APPLICATIONS OF AUTONOMOUS DATA COLLECTION AND ACTIVE LEARNING

Towards Automated Design of Corrosion Resistant Alloy Coatings with an Autonomous Scanning Droplet Cell

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We present an autonomous scanning droplet cell platform designed for on-demand alloy electrodeposition and real-time electrochemical characterization. Automation and machine learning are currently driving rapid innovation in high-throughput and autonomous materials design and discovery. We present two alloy design vignettes: one focusing on a multi-objective corrosion resistant alloy optimization and a study highlighting the complexity of the multimodal characterization needed to provide insight into the underlying structural and chemical factors that drive observed material behavior. This motivates a close coupling between autonomous research platforms and scientific machine learning methodology that blends mechanistic physical models and black box machine learning models. Finally, we reflect on our early efforts in on-demand alloy deposition, highlighting some of the challenges. This emerging research area presents new opportunities to accelerate materials synthesis, evaluation, and hence discovery and design.

Key words: Corrosion resistant coatings, Alloy design, Automation, Machine learning

INTRODUCTION

Integrating experiments and theory-based modeling to understand and design complex multiscale materials is an important and perennial research topic in materials science and engineering. The recent rekindling of interest in applications of machine learning (ML) to materials science problems has intensified this focus on integrating computation and experiments. This is clearly illustrated in the nascent autonomous materials science and discovery community, which has a strong focus on orchestrating automated experimental and computational materials science experiments through adaptive machine learning systems for data evaluation and experimental planning.

The fields of active learning and design of experiments use ML models to inform a series of decisions about which valuable data to obtain, through either experiments in the lab or on-demand physics based modeling of alloys, condensed-phase materials, and molecules. The autonomous materials experimentation community is consistently demonstrating the power of fully closing the synthesize-characterize-predict loop across a diverse and growing portfolio of materials technologies. Notable examples include optimizing the growth rate and thereby quality, of carbon nanotubes by tuning chemical vapor deposition conditions, optimizing the strength of additively manufactured polymer lattices by varying the three-dimensional geometry...
of their superstructure, and autonomously mapping out non-equilibrium processing phase diagrams.

In addition to directly optimizing material properties, there is promising progress in robotization for learning to fabricate materials and chemicals with desired structure and for efficient on-demand acquisition of complex and/or expensive experimental data, for example, synchrotron x-ray measurements of nanoparticle density and accelerated structural phase map acquisition.

This article explores some of the most challenging aspects of closed-loop materials design. Material properties are linked to process and composition via complex relationships, mediated by structure and chemistry across multiple length scales, such as phase distribution, density, microstructure, and surface quality. Integrating enough materials synthesis and characterization capabilities into experimental systems to address such complex materials phenomena is a substantial challenge. A major roadblock to incorporating this complexity into autonomous materials research platforms is addressing the need for quantitative data analysis at scale, which with the advent of high-throughput material synthesis and measurement systems is often a primary research bottleneck. This is an opportunity for mutual progress in autonomous materials research and scientific machine learning, which is the subfield of ML concerned with designing models and algorithms imbued with physics-based bias and model structure. Early materials research in this area focused on explicit incorporation of domain knowledge into model form and incorporating automated reasoning and physical constraints into machine learning algorithms. A particularly important benefit of incorporating explicit physical insight into ML models is the potential for substantial efficiency improvements by directly targeting scientific goals through the acquisition policy of an active learning system, for example, in measuring a magnetic transition temperature via neutron diffraction.

Often, even once desirable composition and structural features are known, the path to actually fabricating usable material with these properties is unclear, presenting an opportunity for creative automated systems to have high impact. For example, the Chemputer project aims to fully automate organic synthetic chemistry with general-purpose hardware and to algorithmically discover efficient synthesis routes for any possible target molecule. Focused on metals, Ref. outlines a diverse set of synthetic approaches spanning composition and thermomechanical gradient techniques, batch casting and additive manufacturing methods, and roboticized microscopy platforms. For addressing complex materials systems dominated by multiscale structure, modular clusters of multiple synthesis and characterization tools may provide a path forward.

