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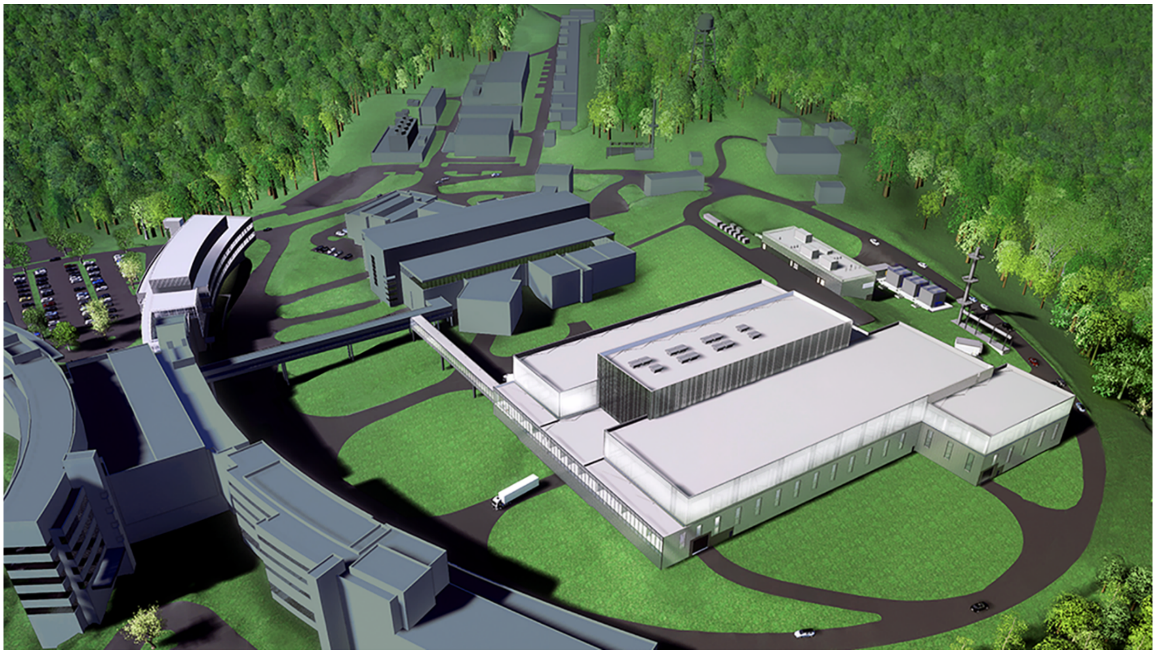
Second Target Station Project: STS/PSD/ESTD Workshop on Batteries and Energy Storage



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STS/PSD/ESTD Workshop on Batteries and Energy Storage

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January 2024

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CONTENTS

ACRONOMY

AI	Artificial Intelligence
CO2RR	CO2 Reduction Reaction
DAQ	Data Acquisition
ESTD	Energy Science and Technology Directorate
FTS	First Target Station
HER/HRR	Hydrogen Evolution Reaction/ Hydrogen Reduction Reaction
HFIR	High Flux Isotope Rector
INS	Inelastic Neutron Scattering
MEA	Membrane Electrode Assembly
NScD	Neutron Sciences Directorate
NSE	Neutron Spin Echo
OER/ORR	Oxygen Evolution Reaction/Oxygen Reduction Reaction
ORNL	Oak Ridge National Laboratory
NRR	Nitrogen Reduction Reaction
PGM	Platinum Group Metal
PPU	Proton Powder Upgrade
PSD	Physical Sciences Directorate
QENS	Quasi-Elastic Neutron Scattering
SANS	Small Angle Neutron Scattering
SE	Sample Environment
SEI	Solid Electrolyte Interphase
SNS	Spallation Neutron Source
STS	Second Target Station

1. EXECUTIVE SUMMARY

Oak Ridge National Laboratory's (ORNL) upcoming Second Target Station (STS) at the Spallation Neutron Source (SNS) provides a substantial leap forward in neutron scattering capabilities, offering high-brightness cold neutron sources that have the potential to revolutionize the study of energy materials. This report highlights the outcomes of a cross-directorate workshop on Batteries and Energy Storage held in September 2023, which brought together stakeholders from the STS Project, the Physical Sciences Directorate (PSD), the Energy Science and Technology Directorate (ESTD), and the Neutron Sciences Directorate (NScD).

The workshop concentrated on exploring neutron science opportunities in batteries and other energy storage systems, leveraging the unique capabilities of the STS. The discussions delved into emergent scientific problems and neutron scattering research directions. Four breakout sessions addressed the challenges and development opportunities associated with these topics.

Advanced batteries and other electrochemical energy storage systems play indispensable roles across our modern landscape, spanning ubiquitous consumer devices, clean transportation, and grid-scale deployments propelling us toward net-zero. These systems pose intricate challenges due to their highly heterogeneous and dynamic nature. They consist of multiple components such as electrodes, electrolytes, and separation membranes connected by interfaces. The interfaces are often dynamic and metastable during charging and discharging, while electronic and ionic transport occurs across multiple lengths and time scales. The STS promises scientific breakthroughs by enabling simultaneous observations of processes across broad spatiotemporal scales in working devices. Additionally, neutrons' sensitivity to light elements, such as H, Li, C, O, and N, is crucial for characterizing many energy storage materials. The increased cold neutron brightness at STS enhances the capability to study materials at lower concentrations and to investigate kinetics with better time resolution, expanding the range of scientifically and technologically relevant experiments. The STS instruments are expected to offer unprecedented capabilities by enabling sophisticated *in-situ* and *operando* experiments and incorporating multiple modalities.

