Computationally accelerated experimental materials characterization - drawing inspiration from high-throughput simulation workflows

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Abstract

Computational materials science is increasingly benefitting from data management, automation, and algorithm-based decision-making in controlling simulations. Experimental materials science is also undergoing a change and increasingly more 'machine learning' is incorporated in materials discovery campaigns. The obvious benefits include automation, reproducibility, data provenance, and reusability of managed data, however, is not widely available. We demonstrate an implementation of a Gaussian Process Regression directly controlling an experimental measurement device in pyiron, a framework designed for high-throughput simulations, as a first step to increasingly combine experimental and simulated data in one framework.

With data from both in the same framework, a heretofore untapped and much-needed potential for the acceleration of materials characterization and materials discovery campaigns becomes available.

Keywords: active learning, gaussian process regression, research data management, automation, autonomous discovery

1 Introduction

Computational materials science has developed from single calculations to high-throughput (HT) simulation campaigns in recent years. This development is based on access to increased computational power and the development of simulation tools that allow for increasingly more complex simulation protocols. But these capabilities present new challenges to managing calculations and the produced data. In computational materials science, these challenges are addressed with integrated development environments such as pyiron [1] or AiiDA [2] and other automation tools [3–8]. Major benefits of such approaches include automation, reproducibility, data provenance, and ensuring accessibility and reusability of existing data.

Experimental materials science is currently undergoing a similar shift from manual, human-guided sample-by-sample synthesis and testing of individual specimens towards HT (combinatorial) synthesis combined with HT characterization. Materials discovery in particular necessitates the screening of a technically unlimited amount of compositions and processing routes for specific functional properties [9].

Samples in HT synthesis and characterization typically come in the form of composition spread materials libraries (CSML) [10]. Conventionally, CSMLs are characterized by measuring chemical composition, structure, mechanical, optical, electrical, etc. by (semi-)automated measurement systems on a given number of pre-defined measurement areas, typically several 100s. This degree of automation reduces the input of scientists and the time for measurements in the characterization and analysis workflow. Similarly, the analysis of the resulting relatively large datasets is challenging and often presents a substantial part of the total time needed for characterization. With several measurements on one CSML sample, combinatorial synthesis and HT characterization have similar challenges for data management [11] as HT simulation campaigns and the benefits provided by integrated development environments (IDEs) are equally desirable for experimental data and workflows. Experimental data is often generated on distributed measurement instruments without or with limited metadata or data formatting standards. And, similar to the approach taken for simulations, increasingly more automation and orchestration is incorporated into experimental materials science, e.g. BluesSky [12], ChemOS [13], or HELAO [14], including the benefits valid for HT simulation campaigns when using integrated development environments. A recent review

of approaches can be found in [15]. What is, however, still missing from all existing experiment-focused approaches is an interface to the computational domain.

Increasingly larger degrees of automation in experimental synthesis and characterization workflows provide the path to use the same optimization strategies employed in simulation workflows to accelerate the discovery and design of materials. Once data from both domains exist in the same framework, leveraging the strengths of both approaches simultaneously becomes possible and provides further opportunities for acceleration. Instead of triggering a calculation based on a suggestion provided by an optimizer, one can easily envision alternatively triggering a synthesis [16] and/or characterization routine to obtain the next (*real*) material's properties in a discovery cycle. Ultimately, a global optimizer in a discovery cycle can then autonomously choose the *best* next step w.r.t. cost, time, or uncertainty: either trigger a simulation or synthesis plus characterization of a real sample. Further potential for optimization and acceleration lies in the possibility directly accessing all previously measured and calculated properties and exploit existing knowledge to guide the optimization in the *current* search space.

On the path to fuse data from simulations and experiments in one framework, we present a first demonstrator using pyiron [1] for experimental data acquisition. pyiron is an integrated development environment (IDE) for creating workflows [1]. It combines a job management system for automation, and a hierarchical data management solution originally designed for atomistic modeling and is implemented in Python. Its modularity allows to add custom jobs that make use of the built-in data management capabilities with minimal overhead. Our demonstrator is designed to show that the concept of an IDE, developed for simulation workflows, can (a) be applied to experimental data acquisition, (b) directly optimize the measurement using an active learning strategy based on Gaussian Process Regression (GPR), (c) analyze the data using automated data processing or data analysis routines [17, 18], and (d) automatically solve the issue of data management and storage.

