

corrigendum

Isotope Substitution Extends the Lifetime of Organic Molecules in Transmission Electron Microscopy

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In the original paper, the time step used in the molecular dynamics simulations is too large to ensure conservation of energy; the combination of high values of kinetic energy transferred to atoms under the electron beam (e-beam) and the very low mass of the hydrogen atom leads to excessively large atomic velocities of these atoms. The use of a 1 fs time step introduces some unphysical artefacts in the simulations with the overall result of artificially lowering the calculated threshold energies. A time step of 0.5 fs is, however, entirely sufficient to reproduce the physically correct behaviour (converged to 0.1 fs).

The simulations, corrected for a smaller time step, differ in one key way compared to the reported results. Contrary to the behaviour shown in Figure 3, there is no hydrogen atom emission following multiple bond stretches. The maximum bond length in each subsequent bond stretch decreases, hence the impact corresponds to one that lies below the ejection threshold if a bond begins to contract. This results in a relatively large increase in the value of the ejection threshold energy for hydrogen, from 6.1 eV (as reported in the paper) to 7.7 eV (obtained with the reduced time step). With reference to labelling used in Figure 2b, there is also significant difference between the positions of H_a and H_b , as there is for D_a and D_b . H_a and D_a can be taken as the atoms that are most vulnerable to atom emission due to impacts from the e-beam. The increased mass of deuterium is almost sufficient to render the 1 fs time step suitable; exactly the same qualitative dynamic behaviour is seen at 0.5 fs, although the threshold energy increases slightly from 7.8 eV to 7.9 eV. The threshold energy for removal of deuterium is therefore still larger than that for hydrogen emission, but the magnitude of this difference is much smaller (0.2 eV instead of 1.7 eV). The 1 fs time step is suitable for the simulations involving impacts on the much heavier carbon atoms.

Figure 1 shows the effect of these changes in calculated threshold energy on the cross-sections of hydrogen emission. Hydrogen emission is not quite as accessible to the e-beam as previously predicted, slightly reducing the expected stability increase provided by deuteration. However, deuteration is still expected to supply significant protection to organic molecules in TEM, as the major component of this stabilisation was not the difference in threshold energy but the halving of the transferable energy from the e-beam to deuterium (94 eV at 80 kV) compared to hydrogen (188 eV at 80 kV). In fact, as this effect was previously overestimated compared to our experimental results, this correction brings our theoretical predictions into line with the experimental measurements (2.2x theoretical increase in stability instead of 2.8x originally reported, compared to the approximately 1.8x increase of the experimental lifetimes in the nanotube and the 2.4x increase in the crystal structures).

Finally, a minor typographical correction to the end of page 2: the ∇ symbol in the text should of course be \hbar , referring to the reduced Planck constant.

Many thanks to Dr. Jani Kotakoski for bringing this issue to our attention.

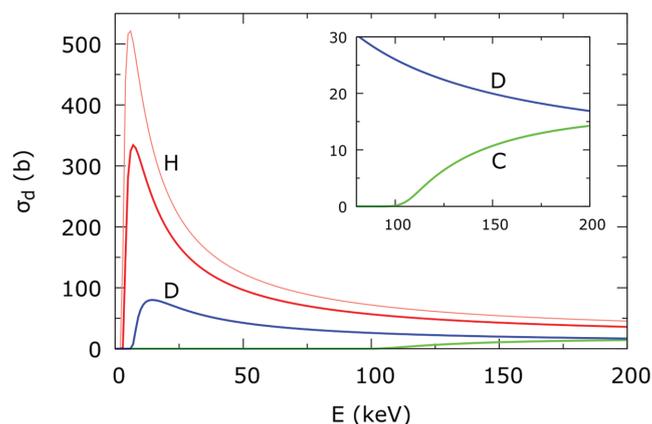


Figure 1. Ejection cross-sections σ_d as a function of electron beam kinetic energy E for protium (1 fs time step, pink line; 0.5 fs time step, red line), deuterium (blue line) and carbon (green line) calculated for the $C-H_a$ and $C-D_a$ bonds in coronene and deuterated coronene.