Pt/Ag Solid Solution Alloy Nanoparticles in Miscibility Gap Synthesized by Co-Sputtering onto Liquid Polymer

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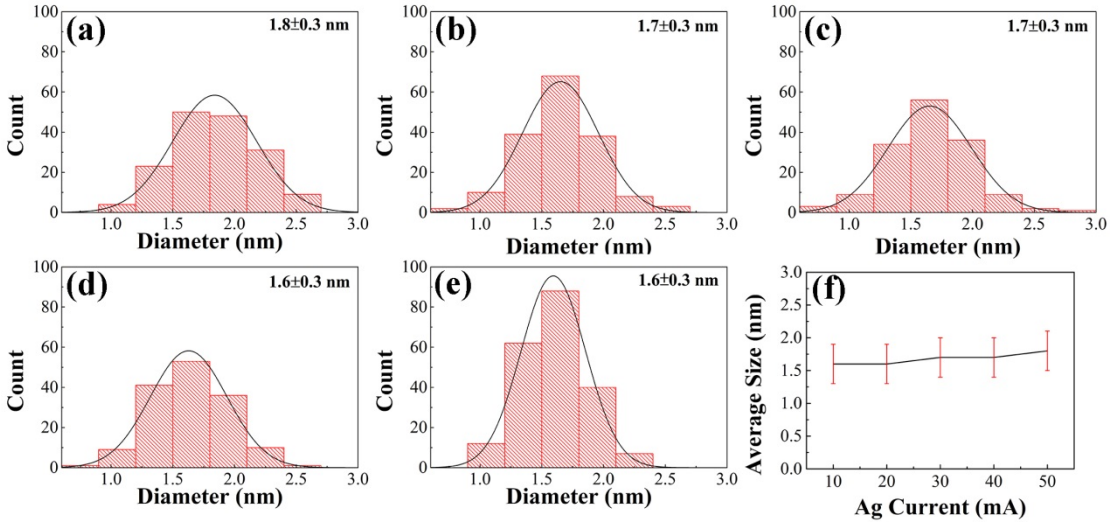

Figure S1. Size distributions of Pt/Ag samples shown in Figure 1: (a) Pt50Ag50, (b) Pt50Ag40, (c) Pt50Ag30, (d) Pt50Ag20, (e) Pt50Ag10. (f) Summary of the particle sizes shown in a-e with the error bar showing the standard deviation of particle sizes.

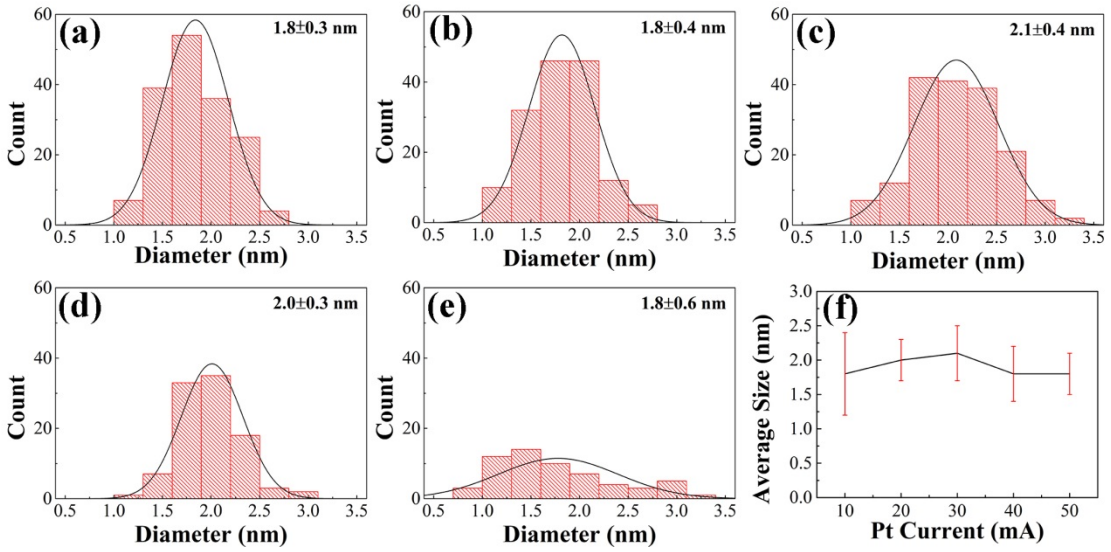

Figure S2. Size distributions of Pt/Ag samples shown in Figure 1: (a) Pt50Ag50, (b) Pt40Ag50, (c) Pt30Ag50, (d) Pt20Ag50, (e) Pt10Ag50. (f) Summary of the particle sizes shown in a-e with the error bar showing the standard deviation of particle sizes.

