

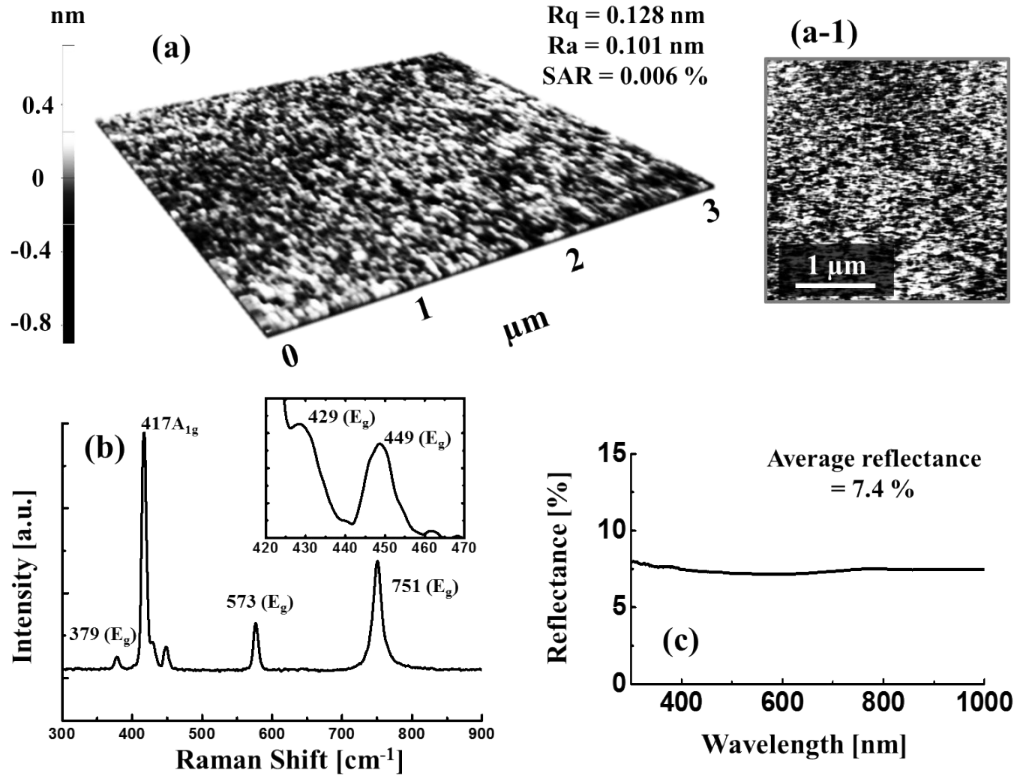
# Supplementary Materials for Publication

## *Compositional Effect on the Fabrication of $\text{Ag}_x\text{Pd}_{1-x}$ Alloy Nanoparticles on c-Plane Sapphire at Distinctive Stages of the Solid- State-Dewetting of Bimetallic Thin Films*

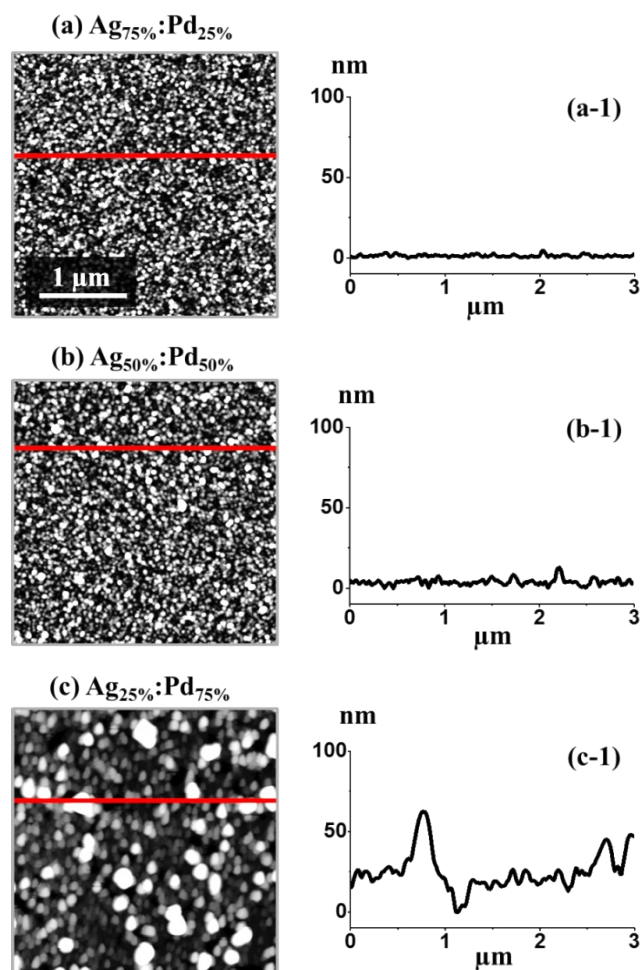
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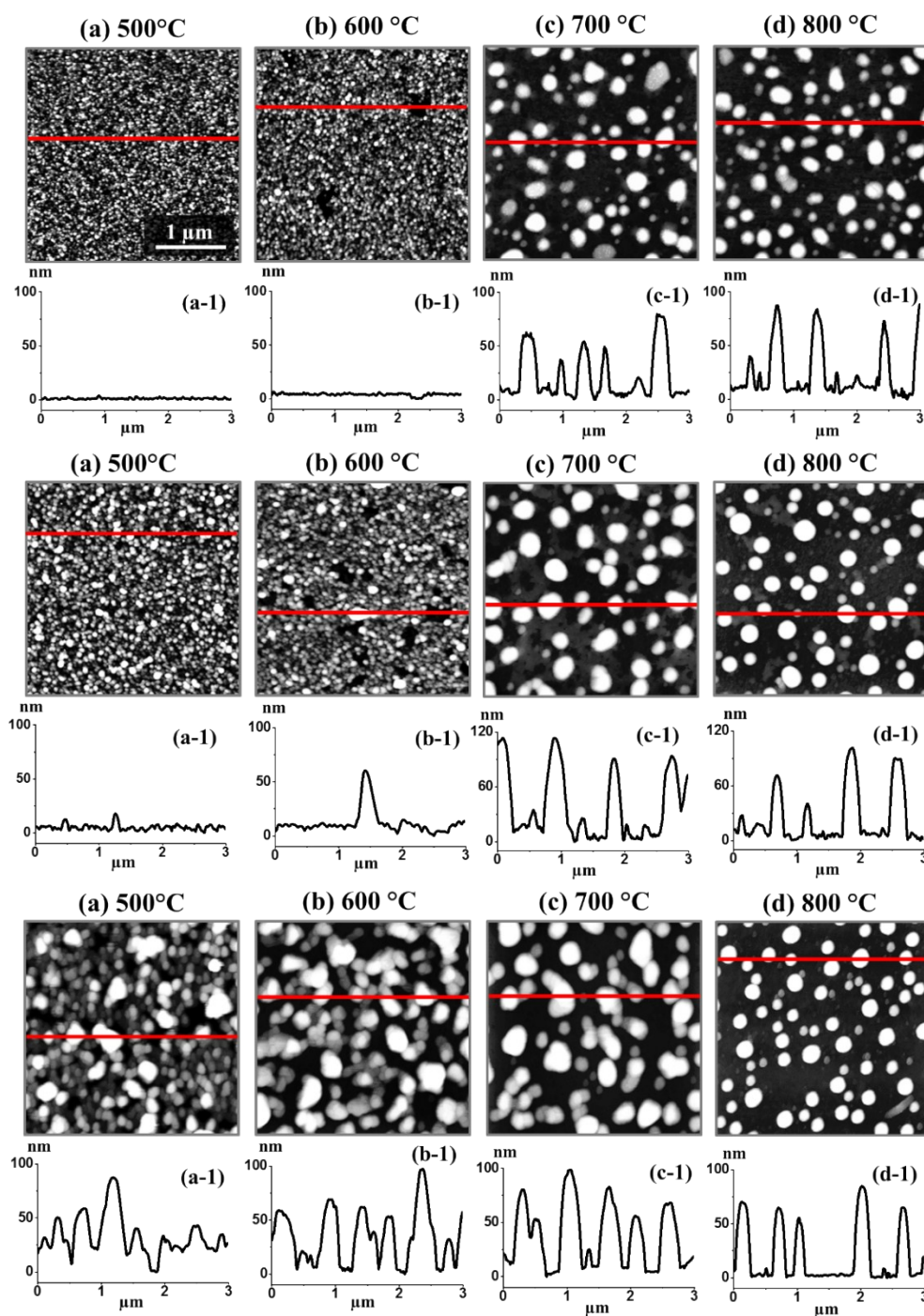
**Note added after first publication:** This Electronic Supplementary Information file replaces that originally published on 7th December 2017, and contains a corrected Table S7 and Fig. S14.



