Towards Autonomous Nanomaterials Synthesis via Reaction-Diffusion Coupling

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Abstract

Reaction-diffusion coupling presents a pathway for producing nanomaterials that are precisely distributed in a reaction medium, with controlled gradients of chemistry, crystallography, and morphology. In this study, we use an automated laboratory to investigate the precipitation patterns of copper hydroxide via reaction diffusion (RD) coupling in a solution-gel system. Depending on the initial conditions, the products form continuous precipitates or oscillatory patterns typical of the "Liesegang" phenomenon. The band structures are characterized using empirical spacing metrics, which convert the complex patterns into scalar values and can thus be used for supervised machine learning in an active learning loop. The machine learning algorithm serves dual roles, providing correlation between reaction conditions and resulting precipitation patterns, which are often beyond the physics-based models, as well as dynamically evaluating the most significant areas of the parameter space. Our goal is to develop an autonomous platform wherein the user can pre-select a target product pattern, and the system converges to it with closed loop feedback. We have demonstrated a complete cycle of this process using the Liesegang precipitation of $Cu(OH)_2$ as a test case.

1 Introduction

Over the past few decades, advances in nanomaterials science have enabled the synthesis of a broad library of products, including defect-free crystals, nanoparticles with tunable sizes and structures, surfaces with tailored chemistries, and atomically precise thin films.[1–4] Examples include the crystalline silicon, synthetic gems, highly emissive quantum dots, magnetic storage media, and thin-film-based solar cells. A common feature in these materials is that they are typically grown at near-equilibrium conditions to facilitate the synthesis process. For instance, mixing reactants in a beaker produces a uniform supersaturation condition throughout the reaction medium that can be reproduced experimentally and described using relatively simple models. In contrast, nature uses a radically different paradigm. By leveraging far-from-equilibrium pathways, nature can create hierarchical materials with advanced functionalities from ordinary building blocks.[5,6] For instance, biominerals consist of calcium carbonate or phosphate nanoparticles that organize into complex architectures which result unexpected physical and structural properties.[7,8] Far from being synthesized in mixed beakers, biominerals form in complex chemical environments governed by gradients of pH, ion concentrations, and spatially confined media.

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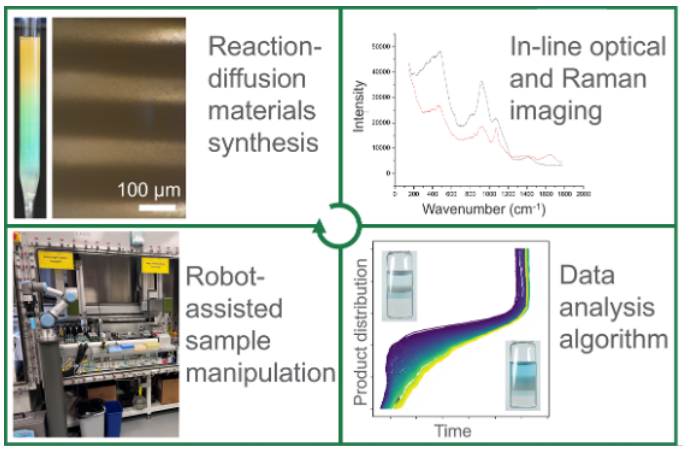

Figure 1: A basic feedback loop describing ML-informed automated synthesis for nanomaterials discovery using reaction-diffusion coupling. ML serves dual roles of characterizing material outcomes as well as informing optimal exploration of parameter space.

