Ref. Ares(2025)4368706 - 30/05/2025

OPINCHARGE

PROJECT
PoC SIMS imaging

05/2025

Funded by
the European Union

Lead Beneficiary	Luxembourg Institute of Science and Technology
Author(s)/Organisation(s)	Andrés Miranda Martínez (LIST), Santhana Eswara (LIST)
Contact Email	andres.miranda@list.lu
Contributor(s)	Tim Dahmen, Akarsh Sudheendra Bharadwaj
Work Package	WP4, Task 4.3 Integration of Adaptive Sampling and reconstruction algorithms in FIB-SIMS chemical imaging and benchmarking of performance.
Delivery Date (DoA)	30/06/2025
Actual Delivery Date	27/06/2025
Abstract:	FIB-SEM/SIMS is a promising tool to study the physical and chemical properties of battery material, Lithium in particular. The main challenge to overcome is the damage caused to Lithium due to radiation. In this context, one way to sort out this situation is to develop advanced low-dose, high-speed imaging methods for FIB-SEM/SIMS. These methods allow for high-resolution imaging of radiation sensitive samples, such as Lithium, by reducing beam damage and acquisition time. The instrument integrates a Ga-FIB column, and a magnetic SIMS module, both controlled by an in-house software developed in LabVIEW and Python. The software provides users control over scanning parameters like image size, dwell time, and sparse percentage This will execute the sparse scanning method that allows low-dose high-speed acquisition by scanning less pixels from the image. The purpose of this document is showing the validation and results obtained with the FIB-SEM/SIMS system for low-dose, high-speed imaging.

Document Revision History			
Date	**Version**	**Author/Contributor/Reviewer**	**Summary of Main Changes**
25/05/2025	V1.0	Andrés Miranda Martínez	First Version
28/05/2025	V2.0	Andrés Miranda Martínez, Santhana Eswara	Second Version

Dissemination Level and Nature of the Deliverable		
PU	Public	No
SEN	Sensitive, limited under the conditions of the Grant Agreement	SEN
Nature	R = Report, E = Ethics or, O = Other	O

OPINCHARGE Consortium			
#	Participant Organisation Name	Short Name	Country
1	LUXEMBOURG INSTITUTE OF SCIENCE AND TECHNOLOGY (LIST)	LIST	LU
2	CENTRE NATIONAL DE LA RECHERCHE SCIENTIFIQUE CNRS (CNRS)	CNRS	FR
3	CENTRO DE INVESTIGACION COOPERATIVA DE ENERGIAS ALTERNATIVAS FUNDACION, CIC ENERGIGUNE FUNDAZIOA (CIC ENERGIGUNE)	CIC ENERGIGUNE	ES
4	DEUTSCHES ZENTRUM FUR LUFT - UND RAUMFAHRT EV (DLR)	DLR	DE
5	DEUTSCHES FORSCHUNGSZENTRUM FUR KUNSTLICHE INTELLIGENZ GMBH (DFKI)	DFKI	DE
6	FEI ELECTRON OPTICS BV (THERMO)	THERMO	NL
7	FUNDACION CIDETEC (CIDETEC)	CIDETEC	ES
8	UNIVERSITAET PADERBORN (UPB)	UPB	DE
9	PEDAL CONSULTING SRO (PEDAL)	PEDAL	SK
10	PAUL SCHERRER INSTITUT (PSI)	PSI	CH
11	FORSCHUNGSZENTRUM JÜLICH (FZJ)	FZJ	DE

NOTICE

Funded by the European Union under grant agreement number 101104032 . Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or European Climate, Infrastructure and Environment Executive Agency (CINEA). Neither the European Union nor the granting authority can be held responsible for them.

OPINCHARGE

Grant Agreement: 101104032 .

Call: HORIZON-CL5-2022-D2-01.

Topic: HORIZON-CL5-2022-D2-01-02.

Type of action: HORIZON Research and Innovation Actions.

Granting authority: European Climate, Infrastructure and Environment Executive Agency.

Grant managed through EU Funding & Tenders Portal: Yes (eGrants).

Project starting date: fixed date: 1 June 2023.

Project end date: 31 May 2026.

Project duration: 36 months.

