Supplementary Information

Fast and Inexpensive Separation of Bright Phosphor Particles from Commercial Sources by Gravitational and Centrifugal Sedimentation for Deep Tissue X-ray Luminescence Imaging

Mohammad Arifuzzaman [1,†], Meenakshi Ranasinghe [1],

Apeksha Rajamanthrilage [1], Sriparna Bhattacharya [3,*], Jeffrey N. Anker [1,2,*]

[1] Department of Chemistry, Clemson University, Clemson, SC 29634, USA

[2] Center for Optical Materials Science Engineering Technology (COMSET), Clemson

University, Clemson SC, 29634, USA

[3] Clemson Nanomaterials Institute, Department of Physics and Astronomy, Clemson

University, Clemson 29634, USA

* Corresponding: bbhatta@g.clemson.edu; janker@clemson.edu

† Present address: Research & Development Scientist, Cardinal CG (Coating Glass)

Technology Center, Spring Green, WI 53588, USA.

Electronic Supplementary Information

Table of contents: This supporting information includes figures depicting the characterization of ball-milled and separated nanoparticles, in terms of X-ray diffraction, spectroscopy, electron microscopy and DLS.

(a) Ball-milled commercial Eu-doped GOS

Legend: BM 0h, BM 1h, BM 2h, BM 3h, BM 6h, BM 7h, BM 8h

Peaks labeled: (100), (101), (102), (003), (110), (111), (200), (201), (202), (113), (203), (210), (211)

Intensity (eps) vs Angle (2Theta)

(b)

BM time	Crystalline Size (A)	Strain
0 h	1806	0.145
1 h	336	0.385
2 h	235	0.537
3 h	165	0.537
4 h	142	0.506
5h		
6 h	137	0.521
7 h	110	
8 h	88	0.01

(c)

Intensity vs λ (nm)

Legend: 8 Hr, 7 Hr, 6 Hr, 5 Hr, 4 Hr, 3 Hr, 2 Hr, 1 Hr, Com 8μm

Figure S1: XRD and x-ray luminescence spectra of long term ball-milled commercial 8 micron Gd$_2$O$_2$S:Eu particles. Structural characterization of the commercial 8 μm Gd$_2$O$_2$S: Eu microparticles before and after ball-milling using **(a)** X-ray diffraction, **(b)** crystallite size and strain calculated from Rietveld analysis. **(c)** Corresponding luminescence intensity of the commercial and ball-milled particles as a function of emission wavelength. The double y-axis represents the luminescence intensity of the following: 0h ball-milled (Com 8μm) particles (**right axis**), and the 1-8h ball-milled particles (**left axis**).

