Supporting Information for

Highly Correlated Size and Composition of Pt/Au Alloy Nanoparticles via Magnetron Sputtering onto Liquid

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Number of Tables: 3

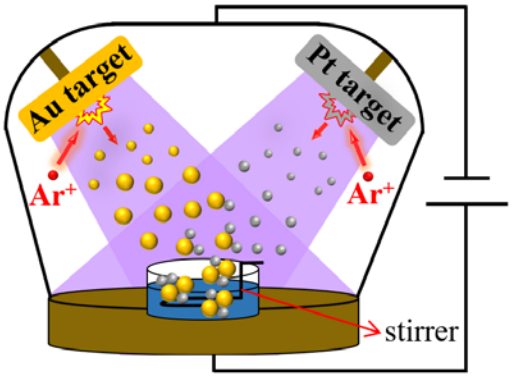

Figure S1. Schematic illustration of the synthesis of Pt/Au alloy NPs via double target sputtering onto PEG.

Table S1. Summary of different preparation methods for Pt/Au NPs.

Method		**Sputtering current /mA**	
		Pt target	**Au target**
I	Sputter only Pt	50	0
II	Sputter only Au	0	50
III	Mixture of (**I**) and (**II**)	50	50
IV	Stepwise sputter	50 (1st sputter)	50 (2nd sputter)
V		50 (2nd sputter)	50 (1st sputter)
VI	Co-sputter	50	50

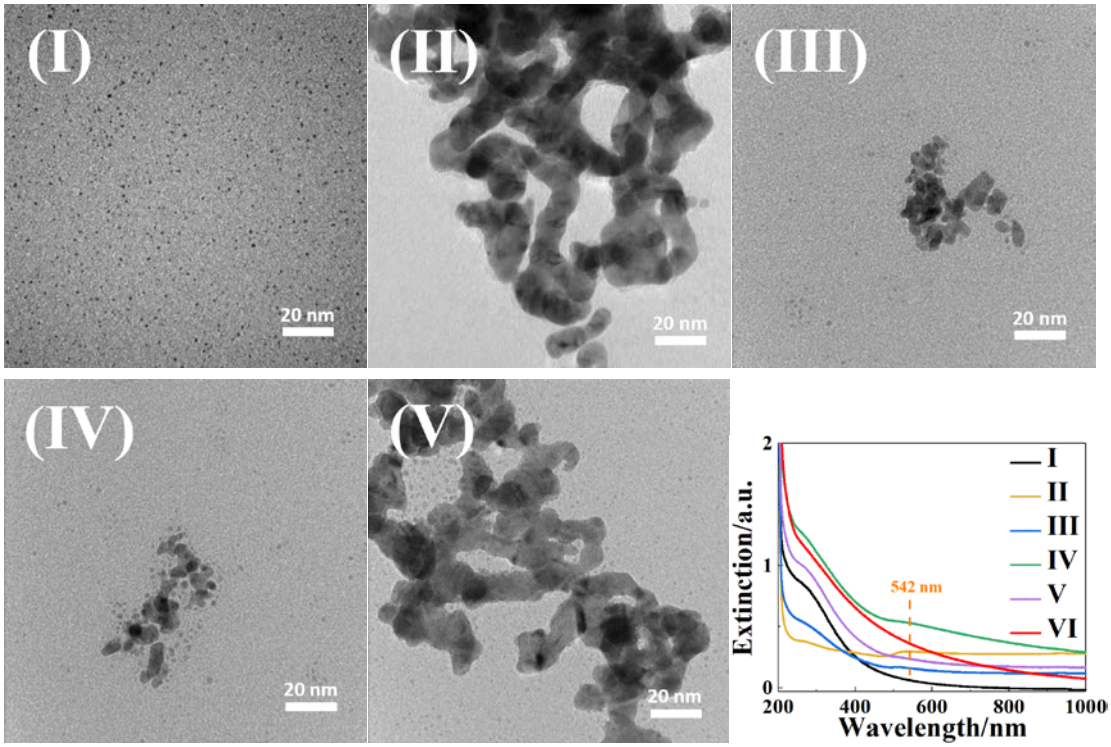

Figure S2. TEM images and UV-Vis spectra of samples shown in Table S1: Pt (I), Au (II), PEG-Pt NPs dispersion mixed with PEG-Au NPs dispersion (III), sputter Pt target for 30 min followed by sputtering Au target for 30 min (IV), and sputter Au target for 30 min followed by sputtering Pt target for 30 min (V). TEM image of Pt/Au NPs (VI) prepared by co-sputtering is shown in Figure 2 in the main text.

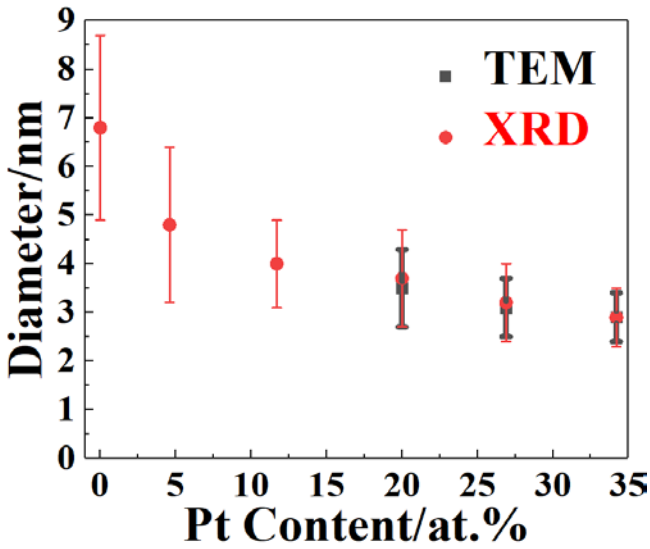

Figure S3. Changes in crystallite diameter and particle diameter with changes in proportion of Pt (at.%) in NPs. Crystallite diameter of Pt/Au NPs, calculated based on the measured 2θ values of (111) and (200) diffraction lines. Samples with Pt < 20 at.% show severe agglomerate structures as for pure Au shown in Figure S2.

