Building a Materials Acceleration Platform (MAP) for battery materials research: Automated module for the synthesis of inorganic compounds

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MEMBER OF BASQUE RESEARCH & TECHNOLOGY ALLIANCE

1. GENERAL INTRODUCTION

1. Introduction

Energy storage scenario

Decarbonification of the society

Renewable energies need the integration of **energy storage systems** in the power grid

- The growing market of EVs is expected to further **amplify the demand of energy storage systems**.

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Batteries are enablers of the energy transition
But they still require:

- ✓ **Higher energy densities** & **power capabilities**
- ✓ Lower **cost**
- ✓ Higher **sustainability**

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High-Throughput and automation needs

Further research needed to address the urgent **demand** of **new battery materials** with **superior energy density**

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Conventional trial-and-error approach:

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Accelerate the discovery of materials!

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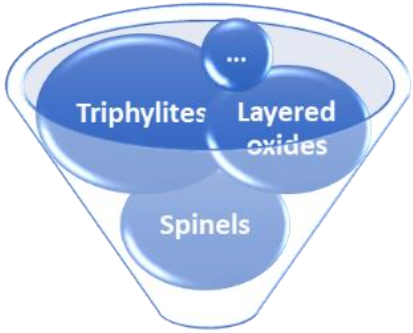
Computational screening (BVEL, …)



Experimental validation

Accelerate the discovery of materials!

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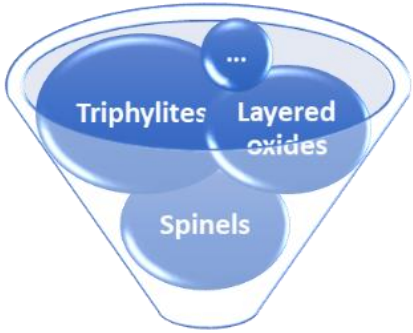

Experimental validation

Acceleration of rate of production of samples

Experimental screening of identified families

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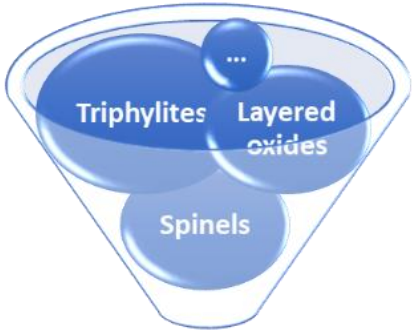

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Materials Acceleration and Innovation platform for Technologies for ENergy Applications (MAITENA)

Enable accelerated, autonomous discovery of new electroactive materials

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Automated synthesis

Solid state

Solvothermal

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- Solid state
- Solvothermal

High Throughput characterization

- XRD
- Electrochemistry

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Automated Data Management

Analysis

Interpretation

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Machine learning experiment planner

- Propose new candidates
- Optimize synthetic properties

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Closed-loop experimentation process integrating the feedback from theoretical calculations, synthesis results, and characterization data.

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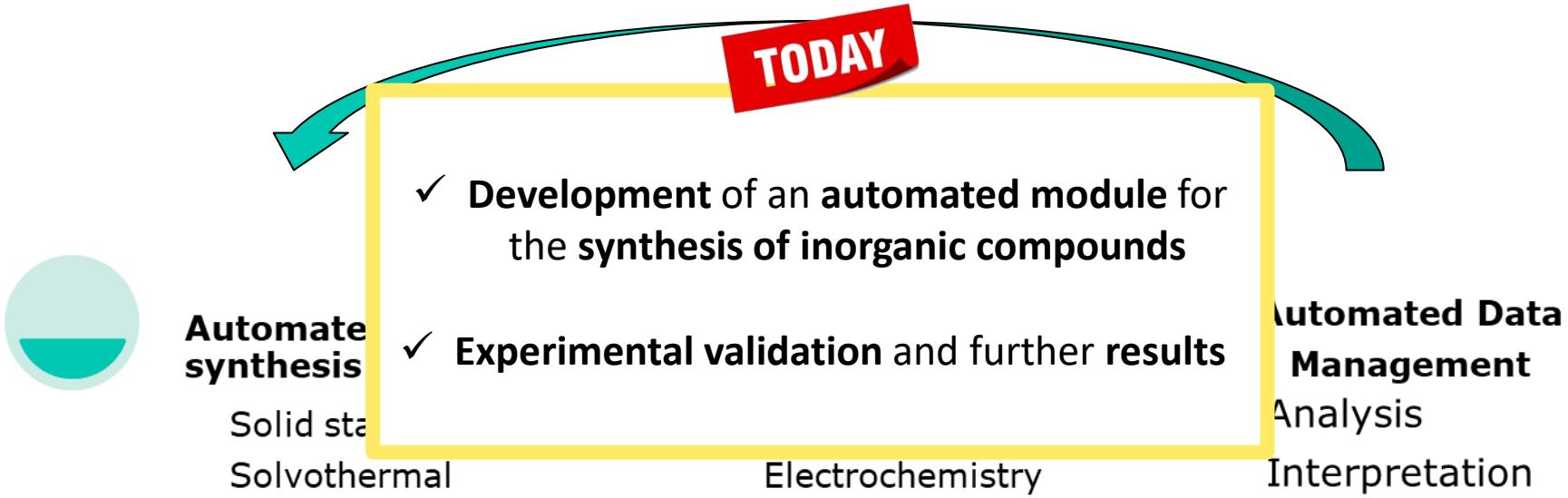
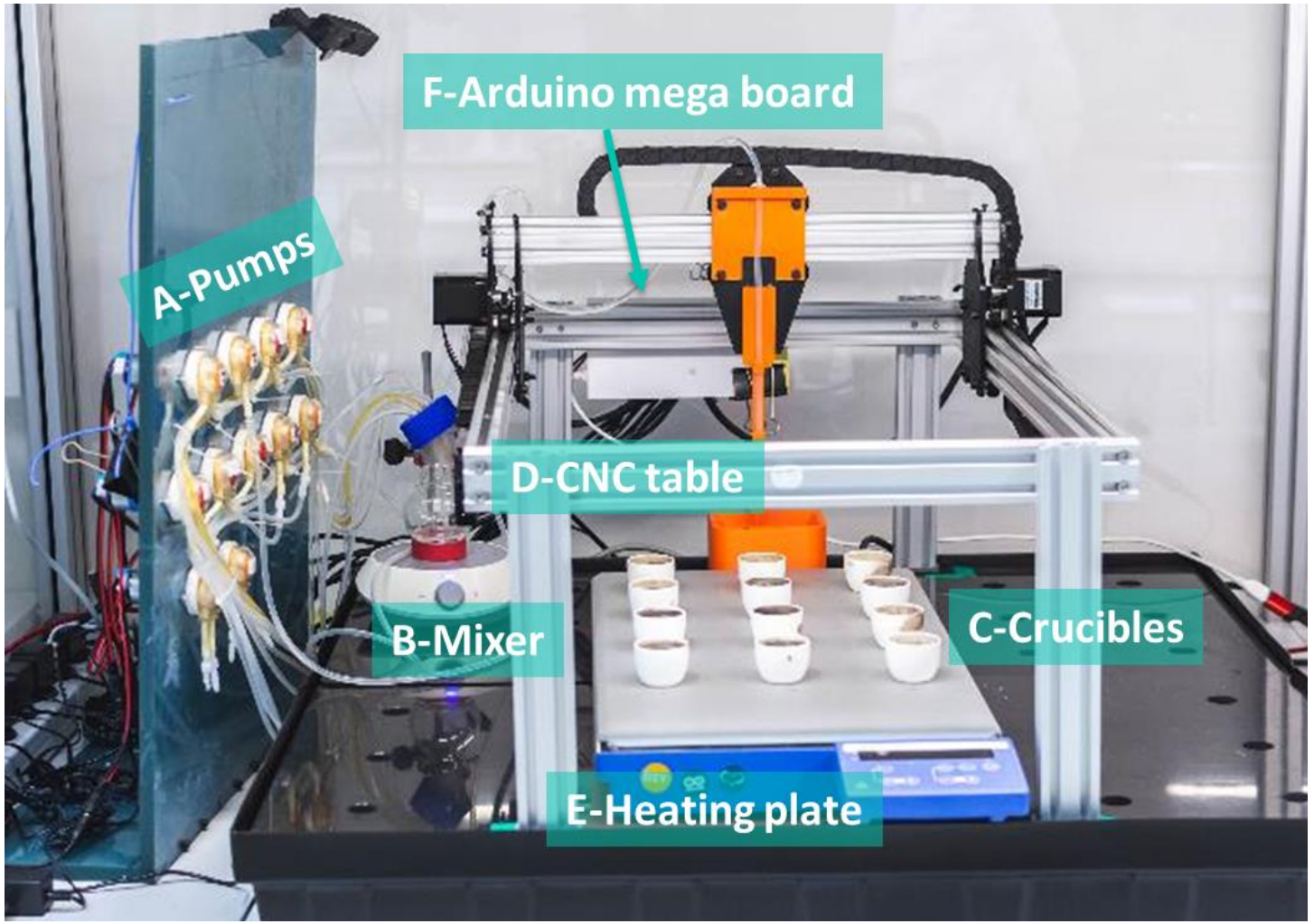


Closed-loop experimentation process integrating the feedback from theoretical calculations, synthesis results, and characterization data.