Optimizing measurements in our context means decreasing the number of actual measurements while predicting/interpolating unmeasured regions in the search space with a defined uncertainty. This directly translates into an acceleration of a characterization cycle, ultimately accelerating the materials discovery cycle. pyiron takes the role of an *orchestrator* running the GPR, querying the measurement device for the next optimal measurement point as well as storing and managing data. The application of GPR works because the composition-structure-property relationships often show smooth trends that can be approximated by relatively simple functions of the composition, which is often the case for material systems that form solid solutions over a wide composition range, such as high entropy alloys [19, 20]. Employment of an adaptive sampling strategy to approximate (instead of fully measure) a property as a function of the composition of CSMLs with a model based on a very

small number of measurements results in an order of magnitude fewer measurements than brute-force automated approaches need as we show here. The potential of integrated experimental-computational materials discovery has been demonstrated for organic materials [21], nuclear materials [22, 23], and is being considered to accelerate the discovery and optimization of urgently needed materials, e.g. for catalysis [24].

Recent developments in the experimental field aim to integrate such workflows in automated, closed-loop discovery cycles, e.g. by using robotic platforms with integrated algorithms for data analysis and hypothesis generation [25]. It is envisaged that such platforms will enable fully autonomous research systems for materials discovery [26–28]. Fully automated robotics platforms may be used in certain cases, however, it will be a long way to integrate several, especially high-quality in-depth characterization techniques (e.g. transmission electron microscopy, atom-probe tomography), and retrofitting existing equipment. Nevertheless, it is possible and useful to create an integration of synthesis-characterization-evaluation cycles at the data level to unlock synergy effects between multiple data sources and to create partially (offline) autonomous research systems. Ideally, computational and human resources should work collaboratively in such environments [17]. The main difference between these systems and our approach, however, is the direct integration of simulations in the same framework as experimental measurements to benefit from all accessible knowledge to accelerate the materials discovery cycle.

Our demonstrator first and foremost shows that it is possible to include an experimental workflow in pyiron. We demonstrate a useful acceleration of property measurement and benefit from the already-implemented data storage solution. This is a step towards on-line data fusion from experiments and simulations with the potential to markedly accelerate materials discovery cycles. The bigger picture is that an IDE such as pyiron has the potential to become an integrated platform for materials science with access to data from simulations and experiments and computer-assisted data analyses.

2 Results

The demonstrator implementation of an active learning loop in pyiron communicates with an offline experimental dummy device that provides resistance measurements based upon query [29]. The implementation has three ingredients: 1) pyiron as a basis to manage data, 2) a bespoke `ResistanceGP` "job"[1] which steers the experiment, similar to a single simulation, and 3) a custom interface to a measurement device. pyiron itself is detailed in other publications [1] and we, therefore, focus on the two other ingredients. A "job" in pyiron is a single calculation. Concretely, the "job" here is an optimized measurement of the resistance on a CSML. Its input comprises the user name, an identifier of the measurement device, a sample id, an initial parameter set including the maximum number of iterations for regression, and in our case

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[1] "job" here is the pyiron-internal name for a defined workflow.

a file path that points to a completed measurement (composition and resistance) to initialize the dummy measurement device which provides the actual *measured* values upon request of the running job. An overview is presented in Tab. 1. We want to stress that the choice is tailored to demonstrate the functionality and is not final. Since the logging is done automatically we can add any number of parameters automatically to a job through communication with a measurement device or a script triggering the characterization. Ideally, each component communicates its settings at runtime, and all settings needed for the reproduction of the results are automatically saved along with the acquired data with minimal manual user intervention. In the future, this could go as far as adding bar codes or radio-frequency identification (RFID) tags to samples, devices, and users for minimal manual intervention.

Table 1: List of input parameters for the pyiron experimental job for optimized resistance characterization of the demonstrator.

Parameter	Description
`exp_user`	Name of the scientist
`measurement_device`	A unique name of the measurement device used
`sample_id`	Identifier of the physical sample
`measure_indices`	User-defined coordinates on CSML to initialize the GP
`sample_file`	Path to data to initialize experimental dummy device
`max_gp_iterations`	Maximum iterations for GP

Within the "job", a custom interface to the measurement device is initialized and a Gaussian process regression based on `GPy` [30] is invoked. Uncertainty sampling is applied in which the algorithm determines the next measurement as a function of chemical composition which is translated into spatial x/y coordinates of the physical materials library. The algorithm chooses the composition for which the model predictions has the highest uncertainty. Five resistance measurements on the CSML are used to initialize the GPR. This initial choice is typically defined by user input but if prior data on a subset of the composition space exists, this can serve as a prior without user input. The job subsequently requests the next measurement on the CSML by evaluating the composition for which the GPR predicts the highest uncertainty in predicting the resistance. It runs until a stopping criterion is reached and the acquired data is automatically stored using the data management solution provided by pyiron. Two possible stopping criteria can be envisioned: 1) maximum value of iterations (currently used); or 2) a threshold for the largest tolerable uncertainty.

