Supplementary Information (SI) for Nanoscale.
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Supporting Information

Portfolio of colloidally stable gold-gold sulfide nanoparticles and their use in broad-band photoacoustic imaging

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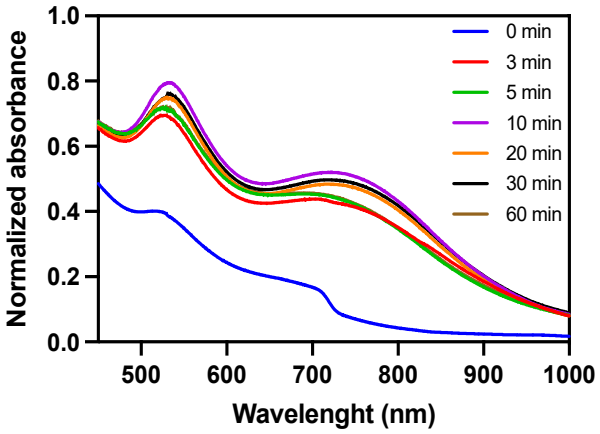


Figure S1 Time dependent absorbance spectra of the reaction mixture following the progress of GGS formation and growth in the absence of a coating material.

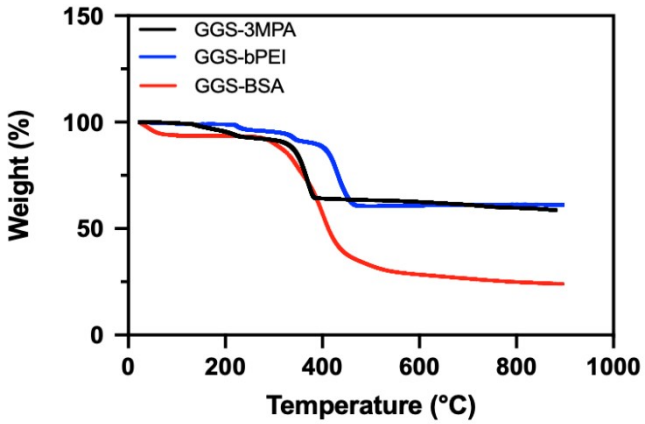


Figure S2 Thermogravimetric analysis of GGS-3MPA, GGS-bPEI, and GGS-BSA.

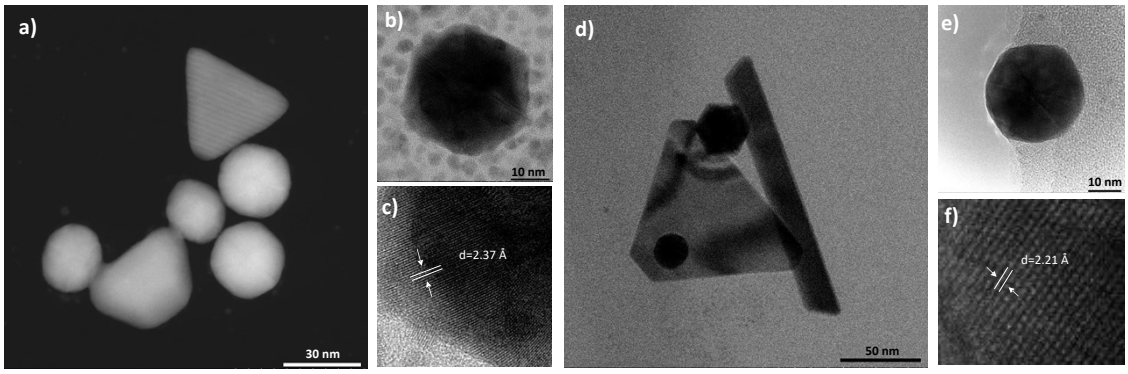


Figure S3 TEM images of GGS-bPEI at different magnifications (a-b) and lattice spacing (c). TEM images of GGS-BSA (d-e) and lattice spacing (f).

