

# PROGRESS REPORT

**DOE Award: DE-FG02-11ER26047/DE-SC0007046**

**Recipient: UNIVERSITY OF TENNESSEE**

**Project Title: Multiscale Mathematics for Biomass Conversion to Renewable Hydrogen**

**Principle Investigator: Petr Plechac**

**Summary of work done at University of Tennessee, Knoxville  
(reporting period ending: 8/31/2010)**

## **I. Research.**

### **Progress and Accomplishments**

The main focus during the period of research at UTK was on developing a mathematically rigorous and at the same time computationally flexible framework for parallelization of Kinetic Monte Carlo methods, and its implementation on multi-core architectures. Another direction of research aimed towards spatial multilevel coarse-graining methods for Monte Carlo sampling and molecular simulation. The underlying theme of both of these topics was the development of numerical methods that lead to efficient and reliable simulations supported by error analysis of involved approximation schemes for coarse observables of the simulated molecular system. The work on both of these topics resulted in publications.

### **Scientific Tasks: Accelerated Monte Carlo simulations for systems with chemistry**

**Publication 1: Hierarchical fractional-step approximations and parallel kinetic Monte Carlo Algorithms** [G. Arampatzis, M.A. Katsoulakis, P. Plechac, M. Taufer, L. Xu, preprint; arXiv: 1105.4673, submitted to J. Comp. Physics]

This paper developed and tested a new approach to simulations of large lattice systems with complex chemistry using parallel architectures. The primary focus was on demonstrating applicability on multi-core architectures and GPUs. We presented a mathematical framework for constructing and analyzing parallel algorithms for lattice Kinetic Monte Carlo (KMC) simulations. The resulting algorithms have the capacity to simulate a wide range of spatio-temporal scales in spatially distributed, non-equilibrium physiochemical processes with complex chemistry and transport micro-mechanisms. The algorithms can be tailored to specific hierarchical parallel architectures such as multi-core processors or clusters of Graphical Processing Units (GPUs). The proposed parallel algorithms are controlled-error approximations of kinetic Monte Carlo algorithms, departing from the predominant paradigm of creating parallel KMC algorithms with exactly the same master equation as the serial one.

We carried out a detailed benchmarking of the parallel KMC schemes using available exact solutions, for example, in Ising-type systems and we demonstrate the capabilities of the method to simulate complex spatially distributed reactions at very large scales on

GPUs. The approach also provides a systematic evaluation of different processor communicating schedules. We discussed work load balancing between processors and proposed a re-balancing scheme based on probabilistic mass transport methods.

This flexibility and hierarchical structure, are key advantages for tailoring our framework to particular parallel architectures with complex memory and processor hierarchies, e.g. clusters of GPUs.

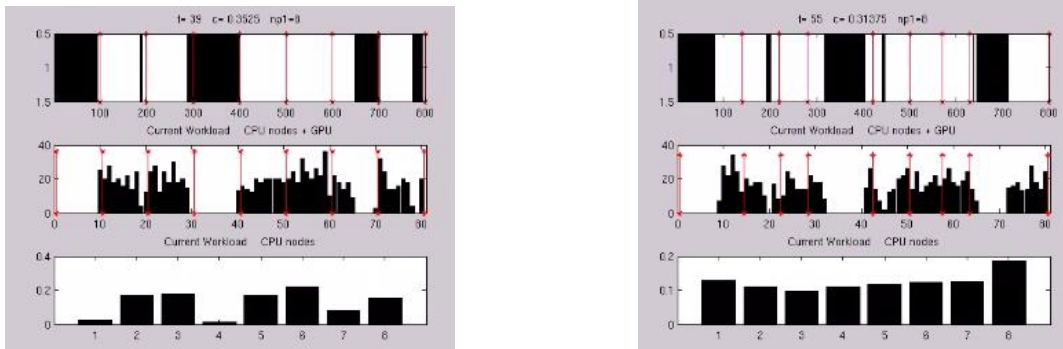


Fig. 1 Dynamic load balancing: example of the workload on sublattices in an algorithm on a 1D lattice: (a) no balancing, (b) dynamic load balancing using probabilistic mass transport framework by refining.

