D5.1 – State-of-the-art experimental matrix, tier 1 experimental plan and workflow

VERSION

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OBJECTIVE

The main objective of WP5 is to develop synergies to initiate a pilot action towards the implementation of a European multi-modal experimental platform using standardized cells/protocols/metadata/data collection, treatment and analysis. The concept will be demonstrated on a selected chemistry using a subset of lab-scale- and large-scale-facility (LSF) techniques. D5.1 is the first step towards this goal by setting the state-of-the art experimental matrix, selecting Tier1 techniques and providing the corresponding experimental plan and workflow. The report describes the WP5 organisation during the first period (M1-M6), where partner competence matrixes and a cluster-type transversal classification were established to map the capabilities in terms of equipment, methodologies, know-how and battery cells. These settings were used to define key priority experiments and their coordinated implementation to generate BIG-MAP lab-scale and Large Scale Facilities data of many types, including operando data, according to the global project workflow.

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1. Introduction

1.1. Objectives of WP5

The objective of WP5 is to develop synergies to initiate a pilot action towards the implementation of a European multi-modal experimental platform using standardized data collection (cells, protocols, metadata etc.), treatment and analysis. The experiments conducted in WP5 will generate data of many types across a large range of length- and time-scales. A wide variety of techniques at lab-scale and at the Large Scale Facilities will be combined to characterize the BIG-MAP battery materials and devices *ex situ*, *in situ* and *operando*. Data will be transferred to central BIG-MAP and all WPs (raw data, processed data, analysed data and output parameters). The workflow in WP5 integrates time-coordinated and site-coordinated experiments, cross-correlated data analysis including fidelity and reproducibility assessments, feed-back loops with other WPs to adapt and design new experiments, as well as demonstrators of on-the-fly control and monitoring using artificial intelligence (AI) and modeling. The first period was devoted to establish the grounds required for an efficient implementation, testing and dynamical refinement of the global methodologies along the project. Several tools were designed to guide the coordinated efforts of all partners in this direction:

◊ Partners know-how and means organized into the “competence matrix”
◊ Classification of available techniques into clusters

These tools enabled the construction of the experimental matrix that classifies the types of data and experiments of interest for battery characterization, as well as the selection of Tier 1 techniques to be used from M6 to M30, while identifying some potential Tier 2 techniques (M20-M36). The experimental plan was defined after careful inspection of partners’ capabilities, identification of first-stage materials and anticipated materials flow, project priorities and initial plans of cooperation and interoperability.

1.2. Interactions with other WPs

Figure 1 graphically illustrates the dense interaction between WP5 and the other WPs. Specifically, four feedback loops can be identified.

◊ WP5 provides in-depth/high through-put/high fidelity data on the selected materials from WP4 and WP6 participating in the process of developing better electrolytes, additives or coatings.

◊ WP5 tests and provides feedback to the standard protocols proposed by WP7 and WP8.

◊ WP2/WP3 and WP5 will design models and experiments, respectively, to improve the atomic and multiscale models.

◊ WP5 will acquire, format, deliver and store data following WP9, WP10 and WP11 recommendations to ensure the formation of relevant data based usable by the AI.

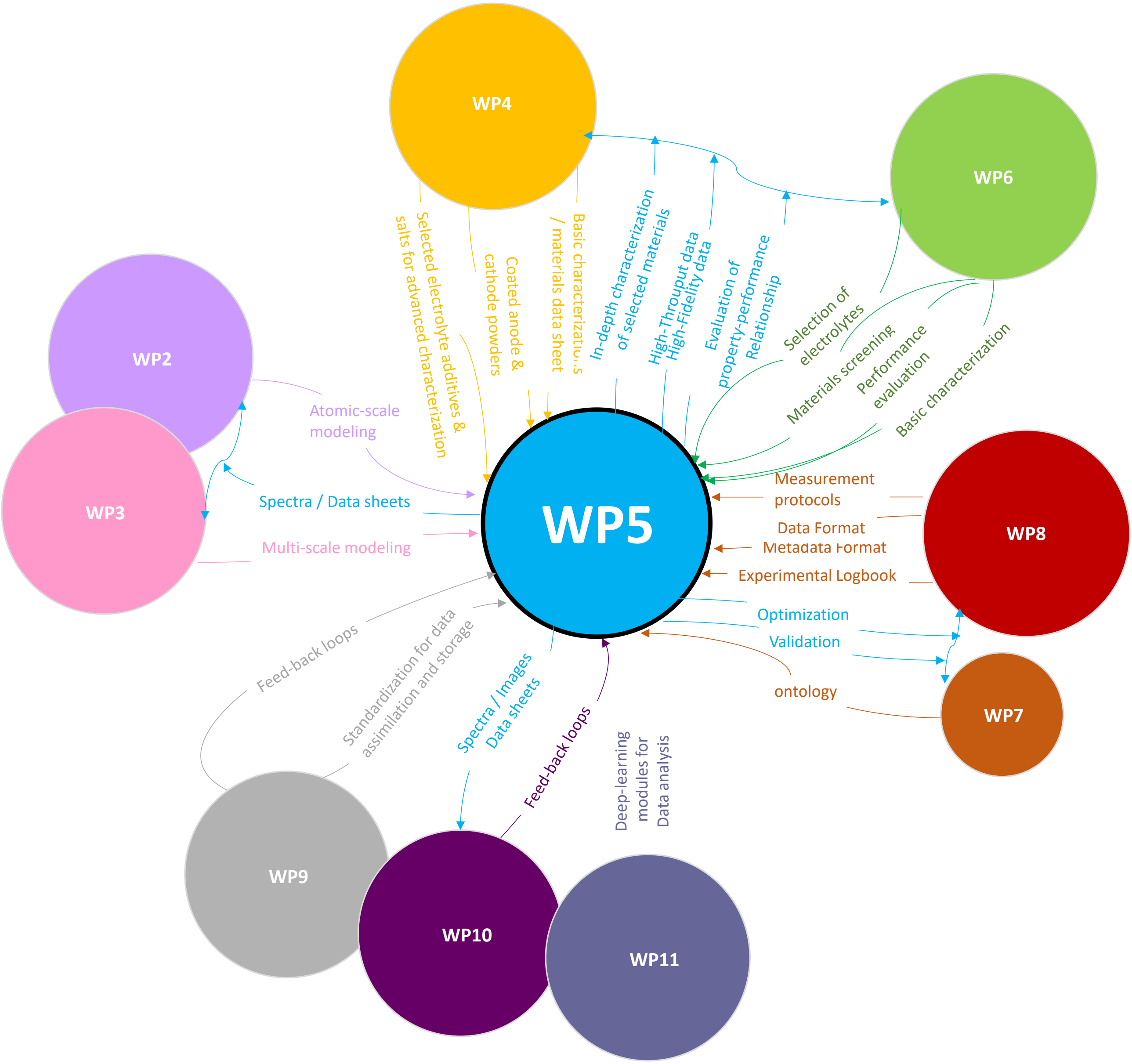

Figure 1 : The role of WP5 in the BIG-MAP and interactions with other WPs.

1.3. WP5 Internal organization

WP5 is composed of four tasks all connected to different work packages (Figure 2).

◊ **Task 5.1** consists in screening, organizing and describing the available techniques in WP5 and results in the construction of the experimental matrix. The selection of criteria and parameters for this benchmark has been performed thanks to a cluster organization adopted in WP5 (described below) and the participation of WP8 and WP7. The experimental matrix will be used by other work packages as guideline to understand the capabilities of WP5. The experimental matrix is to be used to define the Tier 1 and Tier 2 techniques.

◊ **Task 5.2** consists in structuring the workflow which will be dynamically modifying along the project by several inputs such as: experimental feedback from internal data analysis, insights from the modelling work packages (WP2 and WP3), standard testing procedures identified in WP8, and materials for WP4 and WP6. The workflow, together with the definition of the Tier1 and Tier2 techniques will lead to the formation of a set of experimental plans.

◊ Experiment plans are performed, with the data acquisition and pre-processing being the **Task 5.3**. After pre-processing, the data might directly go the other WPs, typically, the modelling WPs (WP2 and WP3), or the WPs responsible for the AI architecture (WP9, WP10, WP11).

◊ The pre-processed data might also be further analyzed in WP5 according to Task 5.4. Again, the output parameters can be transfer to several WP, or back to Task 5.2 for the refinement of the workflow and the definition of the next experiment plan.

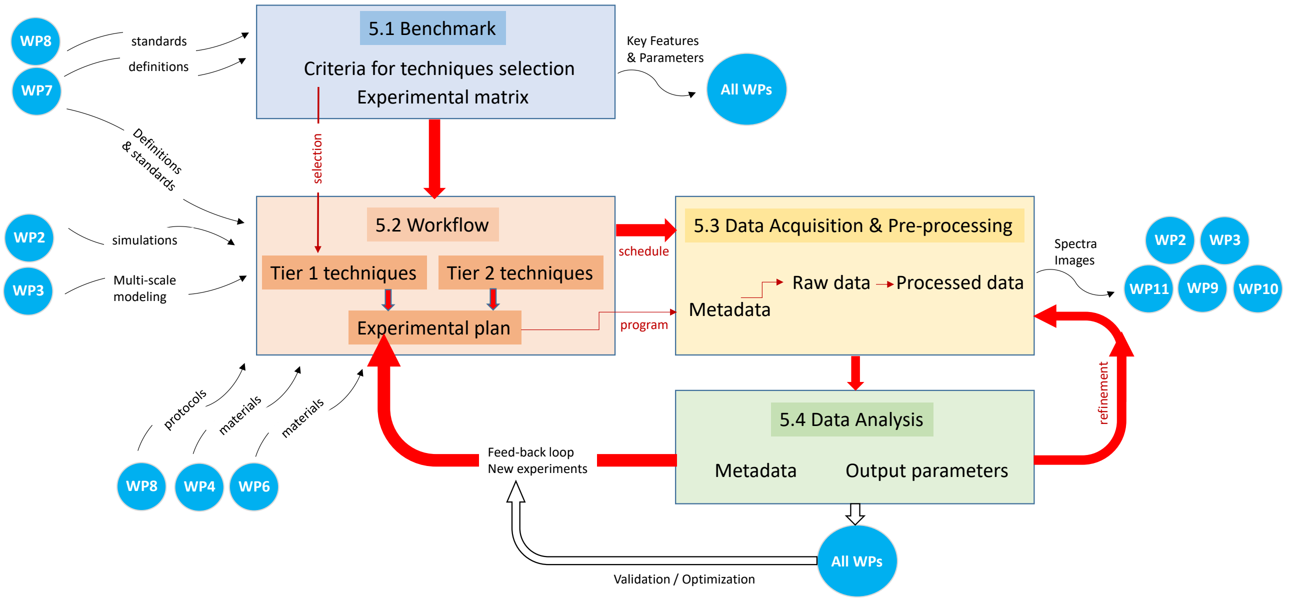

Figure 2: Internal workflow of WP5

The partners and task leads of WP5 are identified and presented in Figure 3.

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Figure 3: Summary of the partners.

1.4. Aim of the deliverable: Task 5.1 and 5.2

D5.1 reports the state-of-the art experimental matrix, selection of Tier1 techniques and corresponding experimental plan and workflow. The deliverable describes the WP5 organisation during the first period (M1-M6), where partner competence matrixes and a cluster-type transversal classification were established to map the capabilities in terms of equipment, methodologies, know-how and battery cells. These settings were used to define key priority experiments and their coordinated implementation to generate BIG-MAP lab-scale and Large Scale Facilities data of many types, including operando data, according to the global project workflow. The deliverable is organized in two parts:

1) **The experimental matrix (Task 5.1 - Benchmark).**
   First, the result of the competence mapping is shown giving an overview of expertise and workforce amongst the different partners of WP5. Second, the cluster organization adopted in response to the wide range of techniques is described. Finally, the experimental matrix is shown with a detailed discussion on the choice of the criteria and parameters organizing it.

2) **Tier 1 techniques experimental plans (Task 5.2 – Workflow).**
   Analysis of the experimental matrix allowed tier 1 experiments to be selected. Experimental plans were gathered from each partner using templates, which were discussed, rationalized, and assembled to produce a detailed experimental plan for next period (M6-M12) and vision for the longer-term development of Tier 1 experiments.

2. Benchmark

This part presents the screening strategy of the available techniques, the competence matrix, and the cluster organization of WP5 resulting from the wide range of expertise.

2.1. Competence Mapping

Due to the large number of techniques, establishing the competence matrix is a complex task. To facilitate the visualization of the diversity present in WP5, a graphical representation is adopted: the competence mapping. To construct the competence mapping, the techniques were gathered and grouped into three high-level different categories, i) laboratory experiments, ii) neutrons experiments and iii) synchrotron experiments.

For each category, the techniques are plotted on a bubble chart, with the size of the bubble being proportional to the number of partners able to carry out the experiment (Figures 4-6). The x and y axis are the typical length scale probed (Å, nm, µm) and the possibility to achieve 1D, 2D or 3D property mapping, respectively. In this representation, a 1D property or parameter is typically a mean value averaged over the volume of the probed material, while 2D indicates the possibility to scan across the thickness of a material and spatially-resolve the property distribution along one dimension. 3D indicates that the full volume (region of interest) is resolved and voxel-type information is obtained. Moreover, the possibility to perform *operando* experiments is indicated by a green contour around the name of the technique. As an example, Figure 4 shows the different laboratory experiments available to the consortium. X-ray diffraction (XRD) experiments, which probe the atomic structure (length scale: Å) without spatial resolution (1D, e.g. averaged bulk crystalline structures within an electrode) can be performed by seven partners, namely Uppsala University, TU Delft, Oxford, University of Cambridge, CNRS, CEA and DTU. As *operando* XRD is quite common in most of these laboratories, the label "XRD" is contoured in green.

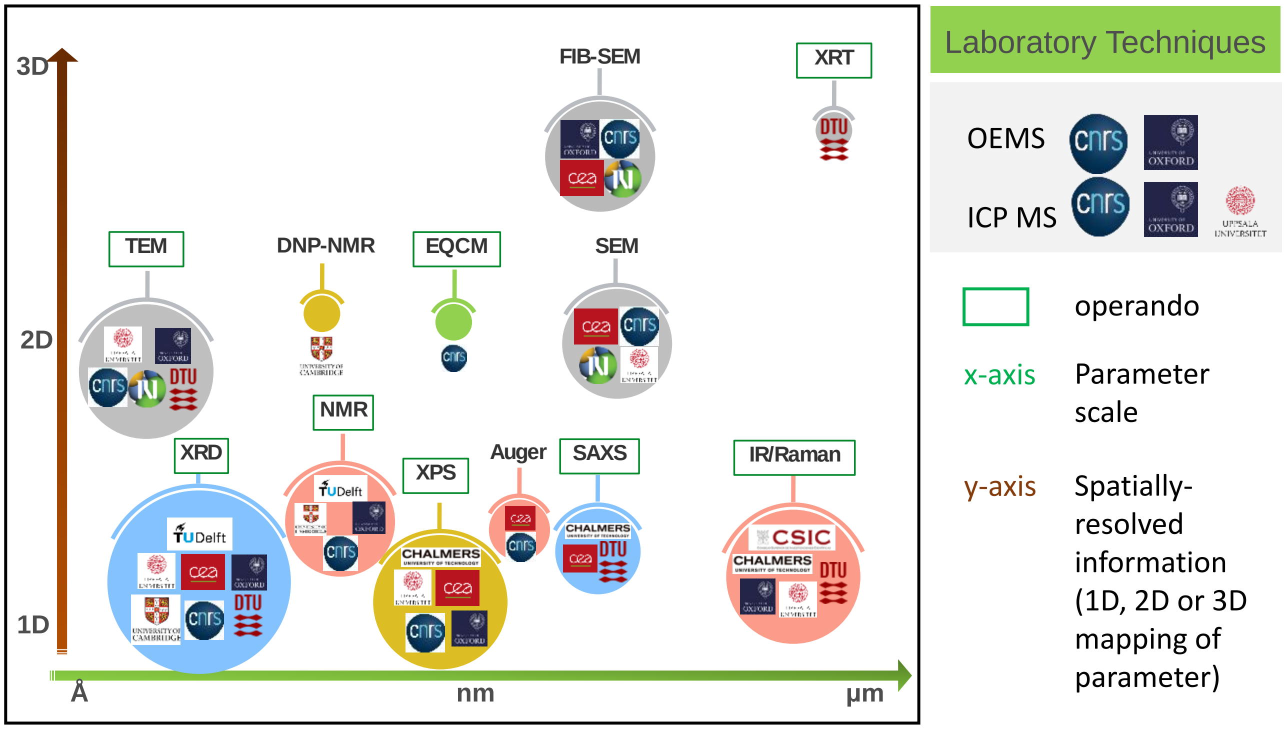

Figure 4: Bubble chart summarizing laboratory scale techniques. The size and the color of the bubbles stand for the number of partners, and the cluster to which the technique belongs. Blue, pink, yellow and grey is for the scattering, bulk spectroscopy, surface spectroscopy and imaging, respectively.

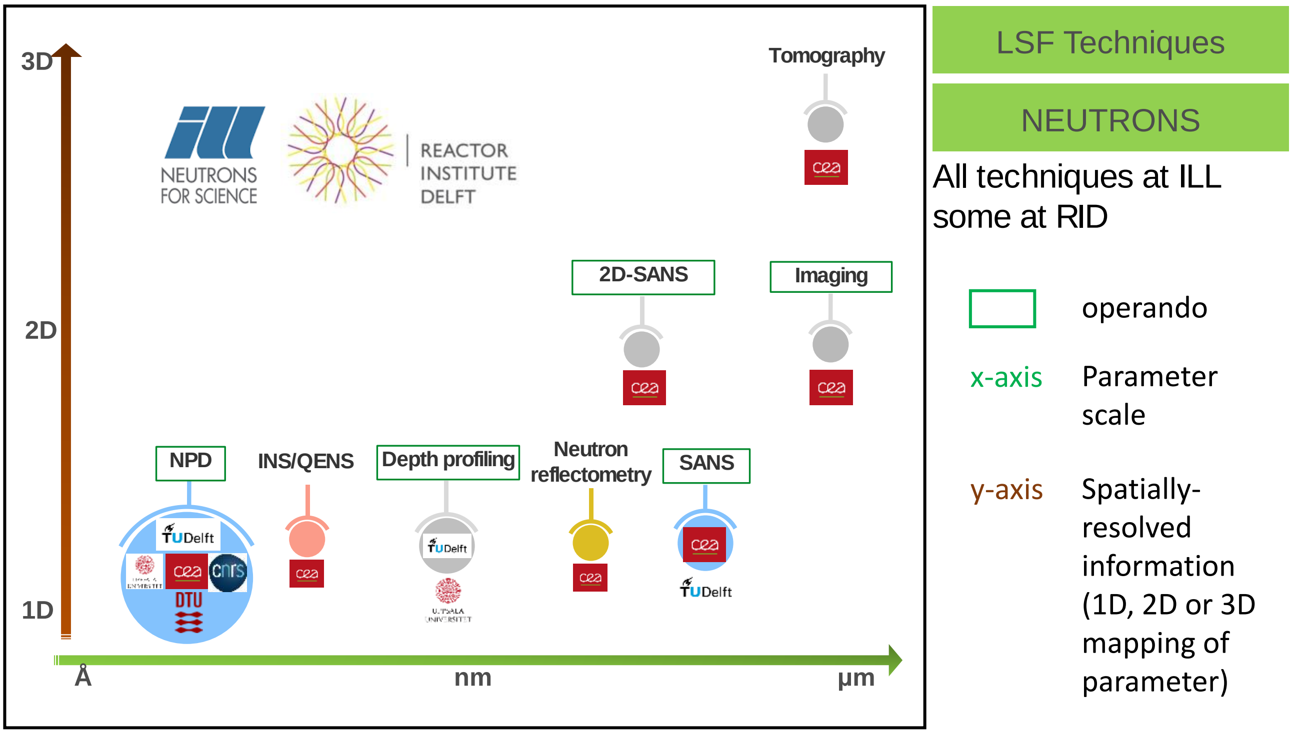

Figure 5: Bubble chart summarizing the neutron techniques. The size and the color of the bubbles stand for the number of partners, and the cluster to which the technique belongs. Blue, pink, yellow and grey is for the scattering, bulk spectroscopy, surface spectroscopy and imaging, respectively.

