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LAMMPS Code Simulation of the Defect Formation Induced by Ion Incidence in Carbon Nanotubes

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A molecular dynamic calculation of the multi-walled carbon nanotube thermal sputtering induced by ion irradiation is carried out. Sputtering results comparable to experimental data are obtained. There are two models of ion and thermal sputtering discussed in the paper. The simulation tested the model of thermal amorphization and revealed that the disordering of multi-walled carbon nanotubes structure occurs as a result of their heating under ion irradiation. Classical molecular dynamic simulation was performed using LAMMPS code. Simulation cell with 14 layers multi-walled carbon nanotube $12 \times 12 \times 30$ nm size contains 285600 atoms. Multi-walled carbon nanotube was irradiated by 80 keV energy Ar^+ ions in cumulative mode. Simulation was performed on the Lomonosov-1 supercomputer. About 24600 nodes-hours were spent on one simulation as a whole. The balancing of MPI flows for a spatial grid of counting nodes occurred according to the scheme $8 \times 8 \times 128$ MPI-stream. LAMMPS code was built with Intel 12.0 compiler. This configuration allowed to speed up the calculation in comparison with the calculation on a single-processor Xeon CPU X5570 2.93 GHz machine by 60 times.

Keywords: ion irradiation, multiwall carbon nanotube, defects, molecular dynamics, sputtering, thermal mechanism.

Introduction

Studies related to the ion modification of carbon nanotubes (CNTs) [8] make it possible to create semiconductor devices (diodes, transistors) [14], sensors [13], filters [3], electrodes for electron emission [5] based on defective nanotubes, devices for controlling ionic beams [2, 17]. In studies on defects in carbon nanotubes [1, 7], little attention is paid to defects that appear as a result of ionic modification, in particular the defect formation mechanisms [4]. Among the defects induced by ion irradiation, vacancies, multi-vacancies and interstitials are most often encountered [9]. Defects formed upon irradiation or induced by ion irradiation from intrinsic defects of nanotubes have unique properties. Some types of defects, such as substitutional atoms, or intercalants can significantly change the electronic band structure of a nanotube by transferring it from one type of conductivity to another. Defects in CNTs are chemically active centers that bind molecules well from the environment, which makes the defective nanotube as a whole chemically more active. Transition of a part of the nanotube atoms from the state with sp^2 hybridization to a state with sp^3 hybridization increases the surface area, which affects the tribological, adsorption and capacitance properties of the nanotube. Modifications of the properties of nanotubes by chemical or plasma-chemical processing of samples, for example, for the creation of gas sensors, are what the work of collective [16] is devoted to. A directional change in the properties of carbon nanotubes by an ion beam to create elements of micro and nanoelectronic devices was carried out by Krasheninnikov, Nordlund et al. [6]. Simulation was accompanied by experiments [12]. The mechanisms of sputtering the surface of multi-walled carbon nanotubes under ion irradiation are discussed in [4, 10]. At the same time, the competition of two processes, thermal and ionic sputtering, is discussed. In this paper, the first numerical calculations of the interaction of ions with a multi-walled carbon tube are presented.

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1. Methods

Calculations of the thermal sputtering of multi-walled nanotubes were performed using the classical molecular dynamics method using the LAMMPS code [11]. Because of the large number of atoms in the simulated system, in order to save computational time, it is advisable to solve the equations of motion by the Verlete method in velocity form. The calculation was performed for a single multiwalled carbon nanotube placed in periodic boundary conditions at all boundaries in a cell with a size of $12 \times 12 \times 30$ nm.

The AIREBO potential [18] is used to describe the interaction between carbon atoms in a nanotube, which allows one to take into account the formation and breaking of new bonds, the reorganization of a network of bonds, and the change in the degree of hybridization. Potential energy obtaining by AIREBO potential consists of V^R – repulsive, V^A – attractive, and V^{LJ} – Lenard-Jones terms:

$$E_{REBO} = \sum_i \sum_{j(>i)} [V^R(r_{ij}) - b_{ij} V^A(r_{ij}) + V^{LJ}(r_{ij})], \quad (1)$$

responds to attraction of atoms at short distances. All coeficient details for repulsive, attractive parts and main potential function were provided in the work [18]. Repulsive and attractive parts of manybody AIREBO potential are used for short distance Feynman-Hellman forces reproducing in terms of classical molecular dynamics. The multi-walled nanotube consisted of 14 layers: the smallest in diameter was $CNT(10, 10)$ 1.4 nm in diameter, the largest in diameter was $CNT(75, 75)$ -10 nm, the length of each single-walled nanotube was 29.2 nm. The distance between the layers is 0.34 nm, the layers are shifted relative to each other so that they form the ABAB-packing of graphite. The total number of atoms in a multi-walled nanotube is 285600. The initial temperature was set to 0.1 K. The nanotube was not thermally tested during the passage of the ion. To reduce the radiation dose and to reduce the counting time, we used heavier Ar^+ atoms. The interaction of Ar^+C was modeled using the Ziegler-Biersack-Littmark (ZBL) potential [19]. The ZBL-potential is well tested for noble gas ions interaction with solids and used in SRIM or TRIM code.

The ion began to move randomly from a distance of 5.5 nm from the axis of the nanotube, with a random initial angle of 3–5 degrees and an energy of 80 keV. Over about 0.01 ps, the ion passed through, then the nanotube was cooled to 500 K for 5 ps, 5 ps, thermostating at a constant temperature, and then another cooling stage for 5 ps. The calculation was performed on Lomonosov-1 supercomputer [15] using nodes with Intel Xeon CPU X5570 2.93GHz (8 cores per node). Calculations were provided at 256 nodes. About 24600 nodes-hours were spent on one calculation as a whole. MPI technology was used for parallelization. The balancing of MPI flows for a spatial grid of counting nodes occurred according to the scheme of $8 \times 8 \times 128$ MPI-streams. LAMMPS code was built with Intel 12.0 compiler. This configuration allowed to speed up the calculation in comparison with the calculation on a single-processor Xeon CPU X5570 2.93 GHz machine by 60 times.

It is known that when passing through a multi-walled carbon tube, the Ar^+ ion can lose energy as a result of two processes: energy transfer to electrons – electronic energy losses, and energy transfer to lattice atoms of a multi-walled carbon nanotube – nuclear energy losses. Thus, as a result of motion, the ion either heats the lattice when interacting with electrons, or destroys it in an elastic collision. In this case, the tube is heated at 300 K. Heating is produced due to

ion-carbon atom scattering. As a result, this leads to an increase in the amplitude of vibrations of the carbon atoms, and as a result, the mobility of defects increases with increasing temperature.

Defect formation at the initial stages occurs due to the formation of collision cascades caused by the incident Ar^+ ion, and after heating up due to rebuilding the network of bonds and changing the bond order of carbon atoms. Results of simulation have shown, that with the accumulation of the irradiation dose, the number of recoil atoms also increases, and hence the defectiveness of the nanotube. Most often mono-and multivacancies are formed at the same time, while the outer layers of the nanotube are sprayed. In addition, it can be seen that the number of atomized atoms is not significant. This indicates that the main mechanism for the modification of the relief is thermal.

Conclusions

This calculation made it possible to test the model of thermal amorphization, according to which the disordering of the structure of multi-walled nanotubes occurs as a result of their heating under ion irradiation. The model did not take into account the heat sink, so from this point of view it is not quite adequate. Later the model will be further developed and based on this model, and it is planned to calculate the number of defects and predict electrophysical properties.

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