Commercial Gd$_2$O$_2$S: Eu (8 micron)
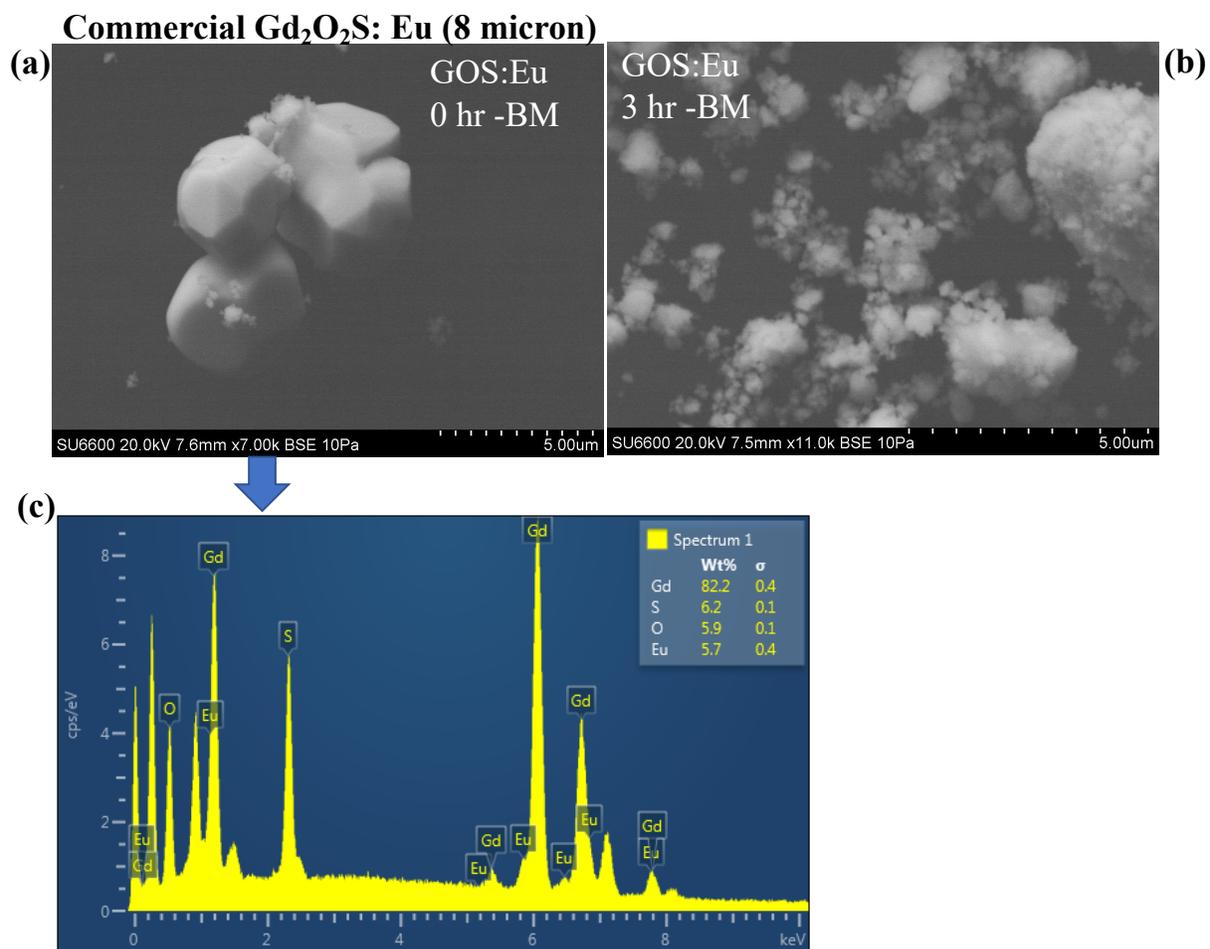
Figure S2: Long term ball-milling of commercial Gd₂O₂S:Eu 8 micron particles. STEM images of the commercial 8 μm Gd$_2$O$_2$S: Eu microparticles before (0h) **(a)** and after (3h) ball-milling **(b)** using STEM. **(c)** Energy dispersive X-ray (EDX) of the commercial 8 μm Gd$_2$O$_2$S: Eu microparticles.
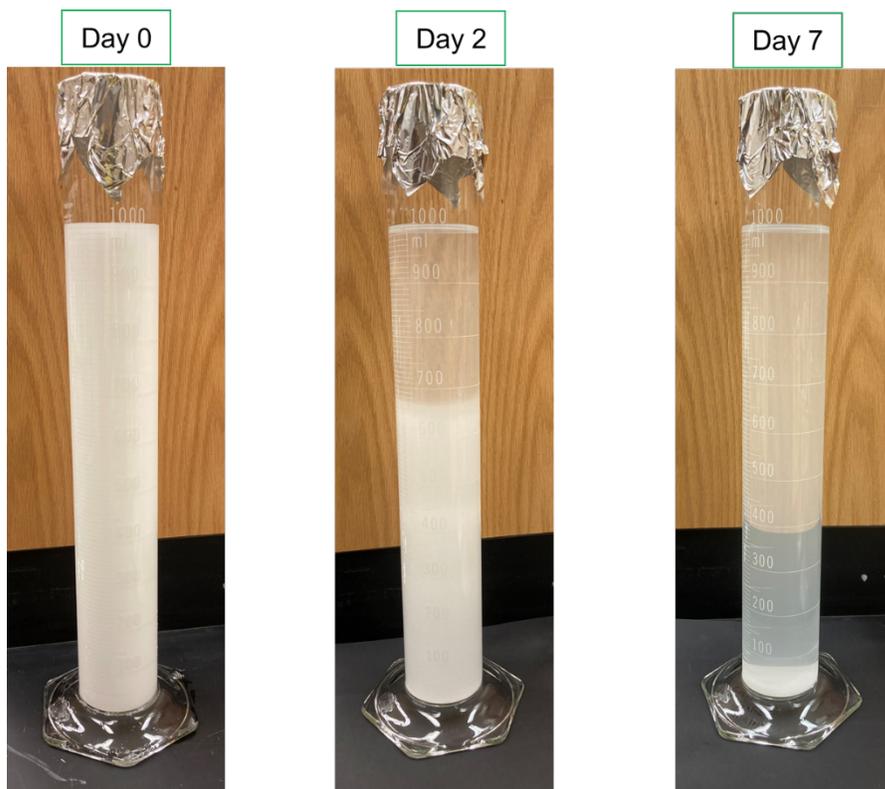
Figure S3: Photograph showing the separation process of nano and submicron sized particles from commercially available