We believe that electrochemistry provides a rich opportunity and testbed for development of automated tools to collect datasets and demonstrate closed-loop materials development. Electrochemistry provides a unique method that enables both synthesis (electrodeposition) and characterization (electrochemical corrosion) using the same experimental apparatus. Furthermore, electrochemical processes are complex and difficult to model using first principles and thus are ideal for modeling using ML methods. A wide variety of high-throughput experiment approaches for electrochemical systems have been explored including scanning droplet cell (SDC) systems. Here we describes our efforts to demonstrate the value of this type of platform through a series of vignettes related to our development and demonstration of a highly automated and closed-loop SDC tool. We hope this work serves to demonstrate challenges and highlight promising directions.

METHODS

Automated Scanning Droplet Cell

Our high-throughput scanning droplet cell platform, described in detail in Refs. and (illustrated in Fig. 1a), integrates a compact electrochemical flow cell with a fully automated bank of syringe pumps and scanning sample stage. The flow cell defines a roughly 4.5 mm diameter circular footprint on the active surface. The cell can be addressed to any point across a planar sample (e.g., a uniform or composition spread thin film, or a bulk alloy sample) and conduct an agile automated serial electrodeposition and/or corrosion experiments under a variety of solution conditions. As such, this tool enables online optimization of electrodeposited alloy composition by programmatically adjusting solution chemistry and applied potential or current (as briefly discussed in Sect. 4). In addition, corrosion assays can be performed as a function of composition or structure on these electrodeposited samples or (as in Sect. 3.1) with composition gradient thin film and bulk samples. Rapid characterization of potential and pH behavior can also be performed.

Figure 1b illustrates the core adaptive alloy electropolating optimization loop. An initial alloy thin film is deposited with candidate settings for solution composition and deposition conditions (applied current, flow rate, etc.). Online process monitoring of variables including measured potential, current, pH, and temperature can provide early indication of automation failure or infeasibility of the candidate process settings. A central challenge of building autonomous materials research platforms is integrating as much online characterization capability as possible. Our system currently performs routine macroscopic surface image acquisition via optical camera (Fig. 2a) and laser (635 nm wavelength) reflectance line
scans (Fig. 2b) to assess the continuity, coloration, uniformity, and qualitative roughness of electrodeposits. Our platform has a modular design, allowing it to readily be rebuilt around and incorporated into a synchrotron measurement station, enabling online acquisition of, e.g., x-ray fluorescence (Fig. 2c), diffraction, and absorption spectroscopy data.

After any online chemical and structural characterization, a wide variety of corrosion resistance assays are possible. The corrosion environment can be tailored along pH and electrolyte composition axes through flow mixing, and in principle a dynamically specified series of electrochemical experiments can be performed.

Finally, the experimental loop is closed by linking processing variables and any measured chemical, structural, and performance quantities through a probabilistic machine learning system, such as a Gaussian process (GP), linking composition to multiple corrosion figures of merit illustrated in Fig. 1b. Active learning algorithms use the model predictions and associated uncertainties to prioritize subsequent candidate experiments to maximize the probability of learning valuable information towards achieving a design or discovery goal.

