

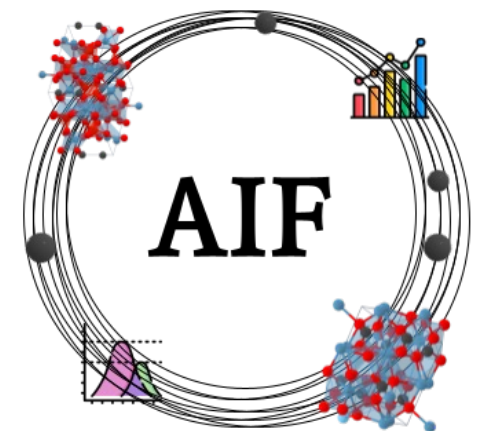
Automated Labs: Data Infrastructure & Framework for Interpretation of Characterization results

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MRS Conference, April 2025



Autonomous labs accelerate discovery. But also bring new data challenges!

Automated synthesis



Robotics



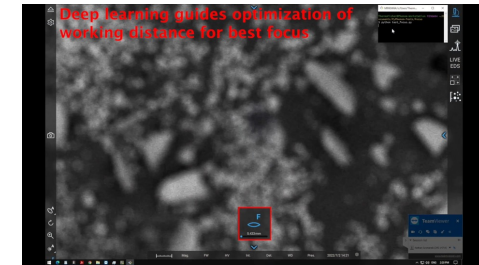
A-Lab at LBNL

Szymanski, N. J et al. An Autonomous Laboratory for the Accelerated Synthesis of Novel Materials. *Nature* 2023, 624 (7990), 86–91.
<https://doi.org/10.1038/s41586-023-06734-w>

Automated characterization (still in progress)

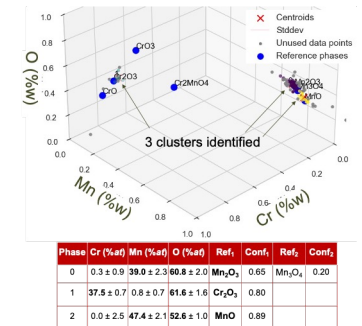
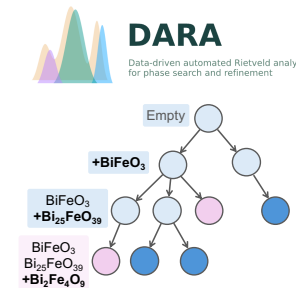
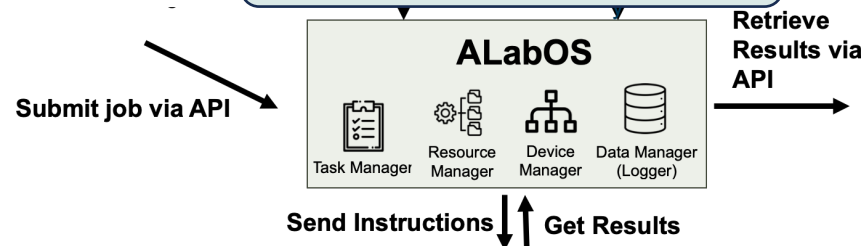


