Highly Energetic Collisions of Xe with Fullerene Clusters

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Outline

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Introduction

- **Fullerene** - The first fullerene($C_{60}$) molecule was synthesized by R. E. Smalley et al. in 1985 by vaporizing graphite using the technique laser irradiation.

- **Carbon nanotubes,**
  Generated in an arc-discharge fullerene reactor by S. Iijima in 1991

- Production of macroscopic quantities of $C_{60}$ by Krätschmer et al. in 1990

Conductivity, Stability, Compatibility
Motivation

Zettergren et al. observed a large number of $C_n (n>60)$ since 2004 by colliding highly energetic inert gases with weakly bound fullerene clusters.

Coalescence products from $C_{60} - C_{300}$ observed in mass spectra in 1992.

Becker, 31st Lunar Plan. Sci. Conf. 1000 1803 ‘00
Experimental studies

high collision energies in the range 10 keV - 400 keV


Higher order oligomers, high density: \((C_{60})_{37}\)

Trimers are probable than dimers with high intensities

Highest intensity at tetramer from low density collisions and hexamer from high density collisions
Theoretical Studies

- Highly energetic collisions

Classical Reactive Dynamics (22.5 keV)

Evidence of a formation of a $\text{C}_{119}$ peanut shaped molecule, only few ps
No statistical analysis of mass distribution patterns
No collisions in 200 keV – 400 keV

Objective

- Compare the mass distribution of highly energetic collisions and understand the post-collisional dynamics of the products formed
Method

Classical Molecular Dynamics

Adaptive Interatomic Reactive Empirical Bond Order (AIREBO)

\[ E_{AIREBO} = E_{REBO} + E_{LJ} + E_{Torsion} \]

\[ V_{ij}^{REBO} = V_{ij}^R + b_{ij} V_{ij}^A \]

Variable Time-step Algorithm

\[ \frac{\sigma_{\Delta E}^2(\Delta t)}{\Delta t} \leq \frac{\sigma_{\Delta E}^2(t)}{t} \equiv D_E \]

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\]

During the ballistic phase before the collision, relatively large time-steps of 0.1-1 fs

Early stages of the collision: $10^{-8} - 10^{-12}$ fs, Later: $10^{-2} - 10^{-4}$ fs

The variable time-step integrator allows to use small time-steps during the collision and to use larger time-steps at other times to reduce the computer cost.

Computational Implementation

The AIREBO potential is capable of modeling carbon reactions with less computational cost than density functional theory and density functional tight binding methods, but still quite demanding at the ns timescale.

The simulations were performed in parallel.

A MPI program was used to distribute the independent collisions across multiple nodes of the computing resource.

The individual molecular dynamics simulations, were restricted to a single processor.

Most molecular dynamics codes, including the AIREBO code used to perform these simulations, do not scale well to fewer than 500 atoms per processor.
Highly energetic Xe Collisions

System and Simulation Details

$(C_{60})_{55}$ fullerene cluster collisions with 200 keV and 400 keV Xe atom

400 keV – 300 collisions
200 keV – 50 collisions

Classical Molecular Dynamics(AIREBO)

impact parameter( b) between
0-26 Å.

Computer cost:
200 keV - 10 node-hours for first 100 ps
400 keV – 40 node-hours for first 100 ps
7 node hours – for 1 ns (post collision dynamics)
200 keV Xe collision with a fullerene cluster
Results

Mass distribution of the carbon products after 200 ps

Probability of $C_n$ and $(C_{60})_m$ decreased with size

400 keV - $(C_{60})_m$ oligomers were observed at $m=2,3,4$ and 12

200 keV - $(C_{60})_m$ oligomers were observed at $m=2 – 12$

Most favorable oligomer is the dimer

Only 3 tetramers from 400 keV, while 12 tetramers from 200 keV

Decay is high in 400 keV
Results (200 keV)

- Preference for $\text{C}_{60+k}$, $\text{C}_{120+k}$ products after 200 ps
- Preference for $\text{C}_{60-k}$, $\text{C}_{120-k}$ products after 1 ns
- Series of even products at $\text{C}_{60}$ experimentally
- No preference for even numbers theoretically, but comparable with series of peaks observed by 12 keV collisions
- Preference for even products at $\text{C}_{180}$ after 1 ns

Results

Post collision dynamics

Dimer

Trimer

Amorphous product and Branched trimer
Conclusions

200 keV collisions is in reasonable agreement with the experimental mass spectral results, where we observed the peaks of fullerene oligomers up to $(C_{60})_{12}$

preference for even sized carbon molecules after 1 ns in the region ($C_{150}$ - $C_{210}$) and more peaks appeared at $C_{60-k}$ and $C_{120-k}$ positions after 1 ns than after 200 ps

Post-collision dynamics and the structures were comparable with “Shrinking of hot giant fullerene” mechanism proposed by Irle et al. in 2006
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Thank you

Questions?