A Computational Challenge Problem in Materials Discovery: Synthetic Problem Generator and Real-World Datasets

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Abstract
Newly-discovered materials have been central to recent technological advances. They have contributed significantly to breakthroughs in electronics, renewable energy and green buildings, and overall, have promoted the advancement of global human welfare. Yet, only a fraction of all possible materials have been explored. Accelerating the pace of discovery of materials would foster technological innovations, and would potentially address pressing issues in sustainability, such as energy production or consumption.

The bottleneck of this discovery cycle lies, however, in the analysis of the materials data. As materials scientists have recently devised techniques to efficiently create thousands of materials and experimentalists have developed new methods and tools to characterize these materials, the limiting factor has become the data analysis itself. Hence, the goal of this paper is to stimulate the development of new computational techniques for the analysis of materials data, by bringing together the complimentary expertise of materials scientists and computer scientists.

In collaboration with two major research laboratories in materials science, we provide the first publicly available dataset for the phase map identification problem. In addition, we provide a parameterized synthetic data generator to assess the quality of proposed approaches, as well as tools for data visualization and solution evaluation.

Introduction
The discovery of new advanced materials has made possible recent technological inventions, from silicon circuits and batteries to solar and fuel cells. As underlined by the Materials Genome Initiative (Patel 2011; White 2012), accelerating the discovery and deployment cycle of new advanced materials is essential to improving human welfare and to achieving sustainable, clean energy.

While most inter-metallic compounds or oxides involving up to two elements have now been studied, it only represents a very small fraction of all possible materials that can be obtained by mixing three elements or more, and by varying the synthesis conditions (e.g. temperature and pressure). Out of billions of candidate materials, there are potentially thousands of materials with interesting physical properties, such as conductivity, light absorbency or catalytic properties, that remain to be uncovered.

The field of materials discovery requires a deep understanding of the underlying crystallographic process that governs a material formation. Experts in materials science have developed elaborate deposition processes to efficiently create so-called composition libraries in a high-throughput regime (Takeuchi, Dover, and Koinuma 2002; Gregoire et al. 2007, e.g.). For example, the High-Throughput Experimentation research program at the Joint Center for Artificial Photosynthesis (JCAP) is capable, at full capacity, of outputting about one million materials a day. Once synthesized, the promising composition libraries need to be characterized using X-ray diffraction and X-ray fluorescence (Chu et al. 2004; Vogt et al. 2004; Gregoire et al. 2009) in order to map the composition and structure of a library. This data collection step requires an X-ray source such as a synchrotron, where the experimentation cost amounts to about $1M for a week of data collection.

Nevertheless, as the data collection reaches a high-throughput regime, the bottleneck of the discovery cycle becomes the data interpretation itself. Indeed, this task remains a laborious manual inspection that relies on materials scientists expertise. The goal of this paper is to encourage computer scientists to propose new computational methods for this data interpretation, by providing synthetic and real data, as well as a problem description and evaluation metrics that transcend materials science research.

This work is in collaboration with two major research centers in materials science. The High-Throughput Experimentation research program at JCAP focuses on automated, high-throughput discovery of materials that can act as light absorbers or catalysts for solar-fuel generation, in collaboration with the Stanford Synchrotron Radiation Lightsource. The Energy Materials Center at Cornell (emc2) aims at improving energy conversion and storage by understanding and exploiting fundamental properties of materials, and conducts experiments at the Cornell High Energy Synchrotron Source (CHESS).

There are several existing sources of tabulated X-ray diffraction data from crystallographic studies of materials. Both the National Institute of Standards and Technology
1. Gibbs Phase Rule, which says that (assuming constant temperature and pressure), that $k_s \leq M$, where $k_s \leq K$ is the number of phases present in sample $s$, and $M$ is the number of different material elements in the system.

2. The compositions at which a phase is observed should be a connected region and its parameters should vary smoothly as a function of composition.

This definition is intended as a guideline, and could be reasonably modified, for example to treat $K$ as an estimated parameter, or to incorporate other knowledge from the materials science or solid state physics literature. It is important, however, that a solution model or its predictions have a reasonable physical interpretation (potentially after post-processing or reconstruction), and are consistent with the underlying physics.

