# Bayesian Optimization in Materials Science: A Survey

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#### Abstract

Bayesian optimization is used in many areas of AI for the optimization of black-box processes and has achieved impressive improvements of the state of the art for a lot of applications. It intelligently explores large and complex design spaces while minimizing the number of evaluations of the expensive underlying process to be optimized. Materials science considers the problem of optimizing materials' properties given a large design space that defines how to synthesize or process them, with evaluations requiring expensive experiments or simulations – a very similar setting. While Bayesian optimization is also a popular approach to tackle such problems, there is almost no overlap between the two communities that are investigating the same concepts. We present a survey of Bayesian optimization approaches in materials science to increase cross-fertilization and avoid duplication of work. We highlight common challenges and opportunities for joint research efforts.

#### 1 Introduction

In many areas of AI, decades of research have resulted in many different approaches for solving hard problems. Perhaps the best example is machine learning, where dozens of different approaches just to classification are available, with little guidance on what to use for a particular problem. While random forests are usually a good choice, there are many problems where other approaches provide better performance. Even human experts struggle to choose the best approach for a given problem without significant experimentation though. Worse, once a particular approach is chosen, its hyperparameters need to be set for optimal performance – or is a different approach actually better once its hyperparameters have been optimized?

This issue is not unique to machine learning – the optimization of the hyperparameters of processes is a ubiquitous problem in AI and many other areas,

including materials science. While this can be done efficiently for some applications, for example because gradients are available or other knowledge about the optimized process can be leveraged, there are many scenarios where nothing is known about the process – it is a black box that can be evaluated, but whose inner workings are not sufficiently understood to aid in the optimization process. Such black boxes are much harder to optimize in practice.

Black-box processes are optimized by repeatedly evaluating them for different hyperparameter settings and observing the effect changes in hyperparameter values have – much like scientists have studied natural phenomena for millennia. In many cases, these evaluations are expensive and it is crucial to minimize their number as the optimization would otherwise use too many resources. Bayesian optimization (BO) is a methodology that allows for just that. It is a sampleefficient optimization method that, in general, does not require a large number of samples to obtain good results and is thus particularly suitable for black-box functions that are extremely expensive to evaluate, as found in many areas of AI, engineering, and materials science, where an evaluation may entail synthesizing and testing a new material and require specialized equipment and skilled operators.

At the heart of modern Bayesian optimization approaches are machinelearning-induced surrogate models, that learn to emulate the black-box process to be optimized based on a small number of evaluations. Such surrogate models have a long tradition in materials science where they helped in the development of new materials and designs long before the advent of machine learning. Traditionally, models were based on first-principles understanding and physical laws that were painstakingly developed by scientists rather than the data-driven approaches we see today.

Bayesian optimization, like surrogate models, is not a new concept, nor did it originate in AI – its origins can be traced back decades to engineering applications. It has gained increasing popularity in materials science in the last decade, similar to popularity gains in AI in the same time period. However, research in both fields proceeds almost entirely independently, with no crossfertilization taking place even though the problems solved are the same in many cases. This survey aims to bridge this gap and increase awareness in the AI community on relevant research and approaches being developed in materials science. We provide an overview of relevant recent research and highlight challenges in materials science that AI research can help with and open problems that may benefit from joint efforts.

# 2 Background

In modern Bayesian optimization, a surrogate model is fit based on the results of evaluating the black-box process at different points in the hyperparameter space. Such an initial design could be random, or based on another design of experiments approach. In most cases, this surrogate model is induced by a machine learning algorithm; Gaussian Processes and random forests are popular choices. An acquisition function then determines the next point in hyperparameter space to evaluate the expensive black-box process at, based on the predictions of the cheap-to-evaluate surrogate model and possibly uncertainty quantifications of those predictions. This acquisition function balances exploitation, i.e. evaluating the neighborhood of points known to yield good performance, and exploration, i.e. evaluating points that are far from regions of the hyperparameter space with known performance to avoid getting stuck in local optima. Commonly-used acquisition functions include expected improvement and maximum uncertainty, which always explores.

The expensive black-box function is then evaluated at the point proposed in this way to obtain ground-truth data that is combined with the data from the initial design. The updated data is used to obtain the surrogate model for the next iteration of the optimization process. The incremental augmentation of the data the surrogate model takes into account iteratively improves and refines it for the areas of interest, yielding more accurate approximations of the black-box function and better optimization results. The Bayesian optimization process can be stopped at any time when the desired quality is achieved or resources are exhausted. While this makes it convenient to deploy in practice, there are no guarantees of convergence to a global optimum except if an infinite number of evaluations of the black-box function are allowed. As we make no assumptions about the optimization landscape, the quality of the result cannot be guaranteed or even assessed except by comparison to the results other methods or repeated BO runs achieve.