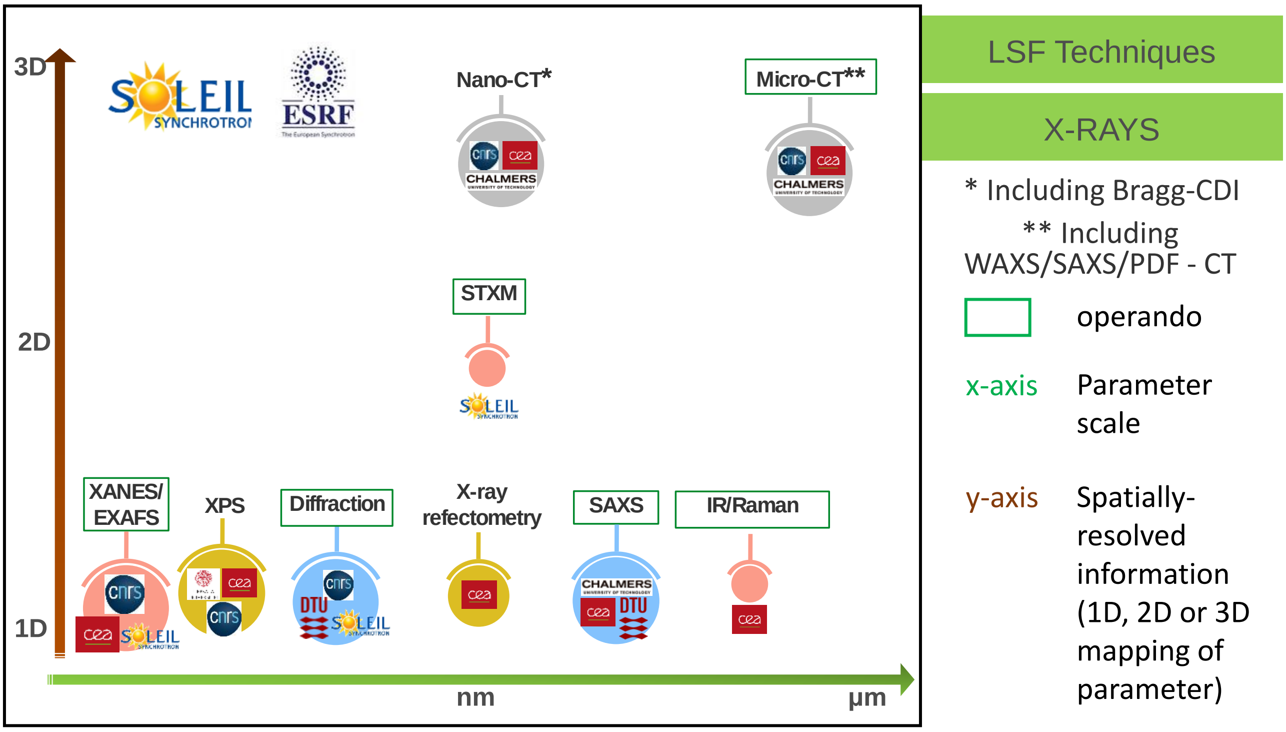

Figure 6: Bubble chart summarizing the synchrotron techniques. The size and the color of the bubbles stand for the number of partners, and the cluster to which the technique belongs. Blue, pink, yellow and grey is for the scattering, bulk spectroscopy, surface spectroscopy and imaging, respectively.

This screening shows quite clearly the large number of techniques available in the consortium with a good distribution between the different length scales and dimensions, together with the possibility for *operando* measurements. Moreover, most techniques can be performed by at least two partners ensuring the possibility to verify the fidelity of the data – an important goal of WP5.

The current representation fails to represent numerous important information such as, for example the depths of probe, and the nature of the information. Typically, XRD and XPS both are 1D techniques probing Å-nm but the latter probes bulk long range ordered structure while the former gives information about the surface local structure. Moreover, the basic principles of both techniques, namely scattering and absorption, are different. Clearly, a more detailed representation is needed, featuring for example the depth of probe or the type of chemical information. To identify the categories, more technical discussion was needed, which lead to the formation of clusters amongst WP5.

2.2. Cluster organization

Four cluster were created and a chair person identified for each of them: P. Norby (DTU) for the diffraction and scattering, A. Matic (Chalmers) for the bulk spectroscopy, K. Edstrom (Uppsala University) for the surface spectroscopy and S. Lyonnard (CEA) for the imaging. The techniques and partners involved are summarized **Figure 7**.

The rationale behind subdividing the WP5 actors into clusters is to allow for more direct interaction between techniques that are a) more naturally combined together and b) offer directly comparable and/or complementary information. Each cluster lead has the responsibility to organize cluster meetings, which take the form of periodic short meetings or workshop-type events. Each cluster has discussed experimental plans, data reproducibility, *operando* cell availability, tier1/tier2 techniques. The cluster substructure thus allows for expertise and resource transfer at a deeper level than is possible at the overall WP5 level. The cluster leads then represent their cluster activities on the WP5 general meetings and act as an intermediate point of contact between individual actors and the WP5 leadership.

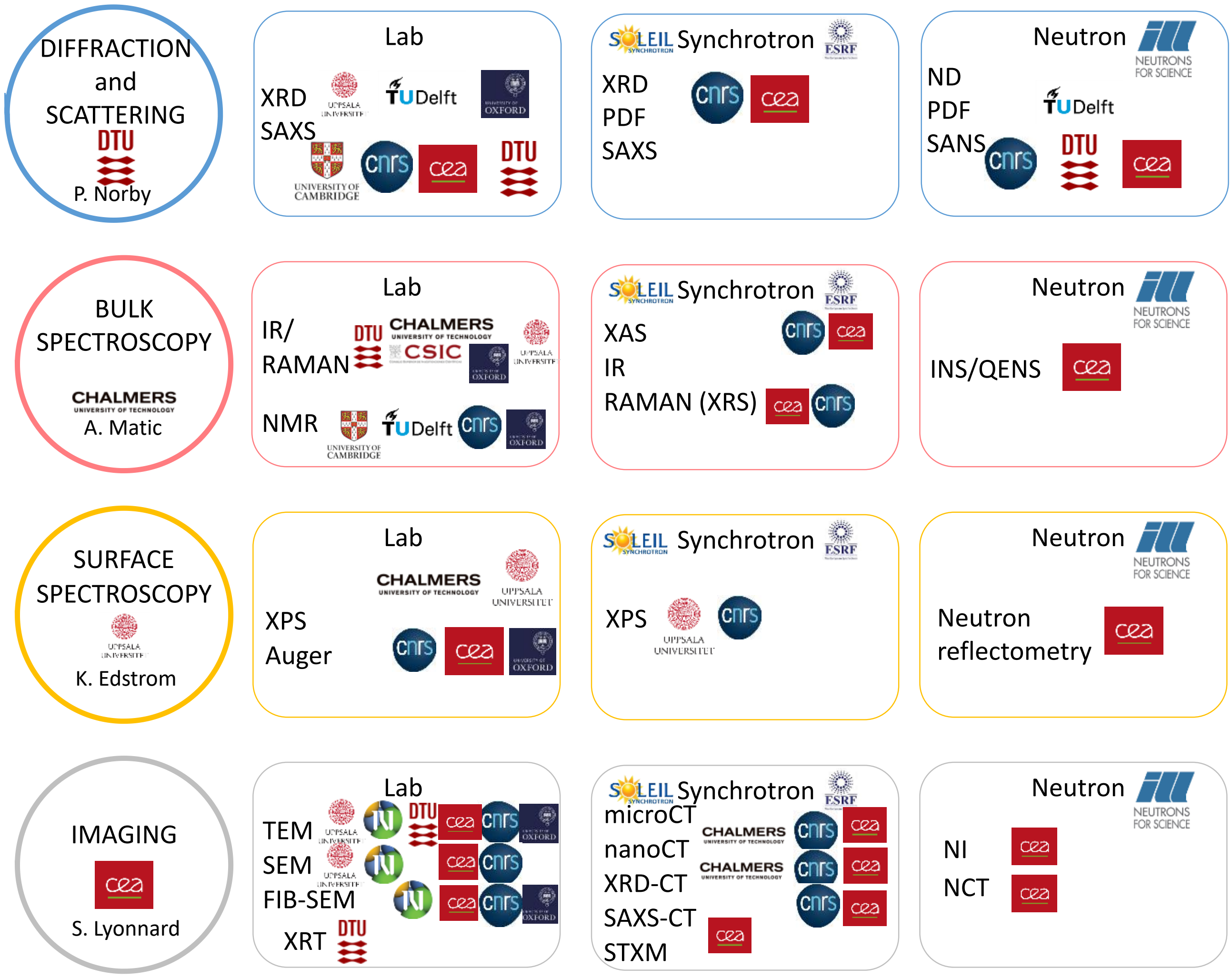

Figure 7: Cluster organization within WP5

2.3. State-of-the-art experimental matrix

The state-of-the-art experimental matrix (hereby matrix) represents an exhaustive report of observables versus techniques available for battery characterization from the raw materials to the complete functioning device. The aim is to collect in the matrix all the information required for establishing the workflow for the case study. In the following the structure and organization of the matrix are explained before presentation of the current iteration of the matrix itself and identified notes for further development throughout the duration of the project.

The matrix takes the form of four tables, one for each WP5 cluster. The four tables loosely correlate to the nature of the observables from the associated techniques.

◊ **Diffraction and scattering** cluster: bulk information on structure from the local and average scales (Å, e.g. Bragg diffraction and PDF) , to the nanostructure (1-100 nm, e.g. SAXS/SANS)

◊ **Imaging** cluster: Two- and three- dimensional distribution of elements with variable resolution (nm to µm) and contrast (e.g. x-ray/neutron/electron absorption contrast or scattering contrast such as in XRD-CT or XPEEM)

◊ **Bulk spectroscopy** cluster: bulk information on the chemical (e.g. Raman) and electronic (e.g. XAS) structure of materials and interfaces.

◊ **Surface spectroscopy** cluster: information on the chemical and electronic information on nm-thick surfaces and interfaces

The division of techniques is not absolute or strict but rather functional and renders the matrix more readable and usable. Multiple techniques are essentially at the interface between the above definitions, e.g. XRD-CT is equally a diffraction and imaging technique. In such cases the assignment to one or the other cluster is arbitrary. It is understood that a successful experimental plan will combine techniques from each cluster/table to achieve a thorough characterization of the case-study chemistry.

Each of the four tables is further structured based on the extend of practical applicability of the techniques to the battery systems under study. Three categories have been introduced depending on the format of the probed sample as follows:

◊ **Commercial**, denotes techniques that can be applied to probe battery cells operando in commercial formats, namely cylindrical, pouch and/or coin cells.

◊ **Realistic**, denotes techniques that can be applied operando to academic cells approximating real batteries and adapted to the specific technique. These generally involve deviations from the industrial format and loadings but approximate well the electrochemical response of real (commercial) systems.

◊ **Model**, denotes techniques that can only be applied ex-situ or on especially prepared systems (e.g. thin films, excavated interfaces). These typically involve significant deviations from realistic geometry and/or electrochemical response.

The classification is evident on the first column of each table and color-coded: red for commercial, orange for realistic and blue for model. This classification correlates with the “maturity” and “fidelity” of each technique and leads naturally to the identification of areas to be developed within the framework of BIG-MAP and beyond (e.g. development of operando XPS capabilities).

Each column in the matrix aims to quantify key aspects associated with each technique entry as follows:

◊ ‘probed area (resolution)’ and ‘penetration depth (resolution)’ columns: Here the geometric dimensions of the interaction volume of the sample from which the observables can be extracted are quantified. The resolution is also included for techniques that can be targeted to different areas and/or depths in the sample; typically associated with imaging or depth-profiling capabilities. These values are meant to be indicative of the technique and not necessarily characteristic of the specific instruments available to BIG-MAP.

◊ ‘detection limit and contrast’ column: the general requirements (chemical/structural etc.) for meaningful signal to be obtained and the main material parameters contributing to the signal, including limiting cases.

◊ ‘observables’ column: The quantities extracted from the data associated with the technique. The format is to be defined and refined in accordance with WP7-WP8.

◊ ‘operando time resolution’ column: The time separating two datasets in an operando cycling experiments.

◊ ‘BIG-MAP availability’: The know-how and equipment within WP5 associated with the technique is listed. This column is further subdivided to ‘Cells’ and ‘Instruments’ and specific effort is made to specifically identify the key individual actors that constitute the point of contact for each technique-partner pairing so as to foster interoperability within WP5 and BIG-MAP in general. (Note: the contact details of individuals have been retracted in the public version of this report).

Diffraction

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables	operando time resolution	refs.	BIG-MAP availability + point of contact		
commercial	X-ray diffraction (XRD)	few mm^2	mm-cm (hard x-ray 60-100 keV)	crystallinity (> 5 nm cryst. size) 1-2 wt.% detection limit atomic number contrast	→Crystal structure →Phase fractions →Crystallite morphology →Strain	ms - min depending on sample, intensity and x-ray energy		**Cells**	**Partner**	**Contact**
								Cylindrical, Pouch	DTU	*[retracted]*
								transmission or reflection geometry coin cells of CR20XX size	ESRF	*[retracted]*
								Modified Swagelok-type cells	ESRF	*[retracted]*
								Instrument	**Partner**	**Contact**
								ID22	ESRF	*[retracted]*
								ID31	ESRF	*[retracted]*
commercial	Neutron Diffraction (ND)	up to 3x5 cm^2	cm	crystallinity (> 5 nm cryst. size) 1-2 wt.% detection limit neutron scattering contrast	→Crystal structure →Phase fractions →Crystallite morphology →Strain	min (standard) ms (stroboscopic)	[1]–[5]	**Cells**	**Partner**	**Contact**
								Cylindrical / Operando	CEA	*[retracted]*
								ILL operando cells	ILL	*[retracted]*
								Instrument	**Partner**	**Contact**
								D2B	ILL	*[retracted]*
								D20	ILL	*[retracted]*
								PEARL	TUD	*[retracted]*
realistic	Laboratory X-ray Diffraction (lab XRD)	1-50 mm^2	Cu (8 keV) ~ 100 µm Mo (17.45 keV) ~1mm	crystallinity (> 5 nm cryst. size) 1-2 wt.% detection limit atomic number contrast	→Crystal structure →Phase fractions →Crystallite morphology →Strain	5-30 mins	[6]–[9]	**Cells**	**Partner**	**Contact**
								Kapton-window dome, (with gas connections) (reflection)	TUD	*[retracted]*
								Be windows, coin cells like	CEA	*[retracted]*
								ELCELL (reflection)	DTU	*[retracted]*
								AMPIX (transmission)	DTU	*[retracted]*
								Glassy carbon window (reflection)	CNRS	*[retracted]*
								Kapton & glassy carbon windows (transmission)	CNRS	*[retracted]*
								Be-window (reflection)	CNRS (LRCS)	*[retracted]*
								Be-window (transmission)	CNRS (LRCS)	*[retracted]*
								Pouch (transmission)	UU	*[retracted]*
								Instrument	**Partner**	**Contact**
								Cu/Co/Mo (Ag in dev.)	TUD	*[retracted]*
								Cu & Mo	CEA	*[retracted]*

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables	operando time resolution	refs.	BIG-MAP availability + point of contact		
								Cu	DTU	*[retracted]*
								Cu/Co/Mo	CNRS	*[retracted]*
								Cu	UU	*[retracted]*
								Cu	UCAM	*[retracted]*
								Cu	UOXF	*[retracted]*
realistic	Synchrotron X-ray diffraction (synchrotron XRD)	few mm^2	Medium to hard radiation (15-100 keV) mm-cm	crystallinity (> 5 nm cryst. size) 1-2 wt.% detection limit atomic number contrast	→Crystal structure →Phase fractions →Crystallite morphology →Strain	$1\ s < \Delta t < 5$ min	[6], [7]	**Cells**	**Partner**	**Contact**
								Be-window (transmission)	Soleil	*[retracted]*
								Kapton-window (transmission)	Soleil	*[retracted]*
								Kapton & glassy carbon windows (transmission)	CNRS	*[retracted]*
								6-position AMPIX cells	DTU	*[retracted]*
								AMPIX-style cell with glassy carbon windows (transmission)	UU	*[retracted]*
								Pouch cell clamped with glassy carbon windows (transmission)	UU	*[retracted]*
								Kapton-window (with gas connections) (transmission)	TUD	*[retracted]*
								Microbeam cell	CEA	*[retracted]*
								transmission or reflection geometry coin cells of CR20XX size	ESRF	*[retracted]*
								Modified Swagelok-type cells	ESRF	*[retracted]*
								Instrument	**Partner**	**Contact**
								BM32	ESRF	*[retracted]*
								ID22	ESRF	*[retracted]*
								ID31	ESRF	*[retracted]*
								ID11	ESRF	*[retracted]*
								ID15A	ESRF	*[retracted]*
								DanMAX (MAX IV)	DTU	*[retracted]*
								CRISTAL	Soleil	*[retracted]*
								SWING	Soleil	*[retracted]*
realistic	laboratory Small Angle X-ray Scattering (SAXS)	< 1 mm^2	micrometers	0.1 wt.% detection limit atomic weight contrast	→Morphology (1-100nm) →Pore Structure →Surface area	Minutes		**Cells**	**Partner**	**Contact**
								Capillary	DTU	*[retracted]*
								Kapton-window transmission	CTH	*[retracted]*
								Ex situ cells	CEA	*[retracted]*
								Instrument	**Partner**	**Contact**
								Mat:Nordic SAXS/WAXS/GISAXS	CTH	*[retracted]*

