Supporting Information for:

Highly Uniform Growth of Monolayer Graphene by Chemical Vapor Deposition on Cu-Ag Alloy Catalysts

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A. **Raman mapping**

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<th>Raman mapping of 30 x 30 μm² area</th>
<th>Scale up Raman mapping (500 x 500 μm²)</th>
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<td>(a) G peak signal pick-up</td>
<td>(d) G peak</td>
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<td>(b) G peak intensity</td>
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<td>(c) 2D peak intensity</td>
<td>(e) 2D peak</td>
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<td>Cu</td>
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**Figure S1** Spectroscopic Raman mapping of graphene characterization. (a) Signal pick-up of the G and intensity maps of the (b) G and (c) 2D band for Cu, Ag200Cu and Ag500Cu respectively with 30 x 30 μm² map area. (d-e) Scale up mapping of Raman spectroscopy with 500 x 500 μm² map area. Intensity maps of the (d) G band and (e) 2D band for Cu, Ag200Cu and Ag500Cu, respectively.

Figure S1 shows the Spectroscopic Raman mapping of graphene synthesized on Cu, Ag200Cu and Ag500Cu for the coverage comparison of graphene. The each view of field is 30 x 30 μm² of graphene film transferred on the SiO₂/Si wafer. The wavelength of the Raman excitation laser was 514 nm (Renishow Equipment) and the Raman map pixel size is 1.0 μm.
Figure S1 (a) shows the G peak signal pick-up of each graphene. If graphene existed at some point, there was G peak signal with red color mapping. For graphene synthesized on Cu, graphene was not fully synthesized with lots of graphene-undetected area. However graphene was synthesized nearly 100% for the graphene of Ag200Cu and Ag500Cu. The partial coverage of graphene synthesized on Cu was reconfirmed with G band and 2D band peak intensity as shown in Figure S1 (b) and (c). Peak intensity difference of G band for graphene synthesized on Cu was relatively severe as compared with graphene of Ag200Cu and Ag500Cu. On the other hand, the peak intensity difference was larger for graphene synthesized on Ag500Cu than for that graphene of Ag200Cu. This result resulted from the increase of the multilayered graphene formation for Ag500Cu. Graphene synthesis was enhanced with the increase of Ag plating thickness coinciding with the result of optical microscopic images in Figure 2.

For the large scale demonstration of graphene coverage, the view of field of 500 x 500 µm² area was performed using Widefield Raman imaging (WRI) system. For WRI system, data acquisition time could be minimized with the optical filter which enables the measurement of selective range of D band (~1350 cm⁻¹), G band (~1590 cm⁻¹) and 2D band (~2700 cm⁻¹). In addition high efficiency and image enlargement were available for WRI system using image grab method. The wavelength of the Raman excitation laser was 532 nm. The Raman map pixel size was 1.5 µm with 5 seconds of data acquisition time. Higher intensity of G and 2D band was represented with dark color and therefore non-coverage region was represented with white color and multilayered graphene was represented with black color.

Graphene coverage results for large area were accord with the Figure S1 (a-c) which showed the results of conventional small area mapping. Non-covered region and intensity difference distribution were larger for graphene synthesized on Cu than for Ag plated Cu.
Therefore for the graphene synthesis at 900 for 40 min, graphene film was not fully covered for Cu. On the other hand full coverage graphene film was formed for Ag plated Cu and graphene synthesis was enhanced with increasing the thickness of plated Ag.

B. Supplemental of EBSD results

Figure S2 EBSD orientation maps of Cu (a) after graphene synthesis on Ag500Cu at 900 °C (b) texture analysis of Ag200Cu after graphene synthesis at 1000 °C; Scale bars (200 μm)