565e Multiscale Systems Engineering with Application to Copper Electrodeposition

Mohan Karulkar, Feng Xue, Timothy O. Drews, Yuan He, Xiaohai Li, Effendi Rusli, Richard C. Alkire, and Richard D. Braatz

New applications are being discovered in materials, medicine, and computers where the control of events from molecular to macroscopic length scales is critical to product quality [1]. This has motivated the creation of multiscale systems engineering techniques [2] that are applicable to processes whose key features cover many orders-of-magnitude in time and length scales. This paper is an update on our efforts in the development and implementation of such techniques in the electrodeposition of copper interconnects during the manufacture of electronic devices, for which a reaction mechanism has recently been published that elucidates the synergistic effect of multiple additives that influence deposit shape evolution [3]. The present paper includes (1) a new algorithm for the optimization of multiscale systems, and (2) the application of the new multiscale optimization algorithm to the estimation of kinetic parameters in the additive mechanism based on experimental data from atomic force microscopy images and current-potential curves.

This paper utilizes a multiscale systems approach that consists of stochastic parameter sensitivity analysis, Bayesian parameter estimation applied to ab initio calculations (when available) and experimental data, model-based experimental design, hypothesis mechanism selection, and multi-step optimization [4]. While the objective of each systems sub-problem are the same as for models described by macroscopic continuum models, the numerical algorithms implementing these methods are different, due to the characteristics of multiscale simulation models such as stochastic fluctuations in simulation outputs, very high computational cost, and the wide variation of structure in the underlying model equations.

The most computationally challenging aspect of multiscale systems engineering techniques concerns tasks in which optimization calculations are required, such as in Bayesian parameter estimation and multiscale design optimization. Most multiscale simulation models produce stochastically fluctuating outputs, so that the direct application of gradient-based optimization algorithms to such models is problematic. Further, a typical multiscale simulation may take 1 day to run using multiple processors on a supercomputer, whereas a typical optimization algorithm may take 100s to 1000s of calls to the multiscale simulation code to converge—resulting in years of computation. Spending years doing computations is not feasible given the desired pace of modern innovations.

This paper presents a new algorithm for the optimization of multi-scale systems. The first step of the algorithm is to perform an initial optimization based on a simulation model in which all stochastic simulation (e.g., kinetic Monte Carlo, molecular dynamics) components are replaced by continuum equations defined by taking the continuum limits of the stochastic simulations. Although the resulting continuum code usually will have fewer parameters (and hence cannot be estimated in this first step) and its outputs will be only a subset of the experimentally observed variables (and hence not all of the experimental data are used in this first step), the computational cost of simulating the resulting set of continuum equations is many orders-of-magnitude faster than simulating the original multiscale simulation. In addition to computing nominal parameter estimates, a multivariate probability distribution is computed that quantifies the uncertainty in the resulting parameter estimates [5]. This information is used to define the region of feasible parameters to optimize in the second step, which is to perform a multi-step optimization algorithm on the full multiscale model [4,6]. The first step determines the right "ballpark" for most of the parameters, and the second step fine-tunes those parameters while estimating those parameters that do not appear in the continuum model.

The new multiscale optimization algorithm is applied to the estimation of kinetic parameters for copper electrodeposition from atomic force microscopy images and current-potential curves. Copper electrodeposition has become one of the most important processes for the fabrication of on-chip interconnects [3,7]. Although copper electrodeposition has been used industrially for almost a century, new challenges arise when it is used for interconnect fabrication, since the interconnects must be fully filled with copper (i.e. contain no voids) and should contain minimal amounts of entrapped electrolyte and additives. Furthermore, as device sizes shrink, precise control of nucleation inside the device becomes important so that single clusters do not close the feature prematurely causing void formation.

The multiscale simulation model for the copper electrodeposition process is comprised of both continuum macroscopic (implemented as a finite volume code) and stochastic meso-scale components (implemented as a coarse-grained kinetic Monte Carlo simulation code). The KMC code is coarse-grained [8,9] so that the simulation domain can be set to the same dimensions and the same pixilation as images obtained from an Atomic Force Microscope (AFM). The finite volume code is a dynamic 2D code that simulates the diffusion and migration of additives and electrolyte species from the diffusion boundary layer to the electrode surface. Recently we have implemented a reaction mechanism into the KMC code which is based on results recently reported by IBM [3]. The parameters in the mechanism were fit to the AFM images and current-potential curves measured for 36 sets of experimental conditions specified by a D-optimal experimental design [10].

Since the hypothesized mechanism has approx. 50 parameters, only the most sensitive inputs are tuned during parameter estimation, which increased the parameter identifiability. The parameter sensitivities were computed by an optimal stochastic finite difference method [11], which can provide improved estimates over standard expressions. In the first step of the multiscale optimization algorithm, the KMC simulation code was replaced by the continuum limit in which the surface diffusion coefficient becomes sufficiently large. At that limit the process can be described by continuum reaction equations whose states are the coverages of the various species on the copper surface. These reactions were integrated simultaneously with the differential-algebraic equations obtained by applying the finite volume method to the electrolyte, resulting in a very speedy continuum approximation for the original multiscale simulation code. The most sensitive parameters in the continuum code were estimated using sequential quadratic programming applied to minimize the differences between the simulated and experimental current-potential curves, with the surface diffusion coefficient being the only sensitive parameter that could not be estimated in this first step of the multiscale optimization algorithm. The parameter estimation algorithm for this first step converged in less than a single day. A hyperellipsoidal uncertainty description computed using standard multivariate statistical methods [5] defined the feasibility region for the most sensitive parameters to estimate during the second step, which were computed using a multi-step algorithm [4] that was based on an algorithm in the literature [6], with several revisions to reduce computational cost (for details see [4]).

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