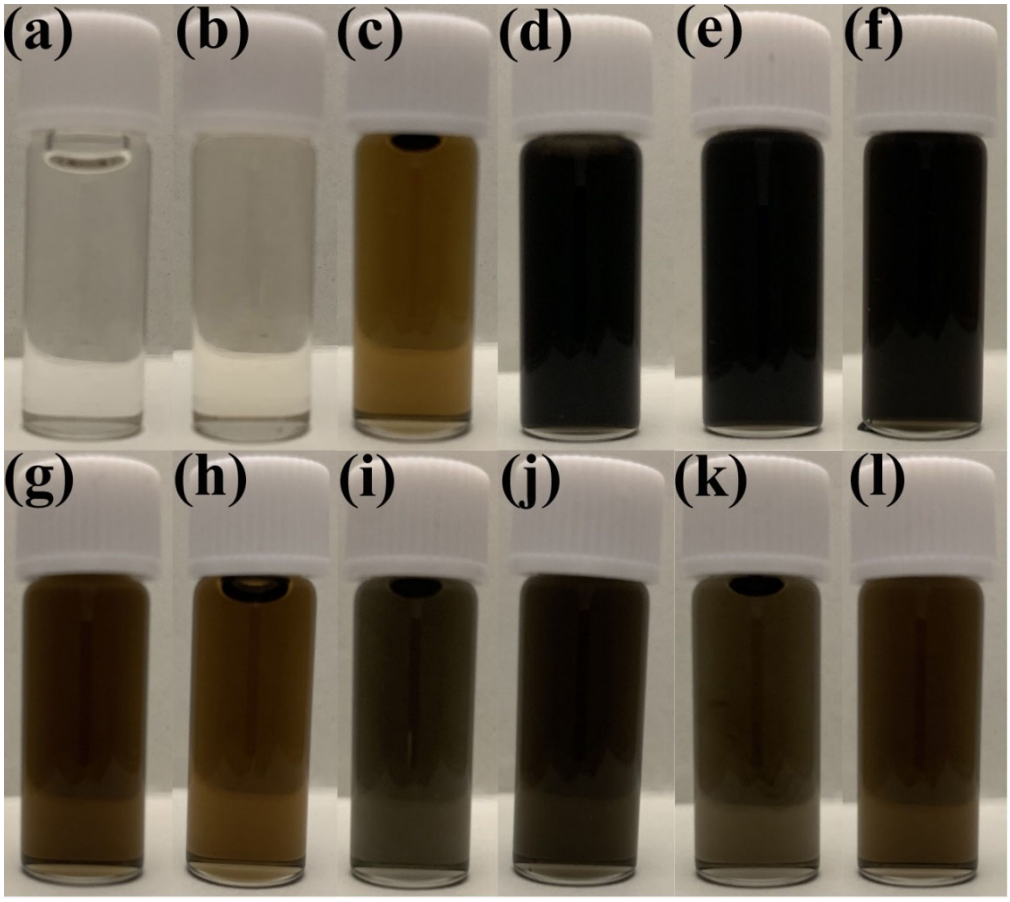

Figure S3. The photographs of samples (a) Ag50, (b) Pt10, (c) Pt50, (d) Pt50Ag50, (e) Pt50Ag40, (f) Pt50Ag30, (g) Pt50Ag20, (h) Pt50Ag10, (i) Pt40Ag50, (j) Pt30Ag50, (k) Pt20Ag50 and (l) Pt10Ag50.

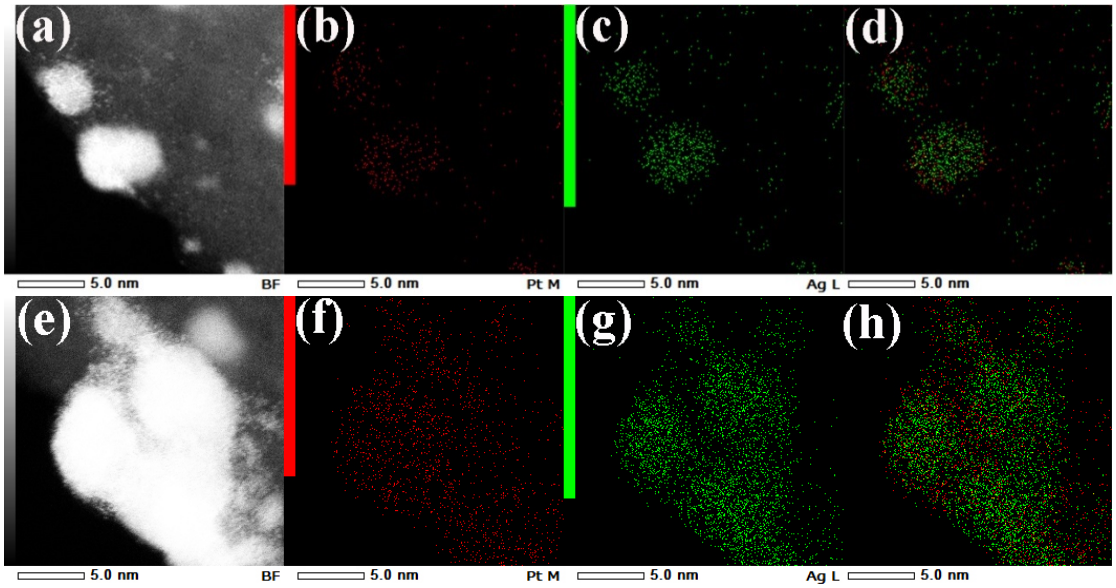

Figure S4. (a, e) HAADF and (b-d, f-h) STEM-EDX mapping images of Pt10Ag50 sample with (b, f) Pt M, (c, g) Ag L, and (d, h) overlapping image of Pt and Ag. The average Pt atomic composition measured from STEM-EDX is 19.8 ± 12.2 at %. Figures (e-h) show an agglomeration of big and small Pt10Ag50 NPs in PEG where the big particle contain more Ag.

