Evaluation of Initial Sampling Strategies for Bayesian Optimization Tasks in Material Science

**Abstract**

Compared to traditional experimental designs, Machine Learning-based optimization strategies, such as Bayesian optimization, have shown promising results in materials development tasks. Before commencing an optimization task, it is crucial to perform initial sampling to collect preliminary experimental data, which then facilitates the construction of a machine learning model and indicates the subsequent optimization direction. Most researchers tend to focus on the model and optimization strategies while overlooking the impact of the initial sampling strategy.

In this study, we propose two initial sampling strategies: Euclidean distance sampling and Knowledge transfer sampling. These two strategies can address different scenarios while initiating an optimization task. If we lack information about the task and need to start from scratch, Euclidean-distance sampling ensures that the initial sampling data is uniformly distributed across the global space. Conversely, suppose we have pre-existing knowledge and databases. In that case, knowledge-transfer sampling can transfer relevant knowledge to assist in the initial sampling for the target task, much like how materials experts select initial experimental settings for new material tasks based on past experiences.

We evaluated our sampling methods on one benchmark function and two material datasets: the palladium-catalyzed aryl halide cross-coupling reaction dataset and the high-temperature alloy creep rupture life dataset. The results demonstrate that Euclidean-distance sampling not only enhances the stability of the initial model but also facilitates subsequent optimization. Knowledge-transfer sampling allows for the leveraging of knowledge from related data, aiding in the discovery of the global optimum.

# Introduction

Machine learning (ML) has recently emerged at the forefront of accelerated materials research. As ML becomes more widely accessible to materials scientists, its integration into experimental design has become increasingly commonplace. In materials experiment design, the need to optimize high-dimensional variables often results in large design spaces, making brute force or traditional methods impractical[1-4].By introducing ML into the loop to guide material optimization tasks, it has been shown to effectively explore complex search spaces and adaptively avoid exhaustive enumeration [5, 6].

Bayesian Optimization (BO), a sequential machine learning approach, has gained widespread application in the exploration of chemical synthesis and materials optimization [7-12]. It is favored for its efficient sampling and optimization capabilities, especially when handling expensive black-box functions.Some recent research studies showed the ML-assisted experiment planning can overperform the conventional design of experiments methods based on the domain knowledge from human experts in many specific case studies, e.g.,optimizing the reaction yield of chemical synthesis [13], discovering new photocatalysts for CO2 reduction [14], and searching for the best fast-charging protocol [15].The BO framework is generally divided into three components: initial sampling strategy, surrogate model, and acquisition function. To enhance the effectiveness and applicability of BO in materials development, current research on classical Bayesian optimization has increasingly focused on the selection of surrogate models and the adjustment of acquisition functions [16, 17]. Liang *et al.* [18] evaluated the performance of BO across five materials science datasets, offering valuable insights into surrogate model selection for materials optimization. Liu *et al.* [19] incorporated previous experimental data as probabilistic constraints, combining subjective human observations with machine learning insights to enable the synergy between domain knowledge and BO. Building on traditional BO principles, Siemenn *et al.* [20] iteratively expanded the sampling search boundary, and customized adaptive acquisition functions to further guide new experiments toward a global optimum.

Yet, few have focused on determining initial sampling methods. Initial sampling provides foundational data for the surrogate model, helping it estimate the surface of the target black-box function, reducing prediction uncertainty, and enhancing the efficiency of the acquisition function [21]. In materials optimization, a well-designed initial sampling can cover the search space and explore the complexity of material features, thereby accelerating the identification of the global optimum, reducing experimental costs, and effectively utilizing limited resources. This makes initial sampling a crucial first step in BO [22].

At present, the predominant initial sampling methods employed in Bayesian Optimization are random sampling and Latin hypercube sampling (LHS) [23-25]. However, both approaches have their limitations: (1) fail to uniformly explore global information: Random sampling, due to its high Randomness, may lead to uneven sample distribution, while LHS operates by independently stratifying each dimension and then combining these strata into sampling points, which means that uniformity is only assured in individual dimensions. (2) unable to leverage relevant knowledge: Conventional initial sampling methods are suitable for scenarios where the target task is entirely isolated and unknown. However, human experts typically do not randomly select initial experiments; instead, they rely on prior experiences gained from previous optimization activities to guide the initial exploration of new tasks [26-30]. Therefore, if relevant data from previous optimization campaigns could be transferred to inform the sampling process of the target task, it would significantly enhance sampling efficiency and optimization results [31, 32].

