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AN EFFICIENT SADDLE POINT SEARCH METHOD USING KRIGING METAMODELS

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ABSTRACT

Simulating phase transformation of materials at the atomistic scale requires the knowledge of saddle points on the potential energy surface (PES). In the existing first-principles saddle point search methods, the requirement of a large number of expensive evaluations of potential energy, e.g. using density functional theory (DFT), limits the application of such algorithms to large systems. Thus, it is meaningful to minimize the number of functional evaluations as DFT simulations during the search process. Furthermore, model-form uncertainty and numerical errors are inherent in DFT and search algorithms. Robustness of the search results should be considered. In this paper, a new search algorithm based on Kriging is presented to search local minima and saddle points on a PES efficiently and robustly. Different from existing searching methods, the algorithm keeps a memory of searching history by constructing surrogate models and uses the search results on the surrogate models to provide the guidance of future search on the PES. The surrogate model is also updated with more DFT simulation results. The algorithm is demonstrated by the examples of Rastrigin and Schwefel functions with a multitude of minima and saddle points.

1. INTRODUCTION

Creating new materials and new processes by design based on a problem-solving paradigm will be a major mission for product and manufacturing engineers in the future. Computational tools are one of the key components to realize such paradigm. Understanding material properties as well as the detailed physical and chemical processes during synthesis and fabrication at the atomistic scales is essential for rational design of materials and manufacturing. For instance, designing functional materials that are used to store information or energy requires a good understanding the phase transition processes. Enhancing the strength of super alloys needs the knowledge of how defects are initiated and propagated in polycrystalline structures. Improving the scalability of nanomanufacturing requires the fundamental knowledge of physical processes or chemical reactions during the synthesis and fabrication. Atomistic simulation can be very helpful for the predictions of the above phase transition or transformation processes.

The general process of simulating a phase transition process is as follows. First, a potential energy surface (PES), which represents the potential of a group of atoms as a function of internal coordinates, is generated. It characterizes the energy levels for various possible configurations of the material system. PES can be regarded as a hyper surface in a high-dimensional configuration space, where the minima correspond to the stable or metastable states. Second, a minimum energy path (MEP) [1, 2], which is the lowest energy path for re-organization of the atoms from one stable configuration to another, is located. The maximum potential energy along the MEP is the saddle point which determines the activation energy. The activation energy is the 'barrier' that the material system needs to overcome to complete the transition. The MEP can be interpreted as the steepest descent path on the PES from saddle point(s) connecting the initial and final stable configurations, also respectively known as the reactant and the product, with local minimum energy level on the PES. Third, the activation energy is obtained by finding the saddle point along the MEP and the transition rate is calculated using the transition state theory [3]. Finally, rare event simulation mechanisms such as kinetic Monte Caro (kMC) method [4] are applied to simulate the phase transition process. Transition rate is one of the most important inputs for kMC simulation. The challenge of accurately predicting a phase transition is the knowledge of the true value of transition rate, which is determined by the energy barrier that exists between the initial and final stable configurations. That is, some activation energy

is required to enable the transition from the initial structure to the final one. Mathematically it is the saddle point on the PES with the global minimum energy barrier that determines the transition rate.

A number of methods [5-8] that search for saddle points on a PES have been developed. However, they require a large number of costly functional evaluations, which is usually conducted by first-principles such as using the density functional theory (DFT). The computational cost limits the application of those methods to large systems. In this paper, we propose a surrogate modeling approach to reduce the number of DFT simulation runs. The surrogate model or metamodel allows us to identify the potential locations of minima and saddle points. This approach will significantly reduce the simulation time as the functional evaluations of the surrogate model requires much less computational time than the one using DFT calculation.

A number of methods [9, 10] have been developed to generate surrogate models with high accuracy based on limited sample points. The most popular ones are polynomial regression [11], support vector regression [12], moving least squares regression [13, 14], radial basis functions interpolation [15], neutral networks [16], Kriging method [17], etc. Among these methods, Kriging, originally developed in the field of geostatistics, gained extensive attentions in recent years. Kriging constructs interpolation models using the concept of stochastic Gaussian processes and predicts the values of interpolated functions. In general, Kriging models include two parts: regression model or mean structure which approximates the interpolation model globally and a Gaussian process model with zero means which represents the local deviation. Kriging models have sufficient flexibility to represent different kinds of functions. In addition, the introduction of stochastic processes enables us to model uncertainty and bias. These two properties make the Kriging method a good candidate for us to construct surrogate models for saddle point search subject to model-form uncertainty in DFT simulation. Our goal is to develop an efficient and robust approach to search saddle points on PES's.

In this paper, a new search algorithm based on Kriging is developed to search local minima and saddle points on a PES. The algorithm uses the information collected in the previous search to construct a surrogate model, which is then used to provide the guidance of future search. After a predefined number of DFT simulations, a surrogate model of the PES is constructed using Kriging interpolation. Then the search process is conducted on the surrogate model until some defined criteria are satisfied. The search result based on the surrogate model will be used to predict the new locations for functional evaluations of the DFT. The search process on the PES resumes. After collecting more data points with DFT simulations, the surrogate model is refined and updated by including those new data points. The search process switches to the surrogate model again. This 'real-surrogate-real' model process continues till some stopping criteria are satisfied. This approach can significantly save computational time by reducing the required DFT simulation runs.

