**Guidelines for the Design of Multiscale Simulation Codes**

*Mohan Karulkar, Yuan He, Richard C. Alkire, and Richard D. Braatz*
University of Illinois at Urbana-Champaign
209 Roger Adams Laboratory, Box C-3
600 South Mathews Avenue
Urbana, Illinois 61801-3602

New electrochemical applications are being discovered in materials, medicine, and computers where the control of events at both molecular and macroscopic length scales is critical to product quality [1]. Recent advances in numerical algorithms and computer speed and memory have motivated investigations as to how the behavior of such systems may be predicted using multiscale simulation. The main objectives of this paper are to provide (1) an overview of the computational aspects that arise in multiscale simulation, and (2) guidelines for the design of multiscale simulation codes for particular applications.

Different numerical methods are most effective for simulating different length scales, which has motivated efforts to simulate overall multi-scale systems by linking multiple simulation codes created at each scale. There are numerous examples where time and length scales are coupled in a serial fashion, where the results from one simulation code are used in another simulation code [e.g., 2, 3, 4]. For quasi-static problems, the quasi-continuum method [5] couples the atomistic and continuum scales by using a system-wide finite element mesh that is refined to atomic dimensions where needed. Many papers propose iterative algorithms to converge codes at multiple length scales to a steady-state or quasi-steady-state solution [6, 7].

The need to simulate *dynamical* systems where a wide range of time and length scales are tightly coupled have motivated efforts to address the more challenging problem of *concurrent* multiscale simulation [8]. An approach applicable to some systems is the use of an effective reactivity to link atomic and continuum scales [9]. The direct numerical simulation approach involves running the simulation codes at each length scale simultaneously, with each code continually passing updated boundary conditions to the other codes [10]. This can be modified to run internal iterations to force convergence of the information passed between simulation codes [11,12], or to update the continuum codes more slowly than the atomistic codes, in accord with their different time scales [13]. The latter approach has been used to simulate the electrodeposition of copper into trenches to form interconnects in electronic devices (see Fig. 1).

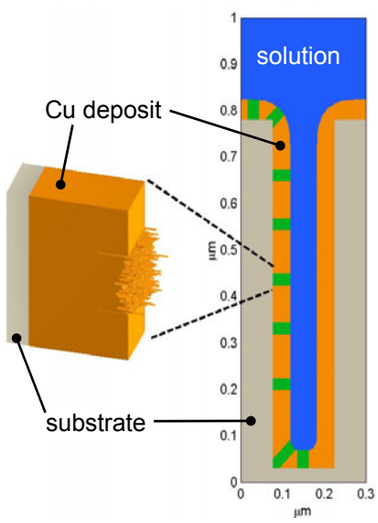


Fig. 1: Simulation of trench in-fill by dynamically coupling (a) multiple (2+1)D KMC codes for simulating reactions and surface diffusion at discrete positions on the surface, (b) a 2D level set method for simulating the changing position of the metal-solution interface, and (c) a 2D finite volume code for simulating the reaction-transport equations in the fluid. The figure at the left shows the surface inhomogeneity computed from one of the KMC codes.

Numerical analysis [14] and control theoretic methods [15, 16] have been used to analyze the numerical stability and accuracy of various algorithms for coupling simulation codes, which has motivated the design of numerical coupling algorithms that increase the numerical accuracy of the overall simulation results by modifying the dynamic information passed between simulation codes or by introducing predictor-corrector iterations. These

analyses provide guidelines on how to best trade off numerical accuracy with computational expense when coupling simulation methods to simulate a particular multi-scale system. For example, the direct numerical simulation approach to coupling codes, which has the lowest computational expense, is restricted to first-order accuracy irrespective of the accuracy of the individual simulation codes. In contrast, a predictor-corrector algorithm can be designed to achieve second-order accuracy, but with higher computational cost.

Expressions for the numerical accuracy and computational expense have been derived for various methods for coupling individual codes to constitute an overall multiscale simulation code, so that an applications engineer can systematically select a coupling method suited to the needs for a particular application. Applications to chemical and electrochemical systems illustrate the various coupling methods and demonstrate their characteristics. One application is the simulation of the electrodeposition of copper into trenches to form interconnects in electronic devices. The simulation of trench in-fill involves the dynamic coupling of (a) multiple (2+1)D KMC codes for simulating reactions and surface diffusion at discrete positions on the surface, (b) a 2D level set method for simulating the changing position of the metal-solution interface, and (c) a 2D finite volume code for simulating the reaction-transport equations in the fluid. Another application involves simulation of the initiation of a corrosion pit on stainless steel owing to formation of a local aggressive chemical environment within a microcrevice formed between a sulfide inclusion and the steel [14]. The simulation involved the dynamical coupling of two continuum codes for (a) the microcrevice region, and (b) the 2D region outside of the microcrevice.