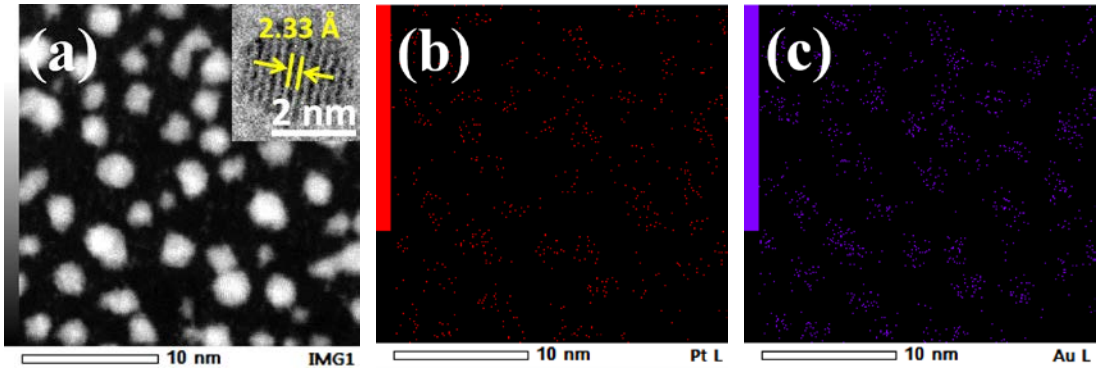

Figure S4. HAADF-STEM image of NPs obtained by sputtering onto TEM grid under sputtering current of Au (50 mA) / Pt (50 mA) for 5 s (a), STEM EDX-mapping images of Pt (b) and Au (c). ((a)-(c) were obtained by JEOL JEM-ARM200F, operating at 200 kV)

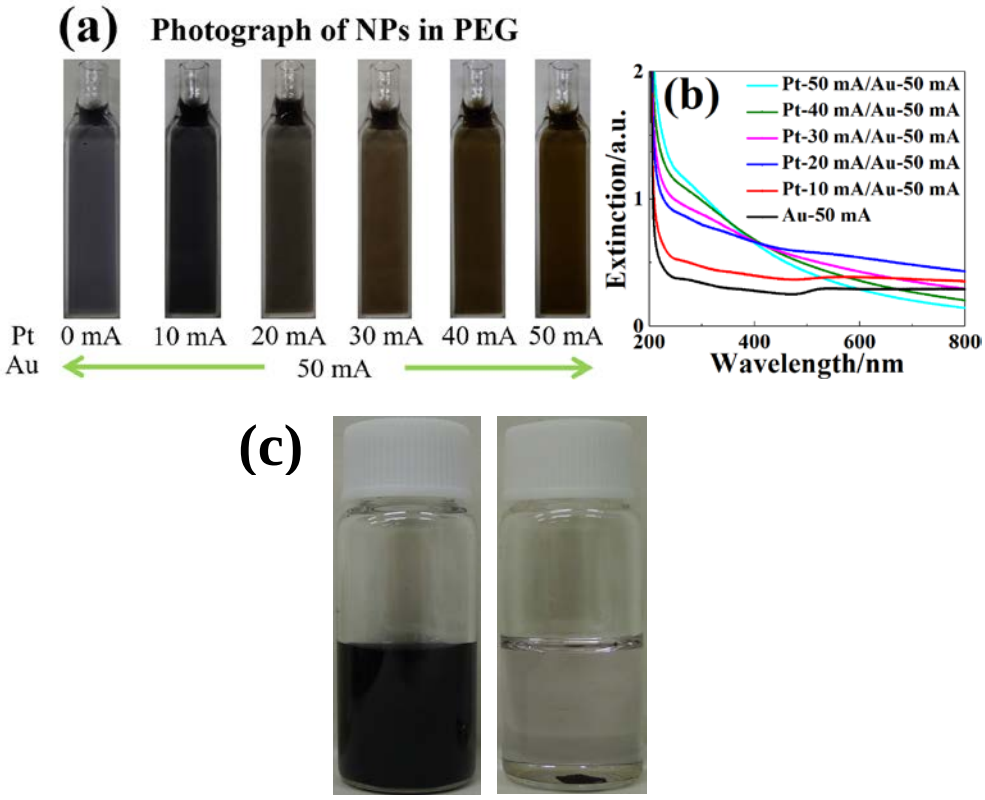

Figure S5. Photograph of PEG-dispersions produced by co-sputtering onto PEG using different sputtering current of Pt target from 0-50 mA while keeping sputtering current of Au target constant at 50 mA under ambient light (a), and corresponding UV-vis absorption spectra of PEG NPs dispersions (b), and photograph of PEG-Au NPs dispersions obtained by sputtering Au target onto PEG using sputtering current of 50 mA, right after synthesis (left) and after keeping in the dark at room temperature for 6 weeks (right) (c).

