

*Identifying Infrastructure and
Collaborative Expertise for
Electrochemical Energy Storage
Applications*

Principal Investigators:

Nigel D. Browning & Laurence J. Hardwick

IDENTIFYING INFRASTRUCTURE AND COLLABORATIVE EXPERTISE FOR ELECTROCHEMICAL ENERGY STORAGE APPLICATIONS

Principal Investigators:

Nigel D. Browning & Laurence J. Hardwick

University of Liverpool, School of Engineering & School of Physical Sciences,
506 Brodie Tower, Liverpool, L69 3GQ. UK

Contributors:

Phoebe Allan (Birmingham)
Houari Amari (Liverpool)
Melanie Britton (Birmingham)
Laurent Chapon (Diamond/Oxford)
James Cookson (Johnson Matthey)
Paul Christensen (Newcastle)
Bill David (ISIS/Oxford)
Victoria Doherty (QinetiQ)
Robert Dryfe (Manchester)
Nuria Garcia-Araez (Southampton)
Clare Grey (Cambridge)
Sarah Haigh (Manchester)
Sajad Haq (QinetiQ)
Andrew Hector (Southampton)
Gareth Hinds (NPL)
B. Layla Mehdi (Liverpool)
Katie Moore (Manchester)
James Naismith (RCAH/Oxford)
Eduardo Patelli (Liverpool)
Sven Schroeder (Leeds)
Barbara Shollock (Warwick)
Susan Smith (STFC-Daresbury)
Andrew Stevens (OptimalSensing)
Pat Unwin (Warwick)
Philip Withers (Manchester).

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EXECUTIVE SUMMARY

A principal goal of the Faraday Battery Challenge is to provide a framework for research and development through which innovative solutions for future efficient, cost effective and recyclable electrochemical energy storage (EES) systems can be conceived and implemented. EES devices, and in particular batteries, are obviously not new (there is evidence that the basic technology is ~2200 years old), and there are many existing chemistries that are being pursued worldwide for both automotive and grid applications – everyone appears to have a favourite combination of electrodes and electrolyte that is sure to provide the best working system very soon! But how do we decide what the best approach should be, given that there will be a myriad of different applications and not enough time, funding and research expertise to try them all? Lord Kelvin famously said “*To measure is to know. If you cannot measure it, you cannot improve it*”, and for EES devices, this means that if we want to make them better, we need to perform the key set of measurements that can identify the underlying chemical mechanisms that can control performance.

In this report, we aim to identify the main characterisation methods that have the greatest potential to identify fundamental mechanisms in EES devices. The infrastructure for these methods is distributed around the UK and is available for researchers to use at large scale national STFC user facilities such as ISIS (neutrons) and Diamond (X-ray imaging and diffraction), midscale national facilities at STFC sites and universities (X-ray diffraction, Electron Microscopy, NMR, Mass Spectrometry), and within single investigator laboratories at universities and industries around the UK (optical methods, electrochemistry, etc). While there is a great deal of characterisation that is already present around the country, one of the clear results of this survey is that the expertise of the researchers to perform state-of-the-art measurements on EES systems and devices is limited to a few widely dispersed institutions. **A major advancement in the UK’s ability to innovate new EES technologies would therefore be achieved by the Faraday Institution supporting mechanisms that both provide access to the best instrumentation and provide training in its use by the experts in the characterisation of EES systems.** Practically this can be achieved through what is known as a “hub(s) and spokes” model in which there is centralised expertise at centres for the “expensive” methods (either limited equipment or limited expertise), and spokes that reach out to satellite centres that can provide unique specialised methods as well as access to resources that are limited in the centralised hubs. This hub(s) and spokes model has the added benefit of being distributed around the UK (thereby providing easy access for many researchers) and of constantly adding new methods and expertise to the characterisation

environment (fed from the spokes to the hubs), overcoming the long-term tendency of any hubs to focus on providing “routine” methods to large numbers of researchers rather than innovating new methods.

By supporting researchers to collaborate extensively in coordinated research at both hubs and spokes, the Faraday Institution can accelerate the establishment and application of unique expertise amongst UK scientists for EES research.

A key aspect of any measurement method is of course to be able to quantify it in some reproducible manner. While this seems like it should be the origin of any scientific approach, in practice it is complicated by the fact that different measurement methods can provide completely different views of any EES system – a blessing if we know how the views overlap, but a curse if we don’t. How, for example, do we correlate together NMR, Mass spectrometry and TEM measurements of the same EES system? Each experiment requires a set of unique expertise and will emphasize a different part of the overall system. When the effect of any potential impurities in the system (random errors) and the “standard practice” of the operator for the instrumentation being used (systematic errors) are taken into account, the difficulties in translating one dataset into another become clear. Coordinating a standard set of measurements, using a standard set of practices and reporting a standard set of metadata to define the experiment, is therefore another key part of using the most advanced UK facilities for characterisation to help innovate new EES devices. This coordinated approach provides strong support for the hub(s) and spokes model described above – the only way to achieve a consensus from all scientists on data reporting standards and to manage a data repository efficiently is to have researchers work together to both define the standards and to implement them. By defining the metadata behind a “certified dataset” from a given technique, the number of different experiments that need to be performed to understand a particular EES system can be significantly reduced – in many cases the most extreme arguments about differing results between researchers are more about systematic errors in the characterisation methods than any fundamental differences in the underlying mechanisms. **The Faraday Institution can therefore optimise characterisation usage, and avoid much of the wasted effort that comes with un-calibrated claims, by setting out a coordinated project and/or policy for data reporting at the outset of the battery challenge.**

Coordinating the characterisation datasets that are being generated by researchers for the Faraday Institution automatically implements another key advancement than can accelerate the innovation of

new EES systems. Machine learning is currently being employed in a wide number of “big data” applications, where it is being used to identify underlying trends that are missed by individual researchers. Machine learning works best when the metadata of the experiments is well defined. By coordinating the methods in the manner described above, key parameters for EES research can be quickly identified by machine learning methods. This will provide major advances in the use of the experimental methods by identifying which experiments should be performed to fill in the missing information – in the case of resource limitations (i.e. limited access to instrumentation) this can provide major advances by significantly reducing the number of experiments that need to be performed to understand the underlying mechanisms of performance. **By integrating scientists that generate data with scientists that analyse the underlying trends in the data into a coordinated approach to state-of-the-art characterisation, the Faraday Institution can accelerate the application of optimised EES systems for each application.**

In summary, the main characterisation methods needed to make advancements in EES systems are mostly already present at the required levels in the UK (there are a few notable exceptions, such as NMR, that are described in this report). In most cases, the limitations in their use are caused by complications in the user access models of the facilities concerned, a sparsity of researcher expertise for EES systems (expertise in the characterisation method is not an issue), and the limited availability of peripheral support instrumentation (such as specimen preparation tools and glove boxes to limit atmospheric degradation of materials). In cases where the instrumentation and expertise is limited, the Faraday Institution can support the purchase of new instrumentation in the long term and can provide access to instrumentation elsewhere in the world in the short term (for example, through research agreements with national laboratories in the USA). **By providing the support that coordinates researchers together in a “hub(s) and spokes” model for the characterisation of EES systems, the Faraday Institution can stimulate the growth in EES expertise in the UK, provide a set of “certified” data that can support a wide ranging research and development environment, and accelerate the insights needed to innovate new EES devices.**

1. OVERVIEW OF CHARACTERISATION FACILITIES IN THE UK

The need for more energy and cost efficient materials and processes for electrochemical energy storage (EES) creates a major challenge for characterization methods (infrastructure), requiring new coordinated approaches to everything from routine benchtop analyses to state-of-the-art imaging and spectroscopy at national user facilities. The origin of the challenge lies in the sheer number of possible new storage chemistries being proposed and the mostly ad-hoc way they are typically characterized – small differences in chemical purities, surface structures or testing conditions can lead to radically different results. In many cases, there is also a difference between the results obtained by expert technologists and novice users, with some researchers and laboratories appearing to “just know how to do it right”. All of these factors make it very difficult to ensure that national targets within the Faraday Institution for the development of new energy storage devices can be realized. The goal here is therefore to identify potential institution wide strategies for characterization of EES materials and devices (use of infrastructures) that provides national access to state-of-the-art facilities with the expertise needed to obtain the best results, and permits results from widely varying multi-scale and multi-modal characterization methods to be directly interpreted, accelerating the development of innovative EES technologies in the future.

To ensure that characterization can be performed to global leading standards and it is available to researchers at every level of academic and industrial research in EES, the whole community must be engaged in identifying the methods most important to EES and the reporting requirements needed for the results to be accepted as valid. In addition, ensuring that there is unnecessary repetition of efforts, data scientists working on compressed methods, machine learning and data mining need to be integrated into the EES community to ensure optimal use of resources for the development of new systems – there is little point in everyone doing the same experiment, while equally there is a need for coordination to contribute to the overall knowledge in the field. This engagement with the community has been achieved here by involving it in a wide survey of EES characterization methods followed by a workshop to discuss methods and practices for EES research (see appendices for details of both the survey and the workshop). The survey aimed to identify both the current standard and potentially novel future methods for EES characterisation while the community workshop involved keynote speakers from international energy storage research programs and leading characterization experts in the UK to identify the priorities and mechanisms for interaction going forward. Together these activities aimed to synergize efforts with the developing UK roadmap for research and innovation infrastructure and provide the following deliverables in this report:

- Identify the key small, medium and large scale infrastructures and expertise that can promote UK innovation in energy storage materials and technologies
- Assess whether the infrastructure is currently available in the UK, could/should be developed or could/should be obtained through international partnership
- Examine capital purchase options, sharing models and access mechanisms that can ensure the widest use of the existing key infrastructures to accelerate UK innovation
- Highlight infrastructure gaps where there is critical need for new investment and how/where it can be implemented for maximum impact

1. Status of Characterisation Facilities in the UK

There are many methods that can be used to characterise electrochemical energy storage (EES) systems. In this section, the goal is to define which methods can have the most impact in the characterisation of current systems, and that have the potential to provide fundamentally new insights into new materials and processes for the future (innovation). Here we break down the methods that are/ can be used into those that require large scale national facilities (such as synchrotrons), those which are generally termed as mid-scale (such as electron microscopy) that are also classified in the UK as national facilities, and small scale individual user facilities typically operated by a single investigator at universities. These imaging/characterisation techniques can be the cornerstone of research in EES materials and devices going forward, providing multi-scale information from the atom to the cell, under operando, in-situ, ex-situ and post mortem conditions. Unique datasets (which can be 2D, 3D or 4D) can be obtained variously from electron, ion beam, neutron and X-ray microscopy (alongside coupled spectroscopy), and all have relative merits in their sensitivity, temporal and spatial resolution, sample preparation requirements and their ability to penetrate matter. The goal is that when performed in concert, these techniques are (and will be even more so in the future) much greater than the sum of their parts: for example, the correlation of X-ray data with neutron data provides the coupled benefits of sensitivity of both nuclear and electron density, similarly the correlation of electron and ion beam microscopy can extend 2D surface investigations to 3D serial sectioning. If the EES community can harness the capabilities

described here, the UK will be well positioned to tackle key challenges across time and length scales, for the wide range of new chemistries, materials and dynamic processes that will be needed for innovation in EES systems in the future.

1.1. Methods Using Large Scale National User Facilities

The UK has pioneered the development of national user facilities since the late 1950s with the establishment of neutron scattering research programmes at Harwell based at the DIDO and PLUTO nuclear reactors. In 1981, the Synchrotron Radiation Source (SRS) at Daresbury was not only the first second-generation X-ray synchrotron source, but also the first dedicated X-ray synchrotron user facility. Today, the co-location of their modern equivalents, ISIS and Diamond, on the Harwell Campus alongside the Central Laser Facility puts the UK in an excellent position to utilise the opportunities and capabilities of these national user facilities across the Industrial Strategy and for the particular benefit of all three strands of the Faraday Challenge. In addition to these large-scale facilities at Harwell, the UK also has access to the ESRF and ILL in Grenoble, France, and is committed to future involvement with the European Spallation Source (ESS) in Lund, Sweden and XFEL, the European Free Electron Laser based in Hamburg, Germany.

There is a massive infrastructure capability on the Harwell Campus with a total of 69 large facility instruments at Diamond (31 X-ray beamlines; 2 electron microscopy centres for life science and physical sciences - eBIC and epSIC) and ISIS (32 neutron and 6 muon) and these are complemented by additional support facilities at Diamond, ISIS and the Research Complex. There is also a substantial cohort of scientists and

engineers who are actively involved in energy research across campus with over 100 members of the Harwell Energy Materials Research Network. Each instrument at Diamond and ISIS costs “£5-10M to design, build and commission, and is managed and operated by a small team of scientists. Furthermore, each instrument is supported by a large infrastructure of engineers, computer scientists and technical support from within the facilities and also elsewhere within STFC. Around 20% of the experiments performed at Diamond and ISIS are related to energy systems and materials, and the major component of this research is in battery science and technology.

New instruments, instrument upgrades and sample environment developments are project managed within the facilities and involve consultation and support from the external academic and industrial user community. Diamond and ISIS are working closely together to improve coordination and already have mechanisms in place for single submission of proposals where access is required for both synchrotron and neutron beam-time. Joint appointments between Diamond and ISIS have been instituted where there is a strong complementarity of instrumentation, for example in the area of X-ray and neutron pair distribution function analysis. There is, additionally, a substantial overlap in data analysis and interpretation that is shared between the facilities and an automatic approach for XPDF (I15-1) and nPDF (POLARIS) measurements is being developed. While sample environment requirements for complementary experiments can differ in scale and complexity between ISIS and Diamond, there is a clear impetus to standardise data analysis procedures and equipment to ensure that precise, accurate and repeatable measurements can be made across facilities.

1.1.1. Diamond Light Source

Diamond, the UK’s third-generation synchrotron, is actively engaged in energy research which represents a large part of the scientific

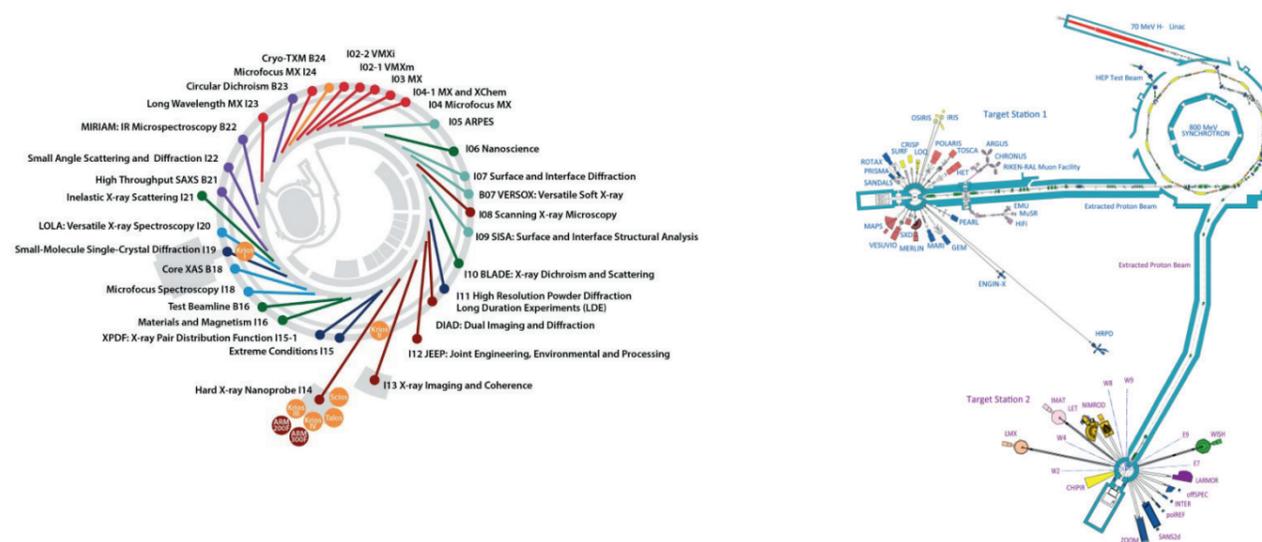


Figure 1: Instruments at Diamond & ISIS. ca.40 of these are relevant to the Faraday Challenge.

activity (about 20%) carried out in collaboration with academic users and industry. Out of the 31 X-ray beamlines (33 by the end of 2019), operating 200 days a year, 15 have programs in Energy research currently (complemented recently by Electron Microscopy with ePSIC), many of them actively involved in the study of EES. Diamond offers:

- 1) A wide range of X-ray scattering and spectroscopic instruments to probe the structure, chemical state and electronic properties of energy materials. This includes:
 - High resolution and time-resolved powder diffraction with in situ/ in-operando capabilities to study the structure of crystalline phases.
 - A powder diffraction facility (LDE: Long Duration Experiments) dedicated to the study of materials that need to be maintained at, or cycled through, various non-ambient conditions for extended periods of time. Several experiments can be run in parallel, including battery cycling.
 - A beamline for Pair Distribution Function to study the structure of partially disordered materials. We are currently building dedicated sample environments such as electrochemical cells.
 - X-ray core level spectroscopies (including soft and hard X-ray absorption spectroscopy soft and hard X-ray photoemission, X-ray fluorescence and resonant inelastic X-ray scattering), including a high-throughput beamline for soft XAS and XPS, near-ambient pressure XPS, micro and nano-focused beams, soft X-ray Inelastic Scattering to study the oxidation, spin state and chemical coordination with elemental selectivity. This infrastructure supports advanced surface, interface and bulk analysis, with capability for in situ and operando process studies. Multimodal experiments combining scattering and spectroscopy are also available in situ/ operando.
- 2) A suite of imaging and microscopy beamlines to map (in 2D, 3D and 3D+time) the density, composition and structure of materials from nanoscale to mesoscale. This includes:
 - Absorption contrast and phase contrast radiography and tomography. Phase contrast can increase the contrast from low