phosphors in DI water simply via spontaneous sedimentation under normal gravity in a 1000 mL graduated cylinder.
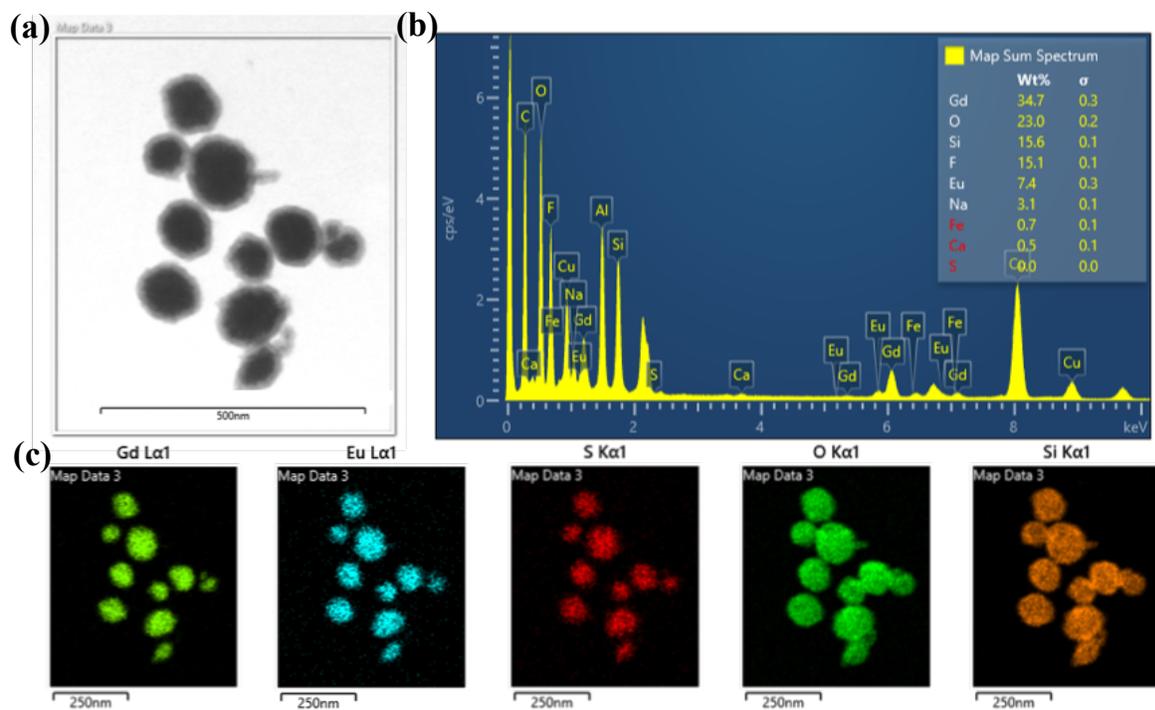
Figure S4: Energy dispersive X-ray (EDX) analysis of separated nanoparticles from commercial Gd₂O₂S:Eu 2.5 μm particles. SEM image (**a**) shows possible Si-coating on the nanoparticles. Elemental analysis using EDX (**b**) confirming the presence of Si. EDX elemental mapping (**c**) on the same cluster of nanoparticles confirming the presence of Gd, Eu, S, O, and Si (starting from left to right).

