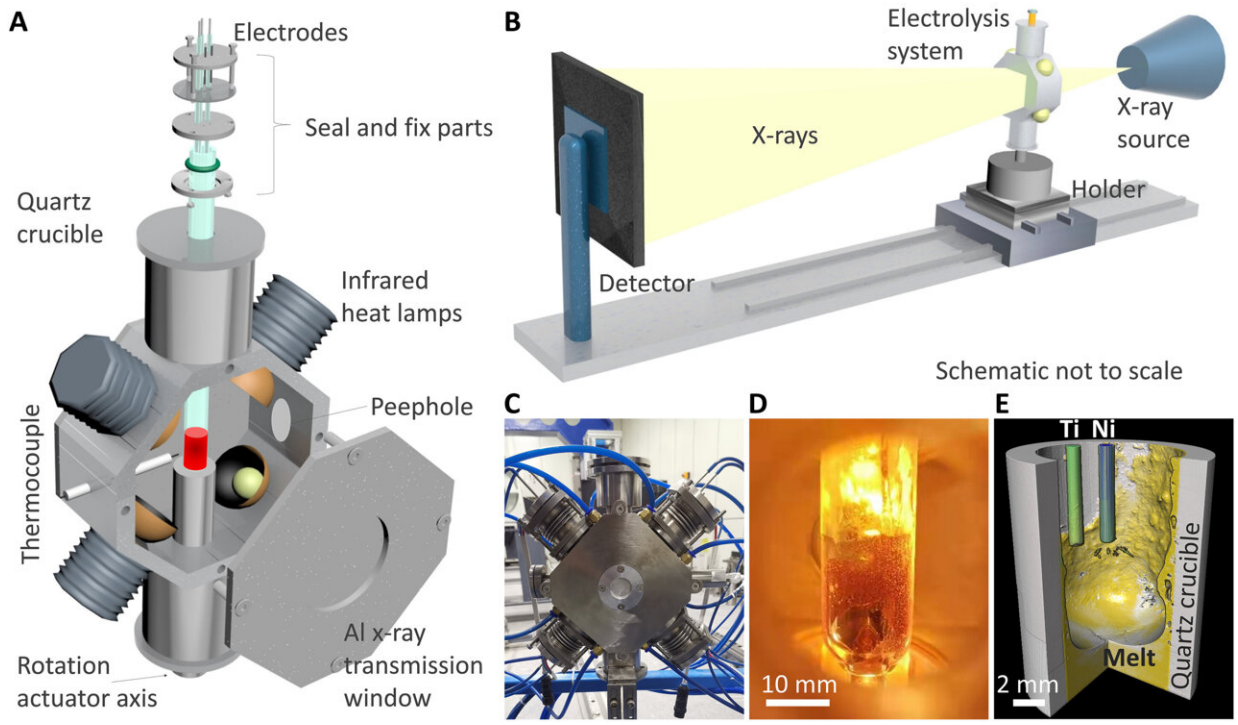
Using 4-D X-ray computer microtomography to observe high-temperature electrochemistry