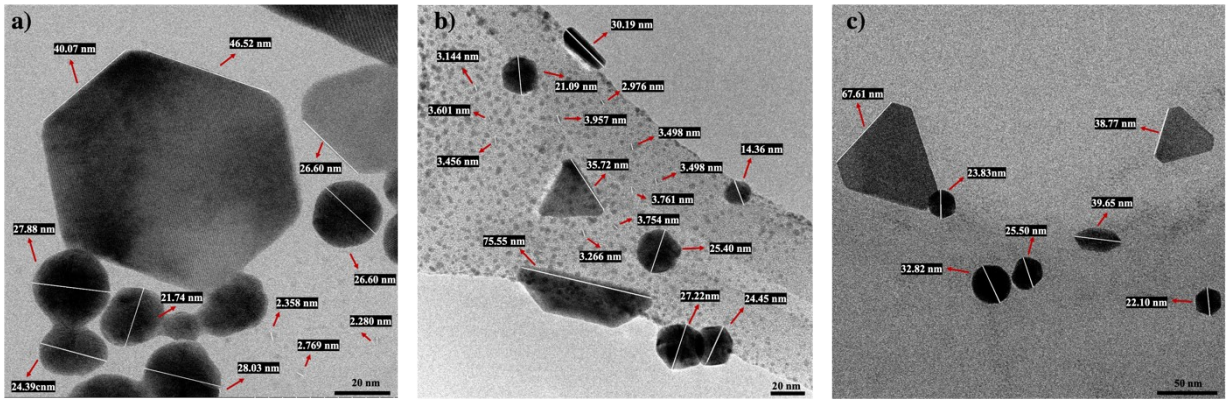


Figure S4 Size analysis of (a) GGS-3MPA, (b) GGS-bPEI, and (c) GGS-BSA NPs from TEM images.

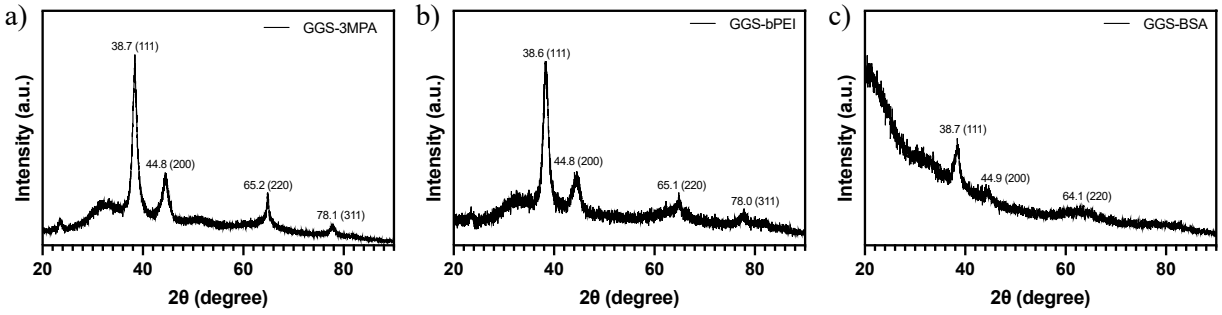
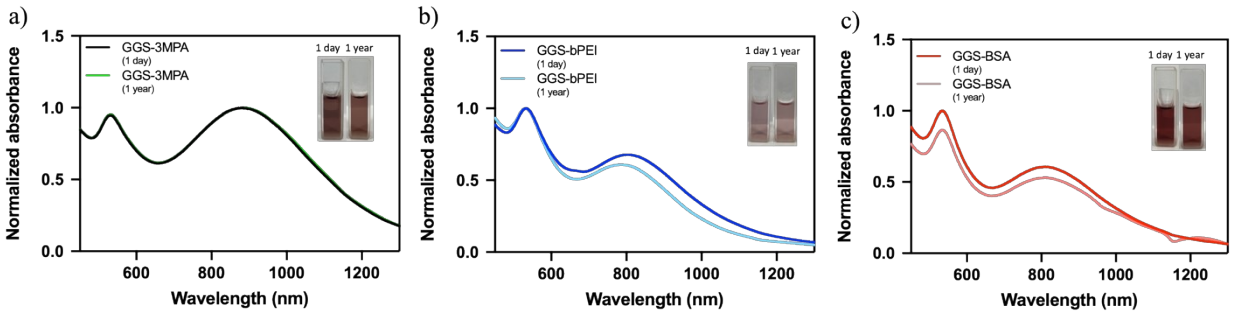


Figure S5 XRD of (a) GGS-3MPA, (b) GGS-bPEI, and (c) GGS-BSA.