### Modeling

Throughout our work, we use the Gaussian process (GP) modeling framework for its flexibility, intrinsic treatment of predictive uncertainty, and facility for automatic hyperparameter optimization. GPs are Bayesian machine learning models that are similar in spirit to a kernel version of Bayesian linear regression; for a comprehensive and accessible introduction, refer to Ref. 41. GP model specification is often described in terms of building a Bayesian prior over functions that might possibly describe the data. This is decomposed into two elements: the mean of the function prior, $m(\mathbf{x})$, where $\mathbf{x}$ is the vector of inputs, and the covariance (or kernel) function $k(\mathbf{x}, \mathbf{x'})$, which controls the
distribution of possible functions that the model can represent. A GP regression model for some target data \( y \sim f(x) + \epsilon \) can be written as \( y \sim \mathcal{GP}(m(x), k(x, x')) + \epsilon \), where \( \epsilon \) represents the Gaussian error term common to most regression models. In this work, we default to the commonly used constant mean GP model with “squared exponential” or “radial basis function” (RBF) covariance function: \( k(x, x') = s^2 \exp(-\frac{1}{2}\|x - x'\|^2/\ell^2) \). The resulting GP model has several hyperparameters that we tune by gradient-based optimization to maximize the model evidence \( p(y \mid x, \theta) \), where \( \theta \) represents the collection of model hyperparameters.\( ^46 \) The principal hyperparameters for an RBF GP are the amplitude parameter \( s \) that controls the overall scale of the model functions, a lengthscale parameter \( \ell \) that controls the level of fluctuation relative to the input space, and the observation noise level \( \epsilon \).

GP models are commonly used in active learning and optimization settings because they are well suited to automatic hyperparameter selection and provide good predictive uncertainty estimates. There are many active learning strategies for selecting experiments based on probabilistic model predictions. The relative performance of these strategies can strongly depend on the characteristics of the data and the active learning task. In the optimization examples presented here, we consistently use the confidence bound strategy,\( ^42 \) which balances candidate selection between high predicted utility and high predictive uncertainty. At iteration \( t \), an experiment is selected from the design space that minimizes the quantity \( \mu(x) - \beta(t) \sqrt{\text{var}(x)} \), where \( \mu(x) \) represents the mean prediction from a GP model, \( \sqrt{\text{var}(x)} \) represents the predictive uncertainty, and \( \beta(t) \) is an iteration-dependent scaling factor that controls the tradeoff between exploring high-uncertainty regions of parameter space and prioritizing promising candidates for global minima. We follow Ref. 43 in using the tradeoff schedule \( \beta(t) = cd \log(2t + 1) \), where \( c = 0.25 \) is a fixed hyperparameter of the confidence bound method, and \( d \) is the dimensionality of the input space. We apply the random scalarization\( ^43 \) method for weighting competing objectives in a way that encourages fuller exploration of the Pareto frontier.

**RESULTS**

**Multiobjective Corrosion Property Optimization**

Figure 3 shows a multiobjective optimization case study based on a high-throughput benchmark dataset of corrosion experiments in a neutral NaCl solution on an AlNiTi composition spread. The details of data collection and analysis, described briefly below, can be found in Ref. 39. Figure 3a illustrates a typical linear scan voltammetry (LSV) result from this series of measurements, showing the measured log current (I, amps) as the potential, \( V \), is increased linearly in time. \( V_{oc} \) is the open circuit potential. We fit a linear model in \( \log(I) \) space to the passivation plateau at positive potentials relative to \( V_{oc} \); the departure from this linear behavior is the transpassive potential \( V_{\text{tp}} \). We determine an effective passivation potential \( V_p \) and passivation current \( I_p \) by locating the median potential within the linear passivation plateau.

Finally, we characterize the “flatness” of the passivation plateau, defined as the inverse slope of the linear model, slope = \( \frac{d \log(I)}{dV} \).

Figure 3b and c shows the obtained \( I_p \) and \( \frac{d \log(I)}{dV} \) for this dataset. To simulate data collection, we interpolate each measured property with a GP model which we can interrogate: for each simulated Bayesian optimization campaign we sample ground truth response functions from the GP posterior fitted to the full dataset, adding observation noise to the simulated measurements to mimic experimental uncertainty. The multiobjective optimization goal is to identify alloys with low passivation current \( I_p \) and a flat passivation plateau slope (i.e., low \( \frac{d \log(I)}{dV} \)).