The workshop identified four key opportunities:

(1) *Deploy innovative* operando *experimental approaches*. Developments in time-resolved *operando* experiments are vital for investigating "real" batteries and energy storage systems. The goal is to operate under conditions resembling real-world devices, providing unprecedented insights into the dynamic processes controlling synthesis and driving chemical transformations. Realizing this capability requires a synergic development of sample environments, experimental protocols, and data analysis along with the instrumentation.

(2) *Integrate multi-modal characterization techniques*. Multi-modal characterization techniques, spanning time, length, and energy scales, provide a critical opportunity to elucidate the multi-physics of complex energy storage systems. Simultaneous measurements are crucial for gaining a quantitative and mechanistic understanding of how atomic-scale structures and reactivity influence the performance of batteries and energy storage systems over time.

(3) *Advance into a new instrumentation design paradigm*. *Operando* experiments with rapid data collection and multimodal characterization necessitate purposeful integration between experimentation and computational modeling. Artificial intelligence (AI)-assisted experimental steering is needed to develop an intrinsic feedback loop that identifies critical information in real-time and redirects subsequent measurement to obtain salient information efficiently from a vast parameter space.

(4) Establish on-site infrastructure for electrochemical experiments. On-site space and apparatus for sample preparation and complementary characterization are essential for electrochemical experiments. Metadata from sample environments and complementary characterization results must be integrated seamlessly into the data acquisition system.

In conclusion, the STS project stands at the forefront of advancing neutron science capabilities, particularly in the critical domain of energy materials. The identified opportunities in instrumentation development underscore the potential for groundbreaking discoveries and the development of innovative solutions for sustainable technologies to meet society's evolving needs.

2. NEUTRON SCIENCE CAPABILITIES

Neutrons possess unique characteristics that make them exceptional probes for materials research. Neutrons are non-destructive, penetrating deeply into materials, allowing us to investigate matter using *operando* experiments with complex sample environments monitoring, for example, structural changes using electrochemical cells. Neutrons are particularly sensitive to light elements (such as H, Li, C, N, and O) and provide scattering contrast between neighboring elements (such as Mn, Fe, Co and Ni), essential for batteries and energy storage materials. Furthermore, neutrons carry magnetic moments, enabling the study of microscopic magnetism. Their wavelength and energy align well with the length and energy scales in condensed matter, making neutron scattering invaluable for probing the atomic world.

This workshop aims to identify opportunities for science using the current and planned suite of neutron instruments at ORNL and to identify capability gaps that might suggest future instruments to be built at the STS. Below are the questions provided to workshop participants to consider during the breakout session discussions:

- What are the emerging and projected energy storage grand challenges where neutrons can provide unique information?
- Are there unique requirements to support energy storage science that should be considered in the current 8 STS instrument designs?
- Are there synergistic or multimodal measurement capabilities that should be incorporated into neutron beamlines?
- What science-driven capabilities are needed beyond the current ORNL instrument suite and the 8 initial STS instruments?
- What advances in neutron data acquisition, reduction, and analysis will be required to exploit STS capabilities fully?
- Will new sample environments be needed to exploit STS capabilities fully? What are they?

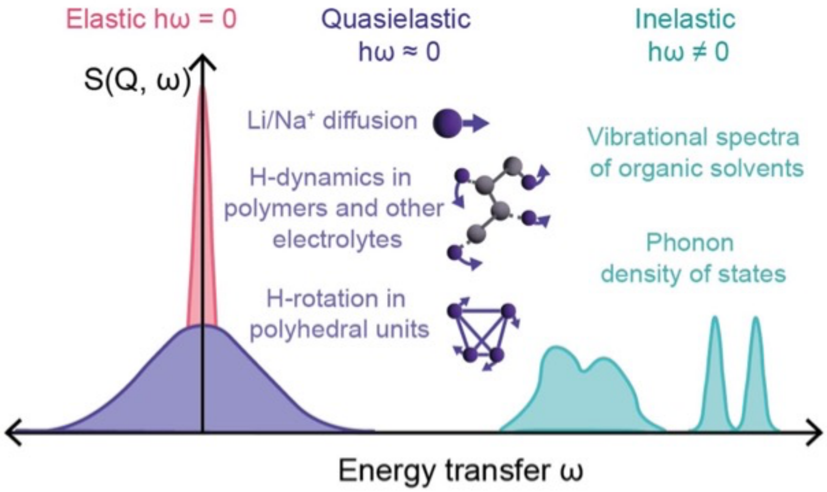

Figure 1 Depending on the energy transfer, neutron scattering processes are grouped into elastic scattering, quasielastic scattering and inelastic scattering, providing insights into various aspects of the structural information about materials under investigation. Battery and energy materials related cases are emphasized here. From Ref. [1].

This chapter reviews current neutron scattering capabilities in the field of batteries and energy storage. Neutron scattering is broadly divided into structural and dynamical characterization techniques (see Figure 1, adapted from Ref. [1]) to investigate two fundamental questions: where the atoms are and what they are doing. Structural characterization techniques rely on the *elastic scattering approximation*, yielding time-average structural information over a duration determined by the instrument's energy resolution. On the other hand, dynamical characterization techniques provide snapshots of time-average structural information over a period corresponding to the finite energy transfer during *quasielastic* and *inelastic scattering* processes. Different energy transfers unveil distinct time windows for averaging structural information, revealing the system's dynamic characteristics.