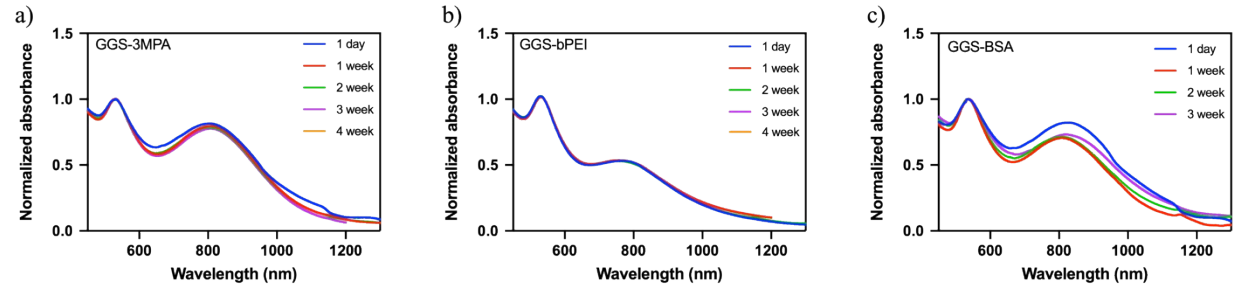


d)

Sample	Dh-number[a] (nm)			Dh-intensity[b] (nm)			Zeta potential (mV)		
	1 day	1 month	1 year	1 day	1 month	1year	1 day	1 month	1 year
GGS-3MPA	3.3	3.3	3.2	64.4	65.0	82.9	-34.1	-33.3	-5.5
GGS-bPEI	4.4	4.1	4.0	101.5	98.2	80.3	+8.9	+9.1	+9.5
GGS-BSA	40.9	40.0	41.6	107.7	103.5	105.5	+25.1	+23.1	+15.3

[a] Hydrodynamic size measured by DLS and reported as the number average. [b] Hydrodynamic sized measured by DLS and reported as the intensity average.

Figure S6 The absorbance spectrum of (a) GGS-3MPA, (b) GGS-bPEI, (c) GGS-BSA, and their (d) hydrodynamic size and zeta potential after the first day of synthesis and one year later. Samples were kept in the dark and +4 °C conditions.

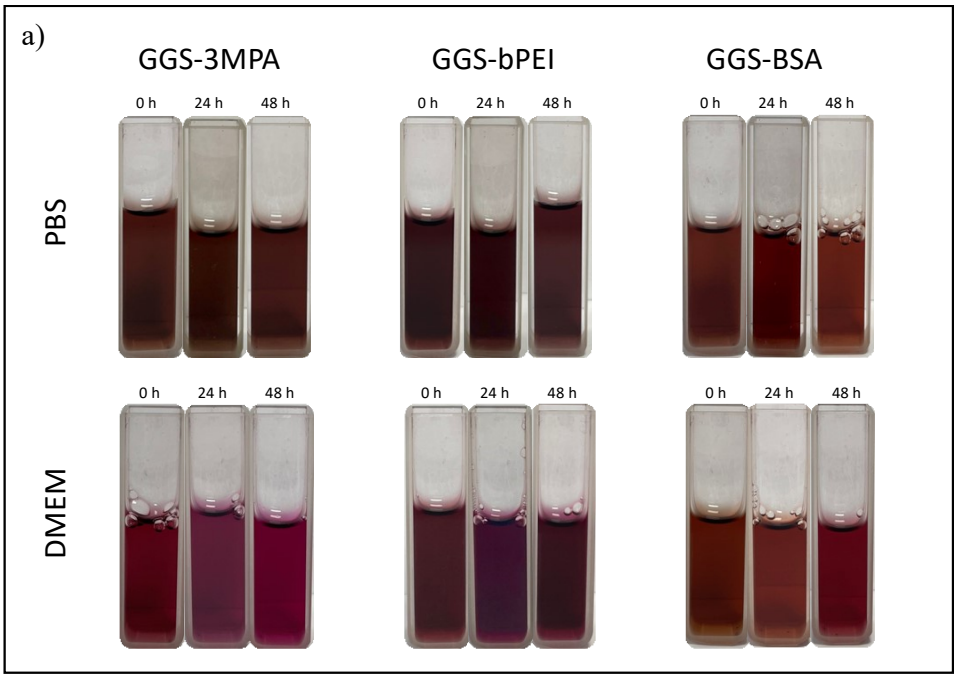


d)