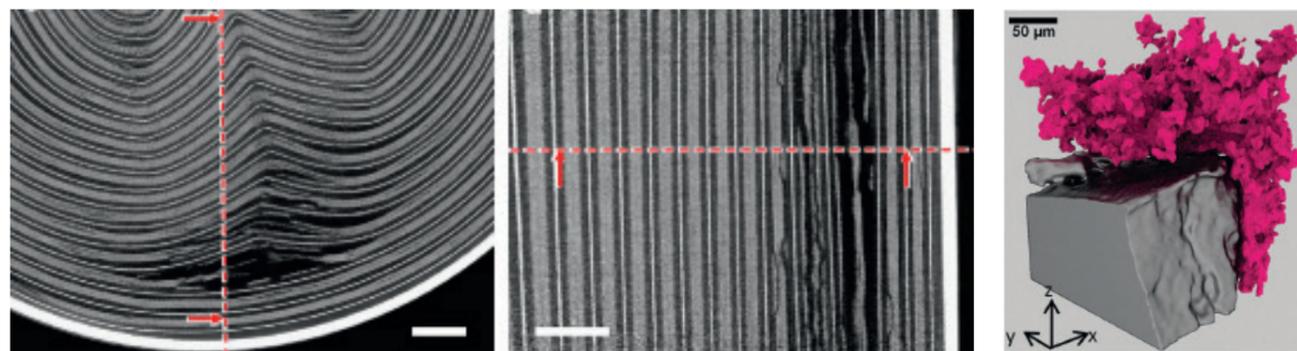


Figure 2: Grey-scale slices from 3D CT reconstructions during in-operando high-frequency tomography of a cell, (a) enlarged grey-scale image of the XY plane 1s before thermal runaway; (b) enlarged grey-scale image of the YZ plane 1s before thermal runaway. Scale bar, 1 mm [2]. (c) The electrodeposition of metallic lithium is a major cause of failure in lithium batteries. Here a phase contrast X-ray CT image of lithium moss formed during galvanostatic cycling is shown [1]. (Images courtesy of Paul Shearing, University College London)

atomic number containing materials such as lithium. The very high x-ray flux also opens the possibility to conduct fast time-resolved in situ studies at micrometer resolution using hard X-ray (to penetrate heavier materials such as steel battery walls), for example to study thermal runaways in batteries [1] (Figure 2). The non-destructive nature of X-ray imaging allows in-situ and operando studies, yielding unique 4D information.

- Hard X-ray nanoprobe with sub 100 nm focussed beam for imaging materials using spectroscopic and/or diffraction mode.
 - Tomo-ptychography with tens of microns field of view and sub 20nm resolution, and Bragg ptychography to map strain in crystalline nanoscale materials.
 - Transmission X-ray microscopy as well as TEM (the epSIC facility).
- 3) The development, in continuing collaboration with ISIS, of new techniques and methods, including data analysis, to support experimental science and the access to ancillary equipment for sample preparation and characterisation directly at Diamond, and elsewhere at complementary facilities on the Harwell Campus at the Research Complex at Harwell (RCaH) and ISIS.

For academic users, Diamond is free at the point of access and beam-time allocation is determined by a peer review process run by international groups of experts. Currently, there are 4 main access routes for Non-Proprietary beam-time (full details available at <https://www.diamond.ac.uk/Users/UserGuide/Proposals/Access-Routes.html>):

- Standard access, i.e. individual proposals, evaluated every six months.
- Block Allocation Group (BAG) access: BAGs consist of groups of users who apply jointly for beam-time spread over a period of two years at a time. BAGs usually consist of a number of researchers from the same research institute or of researchers from smaller institutes in the same region of the UK.
- Programme Access - Long Term Research

- Available for groups to support areas of research that require sustained and guaranteed access to beam-time for their success.
- Rapid access
- Proprietary Access Routes are available for industrial users with different service levels.

1.1.2. ISIS Neutron and Muon Source

ISIS is a world-leading centre for neutron and muon scattering research with a suite of 32 instruments located around two target stations. Together, these instruments are optimised to exploit a broad range of neutron and muon energies and span a wide range of disciplines from magnetism to cultural heritage, engineering to food science, and chemistry to environmental science. The benefits and unique aspects of neutron and muon scattering coupled with the strengths of the facility have been responsible for the emergence of a world-class research programme at the facility, covering topics at the forefront of Physics, Chemistry, Materials Science, Earth Science, Engineering and Biology.

The principal access mechanism to ISIS, 'direct access', is through the two annual peer-reviewed calls that are assessed by seven international panels of experts. ISIS also operates the following access routes which can be exploited by Faraday-related research:

- Rapid Access: full proposals, submitted at any time, requiring more rapid access to beam-time for time-critical studies. Around 10% of ISIS beam-time is currently given out to rapid access proposals.
- Xpress Access: relatively quick (ca. 1hr), simple measurements to, for example, complete a data set or test a sample for neutron/muon response before a full proposal. Samples are mailed in and the experiments run by an ISIS scientist.
- Industrial Collaborative R&D route: for industry to access the facility. This route enables a decision on payment for beam-time (i.e. whether the work is to be kept commercially confidential or published) to be made after the experiment. It is a popular, low-risk route for industrial users.
- Direct purchase of beam-time: normally by an industrial user who knows when they apply for beam-time that they wish to keep the work commercially confidential.

Over the past 35 years, ISIS has developed significant infrastructural support providing a wide range of state-of-the-art sample environment equipment, support labs and technical expertise to support the ISIS science and engineering programmes. There is routine provision of equipment for experiments covering low and high temperature, pressure, gas handling, magnets, motion and temperature control and electronics. Standard sample environments cover temperature ranges from 25 mK up to 2300 K, pressures from vacuum to 25 GPa, magnetic fields up to 14 T. Of specific importance for the Faraday Challenge, ISIS, in conjunction with academic and industrial researchers, has also developed and operates in-situ and operando equipment that enables on-beam, time-resolved measurements of batteries and other

electrochemical devices; dedicated teams support this equipment. ISIS also has four well-found laboratories for off-beam characterisation equipment (X-ray, spectroscopy, and magnetic, electronic and thermodynamics measurements) and sample preparation facilities, all supported by a dedicated laboratories team.

Novel sample environment and technique developments are generally undertaken in close collaboration between ISIS scientists and engineers, and the academic and industrial user community. The development of the Paris-Edinburgh high pressure cell at ISIS is a key exemplar of this approach which led to the construction of PEARL, a dedicated high pressure diffractometer at ISIS. This, in turn, has resulted in similar high impact instruments at neutron facilities around the world and a renaissance of high pressure neutron materials science.

The current development of NMR capabilities on NIMROD is set to have a similar international impact and offers a blueprint for the development of new beamline capabilities for the Faraday Challenge. NeuNMR is an EPSRC-funded project with three principal partners: Manchester, Cambridge and STFC-ISIS, along with two other partners in Johnson Matthey and Queen's University Belfast. The purpose of this collaborative project is to install, commission and operate in-situ NMR equipment on NIMROD to enable simultaneous NMR and neutron total scattering measurements with the aim of better understanding liquid phase heterogeneous catalysis in order to develop catalytic processes. The partners have created a small, focussed and blended team of knowledge and experience. Manchester, Cambridge and ISIS respectively bring expertise in catalysis and chemistry, neutron scattering, and NMR and chemical engineering while Johnson Matthey guides the choice of catalysts and Queen's University Belfast synthesises and prepares model catalysts.

ISIS and Diamond continue to forge closer links in the development of related techniques such as total scattering measurements, small-angle scattering and reflectometry. This includes experimental design, data analysis, and the closer alignment of access mechanisms. Furthermore, ISIS will continue to support its ancillary equipment for sample preparation and characterisation facilities that are directly pertinent to the ISIS science programme, and will also work closely with other complementary preparation and characterisation facilities on the Harwell Campus at the Research Complex at Harwell (RCaH) and Diamond.

A selection of examples of neutron scattering applications for EES research is listed below. Other areas of impact include (i) neutron reflectometry for the study of surfaces and interfaces at the atomic to mesoscale length-scales, (ii) small angle neutron scattering for the study of mesoscale structure from the nanometre to the micron, (iii) molecular spectroscopy using inelastic neutron scattering for the study of molecular vibrations, (iv) quasi-elastic neutron scattering for the study of the dynamics of atoms, ions and molecules and (v) single-crystal inelastic neutron scattering for the fundamental study of battery materials dynamics.

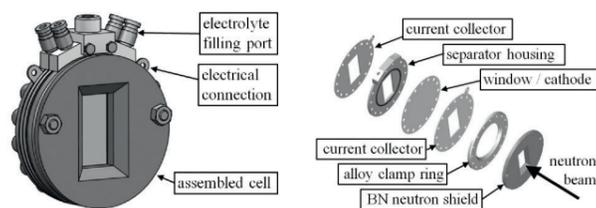


Figure 3: In-situ electrochemical cell for neutron observations.

1.1.2.1. In-situ neutron powder diffraction

Neutron powder diffraction is extensively used to probe the structure-property relationships within materials of technological importance in the fields of energy production and storage. These studies often exploit the sensitivity of the neutron method to the locations of light elements such as hydrogen, lithium and oxygen in the presence of heavier species. In recent years, there has been increased interest in the development of in-situ and in-operando methods which reproduce, as closely as possible, the conditions experienced by the material within its application. These include electrochemical cells which allow time-resolved diffraction studies of battery electrode materials to be performed during charge-discharge cycling and facilities to study oxide-ion conducting ceramic electrolytes under temperature and atmosphere conditions experienced within solid oxide fuel cells. Recent in-situ projects, with an electrochemical cell (see Figure 3) developed in collaboration with the University of Sheffield and Chalmers University, Sweden, have probed the structural changes in lithium-ion and nickel-metal hydride electrode materials during the charge/ discharge process [3-5].

1.1.2.2. Neutron imaging

IMAT (Imaging and Materials Science & Engineering) at ISIS is a new concept in neutron imaging and diffraction instrument for materials science, materials processing and engineering. The special features of the instrument will be energy-selective neutron imaging and the combination of neutron imaging and neutron diffraction. Currently IMAT is under engineering commissioning. IMAT will offer a combination of imaging and spatially resolved diffraction modes such as neutron radiography, neutron tomography, energy-selective imaging, neutron strain scanning, crystallographic structure and phase analysis, texture analysis, and non-destructive testing. A broad range of imaging and diffraction applications will be possible for energy materials, aerospace and transportation, civil engineering, power generation, earth sciences, cultural heritage and agriculture. IMAT will strongly complement the imaging facilities at Diamond.

1.1.2.3. Wide Q-range Total Neutron Scattering

Total neutron scattering is a powerful technique that measures the bulk pairwise structural correlations between atoms that make up a sample, regardless of whether its state is crystalline, amorphous, liquid, or some combination thereof. Performing this measurement over

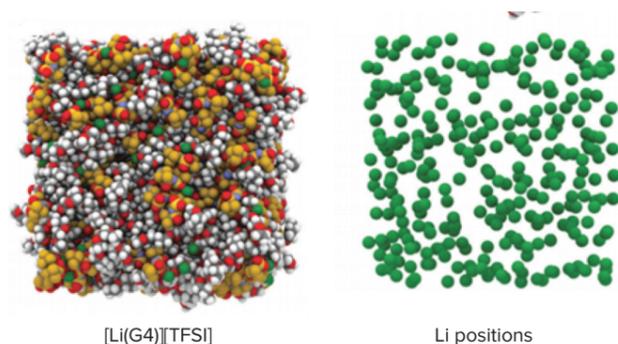


Figure 4: Resolving Li positions in the bulk nanostructure of prototypical good solvate ionic

a wide range of magnitude in the momentum transfer vector of the scattered neutrons, such as from $<0.02\text{\AA}^{-1}$ to $>50\text{\AA}^{-1}$ that is available using the NIMROD instrument of the second target station of the ISIS Pulsed Neutron and Muon Source, can give direct insight into materials structure on length scales up to $\approx 30\text{nm}$, with sub-Ångström resolution for interatomic distances. Although this type of data is inherently complex to analyse and interpret, utilising methods that further refine the technique, such as H/D or $^6\text{Li}/^7\text{Li}$ isotopic substitution, can provide additional chemically specific insight that is often crucial for properly accounting for observed functional structure-property relationships. A particular strength of the method is its highly quantitative nature, in that the measured differential scattering cross sections are calibrated to an absolute scale of barn per steradian per atom. This means that these data can be used to directly benchmark atomistic and mesoscale structural models. Within the EES area, these methods are finding increasing application in the comprehensive characterisation of liquid electrolytes [6], and filled or empty porous or nanostructured electrode materials [7], whether under static or operando conditions inside in-situ electrochemical cells.

1.1.2.4. Muon – probing ionic motion

Positively charged muons at ISIS are used to study the motion of nuclear magnetic moments such as Li and Na in EES materials. The technique probes ionic motion at the atomic level on microsecond timescales. This activity has grown significantly in the past decade following a pioneering study of LiCoO_2 by a team from Toyota Central Research and Development Laboratory [8], who remain highly active users of this facility. Since then several UK University groups and European groups have emerged. In collaboration with the muon group at ISIS, they are developing in-operando cells (Toyota, Stockholm and Glasgow) and extending the technique to other ions such as Mg (Oxford in collaboration with JCESR researchers). Negatively charged muons at ISIS are used to study the elemental composition of materials in a technique akin to X-Ray Fluorescence. The advantages of this technique are controlled probing of depths up to $\sim\text{cm}$ into materials and sensitivity to all elements. Initial studies of EES materials have been performed by academic groups in the UK and Toyota.

1.1.3. Research Complex at Harwell (RCaH)

The RCaH is a laboratory owned by the UK research councils and Diamond, it is based at the Harwell Campus. Its mission is to carry out research that benefits from the large Central Facilities (Diamond, ISIS, CLF) and is of benefit to them. Between them the central facilities cover electrons, neutrons, X-ray photon and light photons. The RCaH works extremely closely with the central facilities with partnership around equipment / experimental rigs; providing lab scale facilities that allow preparation / experimentation before use of central facilities. The RCaH has glove boxes, TEM, SEM, XRD, advanced gas capability, workshop and 3D printing. The RCaH has high quality lab space and operates on a “Hotel” model; that is groups with shareholder funding (UKRI, Diamond) can utilise it free at the point of use. The RCaH has focussed on providing cutting edge laboratory infrastructure for consortia in order that they can most effectively exploit and engage with the central facilities. In recent years, the most high profile of these has been the Catalysis Hub. The Hub at the RCaH sits in the centre of a nationwide network of Industry and Universities. The Hub at Harwell is equipped to support the most challenging *in operando* experiments and is open to all members of the consortium. Breakthroughs have included tomographic reconstructions of catalysis during turnover and neutron imaging of zeolites. Hub at the RCaH contains a small core of highly active researchers and has proven an excellent location for career progression; with several young investigators becoming established at senior level. This model has been adopted by other efforts, including advanced manufacturing and materials. In anticipation of the Faraday Challenge, the RCaH is in the process of creating of two new laboratories, each of 114 m^2 . We have allocated funding to equip one of these labs for use by battery researchers. The RCaH will work with the battery community and coordinate with ISIS, CLF and Diamond to make sure that a suite of equipment is available for in operando research across the campus.

1.2. Methods Using Mid-Scale National Facilities

Mid-scale facilities include instrumentation that is typically too large or too expensive to be operated by a single investigator. Instrumentation in these facilities are usually grouped together to allow technical staff to be employed for their operation and they typically involve some form of re-charge for their operating expenses. In the EES field, the major examples of this type of facility are electron microscopes, nuclear magnetic resonance (NMR) and mass spectrometry instruments (MS). A number of these facilities are classified as National Facilities (originally medium scale facilities) and are partially supported by the EPSRC for wider access; such as XMAS at the ESRF [9], the National EPR Facility at Manchester [10], the UK Solid State NMR facility at Warwick [11] and the National Mass Spectrometry Facility (NMSF) at Swansea [12]. In addition, a significant collection of National Facilities for materials characterisation resides within the Royce Institute for Advanced

Materials [13] (facilities at Cambridge, Imperial College, Liverpool, Leeds, Oxford, Sheffield, Manchester, National Nuclear Lab and Culham Centre for Fusion Energy). These types of facilities experience similar challenges for operation and development of expertise (see section 3). Here we highlight the major facilities in the UK with either the expertise in, or the capacity to work on, EES systems.