For the target black-box function to be optimized (Fig. 1a), performing uniform initial sampling across the global space can provide a rough understanding of the function, helping to avoid getting trapped in local optima and ultimately identifying the global optimum (Fig. 1b).

The conventional initial sampling methods mentioned above are applicable when pertinent data are absent for reference. when relevant data exists, human experts generally eschew these methods . Instead, they leverage insights from previous optimization tasks to guide the initial exploration of new challenges.For BO, if relevant information can be obtained from the optimization of a source function, it can help the target function's optimization task directly focus on the global optimum region, thereby reducing the iteration cost(Fig. 1c). Transferring knowledge across different optimization tasks reflects a natural inclination in human behavior. However, simulating this knowledge transfer process in BO remains a significant challenge.



Figure 1. Schematic illustration of uniform sampling and transfer sampling strategies. (A) The target function. (B) BO with uniform initial sampling. Uniform initial sampling collects comprehensive global information about the target function, allowing for the identification of the global optimum after a few iterations. (C) BO with knowledge transfer from a source function. Transfer sampling directly focues on the likely region of the global optimum, achieving rapid convergence through minimal iterations.

In this work, we explore and discuss initial sampling strategies in two BO scenarios.(1) For scenarios lacking relevant data references, we propose a globally uniform initial sampling strategy, Euclidean Distance Sampling (EDS),. Compared to Random Sampling and Latin Hypercube Sampling (LHS), EDS enables uniform initial sampling across the global space, providing a general understanding of the target black-box function, thereby avoiding local optima and facilitating the discovery of the global optimum (Fig. 1b). (2) When relevant data are available, we employ a transfer learning strategy [33, 34] to transfer knowledge from the source function to the optimization task of the target black-box function, helping BO to directly locate the global optimum (Fig. 1c).It is noteworthy that the two methods described above are tailored to initial sampling strategies and function in parallel. In practical optimization scenarios, the choice of an appropriate initial sampling method should be informed by the specific context of data availability.

This paper is organized as follows: Section 2 introduces the proposed Euclidean Distance Sampling (EDS) and transfer sampling strategy; Section 3 illustrates these methods using the Goldstein-Price function [35]; Section 4 conducts simulated optimization and algorithm testing on two real material datasets—the Buchwald-Hartwig cross-coupling reaction dataset and the alloy creep life dataset. Finally, we provide evidence that the EDS strategy significantly enhances the stability of model performance when initializing the surrogate model, while reducing the risk of converging to local extrema during the BO process. The transfer sampling strategy enables the analysis of the correlation between the source model and the target task throughout the iterative process of BO. By dynamically adjusting the degree of knowledge transfer based on this correlation, this approach effectively assists in the optimization of the target task when the source task is relevant.

# Methodology

In this paper, we explore two primary approaches: (1) the Euclidean distance sampling and (2) the transfer sampling strategy. These methods enable the adaptation of Bayesian optimization strategies in different scenarios, achieving more stable and efficient optimization.

We demonstrate the implementation of these methods on the Goldstein-Price function dataset, the Buchwald-Hartwig cross-coupling reaction dataset, and the alloy creep life dataset. For EDS, we compare it with LHS and Random sampling, examining the impact of different sampling methods on initial model construction and subsequent optimization. For transfer sampling, we compare it with standard Bayesian optimization, analyzing the performance of transfer BO using different source data against standard BO with no source data. Through testing on one function dataset and two materials datasets, we demonstrate the stable effectiveness and broad applicability of these two sampling strategies.