2. DEVELOPMENT OF AN AUTOMATED MODULE FOR THE SYNTHESIS OF INORGANIC COMPOUNDS

2. Automated module for synthesis of inorganic compounds



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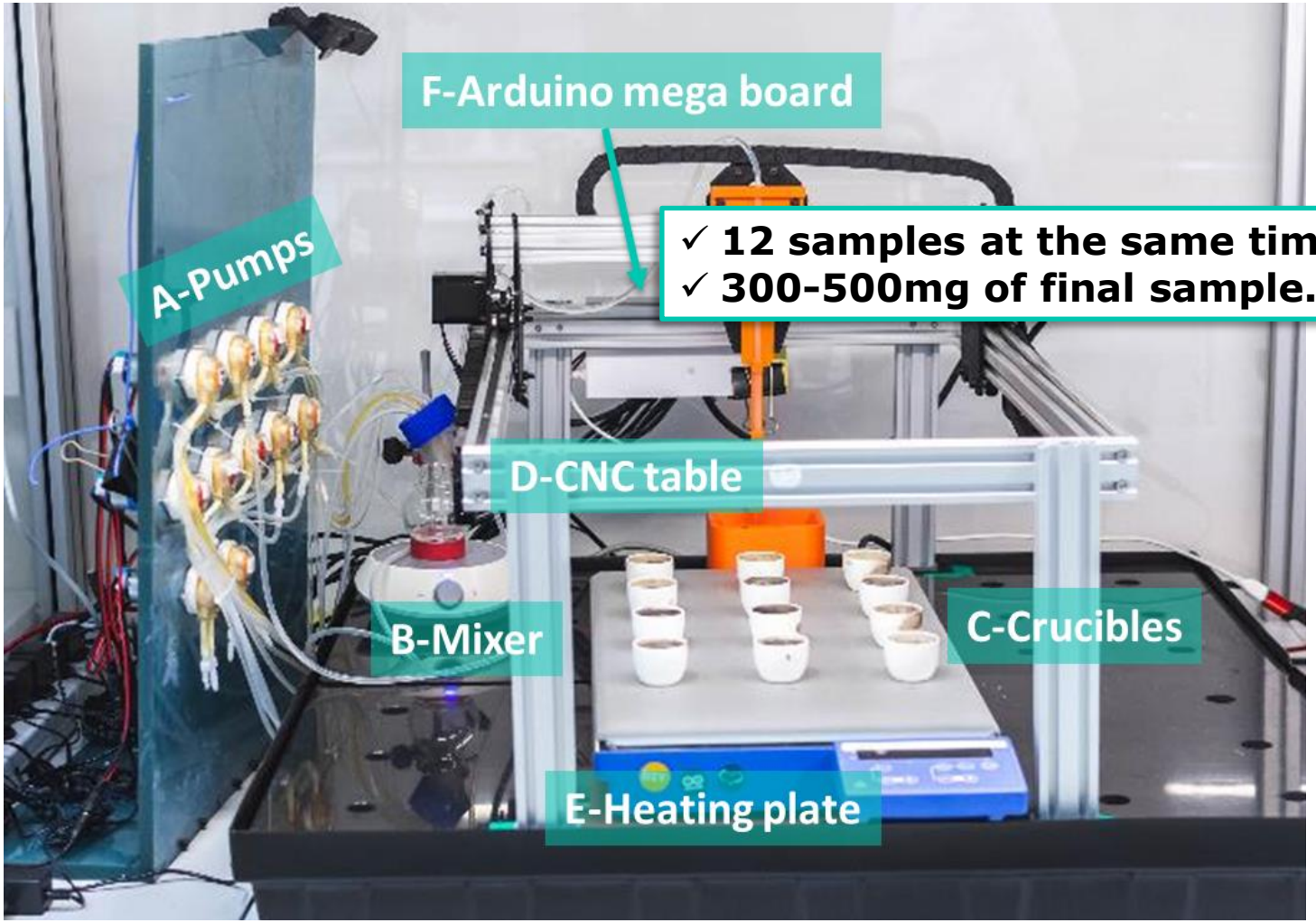

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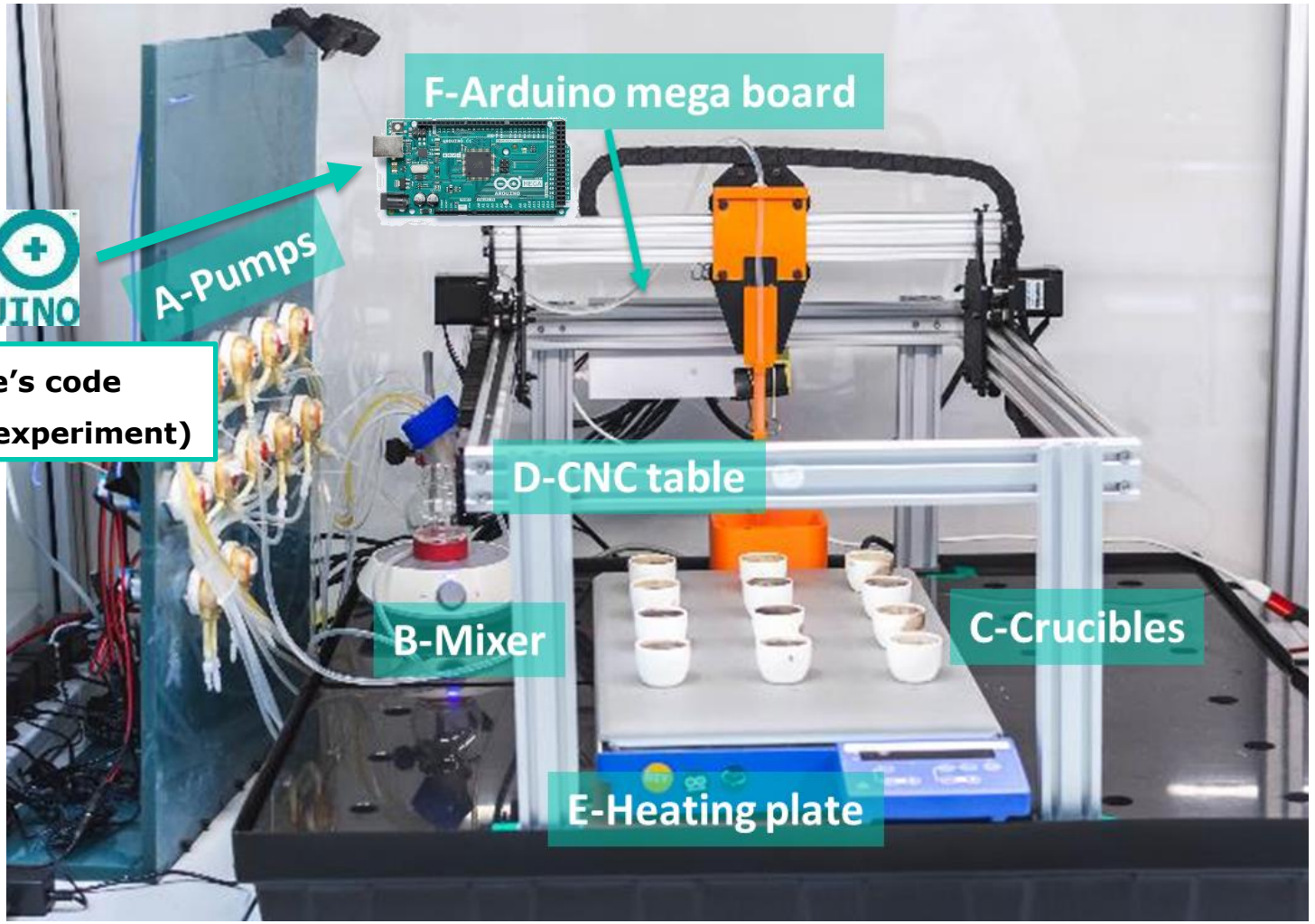

ARDUINO

Module's code (volumes/experiment)

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2. Automated module for synthesis of inorganic compounds

Module's workflow

Reagent solutions

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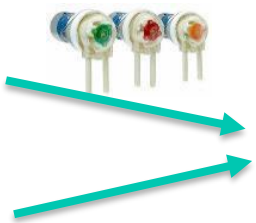
Reagent solutions



Mix of reagents

2. Automated module for synthesis of inorganic compounds

Module's workflow

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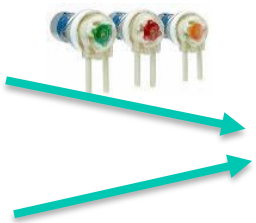

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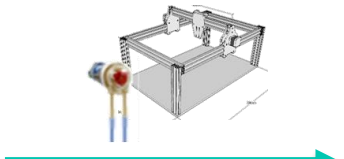
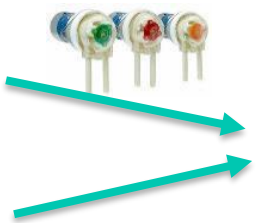


Stir

2. Automated module for synthesis of inorganic compounds

Module's workflow

Reagent solutions



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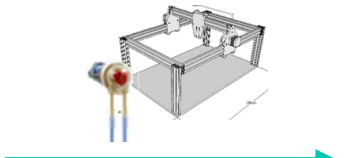

Stir

Wash the system with distilled water

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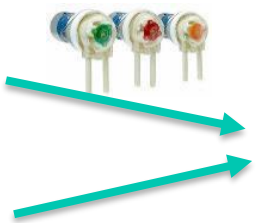

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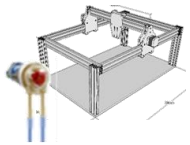


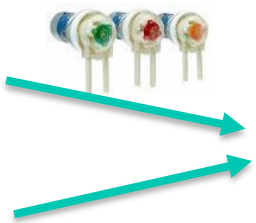
Stir

12 times – 12 crucibles

2. Automated module for synthesis of inorganic compounds

Module's workflow

Reagent solutions



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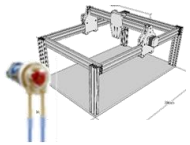


Stir

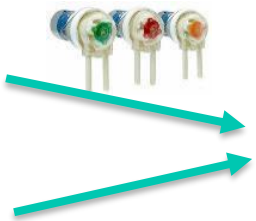
12 times – 12 crucibles

Addition of gelification reagent
(sol-gel synthesis)

2. Automated module for synthesis of inorganic compounds

Module's workflow

Reagent solutions



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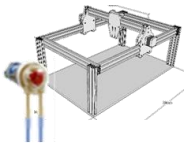