AI, statistics, & modeling-based XRD operation & analysis



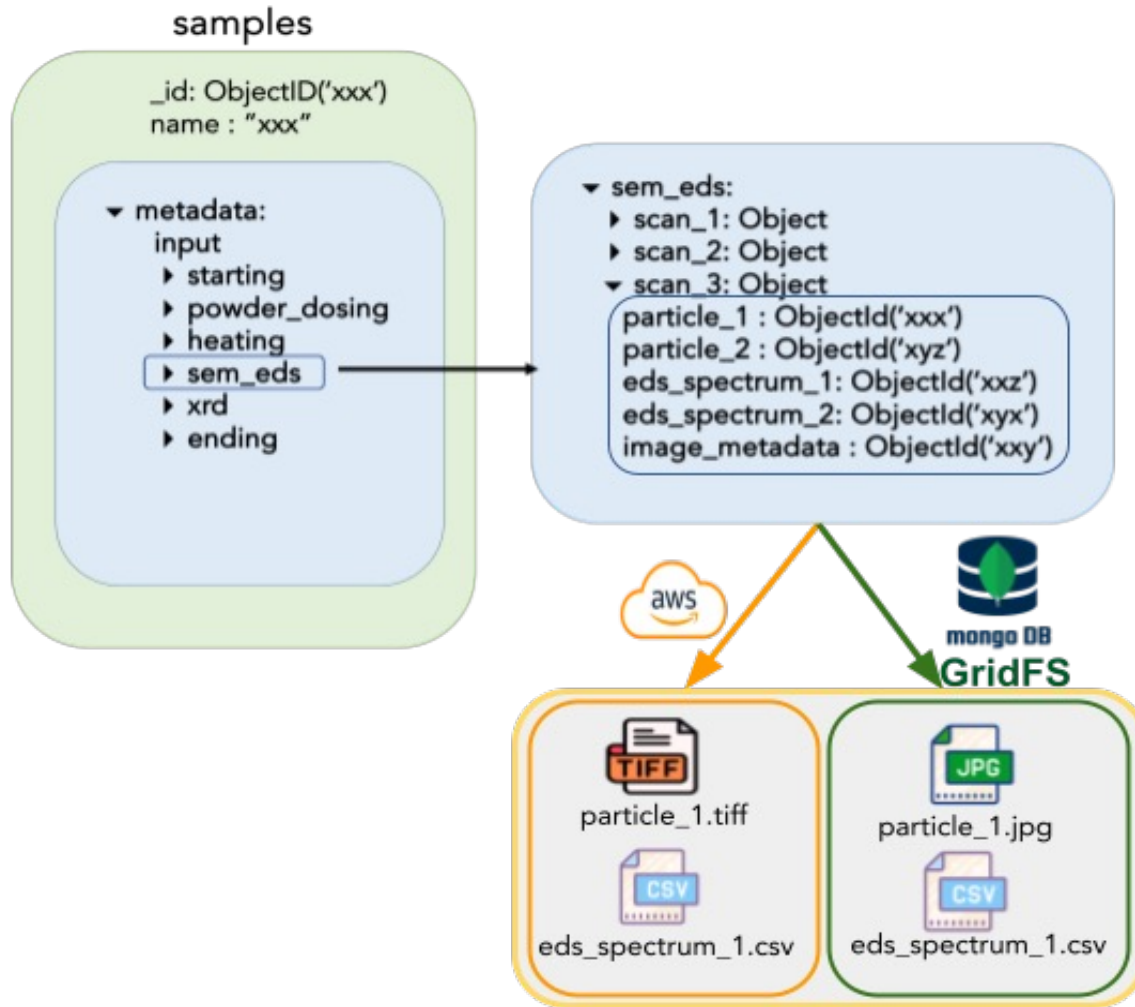
AI-based SEM-EDS operation & analysis

Data management



Fei, Y.; Rendy, B.; Kumar, R.; Dartsi, O.; Sahasrabuddhe, H. P.; McDermott, M. J.; Wang, Z.; Szymanski, N. J.; Walters, L. N.; Milsted, D.; Zeng, Y.; Jain, A.; Ceder, G. AlabOS: A Python-Based Reconfigurable Workflow Management Framework for Autonomous Laboratories. *Digital Discovery* 2024, 3 (11), 2275–2288. <https://doi.org/10.1039/D4DD00129J>.

We have 3 tiers of data storage: document, quick-access files, and archival files



MongoDB(NoSQL) documents
(frequent access) are used to store metadata, analysis, and state. Fast to query and support flexible, nested structures.

MongoDB GridFS
(frequent access) are used to store frequently needed files, images, etc.

AWS storage
(archival, rarely accessed) are used to store large raw data formats.

NoSQL → Enables fast queries by retrieving all relevant experiment data together.

Building an interactive dashboard for analysis

Alab Dashboard

Main Dashboard

Saved Plots

×	TRI_7
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Target:

 CaAg_3O_4

3 Similar Experiments:

TRI 8 : similarity score = 89%

TRI -1 : similarity score = 67%

TRI -4 : similarity score = 67%

1 Experiments with same target:

TRI 8

2 Experiments with same powders:

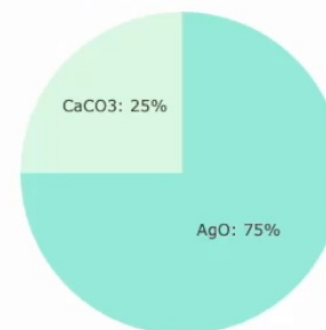
TRI 7 , TRI 8

Available Precursors in Selected Sample

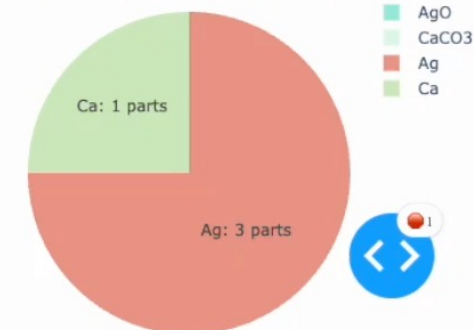
● Not in use ● Used in sample ● Used in Alab