## Euclidean distance sampling

To ensure uniformity in sampling across the search space, we propose a sampling strategy based on Euclidean distance, referred to as EDS. This strategy is implemented through the following steps:

1. Initialize sampling: For the parameter space X, randomly select 3 points as the initial sampling set .

2. Calculate minimum Euclidean distance: For each unsampled point in X, calculate the Euclidean distance between the point and each sampled point in . Then, obtain the minimum distance between the unsampled point and the sampled points:

3. Select the next sampling point: For all unsampled points in X, choose the one with the maximum minimum Euclidean distance as the next sampling point:

4. Update the sample set: Add to , and repeat steps 2 and 3 until the required number of sampling points is obtained.

## Knowledge transfer sampling

In situations where some related datasets are used as source data, we can leverage the knowledge from the source data to assist in sampling for the target optimization task. The difficulty of this process lies in the following aspects: a) How to determine the relevance between the source data and the target task? b) How to utilize the knowledge from the source data to assist in sampling for the target task? c) How to mitigate the impact of the source data during the sampling process as the search evolves?

In this research, we combined the Rank-weight Gaussian process ensemble (RGPE) and the Transfer Acquisition Function (TAF) method. This approach ensures relevance between the selected source data and the target task as the search progresses, automatically updates the weights of the source data, and achieves reliable transfer Bayesian optimization. The process is implemented through the following steps:

(1) Computing Ensemble Weights: If a model can predict the ranking of observation values according to their predicted values, it is useful for optimization. We construct a ranking loss to measure the consistency of the model's predicted ranking with the target ranking. Here,|| = > 1 denotes the dataset used to add weights to the target values in the new round of model fff. The model's ranking prediction loss is measured by the following ranking error loss:

where is an indicator function, and is the exclusive-or operation. The ranking-based loss assures that only the location of the optimum is important, as opposed to the actual values of the predictions. The loss for the target model is computed using leave-one-out cross validation, and is given by

where −k denotes a model fit to all target task observations except for the kth point. Now, the weights are assigned to each model according to the probability that it is the ensemble member with the lowest ranking loss. This probability is estimated by boostrapping S samples from the model predictions on the validation set, i.e. The weight is calculated by:

(2) Transfer acquisition function: The source model and target model both use Gaussian process models. By assigning the calculated weights to the acquisition functions of their respective models, a transfer acquisition function is obtained.

where

By calculating the transfer weight, we can guide the ranking optimization tasks to take further steps, thus realizing the transfer of tasks from source to target.

# Benchmarks

Here, we demonstrate the effectiveness of our method using the Goldstein-Price function (2-dimensional), a well-known benchmark function characterized by multiple local minima and one global minimum. To better mimic the conditions encountered in material data optimization, we generated a dataset of 2,500 function values by sampling each dimension at 50 intervals. The input variables (x) were normalized using min-max scaling, and the output values (y) were logarithmically transformed to prepare the data for subsequent experiments and evaluations.

## Euclidean distance sampling

To analyze the modeling and optimization performance of random, LHS, and EDS, we first extracted 20 samples from the dataset using different methods to build Gaussian Process Regression (GPR) models,and visualized both the distribution of the sample points and the models' predictions of the target function.As depicted in Figure 2A, the "Truth" plot in the left corner represents the actual distribution of the objective function, while the other three plots show the predictions of Gaussian Process models constructed by sampling 20 points using random sampling, Latin hypercube sampling and Euclidean distance sampling respectively. The model prediction based on EDS more accurately capture the general relationship of the target function, while the predictions based on RDM and LHS show larger deviations, particularly in regions where the target function exhibits high variability.

Following this, we incrementally increase the number of samples and compare the changes in the Spearman coefficient across different sampling methods. The Spearman coefficient, which ranges from -1 to 1, measures the strength and direction of monotonic relationships between variables. Figure 2B shows how the Spearman correlation coefficient of the models varies with the number of samples across different sampling strategies. The Spearman coefficient for EDS increases rapidly with the number of samples and surpasses that of RDM and LHS after 20 sample points, indicating that the SE strategy can build an accurate model more quickly. Figure 2C shows the distribution of Spearman coefficients for each sampling strategy after 20 samples and 40 repeated experiments. The distribution for SE is concentrated in a higher range, further validating its stable modeling capability.