An output model should be evaluated using prediction accuracy on the diffraction curve or peak parameters of held-out samples. For synthetically generated data, model parameters can be compared directly with those that follow from the generator parameters.

### Challenges

Many challenges arise as data is collected and as it is processed. The first challenge to overcome relates to the noise in the data. The uncertainty in the diffracted intensities comes from multiple factors, such as measurement errors, background noise, and detector ghosting. There is also uncertainty in the composition values, arising from the analysis of the X-ray fluorescence data.

Another challenge relates to the model assumptions. For example, while thermodynamic equilibrium is usually assumed to solve this problem, it is possible that the equilibrium conditions have not been reached. Other issues might arise from imperfections in the thin-film, variable thickness of the deposited film, or preferred orientation adopted by the crystallites in the thin-film. Finally, for approaches that are based on the discretization of the x-ray patterns into peaks, the peak detection algorithm itself is not a trivial task and might lead to additional uncertainties.
One main challenge is to capture the physical rules that govern the underlying crystallographic process (Long et al. 2007; 2009; Le Bras et al. 2011; Ermon et al. 2012), while allowing the solution approach to scale up to real-sized instances (Le Bras et al. 2011; 2013; Finger et al. 2013). Finally, while structural changes in the crystals typically occur in a linear fashion in composition space, there might be nonlinear changes in shifts, peak widths or relative intensities (peak heights).

Data format
Each data set includes 3 basic types of information:

1. Metadata, such as the chemical elements used in the system. This metadata is useful to retrieve the atomic numbers of the elements, as well as to correlate the proposed phases of a solution with powder patterns of known phases involving these elements.

2. Composition data for each experimental sample, in one of two forms:
   (a) Deposition location, x and y coordinates which are topologically related to the relative concentrations of the constituent elements
   (b) Amount of each element present in the sample (estimated), in scaled absolute units reflecting the thickness of the film as well as relative concentration

3. Diffraction intensity for each sample, in one or both of the following forms. In both cases, the intensity values are scaled by the total amount of X-ray exposure.
   (a) Integrated X-ray counts at each scattering direction in a given range. We provide the filtered curves, which represent a better overall estimate of the signal. Nevertheless, we also provide the unfiltered curves, as it might be more suitable for methods sensitive to artifacts in the shapes of the curves.
   (b) Diffraction peak locations, heights and widths extracted using a wavelet-based peak detection algorithm (Gregoire, Dale, and van Dover 2011).

Listing 1 provides a high-level description of the data format. The metadata contains the system dimension \( M \), the elements involved, the number of samples \( N \), as well as experimentation setup and calibration data such as the beam wavelength \( \lambda \) or detector distance \( d \). Next, the composition data provides the location of the samples on the thin-film (here in the format \([X, Y]\) ). For the diffraction intensity for each sample, the vector \( Q \) corresponds to the range of \( q \) values for which the beam intensities are reported (i.e. x-axis of Fig. 2), and each X-ray pattern is given with respect to these values (i.e. y-axis of Fig. 2). Finally, the detected peaks are listed as triplets \((location, height, width)\).

Visualization and Evaluation Tools
We have developed a graphical user-interface application for exploring and visualizing input datasets as well as solutions to the phase map identification problem1. For visualizing input data, it provides:
1. A map of selectable sample points arranged by deposition or composition-space coordinates (Fig. 1)
2. An interface for viewing diffraction curves from individual sample points (Fig. 2)
3. An interface for viewing diffraction curves from multiple sample points together at once (Fig. 3)

The application can also load solutions provided by a solver.

1Available at http://www.udiscover.it
Figure 2: Diffraction curve (in blue) of a selected sample point (sample point #166) for the instance inst5. The other curves (purple, red, and green) correspond to the component phases involved in that point for a given solution, while the shaded-blue area represents the mixture of these phases, and approximates the diffraction curve.

Figure 3: Heat map of the diffraction curves of the 14 selected points of Fig. 1, where bright colors mean high beam intensity. This visualization helps identify the component phases and their boundaries. The x-axis corresponds to the diffraction angles, while the y-axis refers to the samples in the selected slice.