1.2.1. Electron Microscopy

Electron microscopy typically encompasses such instruments as scanning electron microscopy (SEM) including focussed ion beam (FIB) and Electron Probe Microanalysis (EPMA), Transmission Electron Microscopy (TEM), Scanning Transmission Electron Microscopy (STEM), Helium Ion Microscopy (HIM) and Atom Probe Tomography (APT). These microscopes are used in the preparation and analysis of battery materials to obtain diffraction patterns for phase identification; atomic scale images for nanostructure, interface and defect analysis; electron energy loss spectra (EELS) and energy dispersive X-ray Spectra (EDX) for chemical analysis; 3-D images of nanostructures and atomic scale dopant distributions; surface morphologies and microstructures. The cost of the instrumentation ($\sim\text{£}500\text{k}-5\text{M}$), the annual operating costs ($\sim\text{£}20\text{-}150\text{k}$ per microscope per year), and the expertise needed to operate the microscopes classifies these facilities as mid-scale operations and most universities and many companies in the UK have some form of electron microscopy available. Here, however, we focus on the specialist areas of microscopy that can provide significant insights into the development of battery materials and systems. It should be noted that for these facilities there are many different access models, from free at the point of use, to peer reviewed proposals, to user charges of several thousand pounds per day (access models for mid-scale instrumentation will be discussed in section 3 of this report).

1.2.1.1. (Scanning) Transmission Electron Microscopy

TEM/STEM methods were revolutionized 20 years ago with the development of aberration correctors. Aberration correctors significantly enhance both the spatial resolution and sensitivity of images and chemical analyses performed in the TEM/STEM. As such the leading microscopy centres in the UK feature aberration corrected TEM/STEM [14-25]. While these facilities offer many different imaging capabilities, they tend to focus either on a particular method of expertise or a general set of applications for a particular materials class – such as 2-D materials or semiconductors. For example, the UK SuperSTEM and Leeds focus on atomic resolution imaging and monochromated EELS [14,15], the ePSIC facility at Diamond is developing and applying atomic resolution TEM/STEM methods [16], the Imaging Centre at Liverpool pioneered operando TEM imaging of chemical and electrochemical battery processes [17], Cambridge developed 3-D imaging by tilt tomography [18], at Oxford the goal is advanced ptychographic methods in STEM and atomic resolution imaging in TEM [19], at Glasgow the facilities are geared towards

imaging magnetic structures [20] and at York the main research microscope aims to study catalytic systems in-situ during gas reactions [21]. On the other hand, Warwick [22], Sheffield [23], Manchester [24], Imperial [25] and Birmingham [26] aim to use multi-modal imaging and spectroscopic methods for wide ranging materials challenges. While many of these centres describe an emphasis on “Energy Materials”, this covers everything from batteries to nuclear systems and specific programs on energy storage materials and devices is so far in its infancy.

1.2.1.2. Atomic Resolution Imaging and Spectroscopy

Atomic resolution imaging in both TEM and STEM has been shown to provide direct evidence of the secondary phase formation and the intercalation pathways for ions from the electrolyte into the cathodes typically used for Li-ion [27-31], Li and zinc-air batteries [32-34] and other multi-valent systems [35-37]. Usually experiments are performed before and after electrochemical cycling to determine the rate of intercalation and secondary phase formation. These atomic resolution images were the first to identify that many of the materials used for cathodes in Li-ion batteries were not phase pure, and the distribution of the secondary phases in the initial sample, and during cycling, could have a significant effect on the electrochemical performance of the cell. While the resolution and sensitivity of the microscopes in the facilities described above are slightly different at each, all of the ones listed can perform these types of atomic resolution analyses of battery materials (access must be dealt with locally, but is available at short notice in many places).

1.2.1.3. 3-D Imaging: STEM and Atom Probe Tomography

The atomic resolution analyses described above are limited by the total volume of materials that can be analysed (typically $\sim 1-10\text{nm}^3$ for each experimental image). To improve the understanding of the chemistry over the $10-1000\text{nm}^3$ volume, 3-D imaging methods have been developed. In the case of the STEM, tilt tomography methods pioneered at the University of Cambridge [18], have been shown to be valuable in identifying the distribution of secondary phases

in nanostructured cathode materials [27]. An alternate method of identifying 3-D distributions of elements is atom probe tomography – a form of time of flight mass spectrometry where the atoms are evaporated from a small tip and collected on a position sensitive detector. Although there are specimen preparation difficulties involving cycled battery materials, APT has been shown to be very sensitive to elemental distributions in cathode materials before/after cycling [38]. The main centre for APT in the UK, at the University of Oxford [39] has the capabilities to perform these types of battery analyses.

1.2.1.4. Operando Electrochemical Cells for (Scanning) Transmission Electron Microscopy

One recent advance in scanning transmission electron microscopy (STEM) has been the development of electrochemical liquid stages that can be used to observe the dynamics of the charge/discharge process at electrode-electrolyte interfaces [40-52]. The development of these stages can be traced to direct funding by several U.S. Department of Energy research programs, such as the Joint Centre for Energy Storage Research (JCESR) [53], the Batteries for Advanced Transportation Technologies (BATT) program [54] and Energy Frontier Research Centres (EFRC) [55]. In all of these cases, the technology was co-developed with a small business that now markets the electrochemical stages to fit into all the commercially available microscopes (stages can be purchased from Protochips [56], Hummingbird [57] and DENSolutions [58]). Microscope user facilities at the Universities of Cambridge [19], Oxford [20], Manchester [25] and Diamond [17] are all currently working with versions of these stages and establishing baseline conditions for their operation. Additionally, the expertise behind the original stage development is currently at the University of Liverpool [18] (an example of this work is shown in Figure 5), with new stage modifications and the implementation of novel compressive sensing imaging modes [59] to improve both the spatial/temporal resolution and chemical sensitivity of these observations currently underway. Additional developments at the University of Liverpool (funded by the Faraday Institution) are expanding these electrochemical stages to include solutions optimized for He-ion microscopes (HIM) and scanning electron microscopes (SEM) – these instruments allow for directly correlation of the battery scale condition for wide range

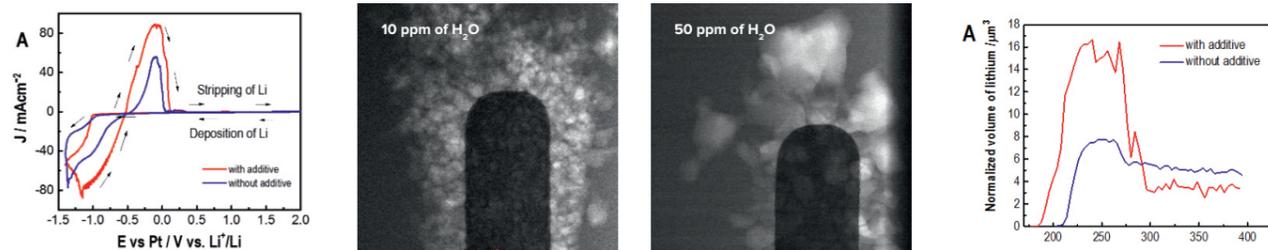


Figure 5: Observation of the deposition of Li on a Pt electrode during the charge/discharge cycling of a LiPF₆/PC electrolyte with 2 different levels of H₂O impurities (50ppm and 10ppm). The differences in the coulombic efficiency (a) can be related to differences in the morphology of the Li deposits (b and c) and the overall amount of Li deposited and stripped (d). This study shows that the microstructure of the deposited Li has a major effect on the overall performance of the battery system [50]. (Images courtesy of Nigel D. Browning and B. Layla Mehdii, University of Liverpool).

of applications among which are degradation and recycling Faraday Institution fast start projects.

1.2.1.5. Scanning Electron Microscopy (SEM) and Helium Ion Microscopy (HIM)

In contrast to TEM/STEM and APT, scanning electron microscopes offer the opportunity to characterise larger volumes of material from the nanoscale to the millimetre scale. Scanning electron microscopes are nearly ubiquitous in UK universities and range from desktop versions to high-resolution field emission systems. In general, images are generated using electrons generated from the interaction of the primary beam with the sample - secondary electrons to obtain topographic information and backscattered electrons to image based on crystallographic and/or atomic number differences. The majority of SEMs are equipped with ancillary equipment to provide information complementary to images. Typical configurations include energy dispersive X-ray (EDX) for qualitative or semi-quantitative compositional analysis, wavelength dispersive spectroscopy (WDS) for quantitative chemical analysis, and electron backscattered diffraction cameras for orientation imaging and phase identification of crystalline materials. These instruments are particularly important as battery materials often have complex 3D microstructures containing a range of pores and insulating materials. Newer SEM instruments often provide information beyond the standard modes, with probe currents from a few pA to up to 700 nA. Innovative in-lens and in-column detectors for secondary electrons and energy selected backscattered electrons have enabled high-resolution imaging ($\sim 1\text{nm}$) at low accelerating voltages and combined with beam deceleration, spatial resolutions of $\sim 0.8\text{nm}$ can be achieved [60,61]. Gas injection systems can be used for charge compensation on insulating samples, removing the need for coating.

An additional challenge exists with characterisation of Li-ion batteries, their sensitivity to air - methodologies for minimising contact with air are currently being developed.

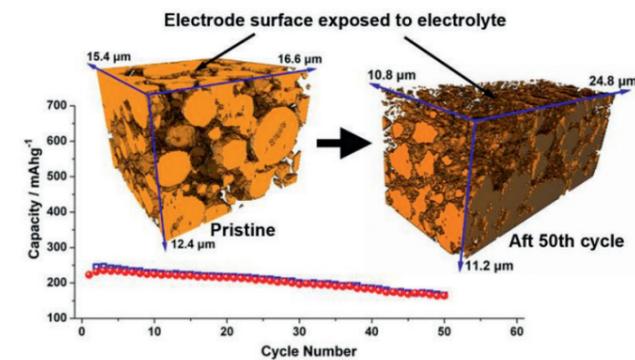


Figure 6: 3D reconstructions of electrodes by serial section FIB-SEM imaging at various cycling stages, in terms of active LLNCM particles before and after 50 charge/discharge cycles. [64]

1.2.1.6. Dual Beam Systems – Electrons and Focussed Ion Beam

Further benefits can be gained from the use of a FEGSEM with EDX/EBSD combined with an ion column. Ga⁺ ion focussed ion beam (FIB) systems with a field emission electron column are becoming increasingly common in university departments, with plasma systems, which offer higher milling speeds, becoming increasingly popular. These dual column instruments combine the capabilities of a FEGSEM with in-situ milling and polishing for near surface and cross-sectional imaging. This combined milling/imaging capability allows for preparation of site-specific cross-sections as well as site-specific preparation of lamellae for S/TEM. Furthermore 50x50x50mm regions can be reconstructed at high spatial resolution by serial section tomography [62]. The advent of plasma FIBS (Royce, Surrey, Glasgow) with their much higher milling rates have opened up volumes 1000x larger than can be viewed by serial sectioning FIB-SEM [63]. The various detectors in FEGSEMs can provide a range of image contrasts highlighting different features, and combined with EDXS and EBSD, gives a comprehensive assessment of microstructure. This diversity of information complemented by EBSD/EDXS allows information to be collected with minimum sample handling.

1.2.1.7. Operando SEM and Helium Ion Microscopes

In-situ SEM experiments are currently being designed and implemented to evaluate the processes in operating cells. Cells need to be compatible with the vacuum conditions in the chamber either at full vacuum in standard instrument or under higher pressures in variable pressure or environmental SEMs. This can be achieved by solid state/polymeric or using low vapour pressure electrolytes such as ionic liquids [65, 66], and more recently specially designed cells that are for liquid cells [67]. New advances in using the ion beam in a FIB system beyond milling [68] opens a range of possibilities in both imaging using the ion beam as the primary source generating secondary ion images and ion induced secondary electron images. A further advantage of using a primary ion beam is adding a secondary ion mass spectrometer to the system that permits the detection and mapping of lithium (as shown in Figure 7). The helium ion microscope (HIM) opens new higher resolution surface imaging with several systems existing in the UK, including one housed at Nexus (National EPSRC XPS Users' Service, Newcastle University) [69-71], one at the London Centre for Nanotechnology (UCL) and one currently being installed at the University of Cambridge. The Helium ions used for imaging can be focussed to smaller probe sizes with smaller interaction volumes giving a spatial resolution of up to 0.5 nm, improved depth of field compared to SEM and the ability to image without charging issues, while Rutherford backscattering provides compositional information.

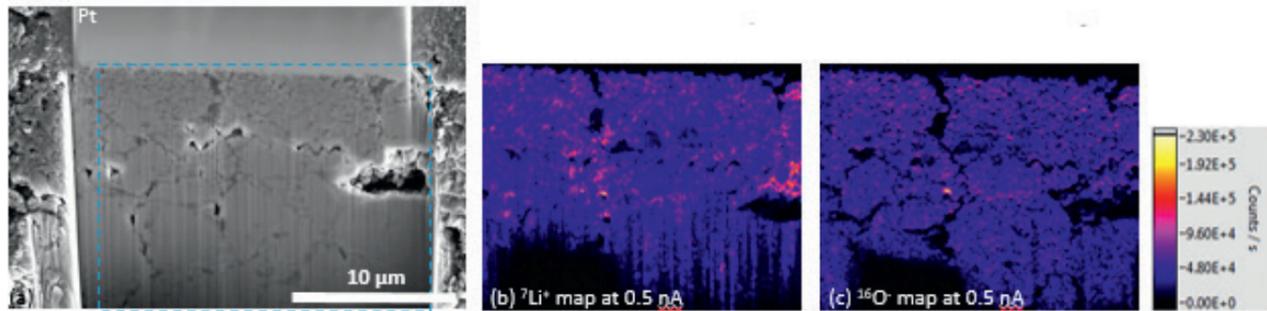


Figure 7: (a) Secondary electron image of 10° ramp into air exposed battery material and corresponding SIMS maps for (b) $^7\text{Li}^+$ and (c) $^{16}\text{O}^-$. (Image courtesy of Dr Daniela Proprentner and Professor Barbara Shollock, WMG University of Warwick)

1.2.18. Electron Probe Microanalysis (EPMA) with Soft X-ray Emission Spectrometry (SXES)

Comparable to SEM, Electron Probe Microanalysis uses a high stability electron source in combination with wavelength dispersive spectrometers to fully and accurately quantify the elemental components of a material down to 10s of ppm, providing spatially resolved elemental characterisation down in to the nm scale. As with SEMs, EPMA instruments may be equipped with a modern, high resolution FEG electron source as well as a range of ancillary equipment (secondary and backscattered electrons, cathodoluminescence, EDX). EPMA instruments routinely incorporate 4 or 5 wavelength dispersive spectrometers, each capable of delivering a spectral resolution orders of magnitude higher than that of EDX. Recent advances in the field of soft X-ray emission detection has produced the EPMA-SXES technique, providing a unique ability to investigate the electron-induced soft X-ray emission of a material at a spectral resolution of approximately 0.3 eV. This high resolution can be used, in some cases, to determine the chemical state of the elements probed. SXES has been previously used to investigate the Li Ka emission (approximately 54 eV) and has been applied to Li battery research where it can discriminate not only the presence and localisation of Li, but also the charge state of the material [72]. The first SXES spectrometer in the UK is attached to the FEG-EPMA housed within the Henry Royce Institute at the University of Manchester.

1.2.2. NMR and MRI

1.2.2.1. Operando Nuclear Magnetic Resonance

Operando nuclear magnetic resonance (NMR) provides detailed information about changes in atomic-level structure and dynamics of battery components in real time during cycling.[73] Recently, the technique has been refined through the development of bespoke *operando* NMR hardware, which facilitates interfacing of the electrochemical and NMR apparatus, and also addresses some of

the practical issues associated with solid-state NMR spectroscopy battery materials which can be challenging in its own right.[74] The changes of local environments of different atoms can now be probed by using small bag cells or plastic cell capsules [73, 74], designed to fit in standard or bespoke NMR probes, appropriate modifications eliminating radiofrequency interference in the electrochemical circuit. These kits are already being used at Cambridge and Lancaster universities to study solid-state batteries and supercapacitors. More recently, this technology has been further optimised with the development of *operando* NMR probes which simultaneously interface with both the NMR spectrometer and the electrochemical cyclers to allow full synchronisation within the experiment (Figure 8).[75] These probes also feature automatic recalibration capabilities meaning that NMR spectra can be acquired over wide frequency ranges and the probe configuration can be continuously optimised to compensate for changes in susceptibility and dielectric properties of the sample during cycling which would otherwise modulate the spectral intensity.[76] This technology allows quantitative analysis of *operando* NMR data acquired over wide frequency ranges, which is critical for fully understanding the microscopic mechanisms of battery operation (Figure 9).[77]

Some of these developments have been spearheaded at the University of Cambridge in collaboration with NMR Service GmbH in Germany who now market *operando* NMR hardware and accessories to the

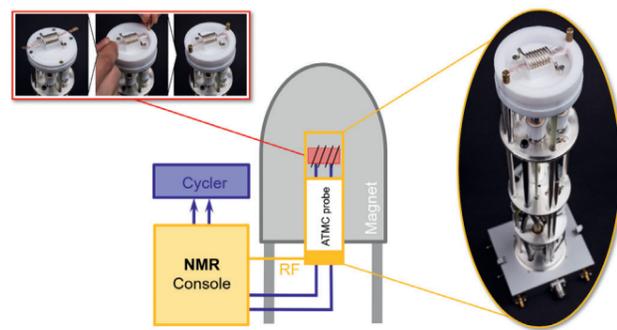


Figure 8: Setup of the automatic tuning/matching cycler (ATMC) *in situ* NMR probe and electrochemical cell inside a magnet [75].