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables	operando time resolution	refs.	BIG-MAP availability + point of contact		
								Cu-anode ; SAXS/GISAXS Xenocs, SAXS/WAXS	CEA DTU	*[retracted]* *[retracted]*
realistic	Synchrotron (ultra-) small angle x-ray scattering (SAXS, U-SAXS)	1cm x 1cm	Full cell (Bulk)	0.1 wt.% detection limit atomic weight contrast	→ Short range order (10 nm to few µm) →Pore Structure →Surface area →rheological properties (rheo-SAXS)	> 1 s	[10], [11]	**Cells** Pouch cells and sample holder for SAXS/WAXS **Instrument** SWING (SAXS and rheo-SAXS) BM02 ID02	**Partner** CEA **Partner** SOLEIL ESRF ESRF	**Contact** *[retracted]* **Contact** *[retracted]* *[retracted]* *[retracted]*
realistic	Synchrotron microbeam SAXS/WAXS	1 cm^2 (400 nm)	Electrode surface	atomic weight contrast	→Morphology (1-100nm) →Pore Structure →Surface area	< 1 hr	[12]	**Cells** Miniaturized cell **Instrument** ID13	**Partner** CEA **Partner** ESRF	**Contact** *[retracted]* **Contact** *[retracted]*
realistic	Neutron Diffraction (ND)	up to 3x5 cm^2 (1 mm)	cm	crystallinity (> 5 nm cryst. size) 1-2 wt.% detection limit neutron scattering contrast	→Crystal structure →Phase fractions →Crystallite morphology →Strain (3D mapping)	1 min < Δt < 30 min	[13]	**Cells** Cylindrical (Al) ILL operando cells (TiZr) **Instrument** D2B D20 SALSA PEARL	**Partner** CEA ILL **Partner** ILL ILL ILL TUD	**Contact** *[retracted]* *[retracted]* **Contact** *[retracted]* *[retracted]* *[retracted]* *[retracted]*
realistic	Small Angle Neutron Scattering (SANS)	55 x 40 mm (10 to 300 mm^2)	Full cell (bulk)	neutron scattering contrast	→ Morphology (1-100nm) → Size & shapes of nanoparticles → volume expansion → interface roughness	2 min (regular beam) 15 min (pencil beam)		**Cells** Ti-cell w/o separator **Instrument** D22 (Simultaneous SANS/SAXS) SANS-2	**Partner** CEA **Partner** ILL TUD	**Contact** *[retracted]* **Contact** *[retracted]* *[retracted]*
realistic	2D SANS	1mm^2	10-20 µm (pencil beam)	neutron scattering contrast	→Morphology (1-100nm)	Few minutes		**Cells** Ti-PEEK based 2D cell (miniaturized, pencil beam) **Instrument**	**Partner** CEA **Partner**	**Contact** *[retracted]* **Contact**

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables	operando time resolution	refs.	BIG-MAP availability + point of contact		
								[D22](#)	ILL	*[retracted]*
Model	Nano-diffraction	1cm^2 (200 nm)	Electrode surface	crystallinity atomic weight contrast	→Crystal structure →Strain	< 1 hr	[14]	**Cells**	**Partner**	**Contact**
								Kapton-window (with gas connections) (transmission)	TUD	*[retracted]*
								a reflection-geometry windowed coin cell holder suitable for XRD/XRF type mapping	ESRF	*[retracted]*
								Instrument	**Partner**	**Contact**
								[ID01](#)	ESRF	*[retracted]*
								[ID11](#)	ESRF	*[retracted]*

Imaging

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables	operando time resolution	refs.	BIG-MAP availability + point of contact		
commercial	X-ray micro-tomography	> 200µm x 200µm (res: < 1 µm)	cm (full cell) (res: < 50 nm)	atomic weight contrast Phase contrast boundary enhancement	Morphological information (H), volume variations (H), pre-existing defects and after ageing (H) Failure mechanisms (H)	µs to s		**Cells**	**Partner**	**Contact**
								Cylindrical cells	CEA	*[retracted]*
								A cell for imaging battery explosion	ESRF	*[retracted]*
								Instrument	**Partner**	**Contact**
								ID19	ESRF	*[retracted]*
								BM05	ESRF	*[retracted]*
								PSICHé	SOLEIL	*[retracted]*
commercial	X-ray diffraction computed tomography (XRD-CT)	1cm x 1cm (300 nm – 1 µm)	cm (full cell)	crystallinity crystal structure contrast	Phase fractions (SOC), Unit cell lattice parameters (H), atomic site parameters (M), atomic thermal parameters (L) lattice strain (L) and size (L) Structural (H), chemical(H) and morphological (H) information	> 10ms	[15], [16]	**Cells**	**Partner**	**Contact**
								Instrument	**Partner**	**Contact**
								ID15A	ESRF	*[retracted]*
								ID06	ESRF	*[retracted]*
								ID11	ESRF	*[retracted]*
commerci al	X-ray Total scattering and Pair Distribution Function-computed tomography (PDF-CT)	1cm x 1cm (res: < 1 µm)	cm (full cell)	crystal- and local structure contrast	Structural (H), chemical(H) and morphological (H) information	> 10 ms	[16]–[18]	**Cells**	**Partner**	**Contact**
								Instrument	**Partner**	**Contact**
								ID15A	ESRF	*[retracted]*
commercial	Neutron imaging	> 4 µm x 4 µm	cm (full cell)	neutron absorption contrast can distinguish isotopes (e.g. ^{6}Li, ^{7}Li)	Lithium distribution & gradients (L), volume variations (L), defects after ageing	ms - s		**Cells**	**Partner**	**Contact**
								Coin cells (small sized)	CEA	*[retracted]*
								operando and ex-situ ILL cells	ILL	*[retracted]*
								Instrument	**Partner**	**Contact**
								D50-NeXT (coupled with simultaneous XCT)	ILL	*[retracted]*
commercial	Neutron tomography	>4 microns	cm (full cell)	neutron absorption contrast can distinguish isotopes (e.g. ^{6}Li, ^{7}Li)	Lithium distribution & gradients (M), volume variations (M), defects after ageing	s to hr		**Cells**	**Partner**	**Contact**
								Coin cells (small sized)	CEA	*[retracted]*
								operando and ex-situ ILL cells	ILL	*[retracted]*
								Instrument	**Partner**	**Contact**

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables	operando time resolution	refs.	BIG-MAP availability + point of contact		
								D50-NeXT (coupled with simultaneous XCT)	ILL	*[retracted]*
realistic	Laboratory X-ray Tomography (lab XRT)			atomic weight contrast				**Cells**	**Partner**	**Contact**
								Instrument	**Partner**	**Contact**
								Nikon XT	DTU	*[retracted]*
								coupled to neutron tomography	ILL (D50)	*[retracted]*
realistic	X-ray micro tomography (µXRT)	100x100 µm^2 (1µm)	cm (full cell)	Absorption contrast phase contrast typically a few percent in density	Morphology of metal anode or composite anode	s to min	[19]–[21]	**Cells**	**Partner**	**Contact**
								operando Swagelok cell	CTH	*[retracted]*
								operando Swagelok cell	CEA	*[retracted]*
								Instrument	**Partner**	**Contact**
								ID19	ESRF	*[retracted]*
								BM05	ESRF	*[retracted]*
								PSICHé, coupled to XRD-CT	SOLEIL	*[retracted]*
								ANATOMIX	SOLEIL	*[retracted]*
realistic	X-ray nano-tomography (nXRT)	50µm x 50µm (20-80 nm)	Full cell (Bulk)	Phase contrast boundary enhancement	Morphological information (H), volume variations (H), pre-existing defects and after ageing (H)	10 min to hours		**Cells**	**Partner**	**Contact**
								operando microbattery cell (in dev.)	CEA	*[retracted]*
								operando cell (in dev.)	CTH	*[retracted]*
								Instrument	**Partner**	**Contact**
								ID16B	ESRF	*[retracted]*
								ID16A	ESRF	*[retracted]*
								ANATOMIX	SOLEIL	*[retracted]*
realistic	Neutron imaging	170x170 mm^2 (res: >4 µm)	Full cell quantify?	neutron absorption contrast can distinguish isotopes (e.g. ^{6}Li, ^{7}Li)	Lithium distribution & gradients (M), volume variations (M)	4 hr		**Cells**	**Partner**	**Contact**
								Operando cell compatible for x-rays (in dev.)	CEA	*[retracted]*
								Instrument	**Partner**	**Contact**
								D50-NeXT (coupled with simultaneous XCT)	ILL	*[retracted]*
								FISH	TUD	*[retracted]*
re	Nano x-ray fluorescence (n-XRF)	50 µm x 50 µm	Full cell (Bulk)	>0.01ppm	Elemental analysis (H)	> 1h		**Cells**	**Partner**	**Contact**

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables	operando time resolution	refs.	BIG-MAP availability + point of contact		
		(res: 20 nm vox: >10 nm)						**Instrument**	**Partner**	**Contact**
								ID16A	ESRF	*[retracted]*
								ID16B	ESRF	*[retracted]*
								ID21	ESRF	*[retracted]*
realistic	X-ray diffraction or pair distribution function computed tomography (XRD-CT / PDF-CT)	5 mm in diameter (res: 1 µm voxel: >300 nm)	full electrode layer (3 mm in diameter) (transmission parallel to stack of battery components)	crystal- and local structure contrast	spatially resolved structural (H) (chemical (H)) and morphological (H) information (diffractogram for each 3D pixel) volume selective analysis of specific battery components Phase fractions and distribution/SoC in 3D Unit cell lattice parameters (H), atomic site parameters (M), atomic thermal parameters (L) lattice strain (L) and size (L)	min	[16], [17]	**Cells**	**Partner**	**Contact**
								cylindrical cell	CNRS	*[retracted]*
								miniaturized version in diameter	CEA	*[retracted]*
								Swage-lock type cell	CEA	*[retracted]*
								Instrument	**Partner**	**Contact**
								PSICHé, coupled to XRT	Soleil	*[retracted]*
								ID13	ESRF	*[retracted]*
								ID15A	ESRF	*[retracted]*
realistic	Small- or wide angle x-ray scattering computed tomography (SAXS-CT / WAXS-CT)	3mm in diameter	5-10 microns^3 voxels		3D repartition of phases; lithiation mapping	~10 mins per slice		**Cells**	**Partner**	**Contact**
								Swagelok type	CEA	*[retracted]*
								in development	CTH	*[retracted]*
								Instrument	**Partner**	**Contact**
								ID31	ESRF	*[retracted]*
realistic	Neutron Depth Profiling (NDP)	1 cm^2	~40 µm (100 nm)	~1 molar Li density	Li-density (M) as a function of depth	min	[22]	**Cells**	**Partner**	**Contact**
								Pouch type cell, using the current collector as window	TUD	*[retracted]*
								Instrument	**Partner**	**Contact**
								NDP	TUD	*[retracted]*
realistic	Ptychography/ Ptychtomography	50um x 50um (res 10nm) voxel >5nm	Full cell (Bulk)	Phase contrast boundary enhancement	Morphological information (H), volume variations (H), pre-existing defects and after ageing (H)	> 10 min	[23]	**Cells**	**Partner**	**Contact**
								Instrument	**Partner**	**Contact**
								ID16A	ESRF	*[retracted]*
								SWING	SOLEIL	*[retracted]*
								HERMES	Soleil	*[retracted]*
	Bragg Coherent diffraction imaging				phase fraction and defects imaging within grains			**Cells**	**Partner**	**Contact**
								under development	CNRS	*[retracted]*

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables	operando time resolution	refs.	BIG-MAP availability + point of contact		
	(BraggCDI)							a reflection-geometry windowed coin cell holder suitable for XRD/XRF type mapping	ESRF	*[retracted]*
								Instrument	**Partner**	**Contact**
								ID01	ESRF	*[retracted]*
								ID13	ESRF	*[retracted]*
								ID10	ESRF	*[retracted]*
								CRISTAL	SOLEIL	*[retracted]*
model	Transmission Electron Microscopy	probe size <0.1nm	transmission, up to 150nm HRTEM: point-to point resolution 0.08 nm; EELS: 0.7-0.4eV energy resolution	crystallinity down to atomic clusters; Heavy and light elements; Li >10 at%; diffraction contrast; thickness contrast; atomic weight contrast.	atomic structure H, crystal structure M, chemical composition M (0.3 at% by EDX), morphological H, texture, chemical gradients, electronic structure (valence) M			**Cells**	**Partner**	**Contact**
								ex-situ / post-mortem	CEA	*[retracted]*
								in dev.	DTU	*[retracted]*
								ex-situ/ post-mortem	NIC	*[retracted]*
								Instrument	**Partner**	**Contact**
								TITAN	CEA	*[retracted]*
								JEM200CF 80-200 kV with QuantumGIF and 100mm^2 Centurio EDX	NIC	*[retracted]*
								(HRTEM)	DTU	*[retracted]*
model	Scanning Electron Microscopy	probe size ca. 1 nm, probed area from nm up to hundreds µm	resolution SEM ca. 1.5 nm, FIB ca. 1 nm; penetration depth SEM 200 nm to 2 µm, FIB cut up to 20 µm	detection limit EDX 0.3 at%; crystallinity down to nm; atomic weight contrast	surface morphology H; chemical composition M (L for thin films);		[24]	**Cells**	**Partner**	**Contact**
								ex-situ / post-mortem	CEA	*[retracted]*
								ex-situ / post-mortem	NIC	*[retracted]*
								Instrument	**Partner**	**Contact**
								Zeiss Merlin Analytical	UOXF	*[retracted]*
								ex-situ / post-mortem (including FIB-SEM)	CEA	*[retracted]*
								Zeiss Supra 35 VP with FIB: FEI HeliosNanolab 650	NIC	*[retracted]*
model	x-ray Laminography					hr		**Cells**	**Partner**	**Contact**
								pouch cell (in dev.)	CTH	*[retracted]*
								Instrument	**Partner**	**Contact**
								ID16b	ESRF	*[retracted]*
								ID19	ESRF	*[retracted]*
model	Neutron Laminography						[25], [26]	**Cells**	**Partner**	**Contact**
								Instrument	**Partner**	**Contact**
								D50-NeXT	ILL	*[retracted]*
			100s nm					**Cells**	**Partner**	**Contact**

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables	operando time resolution	refs.	BIG-MAP availability + point of contact		
	Scanning Transmission X-ray Microscopy (STXM)							**Instrument** HERMES	**Partner** SOLEIL	**Contact** *[retracted]*
model	X-ray Photoemission Electron Microscopy (XPEEM)	25 nm	10 nm (0.1)	chemical and electronic		100ms	[23]	**Cells** **Instrument** HERMES	**Partner** **Partner** SOLEIL	**Contact** **Contact** *[retracted]*
model	Micro-XAS	(500 nm)	<10-500µm		SEI chemical imaging Electronic imaging Spectroscopy	hr	[27]	**Cells** Omicron type sample holder **Instrument** ANTARES	**Partner** SOLEIL **Partner** SOLEIL	**Contact** **Contact** *[retracted]*
model	nano-XPS	(400 nm)	< 10 A		SEI chemical imaging	hr	[27]	**Cells** Omicron type sample holder **Instrument** ANTARES	**Partner** SOLEIL **Partner** SOLEIL	**Contact** **Contact** *[retracted]*

Bulk Spectroscopy

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables + resolution or quality (High-Medium-Low)	operando time resolution	refs.	BIG-MAP availability + point of contact		
realistic	Solid State Nuclear Magnetic Resonance Spectroscopy (NMR)	Full cell	Metals have a specific maximum skin depth that is probed (around 10 µm depending on the field)	Depends on element and chemical environment Convenient nuclei: ^{1}H, ^{2}H, ^{7}Li, ^{19}F, ^{23}Na, ^{27}Al, ^{31}P, ^{51}V Operando: Static experiments, thus lower resolution as compared to MAS possibility to differentiate the different component based on their relaxation time	Element specific chemical environment, Ion mobility (under specific conditions)	min	[28], [29]	**Cells**	**Partner**	**Contact**
								Capsule cells	TUD	*[retracted]*
								Pouch cells	TUD	*[retracted]*
								Capsule cells	UCAM	*[retracted]*
								Pouch cells	UCAM	*[retracted]*
								Instrument	**Partner**	**Contact**
								500 MHz	TUD	*[retracted]*
								200 MHz	UCAM	*[retracted]*
								400 MHz	UCAM	*[retracted]*
								500 MHz	UCAM	*[retracted]*
								700 MHz	UCAM	*[retracted]*
realistic	Raman Spectroscopy	(50x50 µm) res: > 1µm	1-5 µm	Depends on the sample	Electrolyte composition, gradients, carbon structure/ intercalation processes (anodes)	min	[30]	**Cells**	**Partner**	**Contact**
								Operando Raman cell	CTH	*[retracted]*
								EL-cell	DTU	*[retracted]*
								Instrument	**Partner**	**Contact**
								HORIBA LabRam HR Evolution	CTH	*[retracted]*
									DTU	*[retracted]*
realistic	X-ray Raman Spectroscopy (XRS)	1cm x 1cm res: 20um	Thin cell 10-100 µm	Possibility to detect light elements (Lithium)	Chemical state of elemental constituents (H)	> 4 hr	[31]	**Cells**	**Partner**	**Contact**
								Be window (transmission)	SOLEIL	*[retracted]*
								Ex situ cell	CEA	*[retracted]*
								Instrument	**Partner**	**Contact**
								ID20	ESRF	*[retracted]*
								GALAXIES	SOLEIL	*[retracted]*
realistic	X-ray Absorption Spectroscopy (XAS)	2.5µm x 2.5 µm	Depends of transmission/Fluoresence Bulk 0.8 to 8 keV (LUCIA)		Oxidation state 1st and 2nd coordination shell information	2 min < Δt < 30 min	[32], [33]	**Cells**	**Partner**	**Contact**
								Operando cell	CEA	*[retracted]*
								Instrument	**Partner**	**Contact**
								LUCIA	SOLEIL	*[retracted]*
r		400 µm x 1mm	bulk,	> 4% of element		250ms		**Cells**	**Partner**	**Contact**

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables + resolution or quality (High-Medium-Low)	operando time resolution	refs.	BIG-MAP availability + point of contact		
	Hard X-ray Absorption Spectroscopy (hard XAS)		6.5-43 keV		Mn and Ni redox processes		[6], [7]	Kapton & glassy carbon windows (transmission)	CNRS	*[retracted]*
								Be windowl (transmission)	SOLEIL	*[retracted]*
								Instrument	**Partner**	**Contact**
								ROCK	SOLEIL	*[retracted]*
realistic	Resonant Inelastic X-ray Scattering (RIXS)	10 - 100 µm	10 µm	Sulfur, 3d, metals (Co,..) K-edges	Oxidation state Chemical environment	min to hr	[34]	**Cells**	**Partner**	**Contact**
								Be window (transmission)	SOLEIL	*[retracted]*
								Instrument	**Partner**	**Contact**
								GALAXIES	SOLEIL	*[retracted]*
model	Neutron Spectroscopy	beam area at sample 30 x 30 mm^2	cm full cell	Light elements eg Li, H Hyperfine splitting of some elements (e.g. Co, Nd, Ho, V,...)	Mechanisms of local diffusion (H)	min to hr		**Cells**	**Partner**	**Contact**
								ex-situ	CTH	*[retracted]*
								Ex-situ	CEA	*[retracted]*
								Instrument	**Partner**	**Contact**
								IN16B	ILL	*[retracted]*
model	Ex-situ Fourier-transform Infrared spectroscopy (FTIR)	SEI (DRIFTS and ATR) Electrolyte (transmission)	~1 μm	Depends on the sample	Interphase composition (M), Electrolyte spectra	minutes	[35]	**Cells**	**Partner**	**Contact**
								ATR	CTH	*[retracted]*
								Transmission	CTH	*[retracted]*
								Diffuse refl.	CTH	*[retracted]*
								operando in dev.	CTH	*[retracted]*
								Diffuse reflectance (HARRIK)	CSIC	*[retracted]*
								Instrument	**Partner**	**Contact**
								Bruker Alpha	CTH	*[retracted]*
								Vertex 70 FT-IR Spectrometer	CSIC	*[retracted]*

