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Self-Evolving Atomistic Kinetic Monte Carlo simulations of defects in materials

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ABSTRACT

The recent development of on-the-fly atomistic kinetic Monte Carlo methods has led to an increased amount attention on the methods and their corresponding capabilities and applications. In this review, the framework and current status of Self-Evolving Atomistic Kinetic Monte Carlo (SEAKMC) are discussed. SEAKMC particularly focuses on defect interaction and evolution with atomistic details without assuming potential defect migration/interaction mechanisms and energies. The strength and limitation of using an active volume, the key concept introduced in SEAKMC, are discussed. Potential criteria for characterizing an active volume are discussed and the influence of active volume size on saddle point energies is illustrated. A procedure starting with a small active volume followed by larger active volumes was found to possess higher efficiency. Applications of SEAKMC, ranging from point defect diffusion, to complex interstitial cluster evolution, to helium interaction with tungsten surfaces, are summarized. A comparison of SEAKMC with molecular dynamics and conventional object kinetic Monte Carlo is demonstrated. Overall, SEAKMC is found to be complimentary to conventional molecular dynamics, especially when the harmonic approximation of transition state theory is accurate. However it is capable of reaching longer time scales than molecular dynamics and it can be used to systematically increase the accuracy of other methods such as object kinetic Monte Carlo. The challenges and potential development directions are also outlined.

1. Introduction

Simulating materials processes from femtoseconds to millennia is a grand challenge [1]. This challenge is extremely daunting, partially because many long-term material properties and phenomena are driven by a unit event occurring at a split second, and partially because no experimental or theoretical tool has the capability to cover such huge span in time so far. For instance, the defect production event in structural materials for nuclear energy systems is on the order of picoseconds, while subsequent defect evolution and interaction that lead to property changes last for many decades [2–4]. However, the need to bridge the time scale is imperative, not only for fundamental understanding of how unit events and final outcomes are correlated, but also for practical applications, such as predicting the lifetime of nuclear structural materials or a modern battery.

A framework of multiscale simulation methods has been established to address the gap in time and length scale [5,6]. The methods include electronic structure calculations, primarily density functional theory [7–9], atomistic simulations using molecular dynamics (MD), mesoscale models, such as kinetic Monte Carlo (KMC) [10–19], and continuum-level models based on elasticity theory. However, the connection between different methods to enable information passing is still a major difficulty. For instance, the parameters for a new empirical potential may be fitted using the data generated by first principles calculations in order to improve the accuracy and fidelity. However, there is no guarantee the new potential will maintain this accuracy and fidelity for properties beyond those used to fit the potential. Another example is the passing of kinetic defect parameters from MD simulations to object KMC (OKMC) [10–12]. Because only the migration energy barrier and prefactor that are extracted from MD are used as input in OKMC, the details of defect configurations and how they interact with each other are severely simplified and lost because all the defects are treated as abstract objects. Therefore, the predictive capability and fidelity of the overall model is greatly limited. Therefore, the potential to improve the current multiscale simulation framework is tremendous and recent developments in accelerated molecular dynamics [20–23] and atomistic kinetic Monte

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Carlo (AKMC) [13–17] have greatly extended the limit in accessible time and therefore allow more systematic improvement of the framework.

Accelerated molecular dynamics [23], including Hyperdynamics [20], Parallel Replica Dynamics (PRD) [21], and Temperature Accelerated Dynamics (TAD) [22], remains in the framework as MD, the time scale of which is generally limited to nanoseconds due to the need to sample atomic vibrations. Through the introduction of a bias potential, such as those in Hyperdynamics [20], or increasing the temperature, such as in TAD, the transition from state to state is accelerated and some long-term behavior or phenomenon may be revealed. The details of these techniques are reviewed in another article in this issue. In this review, we primarily focus on an alternative approach, which is KMC.

System evolution in KMC simulations is based on the saddle point energies or barriers that separate distinct states of the system. In contrast, evolution in MD requires sampling many local atomic configurations on the time scale of atomic vibrations; therefore KMC offers great potential for accelerating events when barriers are high. Generally speaking, KMC can be separated into OKMC and AKMC. OKMC or event-based KMC considers every defect as an abstract object and the potential system change as an event. Most AKMC have been lattice based, which generally works for point defects, such as vacancies or substitutional impurities, but fails for more complex defect configurations, such as interstitial clusters.

Recently there has been an increased amount of attention and effort in developing on-the-fly atomistic KMC, in which the saddle point configurations and energetics are determined as system evolves. The recent developments include adaptive KMC [13,14], self-learning KMC [24,25], local environment KMC [26], kinetic Activation Relaxation Technique (k-ART) [15,18,27], and Self-Evolving Atomistic Kinetic Monte Carlo (SEAKMC) [16,17]. This type of simulation has several obvious advantages. First, the requirement of pre-determining the potential saddle points has been eliminated, which by itself is a very challenging and time-consuming process. Second, the catalog of potential processes evolves as simulation progresses. More importantly, the atomistic interactions provide significant improvement over assumed simplified or artificial interactions in previous models.

