

Doctoral school of Physics - Eötvös Loránd University (ELTE)

Second Semester report

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(Stipendium Hungaricum)

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Ph.D. Thesis title: **Modifying and parameterization of Brenner potential and its implementation in molecular dynamics simulation (LAMMPS)**

I) Research work

a. Introduction

Historically, the interior of carbon nanotubes (CNTs), particularly, single-walled carbon nanotubes (SWNTs), has not been considered to be chemically reactive [1]. Recently, however, researchers demonstrated sidewall (inner surface) chemical reactions. They believe that the inside of CNTs provides an interesting nano space for the growth of new materials [2, 3]. We aim to compare various reactions in free space and a restricted volume. The formation of benzene from six carbon atoms and six hydrogen atoms is a simple test system in this direction.

b. Molecular Dynamics and LAMMPS

Atomistic simulation research has been increasingly applied in a wide range of areas, including nanoscience and nanotechnology, especially those of an interdisciplinary nature. The heart of atomistic simulations, such as Molecular Dynamics (MD) or Monte Carlo, is force field or interatomic potential [4]. They define the interaction of atoms in a system, and the accuracy of results hinge on the choice of these potentials. So, they are one of the most dictating fundamentals that influence the accuracy of the simulations. The challenge in this purpose at the nanoscale is to have a validated model, which has been checked for calculation of an observable property. Because of uncertainty in the choice of an appropriate interatomic potential, one often must reach an agreement with experiment as a measure of the accuracy or other validated simulations. Thus, it is essential to know which interatomic potential works accurately at the simulation. Another essential factor in simulations is the environment. There are a variety of environments [4]. LAMMPS is a powerful and open source one. It provides a wide range of standard interatomic potentials which is the significant advantage of it.

Among multiple existing force fields, Long-range Carbon Bond-Order Potential (LCBOP), Lennard-Jones (LJ), Adaptive Intermolecular Reactive Empirical Bond Order (AIREBO) and Reactive Force Field (Reax) potentials represent the most common models in our study [1, 3].

They have also been applied widely to atomistic simulations on nanomaterials properties. It is essential in the implementation of interatomic potentials to find their accuracy in different working conditions. Therefore, the parameterization of different models aimed at developing and modifying potentials with better accuracy has been a recurring theme in the literature.

László et al. have already calculated the formation of chains, tubes, ribbons inside a SWCNT with their tight-binding (TB) code [5]. However, in these calculations, only carbon atoms were present. We want to continue these calculations, but using LAMMPS. Although with LAMMPS we could investigate a much larger number of atoms, including also hydrogen atoms, we need to reproduce their current TB results with LAMMPS first. In this direction, we need to carry out lots of calculations frequently, to find the optimal LAMMPS parameters by which we can reproduce the TB results. Parameters mean not only the various switches in LAMMPS commands but also the potential itself (LCBOP, AIREBO, Reax). Only if we can safely control LAMMPS, we can trust the results. In this respect, it is important that we have the TB results as a kind of reference to compare. Having that, we can step to the starting line, and we can increase the number of atoms, including hydrogen atoms, as well.

In our case, the input contains ninety carbon atoms which are made of eighteen pentagon rings inside a (14, 0) SWCNT of 2.5578 nm in length. To create the SWCNT and pentagon rings, Visual Molecular Dynamics (VMD) and ArgusLab are used as well as OpenBabel and PACKMOL to create input file in LAMMPS format. Figure 1 shows this structure.

After preparing the input file, we apply AIREBO and LCBOP pair style potentials for all C-C pairs, separately. In this configuration, the temperature is set to 500 K as a practical temperature for many real-world applications. During the simulation, in both cases, the rings deformed into a tube inside the SWCNT. Despite the fact that TB simulation form ribbon structure instead of tube [5].

According to the AIREBO results, unphysical structures such as triangles occurred while there is no evidence of obtaining triangles in the case of LCBOP (see Fig. 2). Therefore Lcbob pair potential looks to be better.

Next step is to understand how we can get a ribbon instead of the inner tube. The idea is handling the C-C interaction differently between the wall and the inner C atoms (with usual LJ potential) and between C atoms in the wall or the interior of the wall usual LCBOP or AIREBO. We hope that perhaps we will see the formation of a ribbon instead of the creation of an inner tube. Unfortunately, the combination of LCBOP with usual LJ potential did not result in a ribbon (see Fig. 3) We also tried the combination of AIREBO with usual LJ. The result was similarly bad (see Fig. 4). So, we should modify the LJ parameters, in the hope that we can reproduce the TB result. These parameters are the finite distance at which the inter-particle potential is zero position, depth of the potential well and cutoff radius.