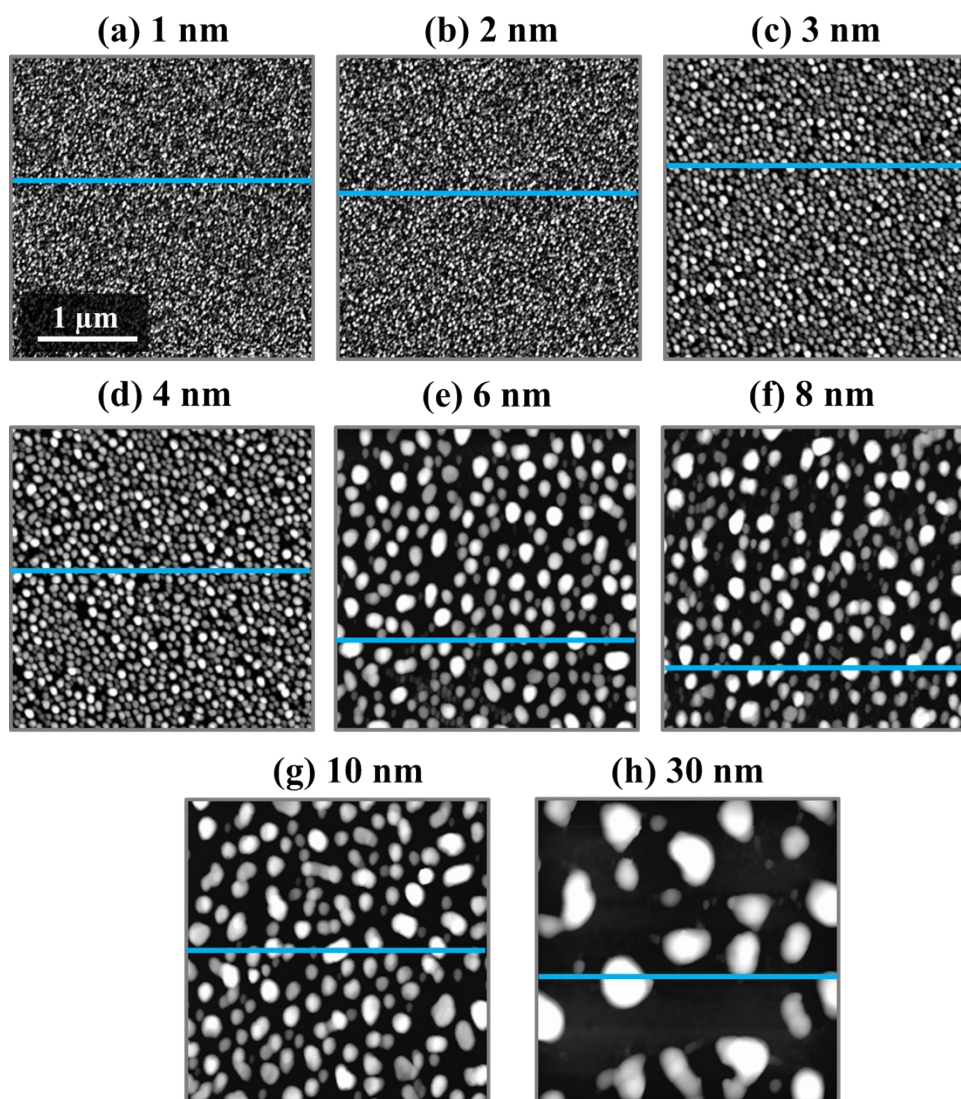
**Figure S1** Bare sapphire after the degassing at  $350^\circ\text{C}$  for 30 mins. (a) AFM side-view of  $3 \times 3 \mu\text{m}^2$ . (a-1) Corresponding AFM top-view. (b) Raman spectrum of the bare sapphire. In total, 6 peaks are observed in the spectrum: 379, 417, 429, 449, 573 and  $751 \text{ cm}^{-1}$ . The  $416 \text{ cm}^{-1}$  peak is induced by  $A_{1g}$  vibrational mode and the other peaks are induced by the  $E_g$  vibrational mode. (c) Reflectance spectrum of the bare sapphire.