We model the corrosion response with independent GPs over composition for the two target variables, \( \log(I_p) \) and \( \frac{d \log(I)}{dV} \). Each GP uses a constant mean function and an RBF kernel defined over composition variables. Measurements are selected with a lower confidence bound strategy,\( ^42 \) we use the random scalarization\( ^43 \) to encourage full exploration of the pareto front. At each iteration, the two objective function weights are drawn from Beta(2, 2), providing a mild preference to avoid compromising on either objective.

Figure 3d compares the performance of the active learning system with a random search strategy. The active learning system shows consistent convergence towards each individual objective in < 10 active learning queries across a benchmark of ten simulated active learning runs. The random search strategy achieves similar results in the long run, but takes nearly twice as many function evaluations to converge and has higher variance, especially in the first ten iterations. The bottom panel quantifies the joint performance on both objectives through the data-dependent term of the Bayes regret, as in: 43

\[ \mathcal{R}_B(T) = \mathbb{E}_{\lambda \sim p(\lambda)} \min_{x \in \mathcal{X}} s_\lambda(f(x)) \].

This is the best observed value \( x \) out of observed values \( \mathcal{X} \), averaging over the random scalarization process \( s_\lambda \), where \( \lambda \) is the objective function weighting drawn from its distribution \( p(\lambda) \) (here a Beta(2, 2)).

In this system, Al strongly suppressed \( I_p \), while Ti is associated with flatter passivation plateau behavior. The passivation slope is a more challenging function to optimize: the length scales of the ground truth GP models for \( I_p \) and the passivation slope are 0.2 and 0.083 (element fraction units). As a result, the active learning system achieves favorable performance on the \( I_p \) objective, particularly with an
experimental budget < 10, but only marginal progress directly optimizing the passivation slope. This apparent difference in optimization difficulty is consistent with a heuristic estimate of the number of experiments needed to fully specify a GP model, following:\(^\text{44}\) the lengthscale of the RBF kernel used in the GP models can be interpreted as the standard deviation of a Gaussian distribution. Dividing the volume of the design space by the volume contained within one standard deviation of this Gaussian distribution yields an estimated experimental cost of eight samples for the \(I_p\) objective and 45 samples for the passivation slope. One limitation of this heuristic is that the passivation current model has a higher estimated noise level, which may increase the number of samples needed to reliably estimate the underlying function. The overall performance measure, the data-dependent Bayes regret \(R_B(T)\), shows that despite the difficulty in finding the optimal passivation slope, the random scalarization algorithm focuses on finding good compromises between both objectives.

Figure 4 illustrates the progress of a multiobjective optimization trajectory towards exploring the full set of tradeoffs between median passivation current and its range over the passivation plateau. These Pareto plots show the observed values of each objective in a multi-objective optimization problem to succinctly illustrate the set of possible design tradeoffs. Though the initial observations are nearly optimal when considering each objective individually, the approximation of the overall Pareto frontier is poor. However, after four active learning queries (Fig. 4b), the Pareto frontier approximation is within the predictive uncertainty of the GP models. Subsequent observations concentrate more densely along the Pareto frontier and reduce the predictive uncertainty of the GP models as they learn better approximations of the target functions.

### Semi-Mechanistic Modeling

Empirical optimization of material structure and processing is an important research area, but black box optimization approaches, those without physics embedded in them, are not well suited to providing the kind of mechanistic insight that drives the development of new materials theory and design heuristics. This is perhaps the most important challenge facing scalable automated science today: bringing together multiple sources of information to decouple the underlying chemical and structural factors that give rise to the aggregate material properties that we observe.

Figure 5 shows (a) the corrosion response (log corrosion current, \(I_{\text{corr}}\)) across a TiNbTa composition spread thin film measured by the SDC alongside (b) crystallite size and (c) microstrain, defined here as \(\text{RMS}(\Delta d/d)\) where \(d\) is the lattice spacing, obtained by Williamson-Hall size-strain analysis\(^\text{45}\) of synchrotron XRD data on the film. The system nominally forms a body-centered cubic (BCC) solid solution, with secondary phase inclusions, determined by subjective inspection of XRD data for unattributed peaks, appearing on the Ta-poor side of the solid black curve.

