SUPPORTING INFORMATION

Probing Distinct Fullerene Formation Processes from Carbon Precursors of Different Sizes and Structures

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Experimental Section

Atomic Force Microscopy (AFM) and Field Emission Scanning Electron Microscopy (FE-SEM). For analysis of multi-layered graphene (MLG), MLG samples dispersed in a solution of water and 2-propanol (50:50, v/v) were deposited onto a silicon wafer. AFM experiments were performed at the National Institute for Nanomaterials Technology (NIST; Pohang, Korea) with a VEECO Dimension 3100 atomic force microscope. FE-SEM experiments were performed at NIST using a JEOL JSM-7401F scanning electron microscope.

UV/Vis spectroscopy. The solutions of triphenylene (Tri) and C_{60} were prepared by dissolving them in toluene. Then, the UV/Vis absorption of prepared solutions were analyzed using Shimadzu UV-1800 (Shimadzu, Japan) spectrophotometer.

Discussion

Penetration depth of 355 nm laser in multi-walled carbon nanotubes.

The intensity of the 355 nm laser as a function of the depth of graphite is estimated by equation (1).

$$\frac{I}{I_0} = e^{-\alpha z} \qquad (1)$$

where $\alpha = \frac{4\pi k}{\lambda}$, λ=355 nm, and k=1.3, which is the extinction coefficient of graphite[1]. The z values at $\frac{I}{I_0} = \frac{1}{e}$ is defined as the penetration depth of laser. This equation was previously shown to provide good estimate of laser penetration depth in graphite, similar to the experimental values[2]. Based on equation (1), the penetration depth of the 355 nm laser through graphite was calculated to be ~22 nm. Because the interval length between the layers in multi-walled carbon nanotubes (MWNT) (0.34 nm) is same as graphite, we assumed that the extinction coefficient of MWNT is equal to that of graphite.

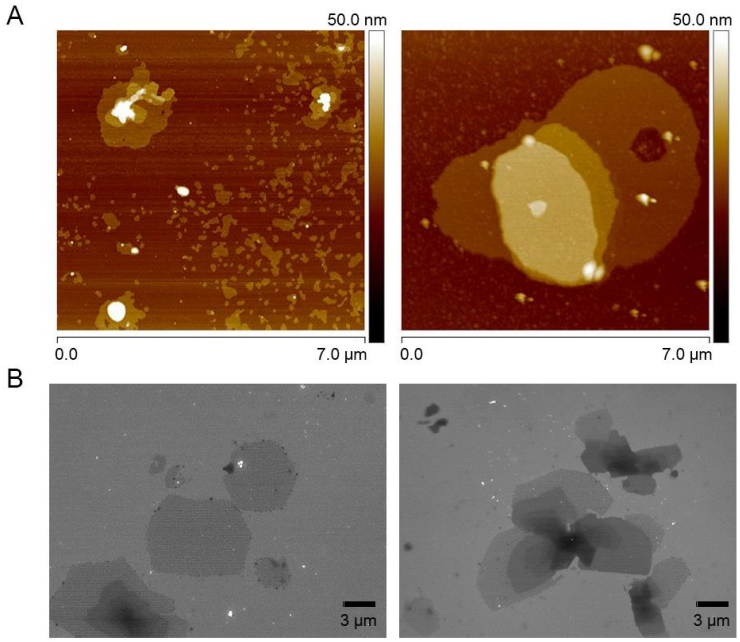

Figure S1. (A) AFM images, and (B) FE-SEM images of MLG used in this study. Thickness of each MLG sheet was ~5 nm, indicating that it is multi-layered. The lateral dimensions were in the order of micrometers.

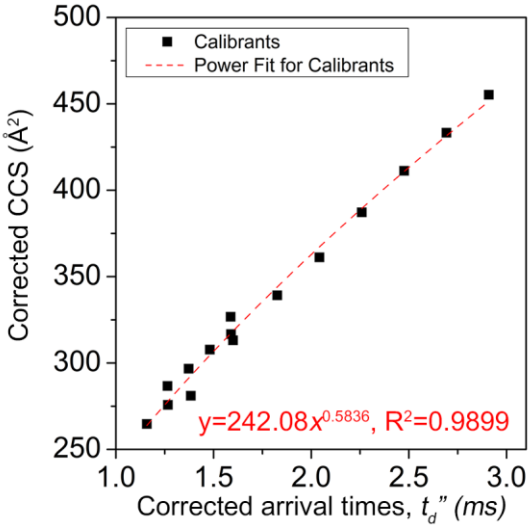

Figure S2. Collision cross-section (CCS) calibration curve obtained using poly-DL-alanine as calibrants.

