Supplementary Information

Large-Area Single-Crystal Graphene Grown on a Recrystallized Cu(111) Surface by using the Hole-Pocket Method

Hoang Danh Phan[#a], Jaehyuck Jung[#b], Youngchan Kim[b], Van Ngoc Huynh[c] and Changgu Lee[a,b*]

[a] Department of Mechanical Engineering, Sungkyunkwan University, Suwon 440-746, Korea

[b] SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University, Suwon 440-746, Korea

[c] Department of Physics, Sungkyunkwan University, Suwon 440-746, Korea.

*Correspondence: Professor C Lee, 85293, Cooperate collaboration center, Sungkyunkwan Univ., 2066, Seobu-ro, Jangan-gu, Suwon-si, Gyeonggi-do, Republic of Korea 440-760/ telephone: +82-31-299-4844/ Fax: +82-31-299-7930/ peterlee@skku.edu

# D. Phan and J. Jung contributed equally to this work.

Cu foil folding process

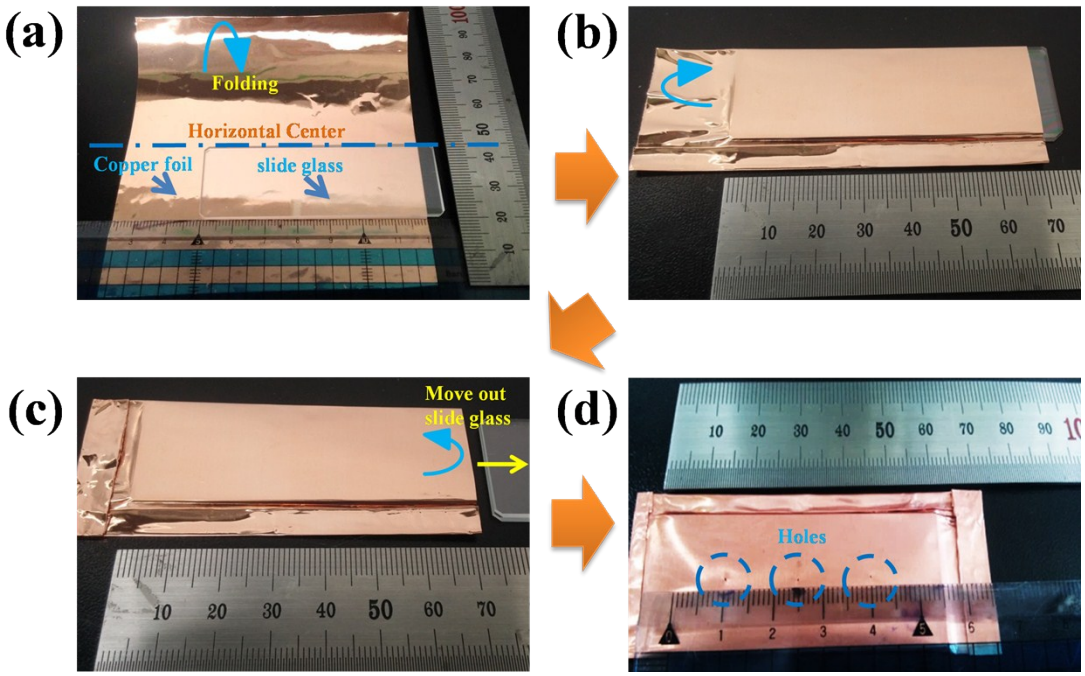

Figure S1. Making process of the hole-pocket Cu foil shape. (a) Prepare the Cu foils and a clean slide glass. (b-c) Fold this Cu foil on the slide glass. (d) Make the holes on the top side of pocket.

CVD system for graphene synthesis.

An atmospheric pressure chemical vapor deposition (APCVD) system with tubular quartz was used to synthesize Graphene films.