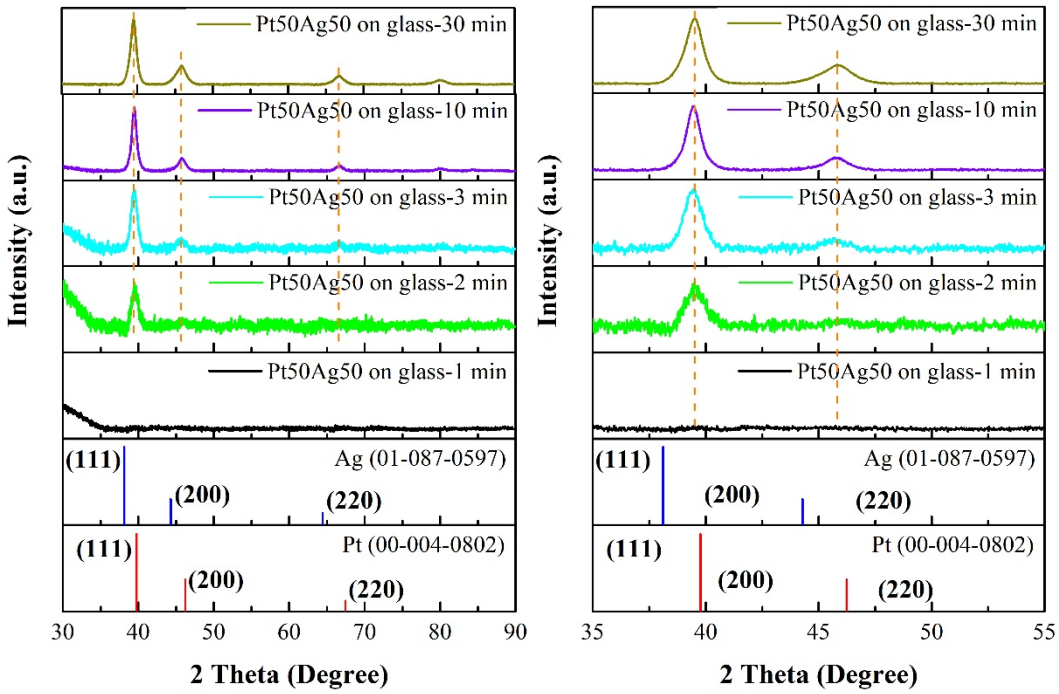

Figure S5. (left) XRD results of Pt50Ag50 samples that sputtered on glass slides with a sputtering time of 1, 2, 3, 10, and 30 min. (right) Enlarged XRD patterns of samples shown on the left.

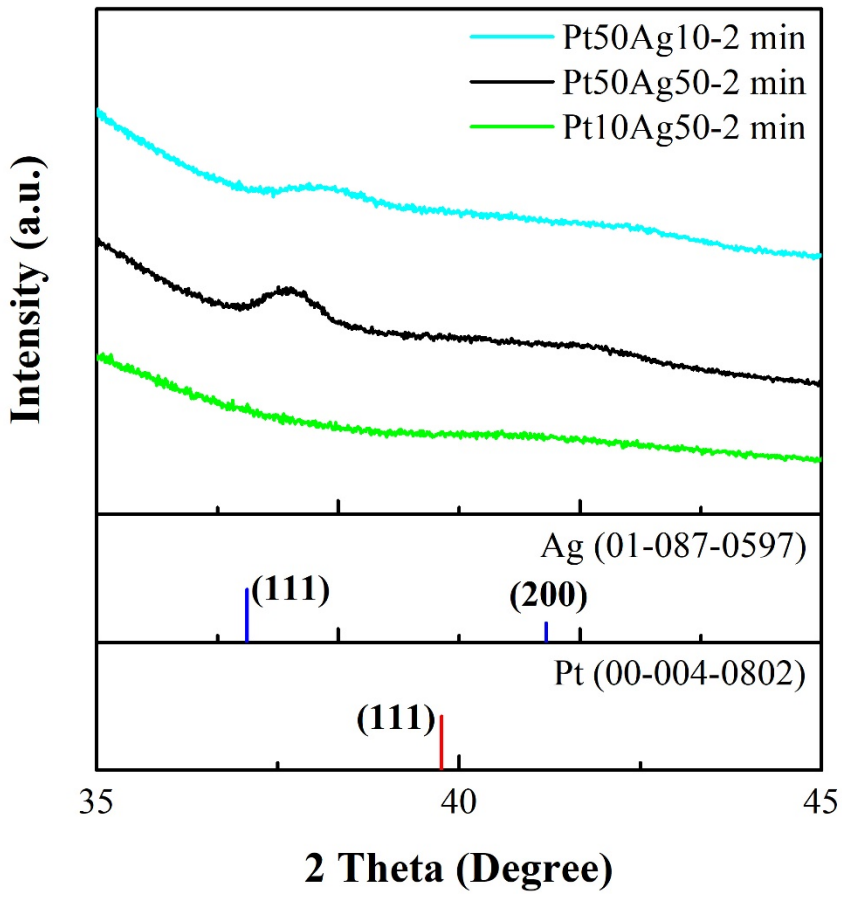

Figure S6. XRD patterns of Pt50Ag50, Pt50Ag10, and Pt10Ag50 samples prepared by co-sputtering on the glass slides for 2 min.

