

Kinetic Parameters of Surface Reactions from Monte Carlo Simulations Combined with Evolution Strategies

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Dynamic, or Kinetic, Monte Carlo (DMC) simulations can yield for all practical purposes exact results for most systems of surface reactions. They are used very successfully with simple models to derive new kinetic concepts and with realistic models to simulate kinetic experiments. They are used, however, very little to derive kinetic parameters (not just activation energies and prefactors, but also e.g. lateral interactions) by fitting DMC results to experiments. One problem is that, because DMC is a stochastic method and its results are noisy, it is hard to determine how accurate one's kinetic parameters reproduce an experiment. The amount of noise can be reduced but only at the cost of a lot of computer time. The stochastic nature also prevents the use of most numerical fitting methods that use derivatives. I am using DMC simulations of kinetic experiments and combining them with Evolution Strategies (ES) to obtain kinetic parameters. ES varies the kinetic parameters, runs DMC simulations with these parameters, evaluates the results of these simulations, and based on these evaluations tries to optimize the fit to experiments. ES is a method from Evolutionary Computation, which consists of methods that mimic natural evolution. (Genetic Algorithms is probably the best-known method of this area of computer science.) ES is robust, which means that it can handle noisy data. It also does a good global search for the best fit. Its main drawback is that it is computationally quite expensive. A single optimization run generally does many thousands of DMC simulations. However, it can easily be parallelized and the ES+DMC combination runs very well on PC-clusters. I show results of the determination of kinetic parameters of various systems from surface science.