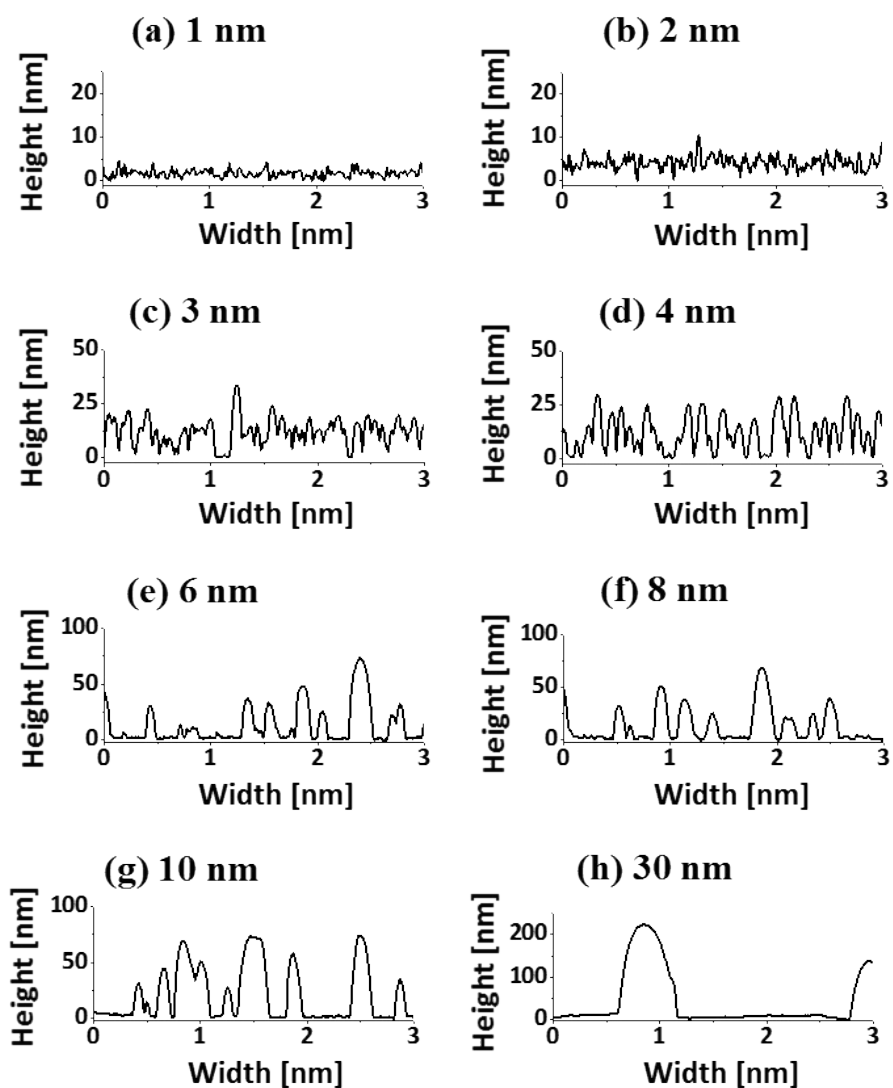
**Figure S2** Ag-Pd bimetallic nanostructures fabricated at 400 °C for 120 s with the deposition thickness of 15 nm and bilayer composition as labelled. (a) – (c) AFM top-views of  $3 \times 3 \mu\text{m}^2$ . (a-1) – (c-1) Corresponding line-profiles.



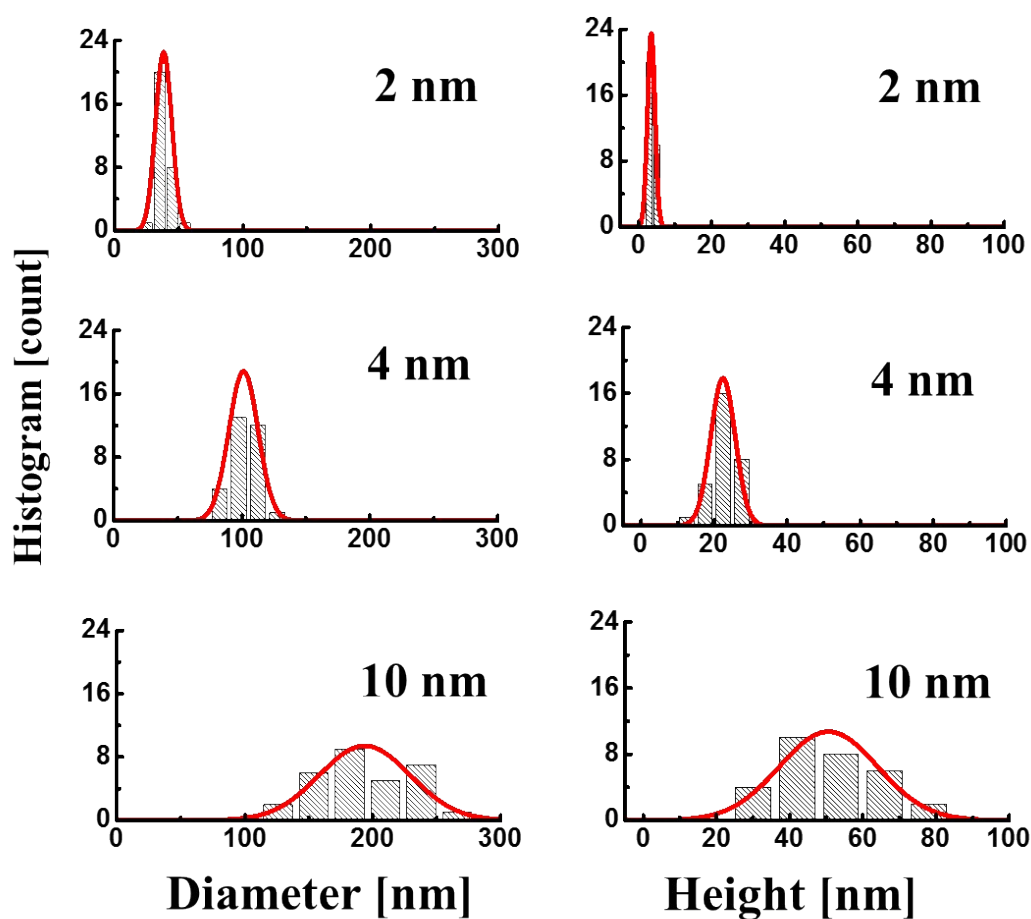
**Figure S3** Ag-Pd bimetallic nanostructures fabricated between 500 and 800 °C for 120 s with the deposition thickness of 15 nm. (a) – (l) AFM top-views of  $3 \times 3 \mu\text{m}^2$ . (a-1) – (l-1) Corresponding line-profiles.



**Figure S5** Ag-Pd bimetallic nanostructures fabricated by the control of deposition thickness between 2 and 30 nm with  $\text{Pd}_{0.25}\text{Ag}_{0.75}$  bilayer composition. The samples were annealed at 650 °C for 120 s. (a) – (h) AFM top-views of  $3 \times 3 \mu\text{m}^2$ .