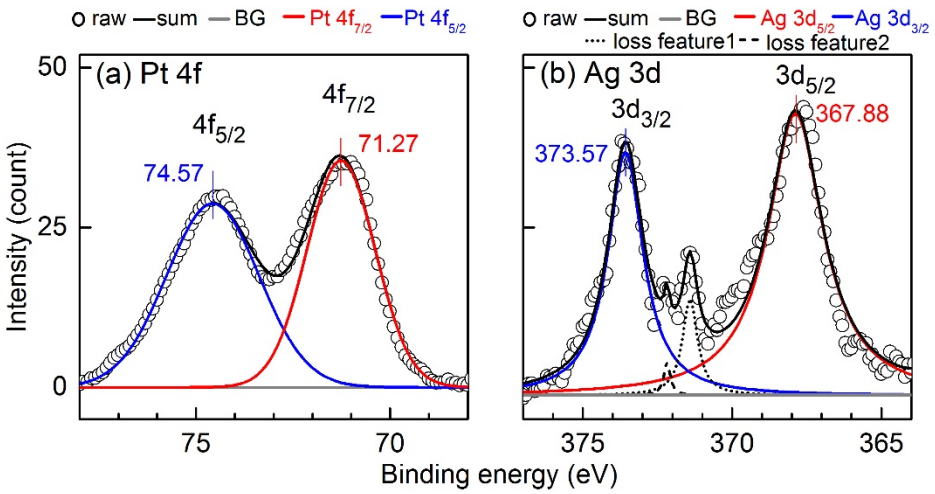

Figure S7. XPS narrow scan spectra of (a) Pt 4f and (b) Ag 3d regions of Pt50Ag50 NPs sputtered onto PEG. Single peak is used to fit each XPS spin-orbit component in the spectra. Larger full-width at half maximum (FWHM) of the high energy component is observed for Pt and reversed phenomenon is observed for Ag. The spectrum near 370 eV of Ag 3d could not be fit well with a single peak.

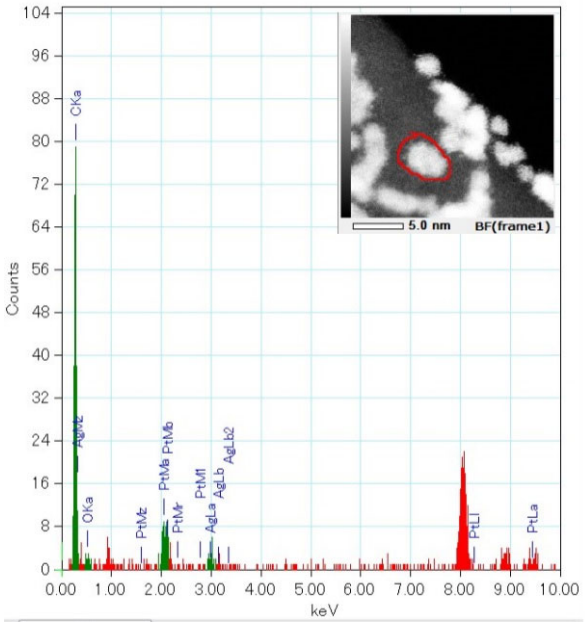

Figure S8. EDX spectra for single particle (in red circle in the STEM-HAADF image) shown in STEM-EDX mapping for Pt50Ag50 in PEG in Figure 9. The signals of Pt Mα and Ag Lα are distinguished from the background.