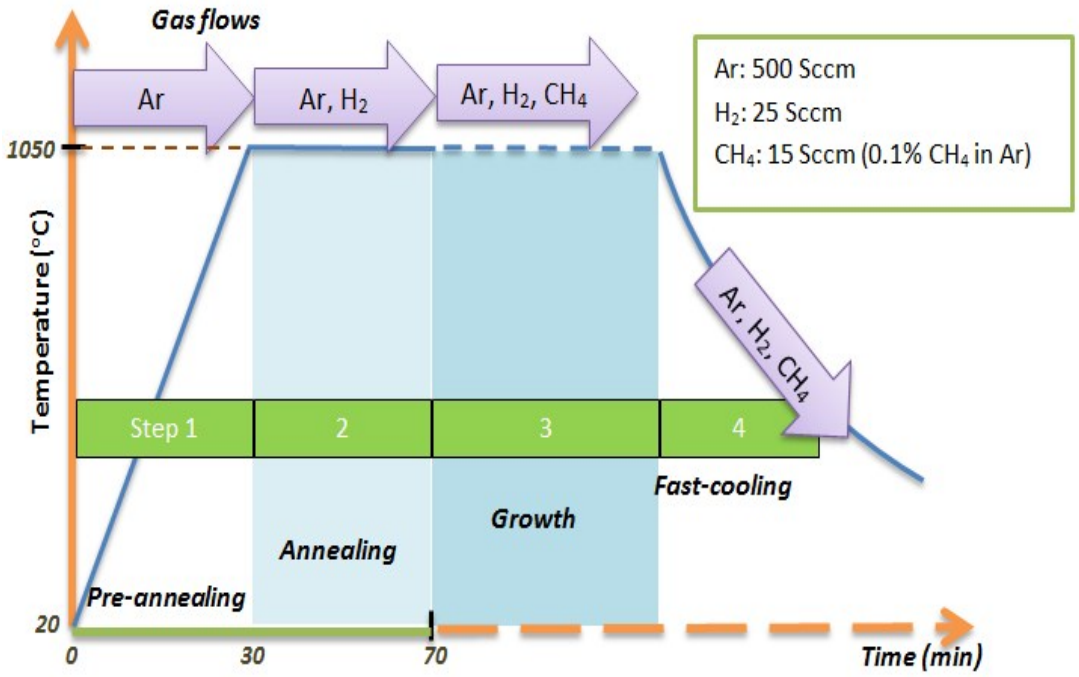

Figure S2. Process of graphene synthesis using CVD.

Hexagonal graphene domains on Cu foil

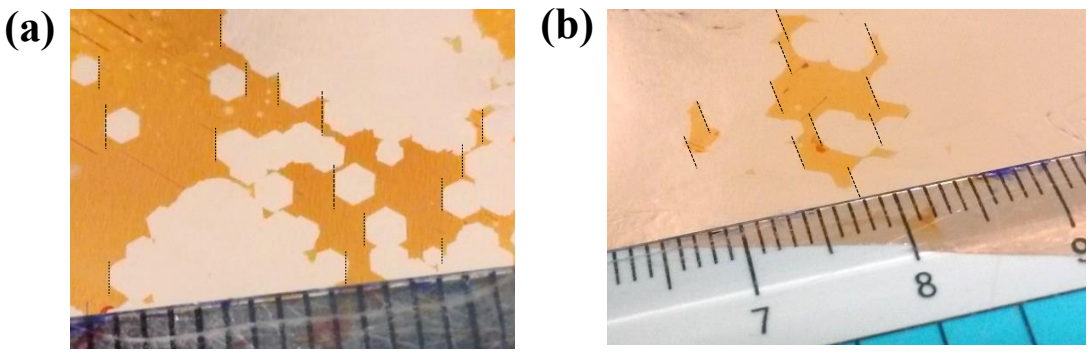

Figure S3**.** Optical images showing hexagonal graphene grown with different time. (a) Graphene domains grown for 3 hours. (b) Graphene domains grown for 5 hours.

Effect of holes on Cu recrystallization

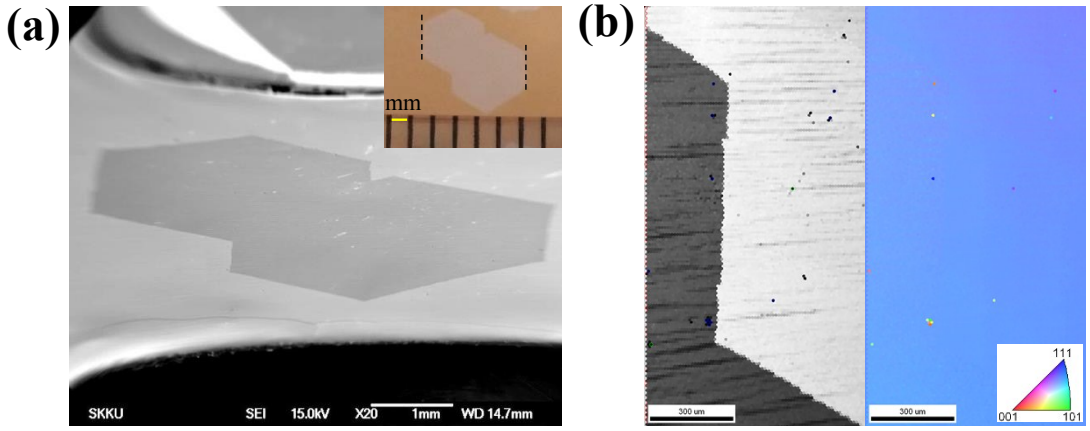

Figure S4. Two graphene domains stitched with the same crystalline orientation (a) Scanning electron microscope (SEM) image graphene domains after 4 hours of synthesis. (b) EBSD image for the orientation of Cu crystal. The yellow box in (a) is the EBSD analysis area.

