D5.5 – Design of workflow for a European experimental multimodal platform completed

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ABSTRACT

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 through the project on a selected chemistry using a subset of lab-scale and large-scale facility (LSF) techniques. D5.5 reports the design of workflow for such European experimental multimodal platform, based on the definition of key concepts and production means, and the development of an automatized selection/optimization process for time- and site-coordinated experiments matching the BIG-MAP data requests.

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1. Aim and Key concepts

1.1 Aim of D5.5

The specificity of the BIGMAP approach towards the European experimental platform is to select, organize and coordinate experiments that can be performed in the various labs of the consortium, including large-scale facilities (LSF) such as ESRF, ILL and SOLEIL. A wide array of techniques and methodologies are available, which need to be efficiently selected and applied for a given purpose. Generally, the main goals are to provide input data to establish the battery interface genome, feed/support/test the AI modules, modelling activities and material developments. Practically, it requires to identify key scientific questions on the BIG-MAP materials (what is the composition and characteristics of the SEI on graphite after few cycles, for instance) and elaborate the experimental plan to best answer the question, taking into account a number of criteria to be defined (*e.g.* what is the availability of a machine, what are the constraints in terms of sample preparation, what is the order of implementation of multi-techniques characterization if needed). The experimental workflow builds on key concepts to define how to run coordinated experiments in BIG-MAP, as well as classifications of means, tools and factors that are needed as a pre-requisite to set and optimize multimodal experimental plans integrated into the BIG-MAP central machinery and providing the desired/requested data.

1.2 Concept - Correlated measurements

A correlated measurement or correlated data-acquisition method is defined by:

• Type-1: The easiest definition of a correlated measurement is when the same sample/region of interest (ROI) is measured. In this case, the correlated *operando* measurements can be performed at the same time on the same electrode using the same set-up providing a set of data to be correlated.

•Type 2: *ex situ* and/or *operando* measurements can also be performed on the same cell and/or the same sample but not at the same time, because of the need to access different sites / labs having the specific machine / equipment needed to acquire the complete set of data. In that case, the correlation is obtained because multiple sets of data are acquired on the same piece of material prepared at the same state of charge (SOC) according to standardized protocols therefore the data can be directly compared.

•Type 3: post-mortem and/or *operando* measurements may need to be performed on different sites, different times, and different samples, but still, the data acquired needs to be analyzed in a correlative manner to provide a holistic multi-scale understanding of materials behaviour. In that case, the workflow, by defining the necessary acquisition sequences required to reach the objectives, enables the correlation at the data analysis level.

1.3 References in the context of BIG-MAP

This deliverable will rely on other deliverables and work performed in the BIG-MAP project, namely D5.1, D11.1, and the online logbook developed by WP8.

D5.1 was submitted at M6 and set the state-of-the art experimental matrix, selected Tier1 techniques, and corresponding experimental plan. This report described the WP5 organization 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 LSF data of many types, including *operando* data, according to the global project workflow.

D11.1 was submitted at M12. It defines and estimates data fidelity and cost. The annex is especially useful in this case as the cost and fidelity are estimated for most techniques described in the experimental matrix (D5.1).

The online laboratory notebook developed in WP8 will contain all experiments performed in WP5 uploaded in a standardized manner, allowing to share metadata (and possibly data) and enabling search tools to access experimental status by keywords. Those keywords may include different types of information, allowing the extraction of parameter-selected maps. For instance:

- All experiments performed by a given partner (e.g., what did Partner X measure so far?)
- All experiments conducted on a given material (e.g., who measured LNO?)
- All experiments using a given technique (e.g., who performed X-ray diffraction?)
- All experiments focusing on a given observable (e.g., who investigated crystal structure?)
- Links between experiments and modelling/AI/materials (e.g., who performed a modelling-triggered experiment?)
- Links between experiments (e.g., who performed a given sequence of connected experiments?)

2. Workflow

New materials and electrolyte compositions are formulated and tested in WP4 and WP6. In parallel, atomistic modelling and simulations are performed by WP2 and WP3. This large dataset composed of basic characterisations, electrochemical data, and theoretical results aims at feeding a multi-fidelity machine learning model guiding the identification of further characterisation to advance material optimisation. These characterisations are performed by WP5 following the workflow detailed in this section. Four main building blocks were identified and are schematically presented in Figure 1: i) definition of the observables and sample characteristics, ii) the identification of possible instrument(s), iii) the determination of the experimental plan(s) optimizing the desired fidelity and cost and iv) the complex interactions between measurement, data processing and data analysis required to deliver correlated, on-the-fly and/or high throughput data to the machine learning model.

Figure 1. General schematic of the WP5 workflow building blocks.

2.1 Definition of observables and sample conditions

This section lists and details the information needed to design characterisation experiments. This information will have to be part of machine learning model outputs.

- Observables, for example, cell parameters of an SEI's crystal structure or chemical composition. A list of the most common observables has already been defined in D11.1.
- Sample shape (model or real electrode, powder of active material, or liquid electrolyte). Indeed, specific techniques can only be applied to a certain type of sample.
- Region of interest. The ROI can be i) the average, ii) a random ROI, iii) a specific ROI. For example, specific ROI can be the top left corner of a pouch cell electrode or a micron-sized zone specified by coordinates. Note that this requires the need to develop techniques/technologies able to mark samples, especially for small ROIs.
- Correlations between the observables and/or ROI. Two observables might need to be correlated. For example, tracking the chemical composition of the SEI together with the crystal structure of the underlying graphite electrode. Correlated experiments are defined herein by using the same sample and, if needed, measurements of the same specific ROI. Correlated measurements, if they can't be performed in the same instrument, need to be performed in sequence. Therefore, sample workflows need to be considered carefully when correlated analyses are required.
- *Operando*/*in situ* characterisation. If *operando* experiments are needed to track the evolution of observables over time or potential, time and potential resolutions need to be defined (number of data points per minute, for instance).
- Data fidelity and time cost required. Amongst the list of available techniques which can be used to monitor observables, some are more accurate, and some take more time. Concepts of fidelity costs are described in D11.1. The fidelity and cost of all available techniques will be dynamically refined along with the project as detailed in the annex of D11.1.