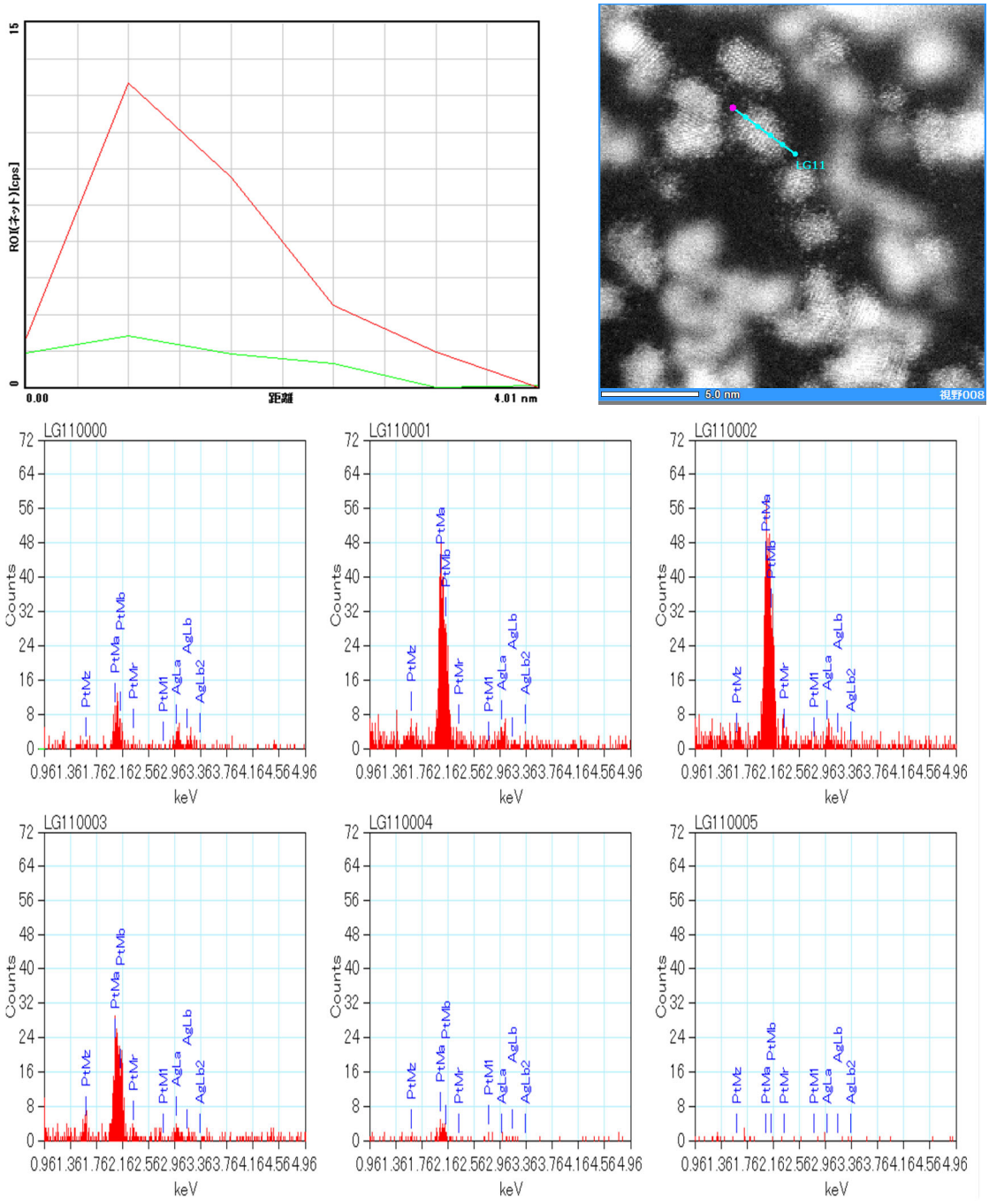

Figure S9. (top-left) STEM-EDX line profile for Pt Mα (red) and Ag Lα (green) of a Pt50Ag50 NP sputtered onto PEG along the green line in the HAADF image (top-right). (bottom) EDX spectra collected at the points shown in the line in HAADF image for taking the line profile.

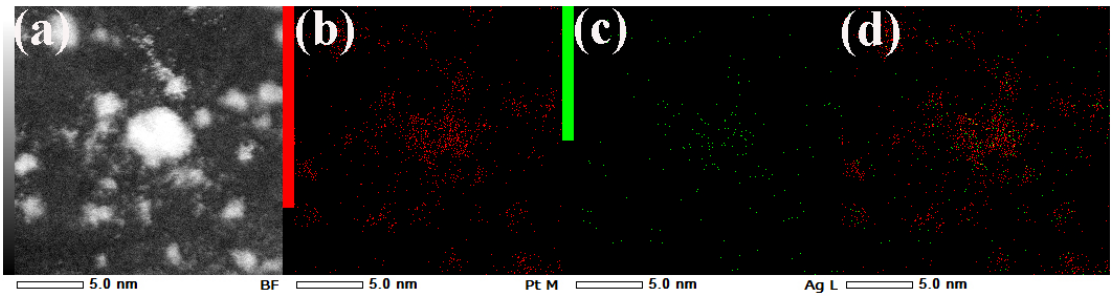

Figure S10. (a) HAADF and (b-d) mapping images of Pt50Ag10 sample with (b) Pt M, (c) Ag L, and (d) overlapping image of Pt and Ag. The average Pt atomic composition of this sample measured from STEM-EDX results is 90.5 at %.

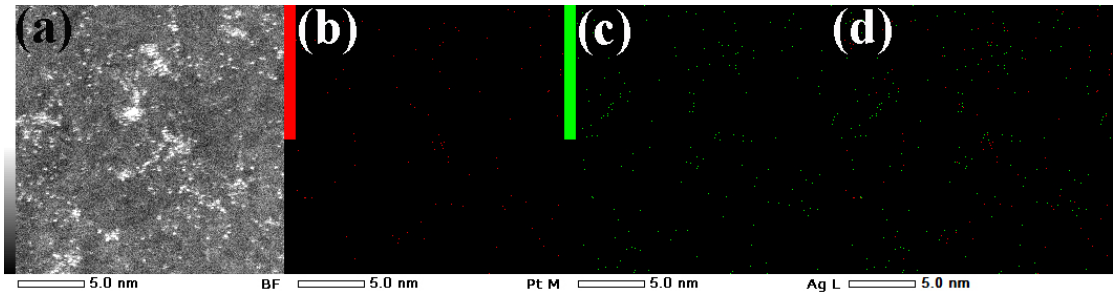

Figure S11. (a) HAADF and (b-d) STEM-EDX mapping images of Pt10Ag50 sample sputtered on TEM grid for 5 s with (b) Pt M, (c) Ag L, and (d) overlapping image of Pt and Ag. The average Pt atomic composition measured from STEM-EDX results is of 13.3 ± 8.0 at %.