An example of a synthetic analogue to biomineralization is periodic precipitation, wherein two reacting components are initially separated and cross-diffuse to create local zones of supersaturation.9,10 In the basic setup, an aqueous solution of the reactants (metal ions) is placed in a cylindrical tube on top of a hydrogel layer loaded with the corresponding counter-reactants (precipitating anions such as hydroxide or carbonate). As cross-diffusion of the reacting species occurs, the precipitating material begins to nucleate in locales where the supersaturation condition has been achieved. Subsequently, nucleation and growth deplete the reactants in the surrounding medium, resulting in the formation of a localized precipitation band. As diffusive supply replenishes the reactants, nucleation events occur at further defined distances from the first precipitation band. The result is a pattern of precipitates known as "Liesegang precipitation rings". More intricate setups based on this principle can leverage reaction-diffusion coupling to create nanoscale patterns of the desired materials.[11]

Using this approach, recent studies have demonstrated the synthesis of inorganic materials,[12] formation of metastable polymorphs, the organization of nanorods into hierarchical structures,[13] and the selective extraction of high-purity minerals from recycled batteries.[14] However, a predictive understanding of the pathways in these systems remains lacking, in part because traditional descriptors such as supersaturation and phase diagrams are not equipped to handle steep chemical gradients under complex spatiotemporal conditions.[15,16] Here the challenge with physics-based modelling is two-fold: Continuum reaction-diffusion models ignore key molecular details that are necessary for capturing the free energy landscapes, while atomistic models have limited timescales and lengthscales, falling short of predicting emergent materials properties at the mesoscale and beyond. Bridging this knowledge gap with data-based modelling promises a new paradigm in materials manufacturing, charting far-from-equilibrium reaction pathways to form products that are typically inaccessible under ambient conditions of conventional synthesis methods.[5,17]

In this context, far-from-equilibrium materials synthesis is conducive for both autonomous research as well as machine learning (ML) driven analytical methods. While synthetic conditions and materials properties are both readily measurable, the rapid breakdown of physics-based models makes traditional iterative lab approaches difficult. By comparison, an automated and unsupervised ML-driven approach offers several benefits (Scheme 1). High throughput as well as high repeatability of automation are ideal for exploring large parameter spaces, particularly for reaction-diffusion systems that are highly non-linear and susceptible to minor perturbations. Moreover, ML serves dual purposes in this application. Using ML feedback in the experimental design loop potentially offers a highly efficient approach as supervised algorithms naturally provide feedback on the most important or fastest changing areas of the design parameter space. Secondly, a method which is agnostic but still capable of capturing the observed phenomenais highly desirable especially when the underlying physics is extremely complex, particularly beyond simple, highly controlled conditions. Applying ML to this problem is robust and more tractable solution than trying to add complexity to known models.

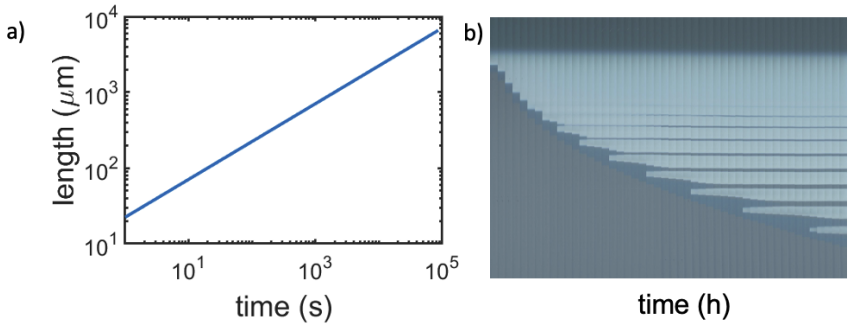

Figure 2: a. Relevant feature length scales expected for a given timescale and a typical diffusion coefficient of 10^{-5} cm^2/s. These relatively slow phenomena provide sufficient latency for real-time computational processing. b. Time-resolved band formation over 48 h.

2 Experimental methods

These experiments were performed using an Unchained brand lab automation platform. Reaction-diffusion experiments were performed in NMR tubes loaded with 600 μL of 0.5% agarose gel prepared by heating to 75 °C with 5-35 mM NaOH for 30 m and setting for 1 h. Various concentrations of salt are prepared and dispensed by the robotic system. The initial parameter space varied this as well as addition of 300 μL $CuCl_2$ varied from 0.1-0.4 M. Up to 96 parallel reaction conditions were explored with experiments allowed to run for 48 h with imaging of each sample taken every 1 h yielding time-resolved band formation and diffusion data.