Sample	Dh-number[a] (nm)					Dh-intensity[b] (nm)					Zeta potential (mV)				
	1 day	1week	2 week	3week	4 week	1 day	1 week	2 week	3 week	4 week	1 day	1 week	2 week	3 week	4 week
GGS-3MPA	1.6	1.8	1.6	1.8	1.5	48.0	55.2	52.6	48.5	66.7	-38.6	-32.6	-35.4	-38.6	-28.3
GGS-bPEI	2.9	3.0	3.1	2.8	3.2	70.2	68.1	63.3	61.2	69.1	+25.8	+15.6	+17.6	+17.8	+10.2
GGS-BSA	3.3	4.6	4.8	6.3	-	91.8	97.3	98.2	174.4	-	+19.5	+11.5	+12.5	+13.4	-

[a] Hydrodynamic size measured by DLS and reported as the number average. [b] Hydrodynamic sized measured by DLS and reported as the intensity average.

Figure S7 The absorbance spectrum of (a) GGS-3MPA, (b) GGS-bPEI, (c) GGS-BSA, and their (d) hydrodynamic size and zeta potential followed for 4 weeks. Samples were kept in the dark and +37 °C conditions.



b)

Sample	Dh-number[a] (nm)			Dh-intensity[b] (nm)			Zeta potential (mV)		
	0 h	24 h	48 h	0 h	24 h	48 h	0 h	24 h	48 h
GGS-3MPA	0.75	0.73	0.85	35.05	34.54	36.11	-10.1	-9.2	-9.0
GGS-bPEI	1.41	1.30	1.43	51.23	59.05	58.17	+3.1	+2.5	+2.2
GGS-BSA	3.23	3.32	4.01	105.11	122.5	120.6	+11.2	+9.6	+9.3

[a] Hydrodynamic size measured by DLS and reported as the number average. [b] Hydrodynamic sized measured by DLS and reported as the intensity average.

Figure S8 a) Pictures of GGS-3MPA, GGS-bPEI, and GGS-BSA solutions in PBS and DMEM. b) Hydrodynamic size and zeta potential of GGS solutions in PBS. Samples were kept in the dark at +37 °C.

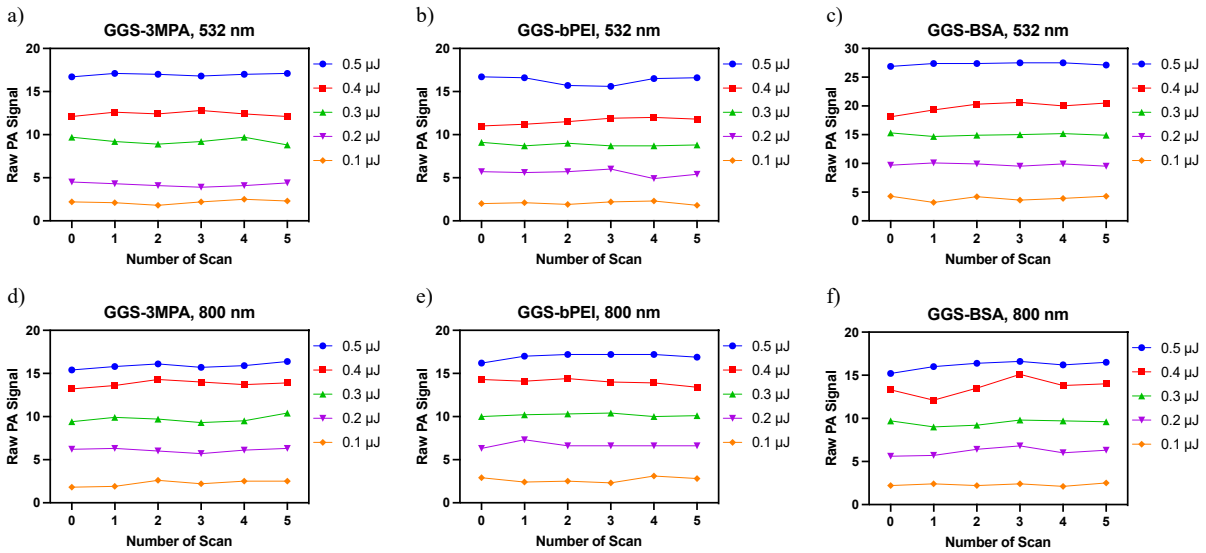


Figure S9 Raw PA Signal of (a) GGS-3MPA, (b) GGS-bPEI, (c) GGS-BSA at 532 nm laser exposure. Raw PA Signal (d) GGS-3MPA, (e) GGS-bPEI, (f) GGS-BSA at 800 nm laser exposure.

