Doctoral Thesis Report

Thesis title:	Development of Parallel Algorithms for Molecular Dynamics Simulation of Heterogeneous Atomistic Systems
Candidate:	Ing. David Celný
Supervisor:	prof. RNDr. Jiří Kolafa CSc.
Reviewer:	Doc. RNDr. Štěpán Roučka, Ph.D., Department of Surface and Plasma Science, Faculty of Mathematics and Physics, Charles University

The thesis presents a computational study of nucleation in gas-liquid phase transitions. Three distinct approaches are applied to the following problems: 1) Model of spherical phase interface in equilibrium based on equation of state 2) Molecular dynamic simulation of cluster formation in supersonic expansion 3) Molecular dynamic simulation of metastable systems undergoing nucleation.

In all the studied problems, the author made a significant contribution to the development of state-of-the-art computational tools and consequently to our understanding of the investigated systems. This is also documented by his 7 published (and one submitted) articles in impacted journals (according to WOS) and by inclusion of his codes in the ms2 simulation code. The number of citations (62 without self-citations) confirms the relevance of the author's work for the scientific community.

Formally, the work is of high quality. All the methods and theoretical concepts are properly introduced in three introductory chapters. The results are clearly described and discussed. The language quality is good, with only a minor number of typographical or grammatical errors.

The presented work shows the ability of David Celný to carry out independent research in the field of mathematical engineering and I recommend this work for defense as a doctoral thesis.

I suggest the following questions and comments for discussion:

1) How well does the SPC/E melting point of 215 K (section 6.4.4. on page 119) agree with experimental data for water clusters? Could the possible difference of melting points explain the different cluster shapes?
2) The parameter b is defined on page 128. But the value of b in the simulations is not clear - is that calculated from rho*_spinodal as mentioned later in the text?
3) The agreement of the present results in Figure 7.13 with the data of Baidakov et al. is remarkable. The outlier point in the T* = 1 liquid data is interesting. Do you have some tentative explanation? Are you planning some further investigation of this feature?
4) Figures 7.11 and 7.13 show the deviation of the calculated pressures from the EoS predictions. The production data for T* = 1 in Figure 7.11 seems to differ from the T* = 1 (vapor) data in Figure 7.13. Please, comment / explain these differences.

5) In Figure 7.11, all the simulated data seems to converge around an upper limit of 7.5%. Is this a real effect, or some artifact of data processing and plotting?
6) In Figure 7.8, there is still a downward trend in the metastable liquid data, even for the largest systems. I understand that these differences are negligible within the standard deviation of the data. Do you think that it would be worthwhile to investigate the behavior for larger systems? Would it be computationally difficult to simulate e.g., 8000 particles?

Minor comments are listed below:

a) Page 14: "alos" should be "also"
b) Page 17: "from of the residual" should be "form of the residual"
c) Page 73: "left hand sized" should be "left hand side"
d) Page 103: Paragraph starts with "paragraph name". More appropriate name should be chosen.
e) Page 111: The cluster shape characteristic is first introduced as ratio of surface areas, but later apparently defined and calculated as ratio cross section areas (or projected areas?) this should be clarified.
f) Page 112: Instead of "−1 J/mol per a molecule" should probably be just "-1 J/mol"
g) Page 131: Formula for Delta V should contain (2a)^3 instead of (8a)^3
h) Page 136: Table 7.1 and the corresponding text on page 135 apparently contain a typo - the liquid density should be 0.6 instead of 0.06
i) Page 137, Section 7.4.2.1.: k should be the number of cells along one axis
j) Page 140: Error bars of p* correspond to simulation duration - what is the conversion factor?
k) Page 144, Badiakov should be Baidakov

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