Effect of annealing condition on Cu(111) recrystallization.

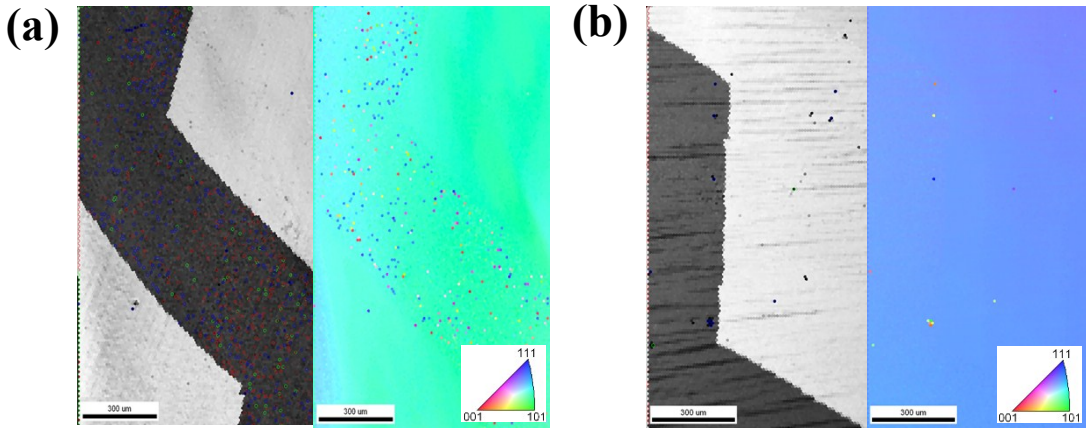

Figure S5. SEM and EBSD images with different growth time for the growth of graphene. (a) Growth time in 2 hours at 1050 ^{o}C. (b) Growth time in 4 hours at 1050 ^{o}C.

Transfer process of Graphene on SiO_2/Si

Figure S6 represents Graphene transfer process on SiO_2/Si substrate after graphene film synthesized on Cu foil by hole-pocket method and continuous graphene film with centimeter scale.

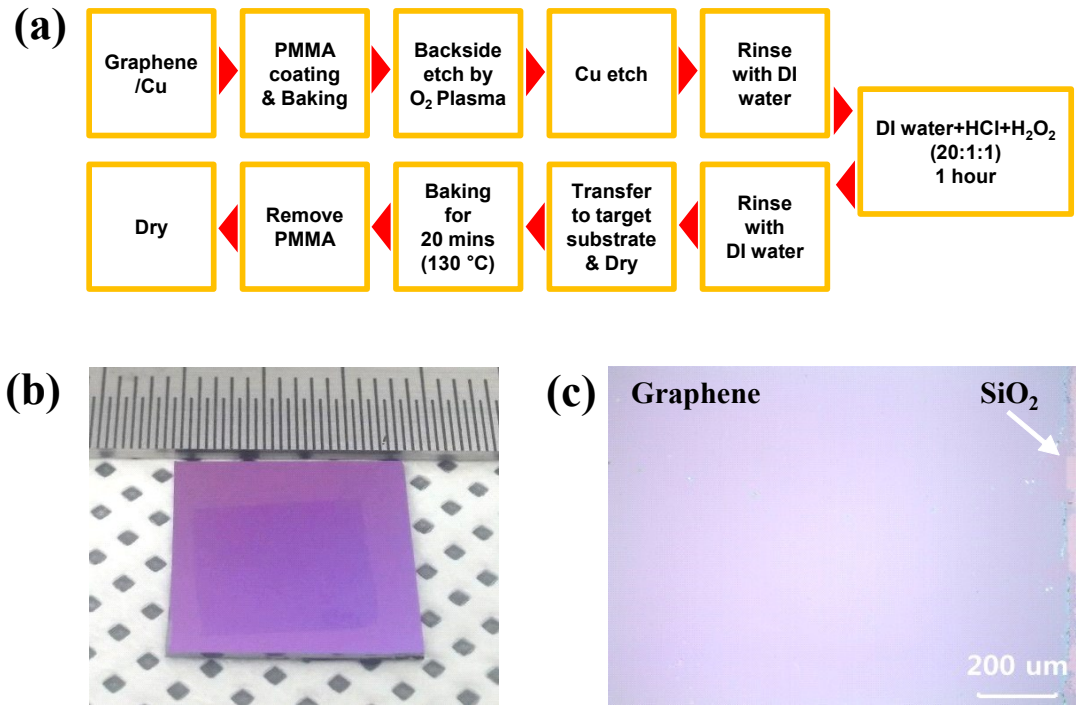

Figure S6. Graphene transfer process and continuous graphene film. (a) a schematic image for transfer process of Graphene. (b) Continuous large-area Graphene on silicon wafer. (c) Optical microscope image of graphene film on Silicon wafer.