February 21 2022, by Thamarasee Jeewandara



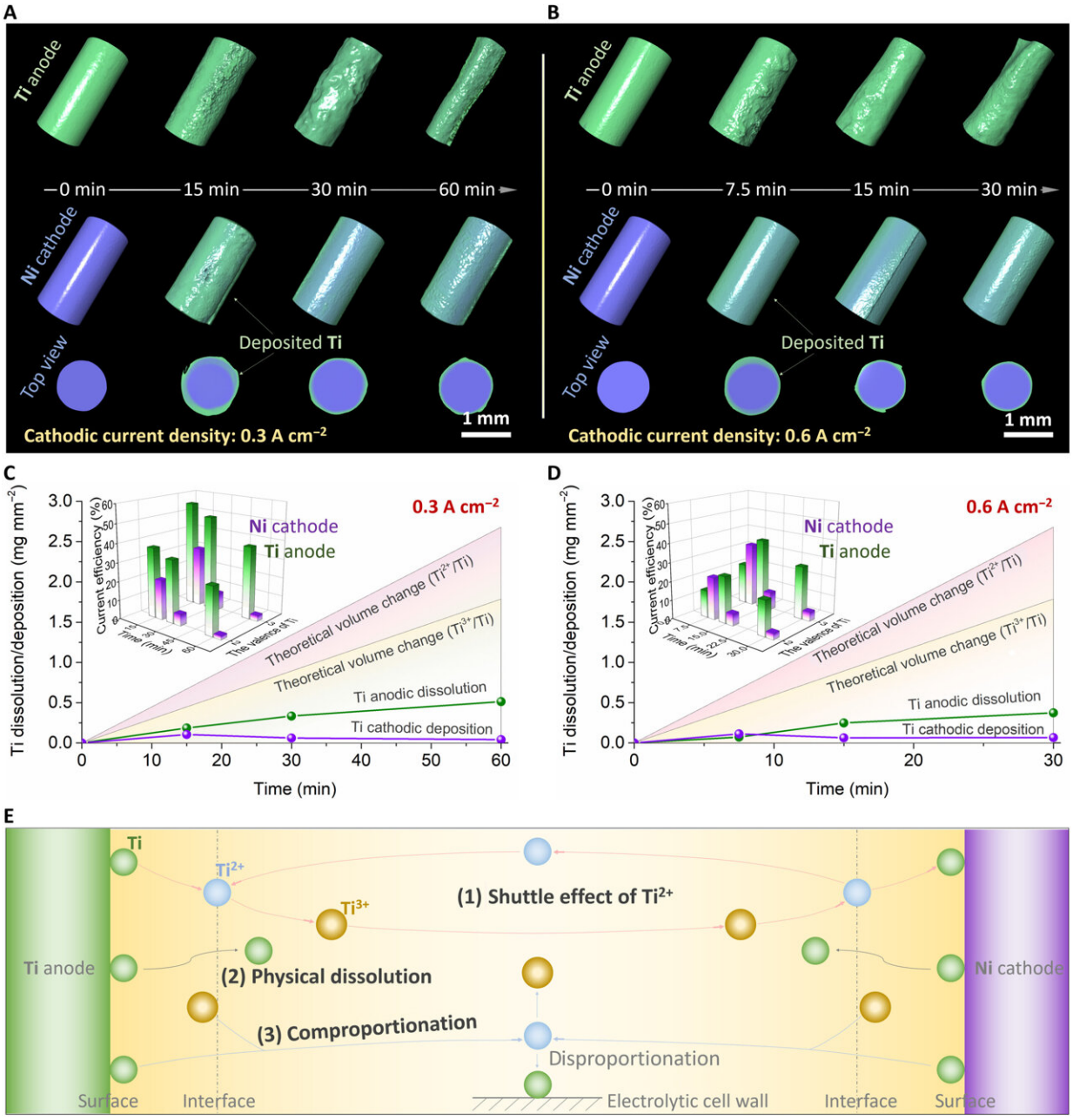
Schematics and photographs of 4D imaging apparatus for high-temperature electrochemistry. (A) 3D scheme of the assembled 4D characterization facility for high-temperature electrochemistry. (B) Schematic of the facility in transmission mode for x-ray μ-CT. (C) Photograph of the 4D studying facility. (D) Photograph of the high-temperature electrolysis cell through a peephole of the facility. (E) 3D reconstructed image of the experimental electrolysis cell at high temperature. Credit: *Science Advances,* DOI: 10.1126/sciadv.abm5678

The concept of high-temperature electrochemistry has broad ranging applications in multiple fields; however, researchers yet remain to conduct real-time observations to gain in-depth understanding of the evolution in such systems. The primary limits include harsh reaction conditions and multiphysics fields. In a new report now published in *Science Advances*, Handong Jiao and a team of scientists in advanced structure technology and metallurgy in Beijing, China, addressed the challenge by developing a high-temperature electrolysis facility. The facility allowed in-situ X-ray computer microtomography (μCT) for non-destructive and quantitative three-dimensional (3D) imaging. The μCT further probed the dynamic evolution of 3D morphology and components of electrodes in 4D. The team visualized the 4D process using reconstructed images to monitor the efficiency of the process, and explore dynamic mechanisms to provide real-time optimization. The 4D analysis platform provided in-depth combinations of traditional electrochemistry with digital twin methods to extract data and facilitate multiscale visualization.

The experiments

High temperature electrochemistry has many applications across metallurgy, nuclear, chemical production and energy industries. The process can facilitate the molten salt/oxide electrolysis to extract and purify metal, with a prominent role in large-scale stationary energy storage transformation. The process of experimentally examining the evolution of internal dynamics in such systems remains challenging due to limited development of the method. To monitor the dynamic evolution under harsh temperature and electrochemical systems, Li et al. developed a specific high-temperature electrochemical facility with built in X-ray microtomography (μCT) for quantitative 3D imaging, including the morphology and components of electrodes within such systems under extreme conditions. The team verified the apparatus via classical electrorefining experiments of titanium in molten salt. They then

performed a 4D study on the electrode structures and chemical components through time. The results combined high temperature electrochemistry with mathematical simulations to quantitatively design and optimize high-temperature electrochemistry.