Table S2. XPS data on the atomic percentage of Pt in the sputtered materials prepared by sputtering onto Si wafer for 30min under sputtering current of Pt target from 0 to 50 mA while keeping sputtering current of Au target constant at 50 mA.

Pt current / mA	0	10	20	30	40	50
Pt at. %, XPS	0.0	3.5	9.8	18.2	27.3	34.1 (33.8*)

*The Pt/Au atomic ratio of the sample generated by sputtering onto PEG at the sputtering current of Pt-50 mA/Au-50 mA was also checked by XPS.

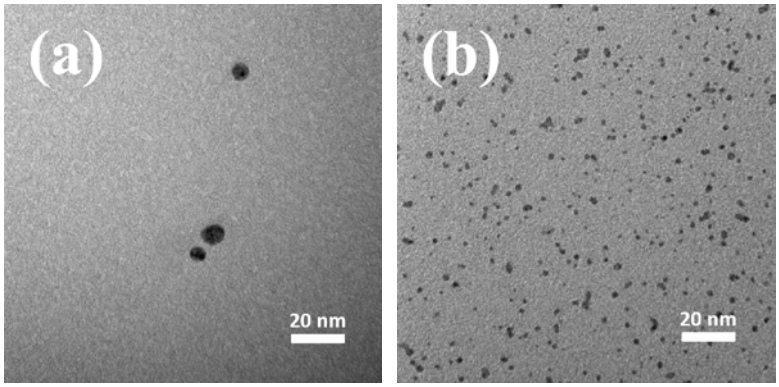

Figure S6. TEM images of PEG-NPs dispersion obtained by Ag-50 mA (a) and Pt-50 mA/Ag-50 mA (b) sputtering onto PEG.

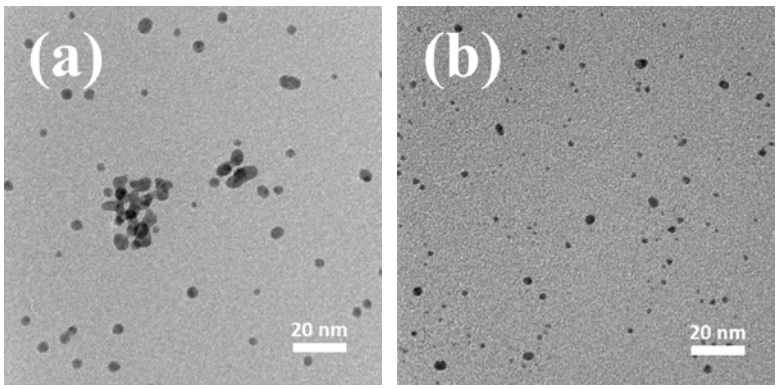

Figure S7. TEM images of PEG-NPs dispersion obtained by Ag-50 mA/Au-50 mA (a) and Cu-50 mA/Au-50 mA (b) sputtering onto PEG.

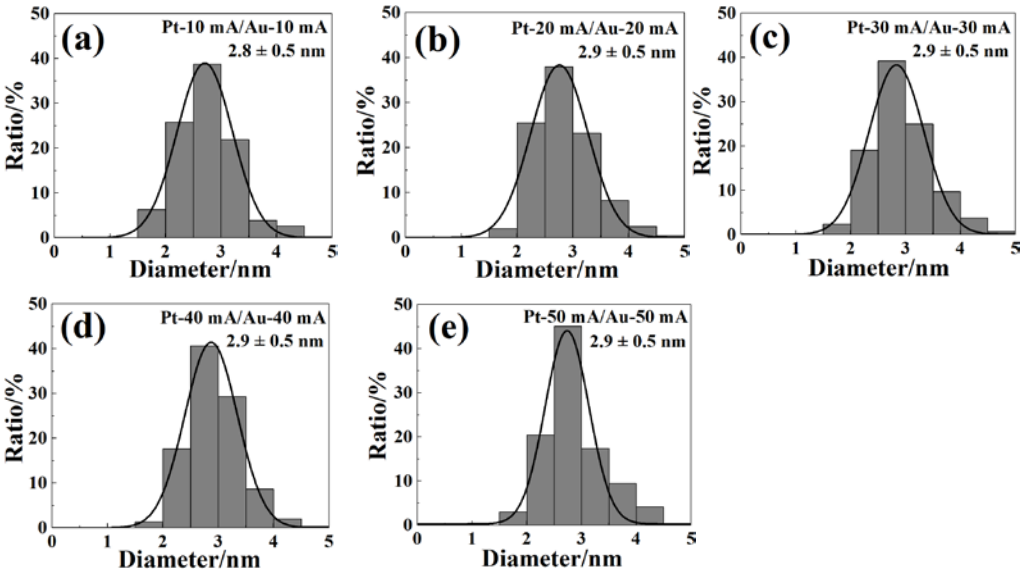

Figure S8. TEM images of PEG-NPs dispersion generated by sputtering onto PEG under various sputtering current of Pt/Au target from 10 mA/10 mA (a), 20 mA/20 mA (b), 30 mA/30 mA (c), and 40 mA/40 mA (d).