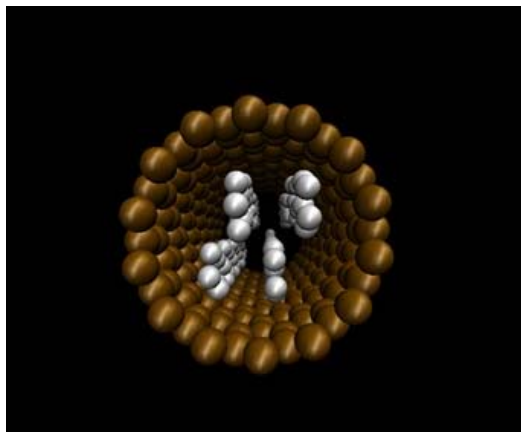


Fig 1. Initial structure. Eighteen pentagon rings which are located inside the (14,0) SWCNT.

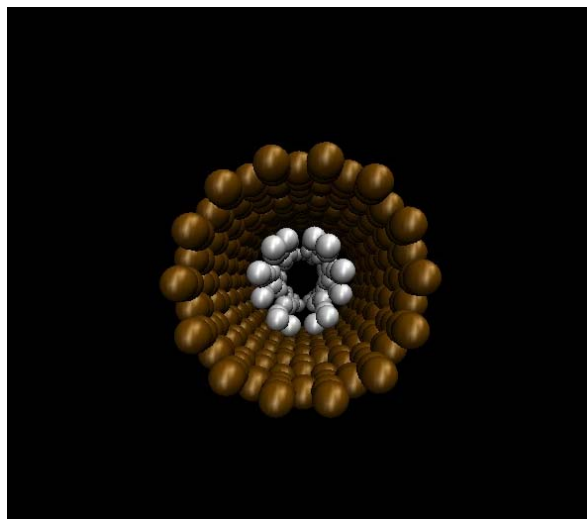


Fig 2. LCBOP for 200-time steps

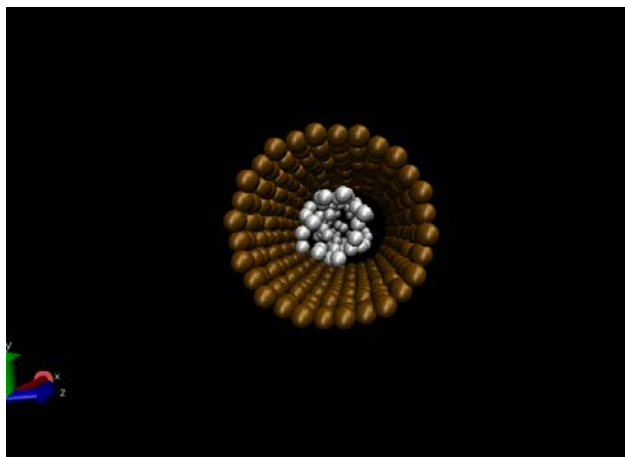


Fig 3. LCBOP with usual LJ potentials for 500000-time steps.

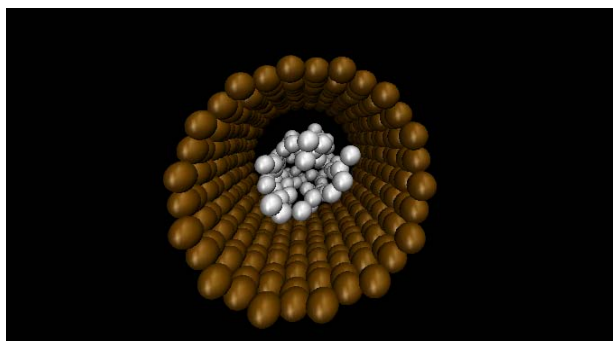


Fig 4. AIREBO with usual LJ potentials for 500000-time steps.

II) Studies in the current semester

Carbon Nanostructures (FIZ/03/015E)

Prof. Kürti Jenő,

In this course, I learned about the fullerenes such as the history of C₆₀, its symmetry, energy levels, band structures, etc. Moreover, I learned how to use the character table of icosahedral symmetry to find the vibrational modes.

Mathematical methods in quantum chemistry I (FIZ/03/034E)

Prof. Surján Péter

The goal of this course was to become familiar with the second quantization formalism in quantum chemistry. To do this, I learned how to derive the relations and equations, and to apply them to get the Hartree-Fock equation in this formalism.

References

- [1] T. W. Chamberlain, J. C. Meyer, J. Biskupek, J. Leschner, A. Santana, N. A. Besley, E. Bichoutskaia, U. Kaiser and A. N. Khlobystov, "Reactions of the inner surface of carbon nanotubes and nanoprotusion processes imaged at the atomic scale," *Nature Chemistry*, vol. 3, pp. 732-737, 2011.
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- [3] A. I. Khan, I. A. Navid, M. Noshin, H. M. A. Uddin, F. F. Hossain and S. Subrina, "Equilibrium Molecular Dynamics (MD) Simulation Study of Thermal Conductivity of Graphene Nanoribbons: A Comparative Study on MD Potentials," *Electronics*, vol. 4, pp. 1109-1124, 2015.
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