Perhaps unexpectedly, the least favorable corrosion rates in Fig. 5a are found in the single phase, near-equiatomic portion of the ternary system. The composition dependence of the crystallite size and microstrain suggest that these microstructural features could play a role in modulating corrosion response. The influence of microstructural features such as grain size\(^\text{46}\) and precipitate size and density\(^\text{47}\) is well established.

To address these complex mechanistic materials science questions and disentangle potentially competing effects, models with finer granularity are needed. For example, the partial dependence analysis of the artificial neural network model in Ref. 48 provides insight into the contribution of composition and microstructure variables to corrosion behavior. However, the modeling approach is not amenable to explicit inclusion of theoretically motivated models.

Our approach is to blend theoretical models, such as the Hall-Petch grain size contribution of Ref. 46, with non-parametric models by specifying additive GPs:

\[
\log(I_{\text{corr}}) \sim \mathcal{GP}(\mu, K_{\text{comp}}) + k/\sqrt{d}
\]
μ and $K_{\text{comp}}$ are a constant mean and standard RBF kernel over composition variables, as in earlier sections of this manuscript. The grain size $d$ in nm is obtained from size-strain analysis, and $k$ is the linear coefficient in the Hall-Petch model, which we optimize along with the other GP hyperparameters.

Figure 6a shows the nominal log($I_{\text{corr}}$), along with the decoupled (6b) Hall-Petch contribution and (6c) composition contribution with grain size set to 40 nm. The composition contribution spans a larger dynamic range of log($I_{\text{corr}}$) values, and the location of the maximum value has shifted and spans a larger composition range compared to the nominal model.

**DISCUSSION**

Composing a complex, semi-mechanistic model as in Sect. 3.2 allows us to explore counterfactual predictions like “what if we could independently increase/decrease the grain size,” as in Fig. 6c. Similarly, competing mechanistic model components could automatically be evaluated, for example, with an alternative Hall-Petch-type model. With the present composition spread dataset, this model is merely descriptive, and it is difficult to validate these kinds of mechanistic hypotheses without the ability to independently vary composition and microstructure variables. Moreover, when the dataset does not contain sufficient variation, the model can be underspecified, leading to difficulty obtaining stable parameter estimates. Creating datasets that can fully span the range of composition and parameter space is in general
intractable with conventional experimental synthesis methods and even high-throughput methods. Meeting this challenge, therefore, represents a huge growth opportunity for autonomous materials science platforms. What is needed are agile automated platforms that (1) use semi-mechanistic models that provide quantitative insight into underlying material attributes that drive behavior, (2) can automatically and with low latency synthesize material with the desired attributes, (3) integrate enough online characterization streams to inform all relevant model components, and (4) use novel planning algorithms that use scientific criteria to plan experiments to optimally evaluate mechanistic hypotheses.

The nascent field of scientific ML will play a strong role in expanding the capability of autonomous materials platforms to broader scientific inquiry. One of the specific challenges to address in materials applications is the sometimes overwhelming complexity of interacting chemical and structural processes that mediate materials behavior. Consider the simple additive GP model in the TiNbTa vignette (Sect. 3.2). The model explicitly attempts to account for grain size effects, but many other important structural characteristics are absorbed by the non-parametric GP over composition. The crystal structure of the primary phase and presence of secondary phases (and their structure, volume fraction, size distribution, etc.) are expected to play a large role in corrosion resistance and other important material properties. Similarly, additional microstructural features like crystallographic texture and defect content and character can heavily influence material properties. Some of the aspects of materials structure have well-developed theoretical frameworks, while others may be treated in a largely empirical fashion. A major goal of materials-oriented scientific ML research should therefore be to integrate many different materials modeling types and approaches into a flexible ML framework that can bridge the gaps between mechanistic insight and empiricism.