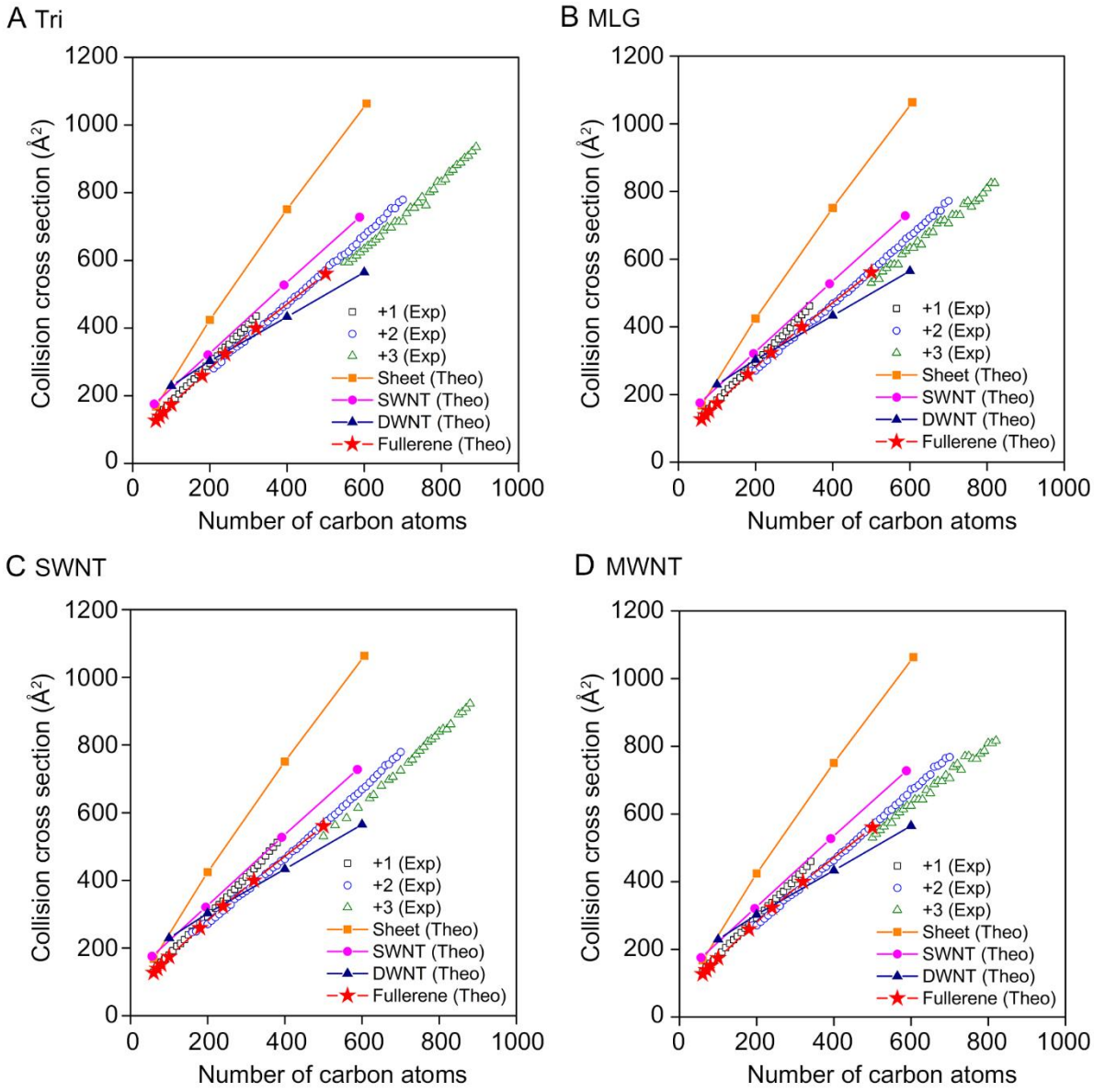

Figure S3. Correlation between the number of carbon atoms and collision cross-section (CCS) areas of carbon clusters from (A) triphenylene (Tri), (B) multi-layered graphene (MLG), (C) single-walled carbon nanotube (SWNT), and (D) multi-walled carbon nanotube (MWNT). The peaks in the spectra which have ratios, S/N>3, are considered. Experimental CCS values of carbon clusters were compared with the theoretical CCS values of various models of +1 charge state. Detailed description of the models is provided in Figure 2B.

Table S1. Theoretical CCS of the structural models presented in Figure 2. The CCS values ($Å^2$) of the models were obtained for +1 and +2 charge state. The models with +2 charge show slightly increased theoretical CCS values. Due to insignificant difference between the values, we adopt the theoretical CCS values of the +1 charge state for plotting Figures 2 and S3.

MODEL		+1	+2
Sheet	C_{60}	167.64	170.72
	C_{200}	424.33	425.21
	C_{400}	751.07	752.28
	C_{600}	1063.40	1063.80
SWNT	C_{56}	175.60	178.29
	C_{195}	320.81	322.46
	C_{392}	527.21	528.76
	C_{588}	727.77	728.46
DWNT	C_{100}	229.59	232.23
	C_{200}	302.57	304.96
	C_{400}	433.36	434.54
	C_{600}	564.99	566.00
Fullerene	C_{60}	126.73	132.00
	C_{70}	138.76	144.33
	C_{80}	151.33	156.74
	C_{100}	174.13	172.79
	C_{180}	259.15	266.27
	C_{240}	323.33	320.79
	C_{320}	399.70	400.58
	C_{500}	560.61	561.52

