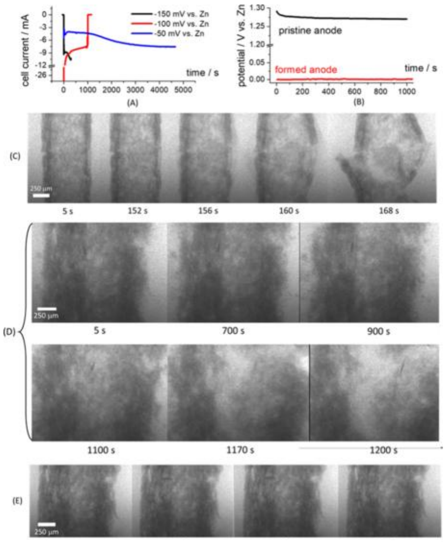

Fig. 3: (A, B) Electrochemical measurements for the cell with sponge electrode. (A) Chronoamperometries during formation (charging) at the indicated potentials, corresponding to *in operando* XμR measurements of Panels (C)–(E). The experiment carried out at −150 mV vs Zn is started with applied potential, but without the electrolyte, that is added at time 0 s. (B) Open-circuit potential (OCP) of the anode in pristine, uncharged (black plot) and formed (charged, red plot) at −50 mV (conditions of blue plot of Panel (A)). (C–E) *In operando* XμR of pristine Zn sponge electrode, polarized at: (C) −150 mV vs Zn (black plot of Panel (A)); (D) −100 mV vs Zn (red plot of Panel (A)); (E) −50 mV vs Zn (blue plot of Panel (A)). Modified after [3]

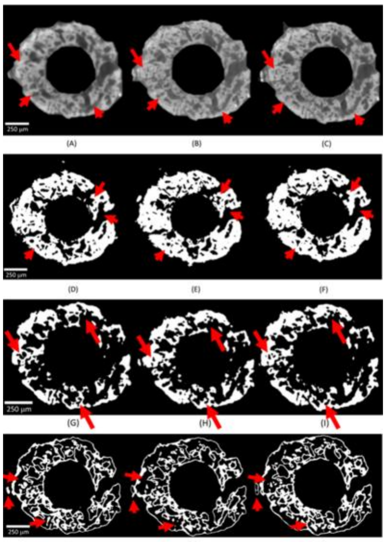

Fig. 4: (A–C) *In-situ* XμCT axial slice of the Zn sponge anode under the following conditions: (A) pristine electrode at OCP; (B) after the formation period; (C) after the discharge period. (D–F) Images corresponding to Panels (A)–(C) segmented for pores. (D) Pristine electrode; (E) after formation; (F) after discharge. (G–I) Images corresponding to Panels (A)–(C) segmented for Zn. (G) Pristine electrode; (H) after formation; (I) after discharge. (J–L) Images corresponding to Panels (A)–(C) segmented for ZnO. (J) Pristine electrode; (K) after formation; (L) after discharge. Modified after [3]

An additional approach to address the challenges of Zn anodes involved the use of ZnO@C nanoparticle as anode active material, with optimization of the slurry formulation and electrode architecture. In particular, two electrode fabrication methods were investigated: blade coating (BC-ZnO@C) and hot pressing (P-ZnO@C), producing electrodes with different thickness (*i.e.*, 100 μm and 400 μm, respectively) to assess the influence of active layer geometry on performance and durability [8]. The BC-ZnO@C electrodes exhibited superior cycling stability and specific capacity compared to P-ZnO@C. XμCT performed on samples before and after cycling allowed to follow non-destructively the morphochemical evolution of the composite Zn anodes in the full-cell ambient (Fig. 5), providing direct visualization of the critical degradation phenomena and allowing a correlation with the electrode architecture parameters [8].