2.2 Definition of the list of possible instruments

Having defined the required inputs, we propose a workflow to identify an exhaustive list of instruments capable of answering the characterisation request (Figure 2). Each potential instrument would be tagged with extra information regarding, for example, the *operando* cell used or the availability of the machine. This list will be used in the next step to finding the best measurement plan according to the desired fidelity and cost.

2.2.1 Exhaustive list of available instruments from a list of requested observables

The starting point would be the list of observables. Using the annex of D11.1, the observables can be matched to techniques with fidelity and cost, resulting in a 'matrix' containing the list of possible techniques for each observable. To move from techniques to instruments (and partners), the experimental matrix in D5.1 listing the different partners having available equipment will be used. This matrix already contains crucial information such as the probed area and penetration depth, measurement time, observables, and the possibility to collect *operando* data, which will be used to refine the choice of the instrument based on the possibility of a measurement and the requested cost and fidelity. For all instruments, a user-defined availability for BIG-MAP research (low/medium/high) and the number of already submitted BIG-MAP samples will be determined and tagged to the instrument. This tag will be used later on to access the time cost of the instrument.

It is important to note that some observables might not be obtained by any technique available in WP5. In this case, the requested observables are saved and used in the production of a report enumerating the most wanted observables. This report could be used to guide the choice of extra partners, techniques, or development that could be carried out in the framework of BIG-MAP stakeholder initiatives.

2.2.2 Assessing the feasibility of the measurements based on request inputs: the filtering process

At this point, the long list of potential instruments will be shortened, taking into account the other inputs of the request while tagging the remaining instruments. To do so, a list of questions needs to be answered for each possible instrument:

- Is the sample shape compatible with the machine, and can the ROI be measured? All machines that cannot perform measurement on the requested sample shape or ROI are removed from the list. For example, a laboratory micro tomography machine cannot be used to assess the morphology of active material in a sub-micron ROI, but scanning/transmission electron microscopy would be suitable. Measuring Li content by neutron diffraction can be done on solid crystalline electrodes but not in electrolyte solutions.

- Does the observable need to be measured *operando/in situ?*
  - Are there available cells? All techniques without *operando* cells should be removed from the list, and the remaining will be tagged with the possible *operando* cells. Note that the fidelity of the different cells will be assessed based on their electrochemical behaviour and signal quality (see below).
  - Can the time resolution be achieved? In other words: Is the integration time of the experiment short enough to capture the developments in the sample during an *operando* experiment? E.g., if a cell is cycled at 1C, the integration time should be of the order of a minute to capture the processes without too much averaging.

- Do observables need to be correlated?
  - Are there instruments capable of measuring all correlated observables? For example, measuring the crystal structure together with the oxidation state of transition metals in cathode materials is possible on some synchrotron beamline offering correlated XRD and XAS experiments. Such instruments could be found by consulting the list of instruments per correlated observables and should be tagged.
  - Do correlated measurements need to be measured *operando*? Correlating *operando* measurement implies measuring the same sample at the same time and hence can only be done in a machine capable of measuring all observables at once, which has been identified in the previous points. Moreover, the cell has to be compatible with the different techniques. This can be checked from the list of *operando* cells tagged to the techniques (see above).
  - Do observables need to be correlated on a specific ROI for an *ex situ* sample? To perform XRD and XPS on a few microns region of the same *ex situ* electrode, special sample holders have to be used. These holders should i) be compatible with different machines, ii) allow to find specific ROI, and iii) be air-tight. Note that air-tight means that using this sample holder, the sample will not be exposed to air at any point from

the preparation to the shipment of the sample to the next partner; hence sample holder might have to be accompanied by transfer chambers. A table summarizing the list of sample holders available from different partners will be created (see Table 1) and dynamically refined in light of the current developments in WP5. Only the correlated techniques that can be used with one of the reported sample holders will be kept in the list, and the sample holder label attached to the machine/partner. Note that the availability of the sample holder should be considered for the time cost analysis using both the user-defined availability and the already submitted sample list. Note that if average information over the sample is needed, no such sample holder is required, indeed the sample can be measured by usual means, i.e., mounted in a machine-specific air-tight holder, measured, dismounted, and transferred under inert atmosphere to the next partners/machine.

Answering 'no' to most of these questions would remove the instrument from the list. The observables, the removed instrument, and the reason for its removal could be saved and used to establish a report identifying the most desired technical development. Special care would be given to observables having an empty list of possible instruments after the filtering process – meaning that they can't be measured at all. For example, *operando* secondary cathode particles cracking might be a popular observable which could, in theory, be done by nanotomography at ANATOMIX (Soleil) or ID16b (ESRF). However, these nanotomography instruments are removed from the possible instrument list, and hence saved in the discarded instrument list, due to the absence of available *operando* cell or time resolution. As a result, there is no instrument capable of measuring the requested observable. The rejected instruments would be tagged and highlighted in a report.

