

From Chains to Rings: Impulse Driven Molecular Growth in C₄H₆ Clusters

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Synopsis We have collided keV atomic ions with neutral clusters of 1,3-butadiene (C₄H₆). We detect efficient molecular growth driven by nuclear scattering induced fragmentation within the clusters, possibly leading to the formation of aromatic rings.

Energetic ions colliding with loosely bound molecular clusters can lead to the growth of new and larger molecular species, even in clusters consisting of very stable and unreactive molecules like Polycyclic Aromatic Hydrocarbons (PAHs) [1, 2] or C₆₀ fullerenes [1, 3]. There the key mechanism is the prompt knockout of atoms from molecules in the cluster by the impacting keV projectile [4, 5]. This produces radical species that can efficiently form covalent bonds with neighboring molecules with little or no reaction barriers [1, 2, 3].

Here we present results for keV ions colliding with loosely bound clusters of 1,3-butadiene (C₄H₆). An experimental mass spectrum from collisions between 3 keV Ar⁺ ions and clusters of butadiene molecules is shown in Fig. 1. We see clear evidence of molecular growth and the formation of new larger molecules with up to at least nine C atoms. Compared to the broad range of growth products observed with PAH clusters [2] we see here that certain specific products C_{n>4}H_x are much more abundant than others. In particular, the peaks corresponding to C₅H₇⁺ and C₆H₇⁺ stand out, indicating that they are “magic” and more stable than other similarly sized species.

We will also present results from our classical molecular dynamics (MD) simulations of the same Ar⁺ + [C₄H₆]_k collisions together with Density Functional Theory (DFT) calculations of reaction product isomers. The classical MD simulations show that the knockout process can indeed initiate growth inside the butadiene clusters similar to what has been observed in PAH [2] and fullerene [3] clusters. Our DFT calculations suggest that many of the reactions products seen in our mass spectrum (see Fig. 1) contain contributions from cyclic structures. This type of molecular processing may be important for the formation and growth of large aromatic molecules, like for example PAHs and Fullerenes in the interstellar medium [6].

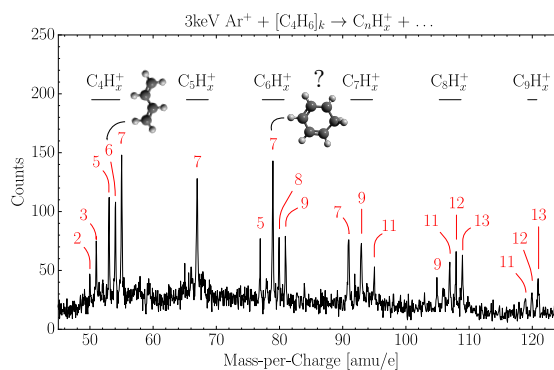
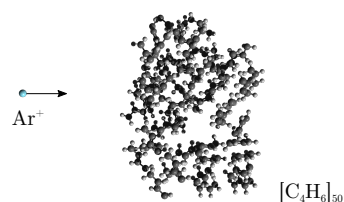


Figure 1. Experimental mass spectrum for 3 keV Ar⁺ ions colliding with weakly bound clusters of C₄H₆. The labels above each group of peaks indicate their molecular formula C_nH_x⁺. The numbers in red indicate the numbers of H atoms for n = 4–9 carbon atoms in each product. The insets show the structures of intact C₄H₆⁺ and the most stable form of C₆H₇⁺, protonated benzene.

References

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