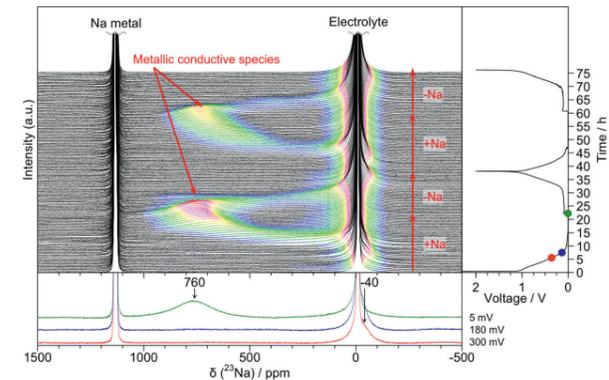


Figure 9: ^{23}Na *in situ* NMR spectra of Na insertion / deinsertion into hard carbon; metallic species forming show a significant continuous shift to higher frequency [77].

wider international research community [78]. Ongoing work includes the development of pulsed-field gradient (PFG) and imaging techniques in order to track the diffusion and migration of electrolyte species over longer length scales. The Universities of Birmingham and Cambridge currently have NMR spectrometers configured for this purpose. Several studies have already demonstrated the power of PFG and imaging for the characterisation of metal dissolution and plating,[79] lithium dendrite formation [80] and supercapacitor charging mechanisms [81]. Current work at Birmingham and Cambridge is expanding the application of these techniques to other systems such as fast ion conductors and redox flow. To fully exploit the power of *operando* NMR, further developments are required in hardware design to allow battery cells to be studied under conditions that are as realistic as possible. For PFG and imaging, this will require integration of the electrochemical circuit into the more complex gradient coil probe. For the study of redox flow batteries, work is required to incorporate fluid flow into the *in situ* cell.

1.2.2.2. Ex-situ NMR

Further technique developments are also required to properly exploit the recent emergence of dynamic nuclear polarisation (DNP) as a method for NMR signal enhancement. DNP can dramatically increase NMR signal sensitivity of surfaces and interfaces, making it ideal for studying solid electrolyte interface (SEI) formation and surface vs bulk intercalation phenomena. However, the UK currently has only one

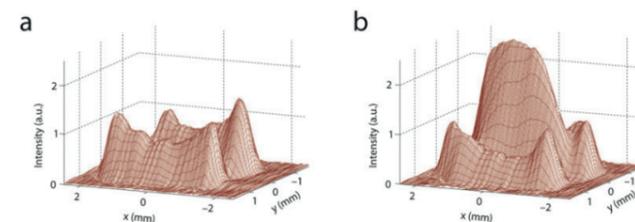


Figure 10: 2D ^7Li MRI *xy* images of a Li-metal bag cell in the pristine state (a) and after passing current (b), with frequency encoding in the *x* direction and phase encoding in the *y* direction.

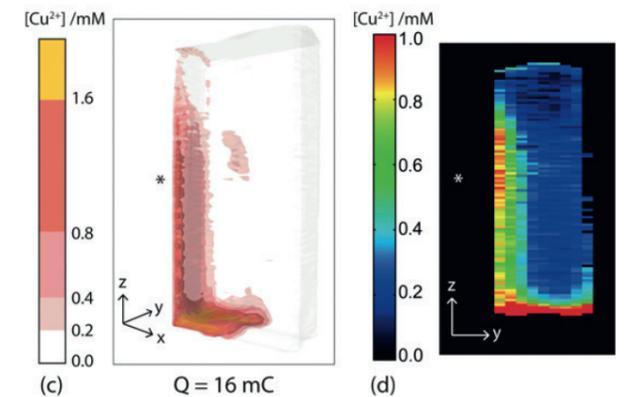


Figure 11: 3D (c) and 2D (d) concentration maps of Cu^{2+} ions following corrosion of metallic Cu, measured using ^1H MRI. (Images courtesy of Melanie M. Britton, U. Birmingham and Clare P. Grey, U. Cambridge).

facility (Nottingham) where these experiments are possible, and access charges are very high (£1000/day) due to the high running costs of the equipment. Increased provision of DNP equipment could be of significant benefit to the battery research community. Other ex-situ NMR capabilities and requirements include access to very fast MAS probes.

1.2.2.3. Magnetic Resonance Imaging of Operando Electrochemical Cells

Magnetic resonance imaging (MRI) has recently emerged as a promising non-invasive, non-destructive imaging method for visualising battery electrochemistry in *operando* [82-86]. It is able to quantitatively map the speciation, distribution and transport of species, which contain NMR active nuclei (e.g. ^1H , ^{19}F , ^7Li , ^{23}Na), in both the electrode and electrolyte, across multiple length scales, in 3D, and, in the case of ^1H and ^{19}F MRI, in real-time. As a result, MRI is able to provide an integrated, holistic view of the electrochemical cell, under working conditions, enabling data to be correlated with the electrochemical properties and behaviour of the cell. Due to the presence of a strong magnetic field and radiofrequency (RF) radiation, MRI must be performed in specialised cells [81, 83, 86, 88], which enable the cell to be aligned with respect to the magnetic and RF fields. MRI facilities are available at the University of Birmingham [89] and at the University of Cambridge [90], to perform diffusion and imaging experiments on NMR instruments operating at 300 MHz and 400 MHz ^1H resonance frequency, respectively, for a range of nuclei including ^1H , ^{19}F , ^7Li and ^{23}Na . Using these instruments, it is possible to acquire 1D profiles, at a spatial resolution of $\sim 10^{-5}$ m, and 2D or 3D images, at a spatial resolution of $\sim 10^{-4}$ m.

1.2.3. X-ray Diffraction, Small/Wide-Angle X-ray Scattering and Related Methods

X-ray diffraction allows interrogation of the crystalline structure of materials, and hence is mainly applied to electrode materials. Typical

effects observed are phase changes during charging and discharging, lattice parameter changes as the material expands or contracts with lithium insertion or extraction, and amorphisation or formation of secondary phases during cycling. Often diffraction studies are limited to phase matching, but fitting of data enables accurate lattice parameter information, crystallite size, crystallographic strain (lattice parameter variations within the sample) and preferred orientation of crystallites to also be extracted. In some cases even data collected at lab scale can be used for full structural refinements to examine atomic scale changes, although often these studies are carried out with synchrotron X-rays or with neutrons.

Ex situ studies of battery electrodes usually require air-free sample handling to preserve the phases present at a specific point of charge or discharge, but the window geometry in these studies can be optimised to the measurement (e.g. a window parallel to the beam direction to minimise scattering). Many *ex situ* electrode studies are carried out with standard powder diffractometers and hence may be possible as single investigator studies. However, use of diffractometers optimised for thin film studies, like those in use for battery studies at ISIS, Oxford and Southampton, enable further information to be extracted. For example, use of grazing incidence geometry on ink-on-foil electrodes allows the signal from the active material to be maximised in cases where often a standard θ - 2θ geometry, as typically used in powder machines, would produce a very strong signal from the metal foil. This geometry has also been used with synchrotron radiation to obtain depth profiling information on thicker electrodes, [91] and could be used to obtain significantly more detailed texture detail e.g. using pole figure measurements and for lateral mapping of phases present in an electrode.

In situ measurements are collected with transmission geometry, often with aluminised polymer bags used to contain electrode assemblies, or with flat plate geometry [92]. The latter can be performed with beryllium, aluminium or aluminised Kapton windows. Some commercial cells are available, e.g. the Rigaku Battery Cell Attachment, [93] with the advantage of integration of the electrochemical treatment into the diffractometer software. However, none of these arrangements are fully optimised. In transmission geometry it is inevitable that signals from all of the other components (windows, counter electrode, separator) are included in the data. Flat plate measurements allow focus on one electrode, but the path length through the window is longer and the available materials are all either absorbing or scattering, and the best option to minimise these effects (Be) is reactive in many battery environments.

Small angle X-ray scattering (SAXS) provides structural information on the overall size, shape and structure of particles and for bulk materials elucidates internal nanostructure across length scales from 0.5 to 500 nm. Just as XRD, the wide-angle X-ray scattering (WAXS) regime offers complementary molecular level structure information on crystalline components, with length scales accessible on the order of 0.2 to 0.5 nm. The use of X-ray sources with photon energies above several keV makes SAXS/WAXS a methods of choice for probing mesoscopic structure in situ and operando, providing valuable information in the

length scale range between molecular and macroscopic dimensions, complementing the information available from spectroscopic, diffraction and microscopy analysis. A wide variety of in situ and operando cells have been reported in the literature, including an increasing number of operando studies on batteries/energy storage systems [94].

For applications requiring single X-ray wavelength measurements, laboratory SAXS/WAXS instruments have recently closed the gap to higher brilliance synchrotron facilities through a number of technical developments; novel optics combined with scatterless-slit collimation make it possible to characterize length scales up to 500 nm. Larger variability in sample to detector distance allows optimizing the angular regime for each sample individually. Highly versatile sample stages permit high-throughput measurements under controlled conditions on the one hand, and in situ process monitoring on the other. Metal-jet X-ray sources can be operated at two different energies, i.e. at 9.2 keV (Gallium alloy) and 24.2 keV (Indium alloy), which not only allows to investigate a wide range of matter from low to high atomic number, but also facilitates the integration of devices with strongly X-ray absorbing housing materials. Such a state-of-the-art instrument for in situ/operando SAXS/WAXS has been funded as a national facility under the EPSRC Strategic Equipment process and is expected to become available for users from Q3 2019. The instrument will operate at Diamond as part of the Diamond-Leeds Collaboration at Harwell. Its capabilities have been specified to complement the synchrotron SAXS/WAXS facilities, which will thus be able to focus more effectively on highest-impact research requiring tunable high brilliance X-rays. The new instrument will be available throughout the year, including synchrotron shutdown periods, and will focus particularly on operando and in situ research, with support from a dedicated sample environment development lab at Diamond [95].

1.2.4. X-ray Photoelectron Spectroscopy (XPS) & Related Core-Level Spectroscopy Methods

By determining the binding energies of core and valence electrons XPS provides elemental analysis and, through chemical shifts of the photoemission lines, chemical and highly localised structure information without requiring long range order of the material under analysis. Often superimposed on XP spectra are X-ray Excited Auger spectra (XAES), which are likewise providing element-specific information chemical states and element concentrations. Use of soft X-ray sources (typically Al K α) results in the detection of emitted photoelectrons with low kinetic energies, which limits the escape depth of the detected signals to at most a few nm. This has established XPS as the most commonly used surface analysis technique in contemporary physical, engineering and life science research. In accord with this, there has been a plethora of XPS studies of electrode surfaces and interfacial deposition/growth in battery and wider electrochemical research. XPS is of course not limited to surface and interface analysis, with chemical shifts also useful for characterising local structure in bulk materials, as widely demonstrated for example for ionic liquids, molecular speciation studies

and other non-crystalline condensed matter. Such dedicated bulk characterisation is achieved by Hard X-ray Photoelectron Spectroscopy (HAXPES), where X-ray sources with higher photon energies are used, including synchrotron sources. Moreover, combination with ion erosion/etching permits depth profiling through layered samples, to depths into the micron range, thereby permitting combined surface and bulk analysis. A laboratory HAXPES system is being installed in the Royce Institute which will be available to all UK users.

Since mid-2017 the EPSRC National Facility for X-Ray Photoelectron Spectroscopy serves the UK user communities with UHV instruments at Harwell, at Cardiff University, at UCL and at The University of Manchester [94]. Various universities operate similar local UHV XPS instruments. XPS has become akin to NMR spectroscopy in that there is an expectation of its availability for materials and device characterisation. The resulting national landscape and possible perspectives are summarised in the EPSRC Roadmap for Photoelectron Spectroscopy [96]. However, exciting opportunities for battery research now arise from the availability of near-ambient pressure (NAP) XPS instrumentation, which overcomes the limits of ultra-high vacuum (UHV) XPS. In NAP instrument pressures up to \sim 20-40 mbar can be routinely maintained. This has suddenly broadened the scope of XPS to hitherto impossible in situ and operando applications, for example maintaining aqueous electrolytes and other volatile components that would previously have evaporated in UHV instruments. The battery and electrochemical research community has only recently begun to explore the possibilities offered by these instruments for studies of materials/devices that are not UHV compatible.

A UK NAP XPS instrument landscape has in recent years evolved around delivery of specialist characterisation, reflecting needs for a high level of local technical expertise and ancillary support infrastructure for different types of samples, materials and devices, and the different in situ and operando sample environments. UK researchers can access a NAP XPS beamline at Diamond ('VERSOX', beamline B07), while NAP XPS instruments now exist at Imperial College, Manchester, Cambridge, Nottingham and Leeds, representing an ecosystem of facilities with complementary capability. Except for the Leeds facility, these instruments combine measurements under NAP conditions with capability for surface science under UHV conditions. The Leeds facility (EnviroESCA) provides a versatile sample table (rather than a traditional sample manipulator) on which samples and devices can be placed for in situ and operando experiments under external control. The NAP analysis chamber eschews UHV operation altogether, avoiding lengthy vent/evacuate cycles, enabling rapid-turnaround NAP measurements and investigations of 'UHV-difficult' samples such as 'wet' soft matter, liquids and electrically insulating materials. For battery research, this permits maintenance of electrolyte in contact with electrodes and membranes. This could enable observations of chemical changes during charging and discharging, lithium insertion or extraction, deposition, segregation or formation of secondary phases during cycling. Using a novel transfer chamber system samples can be transferred for *ex situ* characterisation without any exposure to the ambient atmosphere, for example from cleanroom and glovebox environments. This allows new perspectives for the characterisation

of disassembling cells and examining air- and moisture-sensitive components.

The most widely used related X-ray core level spectroscopies relevant for battery research are the variants of X-ray absorption spectroscopy (XAS). XAS requires continuously tuneable X-ray photon energies, which currently restricts it almost entirely to synchrotron radiation laboratories. The near-edge absorption spectra contain rich fine-structure, usually referred to as near-edge X-ray absorption fine-structure (NEXAFS) at low photon energies ($<$ 1 keV) and as X-ray absorption near-edge structure (XANES) otherwise. The near-edge fine structure stems from variations in unoccupied electronic states below the core level ionisation potentials due to changes in chemical state, coordination geometry and local bonding. The extended X-ray absorption fine-structure (EXAFS) visible beyond the ionisation potential in XA spectra arises from backscattering of the excited photoelectron at nearest atomic neighbours, providing structural information on bond lengths, coordination numbers and the degree of local disorder in molecular and condensed matter, for example for species in electrolytes and at interfaces in batteries. Microfocus beams permit imaging XAS imaging with chemical contrast, including tomography. For battery research, XAS at photon energies above a few keV also lends itself readily to in situ and operando experiments.

A laboratory-based technique that has recently become available is X-ray emission spectroscopy (XES), with UK instruments currently in Sheffield and at Leeds (from 2019). The high-resolution fluorescence spectra obtained from these instruments are sensitive to the chemical state of transition metals, allowing for example monitoring of dopants in Li ion batteries, complementing information from XRD, NMR, XPS and XAS. Other related synchrotron techniques are resonant (RIXS) and non-resonant inelastic X-ray scattering (more commonly referred to as X-ray Raman Scattering, XRS). Harder X-ray RIXS complements XAS and XES by providing additional chemical information by probing photons emitted during the decay of the core-level as a function of photon energy. XRS is currently available to the UK community at the ESRF and permits monitoring low energy absorption spectra (e.g. from Li, C, N, O or Na) in situ/operando, with demonstrated applications monitoring Li batteries.

1.2.5. Laboratory X-ray computed tomography

3D XCT is a non-destructive analytical technique that requires little or no sample preparation. Synchrotron x-ray facilities are well suited to short timescale in situ studies (see section 1.1.1) but laboratory sources can offer a similar range of spatial resolutions without the access restrictions of large scale synchrotron research facilities. Most universities have at least one scanning systems and there are facilities at UCL (3 systems primarily focused on energy storage research), Warwick (including a recently acquired fast CT system), Southampton (4 systems) and the Royce Institute (9 systems primarily focused ion in operando/in situ imaging of manufacturing or in service degradation/testing). The instruments have varying capability including macro (metre scale objects with mm resolution), through cm systems capable

of imaging whole batteries or cells at 10mm resolution to mm samples such as electrodes at sub-micron resolution to sub-millimeter samples at 50nm resolution. Similar to synchrotron imaging systems, the uniquely non-destructive nature of X-ray CT across a broad range of resolutions has enabled the development of a range of in-situ and operando environments for studying EES devices and materials under electrochemical cycling, heating/cooling and under mechanical loads.

In particular, micron scale systems system have been used to time-lapse 3D image working batteries either during charging and discharging (Figure 12) or over battery lifetimes of 100s of discharge cycles [97]. Image data has been successfully used to provide parameterised input to higher order models, or to directly build image based models of battery or electrode performance, providing new insight into the performance/microstructure relationship [98]. A range of performance criteria including energy and power density and cell lifetime are dependent on the nature of the complex microstructure of the electrodes: X-ray CT which has enabled routine characterisation of these materials using both absorption and phase contrast [99] techniques to characterise the breadth of material encountered in EES research. The portfolio of laboratory CT instrumentation is rapidly evolving to mimic the capability of world leading synchrotron facilities leveraging improvements to spatial and temporal resolution available through new lab sources: the complementarity of large scale facilities for 'fast' studies or those requiring mono-chromatic radiation with the capability for longitudinal studies offered by lab CT is a key benefit for the EES research community.