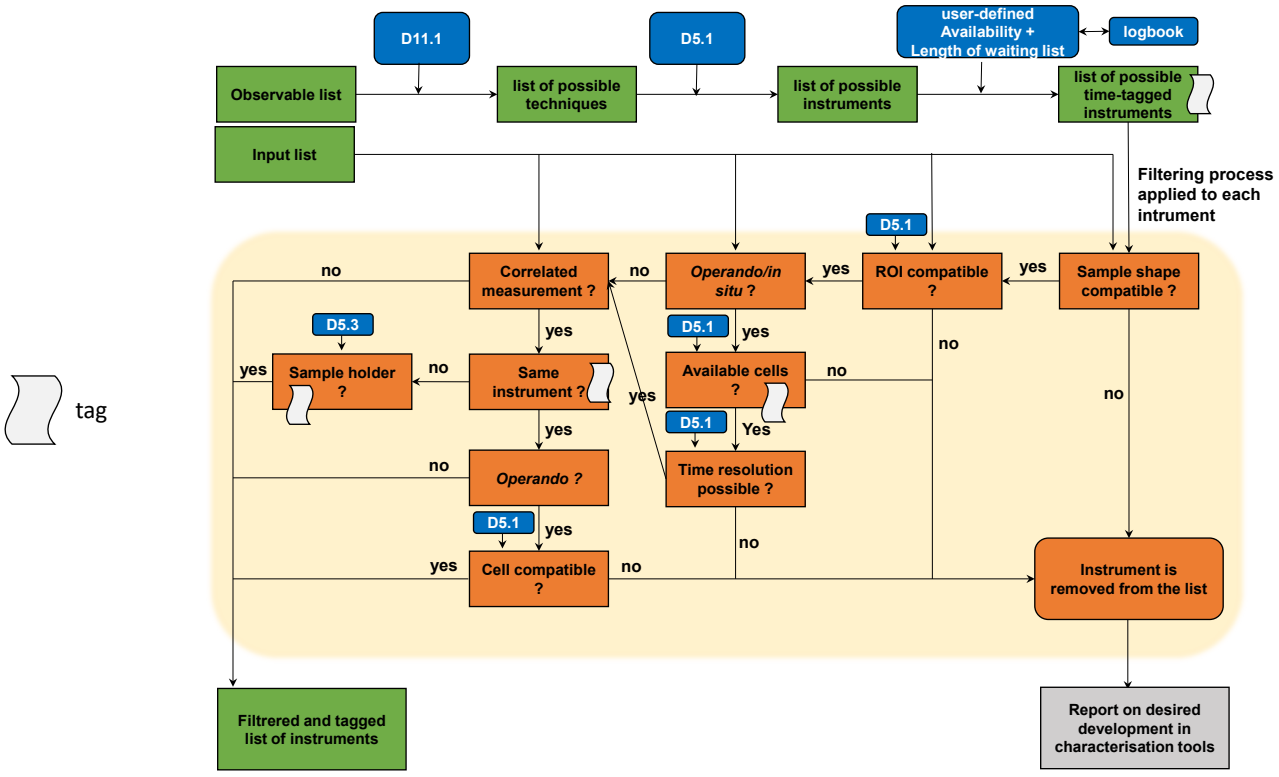

Figure 2. Workflow for the determination of the list of instruments capable of performing the requested characterization.

Table 1. Model for the correlated experiment sample holder list, with an example from the CEA.

Sample holder label	List of compatible instruments and air-tightness			user-defined availability (low/medium/high)	List of planned experiment	sample shape
XPS/SEM transfer chamber	XPS	CEA	Air-tight	medium	updated with logbook	electrode, powder
	SEM	CEA	Air-tight			

2.3 Optimized measurement plan

From the list of tagged and possible instruments, a measurement plan needs to be built. It contains the ideal sequence of measurements according to the required fidelity and cost. First, fidelity and cost criteria need to be defined, allowing to rank the list of possible measurement plans, then the selection process of the measurement plan will be discussed.

2.3.1 Determination of fidelity and cost criteria

We summarize several criteria (Table 2) that need to be taken into account for the determination of the measurement plan. Among the proposed criteria, the sample integrity after measurement (in the 'instrument' tag) requires some attention. Some techniques are destructive (focused ion beam SEM), and some might lead to sample degradation (synchrotron experiments); hence we need to have special care positioning these techniques in the measurement plan. This could be done by updating the experimental matrix in D5.1 with a new category: sample damage (low/medium/high). The sample measurement sequence would be organised to minimize the sample degradation as a first priority by default. If two destructive techniques are needed, then two samples have to be prepared. This leads to the definition of fidelity and cost of sample preparation. This notion covers material synthesis and electrochemical preparation procedure in case of *ex situ* samples, but not sample preparation for specific characterisation instruments – already taken into account in the 'instrument' tag. This type of fidelity and cost needs to be defined together with WP4 and WP6. Concerning the impact of these criteria on the measurement plan, long sample preparation time and low fidelity might reduce the number of parallel characterisations at the benefit of sequential procedure hence reducing the number of samples needed. Regarding the *operando* cell tag, the fidelity will be assessed by both the electrochemical performance of the cell and the resulting data quality. Electrochemical performance assessment could compare cycling data in standard conditions (defined by WP8). The data quality might depend on several factors; for example, the cell casing material might produce a strong signal overlapping with the data of interest, hence complicating its analysis. The data quality of the cell would be user-defined (low/medium/high) for each compatible instrument. For the logistics, although it seems commonplace, a source of loss of fidelity might be sample degradation due to ageing, air-exposure, etc. One of the important sources of delays and exposure is transport, hence the interest in reducing unnecessary travels. These travels would also contribute to the time cost. Delivery time between partners can be roughly estimated (min, hours, days, weeks) depending on the geographical situation of the partners (same lab, same city, same country, different country).

Table 2. Criteria for the fidelity and cost determination.