From the perspective of pyiron, the only difference between a simulation "job" and a measurement "job" is the source of new data. The former source of data results from computational procedures and high-performance computing resources, the latter source of data is the output of a measurement device.

Fig. 1 schematically shows the described extension to pyiron and its seamless integration to the environment.
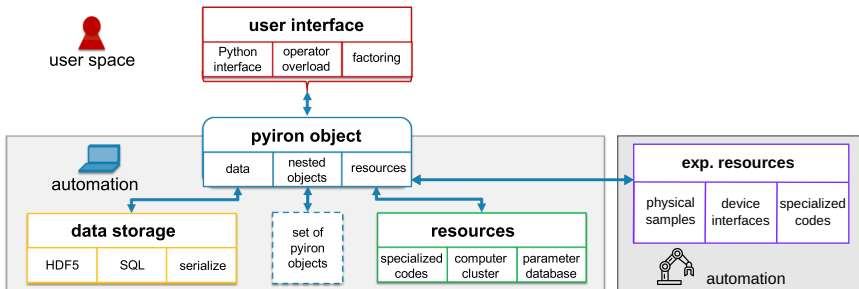
Fig. 1: Internal structure of a pyiron object with the experimental extension on the right. The described extension to experimental workflows makes use of the existing infrastructure and is seamlessly integrated. Adapted from [1] published under CC BY 4.0 license (http://creativecommons.org/licenses/BY/4.0).

The composition and the measured electrical resistance of each point of the CSML used here for the demonstrator is shown in Fig. 2 and published on Zenodo [31].

Composition gradients are created by co-depositing from five pure elemental targets (Ru, Rh, Pd, Ir and Pt) (cf. Fig. 2 a-e). Electrical resistance was measured by a four-point probe on a 342 point grid and is shown in Fig. 2 f. The observed resistance trend is a relatively smooth gradient as a function of the composition, which motivates the use of an active learning scheme to reduce the number of measurements.

Fig. 3 presents the details of the GP-optimized measurement. The indicated numbers show the chosen measurement positions to initialize the GP (red) and subsequent choices (black) of measurements based on the highest covariance value of the update prediction. Mean values and covariance of the predictions are shown for iterations $n = 6$ and $n = 40$. The mean predictions vs ground truth are shown for iterations $n = 6, 12, 24, 36$ as well as the mean absolute error evolution as a function of iterations. It is apparent, that the prediction improves markedly up until ≈ 40 iterations at which point we stop the optimization.