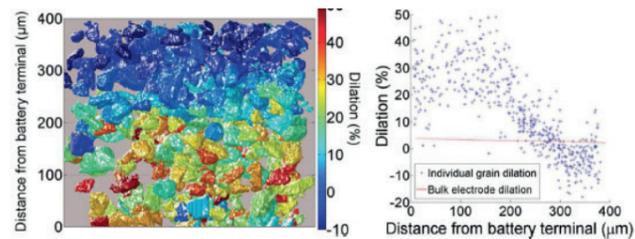


Figure 12: a) A representative central core region of the electrode material in a Li battery coin cell imaged by lab. X-ray imaging showing the dilation ratio of the manganese oxide grains during discharge. b) The dilation as a function of distance from the battery terminal for individual grains. The grains closest to the electrode-terminal interface experience large dilation whereas grains further away experience much smaller dilation. The bulk dilation varies by only 2–3% indicating that most of the grain expansion is accommodated by shrinking of pore and binder phase instead of overall grain expansion. [100]

1.2.6. Nanoscale Secondary Ion Mass Spectrometry (NanoSIMS)

The NanoSIMS 50L is a secondary ion mass spectrometry instrument capable of mapping almost all elements **and isotopes** in the periodic table, from hydrogen and deuterium to uranium, at high spatial resolution (down to 50 nm, although 100 nm is routine) [99]. This is combined with high sensitivity, ppm level for some elements, and high mass resolution, allowing unambiguous identification of ionic species of the same nominal mass. Whilst the spatial resolution is lower than TEM or APT, the field of view that can be imaged is significantly larger and

bulk samples can be investigated making sample preparation much simpler. The technique is surface sensitive, typically sampling from the top few nanometres, but due to its destructive nature it is possible to depth profile into the sample to build up a 3D image of the chemical distributions. The NanoSIMS 50L can simultaneously detect seven ionic species allowing correlation of signals. As the technique does not rely on X-ray emission from Li, and instead detects Li ions directly, the difficulties associated with detecting Li in the SEM can be overcome. Additionally by isotopically labelling samples the NanoSIMS can be used to track reactions or diffusion mechanisms. There are only 4 NanoSIMS instruments in the UK but the Manchester (Royce) NanoSIMS 50L is the only instrument within a materials science department that is equipped with the new RF plasma oxygen ion which allows imaging of electropositive ions at 50 nm, something which was not previously possible, this will allow mapping of lithium at high spatial resolution and sensitivity [102-104]. Other SIMS instruments, such as ToF-SIMS instruments that are more widespread, are also capable of imaging at high sensitivity, can detect the whole mass spectrum simultaneously and can give molecular information but spatial resolution is limited to several hundred nm.

1.3. Methods in Single Investigator Facilities

The majority of characterisation methods used to evaluate energy storage systems are the kind that can be performed in a single investigator laboratory in a university or within a corporate facility. The fundamentals of electrochemistry have been defined for many years and will not be evaluated here (except for the need for calibration and standards discussed in section 2). In this section we focus on some recent developments in laboratory methods that can provide insights into the development of new EES.

1.3.1. Advanced electrochemical methods coupled to in-situ analytical techniques

Most laboratories have electrochemical workstations capable of running many different electrochemical techniques. However, the interpretation of the results is often far from straight forward, and deep understanding of the theory behind is needed, as well as validation of the approaches using model systems, which is often overlooked. For example, GITT has been used to characterize mass transport kinetics, but analysis of a model reversible electrochemical system showed that some assumptions required for the analysis are often not met [105]. Pulsed and cyclic voltammetry are techniques used for sensing the formation of intermediate or side reaction products (eg. polysulfide species in Li-S cells), but validation studies showed that the choice of the sensing electrode material is critical, and several materials previously employed proved unreliable [106]. Impedance spectroscopy is a very powerful technique to characterize key properties of

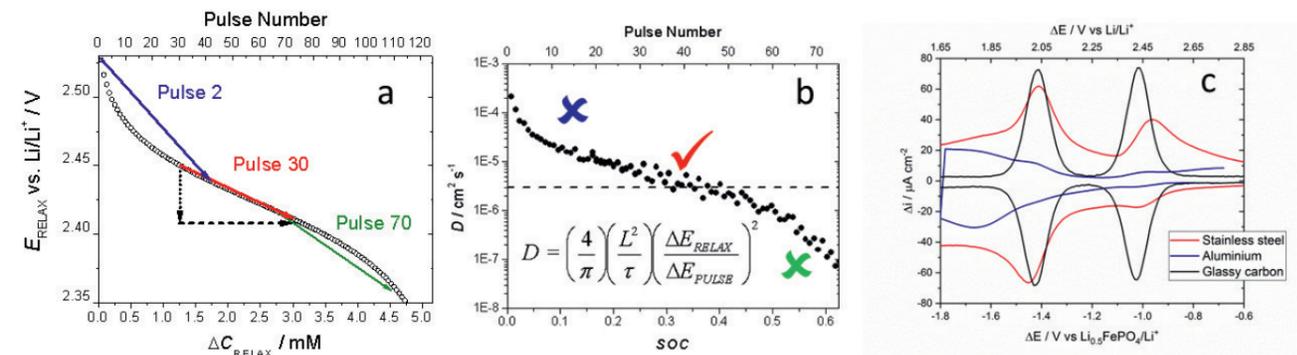


Figure 13: a,b) Validation of the use of Galvanostatic Intermittent Titration Technique (GITT) for the analysis of the mass transport rate of a model system (ethyl viologen), showing that reliable determination of the diffusion coefficient is only obtained for pulse numbers where the variation of potential and concentration are proportional. c) Validation of electrochemical sensors for quantifying the solution redox species (here, ethyl viologen), showing the unsuitability of common materials (stainless steel, aluminium) and the advantage of incorporating a glassy carbon electrode into a Swagelok cell (Images courtesy of Nuria Garcia-Araez, University of Southampton).

energy storage system, but the interpretation of the results is often questionable. In conclusion, validation of results obtained with conventional as well as advanced electrochemical methods is of utmost importance. In addition, electrochemical methods are often not enough to elucidate the complex mechanism of battery reactions (and side reactions), thus (in-situ) analytical techniques are also necessary. For example, in-situ mass spectrometry is a powerful technique to detect even small amounts of gases evolved from batteries [107, 108]. In-situ spectroscopic methods are powerful tools for chemical and phase identification, as described in the following sections.

1.3.2. Vibrational Spectroscopy and Spectro-electrochemistry

Vibrational spectroscopy typically encompasses such instruments as Raman microscopy and infrared spectroscopy. These instrumental techniques are used in the analysis of battery materials for chemical and phase identification; interface analysis; and microstructure distributions. The cost of the instrumentation (~£30k-300k), with modest annual upkeep costs. Every UK Chemistry/Materials department has access to instrumentation, however careful measurement of samples requires no air exposure, thereby sample preparation in a glove box and use of a sealed ex situ measurement cell is required. These techniques can be used to obtain composite maps of electrodes and to probe surface film formation and to follow the progress of lithium insertion/extraction out of host material.

Here, we focus on the specialist areas of vibrational spectroscopy that are combined with an electrochemical cell and can provide significant insights into the development of battery materials and systems. Spectroelectrochemical approaches that investigate the chemistry of electrified interfaces rely mostly on probing of vibrational excitations. Combining linear vibrational spectroscopy (Raman scattering, infrared absorption and ultraviolet–visible) or electron spin resonance with electrochemical methods, is a powerful approach for studying interfacial processes as they can yield valuable information relating to molecular specificity and reactivity of species at the interface. Strong examples

in the literature summarised in recent review [109] demonstrating their scope within this area include studies dedicated to understanding the chemical identity, structural dynamics, interactions between molecules and their environment, and even orientation of the species present at the electrode surface or the interfacial region. The simplicity and convenience of these techniques makes them appropriate for insightful studies in the area of novel energy storage systems.

1.3.2.1. Operando Raman Microscopy

The Raman effect corresponds to the excitation of a molecule, to a real or virtual, excited electronic state and a subsequent emission to a different vibrational state in the electronic ground state. This results in scattered lines either side of the incident frequency scattered light (Rayleigh scattering). These lines are known as Stokes ($\nu = 0-1$) and anti-Stokes ($\nu = 1-0$) lines. Raman spectroscopy is based on scattering processes that are intrinsically weak when compared to optical processes like absorption and fluorescence; therefore, it requires an amplification of the signal. Approximately 1 in 10^7 photons are inelastically scattered, the enhancement of Stokes and anti-Stokes scattering processes is required to provide sufficient signal from these excitations, and the vibrations they correspond to, in order to gain adequate sensitivity in order to observe adsorbed species at electrode interfaces and this is achieved by a multitude of surface enhanced Raman spectroscopy (SERS) techniques.

Of recent particular interest in SERS methodology is shell isolated nanoparticles for enhanced Raman spectroscopy (SHINERS) which allows the exploitation of SERS on electrodes other than gold or silver, such as carbon electrodes (Figure 14). [110] These are nanoparticles coated in a thin shell of silica or alumina. The majority of studies use 55 nm Au nanoparticles, because the preparation is relatively straightforward. It has also been shown that with 633 nm laser excitation wavelength, a common laser in Raman systems, this provides the greatest Raman signal. [111] As well as spherical nanoparticles, nanocube and nanorod SHINS particles have been reported that have tuneable surface plasmon properties, which means the excitation

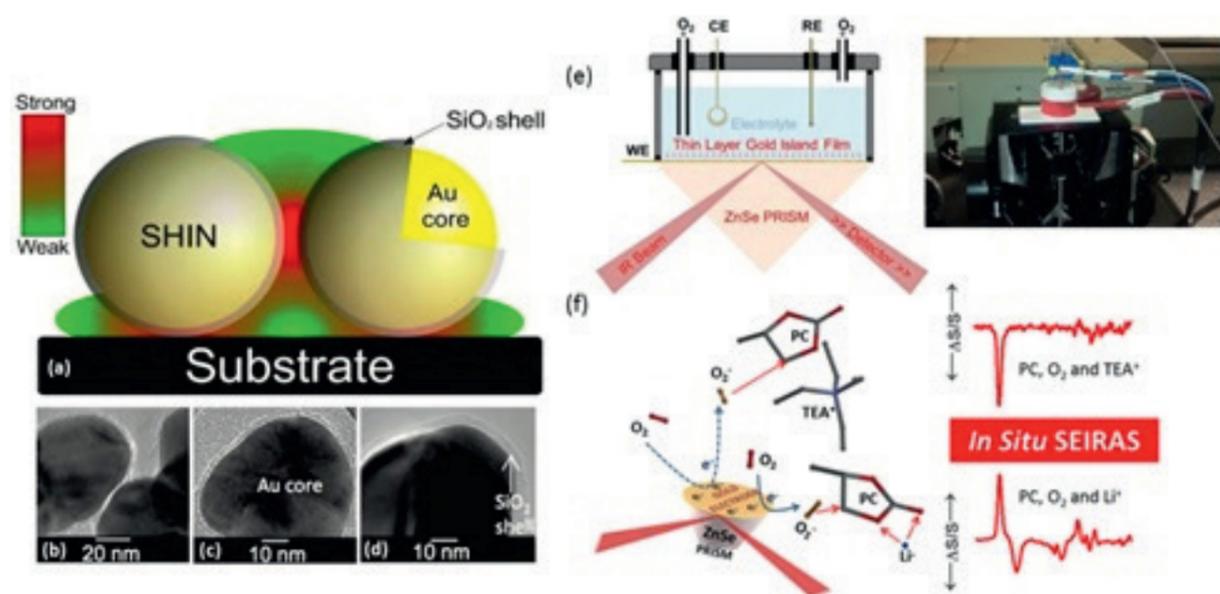


Figure 14: Examples of advanced *in situ* spectroscopic methods for EES investigations (a-d) SHINERS, (e,f) SEIRAS schematic and cell and example SEIRAS spectra on propylene carbonate Li-O₂ system.

wavelength can be modified to near infrared wavelengths. [112] Groups in Liverpool [113], Nottingham [114], Oxford [115] and Southampton [116] are centres that actively publishing SERS studies relating to energy systems.

1.3.2.2. Operando infrared spectroscopy

Infrared spectroscopy is a vibrational spectroscopy technique that relies on the absorbance of the infrared light by a molecule when a beam of infrared light falls on it. The interaction of infrared radiation is associated with molecular vibrations; only those molecules whose electric dipole moment changes during vibration can absorb infrared light. The frequency of the absorbed infrared radiation coincides with the vibrational energy levels of the molecule. The exact frequency at which a given vibration occurs is determined by the strengths of the bonds as well as the mass of the atoms involved. Knowing these absorption frequencies and their intensities would help to get “finger print information” about the structure and composition of the molecule. Fourier transform infrared spectrometers (FTIR) convey throughput advantage since the grating is dispensed with and also with rapid scanning interferometers (or “step-scan” methods) the speed of spectral acquisition can be used for time resolved studies. As such modern FTIR spectrometers have improved the acquisition of infrared spectra so dramatically that FTIR spectrometers have, to a very large extent, replaced dispersive instruments which were first developed in the 1940s. Transmission mode IR spectroscopy has been popular for conventional infrared analyses, whereas infrared characterisation of electrode-electrolyte interfaces normally adopts reflectance methods. Over the past few decades, various designs utilising internal, specular (external), or diffuse reflection of infrared radiation at the

metal-solution interface have been developed. A few examples are infrared reflection-absorption spectroscopy (IRRAS) employing specular reflectance, attenuated total internal reflection infrared spectroscopy (ATR-IR) utilising total internal reflectance, and diffuse reflectance infrared spectroscopy (DRIFTS). Note that there are three different acronyms used for infrared reflection absorption spectroscopy in the literature, viz. IRRAS, IRAS and RAIRS, but IRRAS is more prevalent. The absorption of infrared radiation by the medium through which an infrared beam passes before and after the reflection can hamper the sensitivity to the interfacial region. External reflection spectro-electrochemistry methods (e.g. IRRAS) have to address the problem of attenuation of IR signal by an overwhelming absorption by the electrolyte, while internal reflectance (ATR) techniques have to consider absorption of the radiation by the internal reflection element that limits/determines the spectral range. The polarisation modulation infrared reflection-absorption spectroscopy (PM-IRRAS) has proved successful for studying surface reactions on well-polished and reflective surfaces, polarization modulated techniques also benefit from the surface selection rules.

The simplicity of ATR techniques and their applicability on non-reflective surfaces make them more appealing. There are two possible optical geometries in the ATR mode, the Otto [117] configuration and Kretschmann [118]; surface sensitivity of ATR techniques is also dependent on the configuration. In the Otto-ATR configuration, the electrode is arranged on top of the internal reflection element (IRE) with the electrolyte filled between a small gap (typically a few micrometres) between the IRE and the electrode. In the Kretschmann configuration, a thin metallic film (< 100 nm) of the electrode is formed directly onto the surface of an IRE, then the electrolyte solution is filled on top of the electrode. The effective path length of the IR radiation is related

to the depth of penetration of the evanescent field produced at the interface which is typically < 10 nm. As a consequence the background IR absorption is significantly reduced compared to external reflection techniques. The evanescent wave decays as a function of the distance from the surface, which makes *in situ* probing of the electrode surface extremely challenging in the Otto-configuration because the electrode has to sit extremely close to the IRE in order to probe the electrode surface. Reducing the gap between the electrode and the ATR element often leads to too thin an electrolyte layer that often introduces additional issues due to electrolyte resistance and mass transport issues. Therefore, the Kretschmann configuration is generally preferred for *in situ* studies; the Otto-configuration is widely adopted for *ex situ* analyses where the electrode is analysed after it is removed from the electrochemical cell. Even in the Kretschmann configuration, because the IR beam penetrates into the bulk for several micrometres, some sort of signal amplification on the surface would be necessary to achieve surface sensitivity.

In 1980 Hartstein *et al.* [119] found that very thin discontinuous films (~10 nm thick) of gold or silver coated on an ATR prism with molecular layers results in signal enhancement. This approach has been named “surface enhanced infrared absorption spectroscopy” (SEIRAS). The SEIRA phenomenon can be observed with continuous metal films as well, where a thin layer of metal film was formed on the ATR prism, with a molecular layer then being cast on this. Using a metal film deposited on a silicon ATR prism, Osawa *et al.* [120] demonstrated the scope of ATR-SEIRAS for *in situ* electrochemical studies. [120,121] They developed a simple electroless deposition method to form metal island structure on Si IREs. It is generally accepted that the surface enhancement results primarily from the increased local electric field due to the oscillating plasmonic electrons at the grain boundaries of the nanostructured film. The realization of SEIRA, followed by the pioneering work of Osawa [120,121] exploring both theoretical and experimental aspects of this technique, gave important new scope to *in situ* IR studies of electrochemical interfaces. A key advantage offered by the amplification of the signal (enhancement factors up to 1000) at the interface is that it facilitates monitoring of the surface reactions and structural dynamics at the interface, with considerably higher time resolution than previously achieved and work on metal-oxygen batteries has been reported has taken advantage of this enhancement effect (Figure 14). [122] Aberdeen [123], Birmingham [124] and Liverpool [113] all actively publishing IR studies in energy systems.