In the remainder of the paper, Section 2 gives a brief background of the existing transition path and saddle point search methods. Section 3 gives an overview of kriging. Section 4 introduces the concurrent search algorithm developed earlier, which the new algorithm is developed based upon. Section 5 demonstrates the algorithm using the Rastrigin and Schwefel functions which are benchmarks for global optimization, and Section 6 summarizes the paper.

2. BACKGROUND

2.1 Saddle Point Search Methods

Here, only a brief summary of existing saddle point and transition path search methods is given. Refs. [5-8] provide a detailed review. The existing saddle point search methods can generally be categorized into two groups: single-ended methods and double-ended methods. Single-ended methods start with one configuration to search the saddle point on a PES without locating the corresponding MEP. Double-ended methods are to locate the saddle point and corresponding MEP between two starting states. The major group of single-ended methods is eigenvector-following methods [18] that follow the eigenvector of Hessian matrix with local quadratic approximations of the PES.

The most popular double-ended methods are chain-of-states methods including nudged elastic methods (NEB) [19], string methods [20], and other methods [21]. Chain-of-states methods rely on a collection of images that represent intermediate states of the atomic structure as it transforms from initial to final configurations along the transition path. These discrete states are chained to each other after the search converges, and the transition path and saddle point are obtained.

2.2 Kriging Methods

Kriging was originally developed in geostatistics by a South African mining engineer, Krige [22]. This method is further developed by Matheron [23, 24]. In 1989, Sacks et al. [17] first brought the application of Kriging method to the design and analysis of computer experiments. Currin et al. [25], and Welch et al. [26] further expand the application of Kriging in computer simulation. Morris et al. [27] extend the method to include the first derivative information in model construction. Since then, the Kriging method becomes popular in constructing surrogate models for deterministic but computationally expensive simulations and design optimization [28-31].

Kriging predicts functional values from a limited number of existing ones by modeling the unknown function as a Gaussian process. The response function $Y(\mathbf{x})$ with n-dimensional input $\mathbf{x}$ is composed by a polynomial model and a Gaussian process model with zero mean as [17]

$$y(\mathbf{x}) = \mathbf{f}(\mathbf{x})^T \boldsymbol{\beta} + \varepsilon(\mathbf{x}) \quad (1)$$

in which $\mathbf{f}(\mathbf{x}) = [f_1(\mathbf{x}), \ldots, f_p(\mathbf{x})]^T$ is a vector of p basis polynomial functions, $\boldsymbol{\beta} = [\beta_1, \ldots, \beta_p]^T$ are the corresponding

unknown regression coefficients, and $\varepsilon(\mathbf{x}) \sim \mathrm{GP}(0, \sigma^2\mathbf{R})$ represents the Gaussian process with zero mean and covariance

$$\mathrm{cov}(\varepsilon(\mathbf{x}_i), \varepsilon(\mathbf{x}_j)) = \sigma^2 \mathbf{R}(\theta_\varepsilon) \quad (2)$$

in which σ^2 is the process variance and $\mathbf{R}(\theta_\varepsilon)$ is the $m \times m$ correlation matrix for m data points with θ_ε as the process correlation parameter. For instance, for Gaussian correlation function, the element ij is defined as

$$R(\theta_\varepsilon, \mathbf{x}_i, \mathbf{x}_j) = \exp(-\sum_{k=1}^{n} \theta_\varepsilon^{(k)} (x_i^{(k)} - x_j^{(k)})^2) \quad (3)$$

for n dimensional space with parameter $\theta_\varepsilon = (\theta_\varepsilon^{(1)}, \ldots, \theta_\varepsilon^{(n)})$. For an unknown position $\mathbf{x}$, the vector of correlation is $\mathbf{r}(\mathbf{x}) = [R(\theta_\varepsilon, \mathbf{x}, \mathbf{x}_1), \ldots, R(\theta_\varepsilon, \mathbf{x}, \mathbf{x}_m)]^T$.

Given m data points with inputs $\mathbf{x}^* = (\mathbf{x}_1, \ldots, \mathbf{x}_m)$, the corresponding output vector $\mathbf{Y} = [y_1, \ldots, y_m]^T$, and an $m \times p$ matrix of polynomial values $\mathbf{F} = [\mathbf{f}_1(\mathbf{x}^*), \ldots, \mathbf{f}_m(\mathbf{x}^*)]^T$, θ_ε is determined by the maximum likelihood estimation (MLE), which is to maximize the likelihood function L defined as

$$\ln L = -\frac{n \ln \hat{\sigma}^2 + \ln |\mathbf{R}|}{2} \quad (4)$$

in which $\hat{\sigma}^2 = (\mathbf{Y} - \mathbf{F}\boldsymbol{\beta})^T \mathbf{R}^{-1} (\mathbf{Y} - \mathbf{F}\boldsymbol{\beta})$ is the estimator of the process variance and $\boldsymbol{\beta} = (\mathbf{F}^T \mathbf{R}^{-1} \mathbf{F})^{-1} \mathbf{F}^T \mathbf{R}^{-1} \mathbf{Y}$ is the generalized least square estimator of coefficients. Parameter θ is determined by solving the MLE problem. After θ is determined, the regression coefficient matrix $\boldsymbol{\beta}$ can be calculated and then followed by the variance σ^2. The estimation of functional value at the unknown position $\mathbf{x}$ is $\hat{y}(\mathbf{x}) = \mathbf{f}(\mathbf{x})^T \hat{\boldsymbol{\beta}} + \mathbf{r}(\mathbf{x})^T \mathbf{R}^{-1} (\mathbf{Y} - \mathbf{F}\hat{\boldsymbol{\beta}})$.