For *operando* experiments, we need to distinguish between time-resolved experiments and dynamic characterization. Time-resolved experiments can involve both structural and dynamic characterization of transient states during experiments. For neutron scattering experiments at pulsed neutron sources, the ultimate time resolution is limited by the source frequency for irreversible phenomena and by the neutron pulse width for reversible phenomena.

2.1 STRUCTURAL CHARACTERIZATION

Neutron scattering comprises multiple structural characterization techniques across various length scales, including diffraction, reflectometry, small-angle scattering, and imaging. Diffraction is sensitive to atomic structural changes and phase formation during chemical reactions. Neutron reflectometry (NR) can study interfacial chemistry and ion transport in solid electrodes and electrolytes. Specular NR provides nanometer-scale depth profiles of layer structures, while off-specular NR provides information about in-plane morphology. Small angle neutron scattering (SANS) is powerful for studying nanoscopic and mesoscopic objects from a few nm to hundreds of nm. Neutron imaging provides spatially resolved structural information with a resolution of ~ 10 μm.

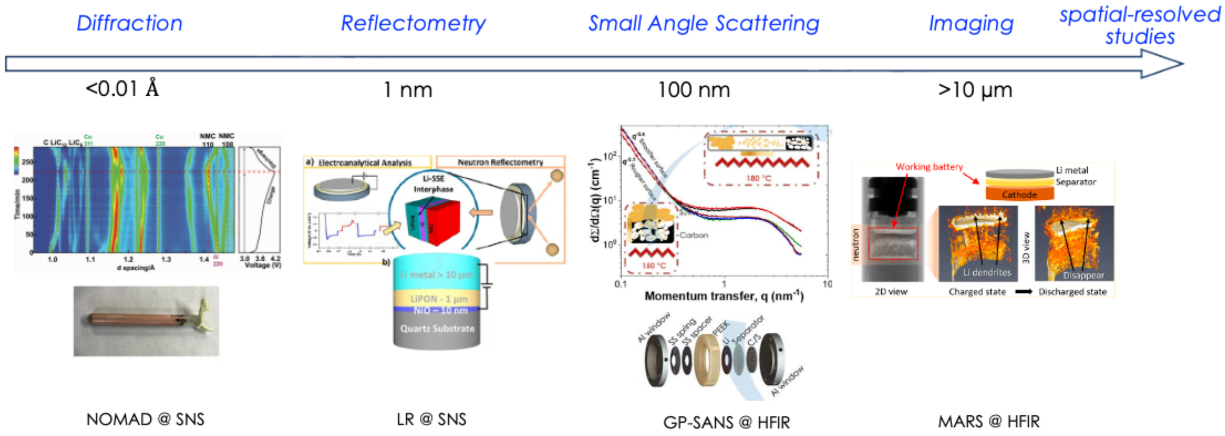

Figure 2 Neutron scattering techniques probe real-time structural changes across extended length scales in operando experiments using electrochemical cells. Adapted from Ref. [2-5]

Battery and energy storage research is an excellent example illustrating *operando* neutron scattering experiments providing dynamic structural information over broad length scales, as exemplified in Figure 2. Neutron diffraction determines atomic position changes down to sub-picometer (0.01-Å) length scale during charging and discharging [2]. Reflectometry detects the evolution of the solid electrolyte interphase (SEI) layer *in situ* with cycling with a sub-nanometer resolution [3]. Small angle scattering shows the evolution of chemical heterogeneity from ~ 1 to 100 nm scale during reaction [4]. Neutron imaging monitors real-time dendrite growth in batteries [5, 6], providing critical insights into degradation mechanisms. Neutron imaging instruments at a Spallation neutron source can benefit from the intrinsic wavelength resolution to enable advanced capabilities, such as Bragg-edge imaging, to gain quantitative spatial information about the atomic and internal structures, including strain profiles, grain orientation, and phase distribution [7]. Special electrochemical cells have been developed for these experiments for various neutron scattering techniques, which reveal micro-structure dynamics ranging over sub-Å to over 10 µm length scales, providing a holistic view of what is happening in working batteries.

2.2 DYNAMIC CHARACTERIZATION

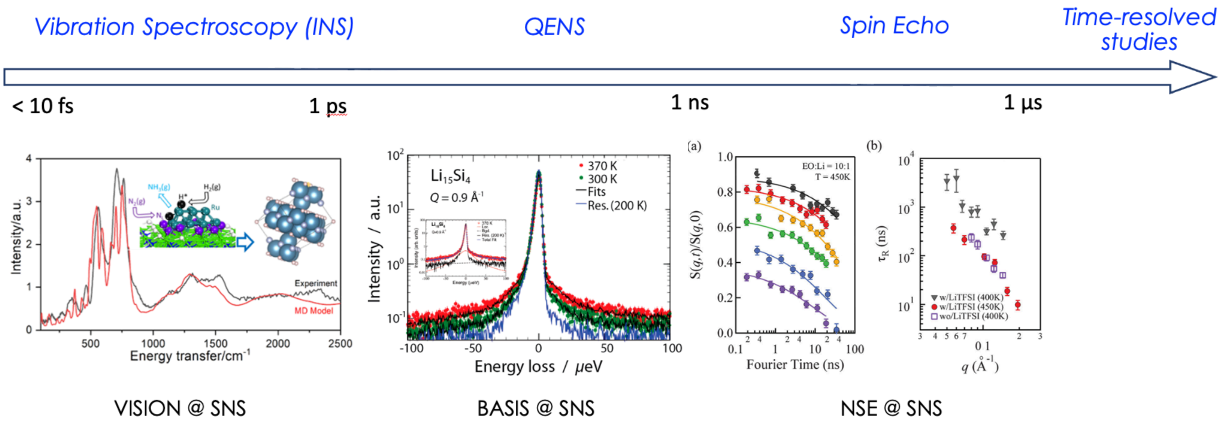

Figure 3 Various neutron scattering techniques provide access to dynamics over a broad time scale in energy materials. Adapted from Ref. [6-8]