Table of Contents

List of Figures

List of Abbreviations and Acronyms	
DAQ	Data Acquisition board
DT	Dwell Time
ESI	External Scan Interface
ETD	Everhart-Thornley Detector
FIB	Focused Ion Beam
FPD	Focal Plane Detector
Ga	Gallium
Li	Lithium
LMIS	Liquid Metal Ion Source
MPC	Microscope Personal Computer
SEM	Scanning Electron Microscopy
SIMS	Secondary Ion Mass Spectrometry
SNR	Signal to Noise Ratio
SPC	Support Personal Computer
TCP	Transmission Control Protocol
TDC	Time to Digital Converter
TIC	Total Ion Count
UI	User Interface

1 Introduction

Traditional scanning approaches in electron (or ion) microscopy are not suitable for observing dynamic material processes when a high-speed low dose scanning is required. Simply decreasing the dwell time (DT) per pixel is not a viable solution, as it reduces the signal-to-noise ratio (SNR), which will cause a degradation in image resolution. Additionally, the radiation dose delivered to the sample plays a critical role; excessive exposure can damage the sample's structure and deteriorate image quality. Hence, precise dose management is crucial for ensuring reliable sample analysis [1].

As an alternative to conventional scanning processes the alternative sparse scanning techniques are a good option to study. From the possible methodologies, random sampling stands out [2]. This method helps reduce the acquisition time and dose by scanning a specific percentage of pixels. This methodology involves a trade-off between reducing acquisition time (which lowers the dose) and maintaining image resolution. On the other hand, the use of a random scanning approach requires the implementation of an image reconstruction algorithm to recover the information lost from the unscanned pixels. The improvement in image resolution will be according to the effectiveness of this reconstruction algorithm.

The significance of this development lies in its application to the study of radiation-sensitive materials, such as lithium. If we aim to analyse the structural and chemical changes occurring in lithium during an operando analysis, it is essential to use a methodology that minimizes the radiation dose delivered to the sample.

This document describes the implementation of a sparse sampling methodology [2] for focused ion beam (FIB) – scanning electron microscopy (SEM) and FIB-SIMS (FIB-SEM-SIMS), allowing us to reduce acquisition time and dose into the sample. Section 2 below describes the methodology, the hardware, and the software; in section 3, the results are presented. Finally, Discussion and conclusions are shown in section 4.

2 Hardware and Software

The instrument used is the Scios form Thermo Fisher. This is a platform that contains several modules that extend its capabilities. For the purposes of this document, only the relevant modules will be mentioned. The Scios is a dual beam electron microscope, equipped with a SEM column, and a Ga liquid metal ion source (LMIS)-based FIB column (Ga-FIB). It contains one secondary electron detector Everhart-Thornley (ETD) and a magnetic sector SIMS (Figure 1) [3]. Internally, the SIMS contains a Focal Plane Detector (FPD) where the ions hits at a specific distance depending on their mass. The readout from the FPD is done by a Time-to-Digital Converter (TDC), which converts analog ion arrival information into digital data with high time resolution, and that digital information is sent to the Support PC (SPC).

Two PCs are used: the Microscope PC (MPC) where the Scios is typically manipulated, and the SPC which is a secondary PC where our software is developed and executed.

The Ga-FIB is controlled by a deflection system. To have control of it, an External Scan Interface (ESI) is used to switch the control of the instrument from the MPC to the SPC.

A Data Acquisition board (DAQ, NI USB-6351) board from National Instruments is used to send two analog signals that control the deflection system. Each signal moves the beam along the X and Y axis.

Funded by the European Union

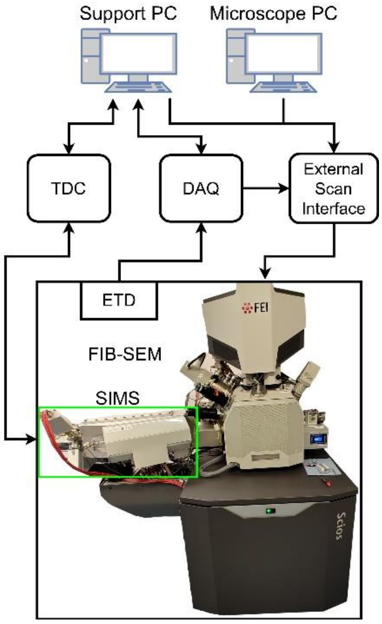

Figure 1. Instrument (Scios) set-up

The software is developed mainly in a graphical programming environment called LabView from National Instruments. Python code is also used to generate the sparse locations that will be scanned after.