Surface Spectroscopy

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables + resolution or quality (High-Medium-Low)	operando time resolution	refs.	BIG-MAP availability + point of contact		
model	X-ray photoelectron Spectroscopy (XPS)	UU (Kratos): 700 x 300 µm; 110 µm dia.; 55 µm dia.; 27 µm dia.; 15 µm dia. res: ~2 µm UU (Kratos): XPS Imaging. 400 x 400 µm, 200 x 200 µm and 80 x 80 µm fields of view. Stitchable (unlimited) to form larger images. res. < 1 µm	UU (Kratos): Al Kα: 6-10 nm Ag Lα: 12-20 nm res: nm 'Unlimited' with Gas Cluster Ion Sputtering (GCIS) - depth profiling. (res - nm)	+/- 5 % typical for XPS	SEI/CEI chemistry (oxidation state) (H), overall composition (H) XPS Imaging: chemical homogeneity of surface	in dev.	[36]	**Cells**	**Partner**	**Contact**
								Kratos operando (electrochemistry, temperature, in testing)	UU	*[retracted]*
								omicron sample holder	SOLEIL	*[retracted]*
								Ulvac-PHI/Omicron/operando	CEA	*[retracted]*
								Instrument	**Partner**	**Contact**
								Quantes Cr Kα	CEA	*[retracted]*
								Versa-Probe II Al Kα	CEA	*[retracted]*
								Kratos AXIS Supra+ (Al Kα, Ag Lα)	UU	*[retracted]*
								PHI 5500 (Al Kα)	UU	*[retracted]*
								in dev.	NIC	*[retracted]*
								TEMPO	SOLEIL	*[retracted]*
								BM25-SPLINE	ESRF	*[retracted]*
model	Nano-X-ray photoelectron Spectroscopy (nano -XPS) Nano-UPS and Angle resolved Photoemission (nano-ARPES)	50nm	Up to 4 nm	XPS and ARPES Imaging: chemical and electronic homogeneity of surface	+/- 5 % typical for XPS			**Cells**	**Partner**	**Contact**
								Omicron sample holder	SOLEIL	*[retracted]*
								Instrument	**Partner**	**Contact**
								ANTARES	SOLEIL	*[retracted]*
model	(near-) ambient pressure XPS ((N)AP-XPS)	100 x 100 µm	~5 nm at 1.5 keV	1-10% in composition (depending on cross section)	SEI chemical composition In-situ / operando	in situ, hours		**Cells**	**Partner**	**Contact**
								Under development	CNRS	*[retracted]*
								Instrument	**Partner**	**Contact**
								TEMPO	SOLEIL	*[retracted]*
								HIPPIE (MAXIV)	UU	*[retracted]*
								SPECIES (MAXIV)	UU	*[retracted]*

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables + resolution or quality (High-Medium-Low)	operando time resolution	refs.	BIG-MAP availability + point of contact		
model	hard x-ray photoelectron spectroscopy (HAXPES)	50 µm	~30 nm at 10 keV resolution < 1 nm	Similar to XPS	Local chemical, electronic properties ; SEI chemical composition ; in-situ / operando	in situ, minutes	[37]	**Cells** Under development **Instrument** GALAXIES In-lab Quantes Cr Kα	**Partner** CNRS **Partner** SOLEIL CEA	**Contact** *[retracted]* **Contact** *[retracted]* *[retracted]*
model	Reflection electron energy loss spectroscopy (REELS)	unknown (probably 10's-100's of microns dia.)	15 nm	unknown	-band gap (H), Hydrogen content (H), conjugation/aromaticity in organics (H), sp2/sp3 hybridisation in carbon (H)			**Cells** Kratos operando (electrochemistry, temperature, in testing) **Instrument** Kratos AXIS Supra+	**Partner** UU **Partner** UU	**Contact** *[retracted]* **Contact** *[retracted]*
	Ion Scattering Spectroscopy (ISS)	250 µm - 700 µm dia. spot size	surface atomic later	likely +/- 5 %	chemical composition of surface atomic layer (complementary to ToFSIMS) (H)			**Cells** Kratos operando (electrochemistry, temperature, in testing) **Instrument** Kratos AXIS Supra+	**Partner** UU **Partner** UU	**Contact** *[retracted]* **Contact** *[retracted]*
model	near edge / extended x-ray absorption fine structure (NEXAFS/EXAFS)	50 x 50 µm res 10um pixel >3um	10nm Electrode surface	>0.01ppm 1% in composition	Short range order information (H), coordination number (H), chemical state of elemental constituents (H) Chemical and electronic structure	2-10mn		**Cells** omicron sample holder **Instrument** TEMPO BM23 ID24 ID26	**Partner** SOLEIL **Partner** SOLEIL ESRF ESRF ESRF	**Contact** **Contact** *[retracted]* *[retracted]* *[retracted]* *[retracted]*
model	soft x-ray absorption spectroscopy (XAS)	100nm	10nm	0.01ppm	Chemical and electronic structure H	100ms – 1mn	[23]	**Cells** Operando electrocell **Instrument** HERMES ANTARES	**Partner** SOLEIL **Partner** SOLEIL SOLEIL	**Contact** *[retracted]* **Contact** *[retracted]* *[retracted]*
model	surface-enhanced Raman spectroscopy (SERS)	res 1 µm	nm	Single molecule detection is possible (in favourable conditions)	Processes at electrode interface	Minutes		**Cells** Model cell for operando SERS **Instrument** HORIBA LabRam HR Evolution	**Partner** CTH **Partner** CTH	**Contact** *[retracted]* **Contact** *[retracted]*
model	soft x-ray Resonant Inelastic X-ray Scattering (RIXS)	10 - 100 µm	< 1 µm					**Cells** **Instrument** SEXTANTS	**Partner** **Partner** SOLEIL	**Contact** **Contact** *[retracted]*

	technique	probed area (resolution)	penetration depth (resolution)	detection limit and contrast	observables + resolution or quality (High-Medium-Low)	operando time resolution	refs.	BIG-MAP availability + point of contact		
model	nano-Auger (AES)	(res:20nm)	<5nm					**Cells**	**Partner**	**Contact**
								ULVAC-PHI/Omicron	CEA	*[retracted]*
								Instrument	**Partner**	**Contact**
								Auger PHI 700Xi	CEA	*[retracted]*
model	X-rays Reflectometry (XRR)	1cm x 1cm res 200um pixel >200um	Thin cell Depth resolution ~2 nm	Possibility to identify the electronic density at embedded interfaces	Roughness (H), interconnection (H)and phases segregated at the interface (H) Interphase thickness (M) and composition (L)	> 10 min	[38]	**Cells**	**Partner**	**Contact**
								cell available, w/o separator (si wafer)	CEA	*[retracted]*
								Instrument	**Partner**	**Contact**
								BM32	ESRF	*[retracted]*
								ID31	ESRF	*[retracted]*
model	Neutron Reflectometry (NR)	2cm x 5cm	~500 nm Depth resolution ~2 nm	1-10% in composition	Li-density (M) as a function of depth (100 nm resolution, max depth 40 µm) atomic and magnetic in-plane structure Kinetics of interface evolution of thin solid films and multilayers	min/hr	[22]	**Cells**	**Partner**	**Contact**
								Instrument	**Partner**	**Contact**
								D17	ILL	*[retracted]*
								ROG	TUD	*[retracted]*

2.4. Cell index

A main objective of WP5 is to harmonize inter- and intra-technique data collection; and a key parameter in that endeavor is the sample environments (cells) utilized for said data collection. This is especially relevant for operando experiments, i.e. when a given technique is combined with simultaneous electrochemical cycling to probe the observables as a function of battery operational parameters. Thus, considerable effort was directed into collecting information on the available cell designs among all partners with the goals of i) harmonization and ii) further development. This was undertaken in parallel to the construction of the matrix and so at present the two (experimental matrix and cell index) are separate entities. Future work will aim at combining them into a single database as outlined in the following section.

In its current iteration the cell index takes the form of a collection of specification sheets, one for each cell, and is appended to the end of the present document (Appendix A).

2.5. Future improvements

Several directions for future improvement have been already identified and are in progress; the main ones are outlined below:

◊ Missing techniques: due to the procedure followed for generating the matrix, the current iteration is biased towards techniques accessible by WP5 partners in terms of instrumentation and expertise. While this covers the majority of the experimental battery characterization space, it is not exhaustive with respect to the characterization techniques that have been applied to batteries in the literature. For example, the following techniques could be included in a future, more exhaustive version of the matrix: optical microscopy, Mössbauer spectroscopy, atomic force microscopy (AFM), secondary-ion mass spectrometry (ToF-SIMS), differential electrochemical mass spectrometry (DEMS), internal sensing of temperature and pressure through optical fibers etc. Such inclusions could also highlight the need for expansion of the consortium in further iterations of the project to include the missing experimental expertise(s).

◊ Evaluation of maturity/fidelity/reactivity: despite attempts to quantify the above parameters for each technique, no adequately objective metric could be identified to quantifiably compare the techniques. Given the sensitivity of the BIG-MAP central objective to said parameters further iterations should aim to address these.

◊ Interactivity/usability: the current iteration of the matrix takes the form of long tables that are not necessarily the most user-friendly/readable despite attempts to the contrary. Further iterations should aim to improve on that aspect by e.g. implementing the matrix into a clickable, interactive database application (app). The cell index and experimental matrix would ideally be integrated in such a database. Interaction with WP9 (Infastructure and Interoperability) could be pursued on that front.

2.6. Matrix references

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3. Workflow

3.1. Selection of Tier 1/Tier 2 techniques

Tier 1 and Tier 2 techniques were defined according to the following considerations:

◊ Tier 1 techniques should provide the key data and parameters needed to obtain a full picture of reaction mechanisms and basic interfacial properties in batteries, allowing the in-depth understanding of battery function and ageing (priority techniques). For instance, XRD, NMR and XPS are workhorse techniques that are widely employed in battery characterization as they give access to crucial parameters as phase identification & transitions, chemical environment and Solid Electrolyte Interphase composition, respectively. Those data are critical inputs to atomic-scale modeling (vibrational spectra, etc.). Similarly, the 3D microstructure of the electrode and its evolution on cycling is key and should be provided early in the project to nourish mesoscale modeling and train advanced AI-based segmentation and statistical analysis.

◊ The order of implementation of Tier 1 and Tier 2 techniques, and their overlap, should allow for a flexible refinement of the experimental program during the project.

◊ Tier 1 plan must account for the maturity of each technique and its availability within the consortium to probe the selected tiered-materials.

◊ Possibilities to perform operando measurements, i.e. availability of existing cells adaptable to the experimental constraints of given techniques is crucial in the early stages of Tier 1 plan.

◊ Possibilities to probe commercial-type or realistic batteries are important to provide data in real or representative cycling conditions.

◊ Fast adaptation to the standards & protocols of BIG-MAP is required to select the first implemented experiments to allow for rapid harmonization of methods and results, as well as efficient transfer to modeling and AI.

◊ Second round techniques, i.e. Tier 2 techniques, are the ones where technical challenges have to be faced, and/or methodologies for data analysis need to be developed or optimized to meet the requirements of the global workflow in the project. For instance, single crystal investigations or tomography at ultimate resolutions require ad-hoc experimental set-ups and conditions, not yet existing or in development.

◊ The Tier 1 and Tier 2 experimental plan must adapt to the sequential delivery of materials. At M6, graphite as anode, LNO as cathode, and standard LP57 electrolyte, has been provided to the partners by the industrial partners and suppliers. Next steps include the delivery of Si-Gr, NMC, new electrolytes with additives and coatings.

The Tier 1 techniques were therefore selected from the experimental matrix on the basis of the above considerations, by identifying the ones that meet most requirements. The tier 1 experimental plan is elaborated in section 3.3.

3.2. Workflow

Having defined the Tier 1 and Tier 2 techniques, the WP5 workflow is presented as a general timeline with the different tasks separated by period of 6 months for reasons of clarity (Figure 8). Obviously, the project is expected to occur through a continuous sequence.

◊ M6 to M12: Both lab-scale and large-scale facilities Tier 1 experiments will start from M6. The first task will be to perform preliminary measurements following the standard operation procedures proposed by WP8 (cycling conditions for *ex situ* samples, sample washing, handling of sample for surface techniques, operando cell assembly…etc). This will come along the development of BIG-MAP cells and sample environments focusing on interoperability between the partners. The conception of these cells will rely on the expertise of the consortium as exemplified by the large number of already available cells (around 40 at the beginning of the project – see appendix). For this challenging task, the availability of a first version of the online notebook will greatly facilitate the communication and the implementation of the standard operation procedures. At M12, the collected data will be shared across the partners in clusters.

◊ M12 to M18: The set of data will be analyzed and compared focusing on the reproducibility and fidelity. Adjustment will be made to obtain a set of reproducible data for all tier 1 techniques by M18, validating the standard operating procedures.

◊ M18 to M24: Moving forward, the reproducible data will be communicated to the other WPs. Interactions with modelling WPs (2 and 3) and material WPs (4 and 6) will lead to the design of a second generation of tier 1 experiments featuring novel materials (coating and electrolyte formulation) and novel measurement conditions driven by modelling. WPs dedicated to the AI (9, 11) will receive the first set of usable data. Finally, WP10, focused on machine learning, will provide useful guidelines to automate analysis, for example for the segmentation of imaging data.
In the meantime, development of tier 2 techniques and the required sample environments will be carried out

◊ M24 to M30: The new set of experiments guided by the modelling and material WPs will be performed with a strong emphasis on integrated sequential analysis of the same sample (or cell) taking advantage from BIG-MAP cells and sample holders. A specific workflow dedicated to the efficiency of sample transfer between the partners will be built and optimized in this period. Tier 2 experiments will start at M24.

◊ M30 to M36: At this stage, the different workflows will be firmly established allowing the project to move to the next steps such as building a demonstrator for high-throughput data acquisition. Also, the close collaboration with modelling and machine learning WPs is will permit to demonstrate a proof of concepts for on-the-fly analysis of the high throughput data acquired.

The experimental plan in section 3.3. elaborates experiments to be done/started in the first period M6-M12.

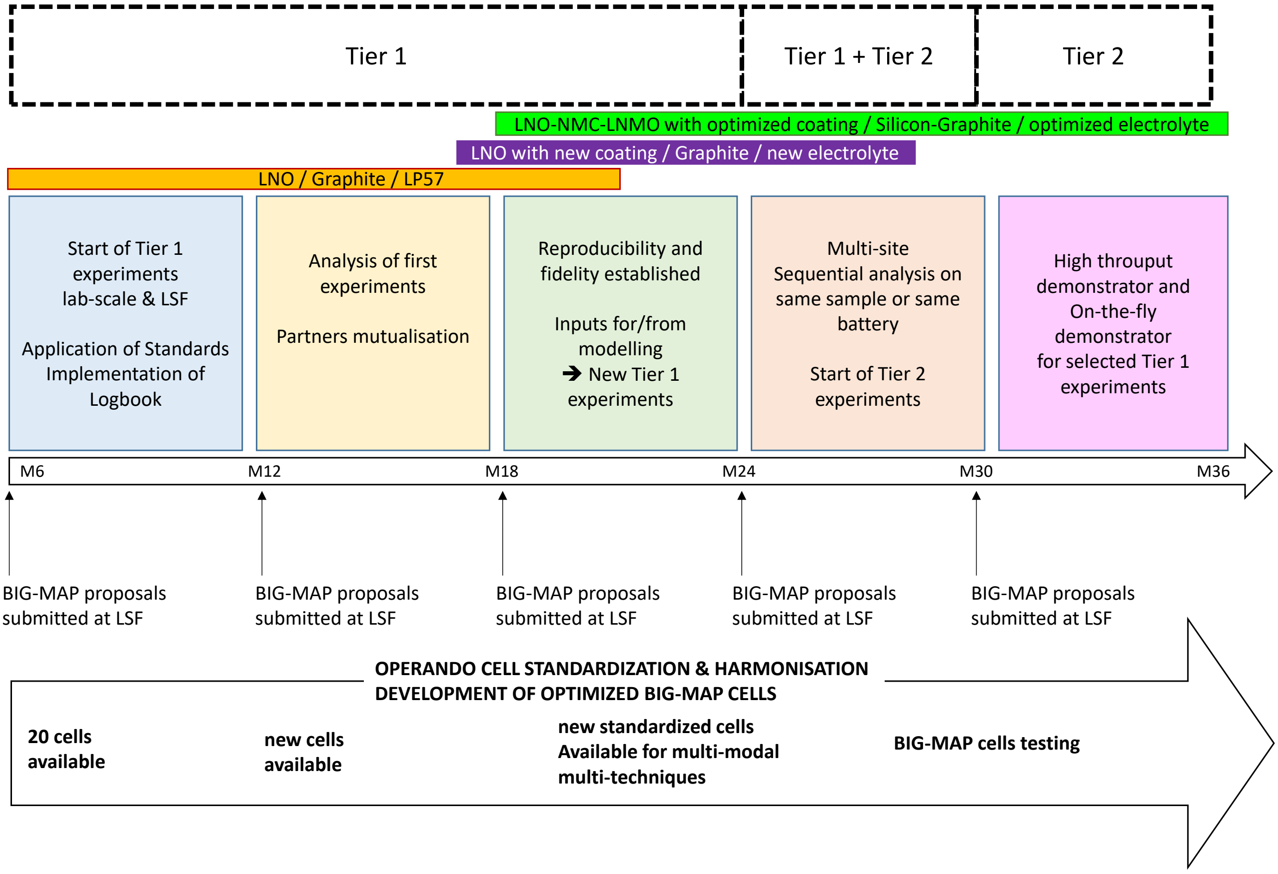


Figure 8: WP5 workflow presenting the timeline of Tier 1 and Tier 2 experiments, the expected material availability, the distribution of the tasks along the project and the planned strategy in terms of operando cell development.

3.3. Tier 1 experimental plan

The Tier 1 experimental plan aims to cover the relevant battery processes, where we distinguish experiments targeting three different themes, (1) (de)lithiation mechanism, the effect of ageing or/and high rate (Table 1), (2) SEI characterization (Table 2) and (3) electrode microstructure, lithiation heterogeneities and single particle dynamics (Table 3). The targeted operando experiments require electrochemical cells (typically experimental technique specific) for which a cell index of available cells has been introduced in section 2.4. The goal is to harmonize the use of operando cells, and to develop improved cell designs within the consortium. To support this development several Tier 1 experiments are proposed, grouped in the fourth experimental theme (Table 4).