Neutron scattering also supports a range of dynamic characterization techniques across a wide range of energy scales, including vibration spectroscopy (INS), quasielastic neutron scattering (QENS), and neutron spin-echo (NSE), as exemplified in Figure 3.

For energy materials research, vibration spectroscopy measures lattice dynamics on energy scales of ~ 1-1000 meV (~ 3 fs to 3 ps). The vibration modes are characteristic signatures that can identify pathways during chemical reactions, e.g., in ammonia synthesis [8]. QENS probes broad-spectrum dynamics. In battery research, these dynamic processes include ionic diffusion (e.g., Li, Na, H, O, etc.) and rotational or relaxation dynamics in electrodes and electrolytes. QENS provides an atomic scale understanding of the diffusion process, including its nature (free diffusion, jump-diffusion, or diffusion in the presence of ion traps) and the geometry of the localized processes. QENS experiments can quantify characteristic times ranging from the picosecond to the nanosecond scale to determine diffusion coefficients and energy barriers for diffusion using temperature-dependent measurements [9]. NSE probes even slower dynamical processes (lower energies) down to microsecond time scales [10].

2.3 MULTIMODAL CHARACTERIZATION

Complex multi-physics necessitates characterization across extended length and time scales. Multi-modal characterization is common in battery and energy storage material research. In some instances, investigating a single system may require multiple neutron scattering techniques to address a research question comprehensively. For instance, combined small and wide-angle scattering experiments explored sodium diffusion in the anode in Na-ion batteries [11]. In another case, QENS and INS spectra were measured to unravel the intricacies of an amorphous Li_XSi anode material [9]. Utilizing synergistic structural and dynamic probes is common in the literature. For instance, a combined approach employing QENS and SANS has been used to gain insights into the limited rate performance observed in solid-state polymer batteries [12].

2.4 SECOND TARGET STATION

Optimal use of a power-upgraded SNS accelerator and advances in target and moderator technologies will make STS the world's leading peak brightness source for cold neutrons. With advancements in neutron optics and shielding techniques and innovative design, STS instruments will gain ~100x better performance, enabling new science, for example:

- Time-resolved measurements to observe materials in action – as they are made, as they perform, as they fail, and as underlying chemical or physical processes unfold.
- High-brightness beams to measure small samples of newly discovered or synthesized materials.
- Simultaneous structural characterization of hierarchical materials from Å to microns.
- Using high brightness and broad perspective (bandwidth) to reveal complex behavior in materials in extreme environmental conditions.

STS provides transformative opportunities to capture the dynamics of complex materials systems in real-time, facilitating the establishment of quantitative links between atomic-level solvation structures, reaction pathways, and nanoscale morphologies. The STS project will construct eight instruments to address key science themes identified in the STS First Experiments Report [13]. Each of these instruments takes full advantage of the unique STS source characteristics. The instruments include,

- BWAVES – broadband spectrometer [14]
- CENTAUR – small-angle and wide-angle scattering instrument [15]
- CHESS – cold neutron spectrometer [16]
- CUPI2D – neutron imaging [17]
- EXPANSE – wide-angle neutron spin echo [18]
- PIONEER –single-crystal diffractometer [19]
- QIKR – kinetics reflectometer [20]
- VERDI – polarized diffractometer [21]

3. EMERGENT SCIENCE NEEDS FOR ENERGY MATERIALS

The key questions guiding the design of new electrochemical energy materials entail understanding the connection between atomic-scale reactions and mesoscopic/macroscopic material structure and transport. For instance, in energy storage materials, processes like charging, discharging, and chemical reactions at the atomic scale impact electrode structure and overall device performance. On the other hand, linking reaction and assembly mechanisms across length scales is crucial for controlling hierarchical assembly and using crystallization reactions to tailor device performance. Due to the diversity of this field, it is impossible to cover all relevant themes in this single-day workshop, and we have focused on several key topics to guide our discussion.

3.1 BATTERIES

In addition to lithium-ion batteries that are (almost) technically mature, there are many potential battery materials that we don’t know how to synthesize or assemble into a battery. Calendar life remains a significant issue for battery technologies based on silicon, sodium, and lithium metal. For example, the state-of-the-art calendar life of Si cells is around 20 months, far below the DOE target of 10+ years [22]. To address these challenges, we need to track the evolution over time of interfaces and structures. Integrating long-duration energy storage with renewable power generation remains a critical challenge.

Addressing this challenge requires developing membranes that can prevent the diffusion of active species, enhance ion transport while reducing over-voltages, improve stability with time and potential, and reduce cost.