Stir

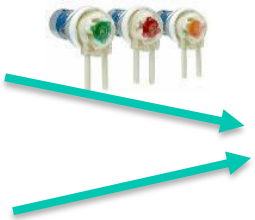
12 times – 12 crucibles

Quartz tray

2. Automated module for synthesis of inorganic compounds

Module's workflow

Reagent solutions



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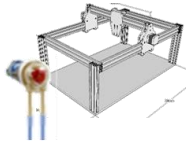


Stir

12 times – 12 crucibles

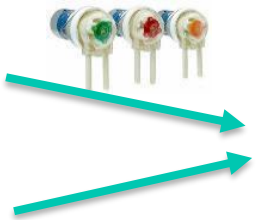
Quartz tray

Thermal treatment

2. Automated module for synthesis of inorganic compounds

Module's workflow

Reagent solutions



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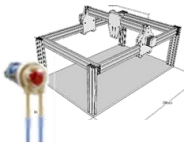


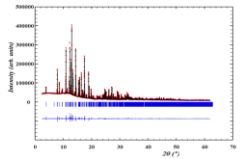
Stir

12 times – 12 crucibles

Quartz tray

Thermal treatment

300-500mg



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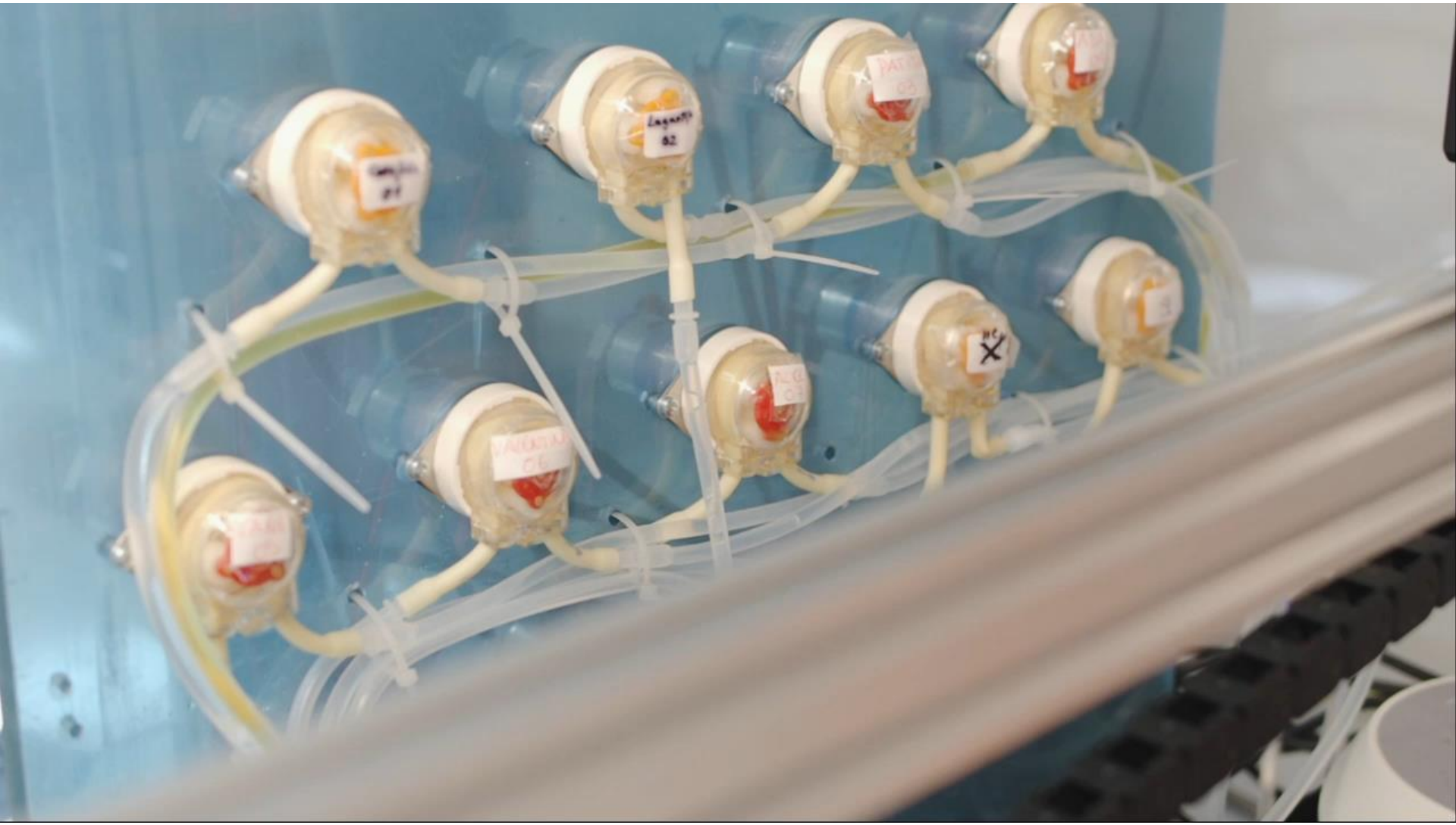

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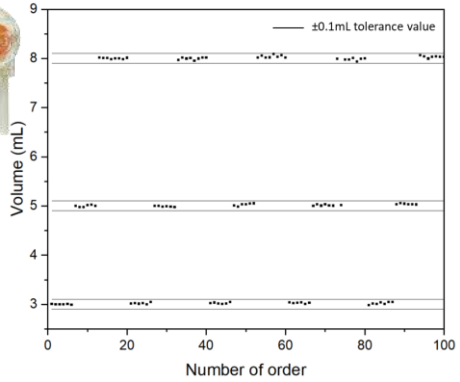
2. Automated module for synthesis of inorganic compounds

Reproducibility considerations

Pumps calibrations and durability



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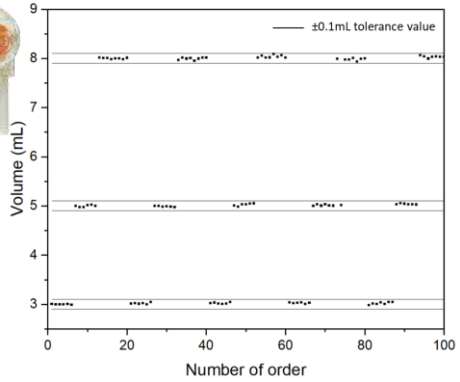
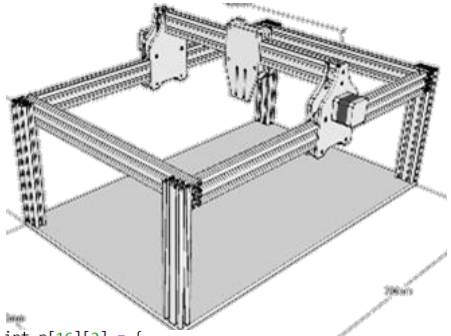


CNC calibrations



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  //              X    Y
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  /*POSICIÓN5*/  {-1480, 730},
  /*POSICIÓN6*/  {-1480, 1180},
  /*POSICIÓN7*/  {-1930, 280},
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  /*POSICIÓN12*/ {-2380, 1180},
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  /*POSICIÓN14*/ {0, 0},
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2. Automated module for synthesis of inorganic compounds

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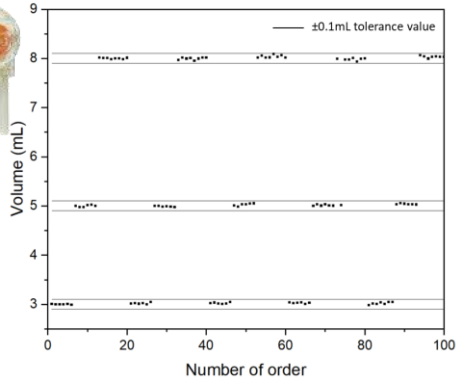
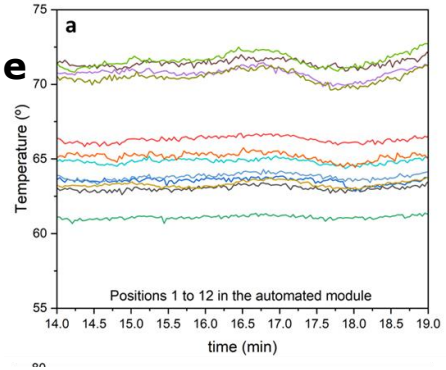
Pumps calibrations and durability



Thermal distribution of the heating plate



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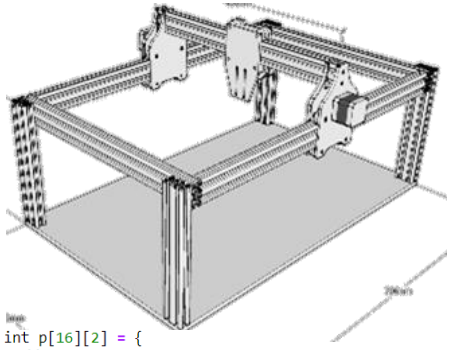

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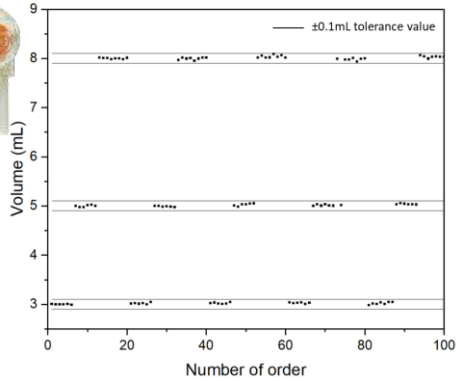
2. Automated module for synthesis of inorganic compounds