Finally, we use 20 initial sampling points to establish the initial model, and based on this, we utilize the Expected Improvement (EI) acquisition function to perform BO. This approach allows us to investigate how different sampling methods influence the subsequent performance of BO. As the number of samples increases, the objective function values progressively approach the global optimum(Figure 2D). Notably, BO based on EDS shows significant optimization effects after just 10 EI sampling iterations and essentially finds the global optimum after around 55 iterations. In contrast, BO based on RDM and LHS requires more iterations to achieve similar results. Statistical analysis of BO optimization results from 40 independent experiments (Figure 2E) reveals that BO using random sampling or LHS sometimes gets stuck around function values of 3.6 and 4.4. In contrast, most of the optimal values obtained using EDS are concentrated below 1.5, indicating that EDS effectively reduces the risk of getting trapped in local optima.



Figure 2: Evaluation of sampling strategies on the Goldstein-Price function. (A) Distribution of sampled points using various strategies. Black crosses indicate sampled data points, while the contour plot presents the GP model predictions in two-dimensional space based on these samples. (B) Spearman correlation analysis of the GP model. This panel shows how data is continuously added from the three sampling strategies to the GP model, enabling observation of changes in the Spearman coefficient on the test set. (C) Histogram of Spearman coefficients when the sample size is 20. The histogram illustrates that the EDS-based model exhibits a more concentrated distribution of coefficients compared to the others. (D) BO optimization trajectories under different initial sampling methods. After acquiring 20 initial data points, the optimization process shifts to expected improvement (EI) for further sampling. (E) Distribution of optimal function values at the conclusion of BO optimization. The EDS-based BO method identifies current optimal values, all below 1.5. All experiments were conducted independently 40 times, with error bars in panels B and D representing the variance of the repeated results.

## Knowledge transfer sampling

To evaluate the effectiveness of transfer sampling methods in optimization, we designed various types of source functions based on the objective function and constructed corresponding source models to provide accessible prior knowledge.The contour plot of the source function is shown in Figure. 3A. Source function 1 is obtained by shifting both x1​ and x2​ of the target test function by 0.1 units and scaling the y-axis to 0.8. Source function 2 is the negative of the y-values of source function 1. Source function 3 is a funnel-shaped function we designed for knowledge transfer from other types of source functions.The blue region of the target function represents the global optimum area that we aim to explore through BO.

We then compared the optimization curves of standard BO without knowledge transfer and transfer BO with different prior knowledge(Figure. 3A). To introduce information from the source function early in BO, we first randomly sampled 3 points and then initiated their respective BO tasks. In each iteration, we add one new sample point.After 30 iterations, both transfer BO 1 and transfer BO 3 successfully identified the global optimum, indicating that transfer sampling effectively leveraged the knowledge from source functions 1 and 3 related to the global optimum, thereby aiding in optimizing the objective function. In contrast, the performance of transfer BO 2 was comparable to that of standard BO, suggesting that transfer sampling could disregard the misleading information from source function 2 and revert to standard BO behavior.

Additionally, we tracked the changes in the weights of different source models throughout the iterative process.Figure. 3C shows the weight variations of the three source models during the transfer BO process. In the early stages of optimization, the weight of source model 1 was greater than that of 3, while the weight of source model 2 remained close to zero, consistent with the conclusions drawn from panel b. As the iterations progressed, the weights of all source models gradually decreased, indicating that as more target data was acquired, the target model became increasingly refined, and the optimization of the target function progressively relied less on the guidance of the source models.