3 Results and discussion

As a model system we investigate the self-organization of copper hydroxide precipitation patterns via RD coupling. Specifically, a solution of copper chloride is placed on top of an agarose gel that is loaded with sodium hydroxide during preparation. Initially, [NaOH] is at least an order of magnitude larger than [$CuCl_2$], such that the ion flux is primarily from the solution into the gel medium. The system is thus supersaturated with respect to copper hydroxide and a precipitate begins to form near the solution-gel interface. The precipitate locally depletes the reactants, preventing further growth until the ions are replenished by diffusion. This interplay between reaction kinetics and diffusion can result in continuous precipitates, oscillatory patterns, or more complex spatial distribution of the product in the reaction medium. Note that the underlying physics of this phenomenon are complicated by the atomistic details of nucleation and growth across concentration gradients which evolve over time, producing generally low crystallinity products with multiple candidate polymorphs.

The formation of precipitate bands, also known as Liesegang patterns, suggests that RD coupling can be used to synthesize products with a precise spatial distribution in the reaction medium. Specifically, the banded patterns obey multiple empirical laws that determine band spacing, thickness, and rate of growth, which show non-linear dependence on the initial conditions. An additional feature of this system is the synthesis timescale, which is limited by the diffusivity of ions in aqueous media (Figure 2). Since the precipitation patterns develop over an approximately 48-hour window there is sufficient time to operate many samples in parallel while also fully processing and training in real time. Image analysis can thus be deployed in real-time to precisely quantify Liesegang bands which can then be directly processed by ML-informed algorithms (Figure 3).

As an initial optimization run, we were primarily interested in targeting the band spacing metric of the reaction. In the literature, the band spacing this is often empirically reduced to an exponential scaling law, with typical scaling of approximately k = 1.06[9,18] for many systems. Image data were scalarized using a script requiring no human post-processing or intervention to extract this parameter for supervised ML.

$$\frac{c_{n+1}}{c_n} = k \tag{1}$$

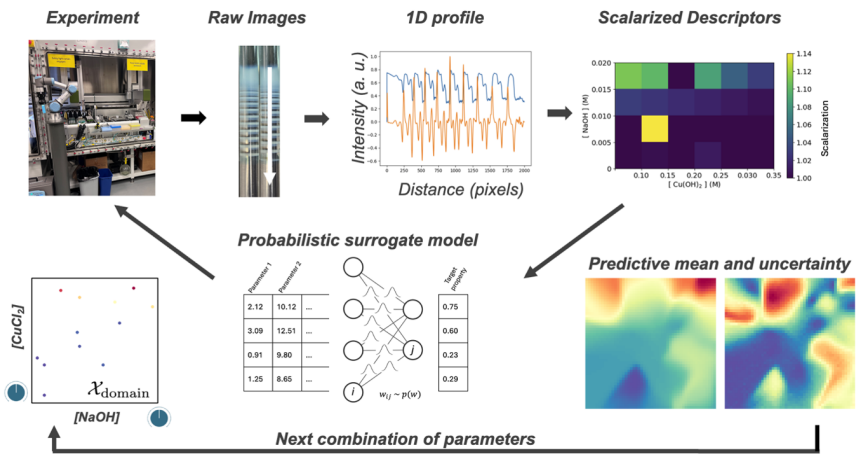

Figure 3: Schematic of the active learning loop used to efficiently search the RD synthesis parameter space specifically to optimize the Liesegang band precipitation and pattern. Experiments are scalarized and processed using active ML to determine the highest value area of the domain space. The loop is then closed when the automated platform performs those experiments.

We established an initial design space investigating the reaction-diffusion precipitation of $Cu(OH)_2$ using 5-35 mM NaOH and 0.1-0.4M $CuCl_2$. The stoichiometric imbalance of precipitating agent as outer electrolyte versus inner electrolyte has been observed in many Liesegang systems,[19] but the exact optimal conditions were not known. To investigate this, the images acquired were automatically processed using a script to determine band positions to an resolution of 9 um/px. the observed data for band formation was fit to equation 1 and the band spacing metric was used as a target for supervised ML. A summary of the extracted data as well as alternate candidate scalarizations are available in the supplementary information. The results of this analysis are shown in figure 4.