Reproducibility considerations

Pumps calibrations and durability



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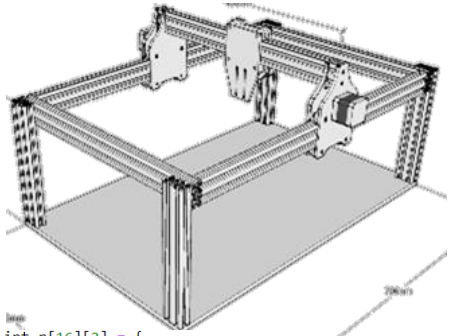

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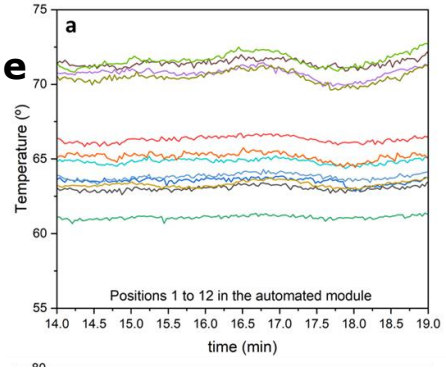
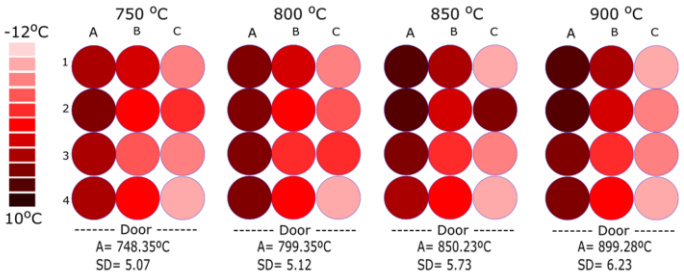
Furnace calibrations

Thermal distribution of the heating plate

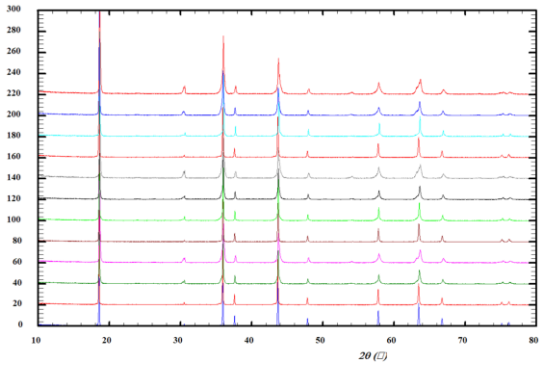




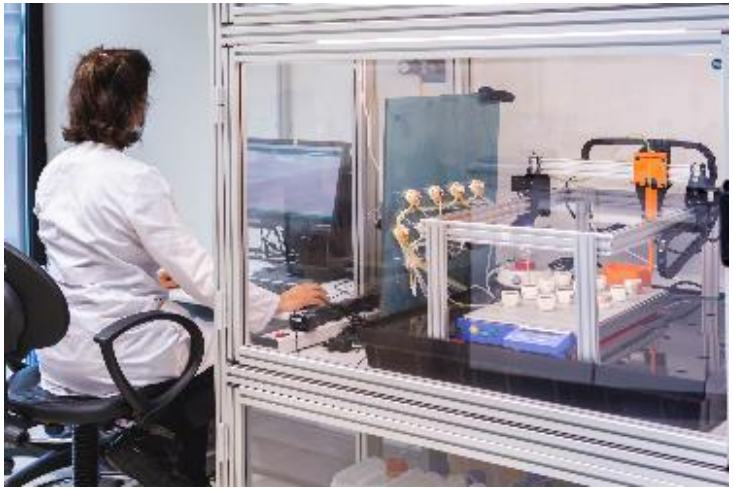
±10°C across different sample positions

2. Automated module for synthesis of inorganic compounds

High-Throughput synthesis

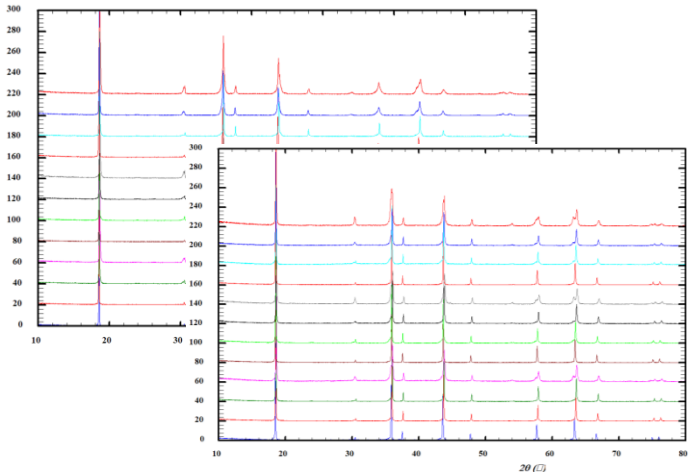


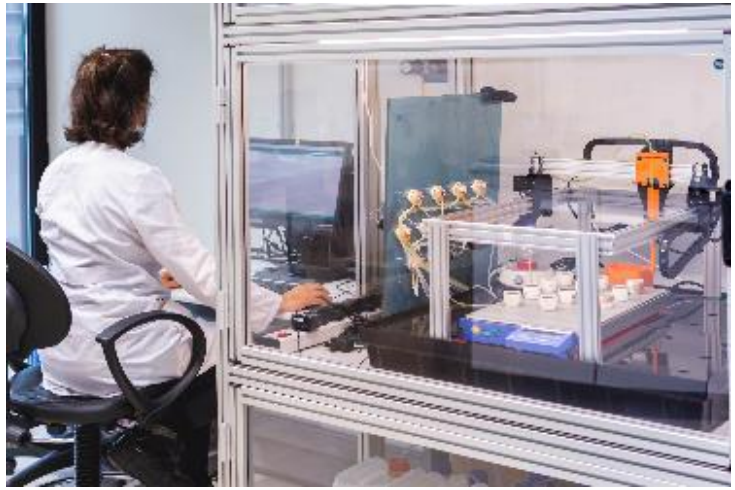
12 samples at the same time



2. Automated module for synthesis of inorganic compounds

High-Throughput synthesis





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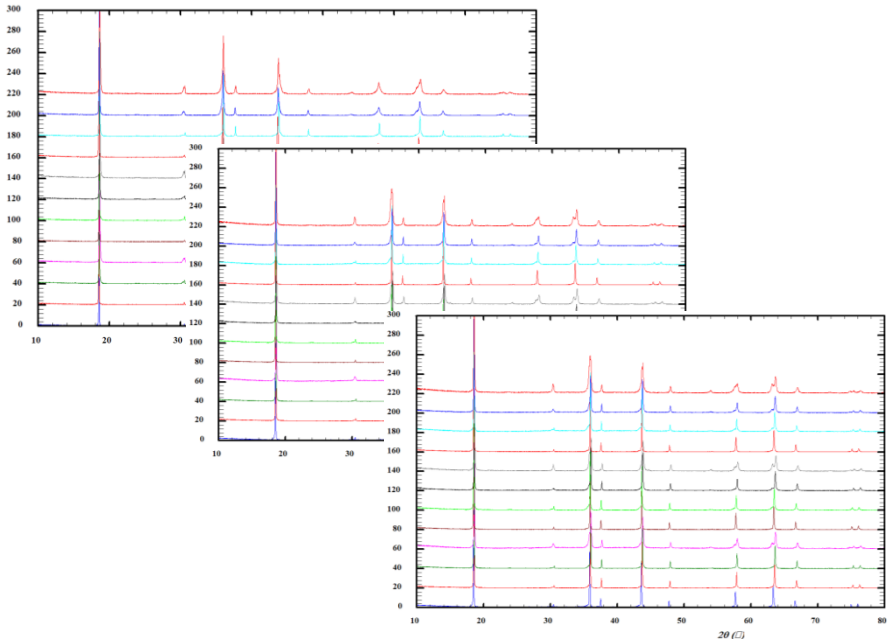