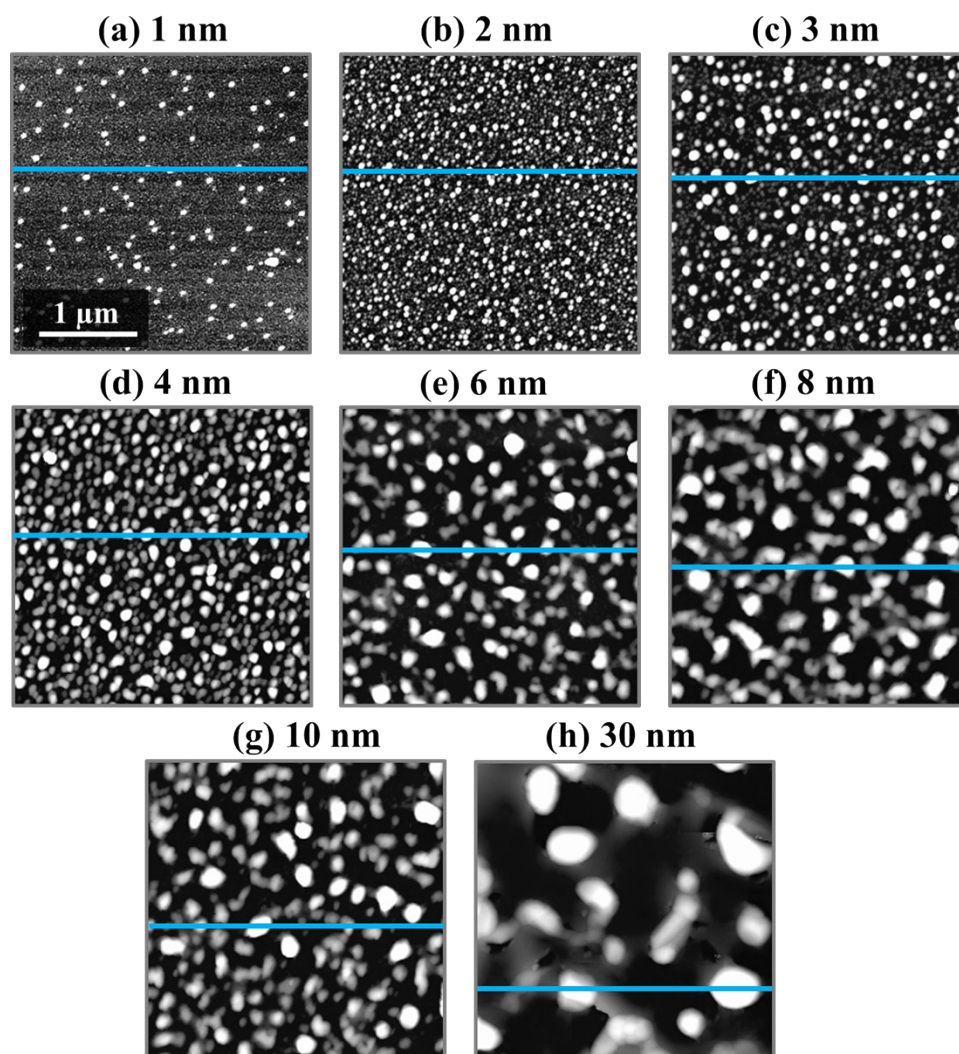


**Figure S6** Cross-sectional line profiles of the AFM top-views shown in Fig. S5.



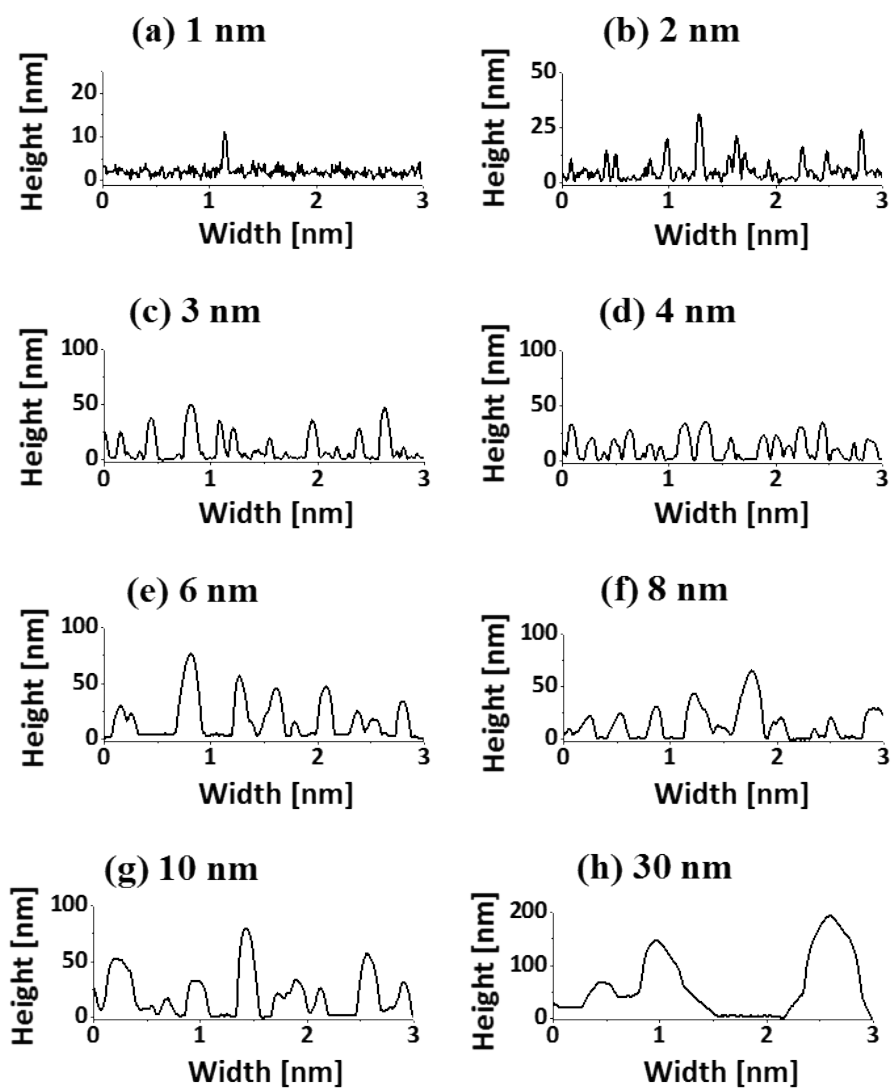
**Figure S7** Quantative analysis for the Ag-Pd bimetallic nanostrucutres fabricated with the control of deposition thickness as shown in Fig. S5. Diameter (left column) and height (right colume) distribution histograms