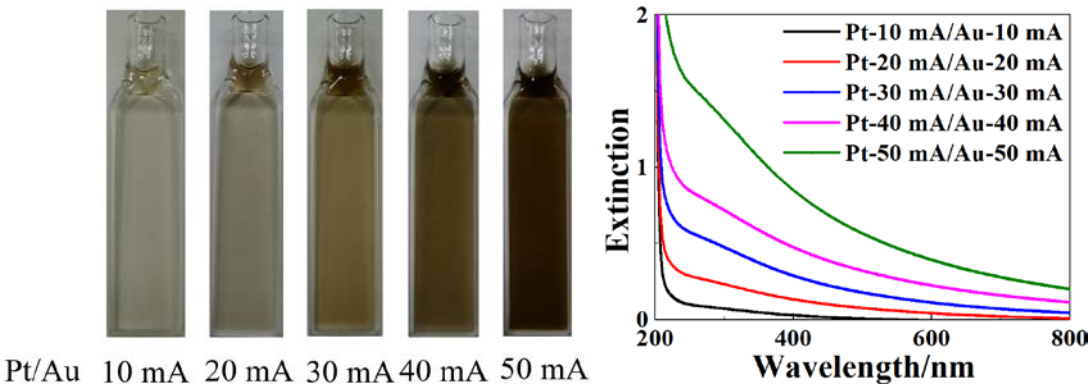

Figure S9. Photograph of PEG-NPs dispersions obtained by sputtering onto PEG under various sputtering current of Pt/Au target from 10 mA/10 mA-50 mA/50 mA. under room light (left), and corresponding UV-Vis absorption spectra of PEG-NPs dispersions (right).

Table S3. Deposition amount of Pt/Au in PEG sputtered using different sputtering current and time.

Sputtering current of Pt-Au (mA-mA)	10-10	20-20	30-30	40-40	50-50
Deposition amount for 3 min sputtering (mg) (Particle size (nm))					0.6 (2.5±0.5)
Deposition amount for 30 min sputtering (mg) (Particle size (nm))	0.5 (2.8±0.5)	1.5 (2.9±0.5)	2.6 (2.9±0.5)	4.1 (2.9±0.5)	5.6 (2.9±0.5)
Deposition amount for 60 min sputtering (mg) (Particle size (nm))					11.2 (2.9±0.9)
Deposition amount for 90 min sputtering (mg) (Particle size (nm))					16.8 (3.7±1.5)
Deposition amount for 240 min sputtering (mg) (Particle size (nm))		12.0 (2.9±0.8)			44.8 (4.5±1.8)

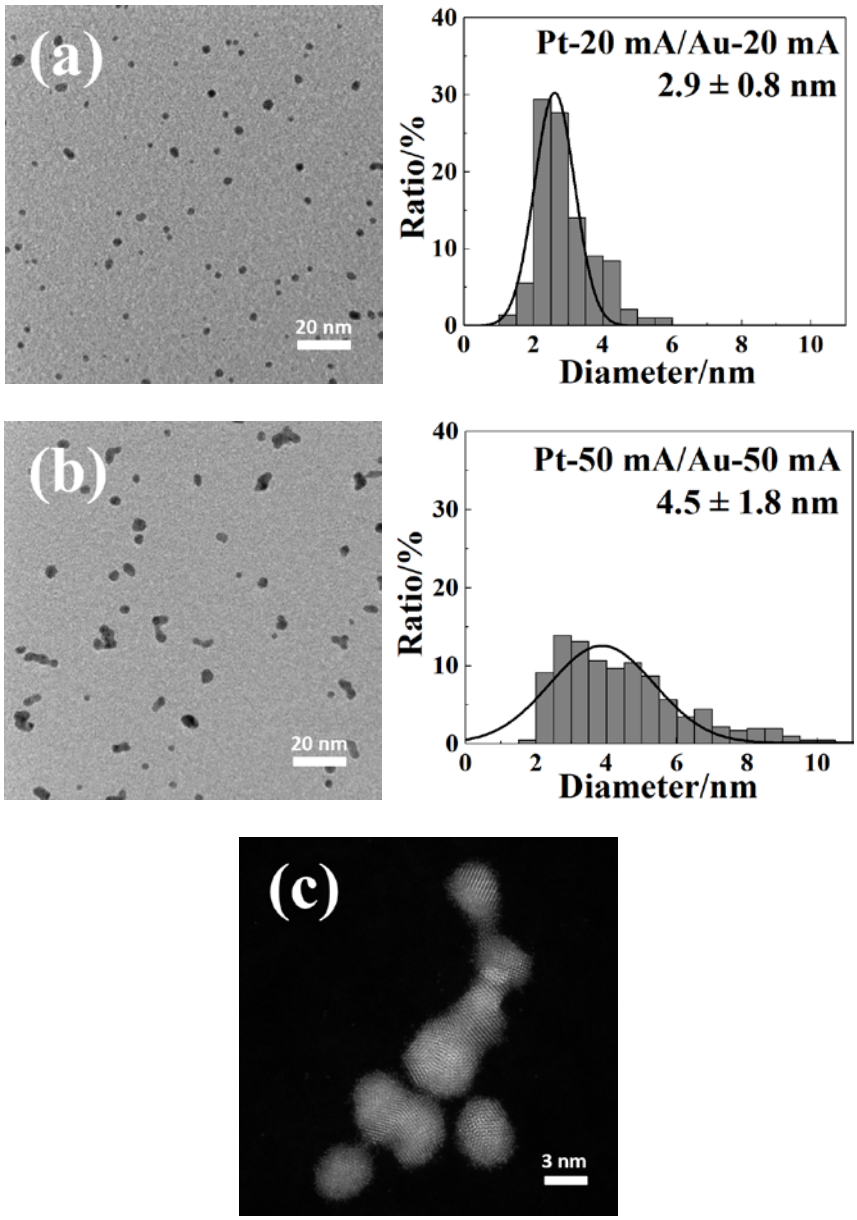

Figure S10. TEM images of PEG-NPs dispersion produced by sputtering onto PEG for 4 h under sputtering current of Pt-20 mA/Au-20 mA (a), Pt-50 mA/Au-50 mA (b), and corresponding STEM image (c).