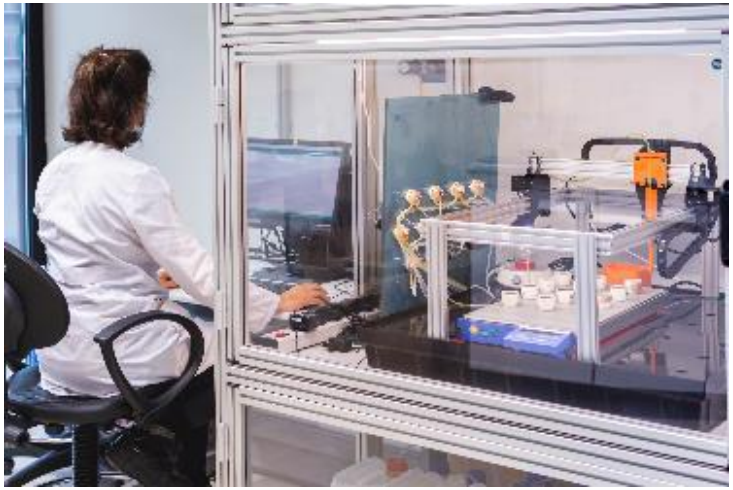

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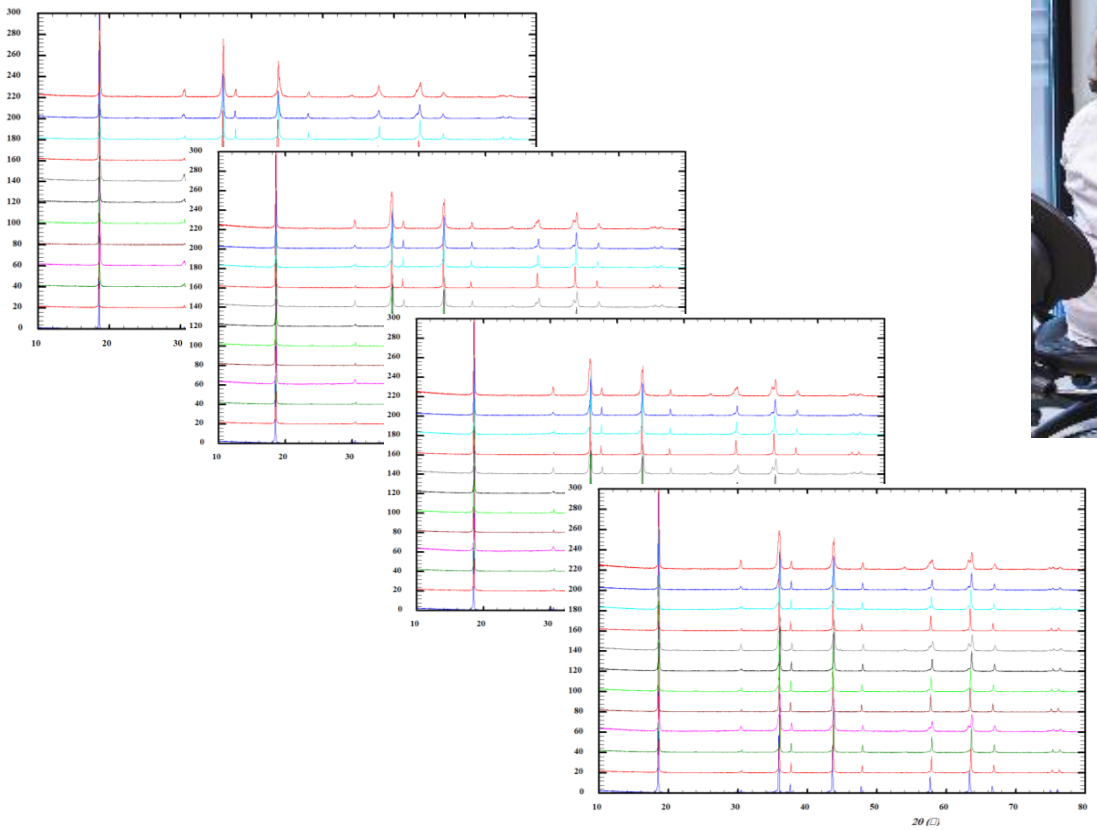
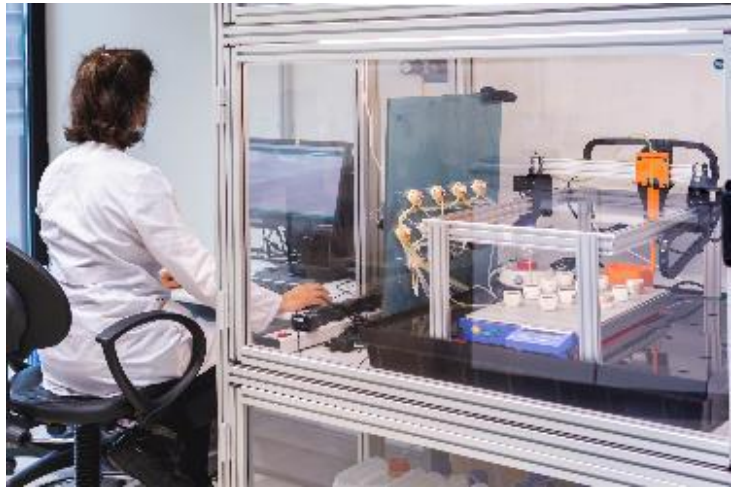
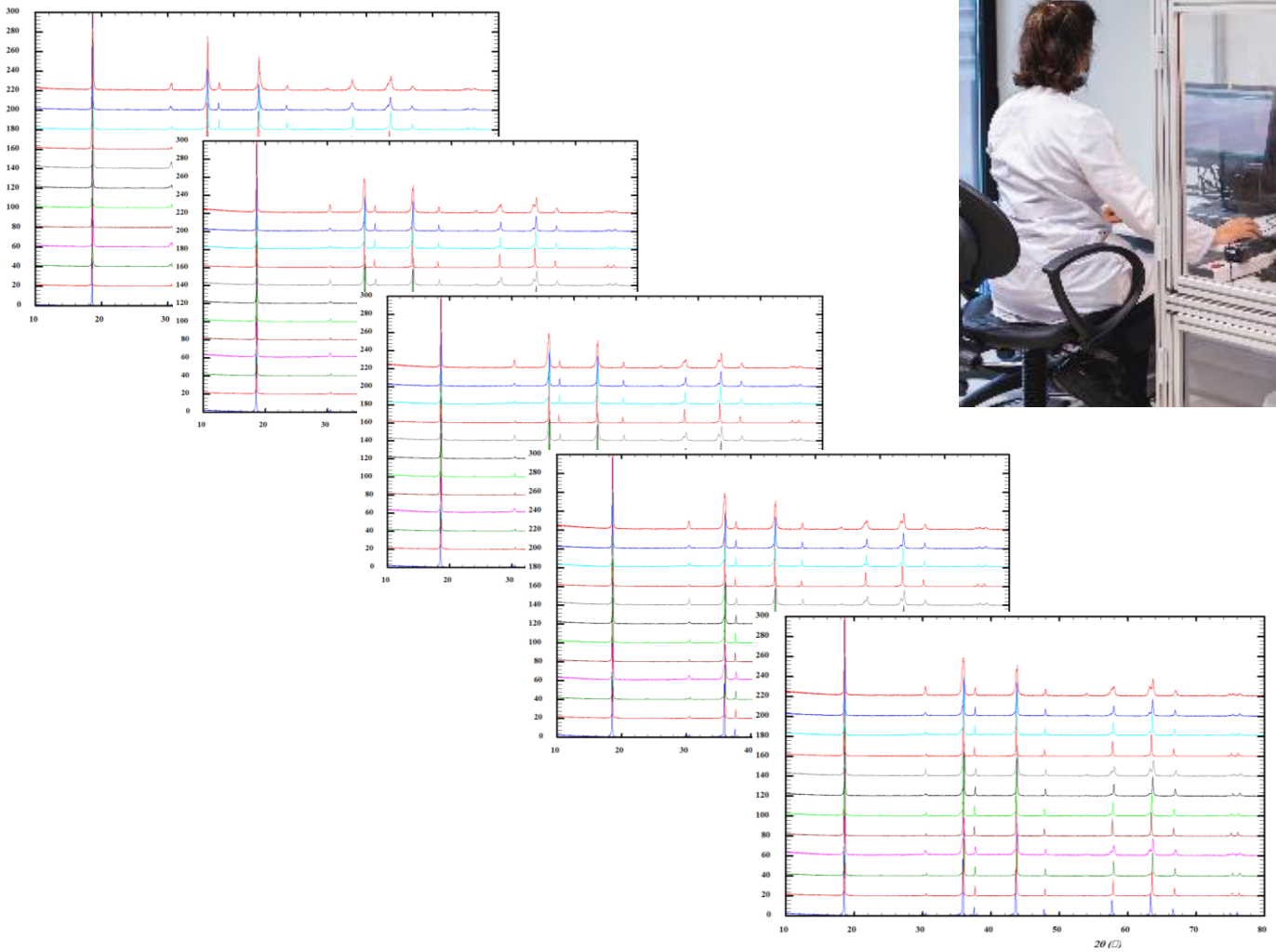
High-Throughput synthesis

