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Atomistic simulation of soldering iron filled carbon nanotubes

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ABSTRACT

The melting and soldering processes of two iron filled carbon nanotubes is explored by means of classical molecular dynamics, in order to develop an understanding of the underlying mechanisms that govern the dynamics of nano-soldering. Molten Fe flows from the open end of the two CNTs, leading to a liquid junction, and eventually to a solid contact. This soldering process is accompanied by partial or total healing of the carbon nanotubes, which after cooling and relaxation form just a single unit which encapsulates the iron, depending on the relative separation, diameters and axial offset of the nanotubes. This makes for a promising scenario for CNT soldering, repairing and healing, and a variety of different tools in the field of nanoelectronics.

1. Introduction

For some time, carbon nanotubes (CNTs) have been the focus of extensive research for physicists, chemists and applied scientists, due to their interest to basic science and because of their various potential and current technological applications. These applications encompass areas as diverse as body-armor, fibers with high tensile strength, logic circuits with CNT transistors [1], inter- and intramolecular logic gates [2], highly bendable transparent thin-film transistors that use CNT based conductors and semiconductors with elastomeric dielectrics [3], etc.

The metal catalyzed nucleation and growth mechanism of CNT has been investigated experimentally [4–6] and simulated theoretically [7,8]. Moreover, the feasibility of fabricating hetero-junctions between metals and CNTs has also been explored [9]. In particular iron-filled CNTs pose an interesting challenge from a basic viewpoint, underlined by the application they have found as magnetic force microscopy probes [10–13], and their potential in the synthesis of nanoparticles [14]. Mechanical properties of Fe filled CNTs were recently simulated for different Fe cores using quasi-static simulations [15], and in general, metal filled CNTs have aroused research interest because of their applicability as very-large-scale integration interconnects due to their high thermal stability, large thermal conductivity and current carrying capacity. Thus, the potential for the use of an Fe filled CNT as a nanoscale soldering device is a subject of special interest. The latter was recently implemented experimentally by Misra and Daraio [16]. They found that two multi-wall CNTs separated by only a few nanometers, were joined by the out-flowing Fe, molten by electron bombardment. In this contribution to metal-filled CNT research we explore, by means of classical molecular dynamics, the melting process and the soldering of two iron filled CNT, for a variety of different scenarios. Kashiwase et al., around the same time, reported soldering nanotubes using an atomic force microscope [17], but this was achieved differently. They used an iron oxide nanoparticle, instead of a nanowire, to join two or more CNTs from the outside.

Very recently a paper by Cui et al. [18] reported simulations, closely related to ours, in which a Ag nanoparticle is employed to solder two fixed axially positioned single-walled CNTs on a silicon surface. In spite of the fact that their model is quite different (they use a metal drop to solder, while we start with a metal filled CNT), it is remarkable that the system evolution and the final configurations are similar.

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2. Model

Here we report on simulation results of how two iron filled CNTs subject to a temperature cycle interact and solder. Several cases are investigated: (i) two coaxial (18,18) CNTs closed on one end and filled with (0 0 1) bcc Fe, initially less than 2 nm apart; (ii) two coaxial (18,18) CNTs closed on one end and filled with (1 1 0) bcc Fe, initially less than 2 nm apart; (iii) two non-coaxial (18,18) CNTs closed on one end, and filled with (1 1 0) bcc Fe, initially less than 2.2 nm apart; (iv) two open ended coaxial (18,18) CNTs, with their position fixed in space and filled with (1 1 0) bcc Fe; (v) two open ended non-coaxial (18,18) CNTs, with their position fixed in space and filled with (1 1 0) bcc Fe; (vi) two closed on one end and filled with (1 1 0) bcc Fe, coaxial CNTs of different diameter, that is one (18,18) and another (29,29); and (vii) two closed on one end and filled with (1 1 0) bcc Fe, non- coaxial CNTs of different diameter, that is one (18,18) and another (29,29).

We generated a capped (18,18) CNT, with a diameter of 2.37 nm, and a length of 5.40 nm along the z-axis. The CNT was filled with a single crystal cylinder of bcc Fe (diameter 1.63 nm and length 5.11 nm), oriented either along the [0 0 1] or the [1 1 0] direction along the CNT axis, and extending up to the rim of the nanotube. We duplicate this configuration to create a mirror image second CNT, either aligned or axially offset parallel to the former along the z axis, with their opposite open ends separated by a 2 nm gap as illustrated in Fig. 1a, in which the Fe is oriented with the [0 0 1] direction parallel to the CNT axis. This implies a total of 3528 carbon and 2400 iron atoms, for a total of 5928 atoms. We followed the evolution of this system under heating, to mimic the effect of an electron beam, using molecular dynamics. MD simulations were run with LAMMPS [19]. For the C–C interactions we used the AIREBO potential [20] with a cut-off of 1.0 nm and neglecting torsional contributions. A CNT simulated with this potential is stable and does not expand significantly up to temperatures well above the bulk Fe melting temperature. For the Fe–Fe interactions we used the EAM potential by Mendelev et al. [21], while the Fe–C interactions were simulated with the Lennard–Jones potential as used by Banerjee et al. [8].