(a)

First 150 mL S8G (sediment)

Size Distribution by Intensity

~1.1 mg in ~45 mL;
Final concentration:
0.024 mg/mL

(b)

Dried aliquot First 150 mL S8G

Size Distribution by Intensity

~1.1 mg resuspended
in 1 mL; Final
concentration: 1
mg/mL
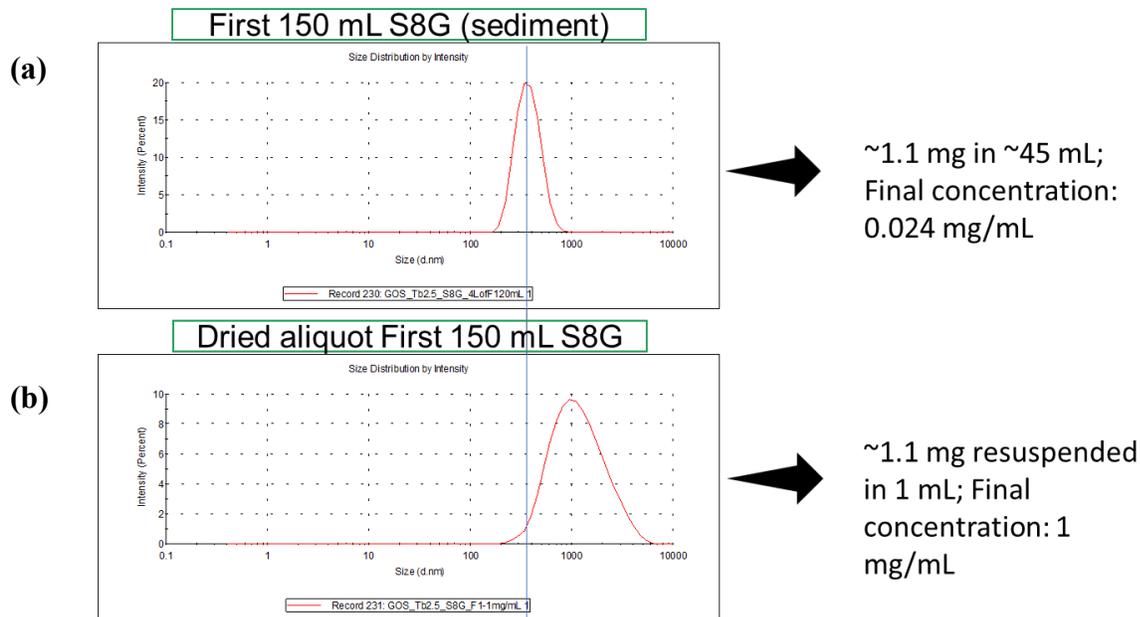
Figure S5: Dynamic light scattering size distribution plot weighted by scattering intensity for separated nanophosphors in DI water of the *F1* layer **(a)** with well-dispersed suspension of diluted nano sized particles (0.024 mg/mL), **(b)** showing aggregation after evaporative concentration to 1 mg/mL.