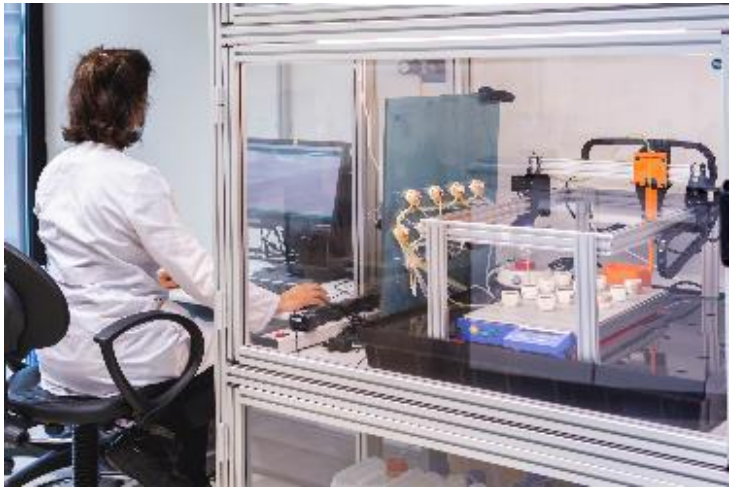




2. Automated module for synthesis of inorganic compounds

High-Throughput synthesis

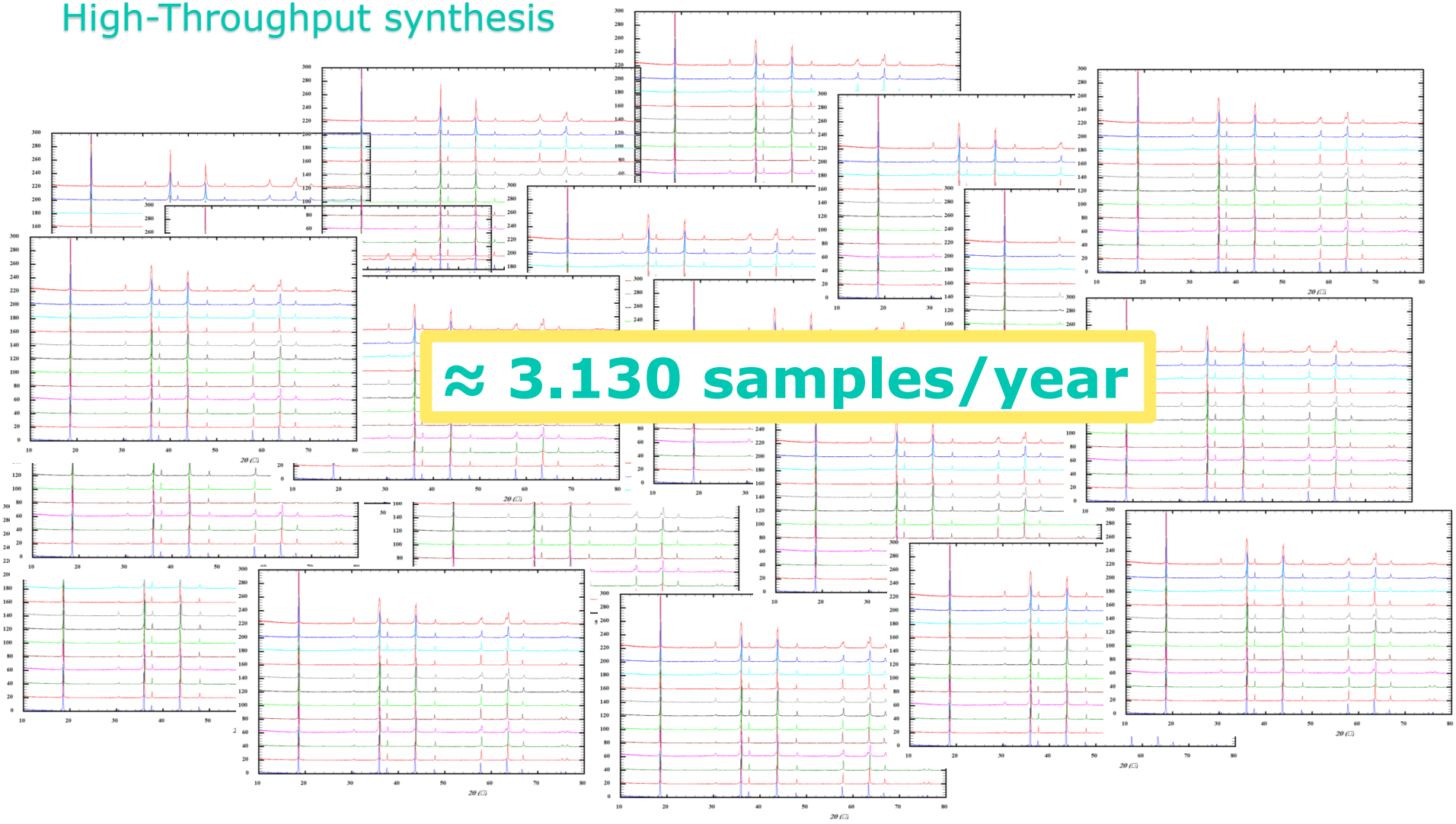




60 samples/week

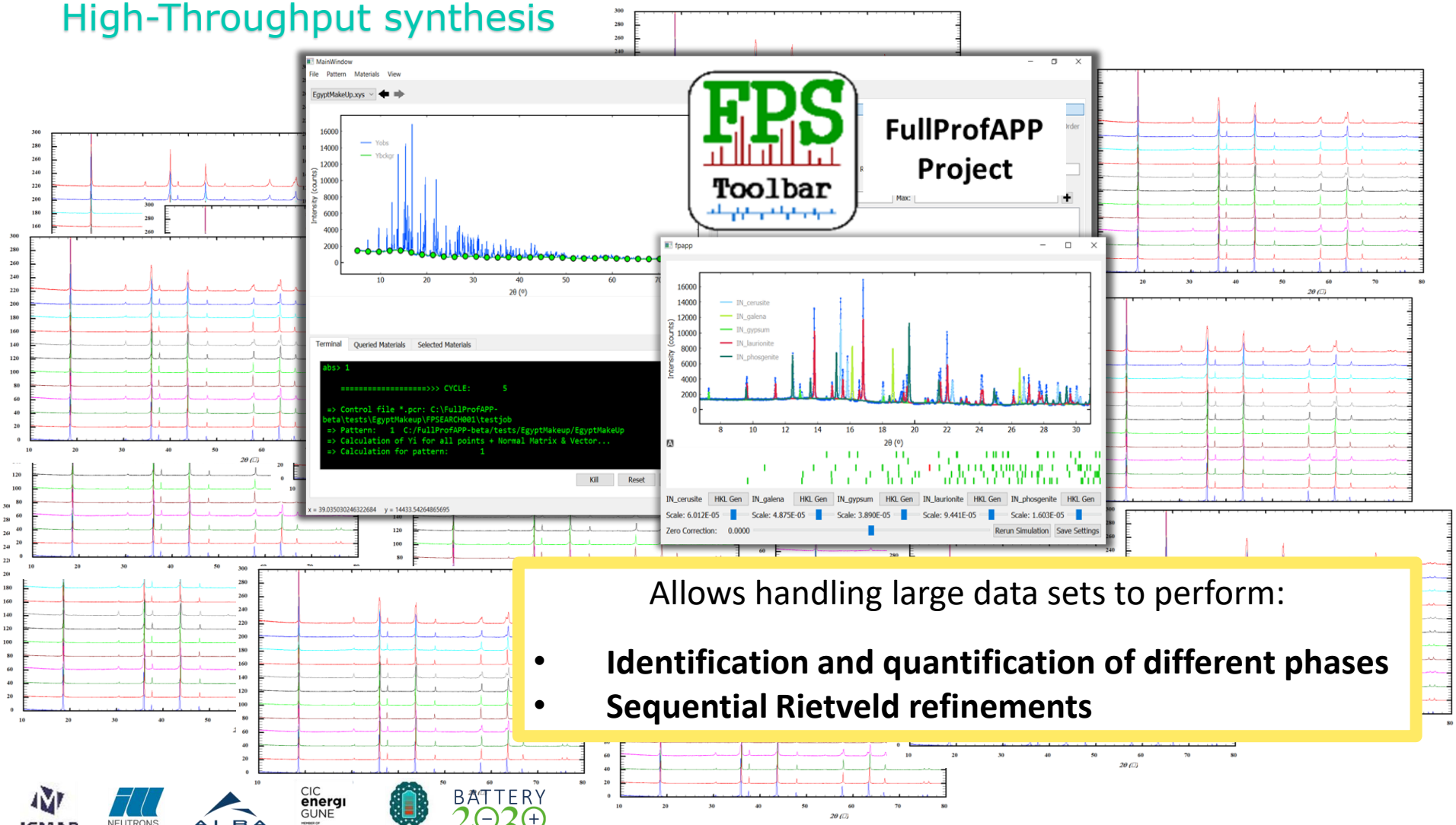
2. Automated module for synthesis of inorganic compounds

High-Throughput synthesis



2. Automated module for synthesis of inorganic compounds

High-Throughput synthesis



Allows handling large data sets to perform:

- **Identification and quantification of different phases**
- **Sequential Rietveld refinements**

ICMAB · NEUTRONS FOR SOCIETY · ALBA · CIC energiGUNE · BIG-MAP · BATTERY 2030+

Collaboration: CIC, ALBA, ILL, ICMAB

Arcelus et al. 2024

3. EXPERIMENTAL VALIDATION AND RESULTS

3. Experimental validation and results

Module validation: reproducibility

- LFMO

$$LiNO_3 + Fe(NO_3)_2 + Mn(NO_3)_2 \xrightarrow{Sol-gel\ synthesis} \mathbf{LiFe_{0.5}Mn_{1.5}O_4}$$

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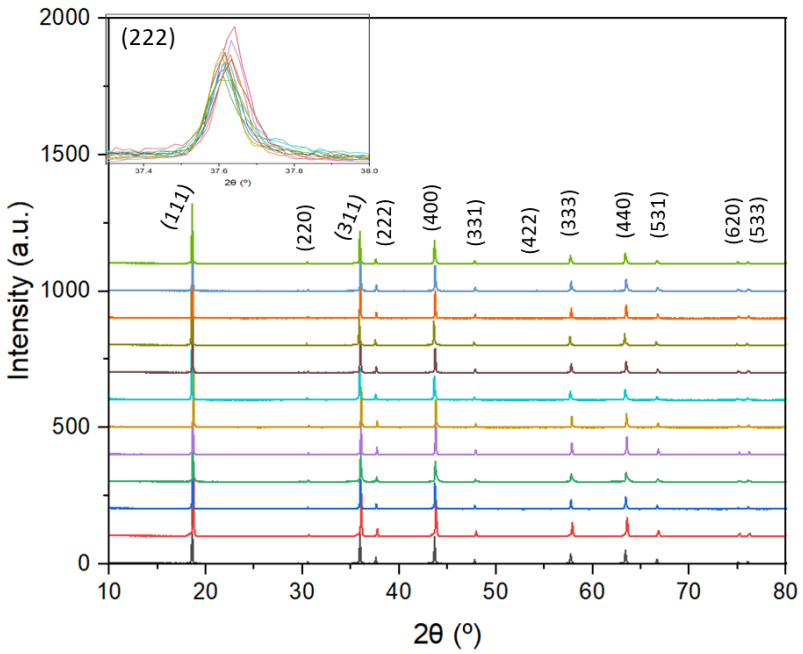

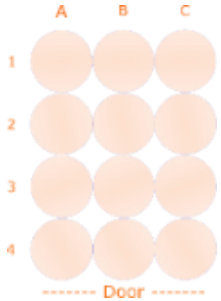

12 positions (module/oven)