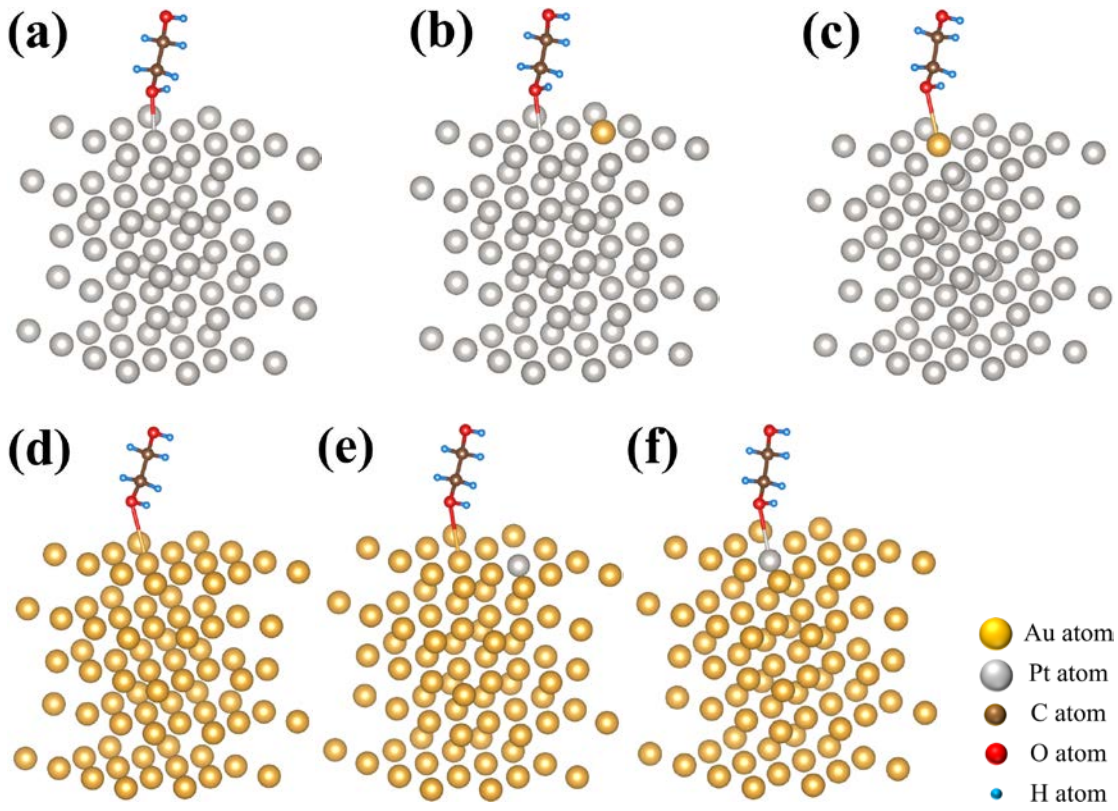

Figure S11. Final relaxed structure for PEG molecule adsorbed on Pt in Pt (111) surface (a), Pt in Pt/Au (111) surface (one Pt atom substituted by one Au atom in structure (a)) (b), Au in Pt/Au (111) surface (one Pt atom substituted by one Au atom in structure (a) (c), Au in Au (111) surface (d), Au in Pt/Au (111) surface (one Au atom substituted by one Pt atom in structure (d)) (e), and Pt in Pt/Au (111) surface (one Au atom substituted by one Pt atom in structure (d)) (f).