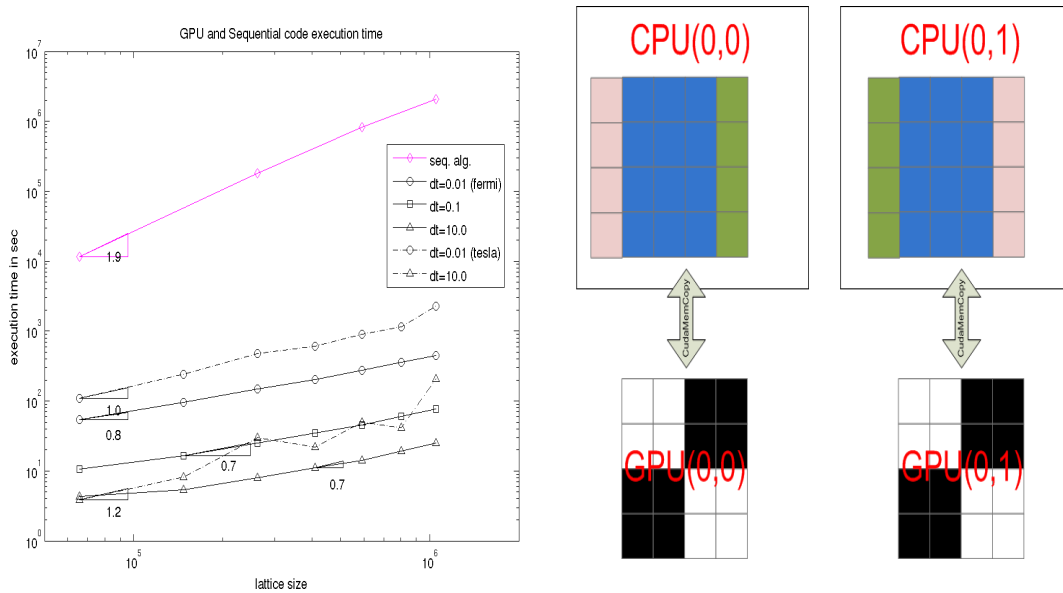


Fig. 2 (a) Efficiency evaluation on NVIDIA GPUs achieving speed-up 10000x. Capability of simulating 1000x1000 lattice models of a heterogeneous catalysis example up to a steady state on a single GPU. (b) Two GPU units and two independent sublattices (B&W) on GPUs.

**Publication 2: Coupled coarse graining and Markov Chain Monte Carlo for lattice**

systems, [[Evangelia Kalligiannaki](#), [Markos A. Katsoulakis](#), [Petr Plechac](#), [arXiv:1006.3781v1](#) (To appear in "Numerical Analysis and Multiscale Computations", in the series Lect. Notes Comput. Sci. Eng., Volume 82, to be published by Springer Verlag.)]

Here we propose an efficient Markov Chain Monte Carlo method for sampling equilibrium distributions for stochastic lattice models with complex spatial particle interactions. The proposed method is a Metropolis-type algorithm with the proposal probability transition matrix based on the coarse-grained approximating measures introduced in a series of works of M. A. Katsoulakis, A. Majda, D. Vlachos and P. Plechac, L. Rey-Bellet and D. Tsagkarogiannis,. We prove that the proposed algorithm reduces the computational cost due to energy differences and has comparable mixing properties with the classical microscopic Metropolis algorithm, controlled by the level of coarsening and a suitably defined reconstruction procedure. The properties and effectiveness of the algorithm are first demonstrated with an exactly solvable example of a one and two-dimensional Ising-type model, comparing efficiency of the single spin-flip Metropolis dynamics and the proposed coupled Metropolis algorithm. The improved efficiency is demonstrated, for example, by speed-up of constructing a phase diagram for lattice-gas simulations with long range interactions, see the CPU times in Table 1..

<b>Constructing a phase diagram</b>	<b>Traditional MC CPU time</b>	<b>Coupled CG-MCMC CPU time</b>
1-D lattice	20.6 min	5.3 min
2-D lattice	477 min	33 min

Table 1: Comparison of CPU times for the developed Coupled Coarse-Grained Markov Chain Monte Carlo method.

**Publication 3: Coarse-graining schemes for stochastic lattice systems with short and long-range interactions** [Markos A. Katsoulakis, Petr Plechac, Luc Rey-Bellet and Dimitrios K. Tsagkarogiannis, arXiv:1003.1506, submitted to Mathematics of Computation.]

In this manuscript we develop coarse-graining schemes for stochastic many-particle microscopic models with competing short- and long-range interactions on a d-dimensional lattice. First, we focus on the coarse-graining of equilibrium Gibbs states and using cluster expansions we analyze the corresponding renormalization group map. We quantify the approximation properties of the coarse-grained terms arising from different types of interactions and present a hierarchy of correction terms. We derive semi-analytical numerical schemes that are accompanied with *a posteriori* error estimates for coarse-grained lattice systems with short and long-range interactions. These semi-analytical methods are based on a multi-level decomposition of the microscopic Gibbs states which is in turn induced by the cluster expansion.

Software development:

**Developing a test suite:** Another effort was directed towards developing a set of standard

benchmarks that can be used for performance evaluation and code validation. Apart from the exactly solvable models of statistical mechanics (e.g., 2D Ising model) a more complex chemistry was tested on the Ziff-Gulari-Barshad (ZGB). The ZGB model is a simplified CO oxidation model, which was one of the first attempts towards a spatially distributed KMC modeling in reaction systems. Although a simplified model compared to the ab initio KMC, where kinetic parameters are estimated by ab initio density functional theory (DFT), it incorporates the basic mechanisms for the dynamics of adsorbate structures during CO oxidation on catalytic surfaces (adsorption/desorption, reactions). It serves as a suitable test example for our parallel kinetic Monte Carlo simulator.

## **II. Presentations and dissemination of results.**

The project maintains the website, now located at

<http://mira.math.udel.edu/ParallelKMC>

which serves as the public portal for dissemination of preprints as well as software releases. It also documents case studies done as part of the project.

The work and results of the project were presented in the meetings:

- DOE Applied Mathematics Research – PI Meeting: Berkeley, CA, Oct 2010
- "Multiscale computations", Banff, Canada, Dec 2009

## **III. Students Supported.**

During the period of the work conducted at UTK two students were supported by the grant in the form of graduate summer internship at Joint Institute for Computational Sciences (JICS): G. Arampatzis and L. Xu. Their work led to a publication reported above.

## **IV. Organizational Tasks.**

On 9/1/2010 the PI (Dr P. Plechac) moved from the University of Tennessee, Knoxville (UTK) to the University of Delaware (UD) where he accepted the position of Professor of Mathematics.