Powder Composition

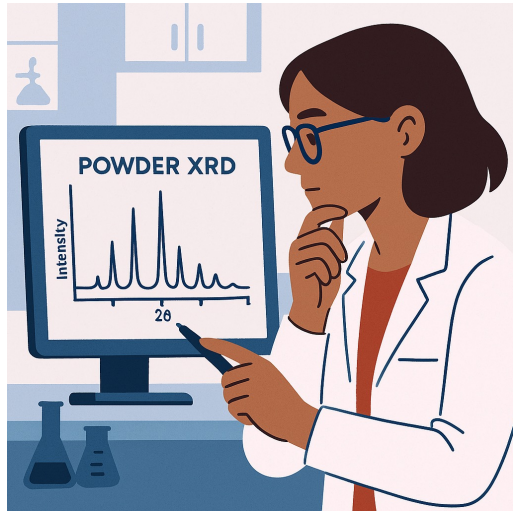


Elemental Composition (Ratios)



More samples, more problems: How to scale characterization?

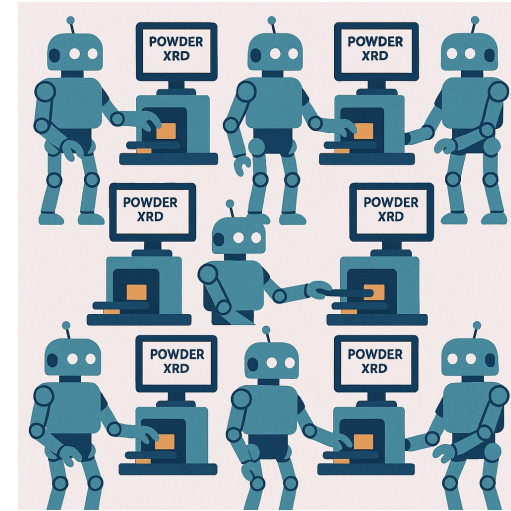
Traditional characterization



Performed **one-at-a-time**, by a dedicated **expert** using their **technical intuition**, **experience**.

Goal is to reach a single (published) conclusion.

Automated characterization



The challenge is to develop an **autonomous framework** for high-throughput characterization.

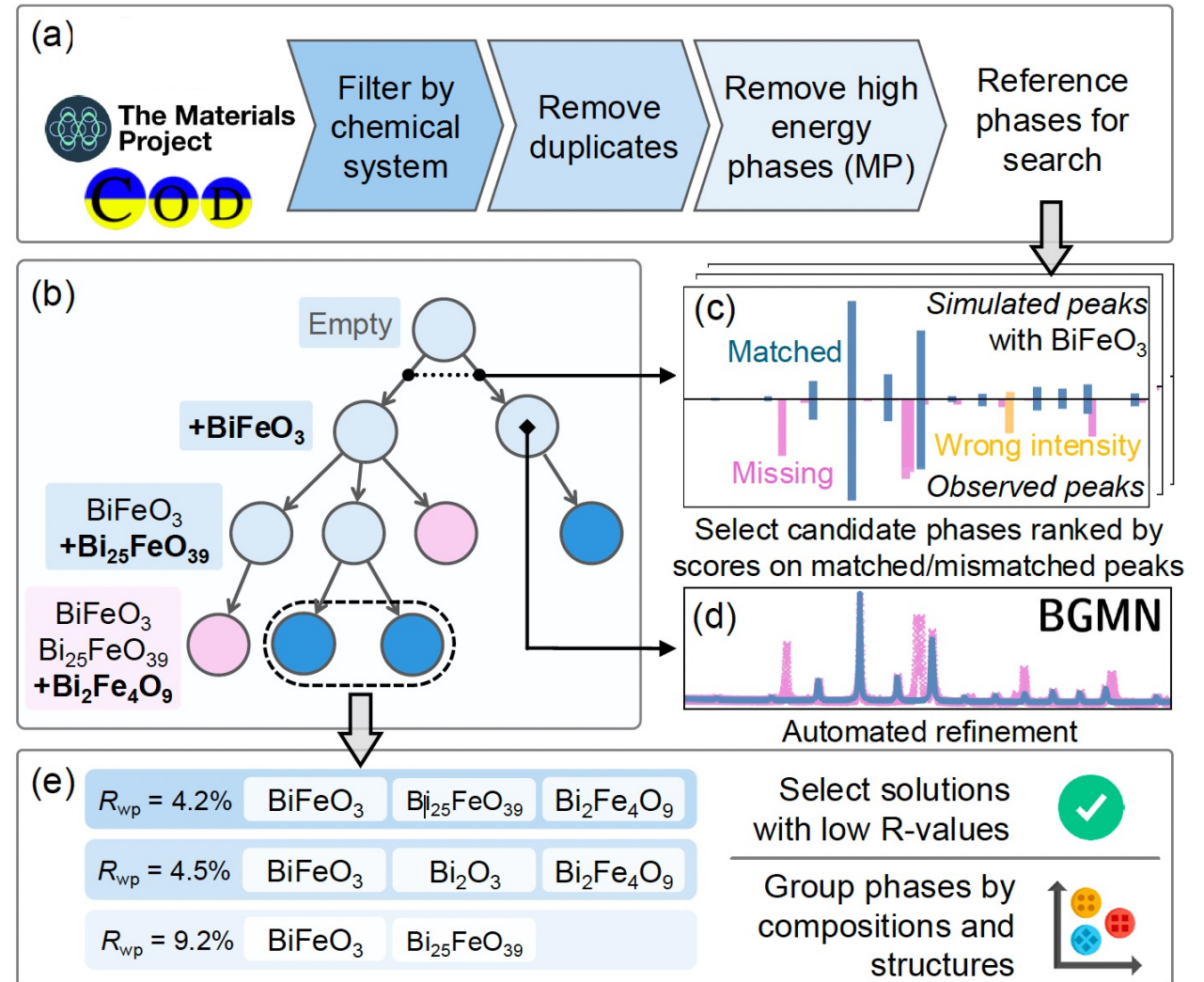
This should **replicate expert reasoning** but scale to a **large number of samples**.