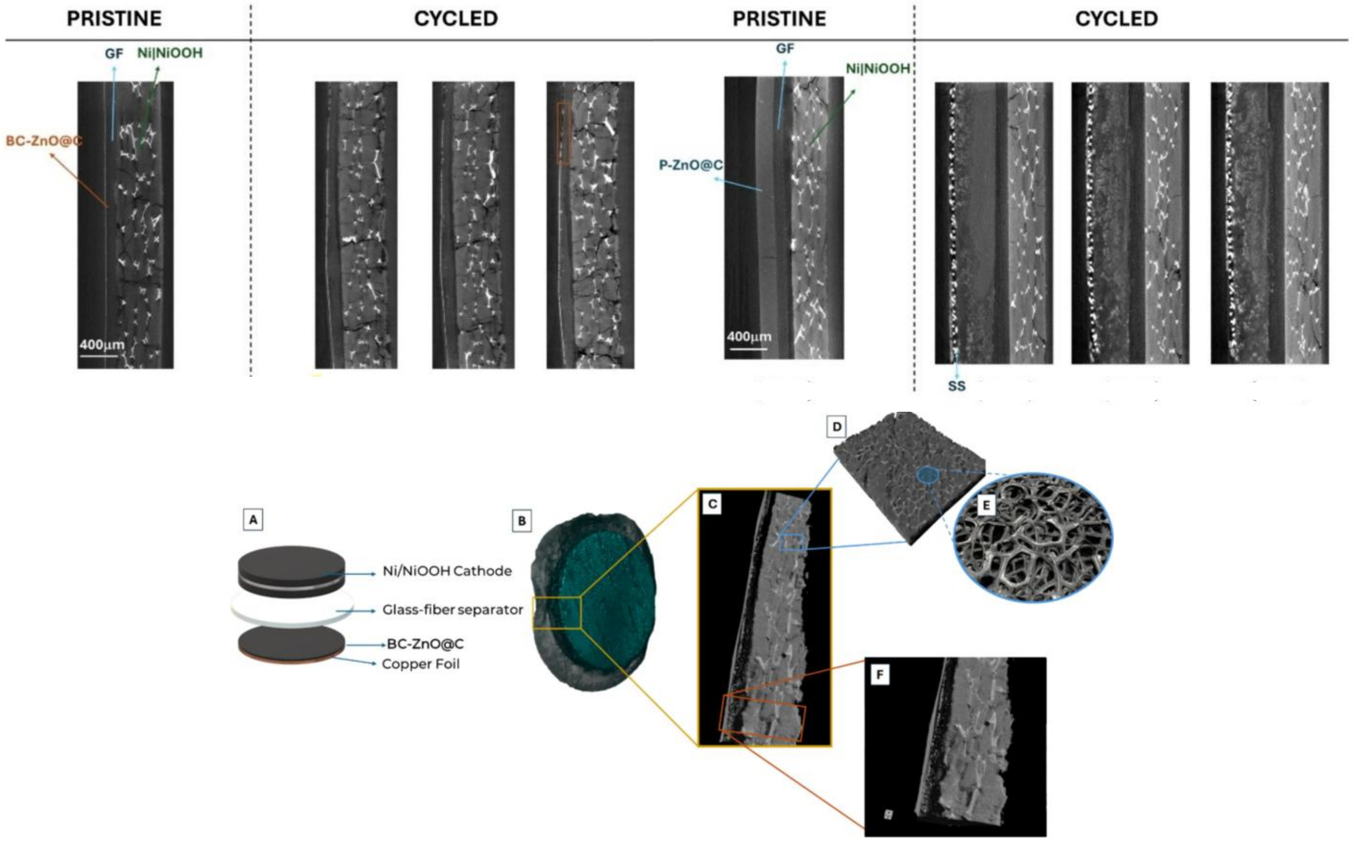

Fig. 5: *top left*) Virtual cross-sections obtained by XμCT analyses showing the copper current collector, the BC-ZnO@C anode (orange arrow), the glass fiber (GF) separator (blue arrow) and Ni/NiOOH cathode (green arrow) of pristine and aged cells; *top right*) Virtual cross-sections obtained by XμCT showing the pristine cell configuration, composed of the self-standing P-ZnO@C anode, GF separator, and Ni/NiOOH cathode, and aged cells after cycling, where the stainless-steel (SS) mesh current collector is visible as part of the anode structure. The comparison of the two highlighted a higher ZnO particle accessibility and a and more uniform ZnO reduction in BC-ZnO@C anodes; *bottom*) (A) Schematic illustration of the battery assembly with BC-ZnO@C anode, (B, C) Volume renderings of the reconstructed full battery analyzed by XμCT, 3D rendering of (D) the Ni/NiOOH cathode and (E) the Ni sponge, (F) 3D rendering showing a portion of the anode and cathode microstructure. Modified after [8]