1.3.2.3. In-situ UV-vis

UV/Vis spectroscopy is the study of the excitation from the ground state to the electronically excited state. The strongest transitions observed below 200 nm corresponding to electronic transitions associated with bands such as C-C and C-H bonds. Saturated compounds, such as carbonyl groups, exhibit n-π* transitions in the region of 150 nm to 250 nm. Most compounds and solvents therefore contain signals from the heavy presence of these bonds in non-aqueous solvents. There are numerous example setups for *in situ* UV-vis measurements in the literature, these include both transmission and reflection

modes.^[11] In using transmission cells the electrodes are required to have a transparency of >45% and metal meshes of high quality can be obtained that are easily purposed as electrodes for these measurements [125]. Sputtering is another good method of obtaining a suitable electrode surface [126]. An additional consideration for these measurements is the path length through the cell, or total amount of electrolyte. The Centre at the University of Liverpool is actively publishing research in this area [111].

1.3.2.3. In situ EPR

Electron spin resonance (ESR) or electron paramagnetic resonance (EPR) spectroscopy, which can be considered as broadly comparable in principle to nuclear magnetic resonance (NMR), excites unpaired electron spin instead of the spin of magnetic nuclei. Although not as widely used in chemistry as NMR it nevertheless holds an important place in the detection and study of species containing unpaired electron spins, for instance in organic free radicals, paramagnetic inorganic metal complexes or in solid state materials and it can also detect free electrons in metals. It is becoming an increasingly present technique across battery research including lithium ion, sodium ion [127] and metal-O₂ batteries [128]. In the latter case its ability to detect unpaired electrons, enables analysis of radical species produced during battery charge or discharge such as superoxide and reduced O₂ species (ROS) and other side products arising from unwanted ancillary chemical or electrochemical processes. Researchers at the University of Manchester are actively publishing research in this area [129, 130].

1.3.3. Scanning Electrochemical Probe Microscopy

Electrochemical probe microscopes make local electrochemical measurements through the use of a small scale electrode or electrochemical cell device that is scanned in close proximity to a sample surface. This family of techniques comprises scanning electrochemical microscopy (SECM), scanning electrochemical cell microscopy (SECCM), scanning ion conductance microscopy (SICM), and hybrids of these techniques, with each other and integrated with additional instrumental methods, e.g. SECM-AFM, SECM-SICM and the use of these methods on confocal fluorescence microscopy platforms. The software for these techniques is available to academic groups via the Warwick Electrochemical Scanned Probe Microscopy (WEC-SPM) platform (<https://warwick.ac.uk/fac/sci/chemistry/research/unwin/electrochemistry/wec-spm/>), and a number of groups internationally are establishing these methods, including for battery research. Recent advances in SEPM means that nanoscale electrochemical probes are more readily employed and there have been efforts to speed up imaging, for example to make voltammetric movies of activity [131]. A further trend has been towards correlative electrochemical multi-microscopy, where data from electrochemical images and movies are correlated with images from other microscopy techniques of the same area (e.g. EBSD, electron microscopy and AFM). These

approaches have revealed considerable new perspectives on active sites in electrocatalysts [132, 133], but have been somewhat underused in battery electrode research. A major challenge is to deploy these methods with stringent environmental control. A recent report demonstrated electrochemical imaging measurements of redox activity of Li_2O_2 using an organic gel probe [134] and there are examples of the use of SECM in a glovebox to study lithium ion batteries (from German groups), but with very large electrode probes. [135]

1.3.4. Electrochemical Impedance Spectroscopy

Electrochemical Impedance Spectroscopy (EIS) is a perturbative characterisation of the dynamics of an electrochemical process, essentially a tool for unravelling complex non-linear processes. EIS exploits Faraday's law to characterise a chemical process in terms of electrical measurements and modelling the electrochemistry in terms of an assembly of electrical components (capacitors, inductors resistors, etc., the "equivalent circuit") representing the various Faradaic and non-Faradaic processes taking place in the system under study. The electrochemical impedance is the response of the electrochemical system to the applied potential, and the frequency dependence of this impedance can be employed to reveal underlying chemical processes based on the equivalent circuit. Electrochemical Impedance is normally measured by imposing a small amplitude sine wave on top of the controlled DC polarisation potential, such that the response of the cell is pseudo-linear. In a linear (or pseudo-linear) system, the current response to a sinusoidal potential will be a sinusoid at the same frequency but shifted in phase. This current signal is then analysed as a sum of sinusoidal functions (a Fourier series). The impedance or response of the electrochemical system (cell) is highly non-linear, comprising real and imaginary parts and this complex response is usually represented in a Nyquist plot. Electrochemical Impedance Spectroscopy (EIS) measurements are more often performed under potentiostatic control than under galvanostatic control. In most cases, potentiostatic and galvanostatic modes are equivalent (PEIS or GEIS techniques) and result in the same Nyquist plots provided that the sine current amplitude is "equivalent" to the voltage sine amplitude. However, in certain conditions, typically when the system evolves during the measurement, results from the two techniques may be different: for example, in battery applications, it is often of interest to determine the variation of internal resistance during discharge/charge, and in this case it may be appropriate to use GEIS.

The team from the School of Engineering in Newcastle University (NU) has a proven tracked record in the characterization of electrochemical energy storage systems based on EIS [136-138]. Through collaboration with electric vehicle OEMs such as Nissan, the NU team has fully developed and industrialized a unique non-invasive testing technique for quality improvement and detecting ageing mechanisms in Li-ion batteries, and this was recently installed on the production line of the Nissan battery manufacturing plant in Sunderland for fault detection and quality control. The EIS battery work at NU involves multidisciplinary and cross-cutting research covering cell, module and pack level. The techniques employed include: state-

of-health estimation, capacity check and fault diagnosis, as well as the development of novel battery equalisation circuitry and battery management systems (BMS). NU's experimental capability ranges from cell level analysis through to the interrogation of automotive battery packs with 4 EIS capable potentiostats having a total of 24 channels operating from 5 to 50V and 5 – 100A, and a 500V, 500A CAN capable vehicle pack testing station. Environmental control is provided by two chambers, one of which is aviation grade, capable of maintaining temperatures from -70°C to $+170^\circ\text{C}$ and 5 – 95% RH. In addition, the energy storage test bed (which includes a 32 kW battery emulator) and Smart Grids Laboratory in NU offer a unique utility-connected interface and ReLiB team at NU have a wide range of cross-cutting experience to support battery research at all scales.

2. CALIBRATED OBSERVATIONS AND DATA DRIVEN EXPERIMENTAL DESIGN

Experimental observations have the potential to provide significant insights into the performance, durability and recycling of EES systems. However, if the characterisation methods contain intrinsic errors in them (caused for example by poorly calibrated equipment or impurities and/or contaminants) then it becomes harder to elucidate trends in the data and to understand how any particular result fits into the overall understanding of EES materials and processes (given the breadth of experiments described in the previous section, no one can be an expert in all the methods being used). To overcome this problem, the Faraday Institution has the opportunity to define the parameters that can be encompassed into data that can be "certified" as providing both the calibration and meta-data to be reproduced by any researcher in the field. In addition to improving the human understanding of these datasets, the "certified" datasets have the potential to increase the efficiency of machine learning methods for both data mining and for designing new experiments. In this section of the report, we discuss the requirements for certified data and how it can be used within the machine learning framework to extract unique information for battery innovations.

2.1. Metrology and Standards

Innovation, built on firm science and engineering foundations, is central to economic growth and societal impact. While the UK is a world leader in scientific research, it lags behind in commercial innovation and in transforming excellent science into new products and services. The Industrial Strategy aims to address this imbalance in a number of strategic areas, including the rapidly growing field of electrochemical energy storage via the Faraday Battery Challenge. Measurement and standards have a critical role to play in ensuring that the appropriate framework for transforming battery science into commercial products and services is in place and in delivering the productivity gains that are essential to the UK's economic performance in this area. This will support the creation, adoption and commercialisation of new technologies, helping UK companies to gain a global advantage in areas of strategic importance.

Measurement is the currency of technical trade, underpinning an estimated £622bn worth of UK commerce that relies on weights and measurement regulations, while over 60% of British Standards refer to measurement or testing. The benefits of measurement interventions have been proven through economic analysis and individual case studies. Econometric studies show that measurement support can boost employment by 10%-15% within two to four years and

organisations who actively seek metrological intervention are up to 72% more likely to generate revenue from products that are new-to-market [139].

There are a number of key themes that need to be addressed at a national level to ensure the UK can take a competitive lead in battery technology worldwide. The first of these is confidence in the performance of UK technology. The establishment of a robust measurement and standards infrastructure will help de-risk investment and accelerate the adoption of new technologies through confidence in performance and safety, in turn delivering strong supply chains. The second is underpinning the quality of UK research. Dissemination of best practice in measurement and testing to Faraday Institution research projects will ensure robustness and inter-comparability of the generated data, accelerating development cycles.

Standards are essential for trade. Setting international standards and associated measurement techniques supports emerging technologies, accelerating innovation and commercialisation. Standards play an important role in the adoption and use of battery technologies. As new battery formats and chemistries emerge, there will be a need to adapt existing standards and create new ones. Significant areas that will need to be considered include the use of new materials, design for end of life, new test methods, charging infrastructure, building regulations, and safety, among others. Setting global standardisation priorities will minimise any market barriers, strengthen industrial collaboration, promote the rapid adoption of UK-led standards and measurement techniques and create a global market platform for UK-based innovators, especially important as we move into a post Brexit environment.

2.1.1. Existing Infrastructure

The National Measurement System (NMS) [140] provides the UK with an infrastructure of laboratories that deliver world-class measurement science and technology and provide traceable and increasingly accurate standards of measurement. Maintenance of the National Measurement System is critical because of the substantial impact it has on every aspect of UK life and its economic success. The NMS enables the UK to compete in global trade and manufacturing by ensuring consistency and recognition of measurement units and standards throughout the world. Internationally-leading knowledge and expertise is passed on to UK stakeholders by a coordinated programme of knowledge transfer.

The NMS is delivered through the Department for Business, Energy and Industrial Strategy (BEIS). The scientific research programmes are

delivered by the UK's measurement institutes:

- National Physical Laboratory (NPL) [141]
- LGC Ltd [142]
- TUV-NEL [143]
- Office for Product Safety and Standards (part of BEIS) [144]
- National Gear Metrology Laboratory (NGML) [145]
- National Institute for Biological Standards and Control (NIBSC) [146]

BEIS invests £65 million a year in the NMS, broken down into four main sector themes:

- Advanced Manufacturing
- Digital
- Energy and Environment
- Life Sciences and Health

BEIS engages sector experts to provide independent advice on the science programmes carried out by the laboratories, which are reviewed regularly. Support for battery technologies has been identified as a key priority in the UK Measurement Strategy [147] and NPL has recently published the results of a detailed consultation with industry, government and academia to prioritise measurement issues that are hampering the establishment of UK manufacturing capability in this area [148]. The outputs of this report have directly fed into the formulation of the NMS battery metrology research programme, which is led by NPL and focused on the development of standard test methods, in situ diagnostic techniques and modelling tools for high energy density batteries. This work falls under the remit of the Energy and Environment and Advanced Manufacturing sector themes and is guided by an Industrial Advisory Group representing all parts of the battery supply chain.

The British Standards Institute (BSI) represents the interests of UK industry and consumers in Europe through its membership of the European Committee for Standardisation (CEN) [149] and the European Committee for Electrotechnical Standardisation (CENELEC) [150] and internationally via the International Standards Organisation (ISO) [151] and the International Electrotechnical Commission (IEC) [152]. BSI manages an expanding portfolio of European and international standards related to batteries, electric vehicles and charging infrastructure. BSI currently has 15 active technical committees in this area, of which three are responsible for the publication of key standards in the field of batteries and automotive electrification. NPL plays a leading role in representing the UK in the technical development and revision of standards in these areas. Similarly, QinetiQ represents UK bodies on Committees at BSI, IEC and SAE and has capability to develop, validate and demonstrate new and novel test methods for these bodies.

Existing standards programmes have been created to support current battery technologies, formats and materials. As new battery technologies are developed, there will be a need to adapt these standards or create new ones. Areas that could give rise to the need for

further standardisation include: battery technology, especially outside lithium-ion; battery testing, labelling and configuration; electric vehicle charging infrastructure; vehicle-to-battery communications, battery re-use, recycle and remanufacture; and diagnostics and testing.

Standards development in some of these areas could make a significant contribution to UK advancements in the field of battery technologies and help to promote confidence in related applications, such as electric vehicles. Further detailed analysis is required to identify where standardisation could most positively impact scale-up in battery and electric vehicle development and strengthen UK aspirations in the field.

There is an inherent relationship between the evolution of the battery supply chain and the development of standards. Standards play an essential role in providing supply chain assurance to buyers, end-customers and consumers that technologies are safe, compatible and meet minimum quality and performance requirements.

Countries such as China, Japan, South Korea and the United States have been developing standards for longer periods than the UK and this seems to correlate with a head start in battery and electric vehicle manufacturing. Early creation of standards tends to reflect a country's degree of industrial maturity in relation to the development of the technology [153]. This also indicates that there are gaps and opportunities in the standards landscape that some nations or independent standards development organisations have started addressing. Creating standards as part of the technology developments would therefore give the UK an opportunity to put these forward as future international standards, ultimately helping to drive UK competitiveness in this area of strategic value [154].

2.1.2. Recommendations for Standards Infrastructure

The following steps are required to establish a robust measurement and standards infrastructure that would underpin the quality of UK battery research and technological innovation:

- Develop **standard test protocols** for commonly used battery characterisation techniques, focusing on quality assurance, calibration, repeatability and calculation of uncertainty budget. This should be done in consultation with a wide range of key stakeholders from academia and industry to ensure the broadest possible uptake of the developed protocols. The outcome will be to establish a common measurement framework for UK research, enabling inter-comparability of data from different organisations and providing confidence in data from government-funded research.
- Disseminate **best practice in measurement and testing** from NPL (and industrial partners such as QinetiQ) to academia and industry through bespoke training courses, publication of best practice guides, secondments and focused workshops. A key component would be the development of a standard data management plan for Faraday Institution research projects, covering issues such as data

provenance, calibration, metadata, security and accessibility.

- Establish a **collaborative research framework** between academia, industry and national laboratories to develop advanced in situ diagnostic techniques, modelling tools and standard test methods. The impact will be to support advances in battery chemistry and engineering and to facilitate reliable and independent test and validation that will enable the market/investors to have confidence in the products.
- Provide **discounted access** to state-of-the-art measurement and testing expertise and facilities at UK national laboratories for academia and industry. This would be of particular value to SMEs, who typically do not have the resources to access such facilities under normal circumstances. A funding model similar to the Analysis for Innovators Programme (run by the NMS with Innovate UK) could be adopted here.
- Conduct strategic analysis to identify where **standardisation** could strengthen UK aspirations in the field of battery technology development. This work would help identify and validate priority areas to support UK capability and accelerate the scale-up of battery technology and the deployment of electric vehicles and charging infrastructure. The exercise should be aligned with the development of any technology roadmaps maintained by the Faraday Battery Challenge and other relevant industry bodies. An assessment of the current formal standards portfolio, including metrics of use, should be conducted as part of this analysis.
- Develop an **industry engagement programme** to support delivery of a high-impact standards programme for the UK, backed by a dedicated community responsible for:
 - o Conducting early-stage technical scoping
 - o Pulling in and directing existing knowledge on battery-related topics from Faraday Battery Challenge projects, the National Battery Manufacturing and Development Facility, and other relevant industry-led initiatives
 - o Creating agile standards projects and/or fast-track standards
- Establish priority areas for **international cooperation** with national standards bodies or independent standards development organisations. This should also consider feeding into and strengthening BSI mirror committees to CEN, CENELEC, ISO and IEC.

2.2. Data Analytics and Machine Learning

Increasing computational power, storage capacity and novel innovative measurement technologies all have the potential to increase exponentially the amount of available data in the EES community [155]. Analysis of big data is a challenging task on its own and it becomes even more challenging when several measurement techniques and

instruments diversify the data, producing different data formats, with different information quality and uncertainty [156]. The EES community can address this challenging task by a more close collaboration with centres of expertise in Data Science, Uncertainty Quantification, Information Modelling and Machine Learning (these centres exist in universities, national facilities and industrial laboratories, such as QinetiQ). This collaboration can provide enormous benefits to Electrochemical Energy Storage applications by ensuring that resources are optimally exploited and that new materials, new methods and new systems are quickly and reliably developed by avoiding unnecessary repetition of efforts, costly experiments, and by quantifying the associated uncertainties (the standards for classification of data described in the previous section provide an ideal starting point for the use of large data methods).

This trend towards larger, diversified and more complex data sets brings with it several challenges. For instance, it would be virtually impossible to process this information manually nor with standard data analysis techniques [157-161]. **There is a growing need for more sophisticated data analysis methods, capable of automatically extract relevant features from the data, discovering new meanings and additional information** [162, 163]. Machine Learning (ML) algorithms offer an opportunity to efficiently and effectively analyse big and diversified data, better boosting knowledge discovery by learning new dependencies and patterns in an automated way [164-169]. Machine learning and uncertainty quantification [170-173] technology can help to better understand the chemical identity of materials, structural dynamics and even interactions between molecules and their environment when suitable data is provided. For instance, convolutional neural networks [174-178] are powerful ML tools, useful for finding patterns in images to recognize objects, defects and scenes. They learn directly from raw image data, using patterns to classify images and eliminating the need for manual non-automated features extraction. Combining these approaches with world-leading expertise in material characterisation can provide enhanced data quality and increase the subsequent business opportunities that use this unique data for battery innovation.

