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White Light-Emitting Diodes Using Thermally and Photochemically Stable Fluorescent Silica Nanoparticles As Color-Converters

Hak-Sung Jung, Young-Jae Ki, Shin-Woo Ha, and Jin-Kyu Lee*

† *Department of Chemistry, Seoul National University, Seoul 151-747, Korea*

E-mail: jinklee@snu.ac.kr, Tel: +82-2-879-2923, Fax: +82-2-882-1080

Table S1. Reaction conditions for size-controlled fluorescent silica nanoparticles.

Entry[a]	Molar concentration (mol/L)				Mole percent of dye-TMS to TEOS (%)	Average size (nm)[c]	Reaction time (h)
	TEOS	Ammonium hydroxide[b]	Water	Dye-TMS ($x10^{-3}$)			
1	0.082	0.14	1.1	0.66	0.80	32 ± 4.4	12
2	0.090	0.14	1.1	0.66	0.73	48 ± 8.3	12
3	0.090	0.21	1.7	0.66	0.73	120 ± 4.6	12
4	0.082	0.14	1.1	0.29	0.35	26 ± 3.6	12
5	0.090	0.21	1.7	0.24	0.27	50 ± 5.0	12
6	0.097	0.34	2.7	0.18	0.19	69 ± 3.4	12

[a] Entries 1–3 are for SiO_2(RhBH), and entries 4–6 are for SiO_2(PR254H); [b] Composition: 28% ammonia in water(v/v); [c] Sizes were characterized by TEM (See Figure 1).

Table S2. Calculated molar ratio of incorporated RhBH and PR254H within SiO_2 by TGA analysis

	Weight % from TGA (%)	Estimated Formula Weight (g/mol)[(a)]	Relative amount from 100 g sample
Amount of RhB	6 %	520 (for RhB part)	6 g (1.2×10^{-2} mol)
Amount of SiO_2	94 %	60 (for fully condensed SiO_2)	94 g (1.6 mol)
Mole percent of RhB[(b)]			7.5×10^{-3} (≒ 0.75 %)
Amount of PR254	2 %	438 (for PR254 part)	2 g (4.6×10^{-3} mol)
Amount of SiO_2	98 %	60 (for fully condensed SiO_2)	98 g (1.6 mol)
Mole percent of PR254[(b)]			2.9×10^{-3} (≒ 0.29 %)

[(a)] Organic parts were counted for RhB and PR254, while other parts were assumed as fully condensed SiO_2; [(b)] Mole percent of organic dyes were roughly calculated from TGA data, which were expected to be same as the percent of Si from RhBH or PR254H comparing to that from TEOS within FSNPs

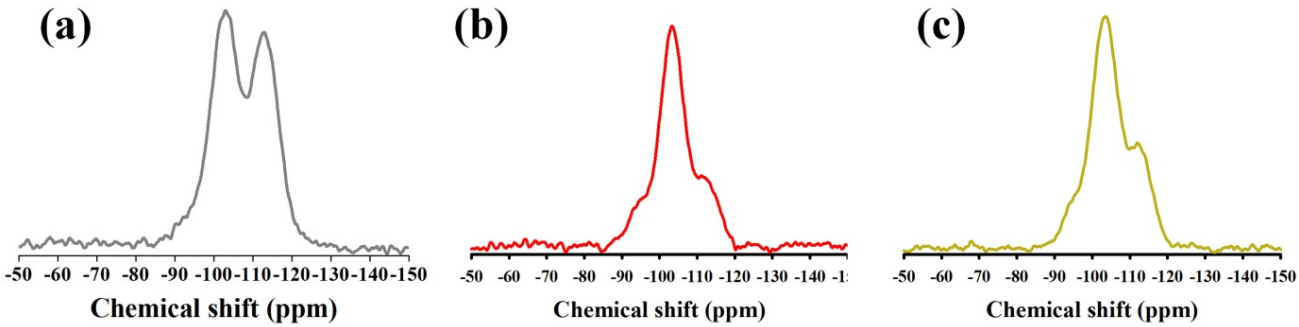

Fig. S1. ^{29}Si-CP-MAS-NMR spectra of (a) SiO_2, (b) SiO_2(RhBH) and (c) SiO_2(PR254H). It was clearly observed that Q^4 peak at -113 ppm in (b) and (c) was decreased significantly while Q^3 peak at -102 ppm did not changed significantly, which meant the incomplete condensation of Si-OH groups due to the steric hindrances from bulky organic dye molecules within silica matrix. It was also noteworthy that no detectable T^2 and T^3 peaks were observed at around -58 and -65 ppm, respectively, from SiO_2(RhBH) and SiO_2(PR254H). It was probably owing to the small amount of Si from incorporated RhBH and PR254H as calculated in Table S1 and S2.