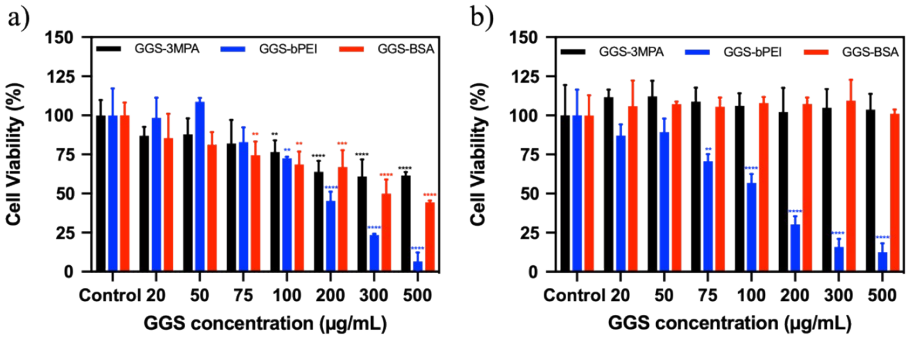


Figure S10 Dose-dependent cell viability of (a) L929 and (b) MDA-MB-231 cells treated with GGS-3MPA, GGS-bPEI, and GGS-BSA after 48 h incubation. Control: Untreated cells. Data were shown as mean ± SD (n=5). Significance levels are indicated as follows: p = 0.0332 (*), p = 0.0021 (**), p = 0.002 (***), and p < 0.001 (****).

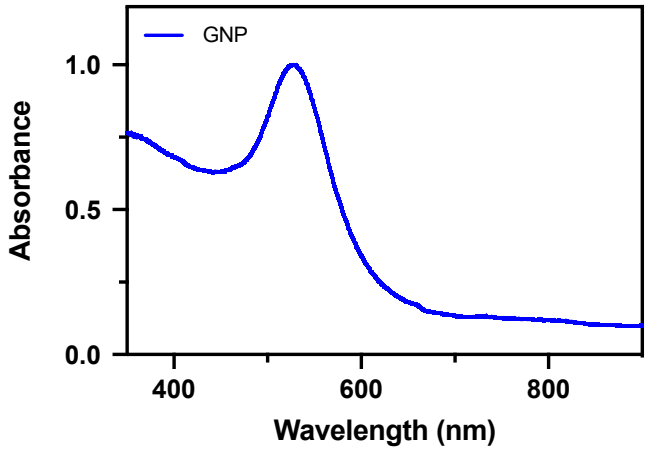


Figure S11 Absorbance spectrum of GNP.