✓ **Reproducible**

➢ Cell parameters: 8.297(2) and 8.307(2)Å

λ= 0.8258 nm

3. Experimental validation and results

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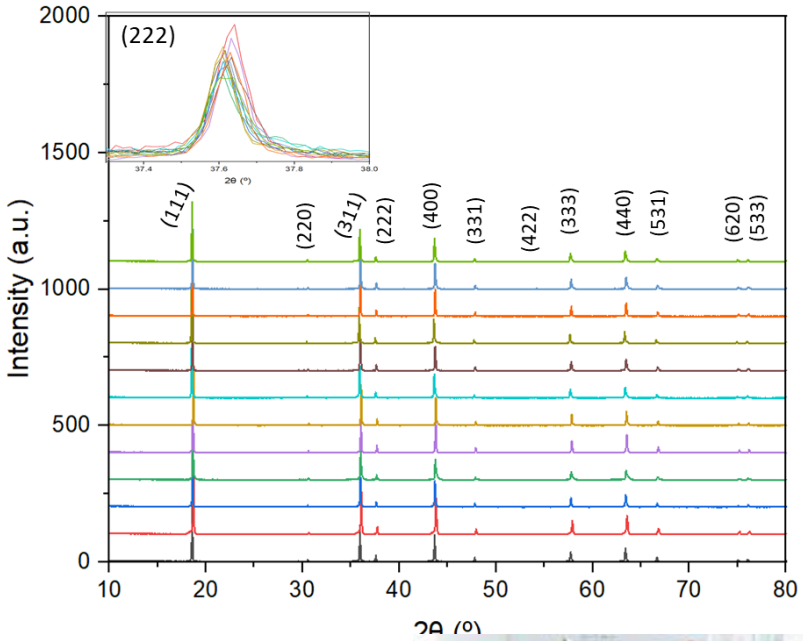

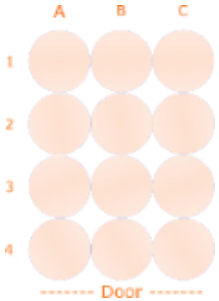

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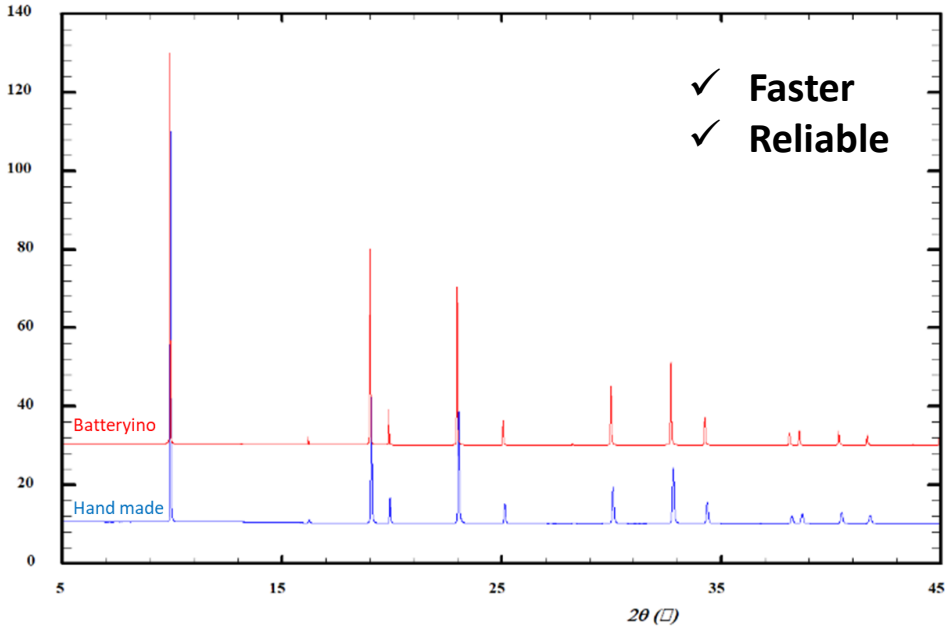


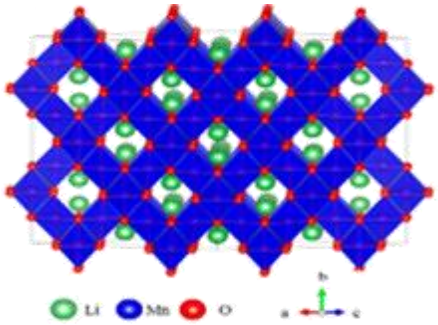
✓ **Faster**
✓ **Reliable**

λ= 0.8258 nm

3. Experimental validation and results

High voltage Manganese-based spinel oxides

- ✓ Very complex crystallochemistry
- ✓ 3D ionic conductivity



3. Experimental validation and results

High voltage Manganese-based spinel oxides

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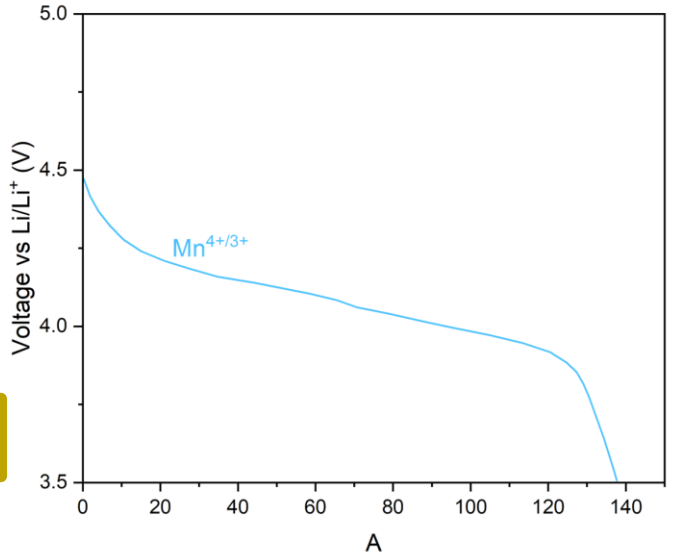
Spinel	Theoretical capacity ($mAh \cdot g^{-1}$)	Discharge operating voltage vs. Li/Li^+ (V)	Critical element's content (%)	Metal cost
$LiMn_2O_4$	148	4.0 and 4.1	33.3	Low
$LiCo_{0.5}Mn_{1.5}O_4$	147	4.0 and 5.1	50	Very high
$LiNi_{0.5}Mn_{1.5}O_4$	147	4.70 and 4.75	50	High
$LiFe_{0.5}Mn_{1.5}O_4$	148	4.0 and 5.0	33.3	Low

Reproduced from Bhaskar et al. 2010, Efterkhari et al. 2003, Yu et al.. 2022, Stüble et al. 2023



- ✓ Well known and commercial
- ✓ Voltage need to be increased

I. Monterrubio et al. 2024, J. Phys. Chem.

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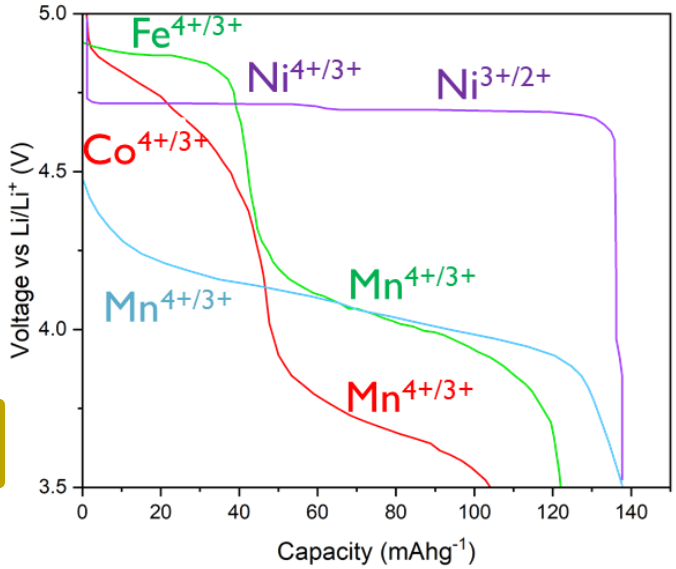

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↓

Mn partial substitutions

3. Experimental validation and results

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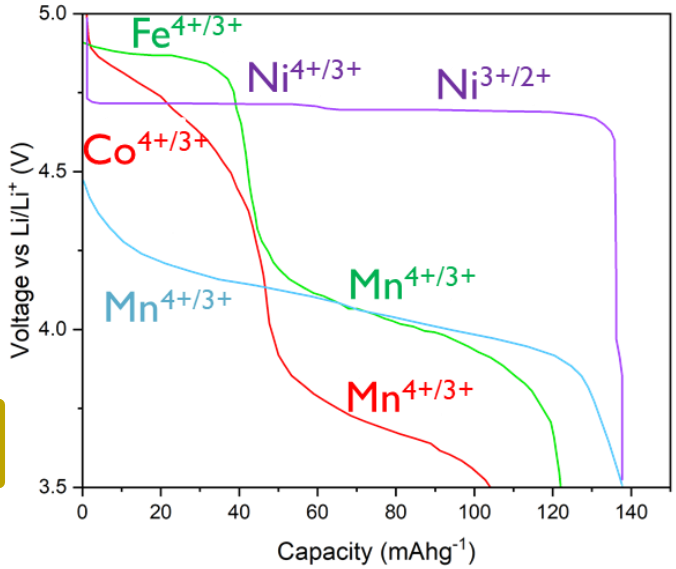

- Co substitution: $ $
- Ni substitution: $

I. Monterrubio et al. 2024, J. Phys. Chem.

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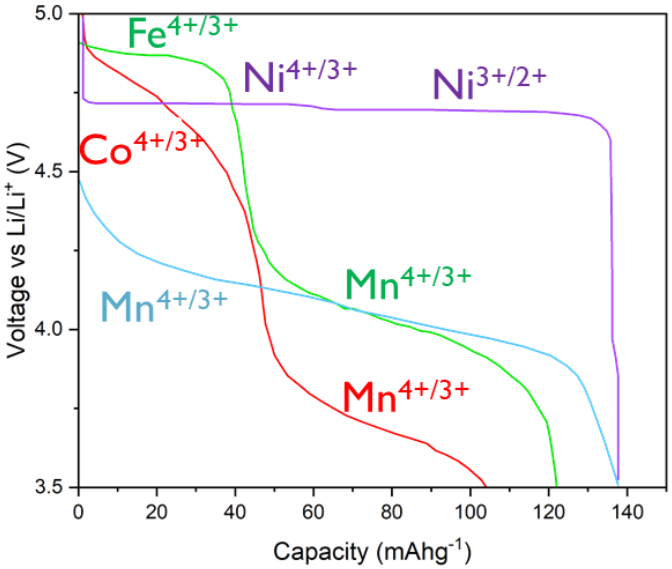

