Electronic Supplementary Information

## Graphene-based crown-cork-like macrostructures

Kenta Hiratochi,<sup>ab</sup> Daisuke Terada,<sup>a</sup> Hiroshi Suga,<sup>a</sup> Mitsuhiro Okada,<sup>b</sup> Kyoko Bando,<sup>b</sup> Tetsuya Kodaira,<sup>b</sup> Takatoshi Yamada,<sup>b</sup> Tetsuo Shimizu,<sup>b</sup> Koichiro Saiki,<sup>bc</sup> and Toshitaka Kubo\*<sup>b</sup>

- <sup>a.</sup> Chiba Institute of Technology (Chiba Tech), 4-11-4 Tsudanuma, Narashino, Chiba 275-0016, Japan
- <sup>b.</sup> National Institute of Advanced Industrial Science and Technology (AIST),

Tsukuba Central 5-2, 1-1-1 Higashi, Tsukuba, Ibaraki 305-8565, Japan.

E-mail: t-kubo@aist.go.jp

<sup>c.</sup> Department of Complexity Science and Engineering, The University of Tokyo, Kashiwanoha 5-1-5,

Kashiwa, Chiba 277-8561, Japan



Fig. S1: Experimental apparatus and temperature profile for graphene growth. (a) A diagram of the CVD setup. (b) A typical temperature profile as applied during the CVD process.



Fig. S2: Elemental analysis of the Fe surface after graphene growth. (a) An SEM image and (b-d) EDS mapping images of the Fe substrate after 30 min of growth. (e) EDS spectra of regions I,  $II_a$ , and  $II_b$  as labeled in the SEM images.



Fig. S3: Bulk structures and elemental analysis of an Fe substrate after graphene growth. (a, b) SEM images and (c-e) EDS mapping images of the Fe substrate after 60 min of growth followed by immersion in HCl to remove the surface region of the Fe substrate in several tenth of micrometers in depth and the C precipitate.



Fig. S4: Elemental analysis of a cross-sectional image of an Fe substrate after graphene growth. (a) An SEM image and (b-d) EDS mapping images of the Fe substrate after 60 min of growth. (e) EDS spectra of regions I and II as labeled in the SEM image.



Fig. S5: Elemental analysis of a cross-sectional image of an Fe substrate after graphene growth. (a) An SEM image and (b-d) EDS mapping images of the Fe substrate after 120 min of growth. (e) EDS spectra of regions I and II as labeled in the SEM image.



Fig. S6: (a and b) Additional views of the TEM image shown in Fig. 2(c). (c) A magnified view of the area indicated by the red square in (b).