Table 1. Tier 1 experiments – (de)lithiation mechanism, the effect of ageing or/and high-rate

(de)lithiation mechanism, the effect of ageing or/and high-rate				
Partner(s)	**Experiment**	**object**	**Observables**	**Time**
UCAM	in-situ NMR	LNO	Ni/Co/X ordering; Li mobility	
UCAM	in-situ NMR	Si	SEI formation – and ageing (stress tests, crossover effects of organics, CO_2, TMs, etc)	
CTH, CSIC	ex-situ Raman and IR	electrolytes and components (salt, solvents, additives), active materials	vibrational spectra	February to May 2021 **M6-M9**
CTH, CSIC	operando Raman	graphite anode	lithiation/delithiation	April to August 2021 **M8-M12**
SOLEIL	operando XAS	Cathode	Redox and local structural changes	July- November 2021 **M11-M15**
CTH, DTU	automated operando data analysis		vibrational spectra	to be published in spring 2021
DTU	Operando synchrotron diffraction	halfcells and fullcells	Dynamics, ageing and degradation and electrode materials	
CEA, DTU	Laboratory operando lab diffraction	LNO	dynamics, aging and degradation of electrode materials, function of rate and T	April-June 2021 **M8-M10**
CEA, ILL	operando neutron diffraction		dynamics, ageing and degradation, electrode phase transformations	Allocated, to be scheduled, May-Oct 2021 (**M8-M14**)
UOXF	OEMS, operando and ex-situ lab diffraction	LNO	dynamics, aging and degradation of electrode materials, function *V*	May-Oct 2021 (**M8-M14**)

Table 2. Tier 1 experiments – SEI characterisation

SEI characterisation				
Partner(s)	**Experiment**	**object**	**Observables**	**Time**
CTH, CSIC, DTU	ex-situ Raman and IR	graphite anode	SEI evolution as a function of cycling	March to June 2021 (**M7-M10**)
UU, CEA, CNRS, SOLEIL	ex-situ HAXPES, XPEEM	(model) graphite electrodes	SEI thickness and composition during lithiation/delithiation	proposal submitted **M6**
CNRS, SOLEIL	Development of in situ / operando XPS, HAXPES	graphite electrodes	SEI thickness and composition during lithiation/delithiation	proposal submitted **M6**
UU, CEA, CNRS SOLEIL, NIC	ex-situ HAXPES, XPS, XPEEM	Ni-rich NMC and LNO	chemical and electronic composition of active material and CEI as a function of rate	proposal submitted **M6**
UU, CEA	in-house XPS, TOF-SIMS, SEM/AFM	post-mortem flat graphite electrodes	complete description of interface	March 2021-February 2021 **M7-M18**
UU, CEA	in-house XPS, TOF-SIMS, SEM/AFM	graphite, NMC, LNO	SEI/CEI as a function of electrolyte e.g. state-of-the-art vs. fluorine-free	March 2021-February 2021 **M7-M18**

Table 3. Tier 1 experiments – Electrode microstructure, lithiation heterogeneities and single particle dynamics

Electrode microstructure, lithiation heterogeneities and single particle dynamics				
Partner(s)	**Experiment**	**object**	**Observables**	**Time**
CEA, NIC	ex-situ FIB-SEM	LNO	electrode microstructure at high resolution	Start March on Pristine, continued on aged until June **M7-M10**
CEA, ILL	operando neutron imaging	LNO (half-cells ; full cells with Gr)	Li concentration gradients, lithiation heterogeneity	May-June 2021 **M9-M10**
TUD	operando Neutron depth profiling	adapted pouch cells	lithium concentration in electrode depth	**M12-M18**
CEA, ESRF	operando SAXS/WAXS microtomography	LNO (half-cells ; full cells with Gr)	electrode microstructure, lithiation heterogeneity, structure	allocated, to be scheduled April-May 2021 **M8-M9**
CTH, DTU	lab SAXS	anode cast electrodes	microstructure	March to May 2021 (**M7-M9**)
CTH	operando lab SAXS	graphite	lithiation/delithiation	April to June 2021 (**M8-M10**)
DTU	operando diffraction (time and spatially resolved)	commercial cells		
DTU	operando diffraction with micrometer resolution	single/few crystals microbattery cells	domain progression, interfaces and lithiation mechanisms	
CEA / ESRF	operando XRD-CT	LNO	Distribution of phases	Proposal on ID15a
UU	combined x-ray diffraction and tomography /ptychography	NA	single particle/crystal non-equillibrium transformations at high rates	

Table 4. Tier 1 experiments – technique and cell development

Technique and cell development				
Partner(s)	**Experiment**	**object**	**Observables**	**Time**
(de)lithiation mechanism, ageing and high-rates studies				
CEA, ILL	development of stroboscopic mode for operando diffraction	commercial cells	high rate structural determination	**M12-M24**
UU	development of operando cell for synchrotron diffraction		high rates (>30C), structural changes w/ sub-second resolution	
CTH	development of operando cell for laminography	NA	high resolution x-ray imaging	Feb. to June 2021 (**M5-M10**)
UU	development of combined neutron diffraction – thermal analysis environment		synthesis of new materials	
UU	development of in-house operando		low rates, in-situ reaction monitoring, accelerated electrode aging	
TUD	neutron diffraction, development and testing of operando cells	NA	validation and comparison of operando cells, proof-of-concept studies	**M12-M18**
TUD	development of in-house (Ag source) transmission operando x-ray diffraction	(commercial) pouch cells	structural evolution, 2D mapping, validation of synchrotron cells	**M12-M18**
CNRS, SOLEIL	development of operando cells for multi-technique characterization with hard X-rays	Realistic and reliable electrocells	structural and redox evolution, oxidation state, local environment	Fixed-term contract or Postdoc to be hired **M9-M27**

SEI/CEI characterisation				
CNRS, SOLEIL	development of electrochemical cell for in-situ surface techniques	model	surfaces and interfaces	PhD hired, **M1-M36**
CNRS, SOLEIL	design of in-situ cell for HAXPES	model	surfaces and interfaces	PhD hired, **M1-M36**
UU, CEA	development of interoperability protocols for surface methods	NA	combination of techniques on the same sample	March 2021-August 2021 **M7-M12**
UU, CEA, NIC, SOLEIL	developments towards operando XPS methodologies	NA		March 2021-August 2022 **M7-M24**
CEA/ILL	operando SANS, development of 2D cell	Anodes	interface studies	May-Oct 2021 (**M9-M14**)
Electrode microstructure, lithiation heterogeneities and single particle dynamics				
DTU	development of in-situ capillary cell	NA	interface and spatially resolved studies	
CEA	Development of microbattery for nanotomography	microbattery /capillary cells	dynamics, ageing and degradation under rate and T	March 2021-August 2021 **M7-M12**
CEA / ESRF / SOLEIL	Optimization of microbattery for nanotomography	microbattery /capillary cells	dynamics, ageing and degradation under rate and T	Proposals submitted on SOLEIL (ANATOMIX) and ESRF (ID16b)
SOLEIL	development of ptychography cell	NA	mechanics and chemistry of individual nanoparticles	

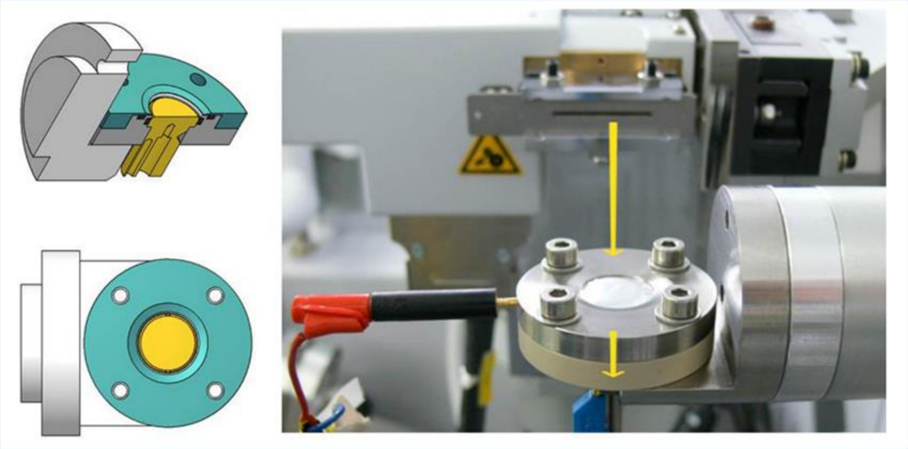
4. Summary

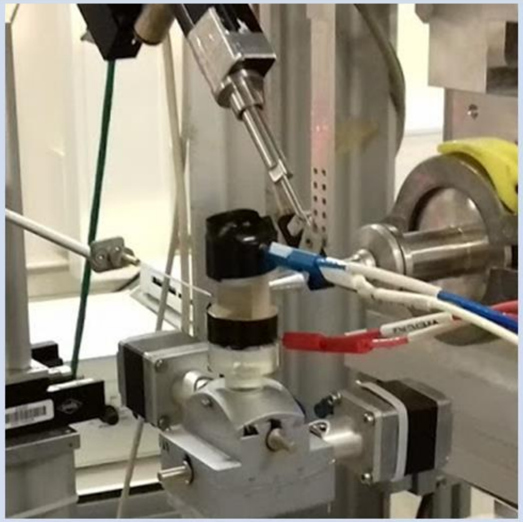
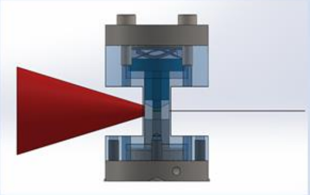
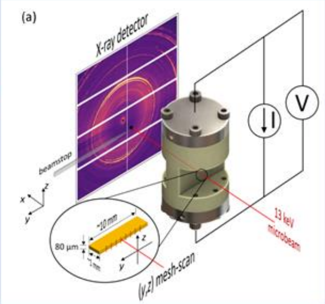
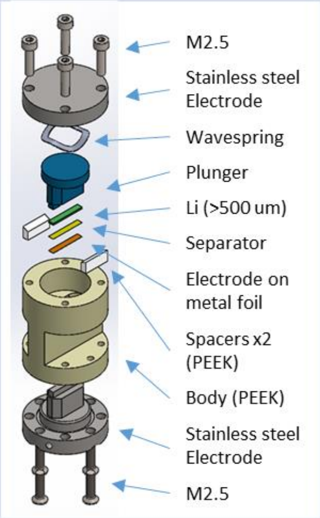
During the first 6-month period, the partners in WP5 have regularly discussed during general and cluster meetings, organized by the WP and cluster leads, respectively. The general meetings were attended by at least one representative from each of the 15 partners, and usually lasted for 2 hours, every month. Early discussions were focused on defining the organization of the work package, and were further dedicated to building collaboratively the content of the experimental matrix, the selection of Tier 1 techniques and the experimental plan.

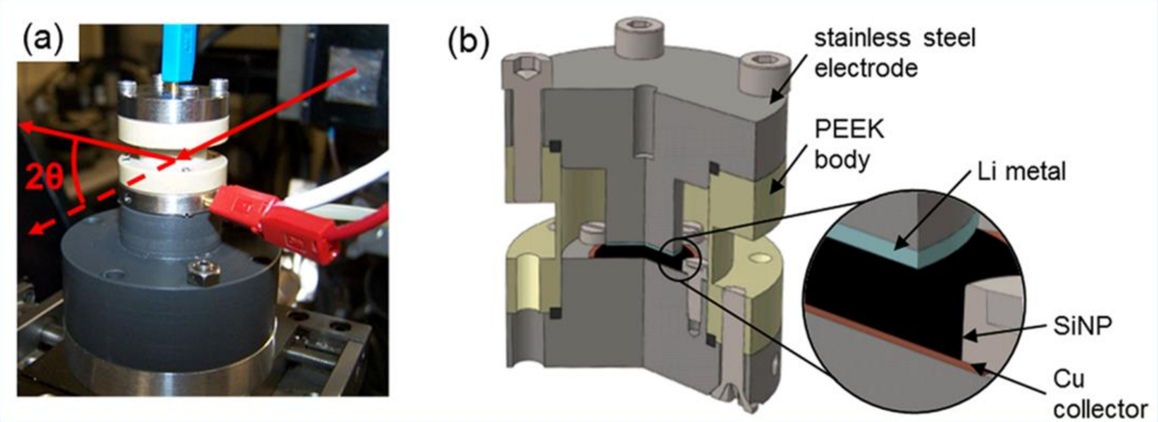
Joint meetings were also organized with other WPs, in particular WP2, WP3, WP4, WP6, WP8 and WP11, to establish the basis of the collaborations and exchanges between experimental characterization, modelling, coatings and electrolyte formulations, and AI-based analysis.

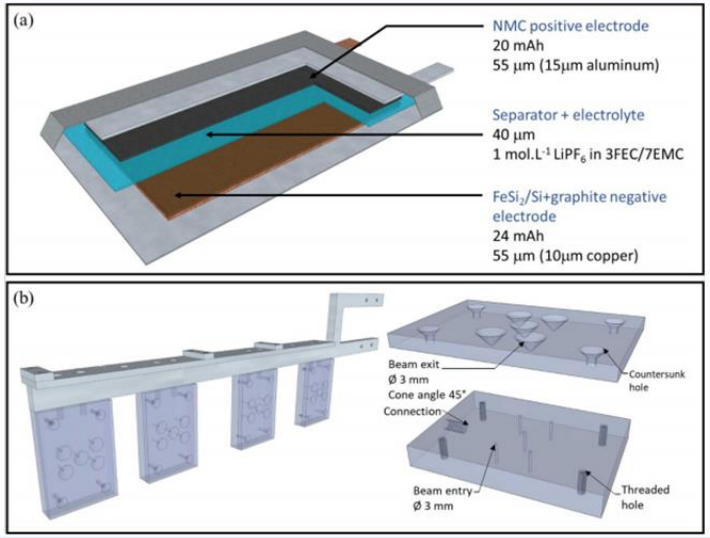
Task 5.1 and Task 5.2 were fulfilled on due time regarding the Tier 1 plan. Task 5.3 is starting at M6, and all partners have already communicated their plan for analyzing the received materials by the extended variety of experimental means available in the consortium. A number of joint proposals were submitted to the Large Scale Facilities in March 2021, expressing the vitality of the WP as well as connections established among the academic partners. Ideas of joint technical developments (cells & methodologies) emerged and will be the topic of forthcoming meetings. As an example, WP5 is planning to organize a dedicated one-day workshop on cell design and standardization. After the first phase dedicated to settle the basis of the cooperation, each cluster will also organize scientific-focused discussions to stimulate the interoperability and organize the workflow in the following months.

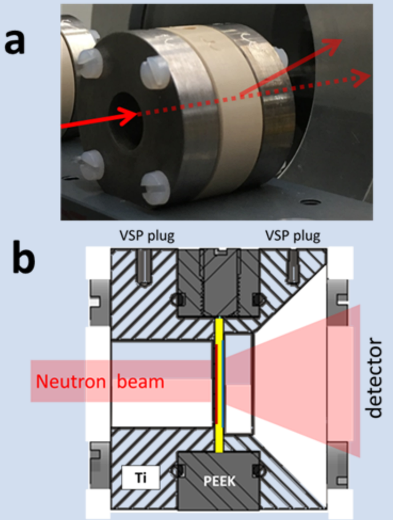
Appendix A – Cell Index

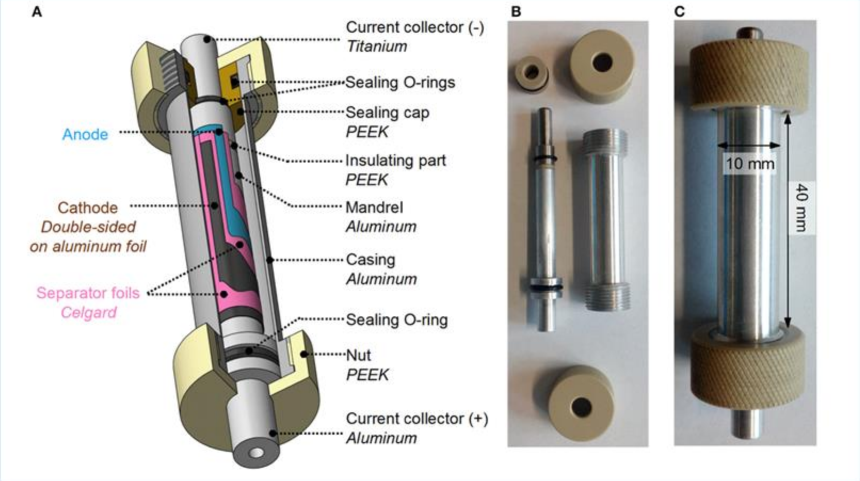
Cluster : DIFFRACTION	Partner & Contact : CEA – C. Villevieille
Cell name	Bleith Be-cell
Homemade / commercial	Homemade
Design	
Technical specifications	titanium and PEEK casing beryllium window (~100 µm) rubber o-ring seal reflection geometry
Primary technique	X-ray diffraction
Optimized for instrument	PANalytical Empyrean diffractometer
Other possible techniques	Any x-ray technique in reflection geometry
Additional information	
References	Seminal : Bleith, P., Kaiser, H., Novák, P. & Villevieille, C. In situ X-ray diffraction characterisation of Fe0.5TiOPO4 and Cu0.5TiOPO4 as electrode material for sodium-ion batteries. *Electrochim. Acta* **176**, 18–21 (2015).