The guidelines for the design of dynamically coupled multiscale simulation codes are illustrated through simple reaction-transport problems which demonstrate the improved numerical stability and accuracy obtained by: (1) using finite volume codes over finite difference codes for simulating continuum domains, (2) using the underlying physical chemistry to decide the direction of the transfer of boundary condition information between individual simulation codes, and (3) introducing filters between individual simulation codes.

**References:**


[1] R. C. Alkire and R. D. Braatz, “Electrochemical Engineering in an Age of Discovery and Innovation,” AIChE J., 50, 2000-2007 (2004); R. D. Braatz, R. C. Alkire, and E. G. Seebauer, “A Multiscale Systems Approach to Microelectronic Processes,” In Proc. of the Int. Conf. on Chemical Process Control, Lake Louise, Alberta, Canada, in press; and references cited therein.

[2] U. Hansen, S. Rodgers, and K. F. Jensen, “Modeling of Metal Thin Film Growth: Linking Angstrom-scale Molecular Dynamics Results to Micron-scale Film Topographies,” Phys. Rev. B, 62, 2869-2878 (2000).

[3] D. G. Coronell, D. E. Hansen, A. F. Voter, C.-L. Liu, X.-Y. Liu, and J. D. Kress, “Molecular Dynamics-based Ion-surface Interaction Modes for Ionized Physical Vapor Deposition Feature Scale Simulations,” Appl. Phys. Lett., 73, 3860-3862 (1998).

[4] D. Maroudas and S. Shankar, “Electronic Materials Process Modeling,” J. Computer-Aided Materials Design, 3, 36-48 (1996).

[5] E. B. Tadmor, M. Ortiz, and R. Phillips, “Quasicontinuum Analysis of Defects in Solids,” Philos. Mag. A - Phys. Cond. Matter Struct. Defects Mech. Prop., 73, 1529-1563 (1996).

[6] D. Hadji, Y. Marechal, and J. Zimmerman, “Finite Element and Monte Carlo Simulation of Submicrometer Silicon n-MOSFET’s,” IEEE Trans. on Magnetics, 35, 1809-1812 (1999).

[7] M. K. Gobbert, T. P. Merchant, L. J. Borucki, and T. S. Cale, “A Multiscale Simulator for Low Pressure Chemical Vapor Deposition,” J. Electrochem. Soc., 144, 3945-3951 (1997).

[8] R. E. Rudd and J. Q. Broughton, “Concurrent Coupling of Length Scales in Solid State Systems,” Physica Status Solidi B - Basic Research, 217, 251-291 (2000).

[9] S. T. Rodgers and K. F. Jensen, “Multiscale Modeling of Chemical Vapor Deposition,” J. Appl. Phys., 83, 524-530 (1998).

[10] D. G. Vlachos, “The Role of Macrotransport Phenomena in Film Microstructure During Epitaxial Growth,” Appl. Phys. Lett., 74, 2797-2799 (1999).

[11] T. P. Merchant, M. K. Gobbert, T. S. Cale, and L. J. Borucki, “Multiple Scale Integrated Modeling of Deposition Processes,” Thin Solid Films, 365, 368-375 (2000).

[12] T. S. Cale, M. O. Bloomfield, D. F. Richards, K. E. Jansen, and M. K. Gobbert, “Integrated Multiscale Process Simulation,” Computational Materials Science, 23, 3-14 (2002).

[13] T. O. Drews, E. G. Webb, D. L. Ma, J. Alameda, R. D. Braatz, and R. C. Alkire, “Coupled Mesoscale-Continuum Simulations of Copper Electrodeposition in a Trench,” AIChE J., 50, 226-240 (2004).

[14] Y. He, J. R. Gray, R. C. Alkire, and R. D. Braatz, “Predictor-Corrector Methods for Dynamically Coupling Multiscale Simulation Codes,” In Proc. of the Topical Conf. on Coupling Theory, Molecular Simulations and Computational Chemistry to the Physical World, Paper 439b, AIChE Annual Meeting, Austin, TX, November 7-12 (2004), and citations therein.

[15] E. Rusli, T. O. Drews, and R. D. Braatz, “Systems Analysis and Design of Dynamically Coupled Multiscale Reactor Simulation Codes,” Chem. Eng. Sci., 59, 5607-5613 (2004).

[16] R. D. Braatz, R. C. Alkire, E. Seebauer, E. Rusli, R. Gunawan, T. O. Drews, and Y. He, “Perspectives on the Design and Control of Multiscale Systems,” J. of Process Control, in press.