There are many interaction potentials for Fe–C, starting with the classical fit by Johnson [22]. However, they are typically meant to describe C as an impurity in Fe and, therefore, greatly overestimate the binding between Fe and the C of the CNT wall. The recent work by Lee [23] using MEAM potentials might help with this problem. However, in this paper we chose the simple potential by Banerjee et al. [8], since it provides appropriate binding energies for Fe inside the NT, and is computationally very efficient.

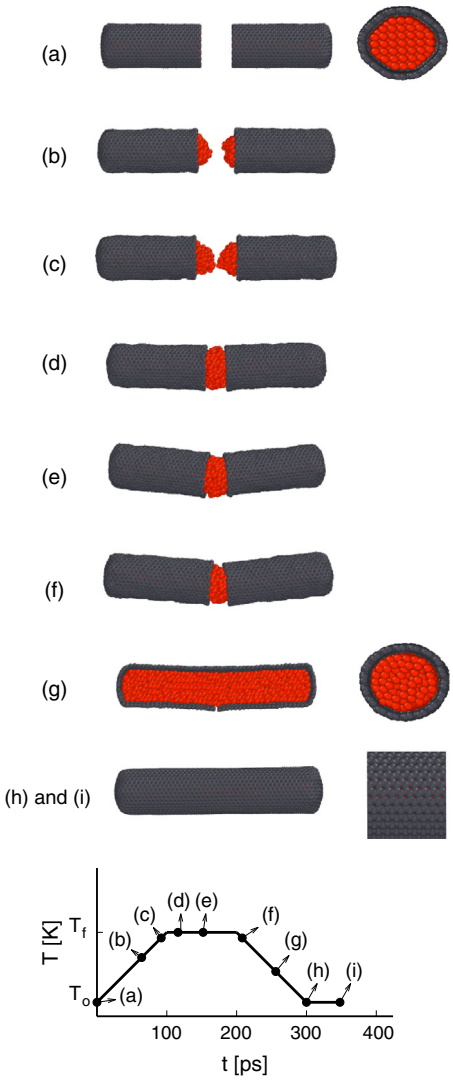

Fig. 1. Snapshots of the soldering process of two iron filled (18,18) CNTs. Initially ($t = 0$) the Fe [0 0 1] direction is parallel to the CNT axis, as illustrated by the frontal view. Initial temperature $T_0 = 10$ K, maximum temperature $T_f = 2000$ K, and temperature ramp rise time $t_r = 100$ ps. The temperature and simulation time of each of the snapshots is given by the ramp displayed at the bottom of the figure. Some images are sliced to show the inner Fe structure. The images to the right in (a) and (g) correspond to cuts perpendicular to the CNT axis. The image to the right of (h) and (i) zooms in on the region where the CNTs have coalesced, and reveals the completion of the healing process. The system consists of 3528 Fe (red) and 2400 C (gray) atoms. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

3. Results

In order to mimic the heating by an electron beam, which was rastered during the experiment, we applied a temperature ramp to the whole system, as is usually done in ion-induced thermal spike simulations [24]. As illustrated at the bottom of Fig. 1 the temperature was increased from T_0 = 10 K (Fig. 1a) to $T_f = 2000$ K, using a linear ramp during $t_r = 100$ ps, in time steps of 1 fs. Next the temperature was kept at T_f during a time t_r, and then lowered back to T_0 following a linear temperature ramp during an additional time t_r. Finally, T was kept at 10 K during $t_r/2$. We also varied T_f from 1500 to 3000 K, and t_r was varied from 10 ps to 10 ns; the final results turned out to be insensitive to all these changes of the molecular dynamics simulations. The Fe melting temperature T_f, as computed for the Fe potential used from phase coexistence simulations [25,26], turns out to be larger than the bulk Fe melting temperature (1772 K). As a matter of fact the precise determination of the melting T constitutes a challenging problem. Several different methods to calculate T_f, other than phase coexistence, have been reported, like the one put forward by Weingarten et al. [27], which focuses on the hysteresis of the melting-solidification process, and a potential energy versus T based procedure by Cui et al. [18]. However, for our investigation, the precise determination of the absolute value of T_f is not the main objective, since the actual melting point for nanoparticles in atomistic simulations strongly depends on heating rate, system size, etc. [25]. Nevertheless, our "melting temperature" gives a reasonable estimate for the threshold temperature at which the Fe nanowire starts flowing out of the CNT.

Liquid flow is observed towards the open ends of the CNTs (Fig. 1b and c) and a contact is established between both ends of the CNTs, mediated by the liquid Fe, as shown in Figs. 1d and e. In Fig. 1f a contact between the two CNTs is established,