As zinc batteries advance toward practical implementation, the scale of anode manufacturing becomes increasingly relevant. Imaging studies on upscaled zinc electrodes demonstrated that larger formats introduce spatial heterogeneities in porosity and compaction not always observed in laboratory samples. These scale-induced architectural variations were shown to facilitate parasitic gas evolution within the electrode bulk, leading to internal gas pathways that promote mechanical instability and premature durability loss. The study of Bozzini et al., [10], showed how XμCT serves as a useful diagnostic for detecting structural criticalities that arise during scale-up, effectively driving R&D of battery components.

Flowing particulate zinc anodes in zinc–air fuel cells were investigated in [11], where a combined experimental-modelling approach was proposed to follow response of the cell under varying operating conditions. The study focused on hopper-type anodes, *i.e.* gravity-fed particulate beds in which zinc granules flow through a funnel-shaped metallic support, ensuring continuous renewal of active material at the reaction interface. Alongside the electrochemical tests, a mathematical model was developed to correlate cell behaviour with particle motion and contact dynamics within the hopper. The combination of XμR and XμCT provided complementary structural information: XμCT allowed the visualization of the three-dimensional arrangement of Zn particles in the hopper anode, while XμR pinpointed that the particles remained in electrical contact with the current collector (Fig. 6).

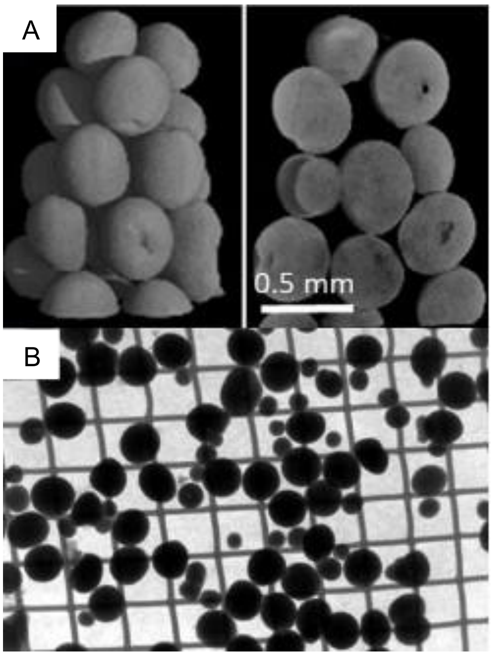

Fig. 6: volume rendering obtained by XμCT (A), showing the arrangement of the Zn particles in the hopper anode and XμR image (B) of Zn particles in contact with the current collector. Modified after [11]

3 Conclusions

This contribution has presented an overview of the recent advances in the characterization of zinc-based rechargeable technologies, highlighting the role of X-ray imaging. Across a wide range of anode architectures, including foil, sponge and composite particles, laboratory and synchrotron based XμCT and XμR enabled direct observation of the structural and chemical transformations governing Zn anode performance. By revealing morphological instabilities, gas-evolution pathways and microstructural heterogeneities, X-ray imaging provided insights that cannot be accessed from electrochemical measurements alone.

Such capabilities are particularly relevant for technologies under development within the framework of the ZnOrgBAT project, where durability, safety, and scalability must be demonstrated using industrially viable materials and designs. The synergic use of electrochemical and X-ray imaging characterization not only clarifies degradation mechanisms but also supports the rational optimization of the chemistry, electrode architecture, and manufacturing strategies, forming a robust methodology for guiding the development of next-generation zinc-based batteries.

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