Tag	Fidelity	Cost	Extra
The instrument	Fidelity of the technique defined in D11.1	Sample preparation, measurement, data processing, data analysis, as established in D11.1	The integrity of the sample after measurement
	User-defined quality of the instrument itself (low/medium/high). Example: r*esolution, signal/noise ratio*	Time cost: User-defined availability (low/medium/high). Number of BIG-MAP samples already submitted	
	User-defined complexity of the sample preparation (low/medium/high)	Technique cost: see D11.1 annexe	
Sample preparation	Low/medium/high (samples from industrial partners might be 'ranked' in high fidelity, while modified samples could be 'medium'. See with WP4 and WP6)	Low/medium/high (samples from industrial partners might be 'ranked' in high fidelity, while modified samples could be 'medium'. See with WP4 and WP6)	None
Sample holder	None	Time cost: User-defined availability (low/medium/high). Number of BIG-MAP samples already submitted without the data processing and analysis time	None
Operando cell	Electrochemical performance	User-defined availability (low/medium/high)	None
	Quality of data, user-defined for each available instrument (low/medium/high)	Number of BIG-MAP samples already submitted	
Logistic	Depending on the number and the location of the involved instruments	Depending on the number and the location of the involved instruments	None

2.3.2 Building and selection of the measurement plan

Building the right algorithm to find the different measurement plans and calculate their respective fidelity and cost is beyond the aim of this deliverable and could be discussed with WP9-10-11, for instance. In this part, we will discuss elements to be considered in the selection process.

Main proposer

Before the machine learning model is able to submit its own characterisation requests and chose the right measurement plan, it will probably be performed by a partner – the main proposer. The main proposer will receive the measurement plans sorted depending on the desired fidelity and cost and will have to choose among these options. Tools could be developed to guide this decision.

Cross-validation of experimental plan using the logbook

As we know, instruments might have punctual technical issues preventing a certain type of analysis, or fluctuating workforce in labs could affect the time scale at which an experiment is performed. To check the capability of partners to perform experiments, the logbook could be used. Comparing the measurement plan with recent similar experiments in the logbook informs on the capability of partners to perform such measurements. If experiments from the measurement plans have never been logged into the logbook, one could imagine contacting the partners to confirm the possibility of the experiments, or at least having a special label on this experiment plan guiding the choice of the measurement plan.

What kind of experiment can be expected depending on fidelity and cost?

Depending on the fidelity and cost chosen, the main proposer should be directed towards different types of measurement. Table 3 summarizes typical experiments in different measurement plans. High fidelity and high cost will target correlated *operando* large-scale facility experiments, while low cost and low fidelity will correspond to single high throughput experiments. In between, measurement plans will consist of single LSF experiments or correlated *operando* lab-scale facilities.

Table 3. The table presents the type of experiments expected in a measurement plan depending on the fidelity and cost.

	High Fidelity	Low Fidelity
High cost	Correlated multimodal *operando* experiments, including large scale facilities	Single large scale facility measurement
Low cost	Correlated or *operando* experiments on laboratory instruments	Single and high throughput laboratory experiments

Validation and planning

Final validation and planning of the experimental plan should be performed by the partners themselves. For this, partners might be contacted sequentially and asked to accept and plan the experiment. Acceptance leads to contacting the next partner, while refusal leads to the calculation of new measurement plans. Already validated experiments would be kept fixed on the new calculated plan.

2.2 Measurement, data processing, and data analysis loop

In the following section, in Figure 3, we describe a measurement plan workflow emphasizing the developments needed to move towards correlated, high-throughput experiments and on-the-fly analysis. However, workflows and especially the density of feedback loops, interaction with other WPs or databases will depend on the type of experiments. For example, data analysis of typical laboratory-based single measurement might not require modelling, but a database will be crucial. However, more exotic spectroscopies (Quasielastic neutron scattering, resonant inelastic X-ray spectroscopy) have limited databases, and hence the analysis of the measured data rely on modelling. In the following, the measurement, data processing, and analysis workflow will include all possible feedback and WPs, which might not be necessary for all cases.