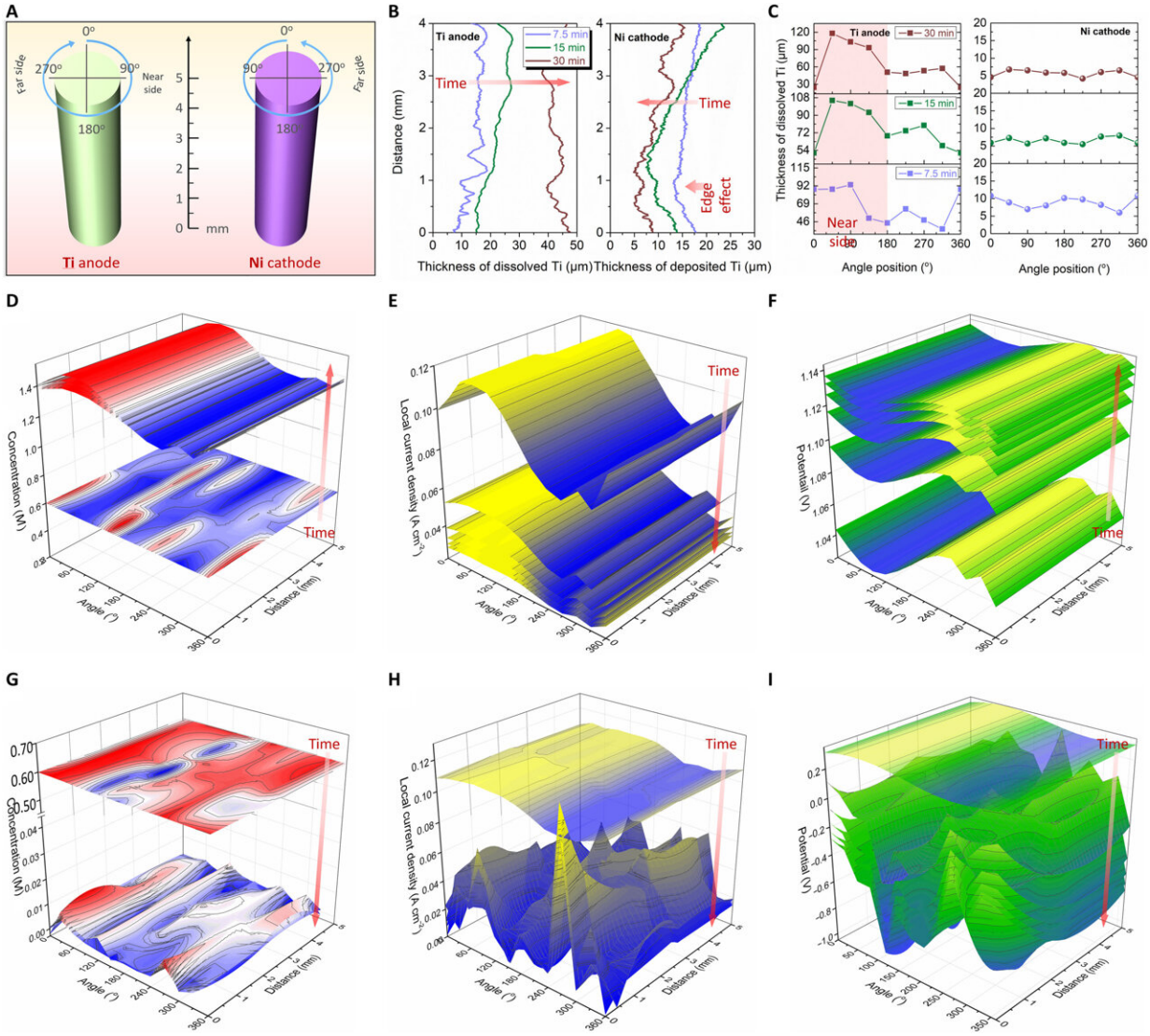


4D imaging and analysis of Ti electrorefining in the molten salt medium. (A and

B) 3D reconstructed images of the Ti anode and Ni cathode at different electrolysis time ranges. Current density, 0.3 A cm−2 (A) and 0.6 A cm−2 (B). (C and D) The change of the dissolution mass of Ti anode and the Ti deposition mass on the Ni cathode at different electrolysis stages. The insets are current efficiencies of Ti anode and Ni cathode at different electrolysis stages. (E) The competitive mechanism of possible reactions at electrodes. Credit: *Science Advances,* DOI: 10.1126/sciadv.abm5678

