Supporting Information

Transformation of Nanodiamonds to Onion-like Carbons by Ambient Electrospray Deposition

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KEYWORDS: nanodiamond, onion-like carbon, ambient electrospray deposition, graphitization, sonic spray

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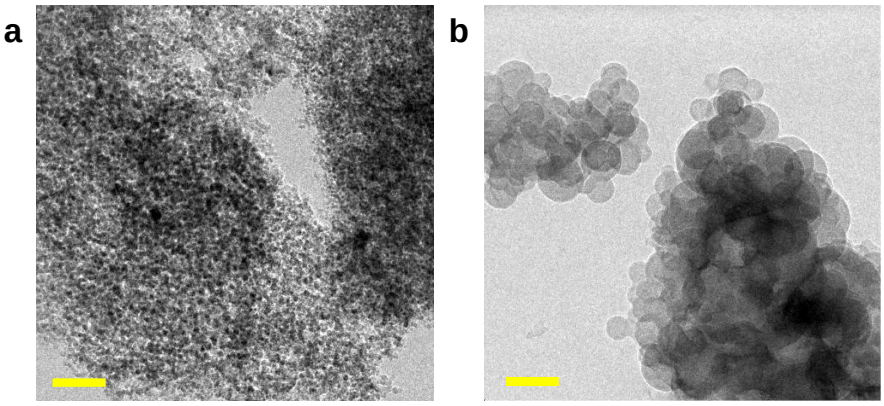

Figure S1. Large area TEM images of ND (1.25 mM aqueous suspension) (a) and OLCs (b) showing an increment in the size of ND upon transformation to OLCs after AESD at 2.6 kV. The scale bars in (a, b) are 100 nm.

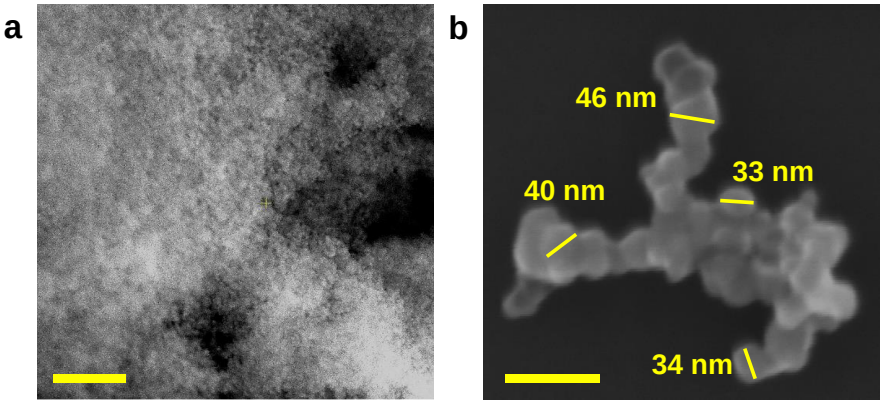

Figure S2. FESEM images of ND (1.25 mM aqueous suspension) (a) and OLCs (b). Size of OLCs varied from 33–46 nm. The scale bars in (a, b) are 100 nm.

