

Supporting Information

Dendritic carbon architectures formed by nanotube core-directed diffusion-limited aggregation of nanoparticles

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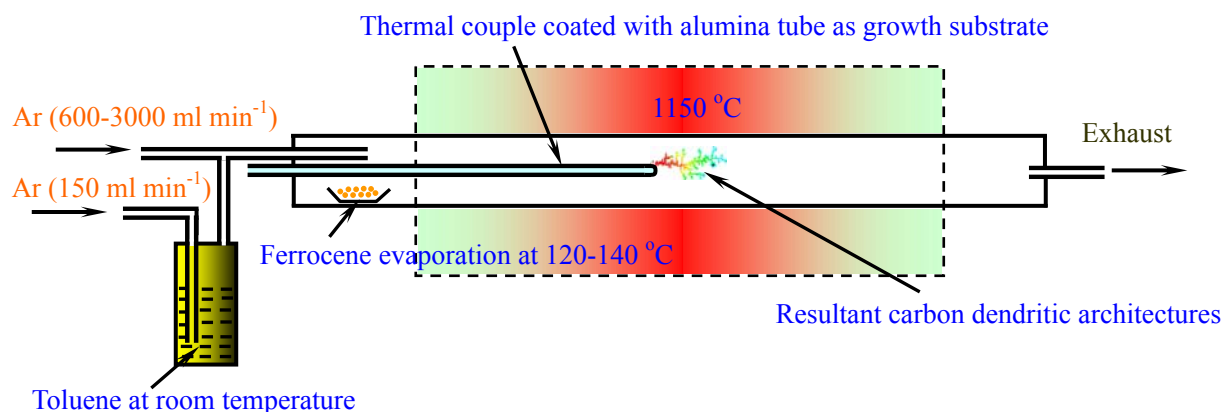


Fig. S1. Schematic diagram of the apparatus used for the growth of dendritic carbon architectures

Monte Carlo simulation details:

The carbon nanotube core directed nanoparticle aggregation model consists of two independent but simultaneous processes, of which a nucleated carbon nanotube acts as a core in the diffusion-limited-aggregation and initializes the branching out. In the second process, nanoparticles are randomly aggregated around the formed nanotube cores and this aggregation results in the thickening of the branches. Due to the independence of those two processes, we used two individual Monte Carlo experiments to simulate the dendritic carbon architecture formations and simplified the basic building blocks as a stick, which combines carbon nanotube core nucleation, evolution and further nanoparticles aggregation.

Nucleation of carbon nanotube core, evolution, and thickening by nanoparticle aggregation on the surface and ultimately develops into the branches by the carbon nanotube core-directed diffusion-limited-aggregation of nanoparticle. The branching out procedure is simulated as following details:

- 1) We start with a stick of a length L at the origin of coordinates as a root of dendritic carbon architecture.
- 2) Another stick represents nucleation of carbon nanotube core and developed branch was generated with its center of gravity $D1$ units away from the origin and its orientation randomly generated.
- 3) Move this stick one unit step toward a random direction and rotate one unit toward a random orientation.
- 4) Repeat step 3 until either this stick touches one of the existing sticks attached to the root or the center of gravity of this stick is more than $D2$ units away from the origin. If this stick touches one of the existing stick attached to the root, then this current stick stops moving and is also attached to the root. If its center of gravity is more than $D2$ units away from the origin, then it is removed from the process.
- 5) Repeat step 2, 3 and 4 until a desired number of units were attached to the root.

In our particular simulation, $L = 10$, $D1 = 30 + \text{height of tree}$, $D2 = 50 + \text{height of tree}$, where *height of tree* is defined as the distance between the origin and the end of the longest branch in the tree.

We simulate the aggregation of nanoparticles around a nanotube core (thickening) represented by an $N \times N$ curved surface area as following:

- 1) Set the initial height (z-value) of each $N \times N$ units to zero, $N=50$ in our simulation.
- 2) A nanoparticle is randomly distributed to one of the $N \times N$ units area and the height value of that corresponding unit increases by one if the height value is below a certain threshold H . $H = 40$ in our simulation.
- 3) Repeat this process K times. K equals 400000 in our simulation and 2600 frames are generated.