3 Discussion

The presented example of a pyiron-controlled experiment with a dummy experimental device in the loop shows the potential for future applications. Instead of brute-force measuring the resistance of all 342 compositions provided by the discretization of the materials library, only 40 compositions are measured and the rest is interpolated including the uncertainty for the interpolation. Consequently, the saved characterization data of the CSML includes 40 actual
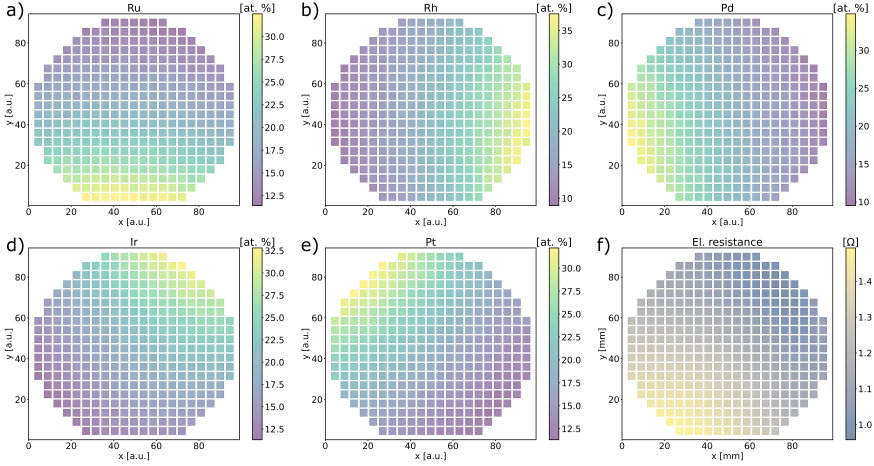
Fig. 2: Chemical composition of the quinary noble metal system Ir-Pd-Pt-Rh-Ru CSML (a-e) in atomic % and electrical resistance (f) for the 342 measurement areas.
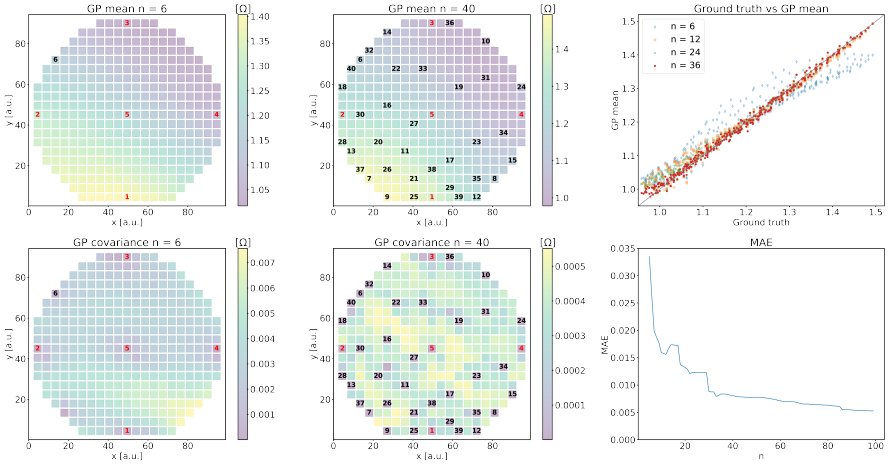
Fig. 3: Iterations of the Gaussian Process Regression for the prediction of the electrical resistance as a function of composition for the Ir-Pd-Pt-Rh-Ru CSML: Mean values and covariance of the prediction for iteration $n = 6$ (a, d) and $n = 40$ (b, e). $n = 40$ represents the final choice with sufficient confidence. Red numbers indicate data points used for initialization of the GP, black numbers indicate the subsequent iterations based on the maximal uncertainty of the prediction; mean predictions vs ground truth for iterations $n = 6, 12, 24, 36$ (c) and the evolution of the mean absolute error (MAE) as a function of iterations (f).

measurements and 302 interpolations (predictions) that are marked as such including the model used for interpolation, translating into an acceleration of the process of one order of magnitude. A typical time to obtain one measurement is $\approx 10\,\text{s}$ including moving the measurement tip to the position. I.e. the measurement of one CSML with 342 measurement points is about one hour. Measuring only 40 points translates to $\approx 7\,\text{min}$. We state this matter-of-factly but the consequences on how materials data could be used for accelerated characterization in the near future need to be stressed. In the context of materials discovery for combinatorial problems, this is the quality of acceleration that is needed.

But the real benefit lies in the central storage and reusability of the data. Imagine a discovery campaign is started in a compositionally (partially) overlapping material system. Any overlap with a previous measurement potentially accelerates the characterization of new systems. Uncharacterized regions of a composition space could even be explored autonomously without the need for measurements at first. Autonomously here refers to an automatable procedure in the framework which routinely checks overlaps in compositions of stored measurements to automatically predict property behavior. An example of such an overlap is schematically shown in Fig. 4a. Two materials libraries exist in the database, one containing a property measurement of elements A and B, a second one containing B and C. The overlap is given by element B. These two measurements in two binary systems comprise the edges of a hypothetical ternary system A-B-C.

With an increasing number of measurements stored in the same accessible database, the larger this benefit becomes and actual sample preparation can then concentrate on "very high uncertainty" regions. A possible pitfall of the models for predicting unmeasured properties is that they all might use different local coordinate systems of elements. In our demonstrator the model is

$$R = f(c_{\text{Ru}}, c_{\text{Rh}}, c_{\text{Pd}}, c_{\text{Ir}}, c_{\text{Pt}}), \tag{1}$$

where R is the resistance and c_i the respective compositions. But a GPR, as presented here, can easily be adapted to different element-based coordinate systems by treating the measured and interpolated known values as given, adapting the coordinate system, and setting any additional element c_{add} to zero:

$$R = f(c_{\text{Ru}}, c_{\text{Rh}}, c_{\text{Pd}}, c_{\text{Ir}}, c_{\text{Pt}}, c_{\text{add}} = 0) \tag{2}$$

