Applications of TAMMBER

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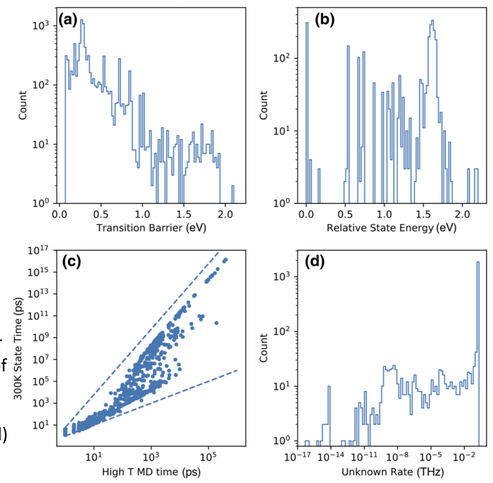
Application to interstitial clusters in Fe

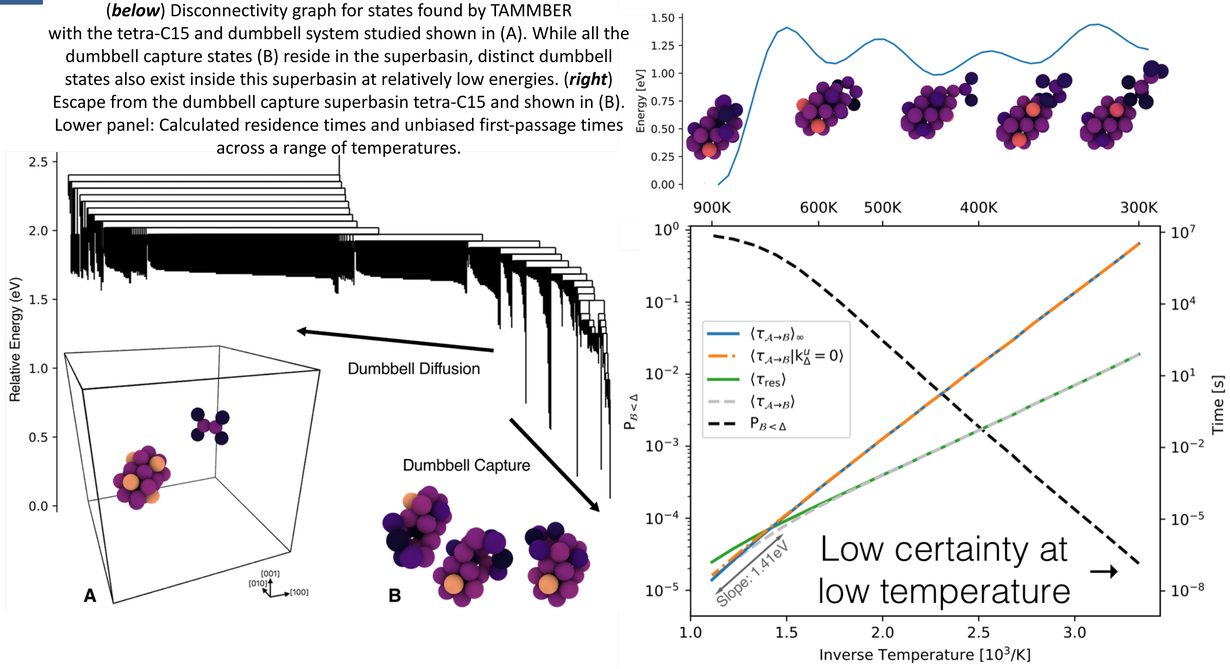
We used TAMMBER to investigate the capture and release of single interstitial atoms defects by very stable 'C15' interstitial clusters in bcc Fe, an important process in post irradiation annealing. The disconnectivity graph shows the two clear super basins corresponding to interstitial diffusion and interstitial capture. By sampling this complex landscape, the dominant capture and release mechanisms can be determined from many potential pathways.

An important feature of TAMMBER is the ability to provide ***uncertainty quantification of the dominant mechanism***, allowing ***mesoscale error propagation***. In this example, we found that, despite running for hours on thousands of cores, the prediction of the rate of interstitial release remains uncertain at low temperatures, as the expected passage time of the dominant mechanism was longer than the network validity time.



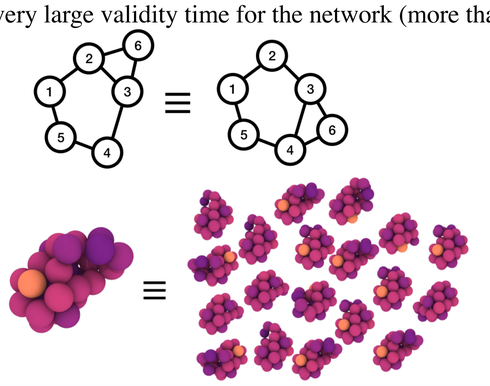
Summary of the TAMMBER simulation for the C15-dumbbell system. (a) Histogram of energy barriers. (b) Histogram of state energies. (d) Effective low temperature time versus high-temperature MD time. (d) Histogram of unknown rates.

(***below***) Disconnectivity graph for states found by TAMMBER with the tetra-C15 and dumbbell system studied shown in (A). While all the dumbbell capture states (B) reside in the superbasin, distinct dumbbell states also exist inside this superbasin at relatively low energies. (***right***) Escape from the dumbbell capture superbasin tetra-C15 and shown in (B). Lower panel: Calculated residence times and unbiased first-passage times across a range of temperatures.