DARA: Data-Driven XRD analysis for automated phase identification



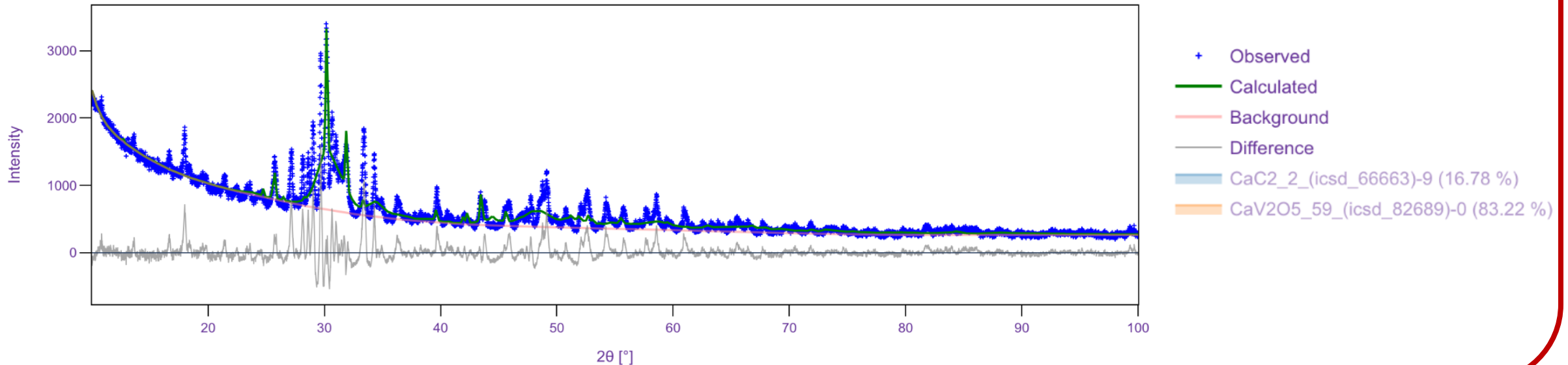
To improve automated characterization in the A-Lab, the team has developed the DARA method.

- Starting with a database of possibilities, the software uses a **tree search method** to traverse all the possibilities of phase combinations.
- A **Rietveld-Refinement-based phase search** (via BGMN) can handle complex patterns (e.g., with peak shifts and broadening)
- Multiple solutions are presented**, ranked by a numerical figure of merit. Phases w/similar patterns (e.g., solid solutions) are grouped together.



