INSTITUTE OF CHEMICAL PROCESS FUNDAMENTALS OF THE CAS

November 20, 2023

Reviewer's Report on David Celný's Thesis

Development of Parallel Algorithms for Molecular Dynamics Simulation of Heterogeneous Atomistic Systems

The title of the thesis reflects in a limited way the content of the thesis. IMHO only Chapter 6 "Simulation of supersonic expansion" and Chapter 3 "Molecular simulation" comply directly with the title of the thesis. The thesis is presented as a summary of various research topics on which the author has worked during his scientific career. Moreover, the thesis is written as textbook on these research topics rather than clearly presenting author's contribution in these particular research topics. Therefore, it is rather difficult to clearly see author's scientific contribution/achievement in these research topics. Hence in the following, my reviewer's report primarily concerns simulations of supersonic expansion. A side note: Despite my criticism of the form of the thesis the thesis is written and presented to a high standard. From the text of the thesis, it is evident that the author has been able to master rather different research areas ranging from molecular thermodynamics and molecular simulations to computer science.

Supersonic expansion allows to generate water clusters which can serve as a proxy of atmospheric particles. Therefore, molecular-level insights into the supersonic expansion play an important role in atmospheric chemistry. Molecular dynamics simulations of water clusters during the supersonic expansion require μs long trajectories and a large number of water molecules employed. Moreover, the simulation systems are highly heterogeneous which in turn makes the traditional domain-decomposition parallelisation inefficient. In addition, the simulation systems evolve at adiabatic conditions which require to use highly accurate, energy preserving integration algorithms. A methodology/protocol for molecular dynamics simulations of a system within the expanding medium was developed by Klíma and Kolafa. The author efficiently parallelised Klíma-Kolafa's simulation method for GPUs and extended the approach from canonical ensemble to microcanonical ensemble. This was accompanied by implementation of the accurate, energy preserving integration algorithm. Both the parallelisation for GPUs and NVE extension are challenging, PhD-level tasks to accomplish. Chapter 6 and its results clearly

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demonstrate that these tasks were successfully and efficiently tackled by the author, which primarily justifies my recommendation for defence of the thesis.

Points for discussion:

1. Can the author provide feedback on my criticism about the form of the thesis since I might miss something?
2. Did the author test another water model, e.g., TIP4P? Generally, if it is known how simulations of water clusters depend on water models?

After thoroughly reading a relevant part of thesis corresponding to its title and despite my criticism, David Celný's thesis represents original research and I recommend the thesis for defence.

prof. Ing. Martin Lísal, DSc.

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