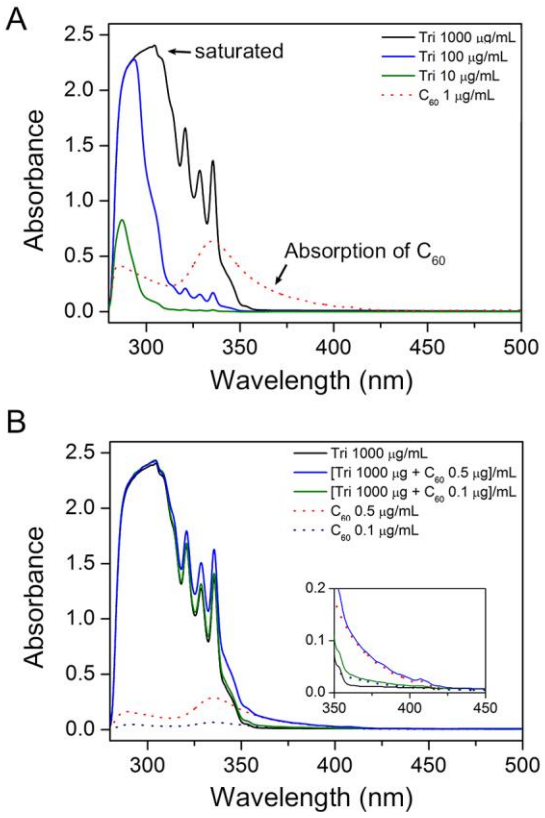

Figure S4. UV/Vis absorption spectra of triphenylene (Tri) precursor, C_{60} fullerene, and [Tri+C_{60}] mixture for the examination of possible innate C_{60} constituents in Tri precursors. (A) The absorption spectra of Tri precursor and C_{60}. C_{60} has absorption at the wavelength range of 370-420 nm as previously reported by other groups,[3,4] whereas Tri precursor shows no absorption at this range. (B) The comparison of absorption spectra of Tri, C_{60}, and [Tri+C_{60}] mixture. The same absorbance of C_{60} and [Tri+C_{60}] at the wavelength range of 370-420 nm indicates that no C_{60} is included in the original Tri precursor.

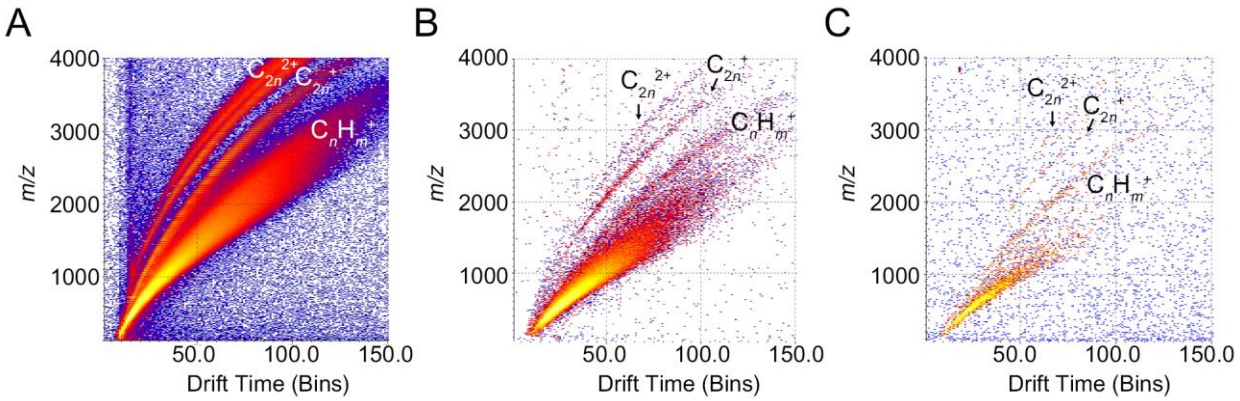

Figure S5. IM-MS spectra of ions generated from (A) triphenylene (Tri), (B) anthracene (Ath), and (C) phenanthrene (Phn). These IM-MS spectra show high abundance of hydrocarbons ($C_nH_m^+$) and traces of carbon clusters (C_{2n}^+ and C_{2n}^{2+}).

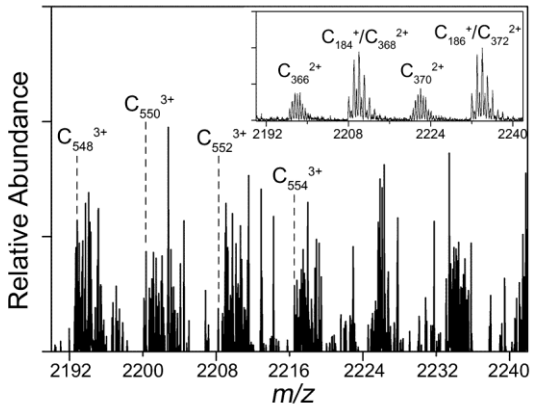

Figure S6. Mobility-separated mass spectrum of +3 charged carbon clusters from LDI mass spectrum of multi-layer graphene (MLG). Inset shows the original mass spectrum over identical *m/z* range.

References

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