For the SIMS data acquisition, scan with different scan patterns and sparse sampling scanning is possible.

Both the user interface (UI) and the software backend were developed based on previously created code for the general control of the instrument.

An example of the UI can be observed in figure 2. The user can set parameters such as image size, DT, and scan pattern for the acquisition. For the sparse sampling case, it is possible to select the sparse percentage (25, 50, and 75%) and if the stratified random sampling will be applied or not. Once the image is taken, it is displayed, and it can be adjusted in terms of brightness and contrast. Information as the acquisition time and histogram is shown too.

The methodology is as follows: the user defines the parameters of the image to be. After selecting the acquisition parameters, the user defines the name and location to save the data within the acquisition controls. Once the name and location for saving the information are defined, clicking the "Acquire" button starts the image capture. Then, the program generates two digital waveforms that are converted into analog signals by the DAQ and the FIB will start illuminating the sample. If it becomes necessary to stop the capture, the "Stop" button will interrupt the measurement. Once the process is completed, the program saves all the data automatically. The acquired image, along with its histogram, are displayed in the interface. The image can be adjusted using two different scales (logarithmic and linear) as well as brightness and contrast through the "Image Adjustments" controls.

The generated data consists of a comma-separated values (.csv) file, an image (in TIFF format), and an additional text file. The .csv files store the acquisition parameters and the total measured values (count) for each pixel in all channels, while the extra text file contains a detailed list of each pixel's position along with the number of events obtained per pixel during the measurement. All these files are saved in the SPC.

Funded by the European Union

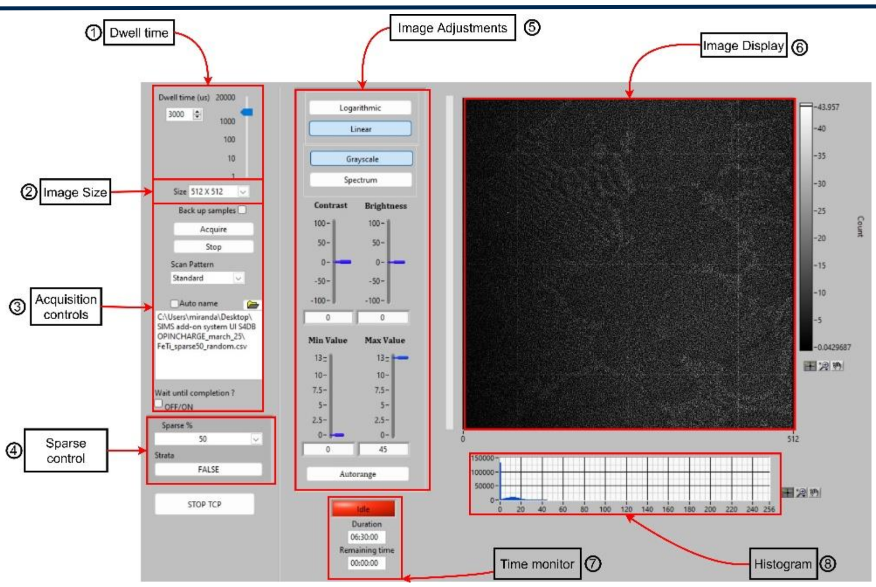

Figure 2. User interface. (1) Dwell time selector, (2) Image size selector, (3) Acquisition control panel, (4) Sparse control panel, (5) Image adjustments panel, (6) Image display, (7) Time monitor, and (8) Histogram display

To make a sparse scan, the software needs to communicate with the python algorithm (provided from DFKI tasks 4.1 & 4.2). Once we start the scan, the LabView program sends the image size, sparse percentage, and a true/false value (to select if we will do a stratified scan) to python using TCP connection. The python code will use this information to generate two arrays to form the coordinates of the pixels to be scanned. After this, python will send back to LabView these arrays to continue the scan process. Figure 3 shows the workflow of this process.

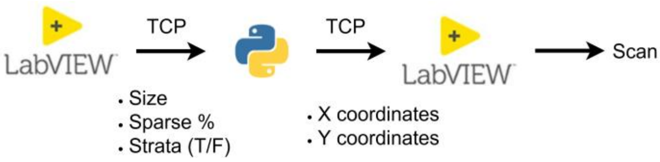

Figure 3. Communication between LabView and python to achieve Sparse Sampling acquisition.