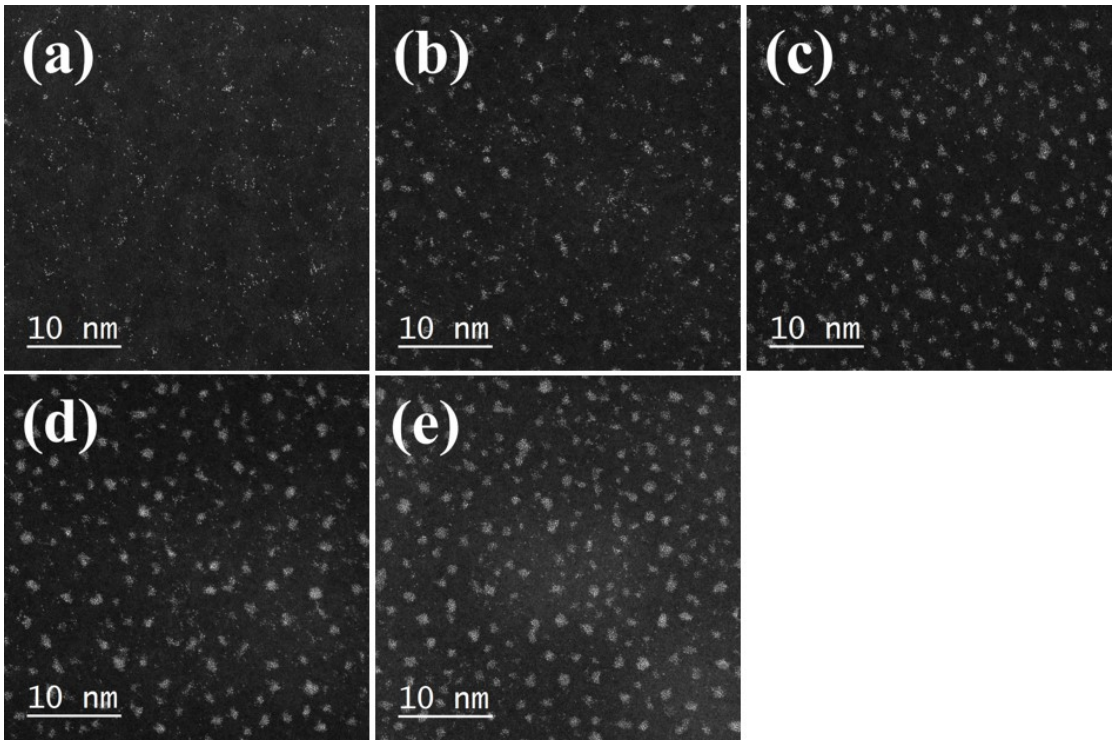

Figure S12. STEM images of Pt NPs obtained by sputtering onto TEM grid for 1s under various sputtering current of Pt target from 10 (a), 20 (b), 30 (c), 40 (d) and 50 mA (e).

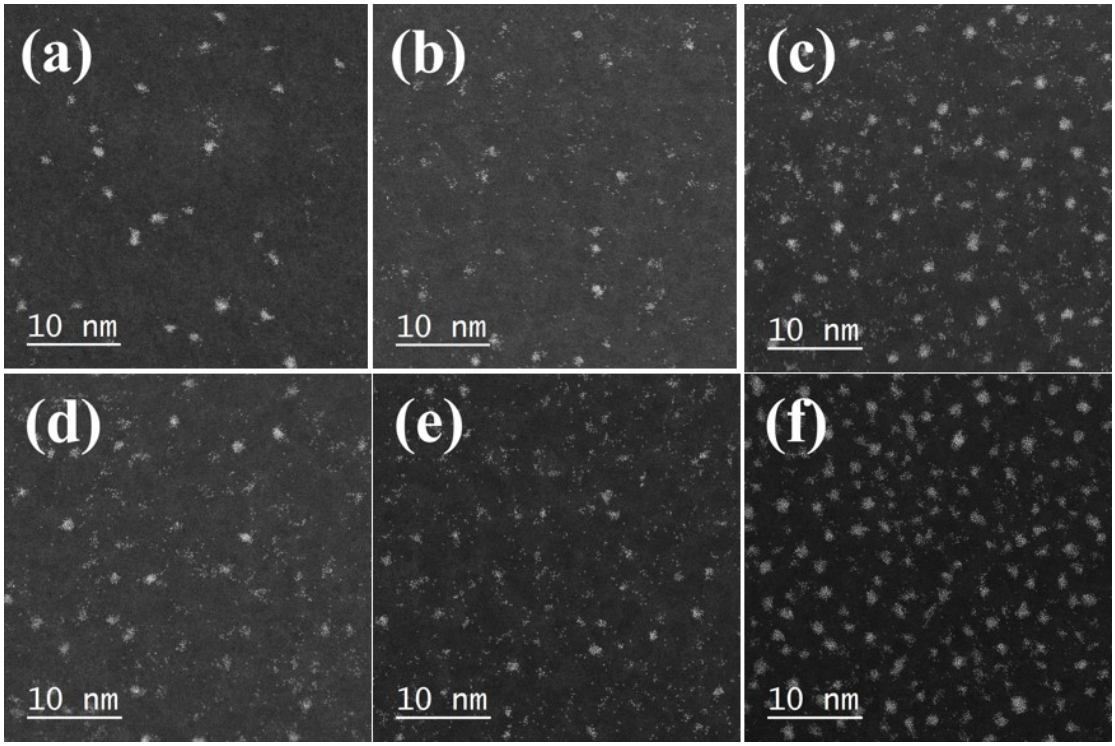

Figure S13. STEM images of NPs obtained by sputtering onto TEM grid for 1s under various sputtering current of Pt target from 0 (a), 10 (b), 20 (c), 30 (d), 40 (e) and 50 mA (f) while keeping sputtering current of Au target constant at 20 mA.

