

Fusion reaction dynamics of fullerene molecules

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Synopsis Systematic molecular dynamics simulations based on the self-consistent charge density functional tight-binding method have been performed for the molecular collisions inside clusters of fullerene molecules, after prompt atom knockouts by the keV ions. The thermodynamic and kinetic stabilities, as well as the bonding features, have been investigated for the most stable dumbbell dimers C_{118}^+ , C_{119}^+ and C_{120}^+ .

During the last decades, experiments have shown that fullerene fusion reactions can take place between C_{60} molecules and C_{58} or C_{59} radicals that are produced upon the fragmentation of C_{60} induced by laser irradiation, photoionization and heating, or ozone oxidation. In recent experiments [1, 2], covalently bound fullerene dimers C_{118}^+ and C_{119}^+ were observed in mass spectra resulting from collisions between clusters of C_{60} molecules and keV He^{2+} - or Ar^{2+} -ions. In this contribution, we present a systematic theoretical investigation [3] on the dynamics of $C_n^+ + C_{60}$ ($n = 58, 59, 60$) collisions following prompt atom knockouts by the fast ions. This study will not only provide additional information to, and a deeper understanding of, the experimental results, but may shed some light on the general mechanisms of fullerene coalescence involving C_{58} or C_{59} fragments reacting with C_{60} . It might also be relevant to the formation and growth of fullerenes.

A large number of molecular dynamics (MD) simulations based on the self-consistent charge density functional tight-binding (SCC-DFTB) method have been carried out, with different initial orientations of fullerene cages and different center-of-mass collision energies. The DFTB method implemented here is based on the second-order expansion of the Kohn-Sham energy in terms of the charge density fluctuation. The re-

sulting energy terms are calculated by applying the tight-binding approximation. The electronic and repulsive parts of the DFTB energy are computed using a set of empirical Slater-Koster interatomic parameters.

From the results of the simulations, we verify the feasibility of forming covalently bound fullerene dimers C_{118}^+ and C_{119}^+ under the experimental conditions, and confirm that the so-formed dimers are stable enough to survive on the experimental timescale. The statistics from the MD simulations indicate a much higher reactivity of C_{59}^+ and C_{58}^+ fragments compared to that of C_{60}^+ . The formation of C_{119}^+ and C_{118}^+ dimers is almost barrierless and the MD simulations demonstrate that these dimers can be readily produced in less than 1 ps at collision energies below 1 eV. This is consistent with the experimental findings. Furthermore, we show that, the dumbbell dimer products C_{119}^+ and C_{118}^+ are both thermodynamically and kinetically stable so that they can survive to be detected on the experimental timescale of tens of microseconds.

References

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