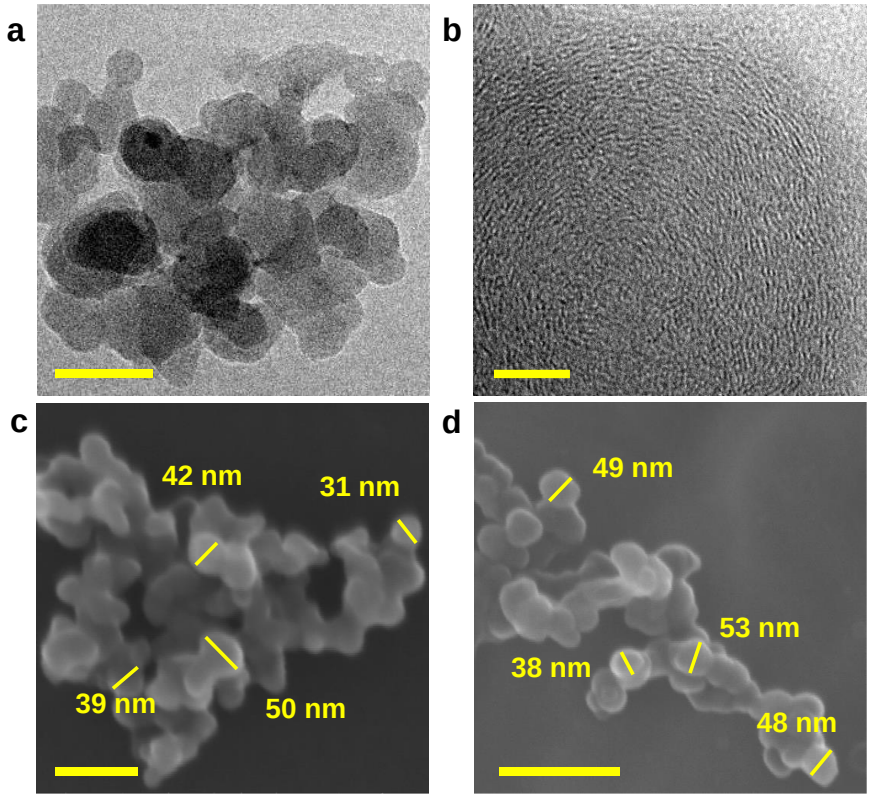

Figure S3. TEM images (a) HRTEM image (b), and FESEM images (c, d) of OLCs obtained after ES of 5 mM ND suspension in water. The scale bars in a, b, c, and d are 50, 5, 100, and 200 nm.

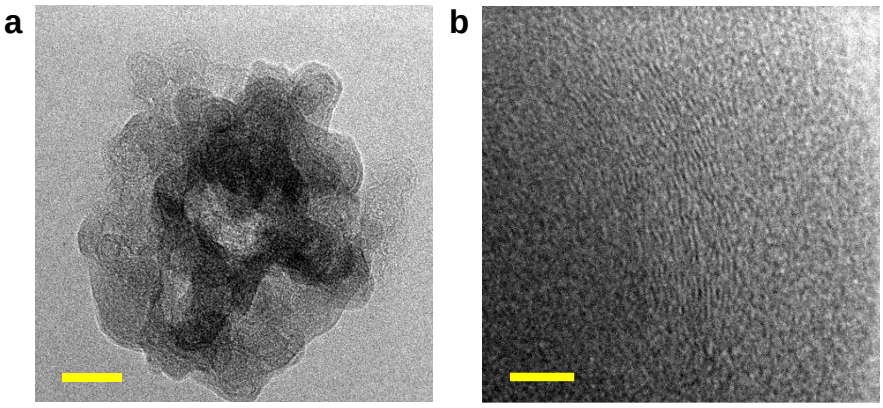

Figure S4. TEM images (a, b) of the product obtained after direct ES on ITO glass slide. The product was scratched from ITO, dissolved in DI water, and drop-casted on a TEM grid. The scale bars in (a) and (b) are 20 and 5 nm, respectively.

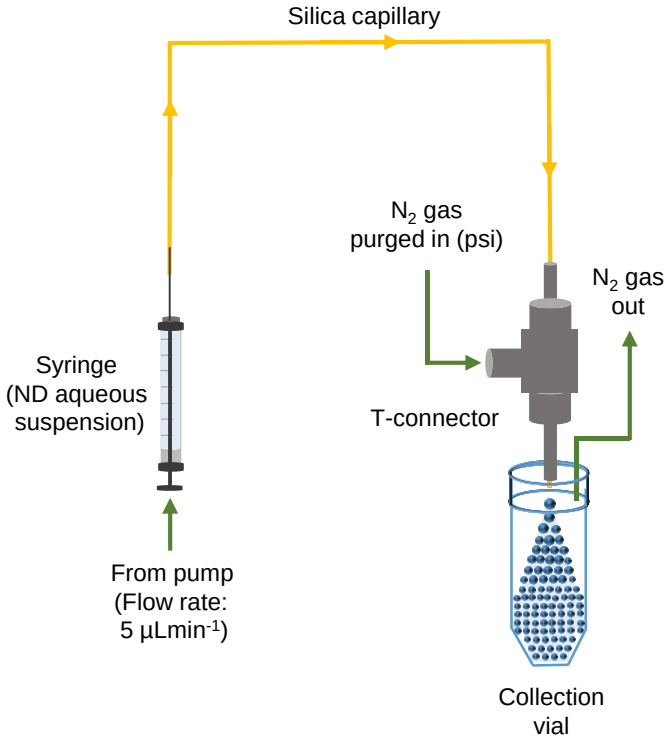

Figure S5. Schematic of the SS set-up.