One of the major limiting factors in the use of high performance computing (HPC) for data analytics and machine learning so far is that there are three different communities that need to be involved - Experimentalists, Theoreticians, and Data Analysts – but as yet there is no common communication pathway between them. Historically overlaps have been along the lines of “please verify my results” rather than using each group's complimentary abilities to drive science by filling in knowledge gaps. Some have suggested that we need to improve communication by having working meetings or online forums where data and problems can be posted for others to look at. An alternate, and more permanent solution is to train new materials scientists to be experts in one field and near expert in another (This also partially solves the issue that most machine learning experts take data analytics jobs with very high pay and are nearly impossible to recruit to fundamental science laboratories). To this end, it should be possible for the Faraday Institution to incentivise PhD and Postdoc positions

that involve a “double-major”. These hybrid-scientists will then be highly skilled in multiple domains and provide the UK with cross-cutting skills for the design and implementation of innovative battery systems.

2.2.1. Existing Infrastructure

Machine learning and deep learning methods are known to be very CPU intensive programs. Nonetheless, there are numerous software frameworks [179-187] and physical infrastructures which provide a valuable support for computationally demanding calculations. For instance, High-Performance-Computing (HPC) centres are available at the Hartree centre and the University of Liverpool (with the new Barkla machine). This infrastructure of super-computers provides all the means to satisfy Machine Learning requirements for computational power, often very high when big data sets have to be analysed and challenging industrial problems have to be solved. A more detailed description of HPC facilities, which are largely available and accessible includes:

2.2.1.1. Hartree Centre

Increasing performance and productivity while minimising energy consumption are challenges faced by all businesses, but especially so when exploiting high performance computing technologies. Hartree Centre aims to deliver significant competitive advantages by tackling these challenges head-on. The UK’s first POWER Acceleration and Design Centre (PADC) aims to improve modelling, simulation and big data analytical capabilities on IBM’s OpenPOWER systems. The OpenPOWER Foundation is an open technical community based on the POWER architecture, enabling collaborative development and opportunity for member differentiation and industry growth. The goal of the Foundation is to create an open ecosystem, using the POWER architecture to share expertise, investment, and server-class intellectual property to serve the evolving needs of customers and industry. Founded in December 2013, the organisation now has more than 150 members worldwide. At the Hartree Centre, there are a wide range of high performance computing platforms such as:

Intel platforms

- Bull Sequana X1000
- Lenovo NeX tScale | 8,192 cores
- Lenovo System x iDataPlex system | 2048 cores
- Intel Xeon Phi | Knight’s Corner
- IBM big data analytics cluster | 288TB

IBM data centric platforms

- IBM Power8 + NVLink + Tesla P100
- IBM Power8 + Nvidia K80

Accelerated platforms and emerging technologies

- Maxeler field-programmable gate array (FPGA) system
- ARM 64-bit platform
- Clustervision novel cooling demonstrator

2.2.1.2. HPC at Liverpool (Barkla with access to GPU for machine learning)

HPC systems are also available at Universities in the UK. As an example, the University of Liverpool has two parallel Linux clusters on campus (Table 1).

| Barkla (newer) | Chadwick (older) |
|--|---|
| 105 nodes 40 cores 384GB of memory | 118 nodes 16 cores 64GB of memory |
| 2 nodes 1.1TB of memory | 1 memory node 128 cores 2TB of memory |
| 46 nodes are for general use | 79 Westmere nodes with 12 cores each |
| 59 nodes for dedicated access | 18 Nehalem nodes with eight cores each |
| All nodes connected by 100 gbit/s OmniPath network | All nodes connected by 40 gbit/s Infiniband |
| 200TB networked storage for ordinary computations | 45TB networked storage for ordinary computations |
| 360TB parallel file storage for computations with large amounts of I/O | 90TB parallel file storage for computations with large amounts of I/O |

Table 1: Computing clusters available at the University of Liverpool

2.2.2. Path Forward

For all of the approaches to machine learning and the application to battery science, it is essential that the three groups of scientists are all involved in the projects being developed (rather than just being the token collaborator to verify results). Integrating these data analytic methods into the design of the initial experiments and the interpretation of the results has the potential to rapidly accelerate the use of characterisation tools to provide unique data that can lead to innovations in battery science. Involving collaborating scientists from academia, national facilities and from industry will further accelerate battery science. In particular:

Information modelling and uncertainty quantification: Dealing with uncertainty is of uttermost importance in solving scientific problems with the right degree of confidence [170-173]. Uncertainty affects computational models for the system simulation [188-189] and affects measurements [190-191]. Uncertainty in measurement can be due to inherent variability in exogenous or endogenous system factors (e.g. variable material properties and non-perfectly-controllable environmental factors such as temperature, humidity, etc.) but it can affect measurements due to poorly defined factors (e.g. poorly estimated parameters, subjective judgements, coarse image resolution, censored data, etc.). Uncertainty must be properly characterised and quantified and ML methods combined with uncertainty quantification techniques offer a convenient way of solving this [1171, 192, 193].

Raw data are often more reliable than processed data: The procedure of editing, cleaning or modifying the raw data is a common practice in many applications and results in processed data. This procedure is

often performed manually by, e.g. filtering the noise, interpolating and remove outliers. However, this procedure often brings with it artificial assumptions on the data, hard to justify, and with a low guarantee to preserve the original quality of the available information. Raw data are often more reliable than processed data and ML techniques allow rigorous analysis of the raw data without adopting unnecessary assumptions for the analysis.

Boost business and product development: ML methods have the right potential to boost business opportunities and lead to more competitive products development cycles. This is due to a more effective way of discovering new information from the available data. There are many research applications, proving success stories of ML approaches. Image analysis [194], inference on interaction patterns [195], unsupervised [196, 197] and supervised learning [174, 197] (e.g. clustering and classification) are few of the problems, which can be tackled by ML methods. However, only a few users have adopted ML methods for industry and for practical applications. This is mainly due to the lack of understanding of new methods and fear associated with automating of the data analysis process. Nonetheless, there is indeed a growing interest within the industry as ML can increase predictive accuracy (e.g. <https://warwick.ac.uk/fac/sci/wcpm/studygroup3/>), provide superior models compared to current standard approaches and provide superior, thus help to provide better and more affordable products.

3. OPPORTUNITIES FOR COLLABORATION & INNOVATION

The research methods described in section 1 outline the characterisation techniques that can have the most impact on EES research and development in the UK. At this point, however, the methods are being carried out in an *ad-hoc* mode of operation – individual researchers or groups are developing and applying methods for particular projects and reporting their results in the usual manner through the scientific literature. As discussed in section 2 of this report, if the research could be coordinated with active communication across the characterisation techniques this will then enhance the standardization process, which will create numerous opportunities to enhance the impact of the results by quantifying the accuracy of each measurement and using machine learning methods to infer new directions for EES materials and systems. The opportunities for collaboration between researchers in the EES field was discussed in the workshop associated with this report, held at the University of Liverpool on July 16th, 2018 (see appendix C & D). In the following sections, we will discuss the results of the break-out sessions at this workshop and the recommendations of the community for characterisation capabilities moving forward.

3.1. UK-wide Connections & Future Developments

In the workshop held at the University of Liverpool on July 16th, there were 5 break-out sessions where the community of scientists involved in characterising EES systems discussed opportunities to improve the utilisation of the methods for the development of new devices. The results of these discussions are summarised in the sections listed below, along with the recommendation of each of the sessions for the pathways forward.

3.1.1. Large Scale National User Facilities

Large scale national facilities such as Diamond, ISIS, CLF and RCaH operate on a user proposal basis, allowing researchers to access facilities free of charge after undergoing a proposal submission and review process. These facilities are geared towards academic researchers performing fundamental studies (the submission process through to time on the instruments can take over 6 months) making it very challenging to get corporate researchers onto the facilities – although mechanisms exist to allow rapid access, the proposal mechanism is primarily an academic one, making a single one-off

experiment difficult to get through the review process. When EES systems are being developed, often it is a rapid single experiment that can make the difference in moving forward with the research or stopping it. There is also a concern that the leap from a benchtop experiment to the full characterisation by ISIS or Diamond can be too large a gap to overcome resulting in wasted efforts on the part of the researchers using the instrumentation and the beam scientists. The establishment of a battery facility in the RCaH overcomes many of these limitations, but it is clear that a specialised effort aiming specifically at EES research across the Faraday Institution research locations is needed. By distributing the effort around the country, so that all the research support is not focused into one geographical/scientific location and that work performed at national/midscale facilities can be used as a feeder into the large scale methods – working for the benefit of both the local researchers at universities and at Harwell. A coordinated approach to the use of large scale facilities through a Block Allocation Group (BAG) supported by the Faraday Institution may be the best approach to achieving the characterisation goals for EES devices, particularly if the BAG contains researchers working on the methods identified as being critical for EES advancement in specialised small to midscale facilities (see next section).

3.1.2. Small to Midscale Facilities at Universities (including National Facilities)

Access to small to midscale facilities in the UK is available through a myriad of different mechanisms. In some cases where there are EPSRC National facilities, access is through the same proposal process as for the large scale facilities described in the previous section (with the associated issues for EES research). In other cases, the facilities are supported on a pay per use hourly rate while in others access is in the form of informal collaborations. Some facilities are easier to access than others (and perceptions rather than facts tend to rule in determining this) while each facility has expertise in different areas (although they don't always identify areas of expertise and simply list the instrumentation within the facility). What is needed for EES research is a register of where the capability exists in the UK and a mechanism supported by the Faraday Institution for researchers to get access to the facilities (the only way to cut through all of the access modes is for this to be facilitated through a centralised agreement). The Royce Institute is making inroads to overcome some of these issues with a register of facilities it houses and its partner's house available through an on-line portal. In this regard, it is support of training for the users of the equipment rather than the capital equipment itself that is seen as the limiting factor for the full utilisation of characterisation tools in

the UK (see section 3.4). There are several methods that have been determined to be important for EES research, including TEM, SEM, NMR/MRI, UV/VIS, DEMS, GITT (and other electrochemical i.e. EIS), Raman, Infrared, XPS (near-ambient liquid/solid), EPR, Tomography, fluorescence, nanoSIMS, XRF, soft x-ray, SAXS, neutrons, PXRD and for all these methods, the key to innovating new EES technologies is the development of operando observations. **No one method at any facility can solve the entire range of intriguing questions regarding battery chemistries, phase changes, length/time domains that are important for new EES systems; coordinating the observations so that the results of each method can be interpreted across listed discipline on the fundamental scale is perhaps the key challenge in the use of characterisation techniques to accelerate EES innovation and is the key challenge the Faraday Institution address.** The full benefit of advances in the small-scale/mid-scale/large-scale facilities can be facilitated by coordinating research across all of these platforms – no one central facility will be able to provide all the methods needed and have the necessary widespread expertise to both develop new methods and to train users in the use of the methods for EES research.

3.1.3. Shared Facilities in the Corporate Sector

One of the issues raised in the discussion of corporate needs, was whether facilities in corporate laboratories could be used as shared resources. While there were many suggestions that this could be a good use of resources for some SMEs (trading access to some capabilities in return for use of others), it was not clear exactly how the issues of intellectual property could be managed in each case – simply knowing how a particular piece of equipment is set up by one company can give information to others as to the direction they are currently exploring scientifically.

An alternative to “shared facilities” as they are defined in the academic sense, was discussed by QinetiQ, with a proposal related to their new facilities for research in batteries. QinetiQ offer integrated capability generation and assurance stretching from research through to custom and prototype manufacture and systems of systems operational assurance. This includes facilities for power and energy research, experimentation, testing, evaluation and manufacture of prototype and bespoke products. The new power and energy facilities contain unique features such as extensive manufacturing equipment within a dry-room, environmental and abuse testing capability that extends to destructive testing and a state-of-the-art gas chromatography/mass spectrometry facility for analysis of trace compounds generated within cells. As the QinetiQ business model involves work routinely performed with organisations whose data is sensitive and confidential and with negotiable IP arrangements, customers can eliminate their capital expenditure investing in new equipment and expertise and move to an operational expenditure or investment model with precise guarding of valuable IP. In order to maximise the pace of advancement in the UK battery supply industry, open access facilities should support the widest possible group of innovators, scientists and other scientists with novel ideas. This will require from the companies to consider split revenue streams and the government to consider subsidising facility

maintenance to enable access by smaller companies and individuals with lower buying power. It will also put emphasis on government ensuring that government-funded activities do not undercut services available in the private sector. Once in place, this will give fast and confidential access to the broadest group to explore potentially disruptive solutions that could place the UK at significant advantage. UK industry should be considered central to Faraday initiatives as companies can pull on experience in particular applications to increase speed of development in the UK battery supply chain. For example, expertise within the automotive or defence sectors. Moreover, industry parties have broad supply chains that can be leveraged for UK benefit, including access and influence in international markets.

3.1.4. HPC and Machine Learning

The discussions in this break-out session focused on how machine learning could help with the identification of new EES technologies. There were many misconceptions in the approaches needed for machine learning by the experimentalists in similar misconceptions on how machine learning could help experimentalists by the mathematicians/computer scientists. Many in the room complained the fact that there is a lack of good postdocs in this area as many are snapped up by competitive salaries in the industry, which are not interested in studying the issues related to fundamental science. Everyone agreed that if we could find a way to train people with both the chemistry skills for EES storage systems and the math/computing skills to advance our understanding of the results, then this would be a big advance for UK research in this area (maybe through double majors or student exchanges). As the communities are widely spread (it is difficult to publish cross-cutting research on machine learning in a chemistry journal and hard to publish analysis of batteries in a machine learning journal due to the knowledge gap and lack of training in both areas of science), the Faraday Institution could play a key role in linking these fields. As for this to work out, there would need to be strong overlap between the metadata that is needed for machine learning and the interpretation of the output in terms of new materials, chemistries and processes, it was recognized that the only possible mechanism would be through a coordinated characterisation program. Only through a coordinated program could the characterisation results (the data format and metadata is often proprietary to the instrument manufacturer) be coordinated on the scale needed.

3.1.5. Establishing Standards

The Faraday Institution projects (and those of the broader Faraday Battery Challenge) are going to generate enormous quantities of data. In order to ensure reliability and inter-comparability, standards will be required. The discussion in this break-out session covered a range of topics relevant to battery research, including standard materials, fabrication procedures, test protocols, metadata, modelling software and hardware fixtures. One concern, particularly for academic researchers, was the loss of knowledge associated with the high

turnover of PhD students and postdocs, which is often exacerbated by a lack of robust documentation of laboratory procedures. It was agreed that standardisation of many of these aspects would be beneficial but this would also have to be weighed against the cost and complexity associated with widespread implementation. The general consensus was that the degree of rigour of the quality systems in place at organisations such as NPL would not transfer easily to an academic environment and some compromise would be required. The key training role of the Faraday Institution was emphasised a number of times throughout the discussion.

With regard to test methods, one of the challenges identified was that they are continually being modified and adapted, making it difficult for standards to keep up. The use of in situ/operando cells is a particular case in point. It was agreed that multi-modal cells incorporating a number of complementary characterisation techniques would be a useful approach. Overall, it was recognised that a suite of characterisation tools would be required to facilitate cross-correlation of test data. Round robin exercises would also be valuable to establish consistency in testing. There is also an opportunity to apply machine learning techniques to the large datasets that will be generated but this will require asking the right questions and understanding the format of each dataset. This is an area in which learning could be transferred from the pharmaceutical industry. It was also stressed that the same formatting and metadata issues apply to modelling as commenting in open source software is often insufficient.

2.2. Instrument Sharing and Capital Purchases

The research capabilities that are described in this report are to the most extent widely available in existing facilities at universities and national facilities in the UK. However, key to the development of coordinated efforts in EES systems is a mechanism to collaborate effectively across institutional and traditional research field boundaries. In this section, a potential mechanism identified by the participants at the workshop is discussed along with a recommendation for near-term instrumentation purchases that can enhance the research being performed across the country.

2.2.1. Shared Equipment

As stated previously in this report, there are many mechanisms that are currently in play for access to the state-of-the-art characterisation tools that can greatly assist in the understanding of EES device functionality, and navigating these different access modes can be a discouraging proposition for anyone trying to analyse a new discovery. No one facility in the UK can provide all the needs for the community in terms of the range of equipment, and there is clearly a need to enhance the expertise in characterisation of EES systems at many of the existing facilities that could support EES research. Any model that is developed

for ensuring access to methods and training in their use must therefore address the whole characterisation endeavour if it is likely to be successful – continuing a piecemeal set of characterisation support will result in more of the fragmented approach that currently exists.

Developing a method that allows for the widespread dissemination of expertise across the country is by no means an easy proposition. Laboratories that have expertise typically guard the difference making expertise as it has tremendous value in applications for research funding. In addition, researchers that have expertise in EES systems often do not want to give that expertise away to characterisation facilities at other institutions for the same reasons. In most cases this results in an uneasy standoff between the synthesis of battery materials and the methods of employing them in batteries and the characterisation of these systems that could lead to understanding new technologies. While the need to respect each individual researcher's independence is clear from the way science functions, this stand-off does not particularly lead to a rapidly advancing national strategy for characterisation that is in the best interest of the country going forward. Any model for collaboration involving shared resources should therefore aim to meet each contributor in the middle and provide benefit to all parties involved in the research.