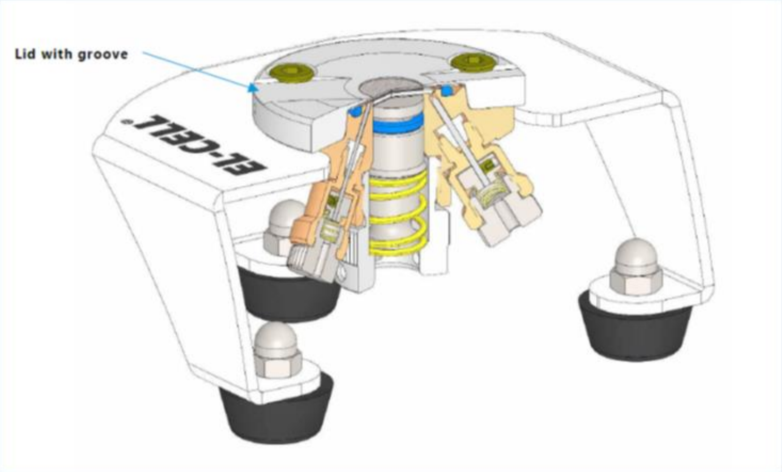
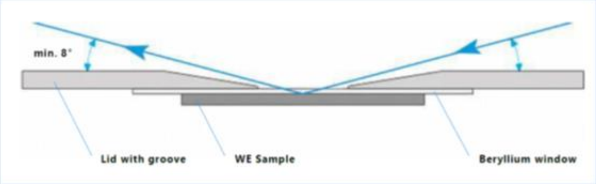
Cluster : DIFFRACTION	**Partner & Contact : CEA - Sam Tardif**
Cell name	Z-scan microfocused beam cell
Homemade / commercial	Homemade
Design	   
Technical specifications	PEEK casing Stainless steel electrodes, plunger and coin-cell wave spring rubber o-ring seal transmission geometry
Primary technique	Operando XRD across electrode depth
Optimized for instrument	ESRF ID13
Other possible techniques	Any transmission XRD
Additional information	Electrode size must be < 2 mm along the beam direction, < 10 mm transverse. No constraints on thickness, but it should thick enough with respect to the probe size (e.g. we used 80 µm for 1 µm probe)
References	Tardif et al., arXiv:2005.04983v1 **[cond-mat.mtrl-sci]** *Under review at Journal of Materials Chemistry A (01/2021)*

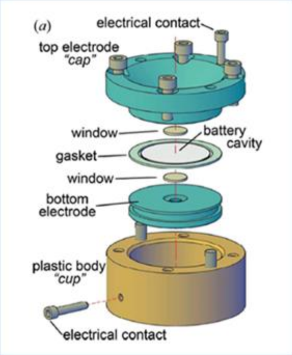
Cluster : DIFFRACTION	Partner & Contact : CEA - Sam Tardif
Cell name	BACCARA cell
Homemade / commercial	Homemade
Design	
Technical specifications	PEEK casing Stainless steel electrodes rubber o-ring seal reflection geometry
Primary technique	Operando X-ray reflectivity and diffraction
Optimized for instrument	ESRF BM32
Other possible techniques	Any reflection technique at sufficiently high energy (typ. ≥ 27 keV)
Additional information	No pressure is applied on the electrode, no separator is present. Electrode surface should be very flat for reflectivity measurements (wafer-type)
References	[Tardif *et al.*, ACS Nano 2017, 11, 11, 11306–11316]

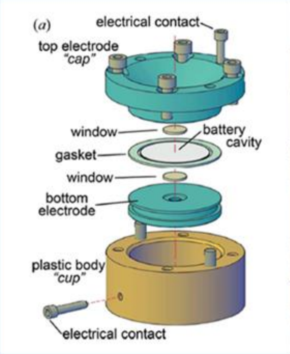
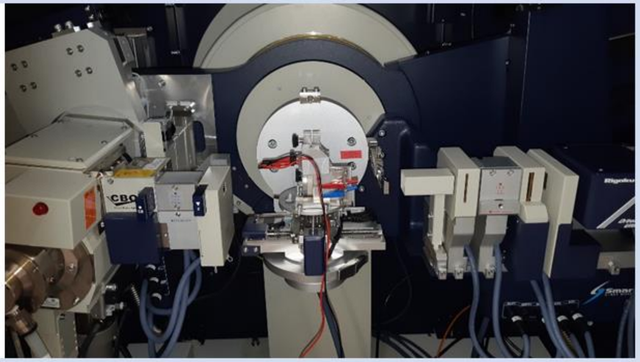
Cluster : DIFFRACTION	**Partner & Contact : CEA - Sam Tardif / S. Lyonnard**
Cell name	SAXS-WAXS cell
Homemade / commercial	Homemade
Design	  
Technical specifications	Standard pouch cell samples Aluminum holder Polyoxymethylene (POM) casing Transmission geometry
Primary technique	Operando Small and Wide Angle X-ray Scattering
Optimized for instrument	ESRF BM02
Other possible techniques	Any transmission technique at sufficiently high energy (typ. ≥ 17 keV)
Additional information	Pressure is applied to the pouch cell but it can be lower at the probe points (hole in the casing) Can be adapted for different pouch-cell sizes
References	Berhaut *et al.*, ACS Nano 2019, 13, 10, 11538–11551

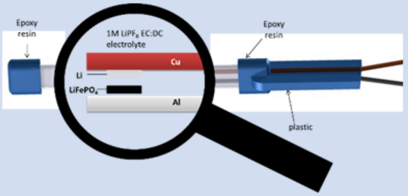
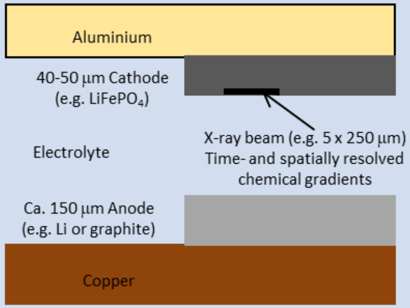
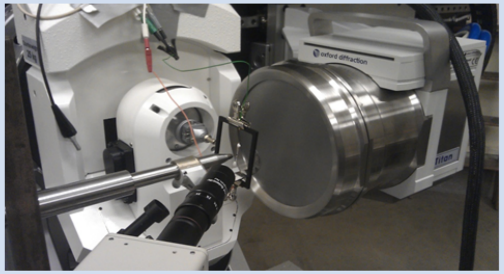
Cluster : DIFFRACTION	**Partner & Contact : CEA - S. Lyonnard**
Cell name	SANS cell
Homemade / commercial	Homemade
Design	
Technical specifications	Titanium body Transmission geometry
Primary technique	Operando Small Angle Neutron Scattering
Optimized for instrument	ILL D22
Other possible techniques	Adaptable to SAXS (with some casing material changes)
Additional information	Electrode directly coated on Ti. No pressure applied on the electrode, no separator. Several cells can me measured in parallel (sample holder available)
References	In preparation

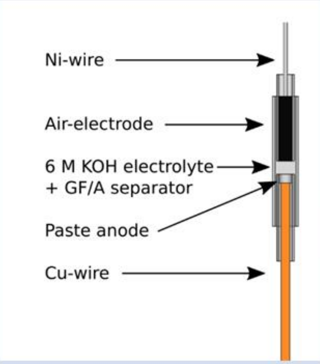
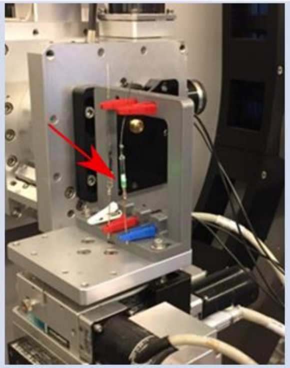
Cluster : DIFFRACTION	Partner & Contact : CEA – C. Villevieille
Cell name	Neutron diffraction cell
Homemade / commercial	Homemade
Design	
Technical specifications	Al body Stainless steel body Transmission geometry
Primary technique	Neutron diffraction
Optimized for instrument	ILL D20
Additional information	Electrode directly coated on Ti. No pressure applied on the electrode, no separator. Several cells can me measured in parallel (sample holder available)
References	Tailored cylindrical cell to study Li-ion battery electrode materials by operando neutron diffraction L. Vitoux, M. Reichardt, S. Sallard, P. Novák, D. Sheptiakov, C. Villevieille Frontiers in Energy Research, section Energy Storage, 2018, doi: 10.3389/fenrg.2018.00076

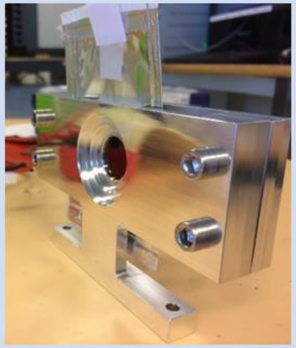
Cluster : Diffraction ans scattering	Partner & Contact : DTU – P. Norby
Cell name	EL cell ECC-OPTO w. X-ray diffraction kit
Homemade / commercial	Commercial
Design	 
Technical specifications	[ECC-Opto-Std | EL-CELL (el-cell.com)](https://el-cell.com) with 10mm X-ray window PEEK casing Glassy carbon window (200 µm) (Beryllium window available) rubber o-ring seal reflection geometry
Primary technique	X-ray diffraction
Optimized for instrument	Rigaku Smartlab, 9kW, Cu-radiation
Other possible techniques	Any x-ray technique in reflection geometry, Raman spectroscopy
Additional information	We normally use glassy carbon windows instead of the original beryllium windows.
References	

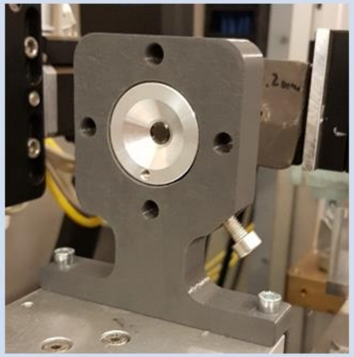
Cluster : Diffraction and scattering	**Partner & Contact : DTU - P. Norby**
Cell name	AMPIX cell -synchrotron
Homemade / commercial	Homemade (Design from APS)
Design	  
Technical specifications	Transmission geometry. Glassy carbon windows 6 position cell holder. Multi channel potentiostat
Primary technique	X-ray diffraction
Optimized for instrument	Designed for synchrotron diffraction and scattering experiments for the DANMAX Beamline at MAX IV, Lund, Sweden. The in situ electrochemical facility at DANMAX consists of a multi cell holder for AMPIX cells (6 batteries), and a Biologic multi channel potentiostat.
Other possible techniques	Total scattering
Additional information	Cell design developed at APS. Used at many synchrotron sources, e.g. APS, Petra III. Modified and adapted at DTU and University of Southern Denmark for the DanMAX beamline at the MAX IV synchrotron, Lund, Sweden.
References	Borkiewicz, O. J., Shyam, B., Wiaderek, K. M., Kurtz, C., Chupas, P. J. & Chapman, K. W. (2012). J. Appl. Cryst. 45, 1261-1269.

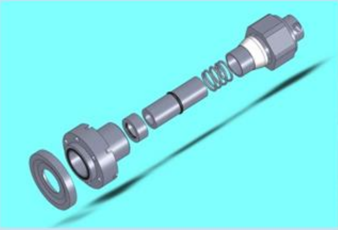
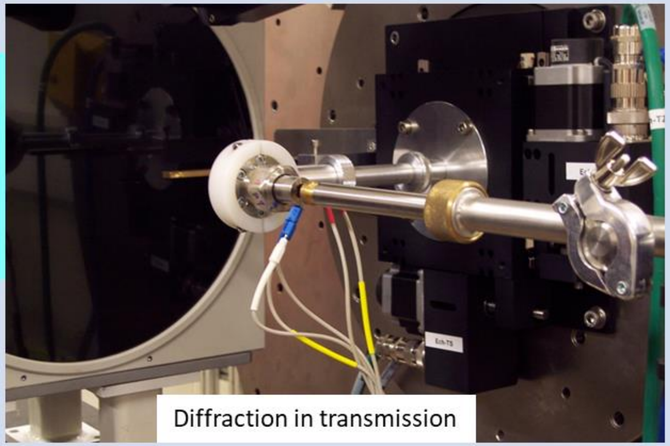
Cluster : Diffraction and scattering	**Partner & Contact : DTU - P. Norby**
Cell name	AMPIX cell – In-house
Homemade / commercial	Homemade (Design from APS)
Design	  
Technical specifications	Transmission geometry. Glassy carbon windows.
Primary technique	X-ray diffraction
Optimized for instrument	Used at in-house diffractometer (Rigaku Smartlab 9kW, Cu-radiation) (Originally designed for synchrotron X-ray radiation)
Other possible techniques	Total scattering
Additional information	Cell design developed at APS. Modified and adapted at DTU and University of Southern Denmark for the DanMAX beamline at the MAX IV synchrotron, Lund, Sweden.
References	Borkiewicz, O. J., Shyam, B., Wiaderek, K. M., Kurtz, C., Chupas, P. J. & Chapman, K. W. (2012). J. Appl. Cryst. 45, 1261-1269.

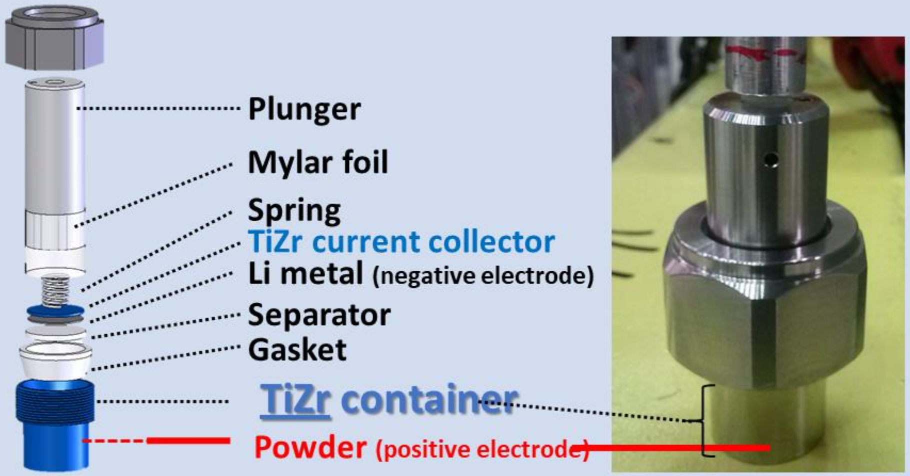
Cluster : Diffraction and scattering	Partner & Contact : DTU - P. Norby
Cell name	Capillary cell
Homemade / commercial	Homemade
Design	  
Technical specifications	Transmission geometry. Glass capillary: 1mm x 4mm Electrodes 0.7-0.8mm wide
Primary technique	X-ray diffraction.
Optimized for instrument	Designed for *in situ* synchrotron X-ray diffraction and scattering experiments. Used with X-ray energies from 12keV to 80keV.
Other possible techniques	Total scattering, spectroscopy, visual microscopy, tomography
Additional information	Used also for time and spatially resolved experiments (investigating gradients across electrode thickness) using a micro beam
References	Johnsen, Rune E.; Norby, Poul “Capillary-based micro-battery cell for in situ X-ray powder diffraction studies of working batteries: a study of the initial intercalation and deintercalation of lithium into graphite” J. Appl. Cryst. 46 (2013) 1537-1543, Young Hwa Jung , Ane S. Christiansen , Rune E. Johnsen , Poul Norby , and Do Kyung Kim *Adv. Funct. Mater.* **25** (2015) 3227-3237.

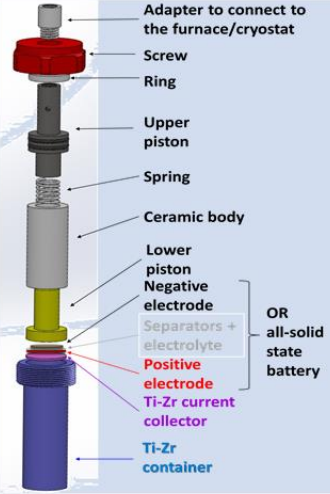
Cluster : Diffraction and scattering	Partner & Contact : DTU - P. Norby
Cell name	Metal-air battery capillary cell
Homemade / commercial	Homemade
Design	 
Technical specifications	Transmission geometry. Glass capillary: ca. 2mm diameter
Primary technique	Hard X-ray diffraction.
Optimized for instrument	Synchrotron X-ray diffraction and scattering experiments.
Other possible techniques	Total scattering.
Additional information	Capillary batteries used for lithium-air and zinc-air batteries
References	Mathias K. Christensen, Jette Katja Mathiesen, Søren Bredmose Simonsen and Poul Norby “Transformation and migration in secondary zinc-air batteries studied by in situ synchrotron X-ray diffraction and X-ray tomography”. J Mater Chem A. 2019;7(11):6459–66.

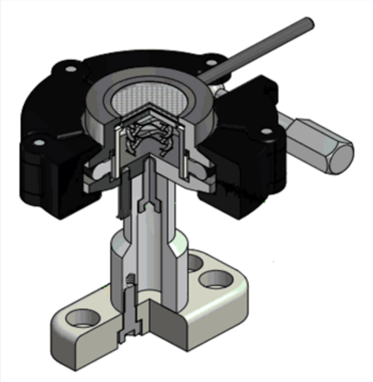
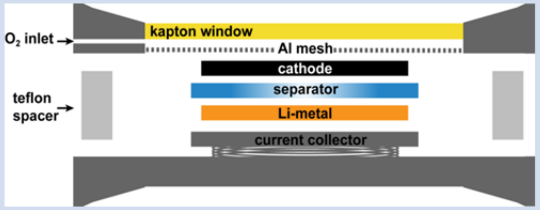
Cluster : DIFFRACTION	**Partner & Contact : Uppsala University – William Brant**
Cell name	Fast cycling pouch cell holder
Homemade / commercial	Homemade
Design	  
Technical specifications	Holder for pouch cells. Glassy carbon windows. Spring wave ensure a set stack pressure. Transmission mode.
Primary technique	X-ray diffraction
Optimized for instrument	P02.1 beam line at Petra III (DESY, Hamburg), but suitable for other synchrotrons as well. Attaches to any xy-stage.
Other possible techniques	
Additional information	
References	

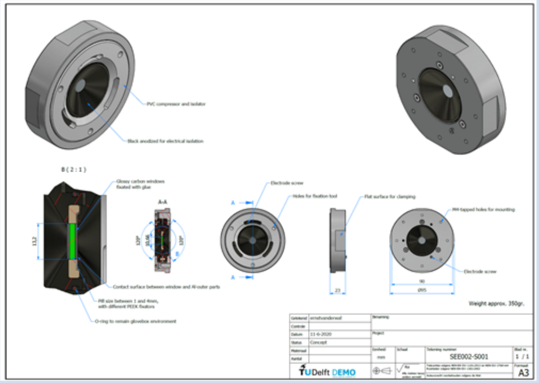
Cluster : DIFFRACTION	Partner & Contact : Uppsala University – William Brant
Cell name	Ampix-type cell
Homemade / commercial	Homemade
Design	  
Technical specifications	Ampix-type coin cell. Glassy carbon windows. Rubber sealings. Includes holder with wave spring to ensure a set stack pressure.
Primary technique	X-ray diffraction
Optimized for instrument	P02.1 beam line at Petra III (DESY, Hamburg), but suitable for other synchrotrons as well. Attaches to any xy-stage.
Other possible techniques	XAS, total scattering
Additional information	Inspired by AMPIX cell developed at the APS. Modified and adapted for ease of use. Can be rapidly implemented on any beamline.
References	