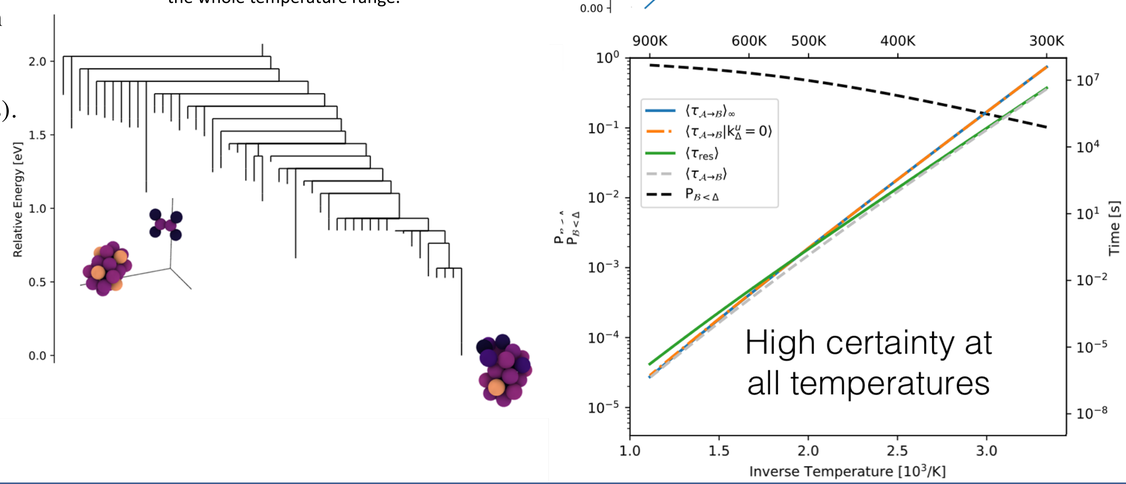


Symmetry aware sampling

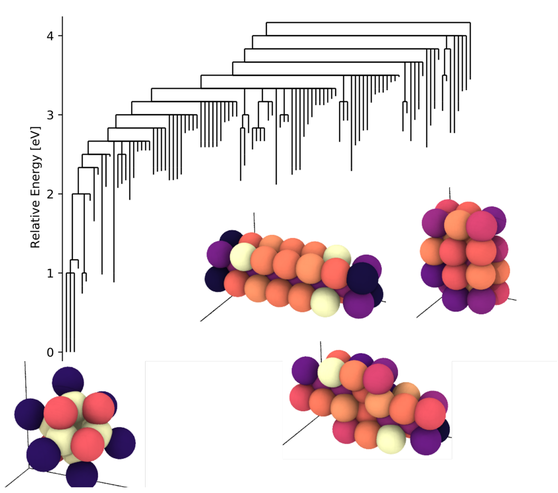
Crystals are highly degenerate under exchange of identical atoms, rotation and translation. Being unaware of this degeneracy while the calculation is performed leads to computational inefficiencies that limit the validity time. However, representing configurations as neighbor connectivity graphs, crystal symmetries become graph isomorphisms. We have exploited this to significantly compress the energy landscape of the Fe system we studied, allowing for a much better certainty (more than 90%) on the dominant process at all target temperatures for a similar computational cost. We also note the very large validity time for the network (more than 10^6 seconds).



(***below***) Symmetry-compressed disconnectivity graph for the tetra-C15 and dumbbell system. (***right***) The reduced number of topologically distinct states greatly improves the computational efficiency, leading to improved certainty on the interstitial release time over the whole temperature range.

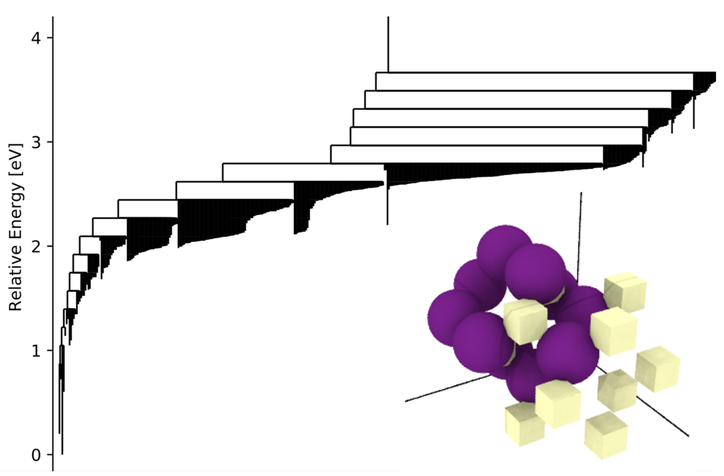


Application to MgO and UO_2



We have begun applying TAMMBER to investigate energy landscape of point defect clusters in ceramics such as MgO and UO_2, which due to the presence of many shallow minima represents an extremely challenging task for current sampling methods. The results of the simulations are presently being analyzed.

For a hexa-interstitial defect in MgO (left), TAMMBER achieved a validity time of about 10^{10} s at 400K. For a Schottky defect in UO_2 (right), the validity time was 0.5 s at 850K.



Next Steps

We are currently exploring ways to optimize specific quality metrics that would explicitly depend on the property under consideration, instead of generically through the validity time.

We are also developing techniques to upscale the atomistic information to the mesoscale, for example through reaction-diffusion equations or by coarse-graining state space. This will be very useful to make connections with cluster dynamics models, while retaining the benefits of the uncertainty quantification that underpins TAMMBER.

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Reference

TD Swinburne and D Perez. "Self-optimized construction of transition rate matrices from accelerated atomistic simulations with Bayesian uncertainty quantification." Physical Review Materials 2.5 (2018): 053802.

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