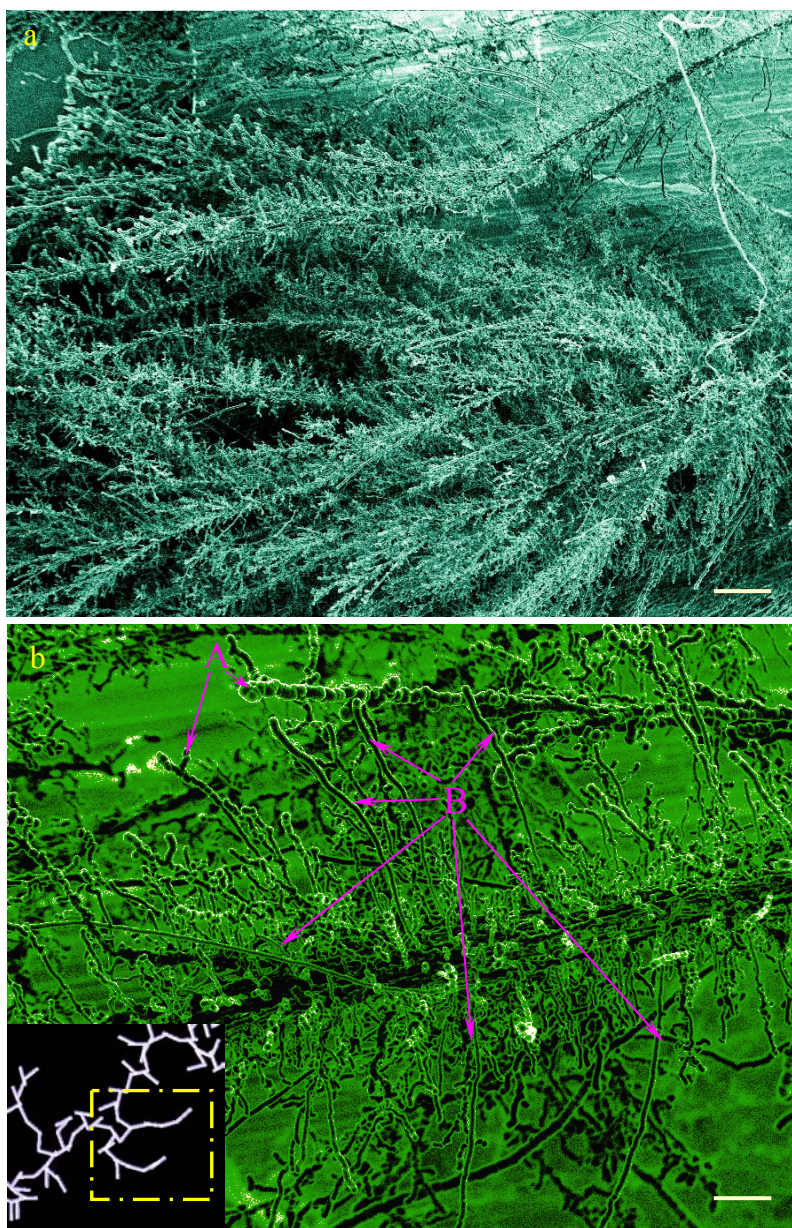


Fig. S2. Macroscopic morphologies of the dendritic carbon architectures evolved by Diffusion-Limited-Aggregation of carbon nanoparticles formed from catalytic pyrolysis of toluene, which is directed by carbon nanotube cores, and followed by subsequent restructuring from surface to bulk. a) Macroscopic morphology of dendritic structure with fractal hyperbranches (scale bar: 200 μm) b) Branches with different morphologies. A. Branches with observably spherulitic nodules; B. Branches with smooth surface and also relatively longer in length. (scale bar: 50 μm) The inset shows Monte Carlo simulation images with the longer branches without branching out due to the continuous development of the core carbon nanotubes.