Conversion cathodes are an emerging research direction that promises cheap and high-power batteries. However, many open questions remain about these materials, including how to prevent sulfur from solubilizing and transporting the electrolyte to the anode, introduce electronic conductivity to fluorides, and achieve percolation through dilute metal networks.

In addition, researchers need to understand better the solid-electrolyte interphase (SEI) layer that forms on the surface of lithium metal anodes. Questions include understanding whether molecular segregation initiates SEI formation and the evolution of the structure of the SEI layer upon applying a potential. Traditionally, SEI studies have been conducted *ex-situ*, meaning that the battery is disassembled, and the SEI layer is washed and studied outside the battery environment. However, this approach does not provide a complete picture of the SEI layer, as it can change when the battery is disassembled — *operando* experiments conducted on working batteries are needed to understand the SEI layer better.

3.2 HYDROGEN FUEL

Hydrogen is a clean and versatile energy carrier that can be produced from renewable resources. To advance hydrogen technologies, new electrocatalysts are needed that are more active, durable, and cost-effective than existing ones, including catalysts for oxygen evolution and reduction reaction (OER/ORR), hydrogen evolution and reduction reaction (HER/HRR), and carbon dioxide reduction reaction (CO2RR). Platinum group metals (PGMs) are the key catalysts for many electrochemical reactions, but they are expensive. PGMs also have low electrocatalytic surface area and require a high loading. New synthesis methods are needed to improve morphology control of PGM and develop new PGM-free catalysts with higher activity [23].

Applying catalytic materials to a membrane for hydrogen fuel cells using an inkjet process is an emerging technology with potential advantages. It is critical to integrate commercial and R&D-developed catalysts into the ink and establish the relationship between catalyst properties and ink characteristics. Different types and quantities of ionomers are needed to bind the catalyst particles together and to the membrane electrode assembly (MEA) substrate. The types and quantities of the solvent need to be optimized for fuel cells, water electrolyzers, and CO2RR applications. The impact of scalable MEA manufacturing methods and the roles of substrates and drying on defect-free coating are also important research directions. Extensive and multimodal layer characterization is critical to identifying the optimal paths for catalyst layer manufacturing.

3.3 AMMONIA PRODUCTION

Ammonia (NH_3) synthesis represents one of the most important industrial processes, but it is burdened with high energy demands. The industry accounts for approximately 1.8% of the world's energy consumption and releases 500 million tons of CO_2 annually, contributing 1.8% of global CO_2 emissions [24, 25]. There is a burgeoning interest in developing more energy-efficient ammonia synthesis methods. A crucial milestone in the ammonia synthesis process involves the creation of a hydride to facilitate the exchange and breaking of the $N\equiv N$ bond, which necessitates using a solid catalyst. Neutron techniques, known for their exceptional sensitivity to hydrogen, offer valuable insights through *in situ* observation. These techniques allow for a deeper understanding of the structural dynamics occurring during the reaction, revealing pathways guided by catalytic reactions that can reduce energy consumption and CO_2 footprint [8].

Electrochemical nitrogen reduction reaction (NRR) would enable a more environmentally friendly ammonia production, including the possibility of point-of-use production. However, the complex reactions at the electrode-electrolyte interface remain to be fully understood [26].

3.4 GENERAL ELECTROCHEMISTRY

Batteries and electrochemical energy storage systems are dynamic and heterogeneous, with multiple components connected by interfaces (see Figure 4, from Ref. [27]). These interfaces often contain metastable materials during charging and discharging and electronic and ionic transport occurs across multiple lengths and time scales. A significant opportunity for innovation lies in understanding how to design and assemble nanoscale materials and structures into mesoscale architectures. New approaches to effectively manage the mesoscale effects of complex reaction pathways in electrochemical systems are crucial. Experiments on operating devices are needed to quantify degradation and failure, particularly measurements that provide dynamic imaging of the degradation process. When simultaneous electrochemical and material characterizations are incorporated, these *operando* experiments can elucidate the causal relationships responsible for degradation.

STS instruments can have an immediate impact on the following research topics: (a) Interfacial properties and changes in interfacial structure during cycling; (b) Kinetics of non-equilibrium phenomena; (c) Solvation of ions, structure and dynamics of solvated ions and solvents, especially for multivalent ions (these may be critical for many energy storage applications) and (d) Changes in the structure of electrodes and electrolytes during cycling (*in situ* experiments would be ideal, but *ex situ* characterization of disassembled cells after a given number of cycles is also valuable). These research topics will benefit from the following capabilities.

- *Spatially resolved techniques to understand interface*. Ions are transferred across multiple phase boundaries as batteries charge and discharge. During this process, interfaces between electrodes and electrolytes and at membranes undergo significant changes due to spontaneous chemical and imposed electrochemical reactions, often leading to degradation. Transformational science can uncover degradation mechanisms and discover pathways to mitigate them. Scientific insights that guide the design of interfaces and associated interphases can improve device performance and extend lifetimes.