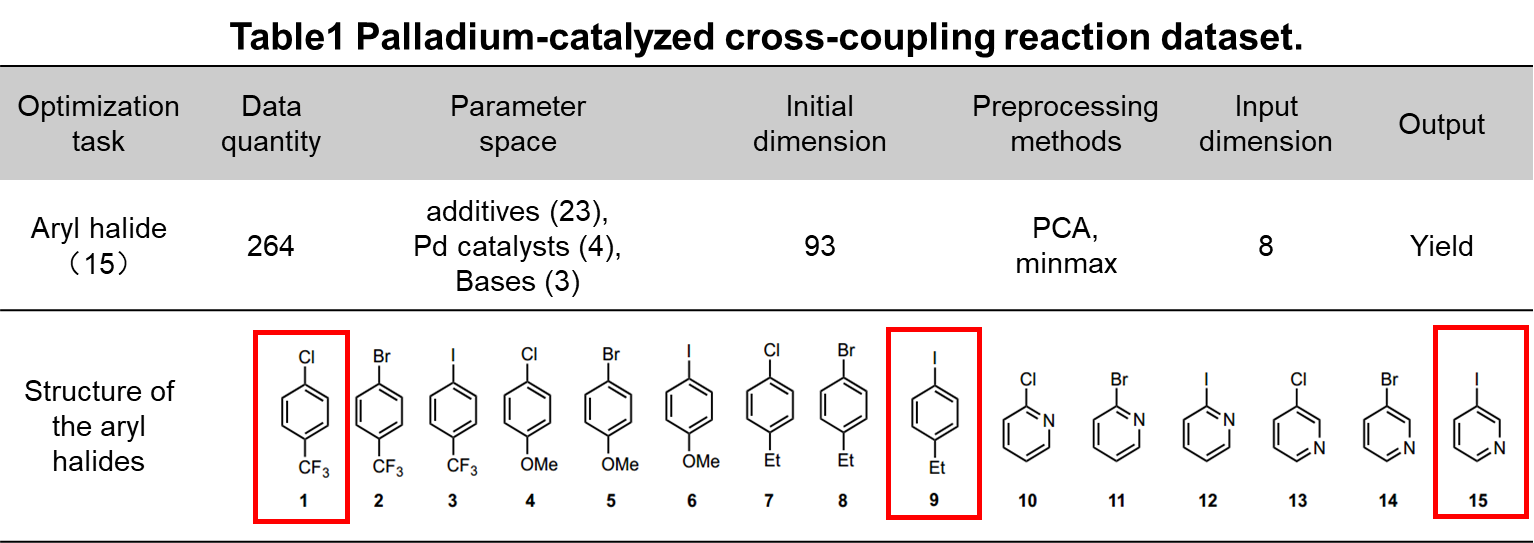
Figure 3: Comparison of standard Bayesian Optimization and transfer Bayesian Optimization with different source models. (A) Contour plots of the target function and three source functions used for transfer learning. The source functions are generated by different methods: Source 1 is produced by shifting and scaling the target function, Source 2 is an inversion of Source 1, and Source 3 is from a completely unrelated function.(B) Optimization trajectories of standard BO versus three transfer BO variants. The left panel compares the optimization paths, with the dashed black line indicating the global minimum of the target function.(C) Evolution of source model weights during the transfer BO process. This panel shows how the weights of the source models change, with higher weights indicating a greater contribution to the optimization.

# Experiments

We selected two real-world materials datasets to evaluate the performance of different sampling algorithms: the palladium-catalyzed aryl halide cross-coupling reaction dataset and the high-temperature alloy creep rupture life dataset. Both datasets are sourced from publicly available materials databases.

## Optimization of Simulated Cross-Coupling Reactions

The Pd-catalyzed Buchwald-Hartwig reaction holds significant value in pharmaceutical synthesis [36-40]. Ahneman et al. [41] conducted high-throughput experiments to obtain yield values for reaction combinations involving 15 different aryl halides. For each halide, the reaction space encompassed 23 additives, 4 palladium catalysts, and 3 bases, resulting in a total of 264 combinations (23 × 4 × 3 = 264). The original study characterized each option using 93-dimensional atomic, molecular, and vibrational descriptors. In our work, we applied min-max normalization and principal component analysis (PCA) for dimensionality reduction based on these original features.



We treated each subset of data as an optimization task, allowing the transfer of knowledge from other aryl halides when searching for the optimal reaction conditions for a specific aryl halide. We randomly selected the optimization tasks for aryl halides 1, 9, and 15 as the target tasks. For the exploration of initial sampling, proxy models were built using initial datasets of the same size obtained through different sampling methods. These models were then used to identify the experimental combination with the highest yield via Expected Improvement (EI). In terms of transfer sampling, data from three randomly selected aryl halides were transferred for knowledge sharing in each target task.