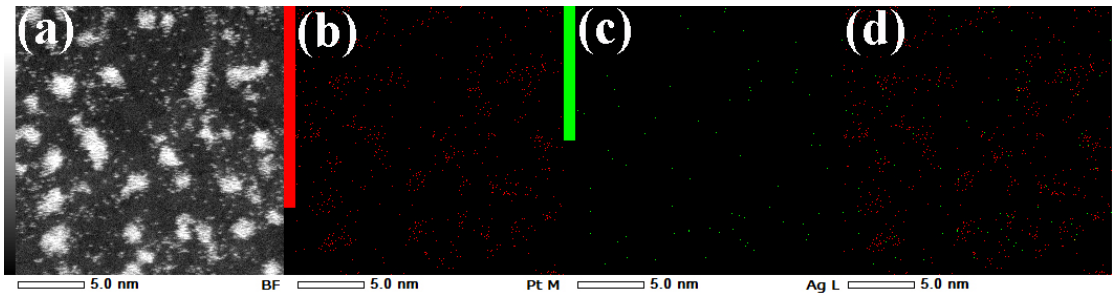

Figure S12. (a) HAADF and (b-d) STEM-EDX mapping images of Pt50Ag10 sample sputtered on TEM grid for 5 s with (b) Pt M, (c) Ag L, and (d) overlapping image of Pt and Ag. The average Pt atomic composition measured from STEM-EDX results is of 83.8 ± 11.2 at %.

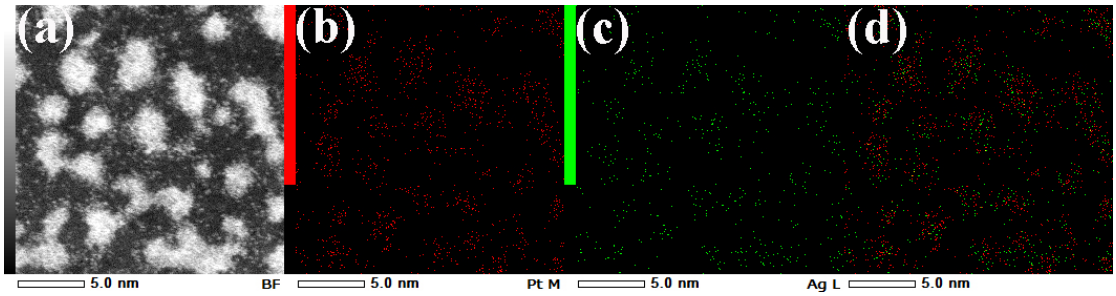

Figure S13. (a) HAADF and (b-d) STEM-EDX mapping images of Pt50Ag50 sample sputtered on TEM grid for 5 s with (b) Pt M, (c) Ag L, and (d) overlapping image of Pt and Ag. The average Pt atomic composition measured from STEM-EDX results is of 50.3 ± 13.0 at %.

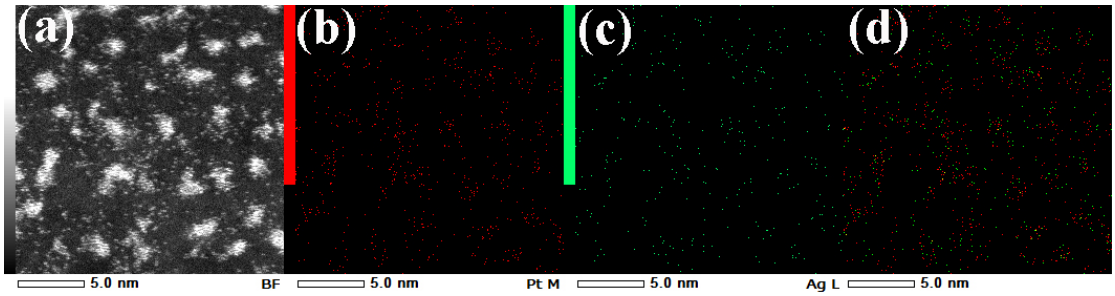

Figure S14. (a) HAADF and (b-d) STEM-EDX mapping images of Pt50Ag50 sample sputtered on TEM grid for 1 s with (b) Pt M, (c) Ag L, and (d) overlapping image of Pt and Ag. The average Pt atomic composition measured from STEM-EDX results is of 50.3 ± 16.3 at %.

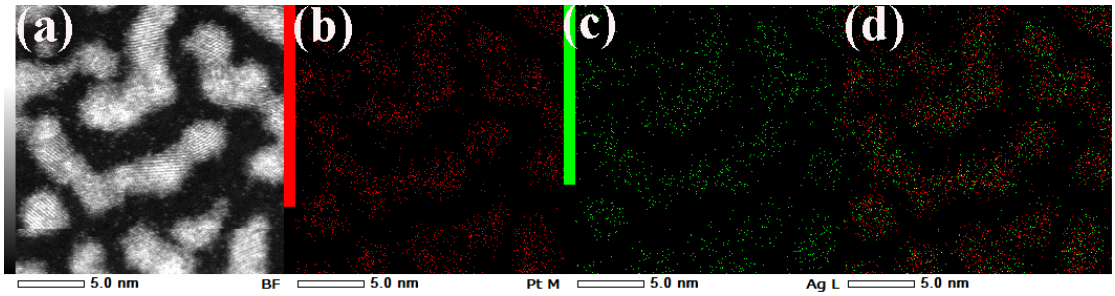

Figure S15. (a) HAADF and (b-d) STEM-EDX mapping images of Pt50Ag50 sample sputtered on TEM grid for 20 s with (b) Pt M, (c) Ag L, and (d) overlapping image of Pt and Ag. The average Pt atomic composition measured from STEM-EDX results is of 44.8 ± 12.7 at %.