In deployment, however, we see instances where chemical intuition is needed for proper interpretation

xrd_data.xy (Rwp=16.19%)



Issue



Synthesis pathway

Precursors: CaCO₃, V₂O₅

Target: CaVO₂

Temperature: 600C

Dwell Duration: 12h

Tube Furnace: Argon

Predicted phases

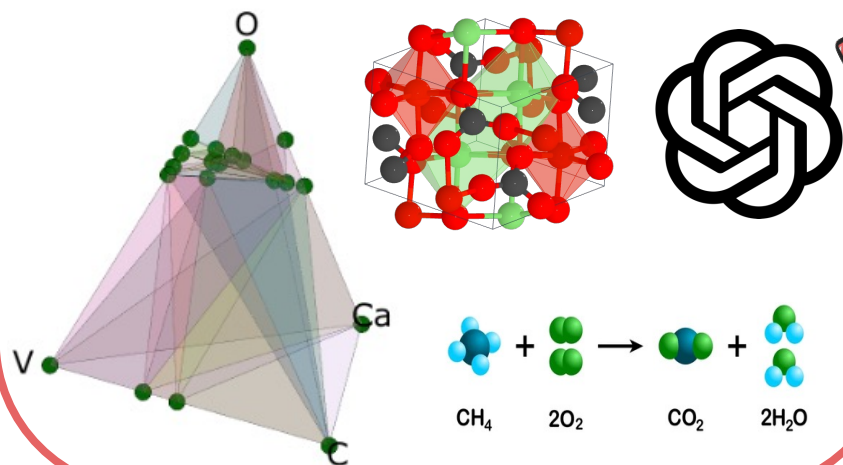
CaC2_2 (17%)
CaVO5_59 (83%)

Carbon rich calcium carbide phase as CaC2 should not be present as we are not reducing to form the carbide. The overall fit quality is also not great so we may have extra skepticism on this result.

How do we automate chemical intuition?

Chemical knowledge:

- Composition balance
- LLM
 - Phase stability
 - Chemical plausibility
 - Precursor information



- Provide the LLM with the **synthesis precursors and procedure**.
- Ask the LLM to rank the likelihood of a synthesis outcome.
- Uses “**few-shot**” **prompting**, so a few examples are provided to the LLM so it understands the instruction.

Do LLMs have such an intuition?

Some of our initial tests indicated the LLMs may indeed have useful chemical intuition

The synthesis data are:

Target: CaV2O5

Precursors: ['CaCO3', 'V2O5']

Temperature: 600°C

Dwell Duration: 12.0 hours

Furnace: Argon

phases = ['CaC2_2', 'CaV2O5_59']

Likelihoods: {{ "CaC2_2": 0.1,
"CaV2O5_59": 0.6
}},

Explanations: {

"CaC2_2": "CaC2 is unlikely to form under the given synthesis conditions. The precursors CaCO3 and V2O5 do not provide the necessary carbon source in a form that would favor the formation of CaC2. Additionally, the temperature of 600°C is not typically sufficient for the formation of CaC2, which generally requires higher temperatures and a more reducing environment."

"CaV2O5_59": "CaV2O5 is more likely to form under the given synthesis conditions. The precursors CaCO3 and V2O5 can react to form CaV2O5 at 600°C, especially in the presence of an Ar+H2 atmosphere, which can facilitate the reduction of V2O5. The dwell duration of 12 hours is sufficient to allow for the reaction to proceed to completion, making the formation of CaV2O5 plausible."

}

Interpretation_Likelihood: 0.3,

Interpretation_Explanation:

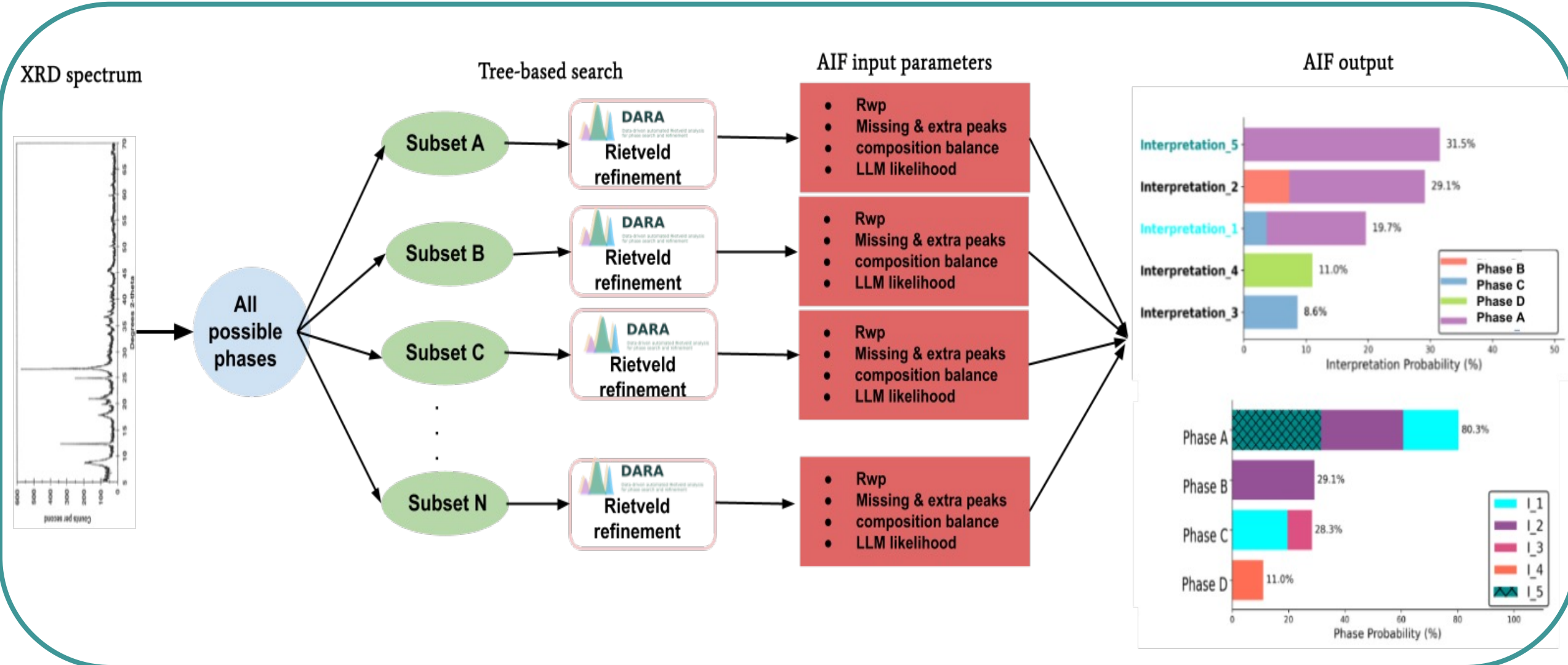
{ "The combination of CaC2 and CaV2O5 as the formed phases is not highly likely. While CaV2O5 can form under the given conditions, the formation of CaC2 is improbable due to the lack of a suitable carbon source and the relatively low synthesis temperature. Therefore, the overall likelihood of this combination of phases being a valid interpretation is moderate to low." }

For the given precursors and conditions, the LLM (GPT-4o) ranks:

- CaC₂ to be an unlikely outcome with p~0.1
- CaV₂O₅ to be a likely outcome with p~0.6
- A mixture of those phases to have p~0.3

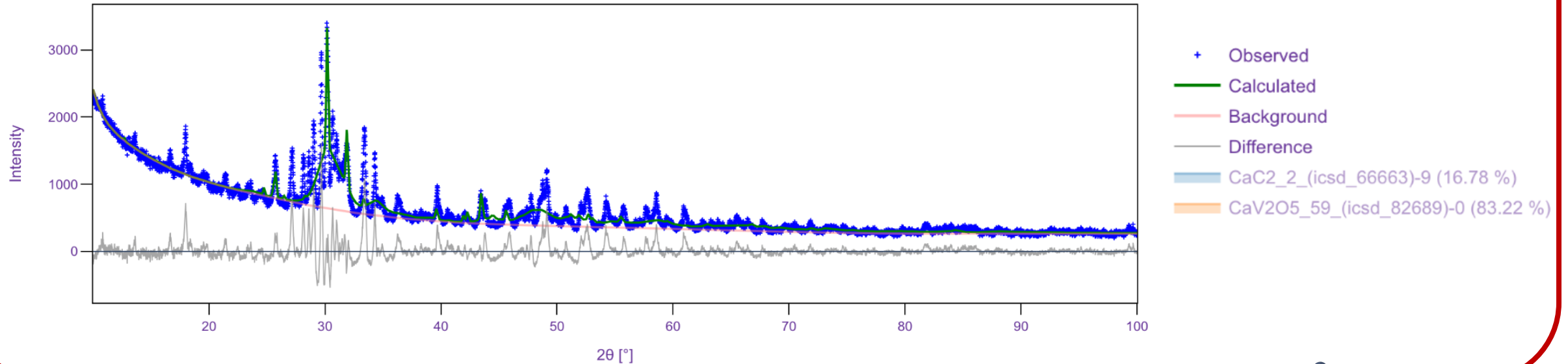
This is the type of intuition we are looking for!