To assess pros and cons of this methodology, the scan time (acquisition time) and the resolution of the images where obtained. The resolution was estimated using the radial profile from the 2D-FFT of each image. Then, resolution is defined as the frequency at which the intensity decreases by 86.5%. Scan time is calculated with the image size and the DT.

3 Results

When working with SIMS data, it is essential to process the data to isolate the specific element(s) of interest. This involves selecting the appropriate spectral channels corresponding to the mass of that element. By defining these channels, we can generate an image that shows the chemical information of the sample, revealing the distribution of the element across the image. In this document, the channels that correspond to Ti were selected to show the Ti distribution in the sample (Ti including all its isotopes).

The sample used to test the scan functionalities in the SIMS system is an alloy with Fe and Ti. We obtained different images collected from the FPD.

First, we scanned the sample by using different patterns (continual, spiral, Hilbert, z-curve, and fixed sparse (figure 4). This allows us to have a better understanding of both the FIB control and data acquisition in the SIMS system. The acquisitions were made with the following settings: 30 keV Ga^+ primary ions, ion current = 50 pA, SIMS polarity: positive, DT = 1 ms, and image size = 512 x 512 pixels. Figure 5 illustrates the histograms for each acquisition. The distributions for most of the cases are similar except for the sparse one, which contains a higher amount of black pixels (value = 0).

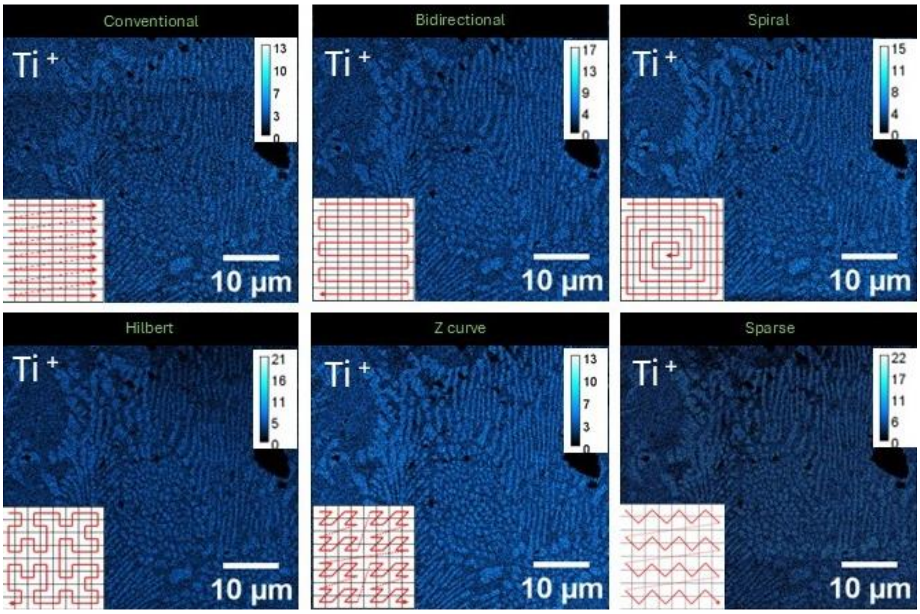

Figure 4. Ti^+ FIB-SIMS images captured using different scan patterns.