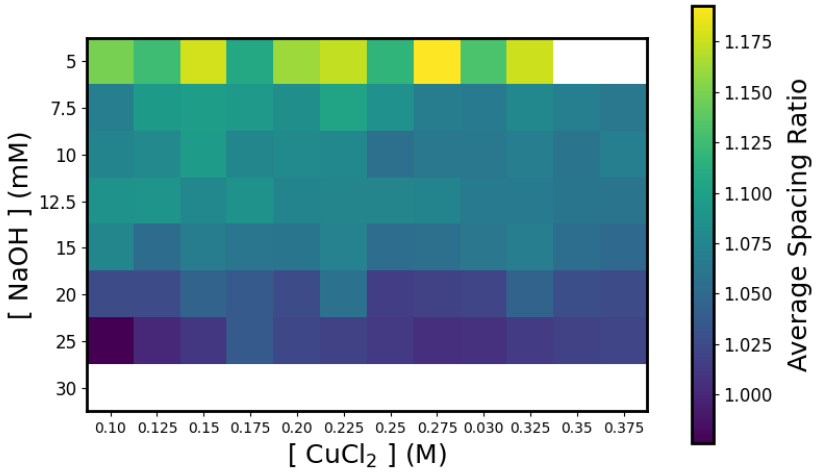

Figure 4: A diagram of the scalarization value k of eq. 1 as a function of the two-parameter input space of $CuCl_2$. and NaOH concentrations. Samples with fewer than two bands are left blank and were considered as sparse points for ML.

Next, we have developed an active learning framework with several probabilistic ML models, based on either Bayesian neural networks (BNNs) or Bayesian Gaussian processes (BGPs), trained to predict the average spacing ratio and other scalarized descriptors from the $CuCl_2$ and NaOH concentrations. Probabilistic models offer many advantages over deterministic models, especially when data is scarce, domain knowledge is limited, and/or experimental noise is unknown. BNNs in particular i) offer all the benefits of neural networks with the ability to provide prediction uncertainties, and ii) can handle data that contain discontinuities and non-stationarities, which are common in physical science problems. Within this loop, we can toggle between explorative and exploitive active learning, where the former is used to efficiently sample the parameter space and improve model performance and the latter is used to target specific band pattern features.

Up to this point, we have operated in the explorative regime, where we have selected the next set of parameters by focusing on regions of the input parameter space in which the ML models are most uncertain. This uncertainty is shown is shown as σ_{pred} in fig 5a. From the uncertainties we choose pairs of [$CuCl_2$] and [NaOH] that maximize the total ML prediction uncertainty of all scalarized descriptors, as shown in Figure 5b. This new set of parameters forms the basis of the next iteration of the design loop, wherein we can continue converge the ML model's predictive ability towards the Liesegang band formation. For example, because the uncertainties were highest in the low concentration NaOH regime the parameters of the next experimental cycle are weighted towards lower concentration NaOH.