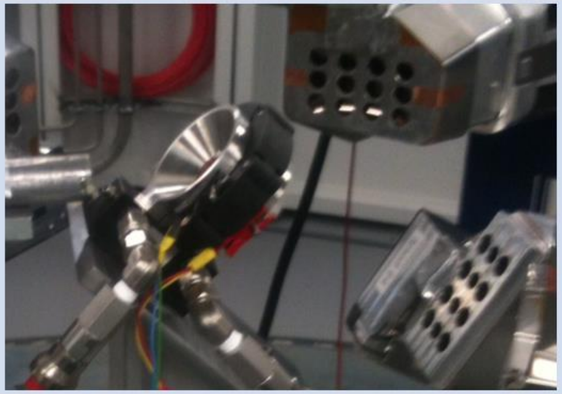
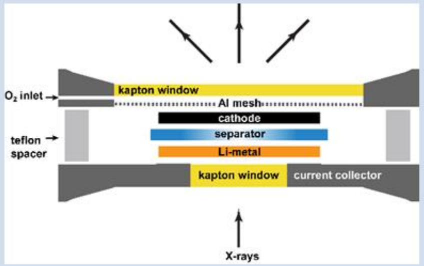
Cluster : Diffraction	**Partner & Contact : SOLEIL – S. Belin**
Cell name	Operando electrocell
Homemade / commercial	Homemade
Design	  Diffraction in transmission  Reflection geometry
Technical specifications	Stainless steel 2 Be windows (200 µm) rubber o-ring seal Transmission and reflection geometry
Primary technique	X-ray Absorption and Diffraction
Optimized for instrument	All hard X-ray XAS beamlines at SOLEIL (ROCK, SAMBA, LUCIA, ODE, DIFFABS, GALAXIES) CRISTAL beamline for Diffraction
Other possible techniques	Any x-ray technique in transmission or reflection geometry
Additional information	
References	Leriche, J.B., Hamelet, S., Shu, J., Morcrette, M., Masquelier, C., Ouvrard, G., Zerrouki, M., Soudan, P., Belin, S., Elkaïm, E., Baudelet, F. "An Electrochemical Cell for *Operando* Study of Lithium Batteries Using Synchrotron Radiation" *Journal of the Electrochemical Society*, 157(5): A606-A610. (2010)

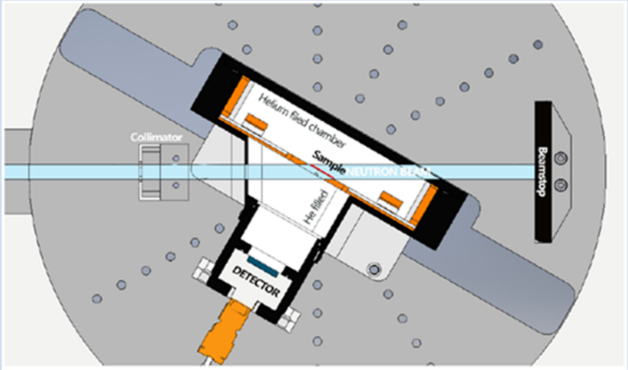
Cluster : Diffraction ans scattering	**Partner & Contact : ILL, E. Suard**
Cell name	ILLBAT#1
Homemade / commercial	Homemade
Design	
Technical specifications	TiZr casing transmission geometry (cylindrical)
Primary technique	neutron diffraction
Optimized for instrument	D20
Other possible techniques	Any neutron diffraction instrument in Debye-Scherrer geometry (transmission, constant wavelength)
Additional information	200 mg electrode in the beam, deuterated electrolytes, four cells available also for off-line tests
References	M. Bianchini, J.B. Leriche, J.L. Laborier, L. Gendrin, E. Suard, L. Croguennec, et al., A new null matrix electrochemical cell for rietveld refinements of in-situ or operando neutron powder diffraction data, J Electrochem Soc. 160 (2013) A2176–A2183. doi:10.1149/2.076311jes.

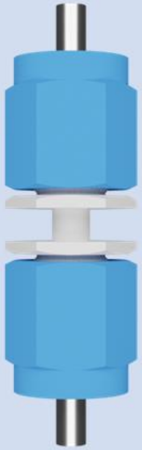
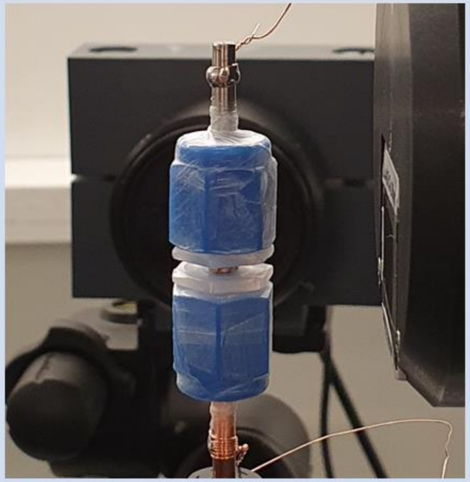
Cluster : Diffraction ans scattering	Partner & Contact : ILL, E. Suard
Cell name	ILLBAT#2
Homemade / commercial	Homemade
Design	 
Technical specifications	TiZr casing transmission geometry (cylindrical)
Primary technique	neutron diffraction
Optimized for instrument	D20
Other possible techniques	Any neutron diffraction instrument in Debye-Scherrer geometry (transmission, constant wavelength)
Additional information	Compatible for All Solid State Batteries, high or low temperatures and pressure applied by spring, 2 celles available for off-line tests
References	

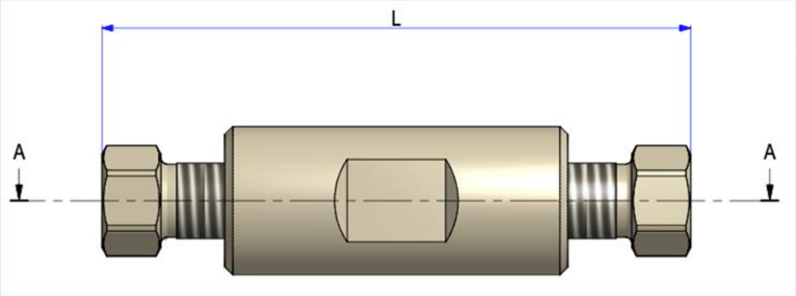
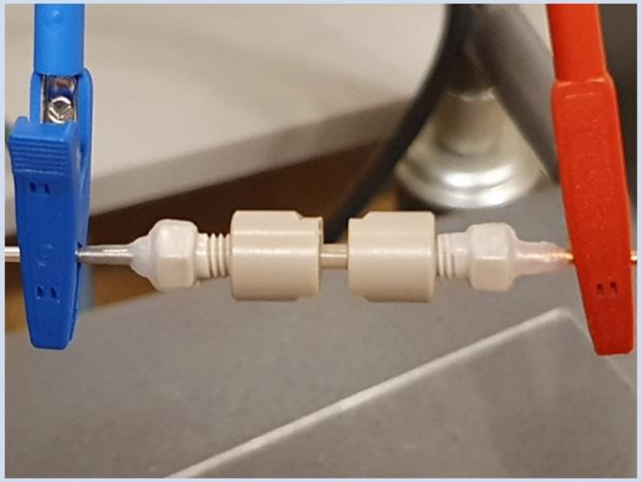
Cluster :	Partner & Contact : TUD, Swapna Ganapathy, Theo Famprikis and Marnix Wagemaker
Cell name	Dome Bragg-Brentano XRD cell
Homemade / commercial	Home made
Design	 
Technical specifications	Two versions, with and without (Swagelok) gas inlet/outlet Rubber O-ring sealing, Kapton window (25 µm), Smallest theta angle ~12 degree, Stainless steel body Can work with carbon paper or mesh metal as current collector, impregnated with electrode material of interest.
Primary technique	XRD in Bragg-Brentano
Optimized for instrument	General purpose for reflection mode XRD, now fits on Panalytical Expert-Pro, and to be updated to Empyrian
Other possible techniques	
Additional information	Especially suitable for monitor weakly scattering species, has been used to monitor Li2O2 an LiOH in Li-air batteries
References	**DOI:** 10.1021/ja508794r

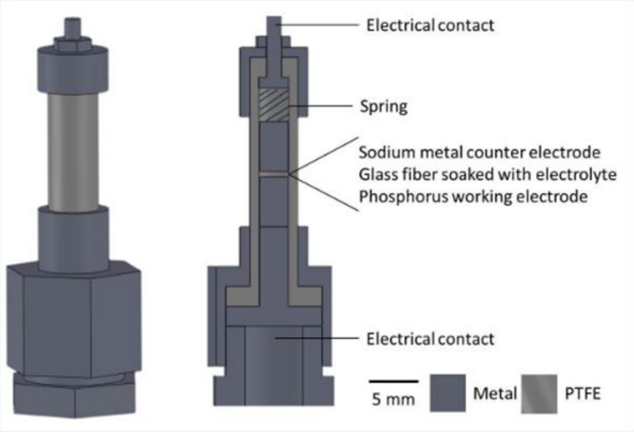
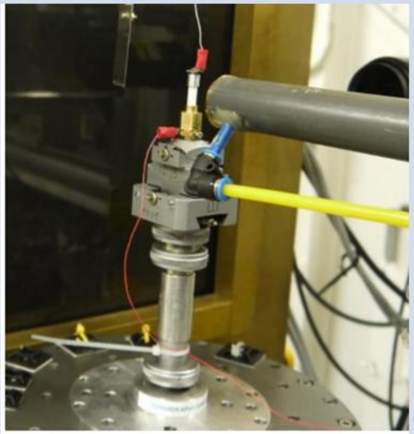
Cluster :	Partner & Contact : TUD, Swapna Ganapathy, Theo Famprikis and Marnix Wagemaker
Cell name	XRD Transmission solid state cell
Homemade / commercial	Home made
Design	
Technical specifications	Al/PVC body PEEK rings to fix the solid state pellet Glassy carbon windows
Primary technique	XRD (labsource or synchrotron) in transmission
Optimized for instrument	General purpose transmission X-ray experiments
Other possible techniques	
Additional information	Is currently being tested
References	None yet

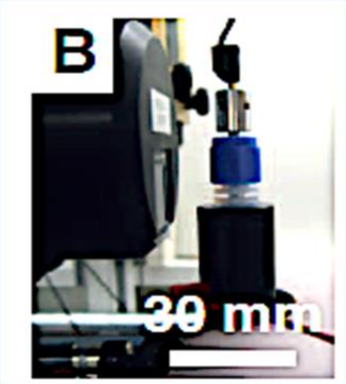
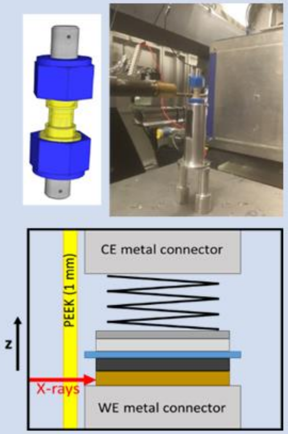
Cluster :	**Partner & Contact : TUD, Swapna Ganapathy, Theo Famprikis and Marnix Wagemaker**
Cell name	XRD Transmission cell
Homemade / commercial	Home made
Design	 
Technical specifications	Stainless steel body, O-ring sealing Kapton windows, pressure through pressurized double kapton window Gas inlet/outlet (swagelok)
Primary technique	XRD (labsource or synchrotron) in transmission
Optimized for instrument	General purpose transmission X-ray experiments
Other possible techniques	
Additional information	
References	DOI: 10.1021/acs.jpclett.6b01368

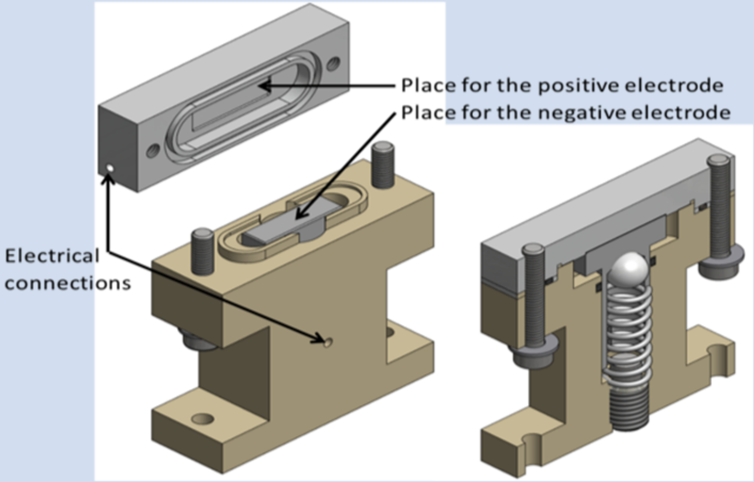
Cluster :	Partner & Contact : TUD, Swapna Ganapathy, Theo Famprikis and Marnix Wagemaker
Cell name	Neutron Depth Profiling Cell
Homemade / commercial	Home made
Design	
Technical specifications	Aluminium body and windows Integrated detector for charged particles, resolution 3.3 keV Works under 1 bar Helium Requires pouch cell battery, with current collector sealed as window
Primary technique	Neutron Depth Profiling
Optimized for instrument	Suitable for any neutron beam, beamsize collimation 1x1 cm2
Other possible techniques	
Additional information	
References	https://doi.org/10.3389/fenrg.2018.00062

Cluster : Imaging	**Partner & Contact : Chalmers – M. Sadd**
Cell name	PFA *Operando* Cell
Homemade / commercial	Homemade
Design	 
Technical specifications	Modified PFA casing, 1/8” diameter Swagelok sealing mechanism 3.125 mm diameter stainless steel rods (current collectors)
Primary technique	X-ray tomography
Optimized for instrument	TOMCAT, PSI (suitable for any synchrotron tomography beamline, including PSICHE and ANATOMIX)
Other possible techniques	XAS
Additional information	
References	

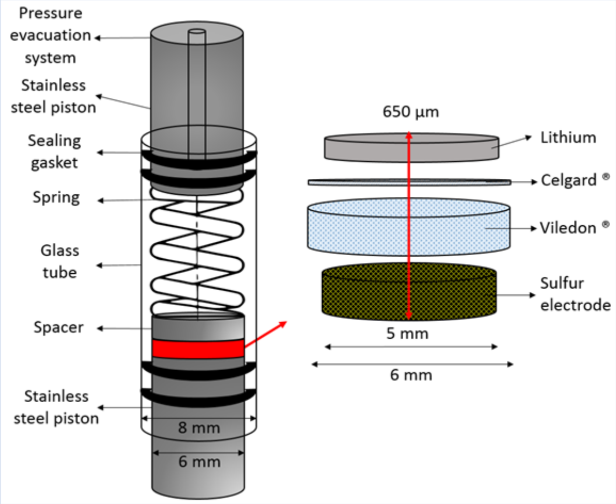
Cluster : IMAGING	**Partner & Contact : Chalmers – M. Sadd**
Cell name	PEEK *Operando* Cell
Homemade / commercial	Homemade
Design	 
Technical specifications	Modified PEEK casing, 1/16” diameter 1.588 mm diameter stainless steel rods (current collectors)
Primary technique	X-ray tomography
Optimized for instrument	TOMCAT, PSI (suitable for any synchrotron tomography beamline, including PSICHE and ANATOMIX)
Other possible techniques	
Additional information	

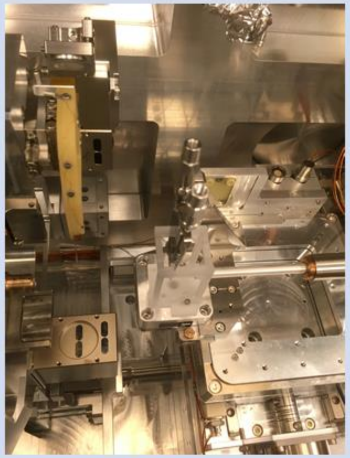
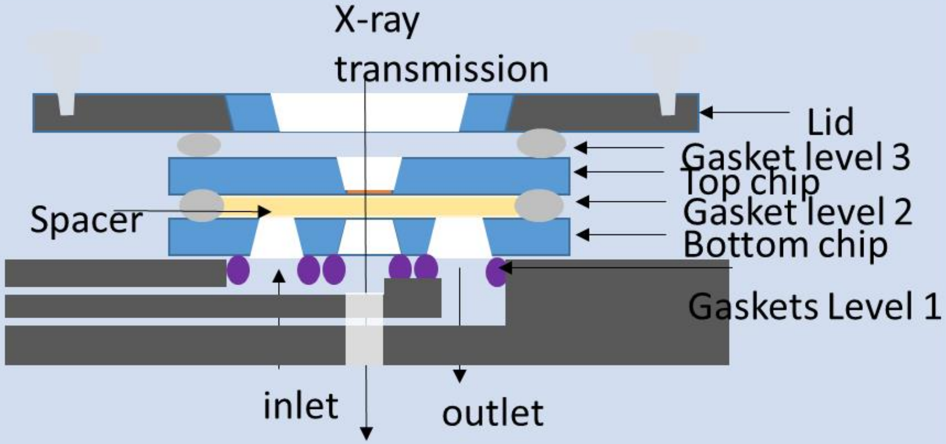
Cluster : DIFFRACTION & Imaging	Partner & Contact : CNRS – J. Sottmann
Cell name	XRD-CT cell
Homemade / commercial	Homemade
Design	 
Technical specifications	PTFE casing, Al plunger PTFE seal Transmission geometry Internal diameter 3mm
Primary technique	X-ray diffraction computed tomogrphy (XRD-CT), Pair distribution computed tomogrphy (PDF-CT), microtomography
Optimized for instrument	ID15 at ESRF (suitable for any synchrotron tomography beamline, including PSICHE and ANATOMIX)
Other possible techniques	
Additional information	
References	Sottmann J, Di Michiel M, Fjellvåg H, Malavasi L, Margadonna S, Vajeeston P, et al. Chemical Structures of Specific Sodium Ion Battery Components Determined by Operando Pair Distribution Function and X-ray Diffraction Computed Tomography. Angew Chem Int Ed. 2017;56(38):11385-9.