The kriging methods can be categorized into two types: the ordinary kriging methods (OKG) and the universal Kriging methods (UKG) based on the order of polynomials used in the mean structure. The OKG assumes a zero or constant mean structure on the entire domain. The UKG constructs the mean structure using first or second-order polynomials. One extension for the UKG is the blind Kriging method [32] and dynamic Kriging method [33], both of which assume that the mean structure is unknown. In the blind Kriging method, the unknown mean model is identified using a Bayesian variable selection technique based on experimental data, while the dynamic Kriging method uses a genetic algorithm to select the optimal basis function locally.

Besides the application in deterministic simulation models, the kriging method also has been extended to approximate random or stochastic simulation models [34-36]. More detailed review of the Kriging methods and its application can be found in [37-40].

3. AN OVERVIEW OF CONCURRENT SEARCH ALGORITHM

Previously we developed a concurrent search algorithm [41] for searching multiple local minima and saddle points along one transition path without the prior knowledge of initial and final stable configurations on a PES simultaneously. The algorithm uses a Bézier curve to represent a transition path. Each control point represents one intermediate state (i.e. atomic configuration) along the transition path. For each polygon formed by the control points, two end control points are relaxed to locate two local minima. One intermediate control point climbs up to locate the saddle point between those two local minima. A constrained degree elevation and reduction scheme was developed to maintain an even distribution of control points. A curve subdivision scheme was developed to break one curve into two curve sections successively to locate multiple transition paths. The searching algorithm consists of three stages: 1) a single transition pathway search, 2) multiple transition pathway search, and 3) climbing process to locate the saddle position.

At the first stage, the algorithm locates two local minima by minimizing two end control points using the conjugate gradient method. The intermediate control points are relaxed along their corresponding conjugate directions with positive eigenvalues of the Hessian matrix for the PES.

At the second stage, the algorithm locates all local minima between two stable configurations obtained from the first stage. To locate all local minima along the path, a curve subdivision scheme is developed to check whether one curve crosses an extra energy basin with a local minimum between two stable states (i.e. end points of the curve produced from the first stage). If one curve is breakable, the algorithm breaks the curve into two curve sections representing two stages of transition at one break point. The break point is minimized using the conjugate gradient method to locate the extra local minimum, while the shape of the two newly created curves are refined following the same procedure as in the first stage. This process continues till each curve crosses only two adjacent energy basins.

At the third stage, the control point with the maximum energy within each of those transition paths climbs up to locate the actual saddle points. Similar to the first stage, a set of conjugate directions are constructed for each intermediate control point. The control point with the maximum energy along each transition path is first maximized along the direction with a negative eigenvalue. Then the control point is relaxed along all other conjugate directions. All the other intermediate control points are only relaxed in all the conjugate directions except the one with negative eigenvalues. Here, the algorithm was not implemented to execute in parallel. However, the optimization processes of the curve section are independent and can be easily implemented as parallel algorithms.

In this paper, a new search algorithm using the kriging method for surrogate model construction is developed, as an extension of the concurrent search algorithm.

4. SADDLE POINT SEARCH ALGORITHM USING KRIGING METHOD

In this section, how the Kriging method is used to construct the metamodel and integrated with the concurrent search algorithm is described. The goal is to improve the efficiency and robustness of the searching algorithm. Different from the classical Kriging methods, which select the point with maximum square error as the next sampling point to improve the accuracy of the surrogate model, here a new sampling scheme is developed to serve the purpose of locating local minima and saddle points.

Similar to the original concurrent search algorithm, the general process of the algorithm includes three stages. The major difference is that the single transition pathway search is integrated with the Kriging method. The climbing process which aims to locate saddle points also incorporate Kriging. The overall flowchart of using the Kriging method for saddle point search algorithm is shown in Figure 1.