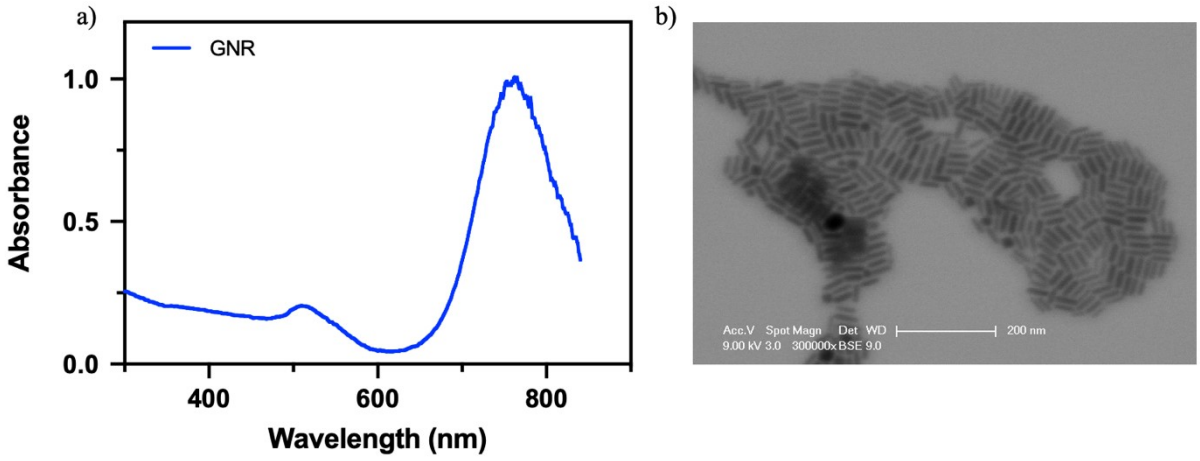


Figure S12 (a) Absorbance spectrum of GNR, (b) TEM image of GNR.

	μ_E=0.25 mm^{-1}		μ_E=0.5 mm^{-1}	
Sample	Slope @532nm	Slope @800nm	Slope @532nm	Slope @800nm
GGS-3MPA	8.7	10.5	8.6	9.1
GGS-bPEI	7.3	8.6	8.0	8.1
GGS-BSA	10.5	10.1	8.6	10.0
GNP	4.2	-	3.7	-
GNR	-	7.9	-	-

Table S1 Photoacoustic signal efficiencies of GGS, GNP, and GNR.

Line & Edge Spread Functions:

We further characterized the resolution power of our PAM setup with a sharp-edged sample scanned with the microscopy system. The results suggest that the FWHM of the minimum resolvable feature is 4 and 6 µm for 532 nm and 800 nm, respectively.

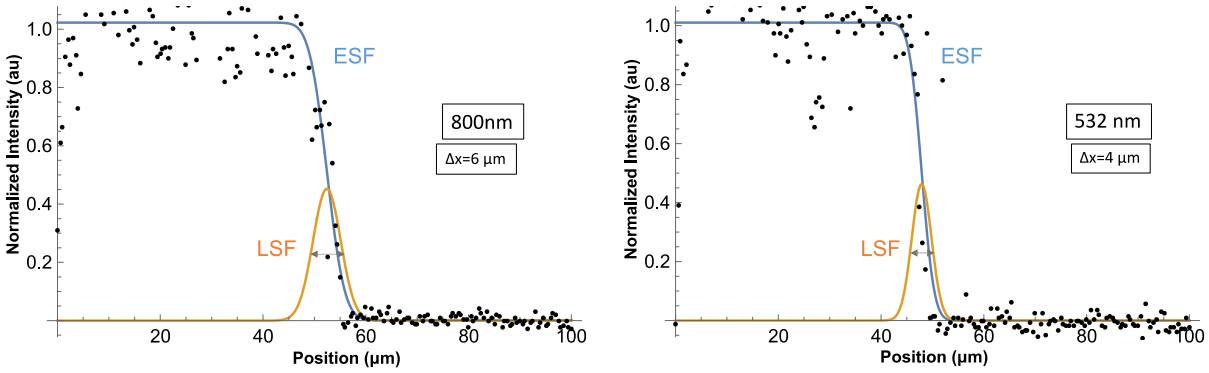


Figure S13 Experimentally measured edge and line spread functions at 800 and 532 nm wavelengths.

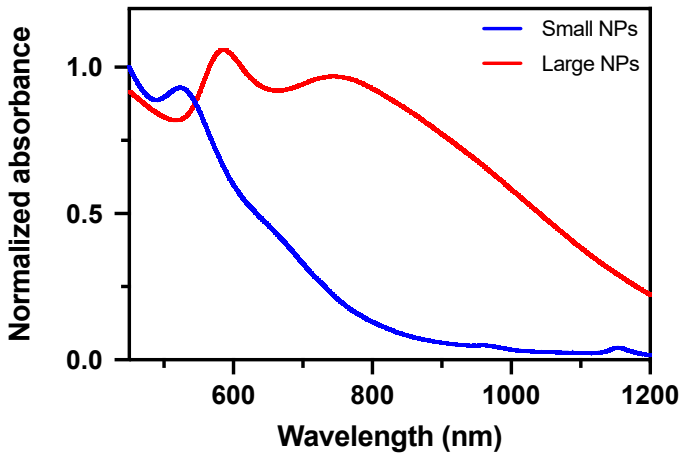


Figure S14 Absorbance spectrum of small and large GGS-3MPA NPs separated via centrifugation.