LC analysis of hole-pocket synthesized graphene

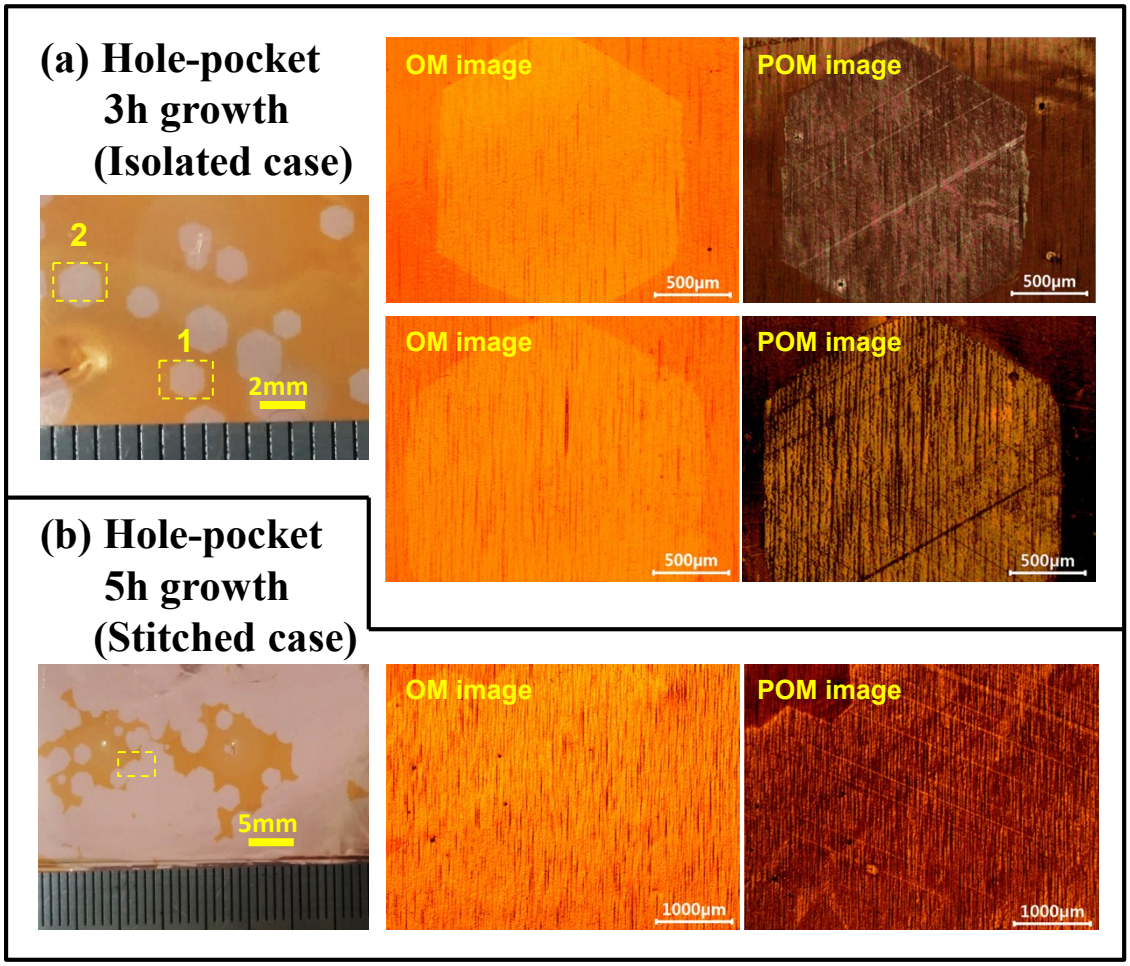

Figure S7. OM and POM images of isolated and stitched graphene domains synthesized using hole-pocket method by controlling growth time. (a) Isolated graphene single domain case with 3h growth. No detectable contrast was found for both domains. (b) Graphene domains merged due to longer growth time. Uniform contrast was observed for large area which indicates single-crystalline orientation in the large area.