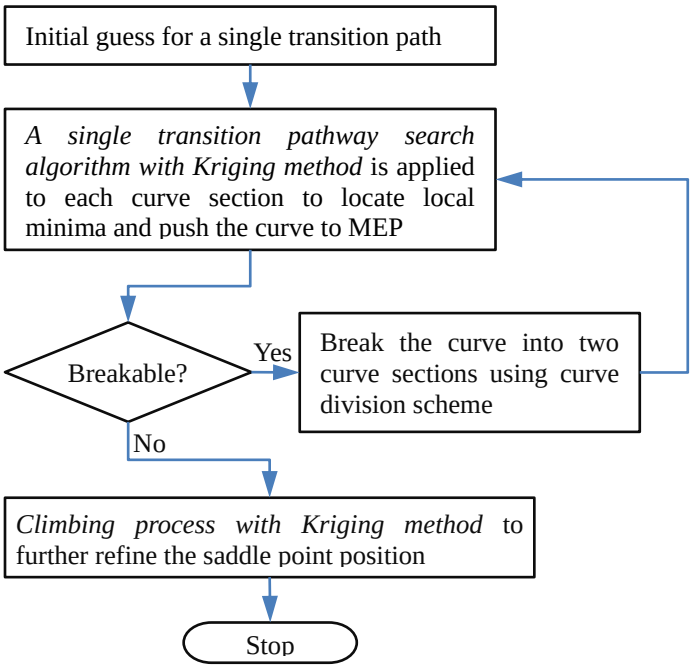

Figure 1: FLOWCHART FOR CONCURRENT SEARCH ALGORITHM WITH KRIGING METHOD

4.1 New Local Sampling Scheme for the Searching Algorithm

The major function of the surrogate model is to help decide the next sampling point in the searching process. In classical Kriging methods, the next sampling point is chosen as the one with the maximum square error. Since our primary goal is not to reduce the prediction error as fast as possible, a new sampling scheme is developed for searching local minima, which uses the criterion of choosing a sampling point with the minimum functional value around end control points.

For interpolation methods such as Kriging, the density of sample points within a region determines the accuracy of the model prediction for that region. Thus sampling needs to occur in the regions where local minima and saddle points are most likely located. The algorithm first updates the positions of control points using the real model with underlying physics, which however is computationally expensive. After collecting a predefined number of sample points which should be at least larger than the required number of design sites to construct the Kriging model, the algorithm then constructs a surrogate model using those sample points. Then the searching process is switched to the surrogate model. Since the surrogate model involves approximation error, there is no need to actually find the local minima or saddle points on the surrogate surface. After one iteration of search which includes moving both the end control points along each conjugate gradient direction and the intermediate control points along their corresponding conjugate directions, the algorithm switches the functional evaluation back to the real model. Then the searching process continues with sampling more points that are closer to the region of interest. This 'real-surrogate-real' iteration continues till the searching process converges.

To further improve the efficiency of the searching process, the new local sampling scheme is developed for the region of local minima which are located by the two end control points in our algorithm. On the surrogate model, for those two end control points, the algorithm does not use the conjugate gradient minimization method to determine their new positions. Instead, a jumping scheme is used to avoid the long local searching process on the surrogate model. For each end control point, the algorithm draws samples uniformly within its local region. The sample point with the smallest functional value is selected as the next sampling point for the end control point. The size of the sample area is decided as follows. First, the center of the sample area is set to be the current location of the end control point. Second, the area is a hypercube with the side length calculated as

$$a = \sqrt{2}c\min(d_{pre}, d_{neighbor}) \tag{5}$$

in which $c \in (0,1]$ is a constant; d_{pre} is the distance between the positions of the end point in current and previous iteration, i.e. $d_{pre} = \| \mathbf{x}^{(i)} - \mathbf{x}^{(i-1)} \|$ where i is the index for the iteration; and $d_{neighbor}$ is the distance between the end point and its neighboring control point, i.e. $d_{pre} = \| \mathbf{x}_{end}^{(i)} - \mathbf{x}_{neighbor}^{(i)} \|$. The definition of the hypercube length assures that the new end control point will not jump to the position which is far away from the closest local minimum. In addition, it prevents the formation of possible loops at the end of the curve. The following two subsections present how the proposed sampling scheme and Kriging are integrated into a single transition pathway search and climbing process.

4.2 Single Transition Pathway Search with Kriging Model

In the concurrent search algorithm, the local minimum is identified using the conjugate gradient method and the path is refined by moving the intermediate control points along conjugate direction, both of which require a large number of functional evaluations for the physical model. To reduce the number of functional evaluations, the new algorithm developed in this paper keeps track of all information about the searching history (i.e. the functional values and gradients at the control points in all previous iterations) and incorporate it into the surrogate construction and searching process by the Kriging method. The algorithm embodies the search history into the construction and refinement of the surrogate model. The memory is then used to help decide the future search.

The general process for the single transition pathway search algorithm with Kriging is as follows. First, the algorithm updates the positions of the end points based on the real model using the conjugate gradient method and the positions of intermediate control points by moving them along conjugate directions until a predefined number of functional evaluations is reached. During this searching process, the functional evaluations in the line search along conjugate directions are conducted based on the real model until the number of evaluations reaches a threshold level. When more functional evaluations are required in the line search, the surrogate model is used instead so that the associated computational cost can be reduced. Second, a surrogate model is constructed or updated using all available sample points and functional values. The whole searching process on the surrogate model is based on the evaluation of the surrogate. Third, when the searching process switches from the real model to the surrogate model, the positions of the end control points are updated with the sample points with the minimum functional values within the corresponding sample areas on the surrogate model. The intermediate control points move along the conjugate directions with all the functional evaluations conducted on the surrogate model. Fourth, after one searching iteration on the surrogate model, the searching process switches back to the real model except that the second portion of the functional evaluations during each line search are still performed on the surrogate model. The overall flowchart of single transition pathway searching with the Kriging method is shown in Figure 2.