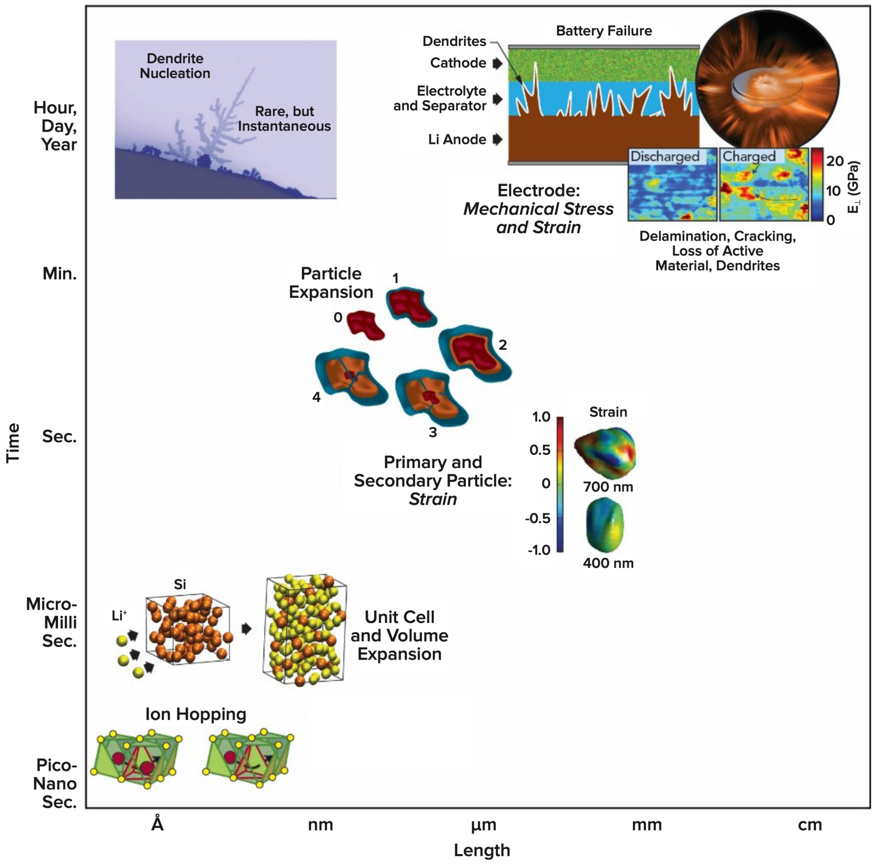

Figure 4 Battery and electrochemical energy storage systems undergo various phenomena at various length and time scales. From Ref. [27].

- *Time-resolved techniques to reveal evolution*. Electrochemistry involves dynamic phenomena that vary during charging and discharging and are heterogeneous over space and time. We must understand the temporal evolution of spatially distributed properties in electrochemical systems to enable predictive design approaches for next-generation energy storage devices.
- *Spontaneous multimodal characterization to bridge physics from broad spatiotemporal scales*. Multimodal characterizations are needed to understand complex electronic, electrochemical, and physical phenomena across time and space. As exemplified by Li-ion batteries in Figure 4, Li-ion insertion and removal induces volume changes and electrode defect formation, leading to stress, strain, degradation, and potential battery failure. Although rare events occur over short time periods and nanometer length scales, they cause effects that evolve over days and significantly impact the entire system, as shown in the upper left corner. A holistic investigation is needed to bridge the fundamental physics on the atomic and molecular scales to functional behaviors on the device level, which is critical to enabling predictive design for next-generation batteries and energy storage devices.

4. NEUTRON SCIENCE OPPORTUNITIES

Unraveling uncertainties in chemical reactions during material precipitation and particle assembly is vital for predicting device performance and designing new materials. Neutrons offer a unique capability to probe chemical actions, especially in systems involving light elements like lithium and the constituents of organic electrolytes. With a temporal resolution of seconds or better, STS will enable researchers to follow chemical reactivity in real-world situations, providing insights into specific nanoscale morphologies and aggregation behavior. Based on inputs from the workshop participants, four key neutron science opportunities have been identified in battery and energy materials for STS.

4.1 OPERANDO EXPERIMENTS

Operando experiments have been identified as one of the most promising directions for STS neutron sciences studies of batteries and energy storage materials. *Operando* electrolysis sample environments are needed for *operando* neutron scattering experiments. To enable early success at STS, a range of development opportunities have been mentioned:

- Adopt standardized sample environments and holders compatible with multiple beamlines to allow multi-modal characterization from identical samples, improving data collection efficiency and quality.
- Develop neutron-compatible electrolyzers that allow users to feed different gases and D_2O into the cell hardware. Design ultra-low loaded electrolyzer electrodes, where neutrons can provide information on polymeric ionomer distribution.
- Enable auto-sample changers to facilitate higher throughput measurements.
- Develop sample environments with optical access (laser or light) to the sample, where light serves as a probe or stimulus. Notable examples include *in-situ* photoexcitation setups at BL-4A MAGREF and photochemistry sample sticks at BL-16B VISION.
- Improve operational reliability, temperature stability and homogeneity, and sample alignment capability.
- Design sample environments with built-in collimators where feasible.

At the same time, sample environments must be seamlessly integrated into the data acquisition (DAQ) system for *operando* neutron scattering experiments. Synchronizing external stimuli, e.g., applied voltages with neutron scattering DAQ is essential. The required time accuracy varies according to the specific experiment. Electrochemical processes usually involve non-reversible phenomena; therefore, the neutron source frequency will limit the time resolution for such experiments. Consequently, millisecond synchronizing accuracy is sufficient. On the other hand, however, a ~10-μs resolution is attainable for fully reversible systems where the neutron source pulse width limits the resolution. This accuracy is crucial when, for instance, pulsed magnetic fields are used to study hidden magnetic phases. At the same time, the meta-data collected from the sample environment apparatus, such as rheometers, shall be saved with the neutron scattering data in a single Nexus file for further data analysis. Although this capability exists for many sample environments at current beamlines, expansion to more sample environments and beamlines is needed.