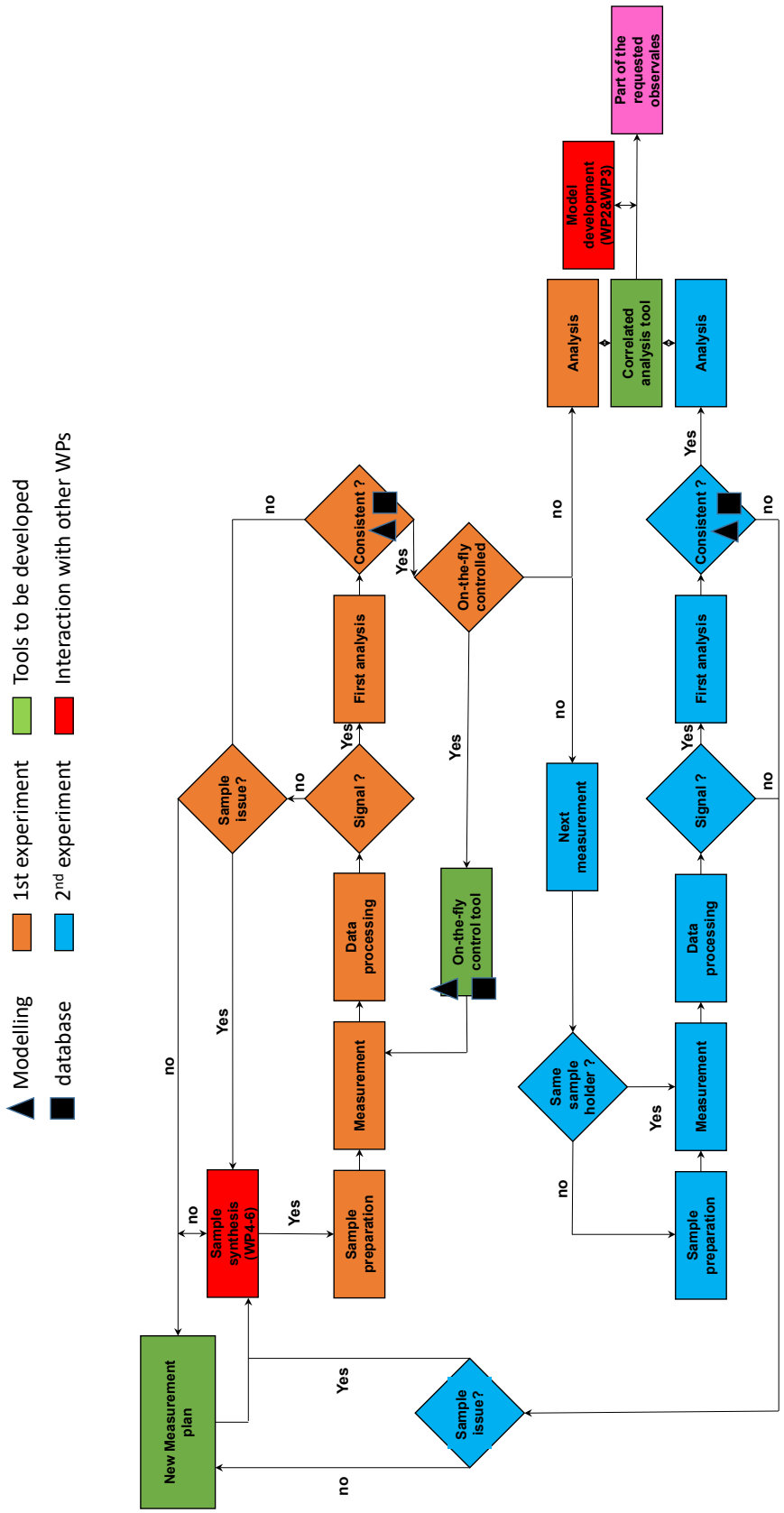

Figure 3. Measurement plan workflow with on-the-fly control loop and correlated sequential experiments.

The proposed workflow contains the following items:

Data analysis steps towards on-the-fly measurements

After validation of the measurement plan, samples are prepared and shipped to the 1st partner of the measurement plan. Measurements are performed and data processed. At this point, the data is checked, and the possibility of obtaining the observable of interest with adequate fidelity is assessed (box 'signal?' in Figure 3). For example, if the XPS signal from SEI components is too low to be measured on a specific sample, the composition of the SEI will not be observed; hence a different technique or sample should be proposed. The first analysis is then performed aiming at determining the observable in a relatively short time and with low fidelity. The result is then compared with model prediction or previous experiments registered in the logbook. In case of large deviations, the measurement plan or the sample is modified. The degree of deviations of the data to the models/previous experiment necessary to trigger the validation loop will need to be defined carefully but would probably be designed to move aside suspicious results. If the data is in general agreement with the expected results, it is considered valid. At this stage, a refinement of the data acquisition could be performed in interaction with modelling and the logbook corresponding to an on-the-fly control of the experiment. For example, depending on the requested fidelity, the number of zones needed to obtain an average microstructural information from microscopy could be determined. While measuring Li heterogeneity *operando*, the list of C-rates could be dynamically updated based on data from other C-rates and modelling. The possibility of performing measurement optimisation on the fly will ultimately rely on the speed at which data processing, analysis, database search, and modelling can be performed and will be detailed in D5.4.

Feedback loop effects, modification of characterisation request or measurement plans

Some measurements might not lead to the requested observables because of instrument or sample limitations. In this case, the characterisation request (sample and experimental conditions) and/or the measurement plan (technique or instrument) should be modified. Changing the characterisation request might have consequences on the entire measurement plan (for example: changing the sample shape), but hopefully, such situations should be rare considering that each experiment has been cross-checked with already existing measurements and validated by experts (the partners responsible for the instrument). Regarding the measurement plan, adding or replacing an instrument should be done without major modification of the rest of the measurement plan, already validated and planned, prioritizing available instruments over high fidelity.

Correlated data analysis

In case of correlated observables, a correlated data analysis strategy has to be implemented. Some tools already exist, such as combined X-ray and neutron diffraction pattern Rietveld refinements or total scattering and extended X-ray absorption fine structure (EXAFS) refinement using reverse Monte Carlo (RMC). These types of datasets can be analysed concomitantly because they describe similar observables – periodic crystal structure and nearest neighbours. Implementing the use of these tools is the first step. Correlating data observing different length scales would be interesting; one could imagine analysing X-ray scattering data in a very large Q range giving information from the micron to the atomic scale. However, such an approach is difficult due to the discontinuity of the models used for the data analysis. WP3 – multiscale modelling - aims to unify models describing different scales and hence ways of analysing correlated multiscale data.

2.5 Development of new tools and workflows

To perform the presented experimental plans, enrich *operando* correlated experiment possibilities, or integrate LSF experiments, developments are currently ongoing in WP5 and will be presented in this section.

2.5.1 *Operando* cell development

Beyond assessing dynamic information, *operando* measurements, by definition, have the advantage of measuring under realistic conditions circumventing the issues of relaxation or washing away species during cell disassembly and sample preparation for *ex situ* measurements. Ideally, all measurements would be done *operando*. However, this is limited by the availability of *operando* cells, hence the need to enlarge the consortium *operando* cell portfolio with cells having good electrochemical performance and compatible with several instruments.

A list of available cells is presented in D5.1, and during a dedicated hands-on workshop organized by ESRF and SOLEIL, their use, pros, and cons have been discussed amongst most of the WP5 partners (Table 4 for the program). It has been agreed that the available cells will be quantitatively compared in terms of electrochemical performance setting up a baseline for further improvement. The development of a BIG-MAP *operando* cell has also been discussed during the workshop. The cell, developed by SOLEIL, will aim at performing correlated diffraction, X-ray absorption spectroscopy, and Raman spectroscopy with high fidelity on the electrochemical performances.