LFMO

- ✓ Higher operating voltage
- ✓ Cheaper
- ✓ More sustainable and less toxic

I. Monterrubio et al. 2024, J. Phys. Chem.

3. Experimental validation and results

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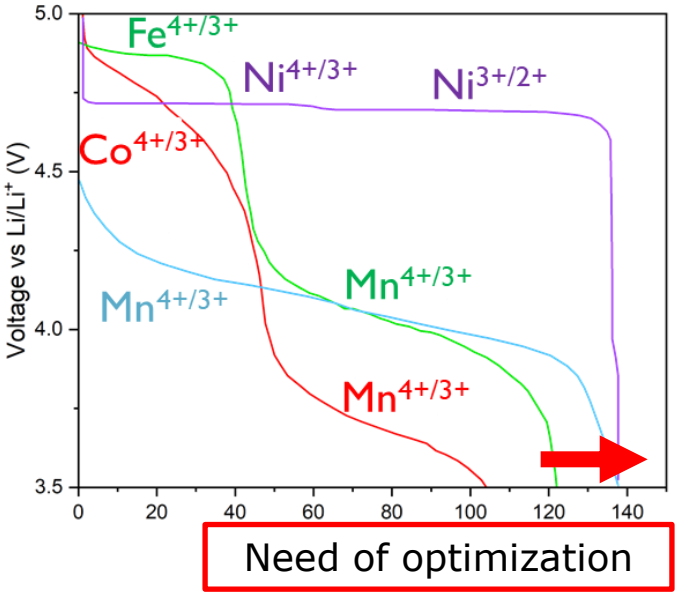

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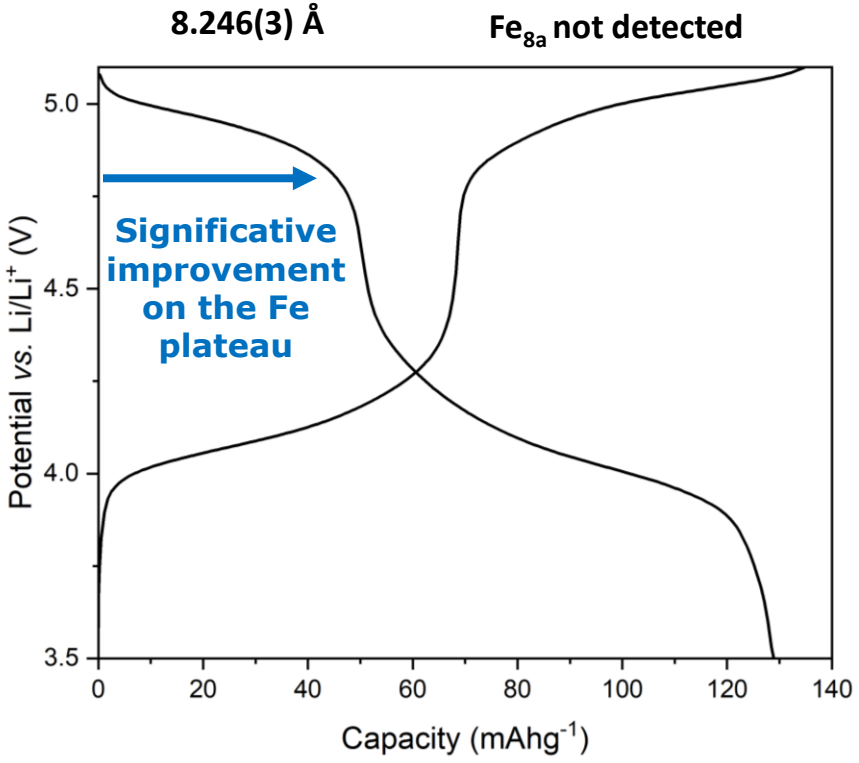
3. Experimental validation and results

Electrochemical characterization

- LFMO:



Cycled in a Swagelok-type cell at C/10 in a 3.5-5.0V voltage window vs. Li/Li^+ using LP30+FEC

Highest values ever reported for LFMO, achieved by the **control of the quantity of antisite defects**

✓ High **discharge capacity: ≈130mAh/g**

✓ Increased **average voltage: 4.4V**

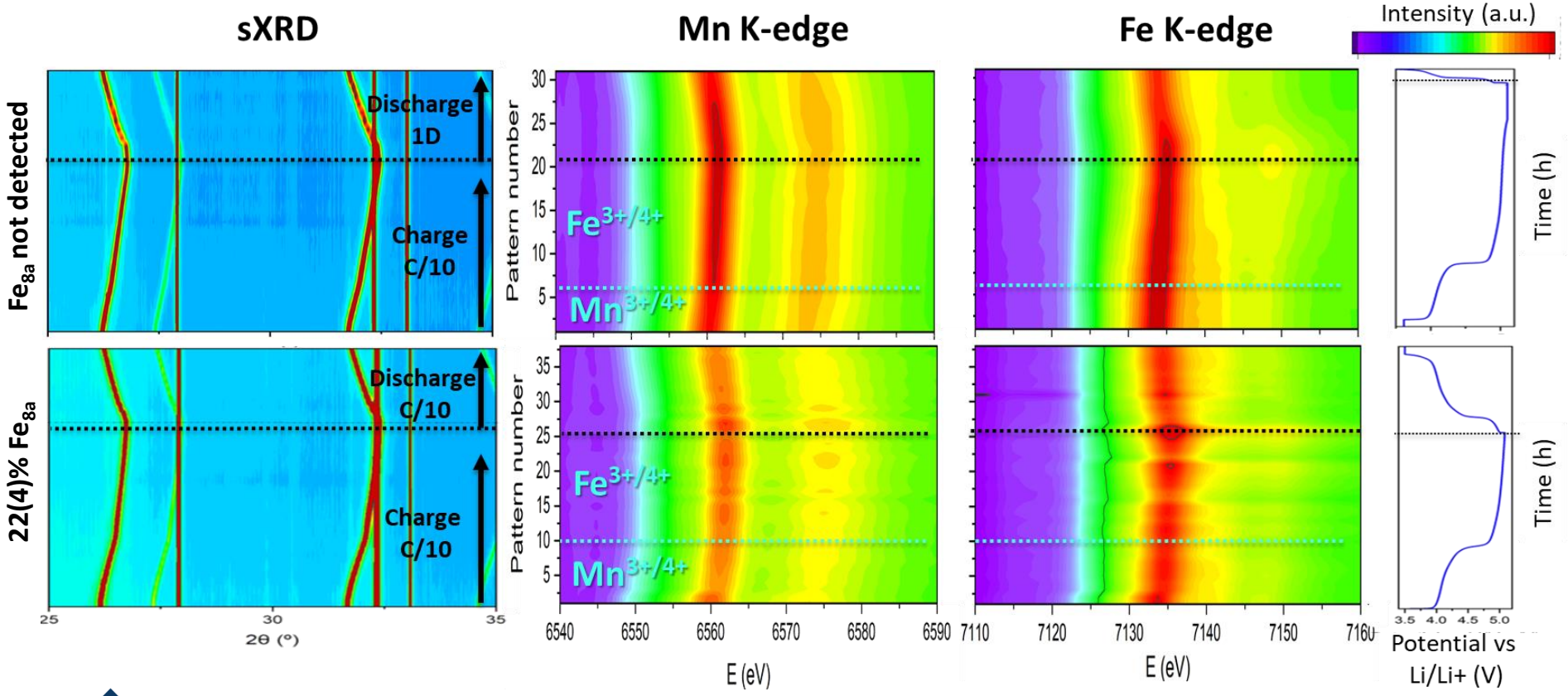
✓ Increased **energy density: 572Wh/Kg**

I. Monterrubio et. al. Submitted 2024

[MM1] ~~CAMBIAR NOMBRES! Si que tiene defectos detectados por XRD (check % de defectos de low)~~

3. Experimental validation and results

Operando sXRD and XAS measurements



Cycled at C/10 and 1D in a 3.5-5.1V voltage window vs. Li/Li$^+$ using $LiBF_4$ in EC:DMC (3:7 by wt.)

4. GENERAL CONCLUSIONS

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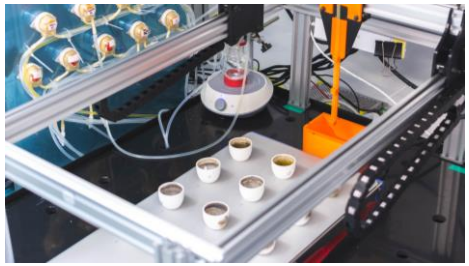
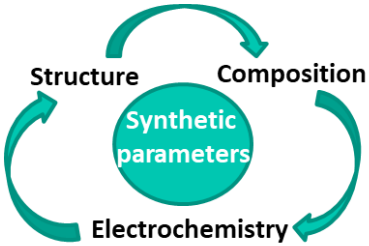
Module:

- ✓ We developed a module to **automatize the mix of reagents** that is reproducible and reliable.



Study of the $LiFe_{0.5}Mn_{1.5}O_4$ composition:

- ✓ Screening of synthetic parameters by synthesizing >150 LFMO samples
- ✓ We can **control the quantity of Fe_{Th}** to **maximize the electrochemistry.**



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ALBA

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Programa Predoctoral PRE_2021_2_0014

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AGENCIA ESTATAL DE INVESTIGACIÓN

BATTERY 2030+

BIG-MAP

Universidad del País Vasco
Euskal Herriko Unibertsitatea

GRACIAS · THANK YOU · ESKERRIK ASKO

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MEMBER OF BASQUE RESEARCH & TECHNOLOGY ALLIANCE

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Making sustainability real

cicenergigune.com