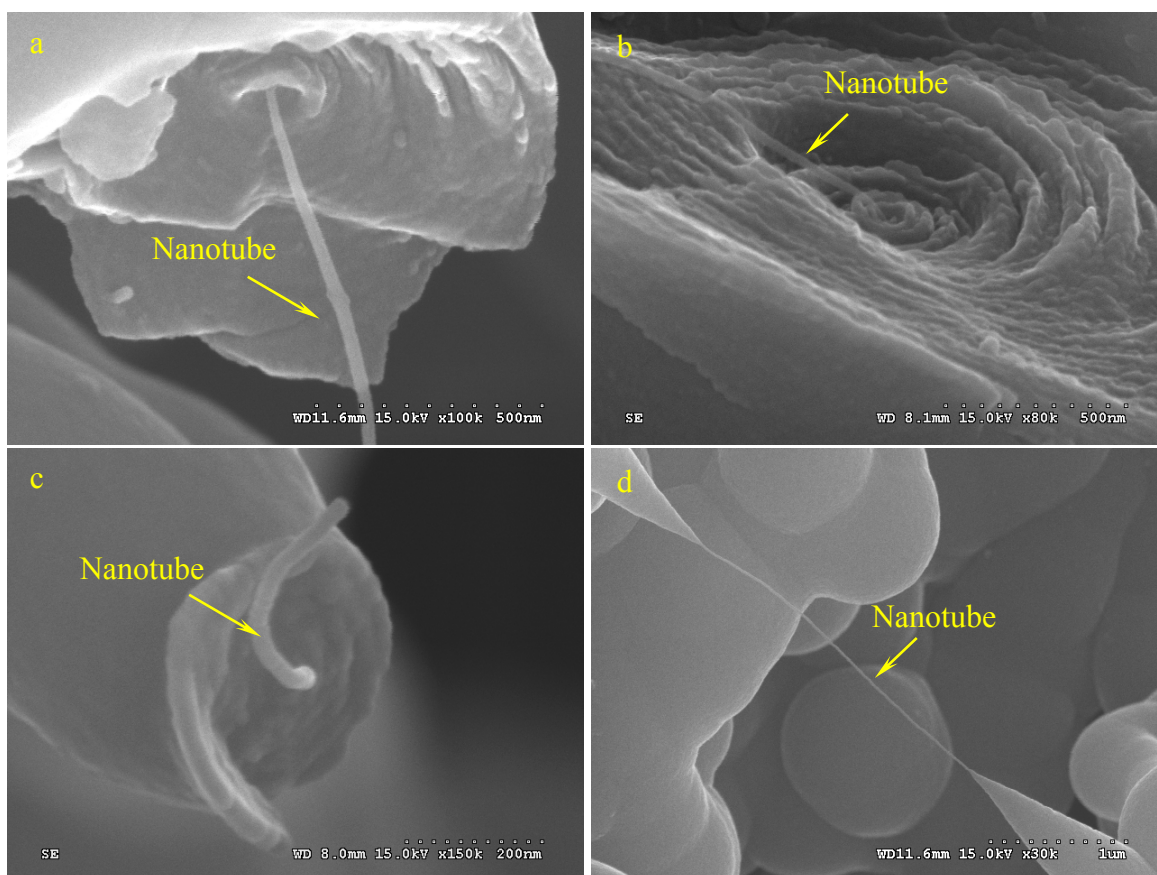


Fig. S3. SEM images to illustrate the presence of carbon nanotube core, the further nucleation on the previous formed branches leading to branching out and subsequent thickening by nanoparticle aggregation.

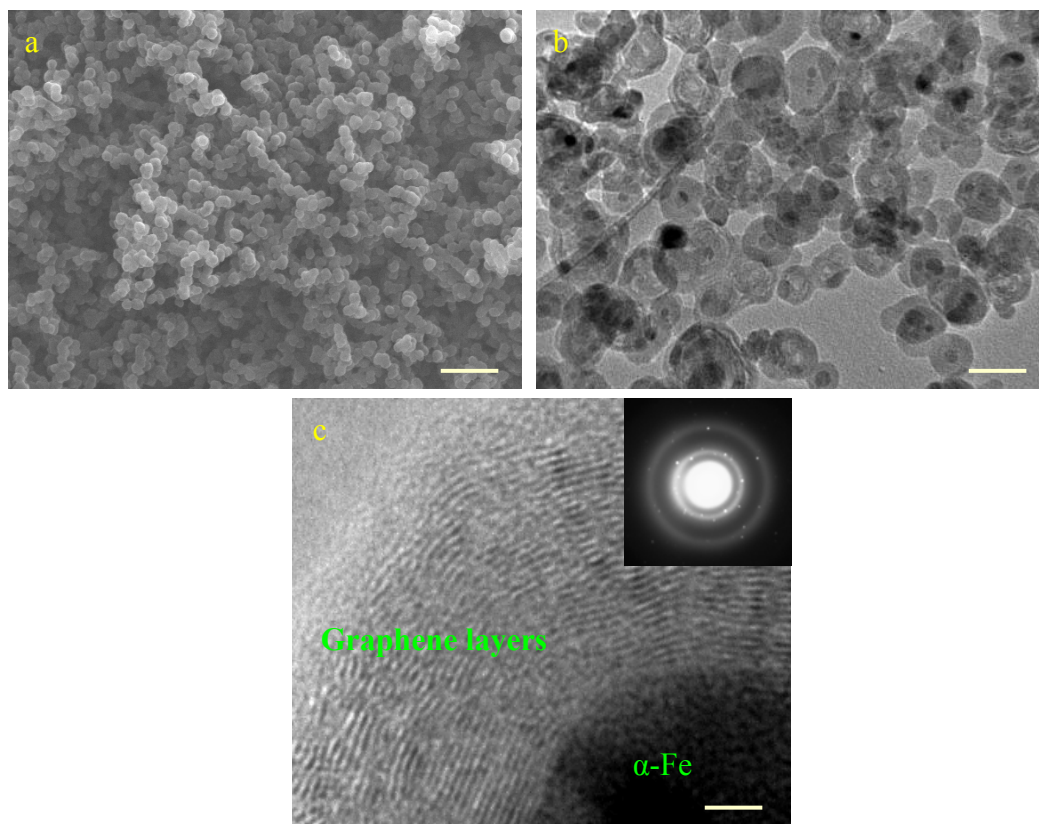


Fig. S4. Nanoparticles collected at the high temperature end of reactor wall. a) SEM micrograph of carbon nanoparticles formed by catalytic pyrolysis of toluene. (scale bar: 200 nm) b) TEM image manifesting core-shell structure. (scale bar: 100 nm) c) High-resolution TEM image revealing the graphitic fringe and Fe-core structure. The inset is a SAED pattern for the nanoparticles. (scale bar: 3 nm)