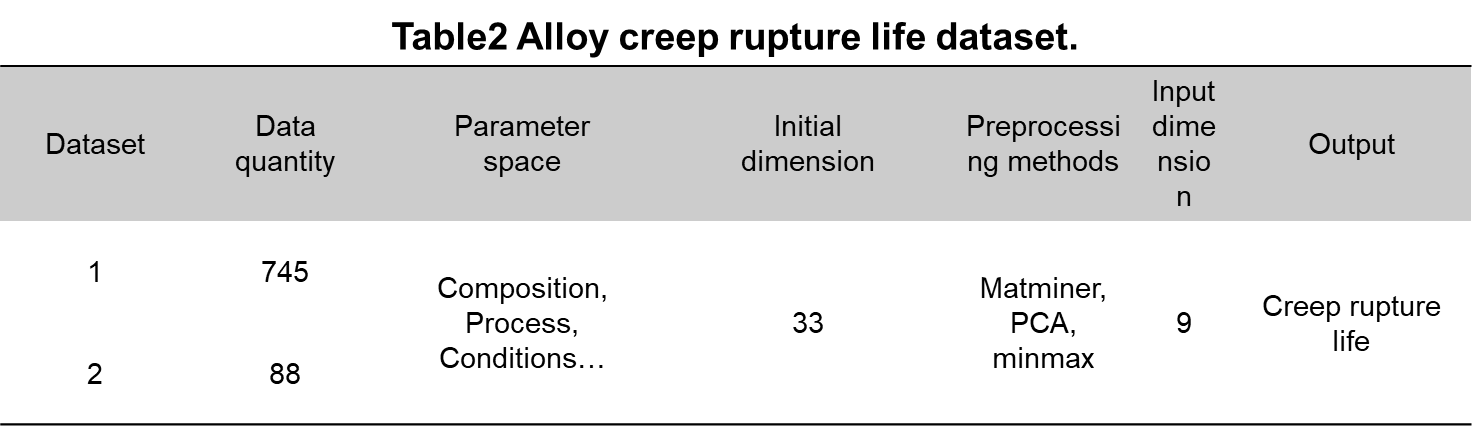
**Figure 4: Comparative analysis of sampling algorithms for different aryl halide optimization tasks.** (A-C) Comparison of initial sampling strategies and comparison of standard BO and transfer BO methods based on aryl halide 1, 9 and 15.(Left) Spearman correlation curves for models on a reserved test set under different initial sampling strategies across three aryl halides. The curves depict the variation in correlation as the number of samples increases for Random, LHS, and EDS. Models trained on datasets generated by EDS exhibit higher performance metrics compared to other methods. (Middle) Yield optimization results from BO using different initial sampling methods. The dashed black line represents the target yield, and the curves illustrate the yield progression as the number of samples increases. BO optimized with EDS-based initial sampling shows reduced variance in the optimization curves, even with comparable mean values across methods. (Right) Optimization trajectories comparing transfer BO using various source datasets with standard BO. The source datasets are generated by randomly selecting different aryl halide numbers and using their corresponding data. Some transfer BO methods show similar optimization performance to standard BO, while others demonstrate significantly faster convergence to optimal yield combinations.

First, we evaluated different sampling strategies on the aryl halide 1 dataset (Figure 4A). The 264 data points were split into a training set and a test set in an 8:2 ratio. Within the training set, data were sampled using Random, LHS, and EDS strategies to build Gaussian Process regression models, and the predictive performance on the test set was evaluated using the Spearman correlation coefficient. Error bars indicate the variance of the metric across 40 independent experiments. When the number of samples exceeded 20, the model based on EDS outperformed those based on Random and LHS. Next, we fixed the initial sample size at 20 to further explore the impact of initial sampling on BO, as shown in the middle panel of Figure 4A. During the first 40 EI-based sampling iterations, the performance differences among the three strategies were minimal. However, when the number of samples exceeded 60, the error bars for BO under the EDS strategy significantly decreased, which we attribute to the early global uniform sampling providing a rough understanding of the optimization task, thereby improving optimization stability. Lastly, the right panel of Figure 4A compares the optimization performance of transfer BO and standard BO using reaction data from halides 2, 4, and 7 as known datasets.. The results showed no significant differences between the two strategies, likely due to low correlation between the source and target tasks, resulting in ineffective transfer of optimization information.