Table 4. Program of the physical 'hands-on' *operando* cell meeting organized by ESRF and Soleil and held the 3rd of November at the ESRF.

Time	Program
9h30 – 10h00	Meeting & welcome
10h00 – 10h20	Gilles Moehl (SOLEIL) – Postdoc & cell design scope at SOLEIL
10h20 – 10h40	Nataliia Mozhhuzkhina (Chalmers) – Goals with the cell design and advancements so far
10h40 – 11h00	Xinyu Li (DTU) – *Operando* XRD on LNO
11h00 – 12h00	**Goal 1** Summary of the experimental requirements/constraints around each technique Establishing a global technique sheet
12h00 – 13h00	Lunch break (ESRF canteen)
13h00 – 13h30	Introduction to the cells in BIG-MAP (short presentations) – 5 to 10 min for each person participating that has a cell for BIG-MAP
13h30 – 15h00	Hands-on discussion with existing *operando* cells (bring physical examples)
15h00 – 15h30	**Goal 2** Summary of the cell requirements (dimensions, materials, versatility…) Establishing a global cell spec sheet
15h30 – 16h00	**Goal 3** Summary of the discussion and extract decisions/guidelines/specs for BIG-MAP cell Validation of the suggested prototype

2.5.2 Assessing the reproducibility of sample preparation, shipment, and washing procedures

Experimental plans described above require, i) reproducible *ex situ* sample preparation, i.e., coin cell (dis)assembly, ii) sample shipment without degradation, iii) reproducibility between similar instruments owned by different partners. In practice, this is far from being trivial. Indeed, different labs have different ways of preparing coin cells storing and shipping samples, some being more effective than others depending on the type of sample. Also, sample preparation before analysis can vary and hence lead to different results (sample washing procedure before surface analysis, transfer chamber for highly air-sensitive samples etc.).

Ex situ sample preparation is well documented by the work of WP8, describing general standards and protocols in terms of battery assembling and disassembling (D8.4). Moreover, most WP5 partners can fabricate coin cells in standard conditions after attending to the battery manufacturing training organised by WP8. Figure 4 shows typical LNO/graphite coin cell cycling results compared between WP8 and WP5 partners (namely the CSE and the CEA). A decent agreement when cycling in standard conditions is achieved (panel a), while a small deviation is observed at a higher cut-off voltage (panel b). Beyond coin cell preparation, more specific procedures will be developed in WP5 regarding sample washing and shipping. Along that line, we are testing an *ex situ* sample workflow for surface characterisation (Figure 5). Surface characterisation techniques have the disadvantage of being quite sensitive to sample preparation, conditioning, and possible bias from characterisation (beam damage). The idea is to have four different partners (CSIC, Chalmers, CNRS, and CEA) prepare several cycled graphite electrode samples and ship them to five partners measuring XPS (Uppsala, CNRS, CEA) and FTIR (CSIC, Chalmers). The electrochemical data, together with the XPS and FTIR spectra, will be used to assess the reproducibility. Depending on the results, sample preparation conditions might be refined or the procedure extended to other instruments.

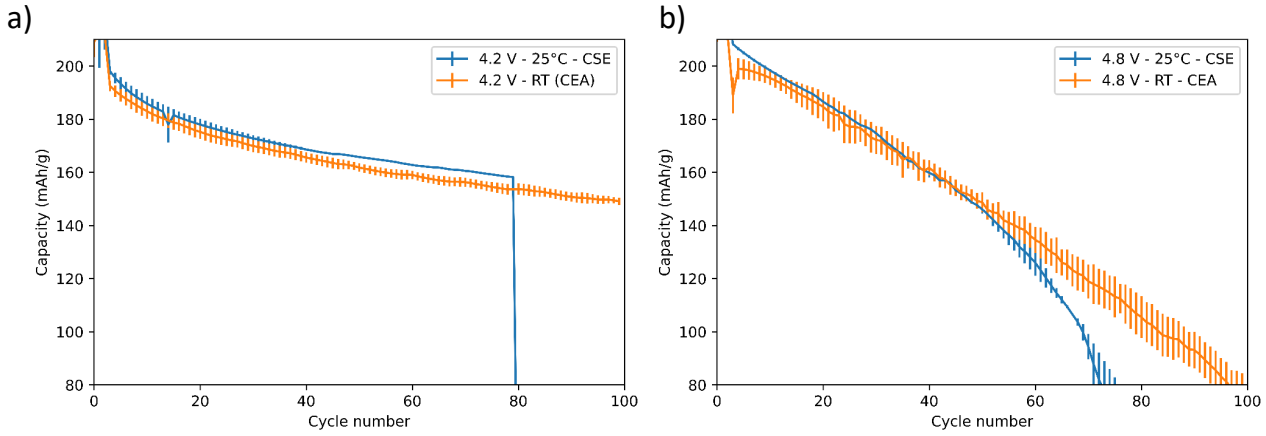

Figure 4. Electrochemical performance of LNO/Graphite cell (3mAh.cm²) using LP57 prepared.