2. Self-Evolving Atomistic Kinetic Monte Carlo (SEAKMC)

The development of SEAKMC is motivated by several facts. First, defects in crystalline materials govern material properties and phenomena. Second, many defects, such as vacancies and interstitials, are localized; their influence is rather short-ranged. Therefore, it is imperative to focus on defects, which are the key to revealing the structure–property relationship in materials. Third, it is found that the successful rate of finding useful saddle points decreases rapidly as system size gets larger. This is because the system energy increases during the initial phase of saddle point searches. As the system size expands, the total system energy correspondingly increases, sometimes to a very high-energy state compared with the initial configuration. This result in saddle points being found with too high energy or not connected to the initial configuration. Taking vacancy diffusion in bcc iron as a simple example, the successful rate of finding a saddle point associated with first nearest neighbor jump is close to 100% when a small volume (2.5 lattice-parameters) is employed during the saddle point search. When this volume is expanded to 5.5 lattice-parameters, the success rate decreases significantly. Almost all the saddle point searches did not lead to the correct configurations. The energy changes during the search for these two different volumes are given in Fig. 1. The implication from this simple case is that without some type of localization technique, even vacancy diffusion in large systems is very inefficient. The situation is worse for complex defects or defect clusters, which require an even larger system size. Without proper handling of this issue, the system size in these on-the-fly techniques is very limited, which can prohibit certain processes or phenomena from being studied. We note that modifications to the activation scheme that limit the number of atoms deformed in the first step of the dimer saddle-search leads to more favorable outcomes, but it still performs badly at large lat-

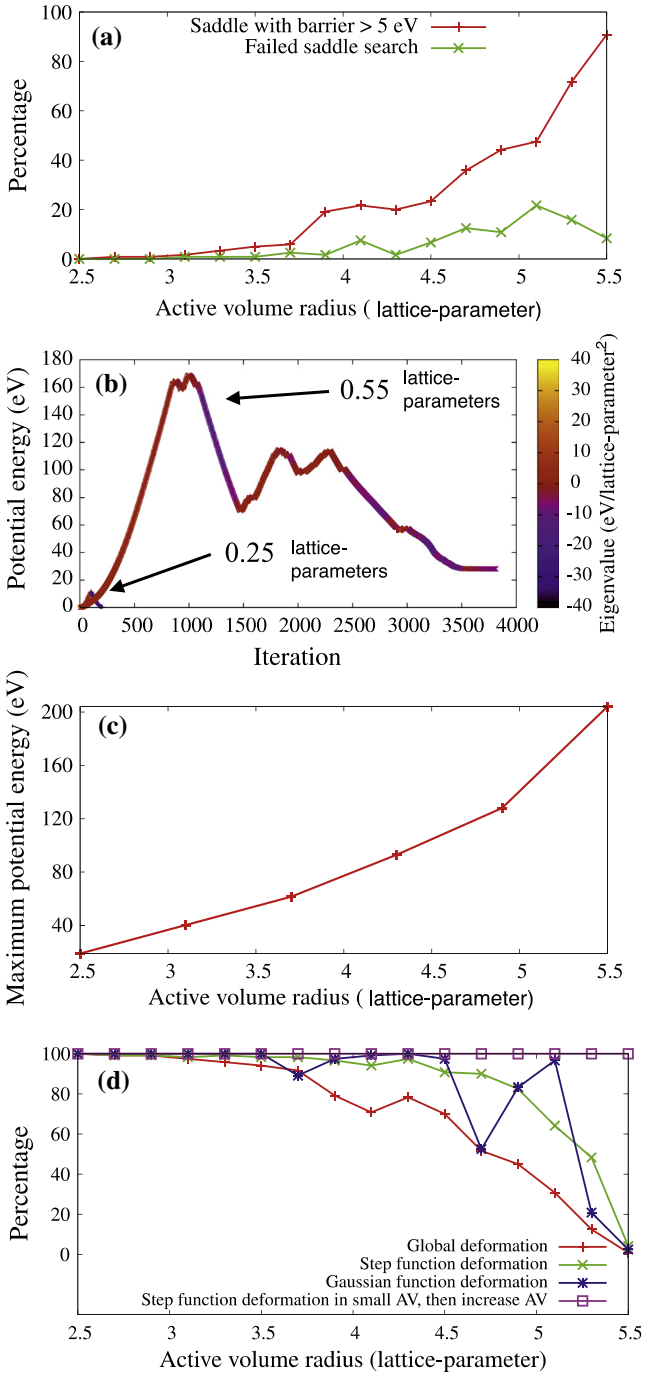

Fig. 1. The effect of active volume on saddle point searches: (a) the percentage of failed saddle searches and configuration with unrealistic high energy barriers using global deformations; (b) the system energy increase for active volume size with 2.5 and 5.5 nm, respectively; (c) the potential energy increase as a functional of active volume sizes; (d) the percentage of successful saddle searches with realistic energy barriers using different activation schemes.