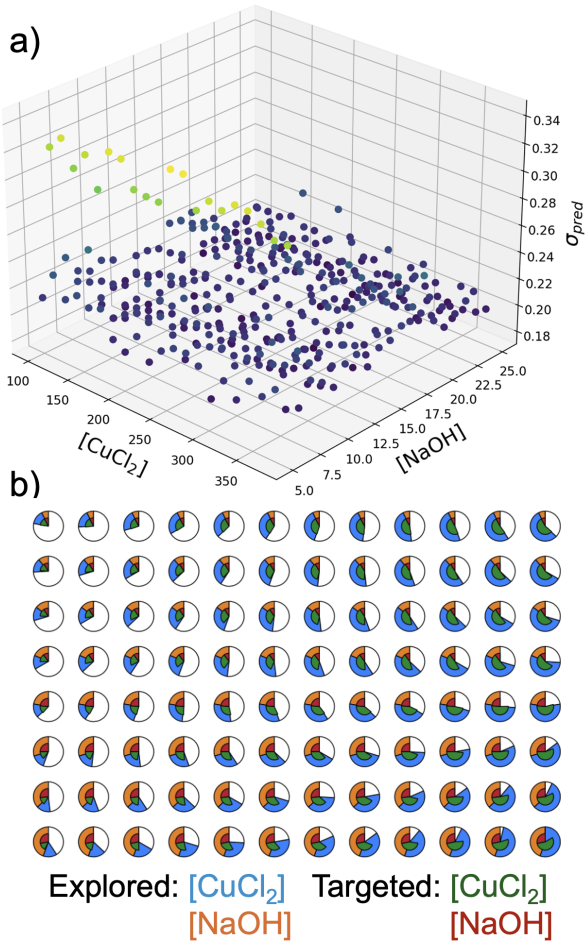

Figure 5: a. Plot of the total ML prediction uncertainty σ_{pred}, summed over all target predictions, over the input parameter space. b. Parameter set selected from regions of high uncertainty in 5a displayed as the ratio of starting reagent concentrations.

4 Conclusion and outlook

We have demonstrated the workflow for combining automated materials synthesis with a machine learning analytical approach, which could eventually enable fully autonomous experimentation. We investigated the complex pattern formation RD precipitation of $Cu(OH)_2$. Using supervised probabilistic machine learning, this workflow dynamically processes the reduced dataset and identifies candidate areas of parameter space for further iterative study, as well as towards the synthesis of the target material with the desired spatial distribution in the reaction medium.

Through the incorporation of these probabilistic ML models in an active learning framework, we are able to i) efficiently sample the RD synthesis parameter space by prioritizing experiments in regions of high ML prediction uncertainty, ii) gain insight into the relationship between these parameters and Liesegang band patterns, and iii) control and tune the band pattern via choice of synthesis parameters.

Future steps will aim to refine this approach to be an increasingly agile tool for materials synthesis and discovery. By adding temporal components to the experiment design and adapting the ML model, we will move towards intra-experimental parameter modification. Dynamic observation of synthetic and material properties would allow for more rapid design as well as expand the landscape of potential far-from-equilibrium pathways across the full parameter space.

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Supplementary Information

SI 1. Extracted band spacing data for 96 grid of paramters. Green dots represent band edge position and blue lines represent fits to eq 2.

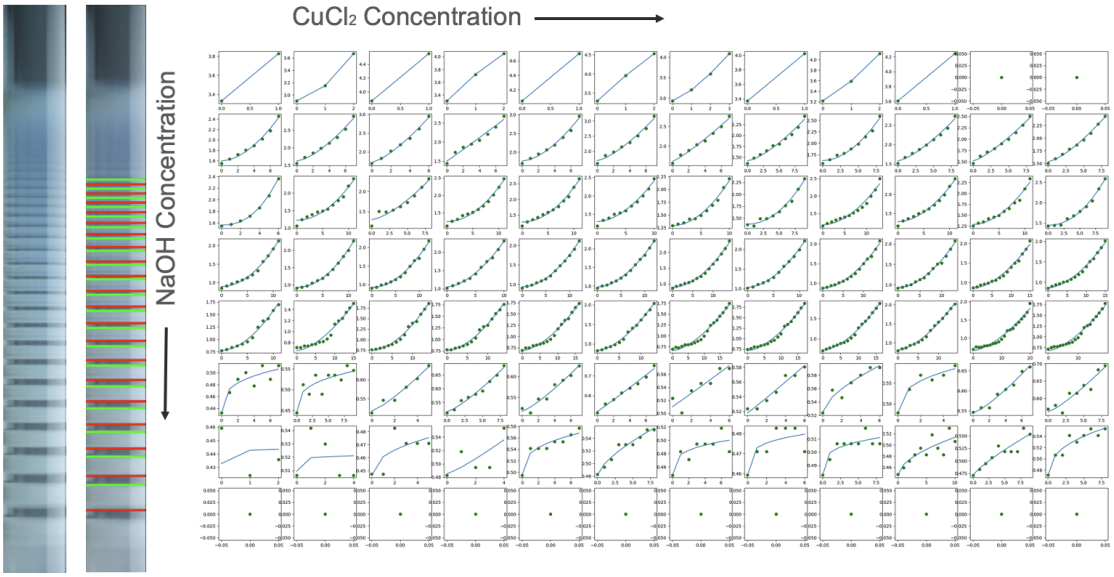

Figure 6: Bands extracted via automated script. Left: example image with band tops and bottoms drawn in as extracted.