4.2 MULTIMODAL CHARACTERIZATION

Integrating multi-modal measurements has been discussed to enhance the characterization of complex materials at neutron beamlines. Although conventional neutron instruments are often optimized for specific techniques due to limited neutron flux, we have started to see more instruments capable of performing multi-modal characterizations simultaneously. Integrated multimode characterization is of great advantage for *operando* experiments. Prominent examples include the NExT instrument at ILL

(Neutron and X-ray Tomography instrument), simultaneous small-angle X-ray and neutron scattering at the D22 instrument at ILL, and the newly designed sample holder at VISION, SNS, enabling concurrent Raman and neutron vibrational spectroscopy. The higher flux available at STS instruments offers opportunities for further integration of multi-modal measurements at beamlines.

Additional suggestions include: (1) X-ray spectroscopy and diffraction can synergize with SANS and other neutron scattering methods, and (2) neutron scattering should be combined with conductivity and/or dielectric spectroscopy measurements.

4.3 NEW INSTRUMENT DESIGN PARADIGM

Workshop participants have recognized that software has been a bottleneck to scientific productivity for existing neutron scattering instruments. At STS, *operando* experiments with rapid data collection and multimodal characterization necessitate purposeful integration between experimentation and computational modeling for real-time experiment steering. This calls for an evolution of the instrument design paradigm.

- Time-resolved reflectometry experiments at BL4-A, SNS exemplify the urgent need for data analysis software development for kinetic studies. Though under active development, existing software needs to catch up to the data collection capabilities of neutron scattering instruments. This challenge will intensify with STS instruments' higher data collection rates.
- The demand for real-time data analysis capabilities, including rapidly converting data into easily digestible forms for real-time decision-making. Integrating AI-assisted tools for real-time experimental planning is essential for future instruments, enabling more autonomous experiments. The exploration of digital instrument twins represents an opportunity for AI model training and user support.
- A mechanism is needed to publicly share the collected neutron scattering data with the broad community for AI-model training. It requires policy changes on data sharing and the development of a protocol to include sufficient meta-data in all collected datasets.

4.4 INFRASTRUCTURE

An on-site wet lab is highly valuable to facilitate on-site sample preparation and complementary characterization. Data collected in the wet lab should be stored in the same IPTS folder as neutron-scattering data to streamline data management and accessibility.

In summary, the STS promises scientific breakthroughs by enabling simultaneous observations of processes across broad spatiotemporal scales in working devices. Additionally, neutrons’ sensitivity to light elements, like H, Li, C, O, and N, and scattering contrast between neighboring elements (such as Mn, Fe, Co and Ni) are crucial for many energy storage materials. STS will play a pivotal role in characterizing reactivity, selectivity, and kinetics associated with reaction and solution separations under realistic conditions. The benefits extend to DOE missions in science and energy, influencing the development of new materials and chemical processes for energy efficiency, production, and storage applications.

REFERENCE

1. Pérez, G.E., et al., *Neutron and muon characterisation techniques for battery materials.* Journal of Materials Chemistry A, 2023. **11**(20): p. 10493-10531.
2. Wu, X., et al., *Structural evolution and transition dynamics in lithium ion battery under fast charging: an operando neutron diffraction investigation.* Advanced Science, 2021. **8**(21): p. 2102318.
3. Browning, K.L., et al., *In Situ Measurement of Buried Electrolyte–Electrode Interfaces for Solid State Batteries with Nanometer Level Precision.* ACS Energy Letters, 2023. **8**(4): p. 1985-1991.
4. Jafta, C.J., et al., *Quantifying the chemical, electrochemical heterogeneity and spatial distribution of (poly) sulfide species using Operando SANS.* Energy Storage Materials, 2021. **40**: p. 219-228.
5. Song, B., et al., *Dynamic lithium distribution upon dendrite growth and shorting revealed by operando neutron imaging.* ACS Energy Letters, 2019. **4**(10): p. 2402-2408.
6. Ziesche, R.F., et al., *Neutron imaging of lithium batteries.* Joule, 2022. **6**(1): p. 35-52.
7. Song, G., et al., *Characterization of Crystallographic Structures Using Bragg-Edge Neutron Imaging at the Spallation Neutron Source.* Journal of Imaging, 2017. **3**(4): p. 65.
8. Yu, X., et al., *In Situ Neutron Scattering Study of the Structure Dynamics of the Ru/Ca2N: e– Catalyst in Ammonia Synthesis.* Chemistry of Materials, 2023. **35**(6): p. 2456-2462.
9. Sacci, R.L., et al., *Lithium transport in an amorphous Li x Si anode investigated by quasi-elastic neutron scattering.* The Journal of Physical Chemistry C, 2017. **121**(21): p. 11083-11088.
10. Do, C., et al., *Li+ transport in poly (ethylene oxide) based electrolytes: neutron scattering, dielectric spectroscopy, and molecular dynamics simulations.* Physical review letters, 2013. **111**(1): p. 018301.
11. Ohishi, K., et al. *Sodium Diffusion in Hard Carbon Studied by Small-and Wide-Angle Neutron Scattering and Muon Spin Relaxation*. in *Journal of Physics: Conference Series*. 2023. IOP Publishing.
12. Chen, X.C., et al., *Origin of rate limitations in solid-state polymer batteries from constrained segmental dynamics within the cathode.* Cell Reports Physical Science, 2023. **4**(8).
13. Adams, P., et al., *First Experiments: New Science Opportunities at the Spallation Neutron Source Second Target Station (abridged)*. 2020: United States. p. Medium: ED; Size: 19 p.
14. Mamontov, E., et al., *A concept of a broadband inverted geometry spectrometer for the Second Target Station at the Spallation Neutron Source.* Review of Scientific Instruments, 2022. **93**(4).
15. Qian, S., et al., *CENTAUR—The small-and wide-angle neutron scattering diffractometer/spectrometer for the Second Target Station of the Spallation Neutron Source.* Review of Scientific Instruments, 2022. **93**(7).
16. Sala, G., et al., *CHESS: The future direct geometry spectrometer at the second target station.* Review of Scientific Instruments, 2022. **93**(6).
17. Brügger, A., et al., *The Complex, Unique, and Powerful Imaging Instrument for Dynamics (CUPI2D) at the Spallation Neutron Source.* Review of Scientific Instruments, 2023. **94**(5).
18. Do, C., et al., *EXPANSE: A time-of-flight expanded angle neutron spin echo spectrometer at the Second Target Station of the Spallation Neutron Source.* Review of Scientific Instruments, 2022. **93**(7).
19. Liu, Y., et al., *PIONEER, a high-resolution single-crystal polarized neutron diffractometer.* Review of Scientific Instruments, 2022. **93**(7).
20. Ankner, J., et al., *Cinematic reflectometry using QIKR, the quite intense kinetics reflectometer.* Review of Scientific Instruments, 2023. **94**(1).