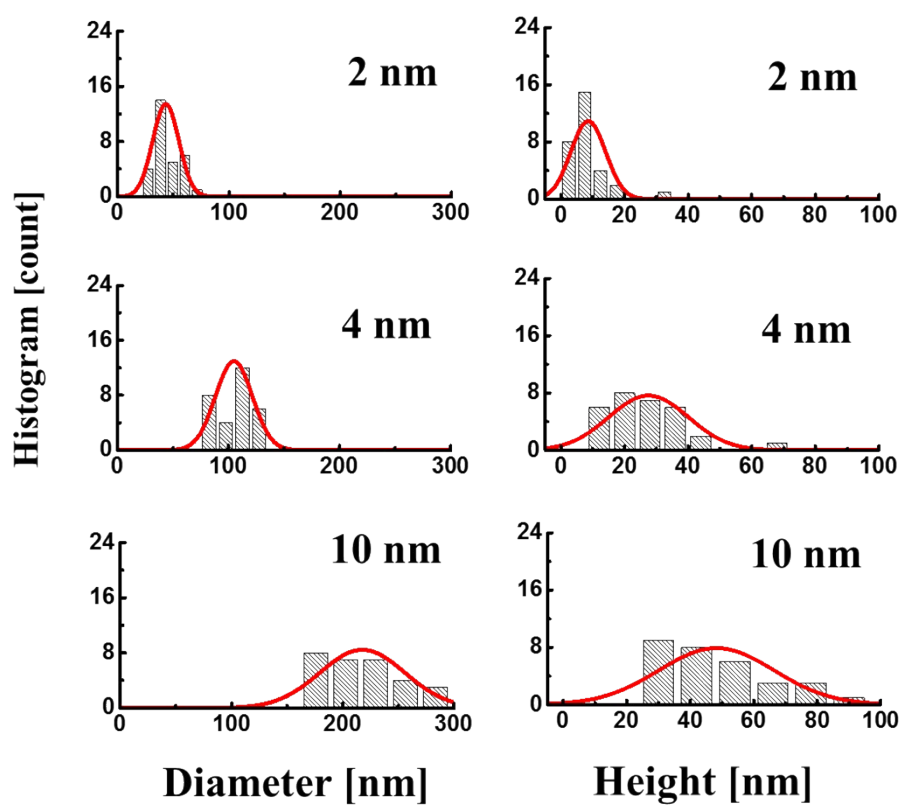


**Figure S8** Ag-Pd bimetallic nanostructures fabricated by the control of deposition thickness between 1 and 30 nm with  $\text{Pd}_{0.75}\text{Ag}_{0.25}$  bilayer composition. The samples were annealed at 650 °C for 120 s. (a) – (h) AFM top-views of  $3 \times 3 \mu\text{m}^2$ .