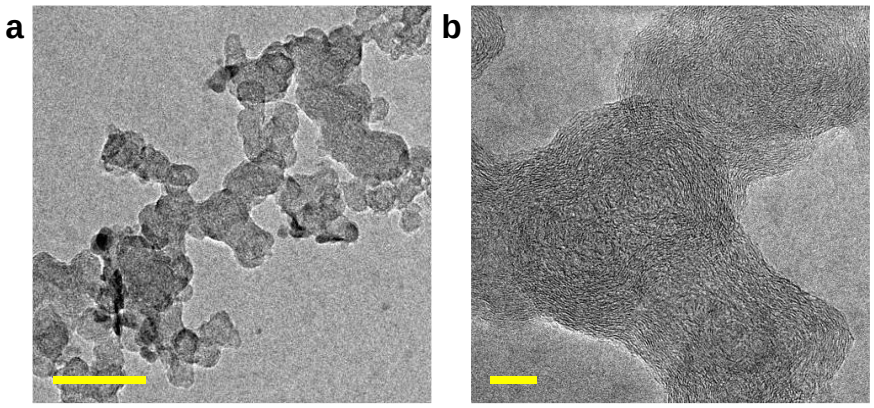

Figure S6. TEM images of OLCs obtained after ES using fused silica capillary. The scale bars in (a) and (b) are 100 and 10 nm.

Table S1. Comparison of the observed Raman peaks with literature[1]

Characteristic feature	Before ES (ND)		After ES (OLC)	
	Observed/ cm^{-1}	Literature/ cm^{-1}	Observed/ cm^{-1}	Literature/ cm^{-1}
D-band	1336	~1326	1349	~1343
G-band	1632	~1647	1624	~1585

Calculation S1. Calculation of increase in sp^2/sp^3 ratio in XPS after AESD

Description	Percentage of sp^2 and sp^3 content	
	Before ES (ND)	After ES (OLC)
sp^2	43.38	63.46
sp^3	45.57	20.05
sp^2/sp^3	0.95	3.16

Calculation S2.

S2a. Evaluation of electric field at the capillary-tip

The electric field at the tip of the capillary with inner radius, r_c (25 μm) placed at a distance, d (8 mm) from the ITO glass slide can be calculated using the relation described below, provided the applied voltage, V_c (2.6 kV) is known.[2]

$$E_c = \frac{V_c}{r_c \ln(4d/r_c)}$$

$$E_c = \frac{2.6 \times 10^3\ V}{25 \times 10^{-6}\ m\ ln\left(\frac{4 \times 8 \times 10^{-3}\ m}{25 \times 10^{-6}\ m}\right)}$$

$$E_c = \frac{2.6 \times 10^3\ V}{25 \times 10^{-6}\ m\ \times 7.15} = 1.45 \times 10^7\ Vm^{-1}$$

S2b. Evaluation of Laplace pressure on the microdroplet in ES and SS

Laplace pressure, ΔP is related to the interfacial tension, γ across the capillary tip with a radius of curvature, r_c using this relation[3]

$$\Delta P = \frac{2\gamma}{r_c}$$

The inner radius of curvature is 25 and 75 µm for borosilicate and silica capillary used in ES and SS, respectively. The interfacial tension or surface tension of water is 75 mN/m, which gave approximate Laplace pressure as 5.8 kPa and 2 kPa.

For ES,

$$\Delta P = \frac{2 \times (72 \times 10^{-3}\ Nm^{-1})}{25 \times 10^{-6}\ m};\ \Delta P \approx\ 5.8\ kPa$$

For SS,

$$\Delta P = \frac{2 \times (72 \times 10^{-3}\ Nm^{-1})}{75 \times 10^{-6}\ m};\ \Delta P \approx 2\ kPa$$

References

(1) Zeiger, M.; Jäckel, N.; Mochalin, V. N.; Presser, V. Review: Carbon Onions for Electrochemical Energy Storage. *J. Mater. Chem. A* **2016**, *4*, 3172–3196.

(2) Kebarle, P.; Tang, L. From Ions in Solution to Ions in the Gas Phase - the Mechanism of Electrospray Mass Spectrometry. *Anal. Chem.* **1993**, *65*, 972–986.

(3) Lee, S.; Kim, D. H.; Needham, D. Equilibrium and Dynamic Interfacial Tension Measurements at Microscopic Interfaces Using a Micropipet Technique. 1. A New Method for Determination of Interfacial Tension. *Langmuir* **2001**, *17*, 5537–5543.