The need to be responsive to multiple researchers and institutions to get the most out of characterisation methods, naturally points towards a Faraday Institution wide project that can set-up the details of sharing costs, intellectual property and publications centrally rather than have each bilateral characterisation experiment have to go through a drawn out negotiation each time. The centralised organisation of such a research strategy also points towards the well-established hub(s) and spokes model for shared resources. Institutions with a wide range of characterisation methods and expertise that are relevant to EES systems become hubs, while satellite facilities or spokes exist to incorporate individual pockets of expertise or unique capabilities that are present in other institutions. Both the hubs and the spokes help develop new methods and bring in users by feeding them from one facility to another depending on the expertise that is required. For this to work well, it is important that there is a centralised agreement on resources and funding, as if you require paid users to maintain your facility, there is no driving force to send them to another facility to do that research – you are better off doing the work yourself, even if you do not have expertise in the area. Again, the well-known limitations in getting the best out of characterisation are all related to the speed and cost of access and this model can overcome many of these issues. It should also be noted that this hub(s) and spokes model also automatically involves a rapid agreement on the metadata that is needed for the experiments to be widely interpreted and facilitate the machine learning approach described earlier in this report – you cannot move people from facility to facility for the best interests of their research if the data they get from one facility is not compatible with data generated in another institution.

This hub(s) and spokes model is employed regularly in the US, where the Department of Energy (DOE) National Laboratories act as the hubs and collaborating universities operate as the spokes. The national

laboratories get funding for the facilities in the hubs depending on the number of users and the output they deliver. The university researchers get funding from DOE in terms of access to the facilities, travel support and direct research funding dependent on the number of publications (amount of new science) that are generated using research facilities at the national laboratories. This approach therefore provides a gain to both the expert beam scientists at the national laboratories and the researchers working on energy materials at the universities. The free flowing research and research staff between the hubs and the spokes also provides a dynamic environment for research where new ideas from around the country are brought together to stimulate new areas of research. A similar approach has also been instigated as part of the Royce Institute, where funding for the core facilities is provided to the researchers with the expertise and funding to use the facilities is provided to the researchers who need the characterisation – this encourages facilities to go out and interact with potential users. A similar approach across the EES characterisation community could be implemented by the Faraday Institution.

3.2.2. Capital Purchases

In many cases the instrumentation necessary to characterise EES systems is present at facilities across the country. However, the main advances leading to innovations in understanding the processes involved at the fundamental level requires the development of Operando methods for these capabilities. Typically turning a high spatial/temporal/spectral resolution measurement into an operando measurement involves the development of a specialised holder and/or stage that costs ~£10-200k depending on the instrumentation being used. This is the same order of magnitude that is needed to provide the specimen preparation and environmentally controlled glove boxes that can maintain EES systems free from degradation/contamination when exposed to the atmosphere (typically a glove box will cost ~£50k depending on the size and the analytical equipment that goes in it). Investing in these types of equipment can go a long way to providing capabilities for EES research that are comparable to the leading facilities in the world.

Of course, there will always be a need to invest in new major equipment purchases – a 10 year old TEM does not have the resolution, analytical capabilities or specimen throughput of a new one. However, the multi-million pound investments in new equipment that focus on EES research could be coordinated through the Faraday Institution and draw on established demonstrations of collaborative developments that benefit EES research to make these recommendations – again this is a major positive of a centrally organised hub(s) and spokes model for characterisation. Although most of the equipment in the multi-million pound category can be readily converted to operando measurements for a fraction of the cost (see previous paragraph), one area where there appears to be a significant lack of state-of-the-art equipment is with NMR. Facilities in the UK appear to be oversubscribed in terms of access. Here, the expertise and investment by the Faraday Institution

in new equipment or providing more access to existing equipment worldwide should be considered.

3.3. Gaps & International Collaborations

While there are the facilities and expertise in the UK to provide all the capabilities described in the previous sections of this report, there are cases where there are limitations to access caused either by the needs of other users or insufficient expert staff time to perform the experiments. In the case of large/mid-scale national facilities, there can be delays in access caused by the proposal process (which can be overcome in part by the BAG scheme described above). In the case of mid-scale facilities at universities, access is either limited to freely available collaboration time or available at a cost – both of which can cause issues if the experiment is urgent. In the case of the smaller facilities, the access to the capabilities is typically only limited by expertise; without an expert user to perform the experiment (usually from the group that designed/runs the equipment) high quality characterisation data is impossible to obtain. The previous sections of this report outlined mechanisms that could improve the access to characterisation capabilities within the UK. However, as an intermediate step in the process of establishing expertise at UK facilities, the Faraday Institution could encourage researchers to access international facilities through joint research agreements and travel grants. Perhaps the best way of achieving this would be for example through collaborations with the National Laboratories in the USA. The US Department of Energy has invested heavily in characterisation of EES systems over the last 10+ years (over \$100M) and has by far the largest collection of methods and expert users in the world. These facilities operate on the same user model as the large/mid-scale national facilities in the UK and if access to the facilities can be negotiated for UK researchers (other than the standard procedure of applying for time, which has already been identified as a major limitation for both academic and industrial researchers) there are major gains that can be made in establishing expertise. Of particular importance would be collaborations; with Oak Ridge National Laboratory (ORNL) for neutron scattering; with Brookhaven National Laboratory (BNL), Argonne National Laboratory (ANL) and Lawrence Berkeley National Laboratory (LBNL) for synchrotron studies; with Pacific Northwest National Laboratory (PNNL), ANL and BNL for NMR observations; with PNNL and ANL for mass spectrometry; with PNNL, LBNL, BNL and ORNL for TEM; and with ANL and ORNL for scanned probe instrumentation.

2.4. Training Opportunities

In many of the discussions of the use of advanced characterisation tools, the subject of qualified users is always raised as a critical issue. As the methods needed for characterising advanced EES systems become more complex (particularly with the operando methods that are discussed here) and the requirements for the materials and processes become more stringent (delivering higher performance devices), the need for expertise will only continue to grow. This need for this expertise becomes even more critical when the discussions of the use of data involve advanced analytics and machine learning (ML). For the analytics and ML to work well, there needs to be well archived datasets conforming to established standards of calibration and metadata. Ideally, the scientists generating the data will be familiar with the process of the analytics/ML to ensure coherency between methods. However, this adds another layer of complexity to the training that is required (and looking at the problem from the other direction, it will require mathematicians trained in chemistry and characterisation methods). Training on this level is not likely to occur without a centrally managed characterisation and data portfolio. With such a managed scheme (as is discussed here with the hub(s) and spokes model) there is an opportunity to achieve a level of senior undergraduate/graduate/technical training that is unique with the worldwide EES community. The expertise in the UK in EES, characterisation, materials chemistry and analytics/ML is (as is shown here) at the world class level, but it is dispersed across multiple institutions. The virtual laboratory approach that the Faraday Institution is proposing where the ability for students/technicians to take advanced courses across the country (moving between laboratories with the world class expertise) would facilitate these interactions. For example, the operando microscopy suite at the University of Liverpool is offers undergraduate, graduate, technical, and supervisory courses in the use and application of TEM methods to study chemical dynamics. Although the content of the courses is essentially the same, the courses are offered over a range of timescales to facilitate professionals wishing to have a quick on-site training course, undergraduates wishing to spread the training over a whole semester and everyone in between. It would be relatively straightforward to establish similar courses across the institutions with the expertise in the methods that would be available to everyone working in the Faraday Institution, thereby accelerating the technical skills across the disciplines for scientists working in the EES field.

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APPENDIX A: QUESTIONNAIRE

Identifying research infrastructure and collaborative expertise for electrochemical energy storage (EES) applications

| |
|-----------------------------|
| Name: |
| Role: |
| Research Department: |
| Research Institute: |

Please give an answer to the following questions:

- 1) What research are you currently undertaking that is/could be of major benefit to EES?
- 2) Do you have enough experimental facilities for future expansion of EES experiments?
- 3) Do you use shared/national user facilities for any part of your research, and if so which ones? If you currently don't use shared facilities, please explain why and under what circumstances you would use such facilities in the future.
- 4) What results/simulations do you rely on for interpreting your experiments?
- 5) How do you calibrate new methods and/or observations in your work to establish reproducible parameters for your developments?
- 6) What do you think is the largest error associated with the experimental results your research generates?
- 7) Do you think there is a benefit in generating "Faraday Institution approved datasets" where the calibration and accuracy of the results are defined by standards?
- 8) What do you think constitutes good, reproducible and widely sharable data for EES research?
- 9) What mechanisms do you think can be used to share information developed through primary characterization facilities to provide useful insights into materials?
- 10) Is there any other information you want to share?

If you are an academic involved in characterisation of electrochemical storage materials

- 11) What is the current level of external access to your research battery characterisation infrastructure?
- 12) Where do you think there are infrastructure gaps where there is critical need for new investment?

If you are from the chemical, materials or batteries sector and are engaged in the development of electrochemical materials for energy storage

- 13) What facilities do you currently have that are delivering ground-breaking / reproducible results?
- 14) Where does the infrastructure for your research lie in the UK? (Both in terms of the hardware and in the thought leadership)?
- 15) How is data generated from these characterization methods and how is it integrated into an overall understanding of the key factors controlling novel EES processes?

Is there a balance that depends on the type of information that is needed?

If there is a balance, how do you weight potentially contradictory results?
- 16) What would you propose as a calibration strategy that could be implemented on the national scale to deal with similar conflicts?
- 17) What infrastructure gaps exist in either your own establishment and/or nationally that would help accelerate innovation in ESS

APPENDIX B: LIST OF RESPONDENTS TO QUESTIONNAIRE

Emma Kendrick, University of Warwick
Carla Shepherd, University of Warwick
Patrick Unwin, University of Warwick
Dharmika Widanalage, University of Warwick
Helen Ryder, University of Manchester
Bob Cernik, University of Manchester
Katie Moore, University of Manchester
Chris Lucas, University of Liverpool
Matthew Dyer, University of Liverpool
M J Rosseinsky, University of Liverpool
Laurent Chapon, Diamond Light Source/University of Oxford
James Naismith, RCaH/University of Oxford
Peter Nellist, Angus Kirkland, Chris Grovenor, James Marrow and Martin Castell, University of Oxford
Kathryn Toghill, Lancaster University
Harry Hoster, Lancaster University
Gareth Hinds, National Physical Laboratory
Alan Drew, Queen Mary University
Robert McGreevy, ISIS
Daniel Auger, Canfield University
Nuria Garcia-Araez, University of Southampton
Sven Schroeder, University of Leeds
Gareth Moody, Croda Lubricants
Philip Withers, Royce Institute/University of Manchester
David Hodgson, PV3 Technologies

APPENDIX C: WORKSHOP AGENDA



UNIVERSITY OF LIVERPOOL

Infrastructure to Knowledge: Advanced Characterisation Methods to Accelerate the Development of Novel Electrochemical Energy Storage Devices

16 July 2018: Foresight Centre, University of Liverpool, 09.30 – 17.00

Workshop Agenda

| | |
|----------------------|---|
| 09.00 - 09.30 | Registration |
| 09.30 – 9.45 | Introduction and Workshop Overview Nigel Browning , University of Liverpool |
| 09.45 – 10.00 | Faraday's Challenge Peter Littlewood , Faraday Institution |
| 10.00– 10.15 | What RCaH can offer battery science Jim Naismith , RCaH and University of Oxford |
| 10.15 – 10.30 | Advanced X-ray characterization of materials at the Diamond Light Source Laurent Chapon , Diamond Light Source |
| 10.30 – 10.45 | Neutron and muon studies of batteries Bill David , ISIS and University of Oxford |
| 10.45 – 11.10 | NMR and MRI studies of batteries: status, challenges and future prospects? Clare Grey , University of Cambridge |
| 11.10 – 11.30 | Coffee Break |
| 11.30 – 11.55 | Fundamental electrochemical characterization of energy storage materials Nuria Garcia-Araez , University of Southampton |
| 11.55 – 12.20 | Experiences with Characterisation Infrastructure in JCESR Lei Cheng , Argonne National Laboratory |
| 12.20 – 12.30 | Q&A |
| 12.30 – 1.30 | Lunch |
| 1.30 – 2.00 | National Energy Policy in a Local Context James Johnson , Liverpool Enterprise Partnership |
| 2.00– 3.00 | Break-Out Sessions (1) 1. Large Scale Facilities Discussion Leads: W. David & J. Naismith |
| | 2. Small to Midscale University Facilities Discussion Leads: L. Hardwick & N. Garcia |
| | 3. Shared Corporate Facilities Discussion Lead: J. Cookson |
| 3.00 – 3.30 | Measurement & Standards Infrastructure Gareth Hinds , NPL |
| 3.30 – 3.50 | Automated Imaging, Simulation, and Experimentation in Battery Development A. Stevens , Optimal Sensing, USA |
| 3.50 – 4.00 | Report on Break-out Session (1) |
| 4.00– 5.00 | Break-Out Sessions (2) 1. HPC and Machine Learning Discussion Lead: E. Patelli |
| | 2. Establishing Standards Discussion Lead: G. Hinds |
| 5.00 | Close and Reception |

THE UNIVERSITY IS A MEMBER OF THE ELITE RUSSELL GROUP OF RESEARCH-LED UK UNIVERSITIES

APPENDIX D: LIST OF WORKSHOP PARTICIPANTS

- Peter Littlewood**, Faraday Institution
- Martin Tillin**, Faraday Institution
- Allan Paterson**, Faraday Institution
- Nigel Browning**, University of Liverpool
- Houari Amari**, University of Liverpool
- Filipe Braga**, University of Liverpool
- Glenda Wall**, University of Liverpool
- Chris Lucas**, University of Liverpool
- Roberto Ferrero**, University of Liverpool
- Layla Mehdi**, University of Liverpool
- Laurence Hardwick**, University of Liverpool
- Yvonne Grunder**, University of Liverpool
- Matthew Dyer**, University of Liverpool
- Asim Mumtaa**, University of Liverpool
- Karl Whittle**, University of Liverpool
- Lin Jiang**, University of Liverpool
- Filipe Braga Nogueira**, University of Liverpool
- Vicki O'Kelly**, University of Liverpool
- Michael Craven**, University of Liverpool
- Yundong Zhou**, University of Liverpool
- Alex Cowan**, University of Liverpool
- Brian Connolly**, University of Manchester
- David Eastwood**, University of Manchester
- Sarah Haigh**, University of Manchester
- Robert Weatherup**, University of Manchester
- Helen Ryder**, University of Manchester
- Robert Cernik**, University of Manchester
- Richard Fields**, University of Manchester
- Thomas Heenan**, University of Birmingham
- Peter Slater**, University of Birmingham
- Melanie Britton**, University of Birmingham
- Paul Anderson**, University of Birmingham
- Daniel Reed**, University of Birmingham
- Phoebe Allan**, University of Birmingham
- Anup Barai**, University of Warwick
- Shanwen Tao**, University of Warwick
- Faduma Maddar**, University of Warwick



Changhui Chen, University of Warwick

Barbara Shollock, University of Warwick

Jonathan Davidson, University of Sheffield

Charles Oseghale, University of Sheffield

Jake Entwistle, University of Sheffield

Kangala Chipasa, University of Sheffield

Mauro Pasta, University of Oxford

Peter Nellist, University of Oxford

Bill David, UKRI-STFC & University of Oxford

Liyu Jin, University of Oxford

Rhodri Jervis, University College London

James Dodwell, University College London

Ruoyu Xu, University College London

Chun Tan, University College London

Nuria Garcia-Araez, University of Southampton

Denis Kramer, University of Southampton

Pooja Panchmatia, Loughborough University

Sina Saremi, Loughborough University

Denes Csala, Lancaster University

Hamid Hosseini, Newcastle University

Amor Abdelkader, Bournemouth University

Alan Drew, Queen Mary University of London

Poopathy Kathirgamanathan, Brunel University London

Leonardo Lari, University of York

Sven Schroeder, University of Leeds

Christos Kalyvas, University of Hertfordshire

Rong Lan, Coventry University

Qiong Cai, University of Surrey

Laurent Chapon, Diamond Light Source

Julia Parker, Diamond Light Source

James Naismith, Research Campus at Harwell

Alexandra Johnston, Advanced Materials Processing Lucideon

Alexandra Johnston, Advanced Materials Processing Lucideon

Stuart Maclachlan, Lucideon

Zoe Walton, Metiscube

Elizabeth Cooper, ICoNiChem Widnes Ltd

Mike Cox, ICoNiChem Widnes Ltd

Gareth Hinds, National Physical Laboratory

Gareth Moody, Croda

Anna Wise, Innovation Lead – Batteries Innovate UK

Susan Smith, Daresbury Lab STFC

Marcus Rutherford, 2DHeat Ltd

Iain Aldous, Stephenson Institute for Renewable Energy

Matthew Davis, Henry Royce Institute

John Conti-Ramsden, CPI Ltd

Rachael Rowlands Jones, KTM Formulation

Mechthild Luebke, Cummins Electrified Power

Victoria Doherty, Energy Distribution QinetiQ

Darren Ragheb, Centre for Process Innovation

Richard Clark, Morgan Advanced Materials

Matthew Haley, ZEISS

Arne Sandschulte

Daniel Atkinson

Sijia Yu

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For further information please contact:

mcteam@liverpool.ac.uk