4.3 Line Search with Kriging Model

Line search is one of the critical components for the searching algorithm as it involves in all stages of the searching process. During the searching of local minima and MEP, line search is used to determine the minimum or maximum along each conjugate direction. Hence, the line search is widely used to search the optimum in a particular direction. For large systems with many degrees of freedom, a large number of line searches are required since a line search is applied to each conjugate direction. Therefore introducing Kriging will significantly improve the efficiency of the algorithm with reduced numbers of functional evaluations during line searches.

During the line search, the second portion of functional evaluations are conducted on the surrogate model. The general process for the line search is as follows. First, the algorithm conducts a couple of trial searches to determine an appropriate step length, which is based on the real model. Second, after the step length is determined, the algorithm refines the position of the control point along the direction using the determined step length. In this step, functional evaluations are also conducted on the real model. Third, the algorithm refines the surrogate model based on the results from the first and second steps and then continues updating the position of the point with functional evaluations being conducted on the surrogate model until the predefined stop criteria are satisfied.

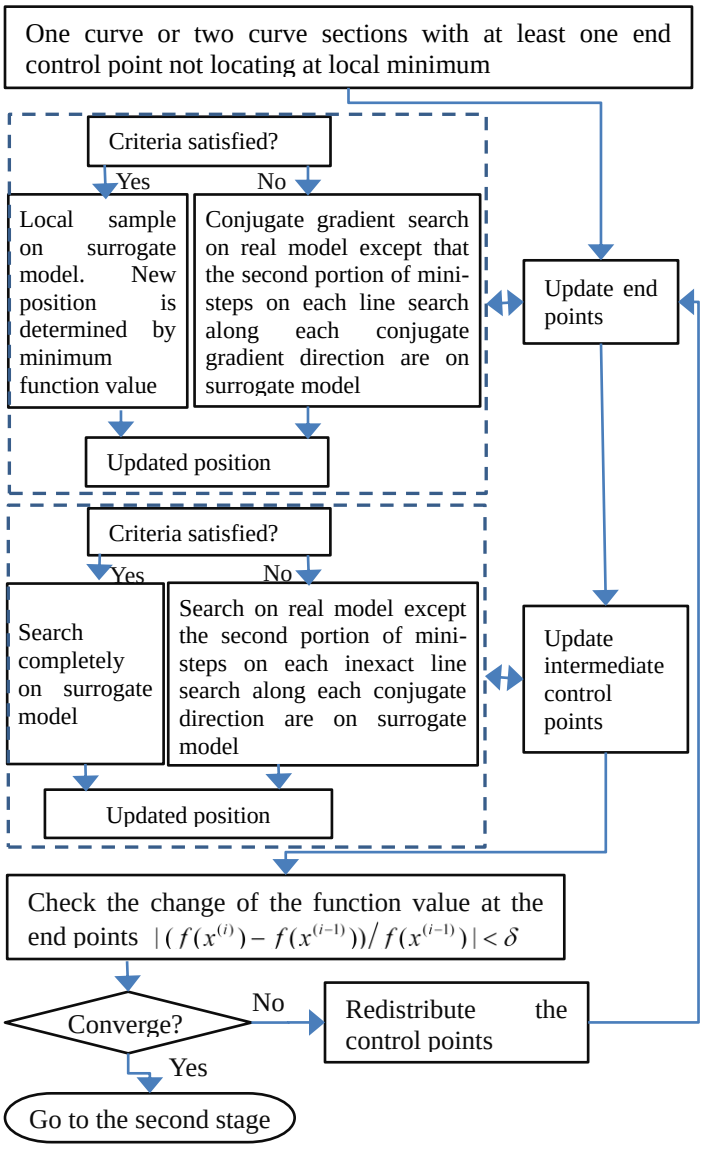

Figure 2: FLOWCHART FOR SINGLE TRANSITION PATHWAY SEARCH ALGORITHM WITH KRIGING

4.4 Climbing Process with Kriging Model

During the climbing stage in the concurrent searching algorithm, the control point with the maximum energy for each path climbs up to the saddle position, while all the other intermediate control points are minimized along their corresponding conjugate directions with positive eigenvalue. Integrated with Kriging, the new climbing process is as follows. First, the algorithm lets the control point with the maximum energy climb up along a conjugate direction and minimize all of the intermediate control points along their corresponding conjugate directions with positive eigenvalues. During the process, the construction of the conjugate directions is conducted on the real model except that some functional evaluations during the line search are conducted using the surrogate model. Second, after the number of iterations reaches a threshold, the algorithm refines the surrogate model and the whole searching process is conducted on the refined surrogate model.

5. DEMONSTRATION

Here the well-known LEPS potential [42], Rastrigin function, and Schwefel function are used to demonstrate the new algorithm. We use these simple functions to help visualize the searching process and results. The Rastrigin and Schwefel functions are among the most used benchmark functions for global optimization, as defined in Table 1. The implementation is done using MATLAB and the Kriging model is constructed with DACE toolbox [43]. In this experiment, the basis polynomial function is set to be the second order and the correlation function is Gaussian correlation function.

The experiments are designed to test the accuracy and efficiency of the algorithm. The general process for the demonstration is as follows. First, we test the accuracy of the algorithm by setting a set of initial positions and test the capability of locating the local minima and saddle points. Second, we test the efficiency of the algorithm by comparing the average number of functional evaluations required to locate one transition path between the new searching algorithm with Kriging and the original concurrent searching algorithm. Since the most time-consuming portion of the saddle point searching algorithm is the functional evaluations on the real model, we use the total average number of functional evaluations as the metric to compare the efficiency in the examples.