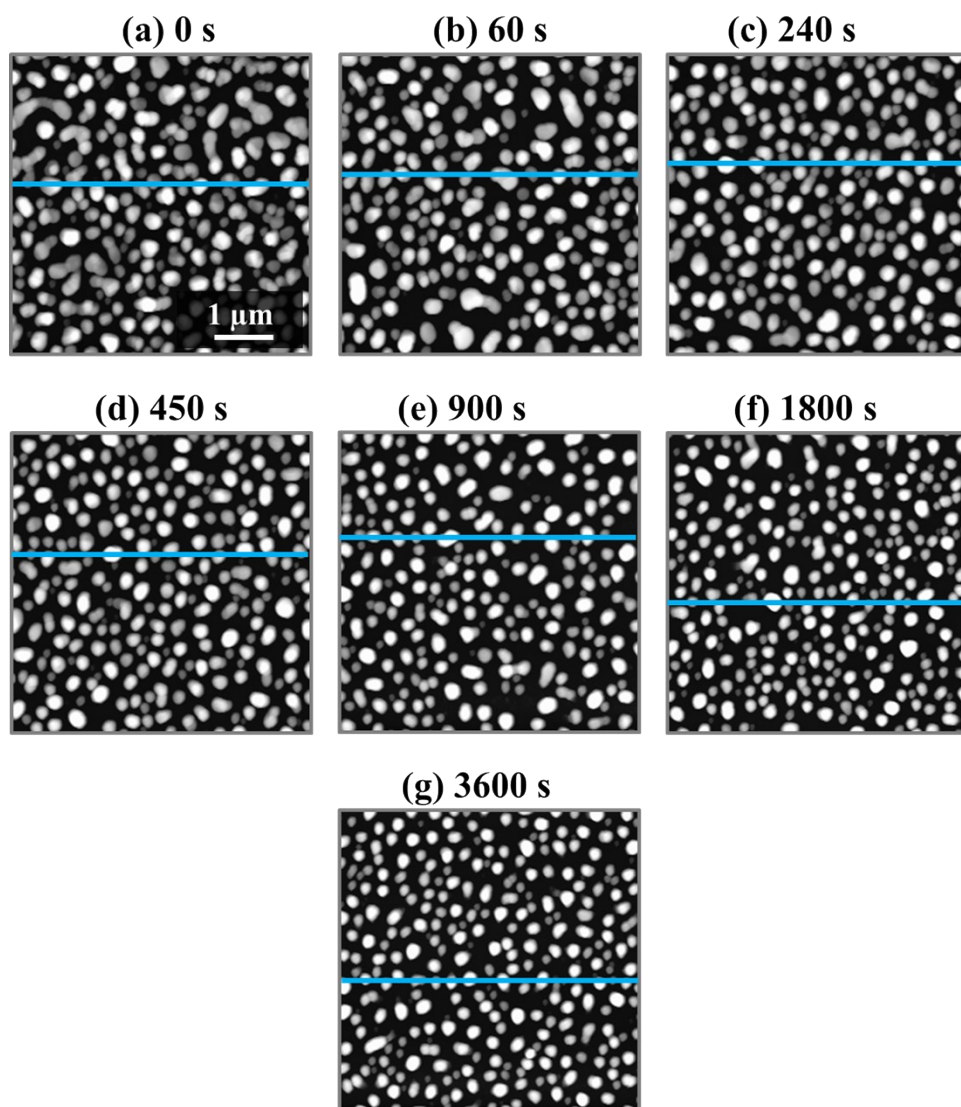




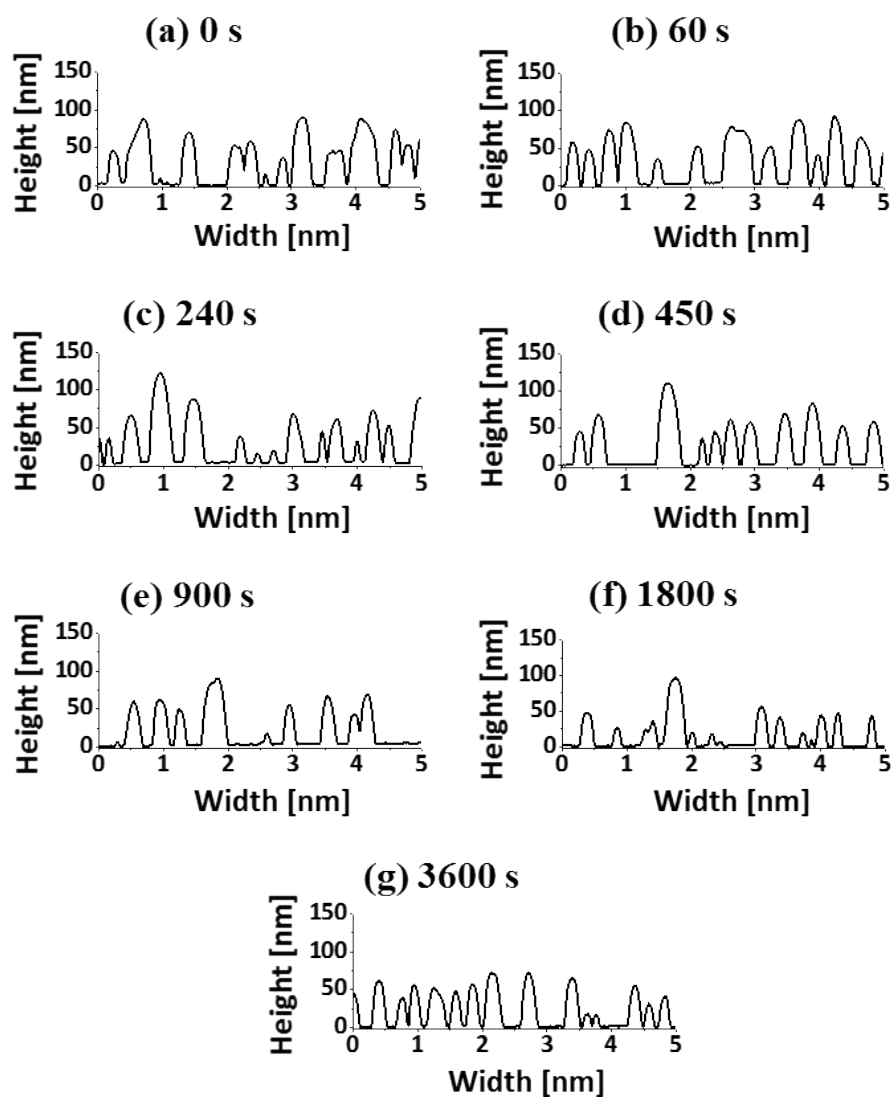
**Figure S9** Cross-sectional line profiles of the AFM top-views shown in Fig. S8.



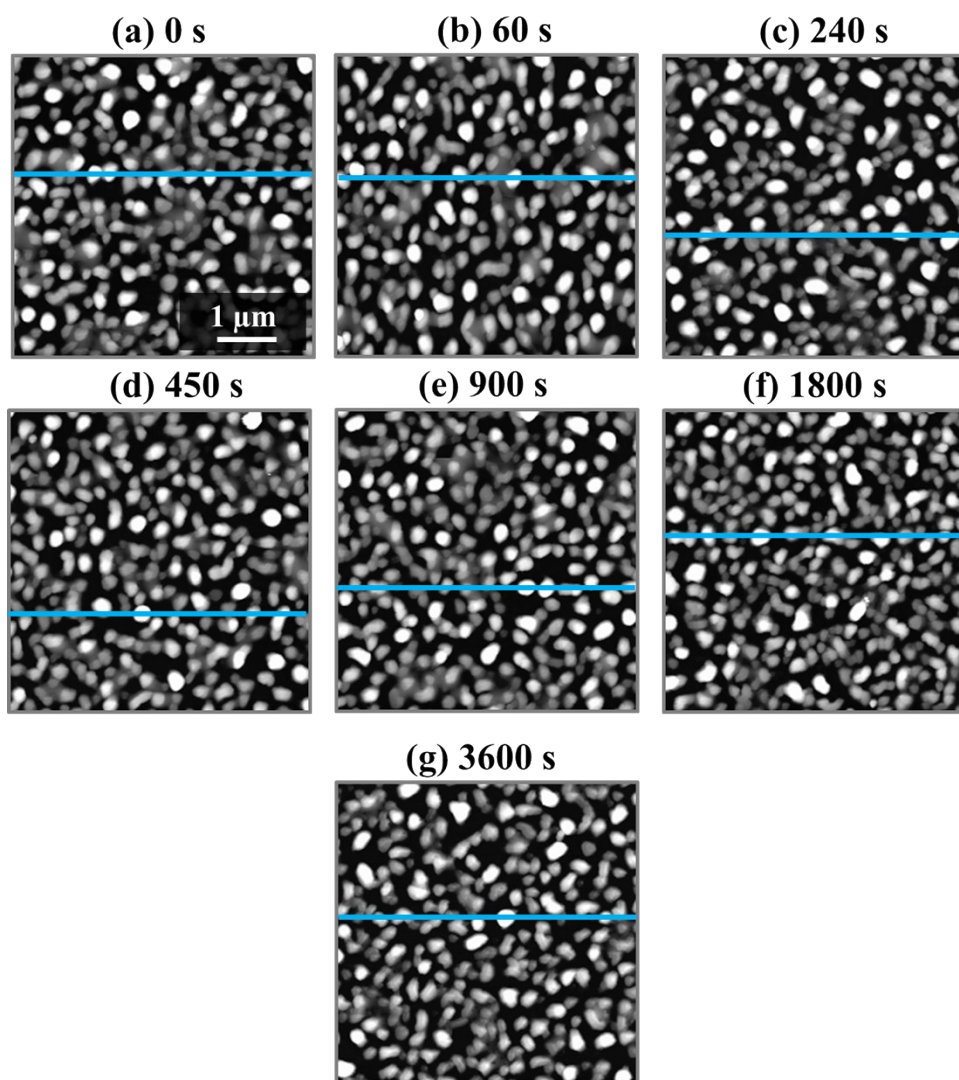
**Figure S10** Quantative analysis for the Ag-Pd bimetallic nanostrucutres fabricated with the control of deposition thickness as shown in Fig. S8. Diameter (left column) and height (right colume) distribution histograms