to fit a model. With such a model, a complete edge or multiple edges new materials system can be used as priors. Fig. 4b shows this schematically for the individual two measurements presented in Fig. 4a. Two CSML with elements A-B and B-C are present as data sets, the characterization of the ternary A-B-C can then be initialized with known properties on the edges. This could even be done automatically. E.g. if samples are tagged with an RFID chip, any existing characterization data and possible priors like energy dispersive X-Ray (EDX) measurements could be looked up in the database. If EDX data is
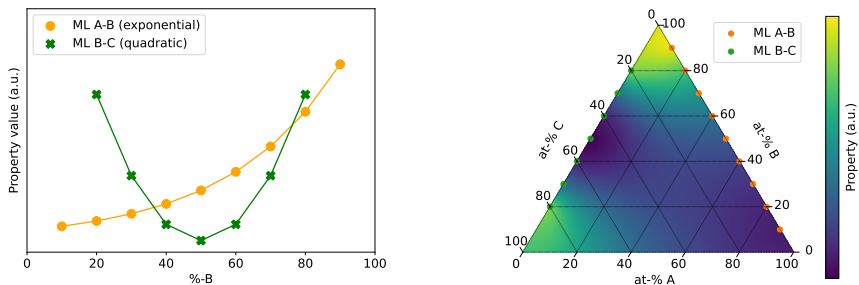
(a) Schematic example property measurements of two materials libraries with elements A-B and B-C.

(b) Schematic of a ternary search space with *edge* data (A-B and B-C) already measured and the rest interpolated.

Fig. 4: Schematic property measurement (a) for two binary systems with each two elements A-B and B-C shown both as a function of the elemental of element B. The property measurement on the library A-B shows an exponential dependence on B-concentration, B-C is quadratic for two binary systems. And (b) the possible property prediction on the ternary prior to any measurements. The characterization of the *full* A-B-C compositional space can be initialized with the existing dataset: The green and orange points would substitute the initial choice of measurements (marked with red numbers) in Fig. 3. This concept is directly applicable to material systems with more elements. A ternary was chosen solely for presentation purposes.

present, this can be used as a prior in GPR, if not, some other sample positions are suggested to be measured first to initialize the GPR.

If characterization data from less complex binary and ternary materials from CSMLs are present, more property predictions for materials with more constituents can be assembled. Given sufficient data and limited expectations about the certainty of a prediction/interpolation, further measurements could even be unnecessary to explore an unknown composition space. In all cases, this requires strict bookkeeping of where material properties originated (measurement, prediction, simulation) and propagation of uncertainty for which frameworks like pyiron are ideal.

Another possibility for informed initialization (improve the "prior") is to directly trigger simulations from an experimental job, which provide a first approximation of the composition-property to be measured and use that as a prior. More related data available prior to any measurement provides a possibility to accelerate the characterization. And the possibility to either trigger simulations or other cheap proxy experiments prior to expensive measurements only exists if both domains are unified in one framework.

Before this can become a standard, several technical, as well as *acceptance* challenges need to be overcome. Practical technical challenges include

that many experimental devices are controlled by proprietary software without APIs, effectively rendering them unusable in the outlined approach. Solutions to this issue are obvious: Either a vendor provides an API through which a device can be controlled or an open-source operating system is run to control the device directly. We expect that this will be solved with time. Operationally, however, and this is a drastic change in how data and data management will be seen: researchers need to be aware and accept that not every data point in the database is actually measured, but most of them are interpolations/predictions. This is in part a communication issue but in our view a larger part an issue of trust in software solutions. Because in the outlined solution a model provides most of the data; in our demonstrator $\approx 9/10$. This model as a mathematically correct concept and, even more important, its correct implementation needs to be ensured. The former is a theoretical problem, which is in principle solved, the latter requires strict standards for implementation through unit tests, continuous integration tools, and benchmarking during software and method development [32–35].

The last point shows a current gap between the simulation and experimental materials science domains. Computational materials scientists are used to using (legacy) code and routinely test and trust software. Models, their implementation in computer code, and computational resources are the *tools of the trade.* Many computational materials scientists go through formal or informal training for coding and software development along with their materials science training which allows them to check their own as well as others' codes and their physical validity. Researchers trained in experimental sciences also undergo a formal training, e.g. in sample preparation and device control, but usually, only a few have the necessary background to assess, use, and change codes and use computational resources which in turn can result in less or no trust in the interpolated results. But the solution for this is also clear: as the experimental and simulation domains are merged on a data level, the formal (or informal) training of researchers also needs to be merged. Experimentalists who use these tools and frameworks need to be trained. And while the initial overhead of time spent for training might be seen as a drawback, the potential for acceleration of this approach in the context of materials discovery is real and becomes larger as more data is collected in the unified framework. Further, recent developments in lab automation [23, 28, 36] will lead to less training spent to operate devices. Adding a new experimental data point will then ideally be as simple as running a simulation, the call of a function within a framework like pyiron.