Integrating this idea into a more general Automated Interpretation Framework (AIF)



Revisiting example with unexpected CaC2

xrd_data.xy (Rwp=16.19%)



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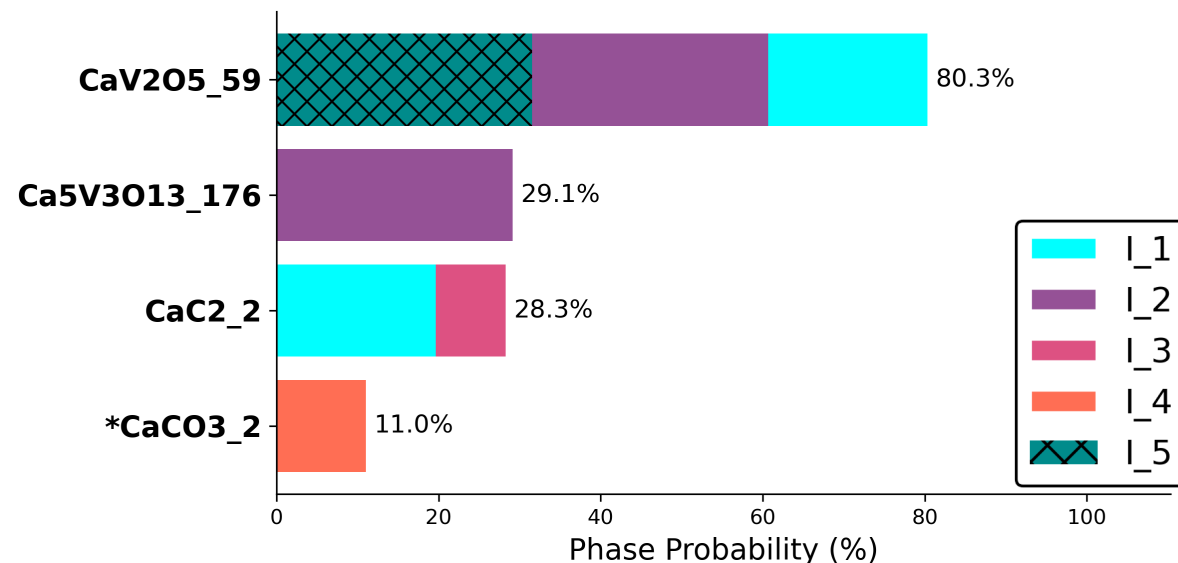
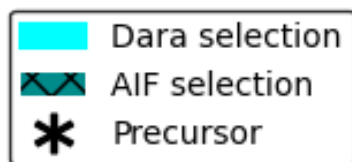
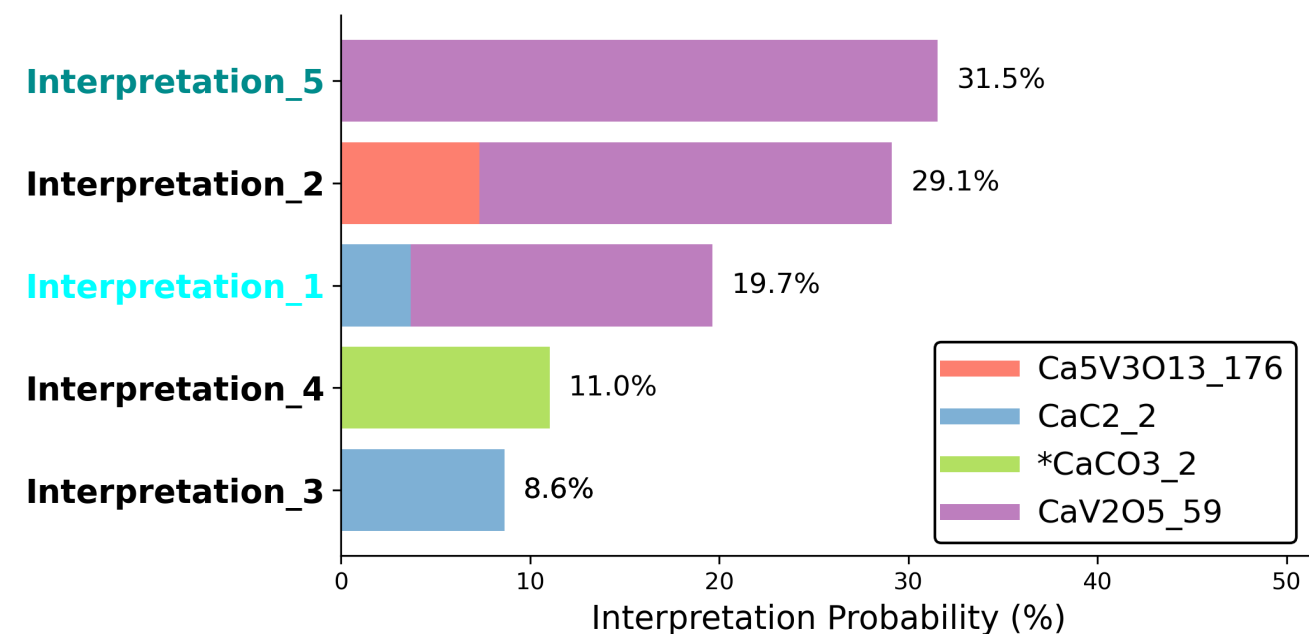
Tube Furnace: Argon