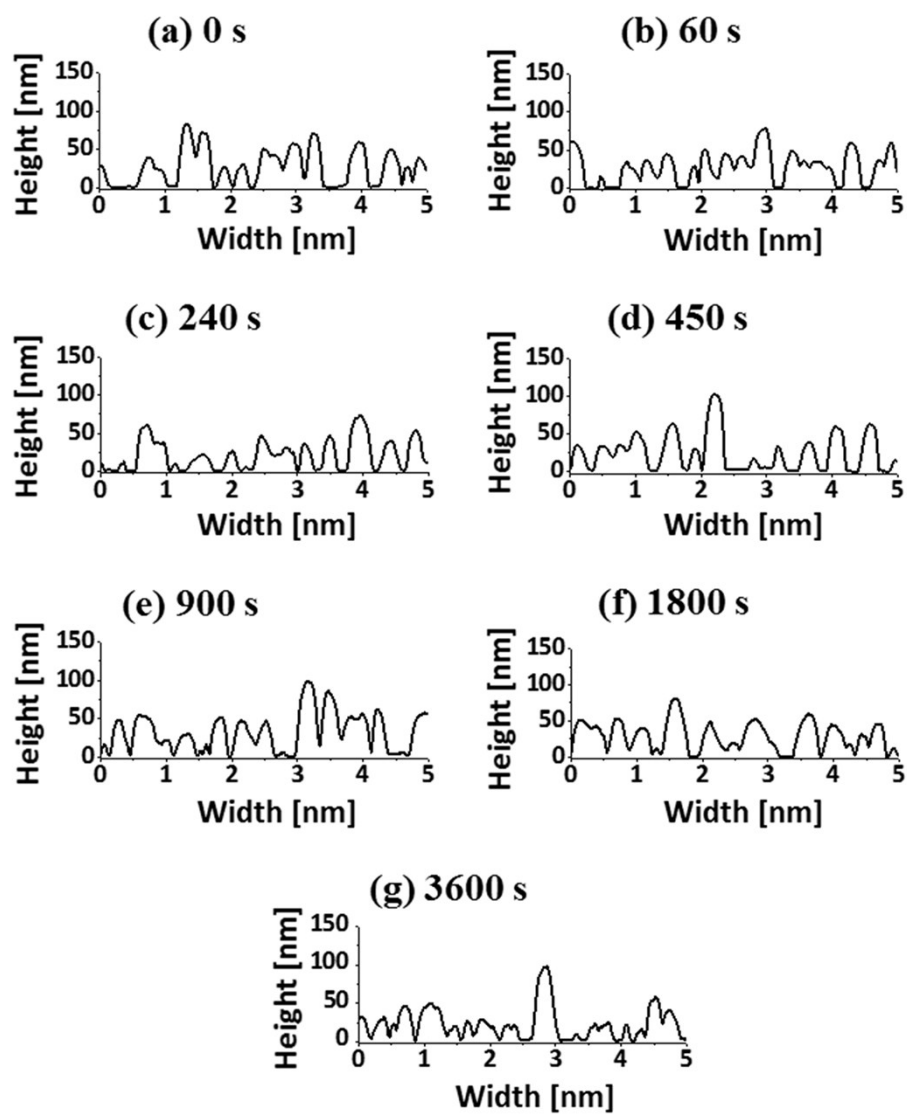
**Figure S11** Ag-Pd bimetallic nanostructures fabricated by the variation of the annealing durations between 0 and 3600 s at 650 °C with the deposition thickness of 10 nm and  $\text{Pd}_{0.25}\text{Ag}_{0.75}$  bilayer composition. (a) – (g) AFM top-views of  $5 \times 5 \mu\text{m}^2$ .



**Figure S12** Cross-sectional line profiles of the AFM top-views shown in Fig. S11.



**Figure S13** Fabrication of Ag-Pd bimetallic nanostructures by the variation of the annealing durations between 0 and 3600 s at 650 °C with the deposition thickness of 10 nm and  $\text{Pd}_{0.75}\text{Ag}_{0.25}$  bilayer composition. (a) – (g) AFM top-views of  $5 \times 5 \mu\text{m}^2$ .



**Figure S14** Cross-sectional line profiles of the AFM top-views shown in Fig. S13.



**Table S1** Summaries of Rq and SAR of the Ag, Pd bimetallic nanostructures fabricated at the control of temperature between 500 and 800 °C for 360 s with the bilayer composition of Ag<sub>0.75</sub>Pd<sub>0.25</sub>, Ag<sub>0.50</sub>Pd<sub>0.50</sub> and Ag<sub>0.25</sub>Pd<sub>0.75</sub> with 15 nm total thickness.

| Temperature<br>[°C] | Ag <sub>0.75</sub> Pd <sub>0.25</sub> |       |          | Ag <sub>0.75</sub> Pd <sub>0.25</sub> |       |          | Ag <sub>0.25</sub> Pd <sub>0.75</sub> |       |          |
|---------------------|---------------------------------------|-------|----------|---------------------------------------|-------|----------|---------------------------------------|-------|----------|
|                     | Rq                                    | SAR   | Coverage | Rq                                    | SAR   | Coverage | Rq                                    | SAR   | Coverage |
| 400                 | 7.23                                  | 1.7   | 97.22    | 1.92                                  | 0.59  | 99.87    | 0.72                                  | 0.12  | 100      |
| 500                 | 13.61                                 | 5.07  | 84.32    | 2.33                                  | 0.72  | 99.64    | 0.69                                  | 0.11  | 100      |
| 600                 | 24.56                                 | 9.83  | 49.28    | 3.67                                  | 0.65  | 94.97    | 0.91                                  | 0.13  | 99.63    |
| 700                 | 28.55                                 | 10.52 | 31.73    | 28.3                                  | 10.68 | 32.15    | 20.46                                 | 9.11  | 32.73    |
| 800                 | 19.46                                 | 9.49  | 23.42    | 26.16                                 | 10.86 | 25.69    | 23.38                                 | 10.73 | 30.16    |

**Table S2** Summary of average reflectance of the Ag, Pd bimetallic nanostructures fabricated at the control of temperature between 500 and 800 °C for 360 s with the deposition of  $\text{Ag}_{0.75}\text{Pd}_{0.25}$ ,  $\text{Ag}_{0.50}\text{Pd}_{0.50}$  and  $\text{Ag}_{0.25}\text{Pd}_{0.75}$ .

| Temperature<br>[°C] | $\text{Ag}_{0.75}\text{Pd}_{0.25}$ | $\text{Ag}_{0.50}\text{Pd}_{0.50}$ | $\text{Ag}_{0.25}\text{Pd}_{0.75}$ |
|---------------------|------------------------------------|------------------------------------|------------------------------------|
| <b>500</b>          | 30.47                              | 38.22                              | 37.53                              |
| <b>600</b>          | 22.56                              | 31.15                              | 36.86                              |
| <b>700</b>          | 18.76                              | 9.31                               | 10.25                              |
| <b>800</b>          | 11.33                              | 9.08                               | 11.23                              |

**Table S3** Summary of Rq and SAR of the samples fabricated at 650 °C for 360 s with various deposition thickness of Ag, Pd bi-metallic alloy of Pd<sub>0.25</sub>Ag<sub>0.75</sub> and Pd<sub>0.75</sub>Ag<sub>0.25</sub>.

| DA<br>[nm] | Pd <sub>0.25</sub> Ag <sub>0.75</sub> |       |          | Pd <sub>0.75</sub> Ag <sub>0.25</sub> |      |          |
|------------|---------------------------------------|-------|----------|---------------------------------------|------|----------|
|            | Rq                                    | SAR   | Coverage | Rq                                    | SAR  | Coverage |
| <b>1</b>   | 2.06                                  | 0.61  | -        | 0.96                                  | 1.49 | -        |
| <b>2</b>   | 4.23                                  | 2.73  | 87.95    | 1.69                                  | 5.36 | 74.95    |
| <b>3</b>   | 8.92                                  | 4.64  | 80.9     | 5.38                                  | 8.08 | 49.48    |
| <b>4</b>   | 9.73                                  | 10.58 | 69.02    | 7.48                                  | 8.77 | 39.77    |
| <b>6</b>   | 13.87                                 | 15.31 | 44.64    | 17.6                                  | 7.42 | 32.57    |
| <b>8</b>   | 14.73                                 | 9.39  | 39.55    | 13.97                                 | 6.79 | 43.16    |
| <b>10</b>  | 14.56                                 | 12.55 | 43.25    | 18.97                                 | 7.09 | 41.85    |
| <b>20</b>  | 15.37                                 | 8.11  | 74.6     | 16.53                                 | 4.48 | 70.45    |
| <b>30</b>  | 41.04                                 | 16.57 | 28.93    | 49.25                                 | 8.66 | 28.39    |