5.1 Test Result for LEPS Potential

The LEPS potential model describes a reaction involving three atoms A, B, and C whose motions are restricted along a straight line. This function is a benchmark function for saddle point search algorithms. Here, we use two different initial positions to test how well the new algorithm works. Figure 3(a) shows the located local minima and corresponding saddle points by using concurrent searching algorithm, while Figure 3(b) shows the results by the integrated Kriging searching algorithm. By comparing the results in Figure 3(a) and (b), it clearly shows that the new algorithm maintains a good accuracy of locating the local minima and the saddle point. The final path identified by the algorithm is very similar to the ones identified by the concurrent search algorithm. Table 2 shows that the total number of functional evaluations for the new algorithm is much lower than the one for the concurrent search algorithm.

5.2 Test Result for Rastrigin Function

We choose a PES defined by Rastrigin function with multiple local minima and saddle points to demonstrate the capability of locating multiple saddle points. The Rastrigin function is a non-convex function which is frequently used to test global optimization algorithms. The function has a global minimum at $\mathbf{x} = (0, \cdots, 0)$ and local minima are uniformly distributed as shown by the contour plot in Figure 4. We test the new algorithm using four sets of initial positions. The results in Figure 4 show that the algorithm accurately locates local minima and saddle points. The identified final paths are also consistent with the ones identified by the concurrent search algorithm.

The total numbers of functional evaluations for the four initial positions are shown in Table 3. To minimize the influence of the initial positions on the complex functional landscape, four different initial positions are applied and the total average number of functional evaluations per final path (N/path) is shown in Table 3. The result in Table 3 shows that in average the number of functional evaluations required to locate one final path for the new algorithm with integrated Kriging is less than half of the one for the concurrent search algorithm. The algorithm works very well for the Rastrigin function.

5.3 Test Result for Schwefel Function

A third example to test the algorithm is Schwefel function which has a relatively non-uniform PES as shown in Figure 5. We follow the same procedure as for the Rastrigin function. The results are shown in Figure 5 and Table 4. The total average number of functional evaluations for the new algorithm is 326 which is lower than the one (461) for the concurrent search algorithm.

Table 1: TEST FUNCTIONS

Function	Definition
LEPS potential	$V^{LEPS}(r_{AB}, r_{BC}) = \frac{Q_{AB}}{1+a} + \frac{Q_{BC}}{1+b} + \frac{Q_{AC}}{1+c} - [\frac{J_{AB}^2}{(1+a)^2} + \frac{J_{AB}^2}{(1+a)^2} + \frac{J_{AB}^2}{(1+a)^2} - \frac{J_{AB}J_{BC}}{(1+a)(1+b)} - \frac{J_{BC}J_{AC}}{(1+b)(1+c)} - \frac{J_{AB}J_{AC}}{(1+a)(1+c)}]^{1/2}$ where $Q(r) = \frac{d}{2}\left(\frac{3}{2}e^{-2\alpha(r-r_0)} - e^{-\alpha(r-r_0)}\right)$ $J(r) = \frac{d}{4}\left(e^{-2\alpha(r-r_0)} - 6e^{-\alpha(r-r_0)}\right)$
Rastrigin	$f(\mathbf{x}) = 10n + \sum_{i=1}^{n}[x_i^2 - 10\cos(2\pi x_i)]$
Schwefel	$f(\mathbf{x}) = 418.9829n - \sum_{i=1}^{n} x_i \sin\sqrt{\lvert x_i \rvert}$

Table 2: TEST RESULTS FOR LEPS POTENTIAL

Algorithm		Pos_1	Pos_2	Total	N/path
Con	N_f	2104	6264	8368	4184
	N_p	1	1	2	
Krig	N_f	643	941	1584	792
	N_p	1	1	2	

Note: "Con" represents the concurrent search algorithm; "Krig" indicates the new search algorithm integrated with Kriging; "Pos_*" means the initial position * which are shown in Figure 3; N_f is the number of function evaluation at each initial position; N_p is the number of identified final paths; N/path is the average number of functional evaluations to locate one final path.

Table 3: TEST RESULTS FOR FUNCTION RASTRIGIN

Algorithm		Pos_1	Pos_2	Pos_3	Pos_4	Total	N/path
Con	N_f	4289	4186	2120	3452	14047	502
	N_p	8	8	6	6	28	
Krig	N_f	1594	1841	1437	795	5667	218
	N_p	8	7	7	4	26	

Table 4: TEST RESULTS FOR FUNCTION SCHWEFEL

Algorithm		Pos_1	Pos_2	Pos_3	Pos_4	Total	N/path
Con	N_f	3037	3799	2742	2875	12453	461
	N_p	7	7	6	7	27	
Krig	N_f	2380	1937	1785	4028	10130	326
	N_p	7	8	8	8	31	

6. DISCUSSIONS AND FUTURE WORK

In this paper, a saddle point search algorithm using Kriging metamodels is developed to improve the efficiency of the searching algorithm. Different from existing saddle point search methods, this method keeps search history in memory. It uses the information collected from previous iterations to guide the search in the next iteration. Kriging models are constructed and updated based on the sample points collected in previous iterations. The cost of the evaluation of metamodels is much less and negligible compared to physics-based simulation such as DFT, which can significantly reduce the number of functional evaluations while the accuracy of the searching algorithm is maintained. The algorithm starts with searching local minima and saddle points with the evaluation on the real model. After collecting enough samples, a surrogate model is constructed. Then the searching process is switched to the surrogate model. After the prediction of the locations of new samplings, the algorithm switches the searching process back to the real model.