Predicted phases

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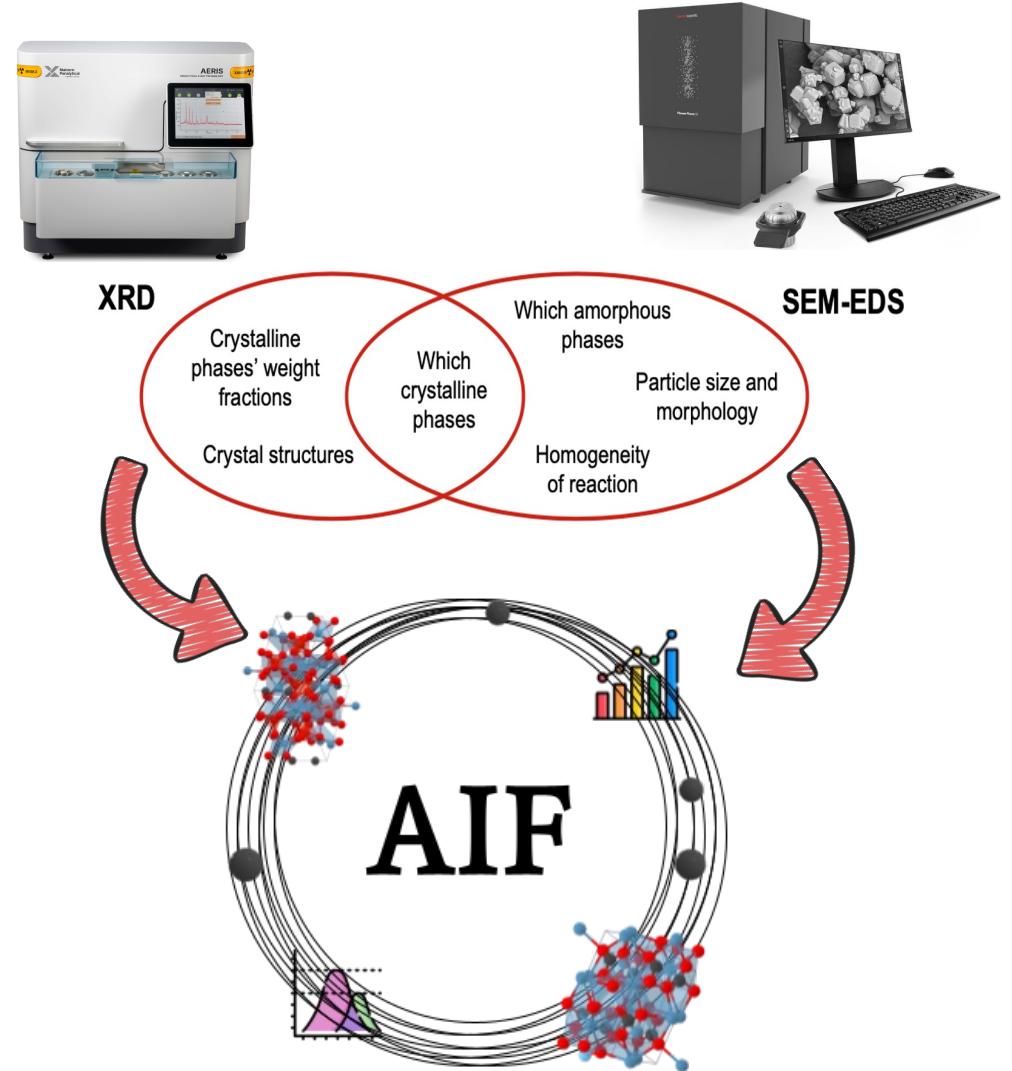
AIF correctly suggests CaC2 is less likely based on chemical priors we introduced



Conclusions and future work

The AIF **probabilistic Bayesian framework** incorporates chemical intuition into automated XRD analysis.

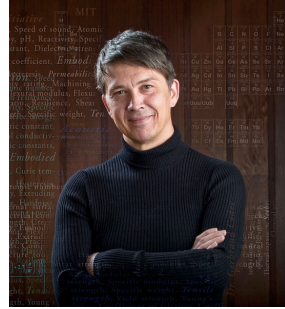
- Future work:
 - **Uncertainty analysis**, including none of the interpretations being correct.
 - **Larger-scale testing** of the framework to demonstrate its applicability is needed.
 - Include framework in joint analyses (e.g. XRD + SEM-EDS).



Acknowledgements



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