**Table S4** Summary of Rq and SAR of the samples fabricated at 650 °C for 360 s with various deposition thickness of Ag, Pd bi-metallic alloy of Pd<sub>0.25</sub>Ag<sub>0.75</sub> and Pd<sub>0.75</sub>Ag<sub>0.25</sub>. D: diameter; H: height

| <b>DA<br/>[nm]</b> | <b>Pd<sub>0.25</sub>Ag<sub>0.75</sub></b> |          | <b>Pd<sub>0.75</sub>Ag<sub>0.25</sub></b> |          |
|--------------------|-------------------------------------------|----------|-------------------------------------------|----------|
|                    | <b>D</b>                                  | <b>H</b> | <b>D</b>                                  | <b>H</b> |
| <b>2</b>           | 38.12                                     | 3.51     | 43.22                                     | 8.59     |
| <b>3</b>           | 83.57                                     | 18.43    | 72.03                                     | 15.81    |
| <b>4</b>           | 101.26                                    | 22.42    | 104.79                                    | 27.56    |
| <b>6</b>           | 125.35                                    | 31.22    | 166.37                                    | 43.11    |
| <b>8</b>           | 166.3                                     | 36.16    | 212.47                                    | 45.14    |
| <b>10</b>          | 193.77                                    | 54.24    | 217.6                                     | 48.25    |

**Table S5** Summary of average reflectance of the samples fabricated at 650 °C for 360 s with various deposition thickness of Ag, Pd bi-metallic alloy of Pd<sub>0.25</sub>Ag<sub>0.75</sub> and Pd<sub>0.75</sub>Ag<sub>0.25</sub>.

| <b>DA [nm]</b> | <b>Pd<sub>0.25</sub>Ag<sub>0.75</sub></b> | <b>Pd<sub>0.75</sub>Ag<sub>0.25</sub></b> |
|----------------|-------------------------------------------|-------------------------------------------|
| <b>1</b>       | 8.75                                      | 8.36                                      |
| <b>2</b>       | 9.72                                      | 8.58                                      |
| <b>3</b>       | 10.74                                     | 11.74                                     |
| <b>4</b>       | 11.84                                     | 12.03                                     |
| <b>6</b>       | 15.03                                     | 14.09                                     |
| <b>8</b>       | 16.5                                      | 13.11                                     |
| <b>10</b>      | 16.3                                      | 17                                        |
| <b>20</b>      | 24.39                                     | 22.35                                     |
| <b>30</b>      | 9.13                                      | 8.64                                      |

**Table S6** Summary of Raman intensities of the samples fabricated at 850 °C for 360 s with various deposition thickness of Ag, Pd bi-metallic alloy of Pd<sub>0.25</sub>Ag<sub>0.75</sub> and Pd<sub>0.75</sub>Ag<sub>0.25</sub>.

| DA [nm] | Pd <sub>0.25</sub> Ag <sub>0.75</sub> | Pd <sub>0.75</sub> Ag <sub>0.25</sub> |
|---------|---------------------------------------|---------------------------------------|
| 1       | 1949.16                               | 2396.05                               |
| 2       | 2156.49                               | 2200.71                               |
| 3       | 1741.27                               | 1887.58                               |
| 4       | 1715.87                               | 1550.67                               |
| 6       | 1261.07                               | 1213.89                               |
| 8       | 1380.83                               | 960.31                                |
| 10      | 1121.68                               | 1047.5                                |
| 20      | 1000.64                               | 593.93                                |
| 30      | 1037.53                               | 755.4                                 |

**Table S7** Summary of Rq and SAR of the samples fabricated with the control of annealing duration at 800 °C with various Ag, Pd bi-metallic alloy of Pd<sub>0.25</sub>Ag<sub>0.75</sub> and Pd<sub>0.75</sub>Ag<sub>0.25</sub>

| Annealing<br>Duration<br>[s] | Pd <sub>0.25</sub> Ag <sub>0.75</sub> |       |          | Pd <sub>0.75</sub> Ag <sub>0.25</sub> |      |          |
|------------------------------|---------------------------------------|-------|----------|---------------------------------------|------|----------|
|                              | Rq                                    | SAR   | Coverage | Rq                                    | SAR  | Coverage |
| <b>0</b>                     | 28.7                                  | 15.52 | 51.37    | 18.44                                 | 6.13 | 61.77    |
| <b>60</b>                    | 29.97                                 | 16.07 | 48.95    | 19.3                                  | 6.57 | 54.7     |
| <b>240</b>                   | 28.91                                 | 15.61 | 48.17    | 20                                    | 6.84 | 51.5     |
| <b>450</b>                   | 26.73                                 | 14.27 | 45.58    | 19.06                                 | 7.19 | 49.57    |
| <b>900</b>                   | 23.86                                 | 11.82 | 38.23    | 18.82                                 | 6.59 | 50.72    |
| <b>1800</b>                  | 21.89                                 | 10.92 | 35       | 15.49                                 | 4.96 | 48.77    |
| <b>3600</b>                  | 21.06                                 | 10.64 | 34.3     | 16.31                                 | 5.04 | 47.43    |



**Table S8** Summary of average reflectance of the samples fabricated with the control of annealing duration at 800 °C with various Ag, Pd bi-metallic alloy of Pd<sub>0.25</sub>Ag<sub>0.75</sub> and Pd<sub>0.75</sub>Ag<sub>0.25</sub>

| <b>Annealing<br/>Duration [s]</b> | <b>Pd<sub>0.25</sub>Ag<sub>0.75</sub></b> | <b>Pd<sub>0.75</sub>Ag<sub>0.25</sub></b> |
|-----------------------------------|-------------------------------------------|-------------------------------------------|
| <b>0</b>                          | 20.3                                      | 18.16                                     |
| <b>60</b>                         | 18.55                                     | 16.82                                     |
| <b>240</b>                        | 18.22                                     | 16.58                                     |
| <b>450</b>                        | 17.6                                      | 16.48                                     |
| <b>900</b>                        | 16.75                                     | 16.44                                     |
| <b>1800</b>                       | 14.05                                     | 16.34                                     |
| <b>3600</b>                       | 12.16                                     | 16.69                                     |