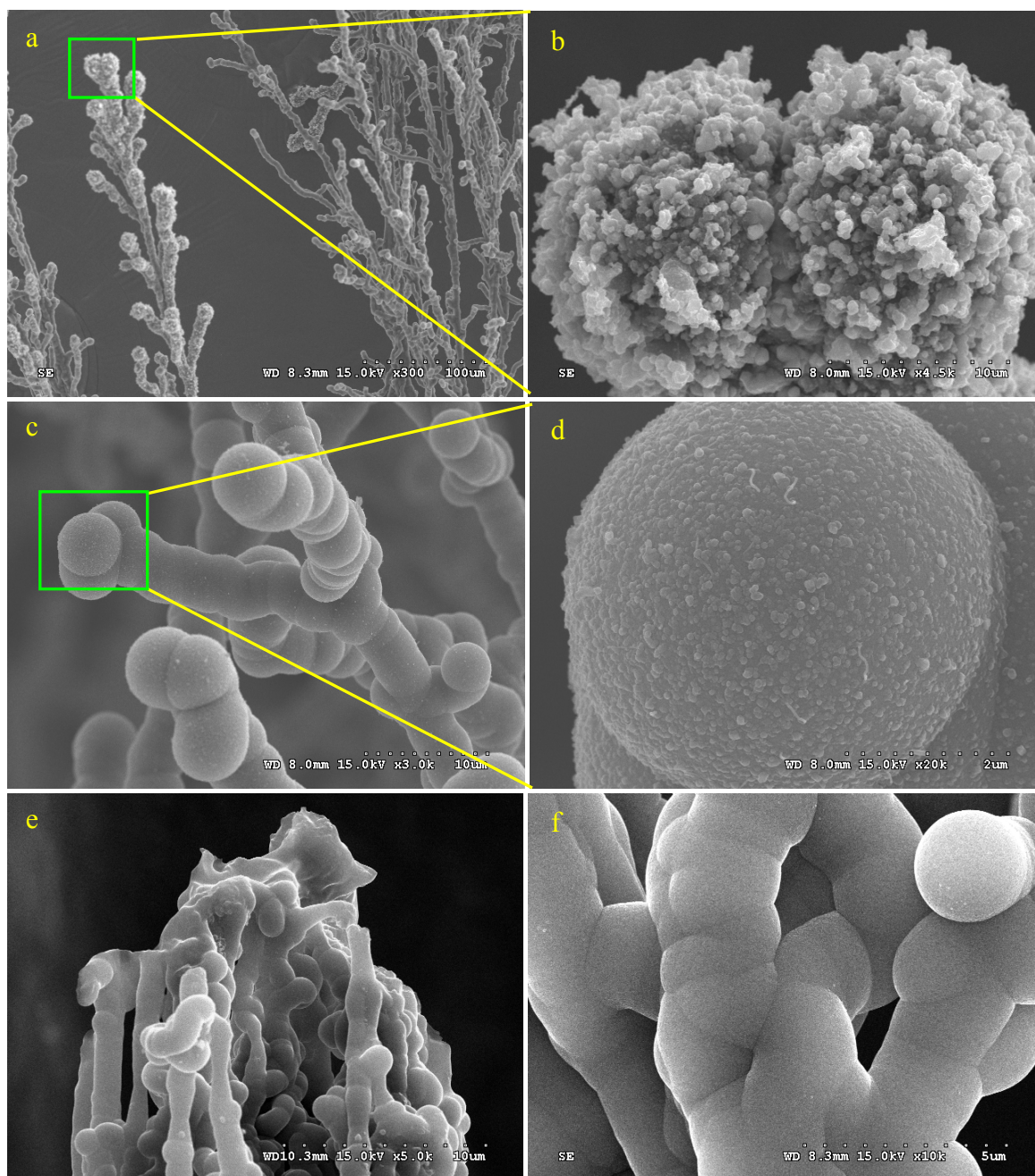


Fig. S5. Morphology and microstructure at different positions of the catalytically grown dendritic carbon architectures to illustrate the evolution of the surface due to restructuring. (a,b) Macroscopic morphology of some newly formed branches. (c,d) Restructuring for a short time to show the trace of the branch is thickening by the aggregated nanoparticles. (e,f) Restructuring for the longest time at the root part to show the smooth surface.