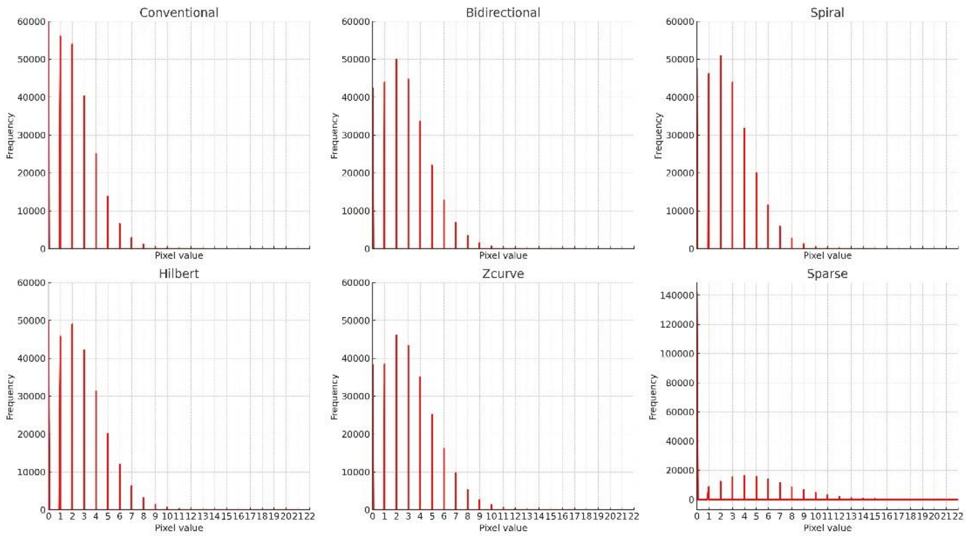

Figure 5. Histograms for every scan in figure 4.

Figure 6 shows the results of the acquisition using the random sampling technique using the same sample. The acquisitions were made with the following settings: 30 keV Ga^+ primary ions, current = 50 pA, SIMS polarity: positive, DT = 3 ms, and image size = 512 x 512 pixels. Three sparse percentages were used: 25, 75, and 75 %, this means the number of pixels scanned per image. For each, stratified random was also scanned. A full scan was acquired using a DT that gives an equivalent dose (and acquisition time) to each sparse percentage. These DT were: 750 µs, 1.5 ms, and 2.25 ms.

A ground truth full image was obtained as a reference using (figure 7).

Figure 8 illustrates the resolution (blue bars) and scan time (red bars) for each image showed in figures 5 and 6. On the left side, the data from the sparse images is ordered from less pixels scanned to all

Funded by the European Union

pixels scanned. It is also specified if the technique used was random sampling or stratified random sampling. The full scans are ordered from the minor to the highest DT (right side of the graph))

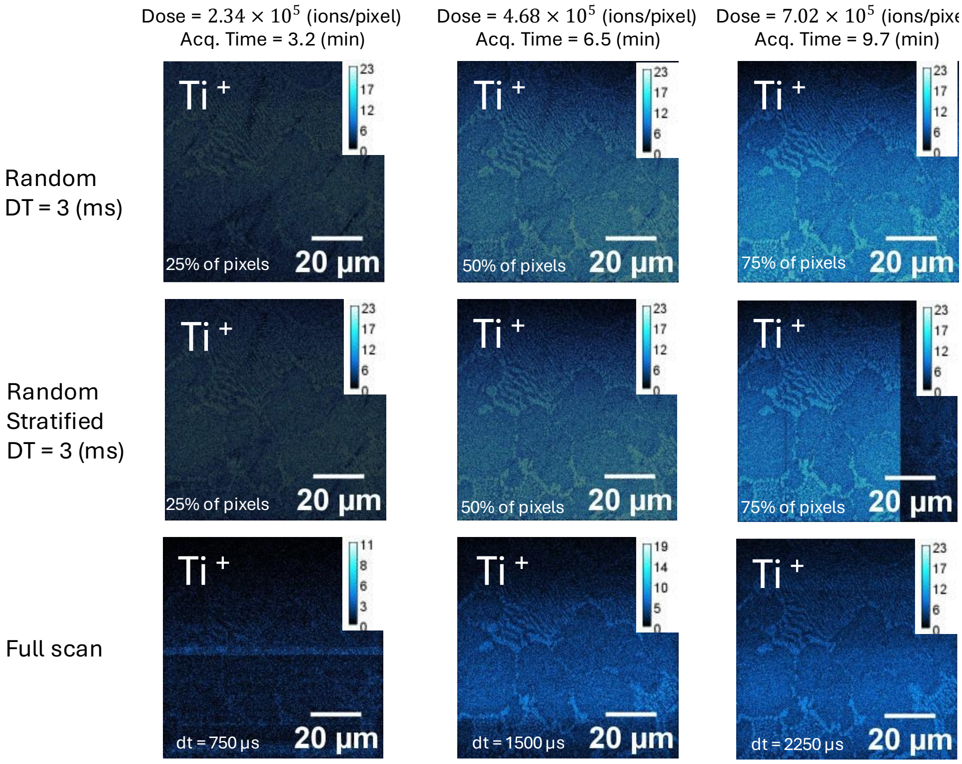

Figure 6. Ti⁺ FIB-SIMS images captured by applying the random sampling scan.