In summary, the use of a unified framework as outlined puts less importance on how data is acquired (simulation, measurement, prediction) and more importance on how data is stored and that it can be reused in subsequent research.

4 Conclusion

We demonstrate that an integrated development environment designed for use with high-throughput simulation is also able to control an experimental procedure, similar to triggering a calculation on a high-performance computing platform. With this approach, the boundaries between data obtained from experiments or simulations vanish because *data* does not care about its source. This approach allows to leverage computational power to accelerate materials characterization, here demonstrated by using Gaussian process regression to reduce the number of measurement points by approximately one order of magnitude while predicting non-measured points with the desired uncertainty. By embedding experimental workflows in pyron we automatically benefit from automation, reproducibility, and data provenance, and at the same time ensure accessibility and reusability of existing data. Accessibility and reusability are key ingredients for further acceleration of characterization because an existing database provides prior knowledge to initialize, e.g., a Gaussian process more strategically instead of manually or randomly. This is the first step towards automatically fusing data from different sources, which allows a big-picture optimization of materials discovery campaigns. Necessary changes in software and hardware accessibility for this vision to become a reality are APIs to experimental devices and training of researchers to create, trust, and use workflow managers for data acquisition.

Ultimately, we envision a (software) layer in which a federation of algorithms (sometimes also referred to as "agents") use results obtained by other agents as prior information for autonomous characterization, thereby lowering the number of needed measurements to ultimately speed up characterization time without compromising (too much) on data precision or sampling. The final characterization of a given material is then a mix of actual measurements and predictions each with its associated uncertainty. Reproducibility and confidence in data is ensured because each step of the procedure is automatically documented by the workflow manager.

5 Methods

pyron extension We extended pyron in the following way. First, we added a new 'job' that inherits the basic functionality and integration to the installed pyron instance automatically. Necessary additions include the definition of user-defined input variables, a recipe for the Gaussian process-based measurement, and functionality to retrieve the measured as well as the interpolated data along with the model. The input variables currently comprise a user, a measurement device name, a sample ID, initial measurement points for the Gaussian process, a sample file, and the maximum number of iterations (Tab. 1). In a future iteration, this user input will serve as a basis for the automatic generation of metadata for datasets and will grow and change over time as more devices and measurement routines are implemented. The version used

in this paper is published here [37] along with an example notebook on how to use it.

Experimental dummy device and wrapper for GPy

The experimental dummy device simulates an API to an experimental setup and is implemented as a simple class in Python and can be found in a utility repository [29]. Its API mimics the call to an actual device and returns the desired property (here the resistance) of a certain composition on a materials library value. This API to a dummy device will be replaced by an API to actual experimental setups once they become available. Additional to the dummy device, a wrapper for the Gaussian processes framework GPy [30] is also included here. The wrapper is a class that allows convenient initialization, update, and prediction of a Gaussian process as implemented in 'GPy'.

Snythesis and characterization of composition spread materials libraries

The CSML was fabricated by co-deposition from five pure elemental targets. The fabrication details are provided elsewhere [38]. The electrical resistance was measured using an automated four-point probe test stand. The setup is described elsewhere [39]. The chemical composition was measured by energy-dispersive X-ray spectroscopy in a scanning electron microscopy Jeol 5800 LV equipped with an Oxford X-act detector. The acceleration voltage was set to 20 kV. The final data set of CSML used in the dummy experimental device with chemical characterization and resistance measurements is published on Zenodo [31].

Data availability The experimental data used in the demonstrator is published on Zenodo [31].

Code availability The code used in this study is available here [37]. The implementation of the experimental dummy device is available here [29].

Author contributions MS, LB, and AL designed the study. NS carried out the measurements and developed a first prototype of the Gaussian Process Regression. MS formalized the prototype and developed the utility library. MS, JN, LB, and NS implemented the custom pyiron job. All authors analyzed the results and wrote the manuscript.

Competing interests The authors declare no competing financial or non-financial interests.

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