Figure 1 illustrates surrogate model and acquisition function for one iteration of a toy example. There are many variations of this general approach and hybrid methods that incorporate techniques from other areas of AI have been developed, for example meta-learning to leverage known good hyperparameter configurations from similar applications. A complete exposition is beyond the scope of this survey. For more details on Bayesian optimization, we refer the interested reader to Jones et al. [1998], Mockus and Mockus [1991] and Forrester et al. [2008] for Bayesian optimization in engineering in particular.

### **3** Bayesian Optimization in AI

The optimization of black-box processes is relevant in many areas of AI; in particular for the automated hyperparameter tuning of algorithms. For many applications, software to solve problems have hyperparameters that allow to choose and tune, for example; a search heuristic, a stopping criterion, or a pruning mechanism. These choices provide added flexibility and enable adaptation of a generic algorithm to a particular problem, but setting them to achieve good performance in a specific setting is a difficult task even for human experts.

There are numerous approaches that apply Bayesian optimization methods to automated hyperparameter tuning in AI, for example Hutter et al. [2011], who introduce the SMAC system and demonstrate its power tuning the hyperparameters of SAT and MIP solvers. Snoek et al. [2012] propose the spearmint



Figure 1: Example Bayesian optimization iteration. The horizontal axis shows the parameter space, projected into one dimension. In the top panel, the solid line is the true objective function; the dashed line is its surrogate-model-based approximation. The gray-shaded areas indicate the uncertainty of predictions of the surrogate model. Red circles represent evaluated configurations used to build the initial surrogate model; green squares configurations evaluated and added to the training data for the surrogate model in previous iterations; and the blue triangle the proposed next configuration to evaluate. The bottom panel shows the expected improvement over the best configuration found so far; the highest peak coincides with the proposed next configuration. Illustration generated with the mlrMBO package Bischl et al. [2017].

system and automatically tune the hyperparameters of a machine learning algorithm, whereas Feurer et al. [2015], Kotthoff et al. [2017] automatically choose the machine learning approach and pre- and post-processing methods in addition to tuning its hyperparameters.

Bischl et al. [2017] propose a general and flexible system for hyperparamter tuning using Bayesian optimization, and Falkner et al. [2018] combine Bayesian optimization with bandit-based methods. The need for hyperparameter tuning in machine learning has resulted in the field of automated machine learning (AutoML), which relies to a significant part on Bayesian optimization and related methods. We refer the interested reader to a recent book Hutter et al. [2019] for more.

### 4 Bayesian Optimization in Materials Science

A large application area of Bayesian optimization in materials science is similar to how it is applied in AI – the optimization of the hyperparameters of computationally expensive processes. In particular, Density Functional Theory (DFT) simulations can compute properties of interest for a given material, but may take weeks to complete. DFT calculations are used to determine the electronic structure of systems composed of many atoms or molecules, in particular the spatially dependent density of electrons, from which properties of the modeled material can be derived. Such simulations rely only on first-principles knowledge in quantum physics and are widely applicable to many different types of materials. DFT and similar first-principles-based simulations are the cornerstone in many areas of materials science. Note that the DFT simulations themselves are not optimized, i.e. BO is not applied to improve the simulation itself, but the underlying material – DFT is used to avoid having to synthesize and experimentally evaluate a material.

Kiyohara et al. [2016], Kikuchi et al. [2018], Bondevik et al. [2019] use BO to optimize the grain boundary structure in polycrystalline materials instead of exhaustively evaluating the design space for the materials. They all demonstrate that results of similar quality to exhaustive evaluation can be achieved at significantly lower cost, with an increase in efficiency by up to two orders of magnitude. Similarly, Ueno et al. [2016] optimize the grain boundary energy over several thousand precomputed hyperparameter configurations, which allows them to compare the performance of different approaches. Talapatra et al. [2018] employ BO in a similar setting by evaluating its performance on a set of a few hundred precomputed results. They optimize the elastic properties of a material and demonstrate that BO is quickly able to identify the optimal hyperparameters in their relatively small approximation of the real search space. Balachandran et al. [2016] investigate the same application and in subsequent publications demonstrate the effectiveness of BO to optimize shape memory alloys Xue et al. [2016], the band gap in perovskites, an important material in the creation of solar cells Pilania et al. [2017], and the band gap in compounds for luminescent materials Lookman et al. [2019]. Ling et al. [2017] demonstrate the utility of BO to optimize the DFT-calculated magnetic deformation of a material, superconductors, thermoelectricity, and the strength of steel. Hankins et al. [2019] apply BO to the problem of creating new materials via a parameterized generator for patterns that describe the structure of the material. They optimize the interfacial area of the material, which can be computed directly and very cheaply compared to numerical simulations, but the large hyperparameter space still necessitates a more efficient hyperparameter optimization approach than exhaustive search. Dehghannasiri et al. [2017] design materials with low energy dissipation by optimizing a dopant (an impurity introduced into a pure material of a different type) and its concentration using BO, showing improvements over random search and pure exploitation.