SI 2. Alternatively an exponential relationship can be used as a scalarization metric yielding 3 scalar values per sample set:

$$d = Ax^B + C \tag{2}$$

Where d is the band distance and x represents the x'th band.

SI 3. Scalar fit for A of eq 2

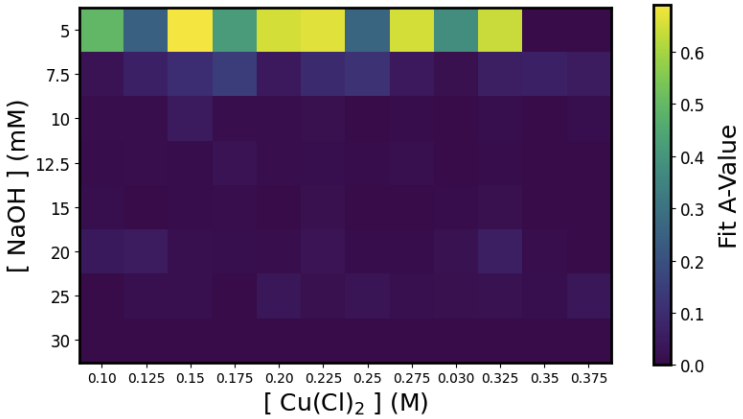

Figure 7:

SI 4. Scalar fit for B of eq 2

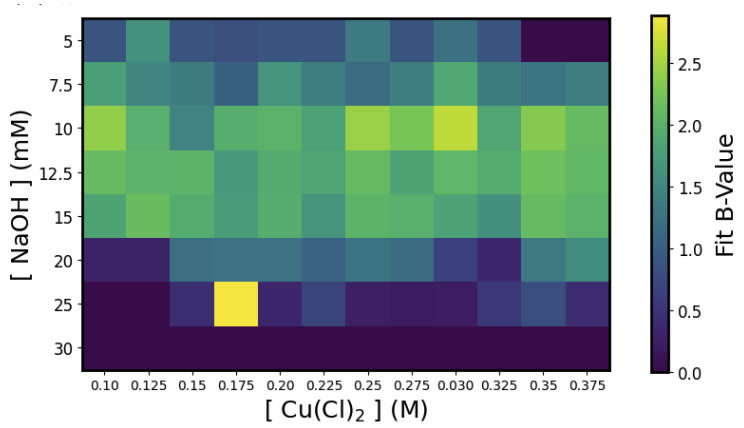

Figure 8:

SI 5. Scalar fit for C of eq 2

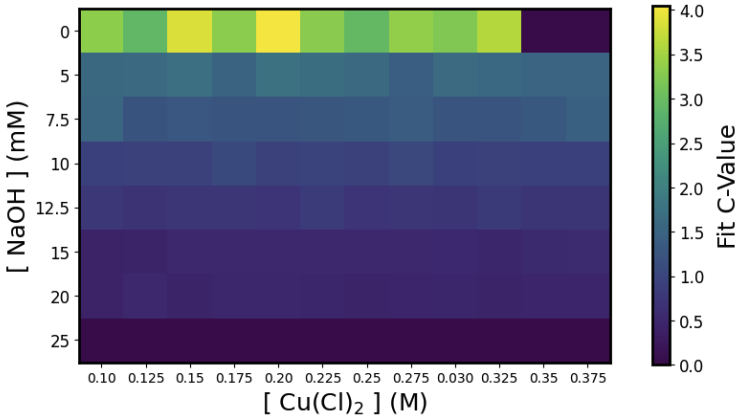

Figure 9:

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