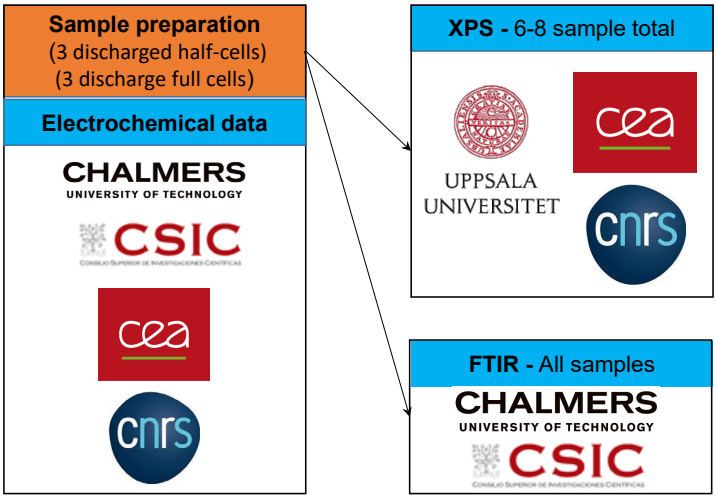

Figure 5. *Ex situ* workflow, sample preparation, and data acquisition.

2.5.3 Accelerating the LSFs workflows

The upgrade of sources, optics, detection systems, as well as the continuous improvement of ultra-specialized instruments and beamlines at LSFs, have allowed significant advances in battery *operando* characterization by pushing experimental limits in terms of sensitivity and resolution. An array of instruments is available at synchrotron and neutron facilities, each being optimized in general to monitor one or two key characterizations. Diffraction and scattering techniques are employed to observe the atomic ordering/crystal/nanoscale structure; spectroscopy techniques allow us to observe the redox valence changes, local dynamics, and local structure; and imaging and tomography techniques probe inhomogeneities and/or distributions of density, redox valence and mesoscale structures.

Battery experiments performed at LSFs usually require access to a dedicated beamline/instrument to probe one (or occasionally several) parameters of specific interest. Research is conducted using standard access modes based on proposal submission where single-shot experiments are the general rule, hence producing high-quality data focalized on one specific sample environment and targeting a specific scientific question at a given length scale (atomic, particle, electrode, or full device). The single-technique experimental workflow usually involves several stages extending over a significant period of time, from the nucleating idea to proposal submission, expert peer-group review, the scheduling and carrying out of the granted experiment, subsequent data analysis, and the final publication of results. To date, it takes ca. more than 6 months between proposal acceptance and further data collection and more than 1.5 years on average before data interpretation/publication of the collected data. Moreover, correlations with modelling, databases, and Artificial Intelligence (AI) are usually limited to the very early (design of an experiment) or the very final (discussion about output parameters) stage of the linear sequential process. Finally, the realization and interpretation of one experiment usually imply decision-making operators, advanced human expertise, and manual operations because data acquisition and its subsequent analysis/interpretation are seldom automatized.

In order to progress beyond single-parameter and/or single-scale investigations, some synchrotron beamlines and neutron instruments offer the possibility of multi-probe characterizations, e.g., to

simultaneously measure XAS and XRD,[1] SAXS and WAXS,[2] SANS and lab-SAXS,[3] NCT and lab-XCT,[4] thus allowing access to co-registered, complementary data in a single-shot (e.g., both morphological and chemical information) and/or covering the multiple length-scales of interest. However, as the situation stands today, most multi-technique studies generally rely on sequential access to different instruments. These experimental workflows are still subject to those bottlenecks encountered in single-technique experiments. Additionally, the timeframe may even be extended due to the difficulty in developing multi-technique-compatible sample preparation procedures, coordinating access to several instruments, the collection/processing/storage of several datasets, and their subsequent correlative analysis - a process requiring relatively recent, non-trivial solutions and mostly limited to statistical and/or qualitative combinations. Therefore, the coupling/correlation and/or combination of LSF experiments remains uncommon, and multi-technique methodologies and workflows for correlative data acquisition and analysis are still in their infancy.

Recently, we have discussed the paradigm shift towards a robust correlative characterization approach in batteries requiring novel types of workflows designed to tackle LSF-specific bottlenecks[5]:

1) The linear sequence of actions extending over months/years to prepare, realize and analyze a specific experiment, including the time-limited access to specific instruments,
2) The active presence of expert users at each step of data collection and analysis. This includes writing the logbook, hence not standard metadata format.
3) The availability of standardized LSF-compatible battery cells to perform *operando* correlative characterization,
4) The generation of big and diversely formatted data volumes,
5) The transfer of results to the research community and accelerated return-on-investment to battery R&Ds.

The automatization and standardization of multi-technique correlative experiments are fundamental to meeting these challenges. Accelerating 1) and 2) requires the implementation of modern tools such as active learning and DL/ML modules, as well as on-the-fly diagnostic and fast feedback loops, ultimately enabling the online control of data acquisition and experimental set-up selection. Accelerating 2) and 4) requires expert databases/repositories, centralized platforms, and apps, new software capable of handling multidimensional datasets and performing autonomous correlative data analysis. Accelerating 1) and 5) requires new access modes to enable fast, reactive, and flexible beamtime allocation, scheduling, and use, ultimately leading to the coordination of multi-site experiments carried out on the same material/device and under the same operational conditions, which implies the development of standardized cell designs and operating procedures, as well as new infrastructures.
A novel type of integrated workflow was proposed (see Figure 6) for correlative (multi-modal, multi-technique) LSF-based research incorporating accelerators acting along the whole experimental chain. These include four key innovations:

1) Standardization of battery cells for multi-modal correlative *operando* characterization,
2) Automated data acquisition systems and standardization of protocols and data management,
3) Dynamic correlative analysis based on AI modules and intelligent batteries, and
4) New access modes to LSF, eventually with interoperable infrastructures.

Those are key aspects investigated and tested in BIG-MAP WP5. The in-lab and LSF workflow must integrate not only the optimized operation of the many technical and scientific means available in the consortium but also define modalities for new interoperable infrastructures enabling, for instance, new beam time access modes to improve global experimental workflow efficiencies and their practical implementation in user-case defined studies.