Seko et al. [2014] perform one of the earliest investigations into whether BO is suitable for optimizing the properties of materials that are evaluated using

DFT. In particular, they focus on the performance of the surrogate model that replaces the DFT calculations, a crucial prerequisite for applying BO. They predict and optimize the melting temperature of solid compounds and find that support vector regression in particular provides high-quality predictions of the DFT results. However, they use Gaussian Processes in a BO framework to optimize the melting temperature, demonstrating better performance than random search. A subset of the authors provide additional applications in a later study Tanaka and Seko [2016] that more explicitly distinguishes between high-throughput approaches where an exhaustive grid search is performed and applications where this is infeasible and the use of BO or a similar technique is necessary.

In contrast to most areas of AI, a lot of materials science is not only computational, but involves physical experiments that synthesize and evaluate materials. While DFT and similar simulations are increasingly accurate, their results are often only approximations of what happens in the real world due to different scales or a lack of first-principles understanding – DFT calculations can only consider relatively small numbers of atoms or molecules that may not accurately represent materials deployed in practical applications. Even more so than for computational simulations, BO is useful to reduce experimental effort in the optimization of materials that requires not only physical resources, but also in many cases specialized equipment and the time of a skilled operator.

Ren et al. [2018] use a manual version of BO to discover metallic glasses. They train a random forest model to predict whether a particular composition of precursor materials results in the formation of a glass based on prior experimental results. The model is then used to manually identify the region with the highest promise for gathering additional data through high-throughput experimentation, allowing for many evaluations in a short amount of time. The experimental results obtained in this way are used to refine the model. The authors iterate this process three times and discover several new glass-forming systems. Häse et al. [2018] use BO to optimize the conditions for a chemical reaction to achieve a certain stability condition. While the optimized experimental conditions could be evaluated through actual experiments, the authors use a simulator to achieve higher throughput and obtain more data. They show that BO is not only able to obtain the desired results quickly, but also does so more robustly than other approaches, namely particle swarm optimization and CMA-ES. Kotthoff et al. [2019] optimize the reduction of graphene oxide to graphene in the context of creating nano-circuits and flexible electronics. The evaluations of the black-box function rely on physical experiments that are performed manually and thus the number of data points is only a few dozen. However, the authors demonstrate that even in this case, significant improvements of the experimental outcome over human experts can be achieved. Wigley et al. [2016] optimize the production of Bose-Einstein condensates with BO, which they refer to as machine learning online optimization, with an order of magnitude fewer evaluations than previous approaches. Ren et al. [2020] optimize the efficiency of solar cells by tuning the growth temperature of gallium arsenide cells. Their customized BO approach is able to achieve improvements after only five experimental evaluations. Vellanki et al. [2017] optimize alloy casting and the production of polymer fibers in a constrained batch process, but do not provide a baseline comparison for their results.

Some approaches combine computational simulations with experiments or different types of simulations with different fidelities and costs for multiple layers of screening. This allows to improve the outcomes of the optimization process while reducing the costs for evaluations of the black box functions. Indeed, this is a major difference to applications of BO in AI, where usually there is only a single way of evaluating the black box function to be optimized. The closest to evaluations at multiple levels are techniques like early stopping, used for example in SMAC, that stop the evaluation if the hyperparameter setting under configuration does not show promise on a subset of the data, e.g. a fraction of the folds for a machine learning problem or a subset of the problem instances for a combinatorial optimization algorithm.