In the tests with aryl halides 9 (Figure 4B) and 15 (Figure 4C), we conducted the same sampling strategy evaluations as described above. The results showed that, in terms of model performance, the EDS method still outperformed Random and LHS, particularly in BO optimization, where the variance of EDS was smaller, further supporting the hypothesis that global uniform sampling improves BO optimization stability. For transfer sampling that introduces relevant information at the beginning of the target task, the effectiveness of transfer varies depending on the source data. The experiments demonstrated that data from halides 8 and 14 helped optimize the reaction combinations of halide 9, while for halide 15, transfer sampling from halide 8 even identified the optimal yield combination within 20 iterations. This indicates that when relevant data is available, transfer sampling strategies have significant potential for practical applications.

## Optimization of Simulated High-Temperature Alloy Materials

The second experimental dataset is the alloy creep life dataset [42], which is sourced from both Refs. and the NIMS public database. The former contains 745 samples, while the latter includes 88 samples, with each sample characterized by 33 features encompassing chemical composition (22 elements), heat treatment processes (9 parameters), and experimental conditions (2 parameters). The target feature is the creep rupture life. Given that creep rupture life values are relatively small, we used the logarithm of this attribute, i.e. (creep rupture life), as the output variable. Our goal is to maximize the creep rupture life. In this study, we used Matminer descriptors to describe the chemical composition based on the aforementioned features. We then combined these composition descriptors with the processing variables, followed by min-max normalization and principal component analysis (PCA) for dimensionality reduction.



Similarly, for the exploration of initial sampling, we treated the two datasets as separate BO tasks to search for the parameter combinations that result in the longest creep rupture life. Proxy models were built using initial datasets of the same size obtained through different sampling methods, and Expected Improvement (EI) was then used to identify the experimental combination with the maximum creep rupture life. For the investigation of transfer sampling, knowledge transfer was performed using the other dataset (outside the target task) as the source data.



**Figure 5. Comparative analysis of sampling algorithms for two alloy dataset optimization tasks.** (A) Comparison of initial sampling strategies and comparison of standard BO and transfer BO methods based on Alloy Dataset 1. (B) Comparison of initial sampling strategies and comparison of standard BO and transfer BO methods based on Alloy Dataset 2.

Figure 5(A) shows the test results on Alloy Dataset 1. This dataset contains 745 data points, which were split into a training set and a test set with an 8:2 ratio. We used only the few dozen data points obtained from initial sampling within the training set for model prediction. Although the Spearman coefficient was not very high at this stage, EDS still showed better modeling performance compared to other methods. Finding the optimal experimental combination among so many data points is challenging. While the mean of BO optimization curves under all three initial sampling methods was comparable, EDS exhibited smaller variance after 150 samples, indicating stronger stability during the optimization process. Regarding the performance of transfer sampling, Dataset 2 contained only 88 data points, so the knowledge available for transfer was limited. As a result, the optimization performance of transfer BO and standard BO was similar, with both outperforming random sampling, further demonstrating BO's effectiveness in optimization problems. Figure 5(B) presents the test results on Alloy Dataset 2. Even in a small sample dataset, EDS demonstrated excellent modeling performance. Particularly after the initial BO sampling, EDS showed outstanding optimization results. This is due to EDS's tendency to sample at the boundaries of each dimension, which allows it to quickly explore extreme values when the target variable changes linearly along a certain dimension. Finally, supported by the larger Dataset 1, transfer BO surpassed standard BO and random sampling in optimization performance after just 10 iterations, showcasing the strong capability of the transfer algorithm in transferring knowledge from large to small datasets.

# Summary ＆ Conclusions

In summary, we proposed corresponding strategies for different scenarios before implementing Bayesian Optimization (BO): EDS (Euclidean distance sampling) for initial sampling without source data and transfer sampling when source data is available. Compared to the commonly used initial sampling strategies, Random and LHS, EDS sampling achieves globally uniform sampling. For scenarios with source data, transfer sampling effectively leverages source data knowledge to aid in optimizing the target task.

Through simulation optimization on test functions, the cross-coupling dataset, and the alloy creep life dataset, our experiments demonstrated that EDS sampling yields more stable model performance than Random and LHS with the same number of samples, positively impacting subsequent EI optimization. Transfer BO using transfer sampling significantly increases the probability of finding the global optimum compared to standard BO with the same number of samples.

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