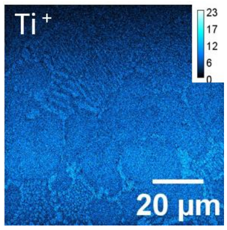

Figure 7. Ti⁺ FIB-SIMS full scan image. DT = 3 (ms). Dose = 9.36×10^5 (ions/pixel)

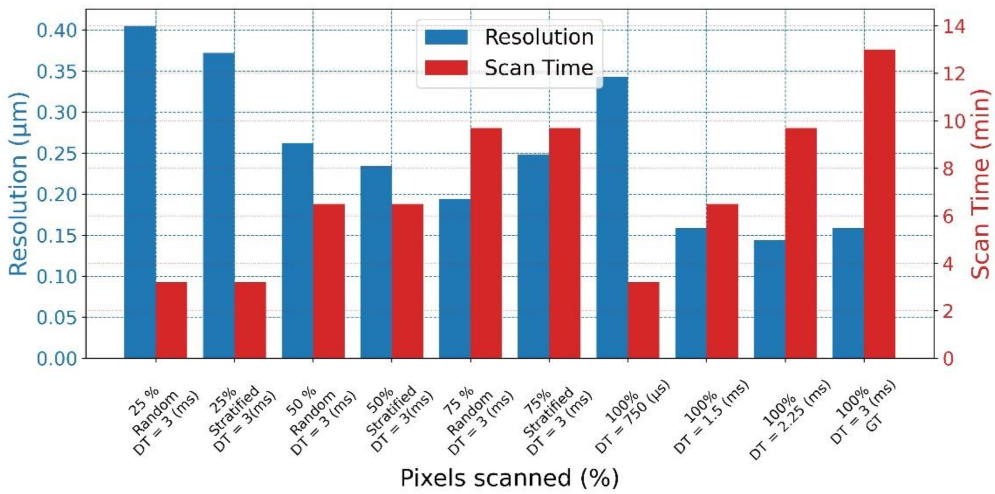

Figure 8. Comparison between resolution and scan time for each case.

Funded by the European Union

4 Discussion and conclusions

The results show significant advancements in low-dose, high-speed image acquisition for FIB-SIMS imaging. With this, the study of the physical and chemical properties of Lithium will become accessible. The sparse sampling technique was successfully implemented and tested to obtain chemical composition using SIMS imaging.

From the graphic in figure 8, it is evident that the acquisition time (therefore also the dose) is decreasing when sparse sampling is executed but sacrificing lateral resolution. When we compare each case separately based on the projected dose on the sample, we observe that performing a full scan provides better resolution than the random sampling cases. It is important to note that random sampling cases result in the loss of image information. However, this technique involves the use of a reconstruction algorithm designed to recover the lost information and enhance the compromised lateral resolution. These reconstruction algorithms are currently under development, and their progress is documented in the report titled "D 4.1. Adaptive-Sparse (AS) Scanning prototype software"

Differences in the resolution between the random sampling and stratified random sampling methodologies were not observed.

Further experimental development is required to validate the methodology with sensitive samples, such as Lithium. On the other hand, applying reconstruction algorithms in post-processing presents a highly promising solution to the trade-off resolution-acquisition time. From the document D 4.1 mentioned before, these algorithms can help to recover lost image information, restoring resolution and structural fidelity. This means that the future of high-speed- low-dose acquisition for SIMS analysis is promising. The next step is to continue with further experimentation and analysis of both methodologies: image acquisition and image reconstruction.

5 References

[1] Nicholls, D., Wells, J., Stevens, A., Zheng, Y., Castagna, J., & Browning, N. D. (2022). Sub-sampled imaging for STEM: Maximising image speed, resolution and precision through reconstruction parameter refinement. *Ultramicroscopy*, *233*, 113451.

[2] Dahmen, T., & Trampert, P. (2019). Sparse and adaptive sampling in scanning electron microscopy. Microscopy and Microanalysis, 25(S1), 29-30.

[3] De Castro, O., Audinot, J. N., Hoang, H. Q., Coulbary, C., Bouton, O., Barrahma, R., ... & Wirtz, T. (2022). Magnetic sector secondary ion mass spectrometry on FIB-SEM instruments for nanoscale chemical imaging. Analytical chemistry, 94(30), 10754-10763.

Funded by the European Union