Gómez-Bombarelli et al. [2016] optimize the design of OLEDs. They prescreen designs using a machine learning surrogate model, then perform DFT calculations on the remaining candidates, and finally synthesize and experimentally evaluate the candidate designs that pass this second screening. While the authors do not use BO directly, they demonstrate the power of a multi-level, multi-fidelity approach in efficiently exploring a large search space. Pilania et al. [2017] combine different types of DFT simulations, one low-fidelity simulation that can be computed quickly, and one high-fidelity simulation, to optimize the bandgap of solids. They use a modified Gaussian Process that is able to take information at multiple fidelities into account directly as a surrogate model and demonstrate better outcomes than when using only the high-fidelity data. Patra et al. [2020] apply the same approach to optimizing the bandgap of polymers and demonstrate similar results, noting that their multi-fidelity surrogate model is able to generalize better to a larger design space than surrogate models that use only high-fidelity data.

The majority of approaches in both AI and materials science consider singleobjective BO, as multi-objective optimization increases complexity considerably and multiple objectives can be combined into a single objective. However, a few approaches apply multi-objective BO to avoid requiring the user to specify how to weight different objectives and being able to choose the trade-off after the optimization from points on the Pareto front. Talapatra et al. [2018] consider the two competing objectives of shear and bulk modulus, which quantify the effect external forces have on the material. The authors use expected hypervolume improvement as the acquisition function and show that they can efficiently identify the points on the Pareto front. Solomou et al. [2018] consider the simultaneous optimization of up to three properties of a particular type of alloy based on computational simulations. They also use expected hypervolume improvement as the acquisition function and demonstrate the effectiveness of their approach. Ragasa et al. [2019] scale up to 10 objectives, but use a custom non-BO approach, though inspired by Bayesian optimization, that iteratively refines the hyperparameter search space based on found Pareto-optimal configurations.

In many applications, it may be desirable to batch evaluations of the black

box function to be optimized, i.e. have the BO process predict multiple hyperparameter configurations at once. This can facilitate the parallel evaluation of configurations to make optimal use of available resources. This is common to both AI and materials science, though there are few approaches that do it. Häse et al. [2018] propose batches of configurations to evaluate by optimizing the acquisition functions for different values of a parameter that trades off exploration and exploitation. The authors demonstrate that this batch evaluation improves overall performance, as the correct setting for this parameter is unclear and varies over time. A unique issue in BO for materials science that does not usually come up in an AI setting is that the first hyperparameter configuration to evaluate may constrain subsequent configurations. Vellanki et al. [2017] give the examples of heat treatment of alloys, where multiple samples can be processed at the same time in an oven but at a fixed temperature, and the production of polymer fibers, where different values for polymer flow and coagulant speed can be evaluated at the same time but within a fixed geometry. They propose a nested BO approach that optimizes the hyperparameters that are subject to constraints in an outer loop and, given the optimized values, the unconstrained hyperparameters in an inner loop.

In many applications of BO in materials science, standard BO approaches, usually with Gaussian Processes as surrogate models and expected improvement as acquisition function, are used. There are a few exceptions; for example Häse et al. [2018] use Bayesian Neural Networks as a surrogate model with a custom acquisition function and Pilania et al. [2017], Patra et al. [2020] use custom Gaussian Processes that can take information at multiple fidelities into account, with an acquisition function that takes the cost difference of the different fidelities into account. Dehghannasiri et al. [2017] use mean objective cost of uncertainty as the acquisition function, which quantifies how much worse an outcome is because of uncertainty. Ren et al. [2020] replace the traditional BO approach with multiple layers of surrogate models of Bayesian networks infused with background knowledge that constrains the (intermediate) outputs to physically feasible ones, combined with a neural network that serves as a surrogate for expensive numerical simulations. Vellanki et al. [2017] propose a nested approach that runs a series of BO processes to propose a batch of hyperparameter configurations to evaluate. Talapatra et al. [2018] use Bayesian model averaging inside the BO process to select the most suitable surrogate model at the same time as optimizing the black-box process. Ueno et al. [2016] augment BO with automated hyperparameter tuning and a few changes to be able to sample and optimize the surrogate model more efficiently with thousands of features.

Most publications compare their approaches only to a grid or random search, but some study the effect variations of the BO process have. Ling et al. [2017] propose a BO framework that uses random forests as surrogate models with three different acquisition functions and demonstrate that using likelihood of improvement instead of expected improvement performs better on their case studies. Balachandran et al. [2016] on the other hand compare the performance of different surrogate models with the same acquisition function. They note that models that quantify the uncertainty of their predictions show better performance than those that do not, and find that support vector regression with an RBF kernel gives the best surrogate models, similar to the findings of Seko et al. [2014].

In most cases, BO approaches in materials science are developed without awareness of similar efforts in AI. A notable exception is Häse et al. [2018], who compare to SMAC [Hutter et al., 2011] and spearmint [Snoek et al., 2012], as well as non-BO optimization methods. They demonstrate that their proposed system Phoenics achieves better performance, differing from the other BO approaches in a different surrogate model (Bayesian Neural Networks) and an acquisition function based on kernel densities.

