Simultaneous growth of monolayer graphene on Ni/Cu bimetallic catalyst by atmospheric pressure CVD process

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Point-based Raman curves for graphene grown at the inner surface of Ni and the inner surface of Cu of the used Ni-Cu bilayer catalyst at CVD temperature of 950°C for 5 mins on are shown in Fig. S1.

Fig. S1 Representative Raman spectra of graphene grown at (a) inner surface of Ni and (b) inner surface of Cu of Ni-Cu bilayer catalyst at CVD temperature of 950°C for 5 mins.
**AFM analysis** was performed with Park System XE-100 on the graphenes grown at inner surface of Ni and inner surface of Cu and deposited onto silicon wafer.

![AFM images for graphene grown on inner surface of Ni (left), and inner surface of Cu (right) and deposited onto silicocon wafer (CVD reaction conditions: temperature 950 °C and duration 5 min).](image)

**Fig. S2** AFM images for graphene grown on inner surface of Ni (left), and inner surface of Cu (right) and deposited onto silicocon wafer (CVD reaction conditions: temperature 950 °C and duration 5 min).

**Controlled experiment 1.** The purpose of the controlled experiment 1 is to verify that the carbon precursor at the interface of Ni-Cu bimetallic catalyst are mainly came from the diffusion through Ni foil. The Cu-Silicon-Ni trilayer catalysts (Figure S1a) and Cu-Ni-Silicon trilayer catalysts (Figure S1b) was prepared and firmly pressed with hydraulic press to displace the air between and maximize the contact between all foils respectively. The CVD was carried out with 80 sccm H\textsubscript{2}, 20 sccm CH\textsubscript{4}, and temperature for 1000 °C and last for 10 min. Longer period and higher temperature used is to confirm that the leak is still insignificant even though under harsh condition.

![Diagram showing the built-up of the (a) Cu-silicon-Ni and (b) Cu-Ni-silicon catalysts used for the control experiment 1.](diagram)

**Fig. S3** The built-up of the (a) Cu-silicon-Ni and (b) Cu-Ni-silicon catalysts used for the control experiment 1.
Controlled experiment 2. In the controlled experiment 2, a Ni foil is used to cover a Cu foil. The CVD condition was set at 950 °C and for 5 min, being the conditions optimized for the inverse bi-metallic system.

Fig. S4 (a) The built-up of the catalytic system used for the control experiment 2. Typical Raman spectra of graphene grown at (b) inner surface of Ni and (c) inner surface of Cu of the bilayer catalyst from Ni wrapped over Cu and transferred to silicon wafer.
Graphitic material formed at 1030 °C

Fig. S5 Optical Micrograph of graphitic material. Optical micrograph of the web-like graphitic carbon grown at the inner surface of Ni at 1030 °C for 3 min.