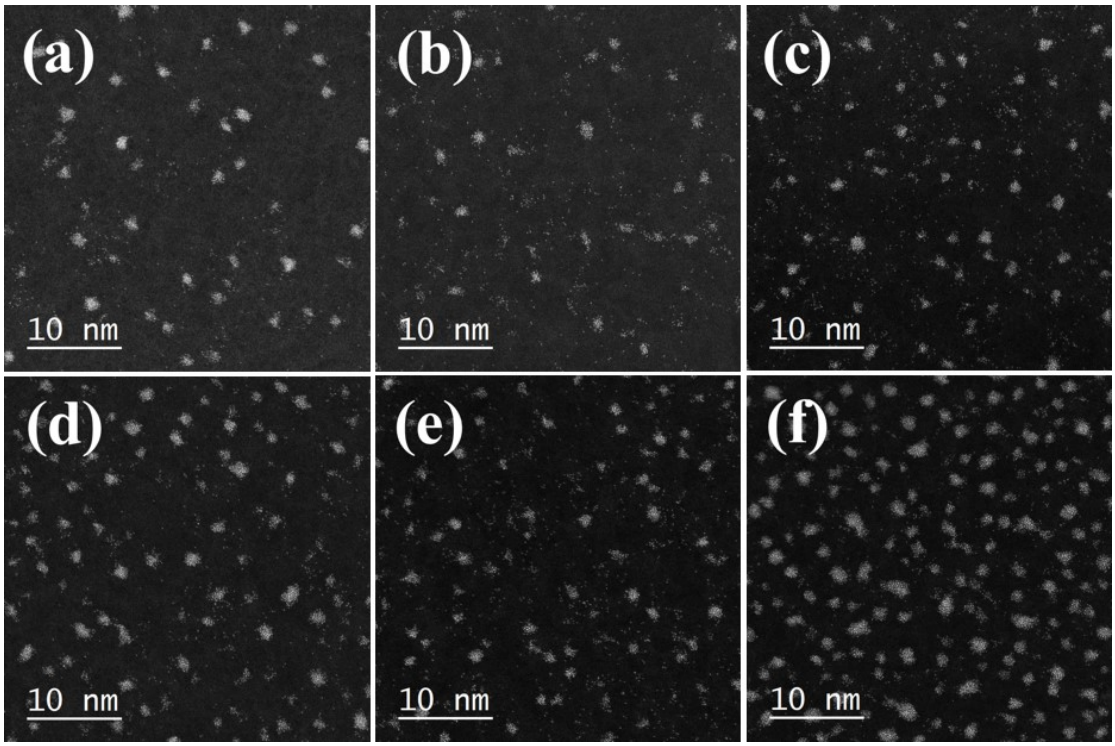

Figure S14. STEM images of NPs obtained by sputtering onto TEM grid for 1s under various sputtering current of Pt target from 0 (a), 10 (b), 20 (c), 30 (d), 40 (e) and 50 mA (f) while keeping sputtering current of Au target constant at 40 mA.

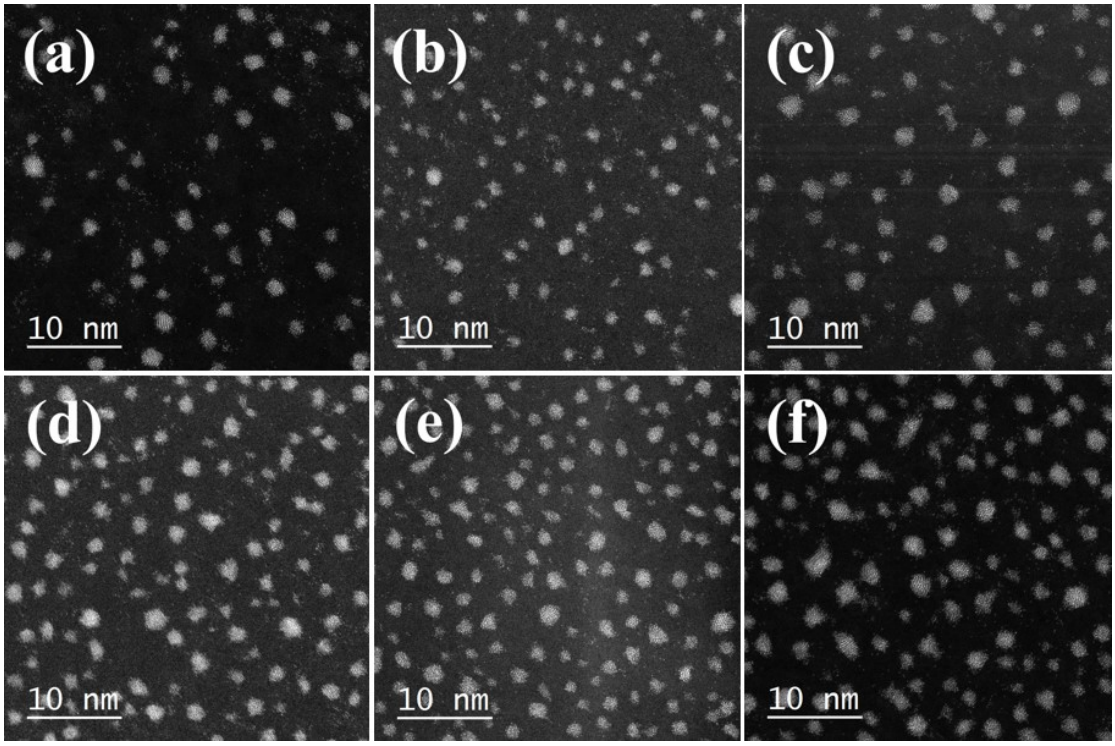

Figure S15. STEM images of NPs obtained by sputtering onto TEM grid for 1s under various sputtering current of Pt target from 0 (a), 10 (b), 20 (c), 30 (d), 40 (e) and 50 mA (f) while keeping sputtering current of Au target constant at 50 mA.