21. Garlea, V.O., et al., *VERDI: VERsatile DIffractometer with wide-angle polarization analysis for magnetic structure studies in powders and single crystals.* Review of Scientific Instruments, 2022. **93**(6).
22. McBrayer, J.D., et al., *Calendar aging of silicon-containing batteries.* Nature Energy, 2021. **6**(9): p. 866-872.
23. Cao, B., et al., *Mixed close-packed cobalt molybdenum nitrides as non-noble metal electrocatalysts for the hydrogen evolution reaction.* Journal of the American Chemical Society, 2013. **135**(51): p. 19186-19192.
24. Olabi, A., et al., *Recent progress in Green Ammonia: Production, applications, assessment; barriers, and its role in achieving the sustainable development goals.* Energy Conversion and Management, 2023. **277**: p. 116594.
25. Society, R., *Ammonia: Zero-carbon Fertiliser, Fuel and Energy Store: Policy Briefing*. 2020: Royal Society.
26. Blair, S.J., et al., *Lithium-mediated electrochemical nitrogen reduction: Tracking electrode–electrolyte interfaces via time-resolved neutron reflectometry.* ACS Energy Letters, 2022. **7**(6): p. 1939-1946.
27. Crabtree, G., et al., *Report of the Basic Research Needs Workshop on Next Generation Electrical Energy Storage, March 27 – 29, 2017*. 2017: United States. p. Medium: ED; Size: 172 p.

APPENDIX A: WORKSHOP AGENDA

STS/PSD/ESTD Workshop on Batteries and Energy Storage

8:30 am – 4:30 pm, September 20th, 2023

Contact: Talia Dix <holdertm@ornl.gov>, Office: 865-241-7667 | Mobile: 865-680-1955

Time (EDT)	Event	Speaker
8:30 - 8:40 am	Welcome and charge questions	Leighton Coates
Morning Session I: STS Instruments ***8:40 - 10:10 am Chair: Leighton Coates***		
8:40 - 9:10 am	STS Project Overview	Ken Herwig
9:10 - 9:25 am	STS Reflectometry Capabilities - QIKR	John Ankner
9:25 - 9:40 am	STS Small Angle Scattering and Imaging Capabilities – CENTAUR and CUPI^{2}D	Shuo Qian
9:40 - 9:55 am	STS Diffraction Capabilities – PIONEER and VERDI	Yaohua Liu
9:55 - 10:10 am	STS Spectroscopy Capabilities – BWAVES, CHESS and EXPANSE	Gabriele Sala
10:10 - 10:30 am	Break	
Morning Session II: Science Talks ***10:30 am - 12:00 pm Chair: Mali Balasubramanian***		
10:30 - 11:00 am	Future of Electrochemistry and Neutrons: In my opinion	Gabriel Veith
11:00 - 11:30 am	ORNL’s H2 R&D: Neutrons and Hydrogen Science	Alexey Serov
11:30 am - 12:00 pm	Dynamic Studies of Electrochemical Processes with Neutron Reflectometry	Mathieu Doucet

Lunch Talk		
12:00 -1:15 pm	Soft Matter Sample Environment for Neutron Scattering	Luke Heroux

Afternoon Breakout Sessions ***1:15 - 4:30 pm***		
Time (EDT)	Event	Lead
1:15 - 2:15 pm	2 parallel breakout sessions – I *What are the emergent science problems regarding Energy Storage and Conversion and Hydrogen @ scale that STS can address?*	Gabriel Veith Mali Balasubramanian
2:15 - 2:45 pm	Joint discussion and regroup	
2:45 - 3:00 pm	Break	
3:00 - 4:00 pm	2 parallel breakout sessions – II *How can we solve these problems using neutrons and the STS?*	Yaohua Liu Shuo Qian
4:00 - 4:30 pm	Joint discussion and summary	

APPENDIX B: WORKSHOP PARTICIPANTS

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