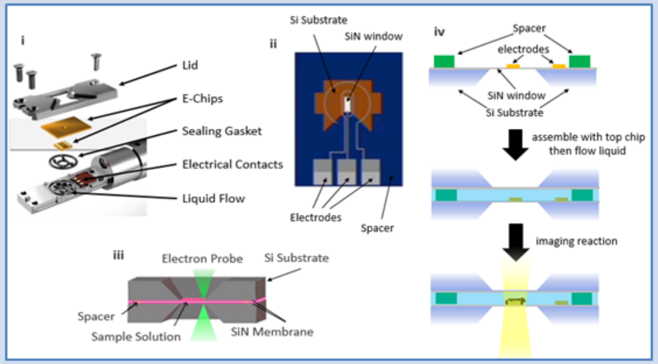
Cluster : Imaging	**Partner & Contact : CEA – S. Lyonnard / C. Villevieille**
Cell name	ScattTomo
Homemade / commercial	Homemade (for liquid and solid state electrolyte)
Design	 
Technical specifications	Swagelok body type Transmission geometry Titanium current collector
Primary technique	SAXS/WAXS tomography
Optimized for instrument	ID31 (high energy), (suitable for any synchrotron tomography beamline, including PSICHE and ANATOMIX)
Other possible techniques	Other microtomography techniques
References	Seminal : Influence of conversion material morphology on electrochemistry studied with operando X-ray tomography and diffraction C. Villevieille, M. Ebner, J. L. Gómez-Cámer, F. Marone, P. Novák, V. Wood Advanced Materials 27, 2015, 1676

Cluster : Imaging	**Partner & Contact : CEA – Claire Villevieille / S. Lyonnard**
Cell name	Neutron imaging
Homemade / commercial	Homemade
Design	
Technical specifications	PEEK casing Titanium electrodes rubber o-ring seal Transimission
Primary technique	Neutron imaging
Optimized for instrument	D50 at ILL
Other possible techniques	
Additional information	
References	To be submitted

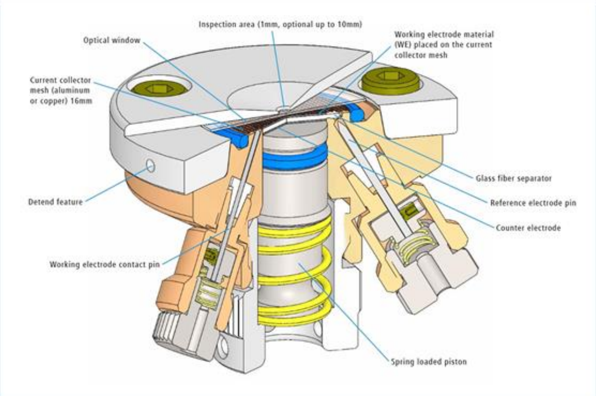
Cluster : Imaging	**Partner & Contact : CEA – C. Villevieille/ S. Lyonnard**
Cell name	Nano-tom
Homemade / commercial	Homemade
Design	 
Technical specifications	Plastic core Titanium current collector Internal diameter if 1.2mm
Primary technique	Operando Nanotomography
Optimized for instrument	ESRF ID16B
Other possible techniques	Any transmission technique requiring very small dimension
Additional information	
References	To be submitted

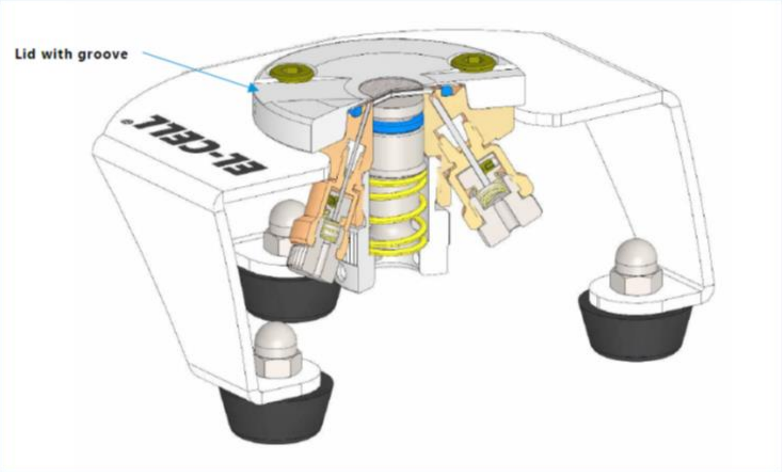
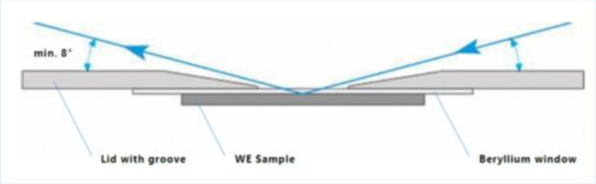
Cluster : Imaging	**Partner & Contact : CEA – S. Lyonnard / C. Villevieille**
Cell name	XRD-CT cell
Homemade / commercial	Homemade
Design	
Technical specifications	PEEK casing Stainless steel electrodes, plunger and coin-cell wave spring rubber o-ring seal transmission geometry
Primary technique	X-ray absorption tomography and X-ray computed tomography
Optimized for instrument	ESRF ID15, (suitable for any synchrotron tomography beamline, including PSICHE and ANATOMIX)
Other possible techniques	Any transmission XRD
References	G. Tonin, G. B. M. Vaughan, R. Bouchet, F. Alloin, M. Di Michiel and C. Barchasz Journal of Power Sources 2020 Vol. 468 Pages 228287

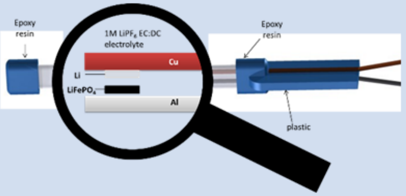
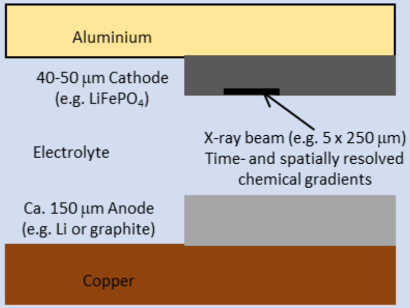
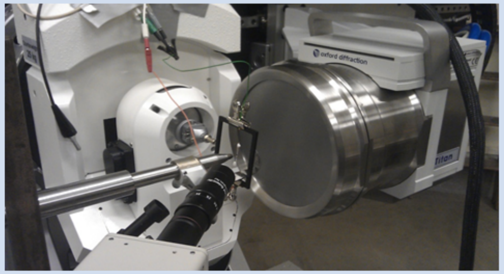
Cluster : Imaging	**Partner & Contact : SOLEIL – R. Belkhou & S. Belin**
Cell name	STXM microscopy Operando electrocell
Homemade / commercial	Commercial prototype
Design	 
Technical specifications	Stainless steel 2 SiN windows (50 nm thick) 2 rubber o-ring seal Transmission measurements
Primary technique	Soft X-ray XAS spectroscopy and microscopy (STXM)
Optimized for instrument	Soft X-ray microscopy and spectroscopy
Other possible techniques	Sub-10nm resolution Ptychography
Additional information	
References	Prototype develloped with NORCADA company. Publication and pattent are under consideration

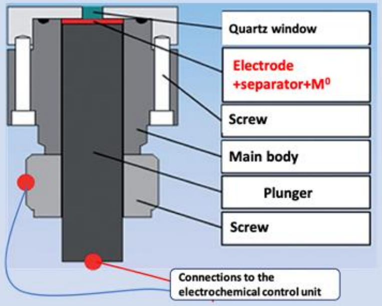
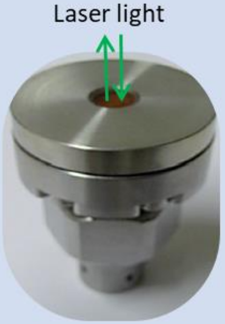
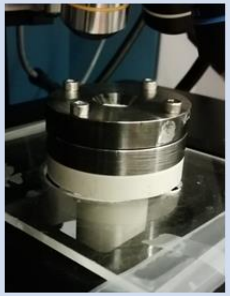
Cluster : Imaging	**Partner & Contact : Oxford, P. Adamson**
Cell name	Protochips Poseidon in-situ electrochemistry TEM holder
Homemade / commercial	commercial
Design	
Technical specifications	Liquid cell E-chips, Peek,
Primary technique	TEM
Optimized for instrument	
Other possible techniques	
Additional information	
References	Pu S, Gong C, Robertson AW. 2020 Liquid cell transmission electron microscopy and its applications. R. Soc. open sci. 7: 191204. http://dx.doi.org/10.1098/rsos.191204

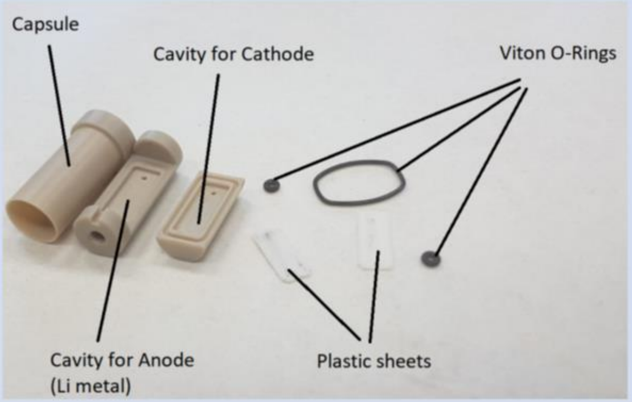
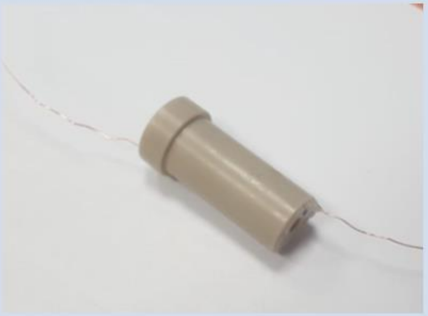
Cluster : Imaging	**Partner & Contact : Oxford, P.Adamson**
Cell name	X-ray tomography tube cell
Homemade / commercial	Homemade
Design	
Technical specifications	Delrin, Tufnol or PTFE tube for different purposes rubber o-ring seal spring loaded
Primary technique	X-ray computed tomography, Neutron computed tomography
Optimized for instrument	Both synchrotron and lab-based XCT (suitable for any synchrotron tomography beamline, including PSICHE and ANATOMIX)
Other possible techniques	transmission diffraction, NMR
Additional information	
References	

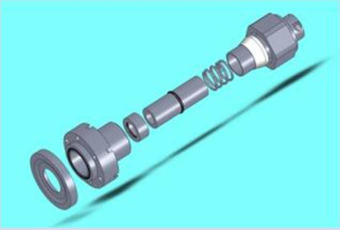
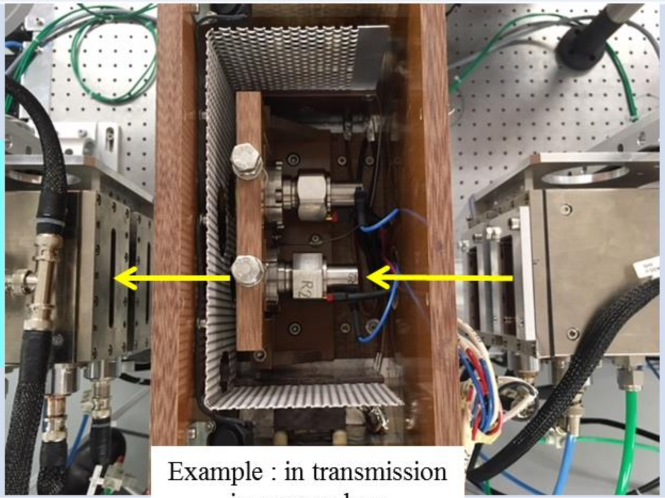
Cluster : Bulk Spectroscopy	Partner & Contact : Chalmers – Nataliia Mozhzhukhina
Cell name	EL-CELL Raman
Homemade / commercial	Commercial: https://el-cell.com/products/test-cells/optical-test-cells/ecc-opto-std/
Design	
Technical specifications	borosilicate glass window, EPDM O-rings, stainless steel 1.4404 and PEEK, three-electrode cell, reflection geometry
Primary technique	Raman spectroscopy
Optimized for instrument	LabRam HR Evoliution, Horiba
Other possible techniques	
Additional information	Electrode material has to be coated either on mesh or on separator.
References	

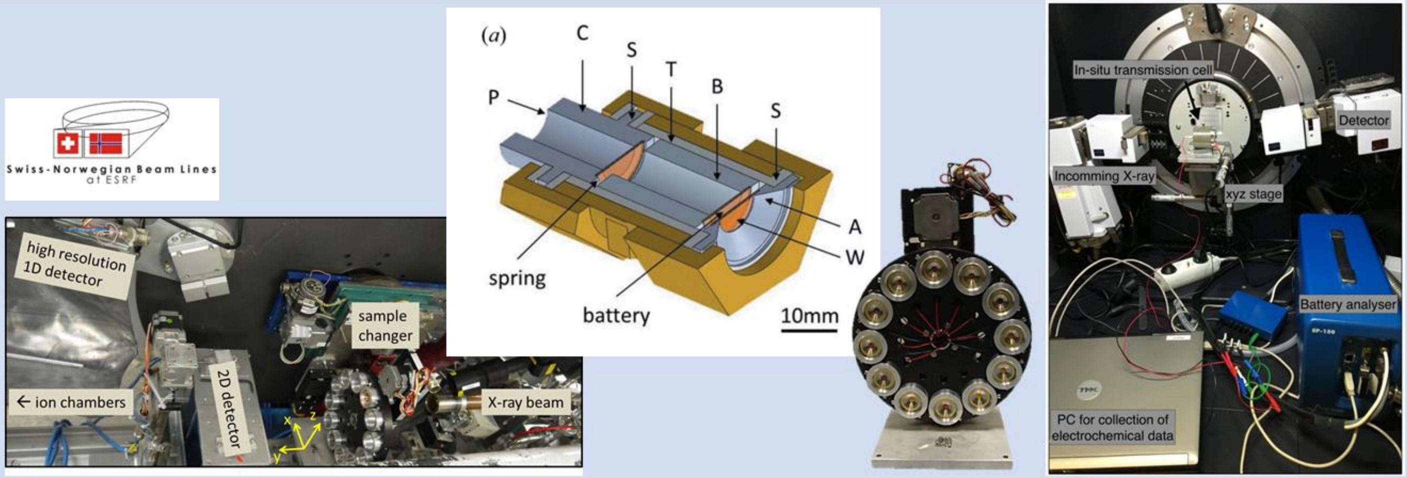
Cluster : Bulk spectroscopy	Partner & Contact : DTU – P. Norby
Cell name	EL cell ECC-OPTO w. X-ray diffraction kit
Homemade / commercial	Commercial
Design	 
Technical specifications	ECC-Opto-Std | EL-CELL (el-cell.com) with 10mm X-ray window PEEK casing Glassy carbon window (200 µm) (Beryllium window available) rubber o-ring seal reflection geometry
Primary technique	X-ray diffraction or Raman spectroscopy
Optimized for instrument	Rigaku Smartlab, 9kW, Cu-radiation
Other possible techniques	Any x-ray technique in reflection geometry, Raman spectroscopy
Additional information	We normally use glassy carbon windows instead of the original beryllium windows.
References	

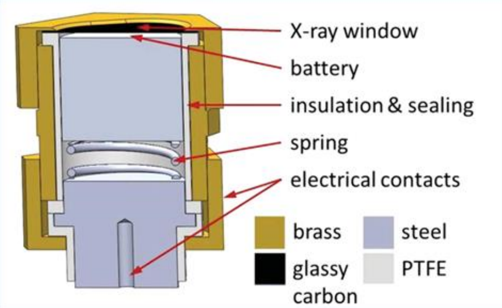
Cluster : Bulk spectroscopy	**Partner & Contact : DTU - P. Norby**
Cell name	Capillary cell
Homemade / commercial	Homemade
Design	  
Technical specifications	Transmission geometry. Glass capillary: 1mm x 4mm Electrodes 0.7-0.8mm wide
Primary technique	X-ray diffraction but could be used for X-ray Raman
Optimized for instrument	Designed for *in situ* synchrotron X-ray diffraction and scattering experiments. Used with X-ray energies from 12keV to 80keV.
Other possible techniques	Total scattering, spectroscopy, visual microscopy, tomography
Additional information	Used also for time and spatially resolved experiments (investigating gradients across electrode thickness) using a micro beam
References	Johnsen, Rune E.; Norby, Poul “Capillary-based micro-battery cell for in situ X-ray powder diffraction studies of working batteries: a study of the initial intercalation and deintercalation of lithium into graphite” J. Appl. Cryst. 46 (2013) 1537-1543, Young Hwa Jung , Ane S. Christiansen , Rune E. Johnsen , Poul Norby , and Do Kyung Kim *Adv. Funct. Mater.* **25** (2015) 3227-3237.

Cluster : Bulk Spectrosocpy	Partner & Contact : Chalmers – Nataliia Mozhzhukhina
Cell name	Montpellier cell.
Homemade / commercial	Homemade.
Design	  
Technical specifications	Two electrode cell in a coin cell configuration with fitted optical window. Two cells available: one completely of stainless steel, and another partly ss, partly PEEK. Reflection geometry, quartz window.
Primary technique	Raman spectroscopy.
Optimized for instrument	LabRam HR Evolution, Horiba.
Other possible techniques	
Additional information	Electrode material has to be coated either on the mesh or on separator.
References	

Cluster :	**Partner & Contact : TUD, Swapna Ganapathy, Theo Famprikis and Marnix Wagemaker**
Cell name	Capsule cell for operando solid state NMR
Homemade / commercial	Commercial (NMR service)
Design	 
Technical specifications	Outer PEEK casing Viton o-ring seal PTFE spacers Cu wire as current collectors
Primary technique	Solid state NMR
Optimized for instrument	Optimized for 10 mm RF coil (NMR service operando NMR probe)
Other possible techniques	
Additional information	
References	Pecher et al. Chem. Mater. 2017, 29, 213–242

Cluster : Bulk Spectroscopy	**Partner & Contact : SOLEIL – S. Belin**
Cell name	Operando electrocell
Homemade / commercial	Homemade
Design	  Example : in transmission in a warm box  Reflection geometry
Technical specifications	Stainless steel 2 Be windows (200 µm) rubber o-ring seal Transmission and reflection geometry
Primary technique	X-ray Absorption and Diffraction
Optimized for instrument	All hard X-ray XAS beamlines at SOLEIL (ROCK, SAMBA, LUCIA, ODE, DIFFABS, GALAXIES) CRISTAL beamline for Diffraction
Other possible techniques	Any x-ray technique in transmission or reflection geometry Combining with Raman spectroscopy by changing one window
Additional information	
References	Leriche, J.B., Hamelet, S., Shu, J., Morcrette, M., Masquelier, C., Ouvrard, G., Zerrouki, M., Soudan, P., Belin, S., Elkaïm, E., Baudelet, F. "An Electrochemical Cell for *Operando* Study of Lithium Batteries Using Synchrotron Radiation" *Journal of the Electrochemical Society*, 157(5): A606-A610. (2010)

Cluster : DIFFRACTION & Bulk Spectroscopy	**Partner & Contact : CNRS – J. Sottmann**
Cell name	CAEN cell
Homemade / commercial	Homemade
Design	
Technical specifications	PTFE and brass casing, stainless steel plunger Kapton and glassy carbon windows PTFE seal Transmission geometry Sample changer for 12 cells, cells can be changed quickly
Primary technique	X-ray diffraction, X-ray absorption spectroscopy
Optimized for instrument	Synchrotron (ESRF: BM31, SOLEIL: CRISTAL and ODE) and Bruker D8 with Mo radiations
Other possible techniques	SAXS
Additional information	Current collector foil in combination with Kapton windows required, battery configuration as in coin cell
References	Sottmann J, Homs-Regojo R, Wragg DS, Fjellvåg H, Margadonna S, Emerich H. Versatile electrochemical cell for Li/Na-ion batteries and high-throughput setup for combinedoperandoX-ray diffraction and absorption spectroscopy. J Appl Crystallogr. 2016;49(6):1972-81.

Cluster : DIFFRACTION	Partner & Contact : CNRS – J. Sottmann
Cell name	CAEN cell
Homemade / commercial	Homemade
Design	  
Technical specifications	PTFE and brass casing, stainless steel plunger glassy carbon window (Be possible but not tested) PTFE seal Reflection geometry
Primary technique	X-ray diffraction
Optimized for instrument	Rigaku Miniflex with Cu radiations
Other possible techniques	Any x-ray technique in reflection geometry
Additional information	Sample height manually adjusted by screw in cell holder, battery configuration as in coin cell or other common lab scale test cell or other common lab scale test cell
References	Sottmann J, Pralong V, Barrier N, Martin C. An electrochemical cell for operando bench-top X-ray diffraction. J Appl Crystallogr. 2019;52(2):485-90.