# 5 Research Directions

The application of Bayesian optimization in materials science is guided by the domain. In particular, this means that achieving performance improvements is perhaps not as important as in AI, and there is more emphasis on understanding the models and being able to incorporate additional constraints that encode the physical knowledge governing the application, as many of the approaches mentioned above do.

Explainable AI has emerged as an important research area in AI with the increasing deployment of AI models that facilitate decision making. In particular deep neural network models are difficult to understand, with real-world ramifications on their performance. Almost all publications in materials science incorporate some analysis of what surrogate models have learned, often with respect to the importance of the features used to characterize the optimized process (e.g. Kotthoff et al. [2019]). To the best of our knowledge, these approaches are relatively straightforward and more sophisticated techniques developed by the AI community, for example the SHAP framework Lundberg and Lee [2017], have not been applied yet.

Another aspect that is emphasized more in materials science than in AI is the quantification of the uncertainties of a surrogate model, which Ling et al. [2017] specifically develop for random forest surrogate models. This may be the reason that Gaussian Processes remain the most popular surrogate model, although different authors have observed that support vector machine models achieve better predictive accuracy. This is a potential research direction for the AI community that would benefit materials science directly.

Lookman et al. [2016] highlight the successes machine learning in general has had in materials science, but conclude that more applications are required to give the community guidelines on how to apply BO and machine learning in the wider sense in the context of materials science. While, as outlined above, a standard incarnation of BO with Gaussian Processes as surrogate models and expected improvement as acquisition function is emerging as a reasonable place to start, there is likely no single approach that will perform best in all settings – a fact that has been known in AI for decades and leveraged in the emerging field of automated machine learning Hutter et al. [2019] for example. While some approaches in materials science are already taking advantage of these powerful techniques, a more ubiquitous integration of AutoML techniques into materials science tools would likely increase the uptake of BO in this area.

de Pablo et al. [2019] identify a lack of systematic methods for reporting the performance of machine learning approaches and baselines to compare to as one of the challenges in materials science. This is again an area where AI research can help, as the same issues are relevant and have been investigated longer. The authors note that "[e]stablished approaches from the statistics and computer science communities combined with new methods developed specifically for materials data issues must be disseminated to the materials community[...]" – a challenge for our community to reach out more and organize interdisciplinary events. Similarly, they note that most research in machine learning focuses on large datasets which are usually not available in materials science. Especially since the advent of deep learning, such small data problems have been somewhat neglected by the AI community and there are opportunities to develop new approaches at the intersection of materials and AI.

Kalidindi [2019] mentions similar concerns and in particular notes that a significant issue in materials science is the fragmented storage and availability of the relevant data. Similar issues have been tackled by the AI community and resulted in standard data formats and repositories that may be applicable in a materials context as well, for example the OpenML project Vanschoren et al. [2013]. The author further highlights the challenge of integrating the governing physical laws into a BO process and proposes a framework to tackle this. Similar efforts to include background knowledge in machine learning and data mining have been undertaken in AI, for example in the ICON project Bessière et al. [2016], but no joint efforts exist to the best of our knowledge.

While most research proceeds independently in AI and materials science, there are a few joint efforts, notably Vellanki et al. [2017], which propose constrained batch optimization and evaluate their approach both for a machine learning and materials science applications. This demonstrates that both fields can benefit from advances and makes a strong case for increased collaboration between AI and materials science researchers.

# 6 Conclusion

We have presented an overview of applications of Bayesian optimization in materials science, where researchers want to optimize poorly-understood black-box processes to achieve desired properties of materials. Similar to applications of BO in AI, the focus is on minimizing the number of evaluations of the expensive black-box function while maximizing the quality of the result, and similar results have been achieved, firmly establishing Bayesian optimization as a state-of-theart method that should be in the toolkit of every materials science researcher.

A lot of recent progress in materials science has been enabled by Bayesian optimization. However, a lot of work remains to be done to improve BO in

practice and scale to more difficult problems, in particular ones with multiple objectives and large numbers of features, and integrate data-driven methodologies with first-principles knowledge.

Materials science presents some unique challenges to applying Bayesian optimization that can stimulate research in AI and, ultimately, benefit applications of BO in AI as well. Research to date has progressed almost independently in both application domains, and we hope that this survey will stimulate interdisciplinary efforts and joint projects that advance either and both fields.

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