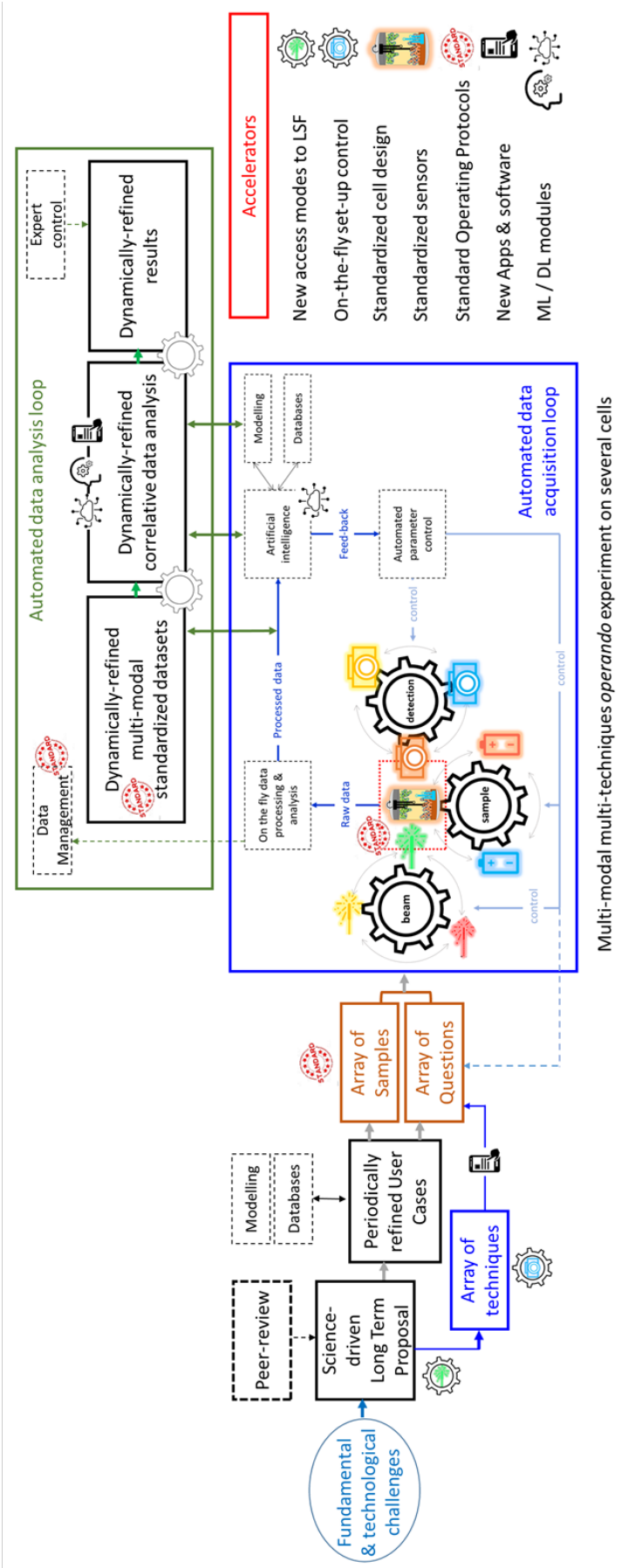
Accelerators
New access modes to LSF
On-the-fly set-up control
Standardized cell design
Standardized sensors
Standard Operating Protocols
New Apps & software
ML / DL modules
Automated data analysis loop
Expert control
Dynamically-refined results
Dynamically-refined correlative data analysis
Dynamically-refined multi-modal standardized datasets
Data Management
Automated data acquisition loop
Modelling
Databases
Artificial intelligence
Feed-back
Automated parameter control
Processed data
Raw data
On the fly data processing & analysis
control
detection
sample
beam
Multi-modal multi-techniques operando experiment on several cells
Array of Samples
Array of Questions
Modelling
Databases
Periodically refined User Cases
Array of techniques
Peer-review
Science-driven Long Term Proposal
Fundamental & technological challenges

Figure 6. Multi-modal multi-technique correlative characterization workflow, where data acquisition (blue box) and analysis (green box) are accelerated by implementing on-the-fly monitoring, online data acquisition feedback loops, dynamically-refined storage, and processing of standardized datasets by correlative analysis. The automated data acquisition loop allows to control and modify on request the beam characteristics and experimental conditions as well as the sample (change the region of interest in the probed battery, and/or move to next battery; ultimately, propose new materials combinations and robotically fabricate new cells). The workflow relies on the development and integration of accelerators applied during all key steps: from new access modes to LSF to new standards for battery and set-up protocols, apps, software, and ML/DL modules. The as-designed *operando* experiment provides high-throughput (multiple-sample data in fast mode) and/or high-fidelity data (selection of best conditions by automated loops for high-resolution / high-density mapping of selected parameters). The human expertise-driven decisions and actions are limited to the verification of automatisms and post-experiment analysis control.

3. Conclusions

In D5.5 we have presented the architecture of an automatized experimental workflow designed to operate a European experimental platform dedicated to battery research by defining/applying new concepts and tools to the BIG-MAP cases and innovations.

The practical implementation of such workflow relies on online laboratory notebook usage and development and specific search and application tools, e.g., dedicated apps or modules. This requires tight connections to WP8 and WP9-10-11 of BIG-MAP and a central orchestration of the requests/actions and will be further explored in the second period of the project.

By now, the required concepts and organization have been identified integrated into the proposed mechanism for defining and realizing experimental plans according to quality/fidelity/feasibility criteria. A specific application of the workflow is currently ongoing regarding *ex situ* electrode characterization (based on expert-decision making) and will be used as beta-testing to continuously refine the methods and target large-scale automatization.

4. References

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