The results show that the algorithm works well with the simple examples such as LEPS potential, Rastrigin, and Schwefel functions. In the future work, we will apply the algorithm to large material systems with higher dimensions. In addition, in the current algorithm, the jump process is the most time consuming portion of searching procedure on the surrogate model. We will investigate the possible ways to improve the efficiency further, although the evaluation of surrogate models is much more efficient than the evaluation of physical models with simulation.

In this paper, we only demonstrate the efficiency of the new search algorithm. The future efforts will also include the improvement of robustness of searching algorithm given that the physical models also contain model errors. In other words, the true value of physical quantity such as potential energy is unknown. The physics-based simulation is just an estimation of the true one. However, uncertainty introduced because of the numerical treatment in the modeling and simulation process. We will include the stochastic error prediction in the future surrogate model construction and thus the sampling strategy will be adjusted accordingly.

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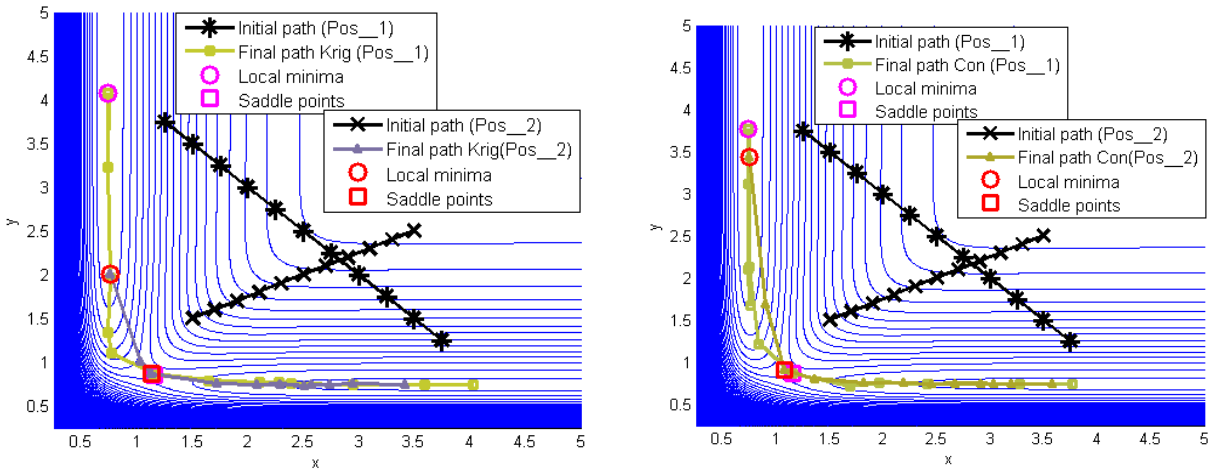
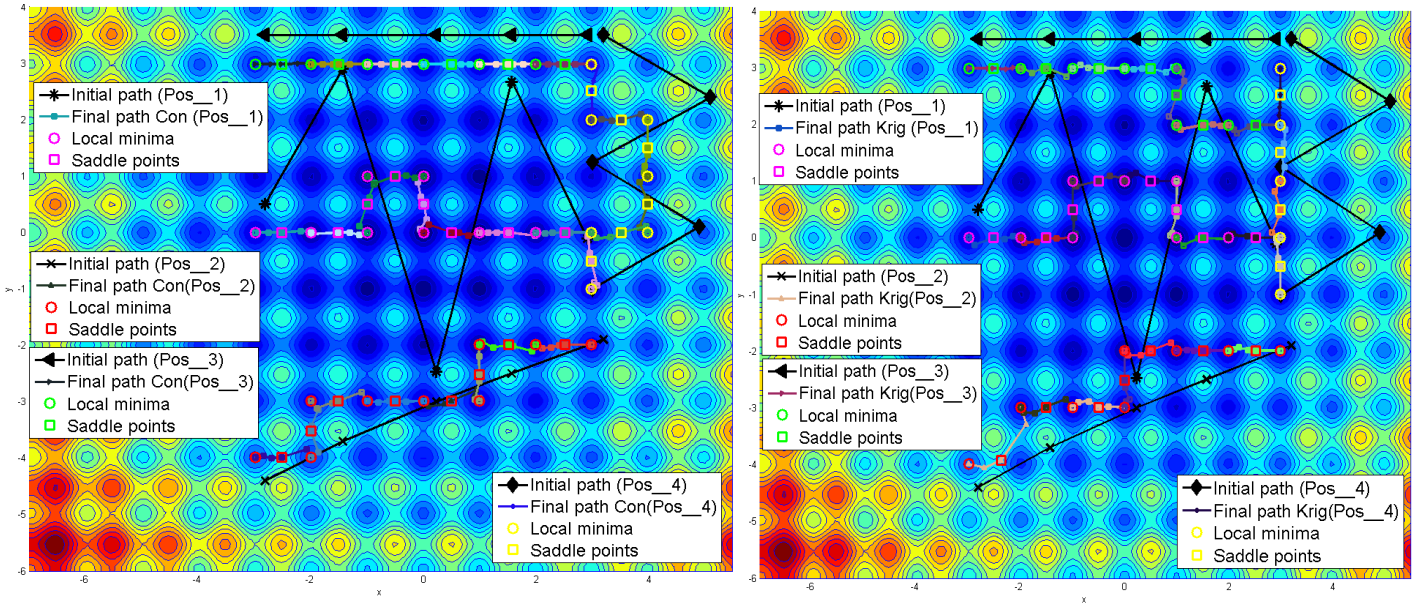