(a)

Sample Name:	GOS_Eu2.5_S8R_F1 150 mL 3		
SOP Name:	mansettings.nano		
File Name:	Example Results.dts	Dispersant Name:	Water
Record Number:	223	Dispersant RI:	1.330
Material RI:	1.80	Viscosity (cP):	0.8872
Material Absorbtion:	0.001	Measurement Date and Time:	Thursday, July 30, 2020 12:19:50 PM

Temperature (°C):	25.0	Duration Used (s):	60
Count Rate (kcps):	299.2	Measurement Position (mm):	4.65
Cell Description:	Disposable sizing cuvette	Attenuator:	9

			Size (d.nm):	% Intensity:	St Dev (d.nm):
Z-Average (d.nm):	277.3	Peak 1:	301.6	100.0	90.27
PdI:	0.133	Peak 2:	0.000	0.0	0.000
Intercept:	0.880	Peak 3:	0.000	0.0	0.000
Result quality :	Good				

Size Distribution by Intensity

PdI for peak 1 = (90.3/301.6)^2 = 0.09

Record 221: GOS_Eu2.5_S8R_F1 150 mL 1   Record 222: GOS_Eu2.5_S8R_F1 150 mL 2
Record 223: GOS_Eu2.5_S8R_F1 150 mL 3
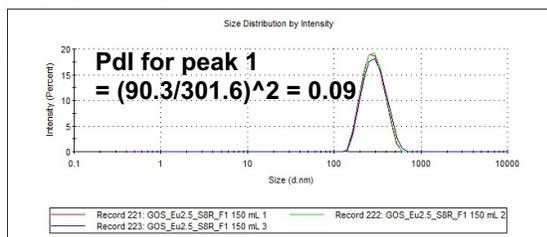
(b)

Sample Name:	GOS_Tb2.5_S8G_4LofF120mL 1		
SOP Name:	mansettings.nano		
File Name:	Example Results.dts	Dispersant Name:	Water
Record Number:	206	Dispersant RI:	1.330
Material RI:	1.80	Viscosity (cP):	0.8872
Material Absorbtion:	0.001	Measurement Date and Time:	Saturday, July 25, 2020 10:33:17 AM

Temperature (°C):	25.0	Duration Used (s):	80
Count Rate (kcps):	160.3	Measurement Position (mm):	4.65
Cell Description:	Disposable sizing cuvette	Attenuator:	10

			Size (d.nm):	% Intensity:	St Dev (d.nm):
Z-Average (d.nm):	355.3	Peak 1:	376.0	100.0	83.85
PdI:	0.068	Peak 2:	0.000	0.0	0.000
Intercept:	0.852	Peak 3:	0.000	0.0	0.000
Result quality :	Good				

Size Distribution by Intensity

PdI for peak 1 = (83.9/376.0)^2 = 0.05

Record 206: GOS_Tb2.5_S8G_4LofF120mL 1
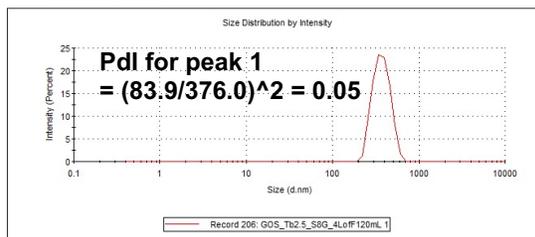
Figure S6: Polydispersity index (PdI) obtained through DLS measurements for the *F1* layers of the separated **(a)** Gd_2O_2S:Eu and **(b)** Gd_2O_2S:Tb nanophosphors. The insets show the PdI values calculated for just peak 1 from the square of the standard deviation divided by the square of the mean, for the separated Gd_2O_2S:Eu and Gd_2O_2S:Tb particles, respectively.