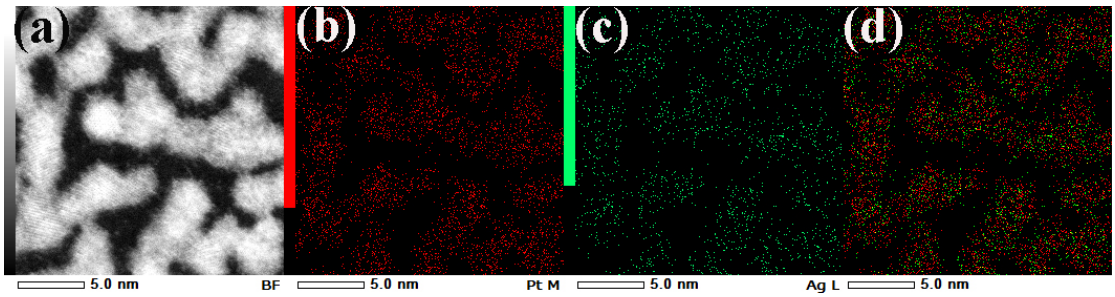

Figure S16. (a) HAADF and (b-d) STEM-EDX mapping images of Pt50Ag50 sample sputtered on TEM grid for 30 s with (b) Pt M, (c) Ag L, and (d) overlapping image of Pt and Ag. The average Pt atomic composition measured from STEM-EDX results is of 49.3 ± 10.6 at %.

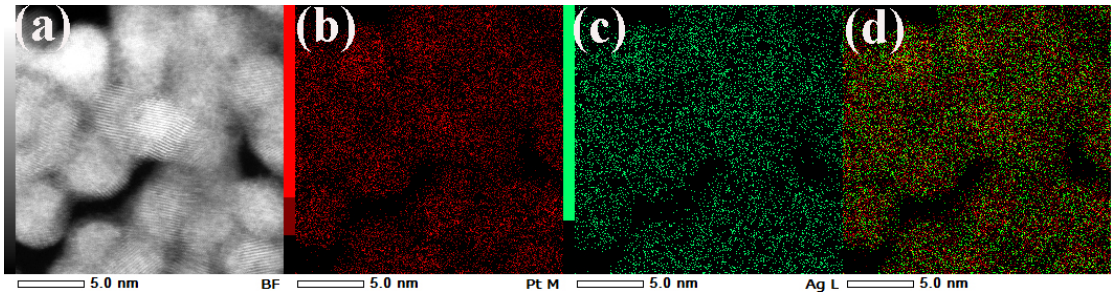

Figure S17. (a) HAADF and (b-d) STEM-EDX mapping images of Pt50Ag50 sample sputtered on TEM grid for 2 min with (b) Pt M, (c) Ag L, and (d) overlapping image of Pt and Ag. The average Pt atomic composition measured from STEM-EDX results is of 47.3 ± 3.4 at %.