4D facility of high-temperature electrochemistry in the lab

The homemade in situ 4D characterization apparatus for high temperature electrochemistry contained a quartz tube electrolysis cell fixed vertically to the rotation actuator axis of the X-ray system. The team controlled the molten salt medium and electrodes by using an electrochemical measurement system based on metal wires. They then heated the cell by using four halogen lamps fixed to the heating furnace and settled the furnace chamber under vacuum conditions to reduce the temperature outside of the furnace. The researchers matched the distance between halogen lamps and cell to alter the heated zone according to experimental requirements. The electrolysis quartz cell provided an optically transparent view, although it was challenging to clearly observe the inner cell via an optical camera due to thermal radiation from high-temperature molten salt at elevated temperatures. The 4D facility based on X-ray μCT could reconstruct a reliable 3D complex structure of inner electrochemical cells with high spatial resolution from microscopic to macroscopic scales.



4D evolution process and mechanism of Ti electrorefining. (A) Demonstration of electrode surface information extraction. (B) The evolution processes of the thickness of the dissolved Ti (left) and the deposited Ti (right) from the bottom to the top of the electrodes as obtained from the 3D reconstructed images. (C) The evolution processes of the thickness of the dissolved Ti (left) and the deposited Ti (right) on different angles of electrodes obtained from 3D reconstructed images. (D to F) The evolution of concentration field, electric field, and current density on the surface of the Ti anodes calculated from the simulation results. (G to I) The evolution of concentration field, electric field, and current density on the surface of the Ni cathode calculated from simulation results. Credit: Science Advances, DOI: 10.1126/sciadv.abm5678