Listing 2 gives a description of the format of a solution. First, it provides the pattern of each one of the $K$ phases as a vector of intensities. Next, it provides the phase concentrations, namely the fraction of the phases that are involved in each one of the $N$ samples, and how the phase patterns should be shifted in each sample.

```
// Number of phases
K=3

// Phase patterns (basis)
Q=10.0,10.1,10.2,10.3,10.4,...
B1=153.02,164.52,127.68,87.34,...
B2=166.70,178.54,153.51,98.02,...
...

// Phase concentrations at each sample
C1=0.00,0.72,0.85
C2=0.00,0.87,0.79
C3=0.00,0.92,0.71
...

// Phase shifts at each sample
S1=0.00,1.00,1.02
S2=0.00,1.01,1.02
S3=0.00,1.01,1.01
...
```

The solution can then be visualized with:

1. A map showing the estimated concentration (and other parameters) of each phase, arranged by deposition or composition-space coordinates (Fig. 4)
2. An interface showing the actual, estimated, component, and residual diffraction curves for each sample point (Fig. 2)

The application can calculate an evaluation measure based on absolute error, however other error models are possible, and therefore it is preferable for quantitative evaluation to design a custom measure (incorporating model complexity) to accompany each particular solution structure.

**Synthetic Data Generator**

In order to assess the quality of proposed approaches and validate them, we have created a synthetic data generator. This generator can create basic artificial binary systems ($M = 2$), as well as complex artificial ternary systems ($M = 3$), and a theoretical ternary Aluminum-Lithium-Iron (Al-Li-Fe) system, where the component phases (the patterns and their parameters) have been theoretically calculated.
The generator has a set of user-specified parameters that allow controlling the complexity of the generated instances, as follows:

1. **Underlying system** that governs the number of phases $K$ and their concentration on the film.
2. **Spacing** (in atomic percent) of the data points, which determines the total number $N$ of points.
3. **Total number of peaks** of the component phases, up to the theoretically defined number of peaks.
4. **Number of diffraction angles** of the X-ray patterns, which corresponds to the x-axis precision of the patterns.
5. **Noise level** as a total number (or total amount) of removed peaks from the original constructed patterns.

Using this generator, we provide a benchmark of 100 instances, with varying complexity\(^2\).

### Real World Data

While the previous datasets are made of artificial and theoretical diffraction patterns, the datasets we present in this section have been collected empirically. Nonetheless, for standardization purposes, it follows the same format as described in the previous section. The instances of this dataset have been acquired under various condition. Table 1 shows the dataset of real instances and their parameters. Each instance is characterized by its system dimension $M$, which represents the number of different elements, its design stoichiometry at the center of the film (i.e. the relative proportion of the $M$ elements), the type of substrate it was obtained on, the total number $N$ of sample points, and the gas used in the vacuum chamber during the deposition, if any.

While this dataset is meant to be a stable benchmark and a first building block towards a unified and standardized discovery cycle methodology, it is also meant to be expanded as more data gets collected.

### Conclusions

We provide the first publicly available dataset for the phase map identification problem. This problem is central to the discovery cycle of new materials, as it aims to provide structure and composition maps that can be correlated with interesting physical properties within an inorganic library. In addition, we provide a synthetic generator in order to evaluate the quality of proposed approaches. Finally, we propose a graphical user interface for the visualization of the data and of its solutions. We hope this paper will motivate computer scientists to propose new computational methods for the phase map identification problem.

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### References


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\(^2\)Available at [http://www.udiscover.it](http://www.udiscover.it)
Table 1: Real instances and their parameters, where $M$ is the number of different material elements in the system (the system dimension), and $N$ is the number of sample points. The design stoichiometry corresponds to the relative proportion of the $M$ elements at the center of the film. The table also provides the type of substrate the data was obtained on, as well as the gas used in the vacuum chamber during the deposition, if any.

<table>
<thead>
<tr>
<th>Instance</th>
<th>$M$</th>
<th>Elements</th>
<th>Design Stoich.</th>
<th>Substrate</th>
<th>$N$</th>
<th>Insitu Gas</th>
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<td>inst1</td>
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<td>O$_2$</td>
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<td>O$_2$</td>
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