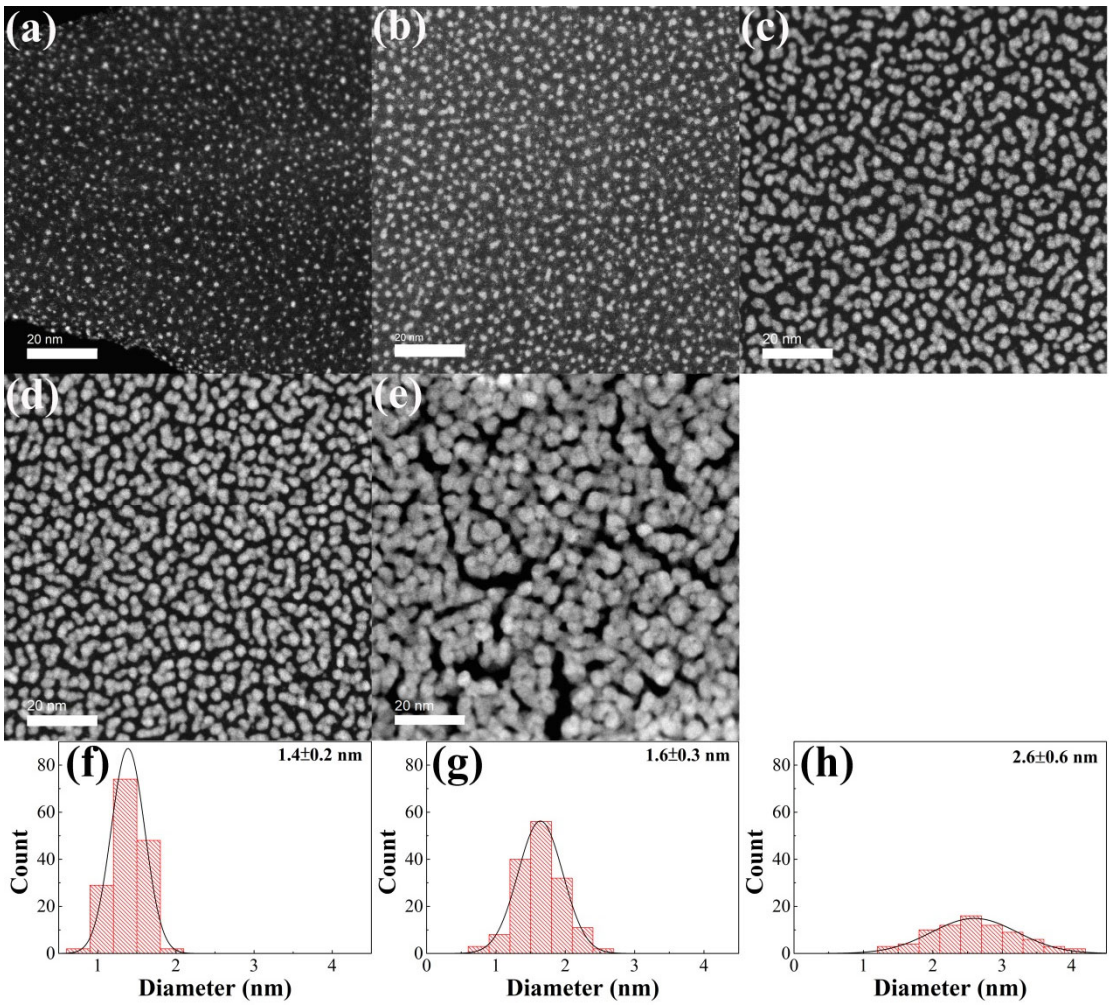

Figure S18. STEM-HAADF images of Pt50Ag50 sputtered on the TEM grids with a sputtering time of (a) 1 s, (b) 5 s, (c) 20 s and (d) 30 s, and (e) 2 min. The scale bar is 20 nm. The size distributions are given for NPs obtained by sputtering for (f) 1s, (g) 5 s, and (h) 20 s. The average sizes of the samples are 1.4, 1.6, and 2.6 nm, respectively. Severe aggregation, necking, and growth to film like structure were observed for samples obtained after sputtering for 30 s and 2 min.

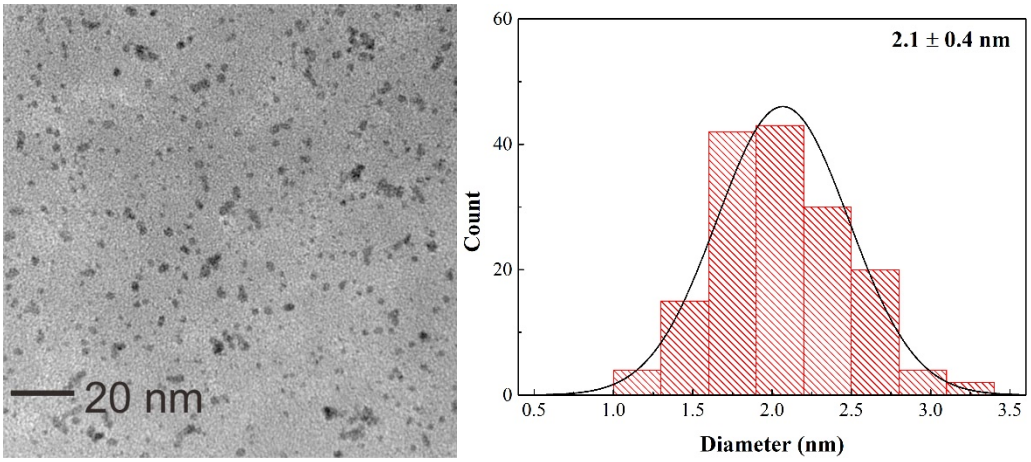

Figure S19. TEM image and size distribution of Pt50Ag50 NPs sputtered onto silicone oil for 30 min.

Table S1. Amount of sputtered Pt and Ag for 1 h sputtering* on Al foil

Sample	Sputtering current (mA)		Amount of Pt	Amount of Ag
	Pt target	Ag target		
Pt50	50	0	3.12 mg (16.0 µmol)	
Pt10	10	0	0.34 mg (1.7 µmol)	
Ag50	0	50		1.36 mg (12.6 µmol)
Ag10	0	10		0.17 mg (1.6 µmol)

*The sputtering was conducted using only one head of the double head target

Table S2. XPS peak position of Ag 3d and Pt 4f core levels of NPs sputtered on Si

Sample	Peak position (eV)			
	Pt $4f_{7/2}$	Pt 4f5/2	Ag $3d_{5/2}$	Ag $3d_{3/2}$
Pt50Ag10	71.4	74.7	367.6	373.6
Pt50Ag50	70.9	74.2	367.6	373.6
Pt10Ag50	70.9	74.2	368.0	374.0
Pt50	71.6	74.9	-	-
Ag50	-	-	368.2	374.2