XENON RELEASE UPON THERMAL ANNEALING OF NANODIAMONDS: A MOLECULAR DYNAMICS STUDY

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Introduction: Nanodiamonds are one of the most important and abundant grains in primitive chondritic meteorites. They contain trace xenon and other noble gases which are isotopically anomalous, indicating that the nanodiamonds are pre-solar. Stepwise heating of the nanodiamonds reveals that the xenon release is bimodal, with major peaks around 500 and 1400 °C. The low-temperature component (Xe-P3) resembles terrestrial abundances, while the high-temperature component (Xe-HL) is enriched in both heavy and light isotopes. It is generally accepted that the origin of the heavy and light isotopes are the *p*- and *r*-processes of nucleosynthesis in type-II supernova. One of the key questions is how the noble gases became incorporated into the nanodiamonds. Ion-implantation – the most popular explanation for trapping noble gases in nanodiamonds – was examined by Verchovsky et al. [1] and Koscheev et al. [2] who implanted low energy ions into ultra-dispersed synthetic nanodiamonds (UDD). Although only a single isotope was implanted, both sets of experiments found a bimodal thermal release pattern for xenon, argon and krypton. To interpret these experiments and explain the structural origin of the bimodal distribution, we performed Molecular Dynamics simulation of xenon implantation and thermal release processes. To the best of our knowledge, these are the first atomistic simulations of this process.

Modelling: A single xenon atom was implanted into a 3.9 nm diameter nanodiamond using the Molecular Dynamics (MD) approach. This size is similar to UDD and meteoritic nanodiamonds. Implantation energies of 100 to 800 eV were used, and subsequently the nanodiamonds were heated for up to 1 ns to determine the temperature at which xenon release occurs. Multiple implantation directions were used to collect meaningful statistics. Since the MD timescale is very much shorter than the experimental equivalent, temperature-acceleration was used. To calibrate the MD temperatures, an Arrhenius approach [3] was employed, using the graphitization of nanodiamonds into carbon onions as a reference point. Carbon-carbon interactions were described using Environment Dependent Interaction Potential (EDIP) and xenon-carbon interactions were described using a Lennard-Jones potential. Close interactions between all atom types were described using the standard Ziegler-Biersack-Littmark potential. These combinations have been successfully employed to model radiation damage cascades in graphite [4] and diamond [5] as well as destruction of nanodiamonds by xenon bombardment [6].

Results: To calibrate the Arrhenius relationship, the unimplanted nanodiamond was heated for 1 ns at a range of temperatures. At about 3250 K a pure carbon onion was formed, a transformation which occurs at ~1500 °C in laboratory experiments. Based on this equivalence, an activation energy of 9.5 eV was inferred. For all subsequent analysis, this value was used to convert simulation temperatures into their experimental equivalent.

In the next phase a large number of initial configuration (more than 220) were constructed by implanting a xenon atom with various directions and energies into the nanodiamond. For low implantation energies the xenon was located just below the surface, while for higher energies the xenon was implanted deep into the central region.

Annealing the nanodiamond at many different temperatures revealed that a xenon within ~3 Å of the surface could be released at around 930 K, corresponding to an experimental value of 480 °C. In contrast, a xenon located between 1 and 12 Å from the surface could be released at 3080 K, corresponding to an experimental value of 1450 °C. At other temperatures xenon release was relatively unlikely. This result almost exactly reproduces the experimentally observed bimodal distribution, giving confidence that the simulation approach is correct.

The simulations reveal that the high-temperature peak is associated with a layer-by-layer graphitization of the nanodiamond which provides an escape path by which the xenon can leave the nanodiamond. However, if the xenon is too deeply buried then the xenon cannot escape, regardless of the temperature. Even at 7 Å below the surface, there is only a 50% chance that the xenon will be released. In contrast, the low-temperature peak involves a completely different pathway in which the nanodiamond structure is retained, and only a few near-surface atoms rearrange to release the xenon. The simulations also reveal differences between the 100 and 111 faces of the nanodiamonds. The latter graphitize more easily, sometimes trapping the xenon and increasing the release temperature.

References:

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