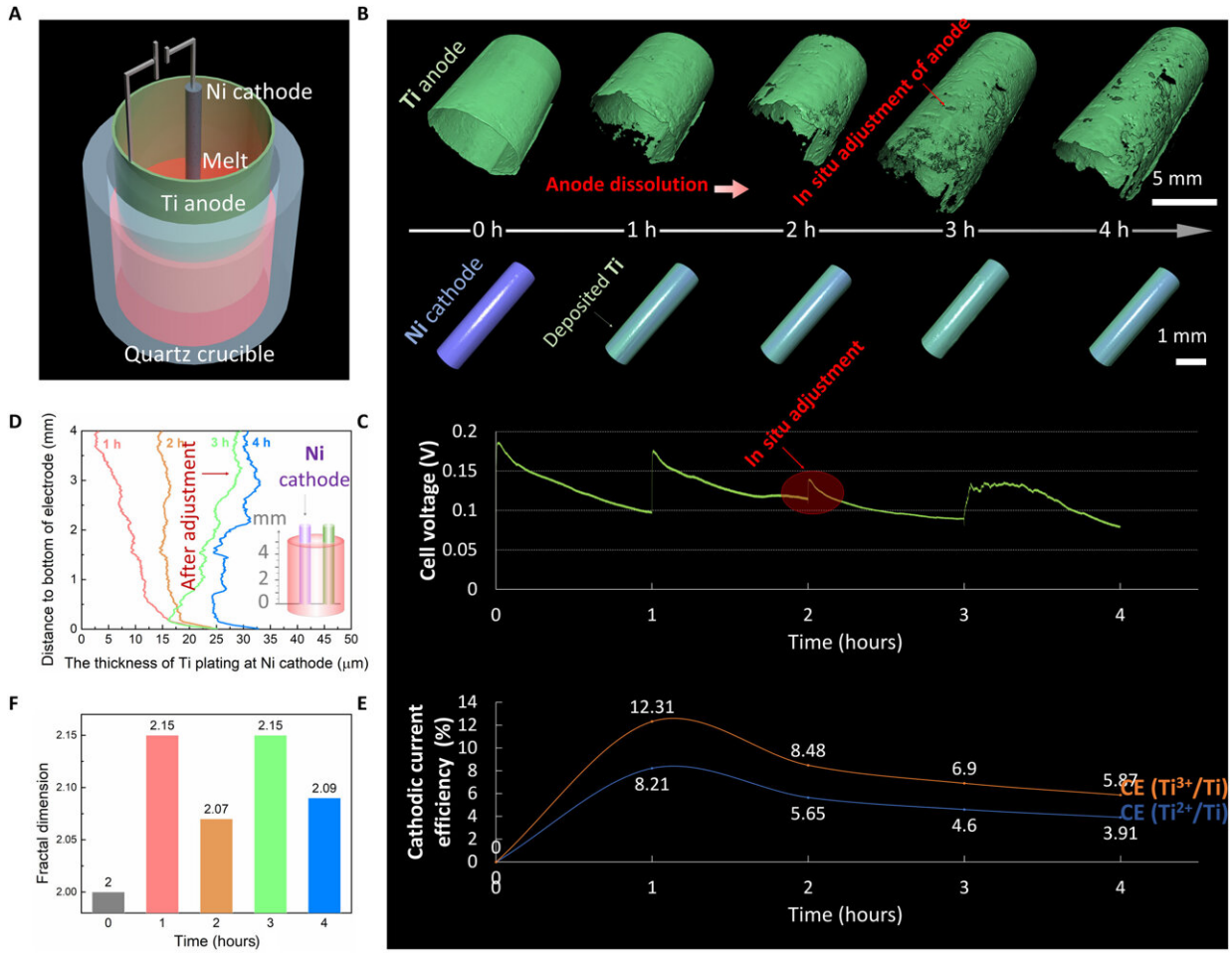
4D analysis

To show the capability of the 4D facility to conduct high-temperature electrochemistry, Jiao et al. performed a typical experiment of titanium (Ti) electrorefining in molten salt. The group had previously studied the extraction and purification of titanium using molten salt electroanalysis, with mechanisms of efficiency, purity and yield data yet to be understood. In this work, the scientists used a two-electrode system with titanium and nickel wires with same area and current density to form the anode and cathode. They then conducted scanning and electrolyzing in a cyclic process and obtained voltage-time profiles for the titanium electrorefining process under different current densities, then reconstructed 3D images of the Ti anode and Ni cathode at different electrolysis time ranges. Based on the outcome, Jiao et al. noted the average thickness of Ti plating on the Ni cathode to increase during the first electrolysis stage, to then drop slowly with time of electrolysis, which was unusual. The team therefore studied the competitive mechanism of chemical and electrochemical reaction at the interface and on the surface of electrodes to implement strategies to improve current efficiency.

Titanium dissolution and deposition

The scientists combined real-time experimental outcomes and simulations to reveal the evolution mechanism and extracted the local evolution information about the electrodes and noted the varying thickness of Ti anodic dissolution and Ti cathodic deposition surrounding the electrodes. The researchers noted the anodic dissolution and cathodic deposition of Ti to not be uniform. They revealed the evolution mechanism via multi-physical field simulations. Similarly, they obtained local information about the electrode surface to show the

evolution of the 3D concentration field, current density and electric field surrounding the electrode along with electroanalysis time. Jiao et al. conducted additional experiments to understand the evolution of the 3D physical fields surrounding the Ni cathode. Based on the outcomes of the 4D characterization process, they optimized the electrorefining process in situ.



In situ optimization of Ti refining process. (A) 3D views of the novel electrolysis system of Ti refining. (B) 3D reconstructed images of the Ti anode and Ni cathode at different electrolysis time ranges. Current density, 0.2 A cm−2. (C) Cell voltage-time profile of electrolysis process. (D) The evolution process of the thickness of the deposited Ti from the bottom to the top of the Ni cathode

obtained from 3D reconstructed images. (E) The current efficiency of the Ti cathodic deposition on a Ni cathode at different electrolysis stages. (F) The roughness of the Ni cathode at different electrolysis time spans. Credit: Science Advances, DOI: 10.1126/sciadv.abm5678

Outlook

In this way, Handong Jiao and colleagues successfully formed an in situ X-ray micro-computer-tomography method for high-temperature electrochemistry. The product offers new 4D characterization of the phenomenon for the first time. By using a titanium electrorefining experiment at high temperature, Jiao et al. verified the concept. They monitored the evolution of electrolysis for hitherto unknown quantitative details on electrode morphologies and current efficiency relative to different stages of electrolysis. The team combined the experimental results and multiphysics simulations to understand the mechanism of evolution in detail. The resulting techniques can be extended to high temperature electrochemical industries and industrial systems.

More information: Handong Jiao et al, A 4D x-ray computer microtomography for high-temperature electrochemistry, *Science Advances* (2022). DOI: 10.1126/sciadv.abm5678. www.science.org/doi/10.1126/sciadv.abm5678

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