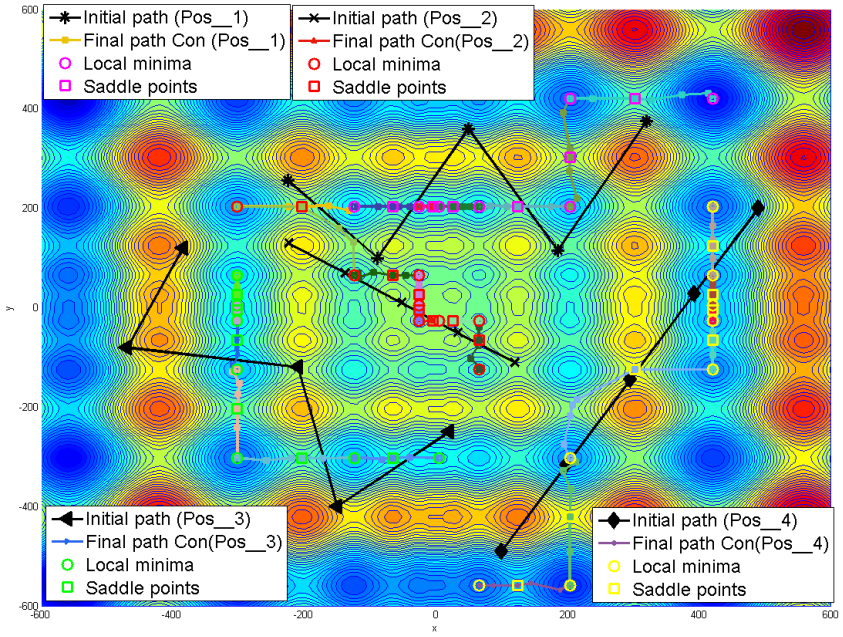
(a) (b)

Figure 3: TEST RESULTS FOR LEPS POTENTIAL (A) CONCURRENT SEARCHING ALGORITHM (B) INTEGRATED KRIGING SEARCH ALGORITHM

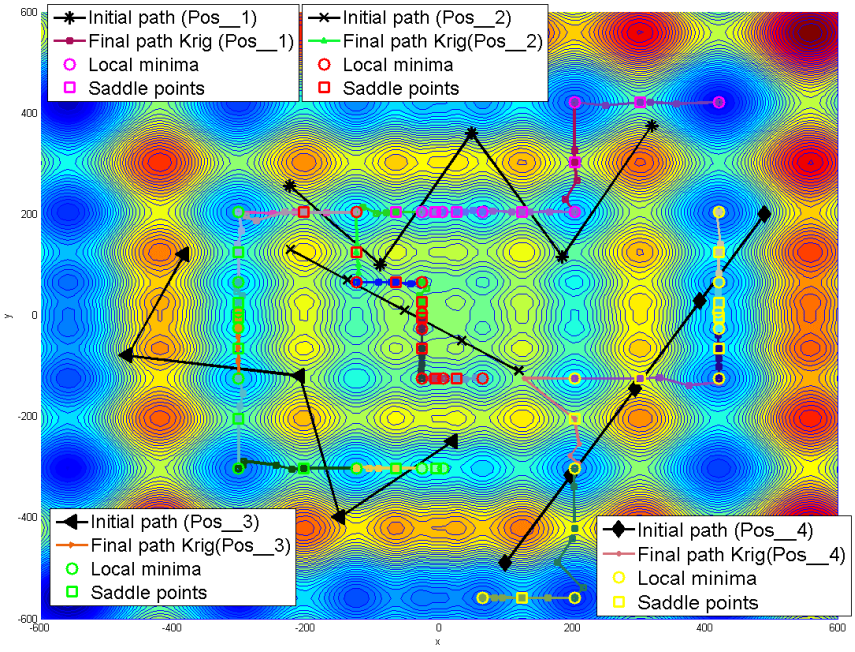


(a) (b)

Figure 4: TEST RESULTS FOR RASTRIGIN FUNCTION (A) CONCURRENT SEARCHING ALGORITHM (B) INTEGRATED KRIGING SEARCH ALGORITHM



(a)



(b)

Figure 5: TEST RESULTS FOR SCHWEFEL FUNCTION (A) CONCURRENT SEARCHING ALGORITHM (B) INTEGRATED KRIGING SEARCH ALGORITHM