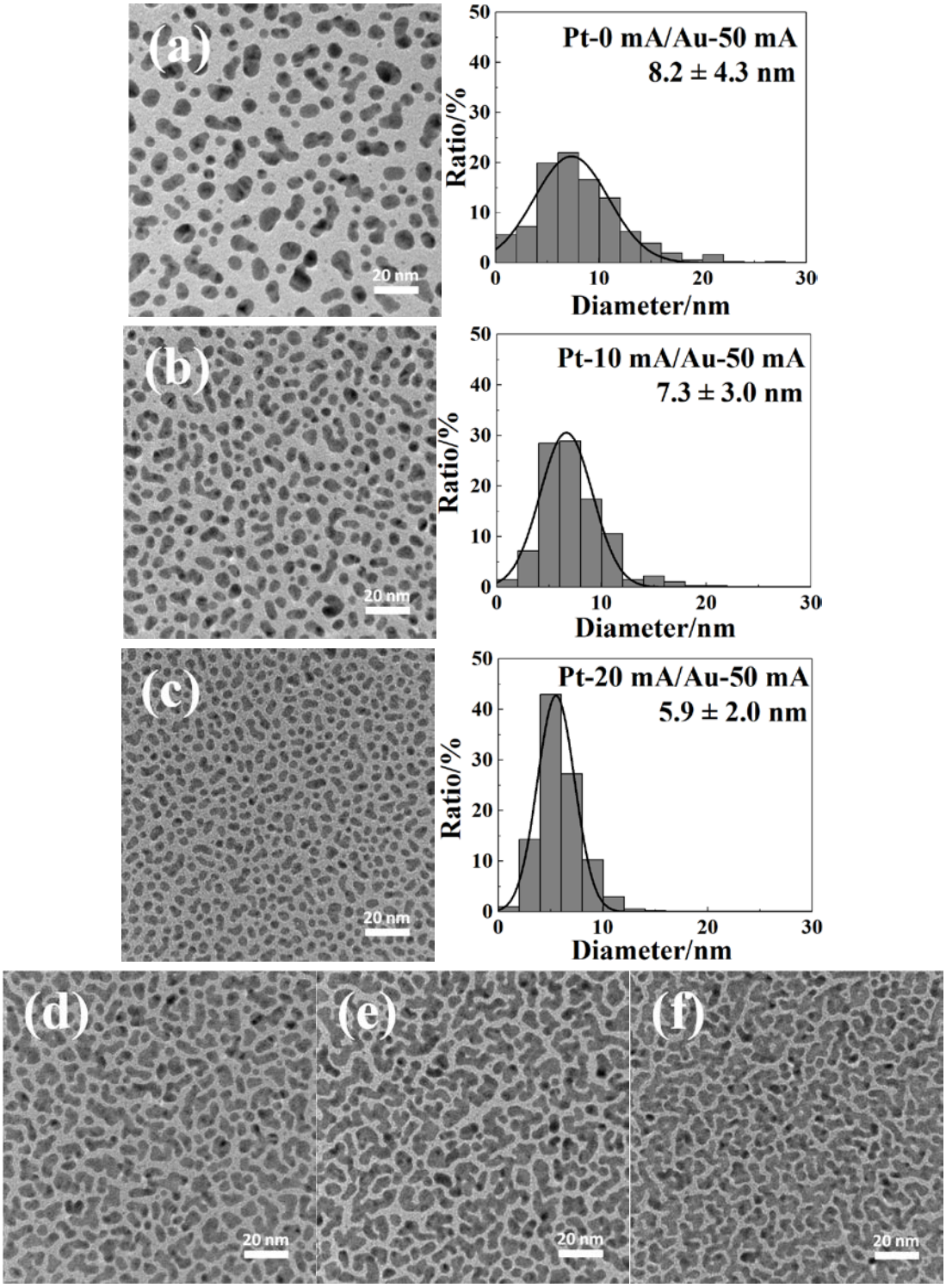

Figure S16. TEM images of NPs obtained by sputtering onto TEM grid for 30 s under various sputtering current of Pt target from 0 (a), 10 (b), 20 (c), 30 (d), 40 (e) and 50 mA (f) while keeping sputtering current of Au target constant at 50 mA.

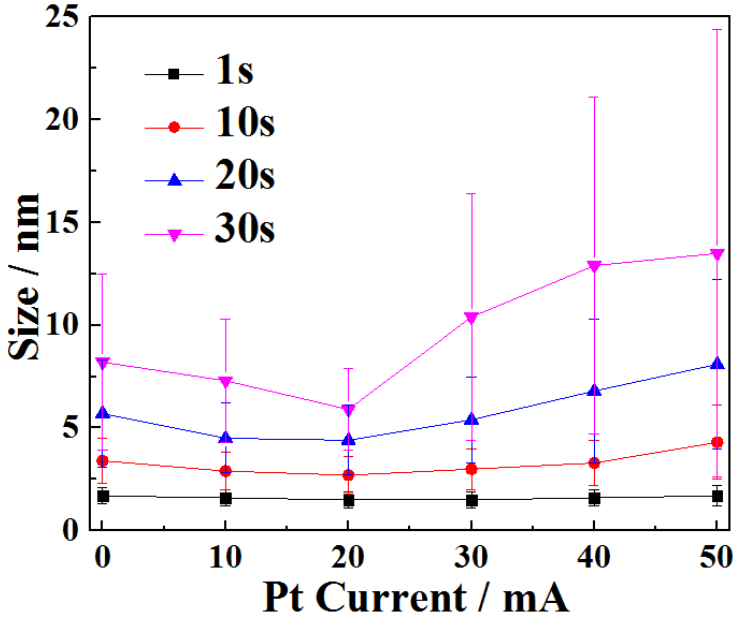

Figure S17. Relation between particle size and sputtering current of Pt sputtered onto TEM grid under the 50 mA of Au for 1 s (black), 10 s (red), 20 s (blue) and 30 s (pink).