



Transformation of Doped Graphite into Cluster-Encapsulated Fullerene Cages

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Introduction

An ultimate goal in carbon nanoscience is to decipher formation mechanisms of highly ordered systems. Carbon cages, known as fullerenes, that encapsulate clusters of atoms (e.g., $M_3N@I_h-C_{80}$) are nanomaterials attractive for use as safe MRI contrast agents and other biomedical diagnostics, renewable energy technologies, and molecular electronics. However, it is entirely unknown how they are built on the molecular level because the reactions that result in their self-assembly are very fast and take place at very high temperature (in a plasma).

Experimental

Ion Cyclotron Resonance user facility's cluster instrument – 9.4 T active magnet

Results and Discussion

Graphite doped with metal and nitrogen was vaporized by means of a high energy laser to synthesize 80 carbon-atom-containing fullerene cages that encapsulate a M_3N (M = metal) cluster through ultrafast self-assembly processes in the gas phase. Nanocarbon reactions involved in the transformation of graphite into spherical cages were chemically sampled and tracked by the MagLab's powerful FT-ICR mass spectrometer. It was discovered that these technologically important nanocages are formed through a distinct 'bottom-up' mechanism. Further, specific structures such as the small, fused pentagon-containing species, $Sc_3N@C_{68}$, were also probed under those same formation conditions to further elucidate new chemical transformations.

Conclusions

This work discloses intrinsic chemical processes that are a fundamental property of carbon under the 'harsh' conditions typical of synthesis and will help tackle the challenge of nanocarbon structure formation for unique caged and hybrid carbon materials. Moreover, this work may be used as a benchmark to guide future nanocarbon explorations.

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References

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