To actually parameterize such models in an online fashion, continued creativity and innovation in automated online synthesis capability are needed to actually make suitable samples. This may constitute an active learning task of its own. For example, a priori specification of alloy electrodeposition conditions to obtain a targeted composition is a challenging task. There are competing thermodynamic and kinetic factors at play, and while there is a rich landscape of theoretical models, it is difficult to predict which mechanism will dominate, especially in a previously unexplored system. This is further complicated by a need to learn how to control microstructural factors like grain size, texture, and phase distribution independently from alloy composition, to the extent possible.

Figure 7 illustrates these issues using electrodeposition data from (7a) and (7b) Cu metal and (7c) preliminary alloy electrodeposition studies. The goal of the copper study is to learn the feasible processing envelope, depositing copper metal for 5 min at constant potential from 0.25 mol/L CuSO$_4$ onto a gold thin film. The GP confidence bound method is used to optimize (7a) the laser reflectance of the deposit (as in 2b) as a proxy for roughness and to minimize the average dissolution current over a 2-min constant potential hold at 100 mV vs Ag/AgCl reference in 1 mol/L H$_2$SO$_4$. The dashed line shows the predictions of a GP classification model for deposition coverage (sufficient/poor); the classification uncertainty is included in the active learning acquisition policy.

The NiCo alloy deposition study in Fig. 7c illustrates some of the challenges in electrodepositing alloys with targeted composition. Alloys were deposited at varying constant potential setpoints in solutions of 0.02 mol/L CoSO$_4$; 0.025 mol/L NiSO$_4$ (blue), 0.02 mol/L CoSO$_4$; 0.1 mol/L NiSO$_4$ (orange), and 0.02 mol/L CoSO$_4$; 0.15 mol/L NiSO$_4$ (green). Alloy compositions were determined offline though energy-dispersive spectroscopy (EDS) line scans across each deposit. The solid curves show expected alloy composition based on a simple linear
deposition current combination model based on single-component potential-current calibration curves. The EDS data span nearly the full binary composition range, but show substantial deviation from the naive expectations, including near insensitivity to deposition potential at the lowest solution concentrations. With online composition feedback, an active learning system may be able to quickly learn the nonlinear interactions between solution composition and deposition driving force to obtain desired alloy compositions.

In addition to developing innovative on-demand synthesis technologies, for maximum impact these platforms need to be tightly integrated with a diverse array of materials characterization technologies. For example, in the NiCo study, we not only want to learn to electrodeposit alloys with high-quality surfaces, but we may also wish to independently control grain size or target specific structural phases (either stable or metastable). Conventionally, the materials synthesis and characterization tools needed to produce all these data are decoupled from each other, and temporal latency between steps is high. Modularization and miniaturization are therefore a high priority for expanding the scope and impact of automated materials science. Wherever modular integration of critical characterization is not yet feasible (e.g., transmission electron microscopy), adopting and improving batch active ML algorithms will be important for integrating this information into research feedback loops. Additionally, quantitative high-throughput measurement of many important materials properties presents yet another category of important scientific ML sub-problems. For example, automated high-throughput phase identification and phase fraction analysis is still a major challenge, particularly in systems where minor and trace levels of secondary phases can potentially play a large role on effective material properties.

Finally, with interpretable scientific ML algorithms and the capability to make and characterize samples on demand, advances in scientific planning algorithms are ripe for high impact. An important first step might include algorithms for planning experiments to identify which latent structural factor is driving material performance. This could be coupled with model visualization and interroga-
DATA AVAILABILITY

The raw data required to reproduce the analyses in this manuscript are made available at Ref. 50.

CODE AVAILABILITY

The automation software for the scanning droplet cell platform is available at https://github.com/usnistgov/autoSDC. Code for the analyses in the manuscript is available at https://github.com/usnistgov/autoSDC-JOM.

CONFLICT OF INTEREST

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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