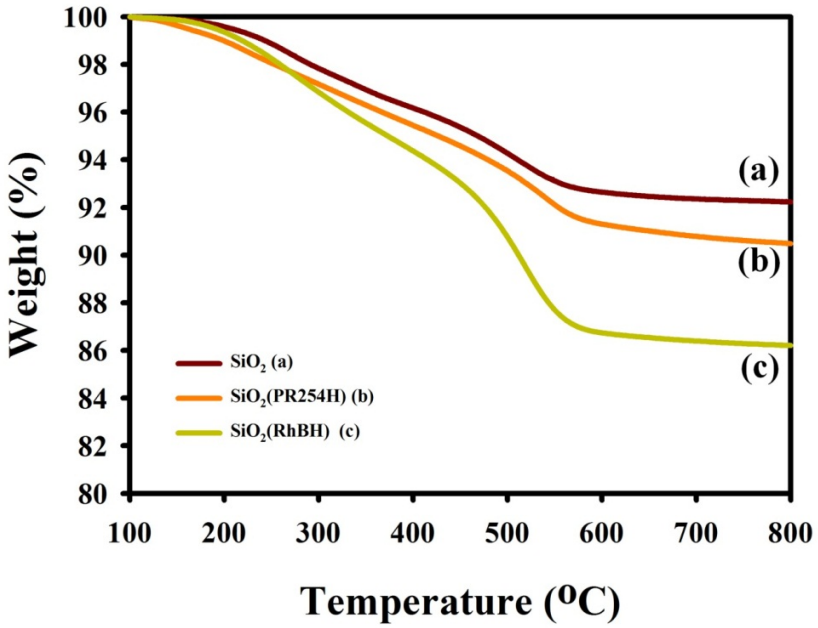

Fig. S2. TGA comparison of (a) SiO_2, (b) SiO_2(PR254H) and (c) SiO_2(RhBH); indicating weight losses of 8%, 10% and 14%, respectively, in the temperature ranges from 100 to 600 °C. An additional 2% and 6% weight loss detected in FSNPs could be attributed to the thermal degradation of organic dye molecules in the silica matrices. Therefore, the mole percentage of organic dyes to SiO_2 matrix could be calculated as summarized in Table S1 (0.29 and 0.75, respectively), which were relevant to the mixing percent of dyes to TEOS in Table S1 (0.27 and 0.73 for PR254H and RhBH, respectively).

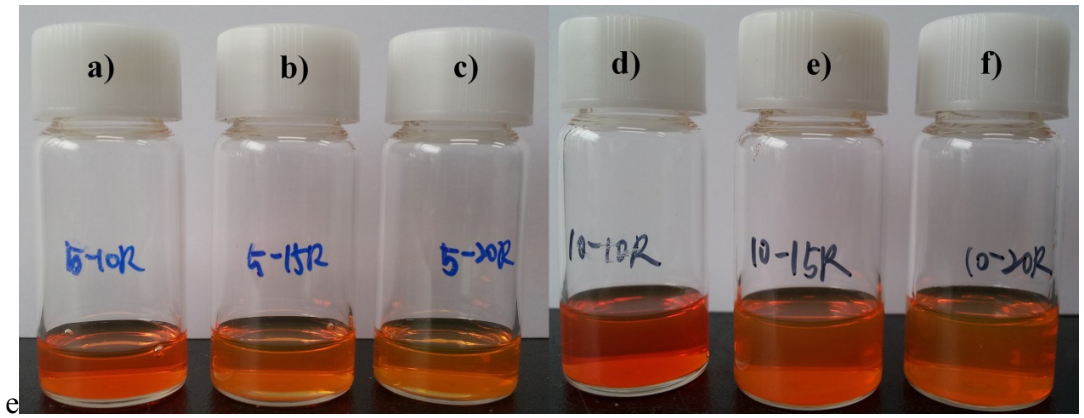

Fig. S3. Photographs of various mixing conditions of FSNPs with silicone polymer in acetone. (a) 5 wt% and 1:10 ratio of SiO_2(RhBH) to SiO_2(PR254H); (b) 5 wt% and 1:15 ratio of SiO_2(RhBH) to SiO_2(PR254H); (c) 5 wt% and 1:20 ratio of SiO_2(RhBH) to SiO_2(PR254H); (d) 10 wt% and 1:10 ratio of SiO_2(RhBH) to SiO_2(PR254H); (e) 10 wt% and 1:15 ratio of SiO_2(RhBH) to SiO_2(PR254H); (f) 10 wt% and 1:20 ratio of SiO_2(RhBH) to SiO_2(PR254H).

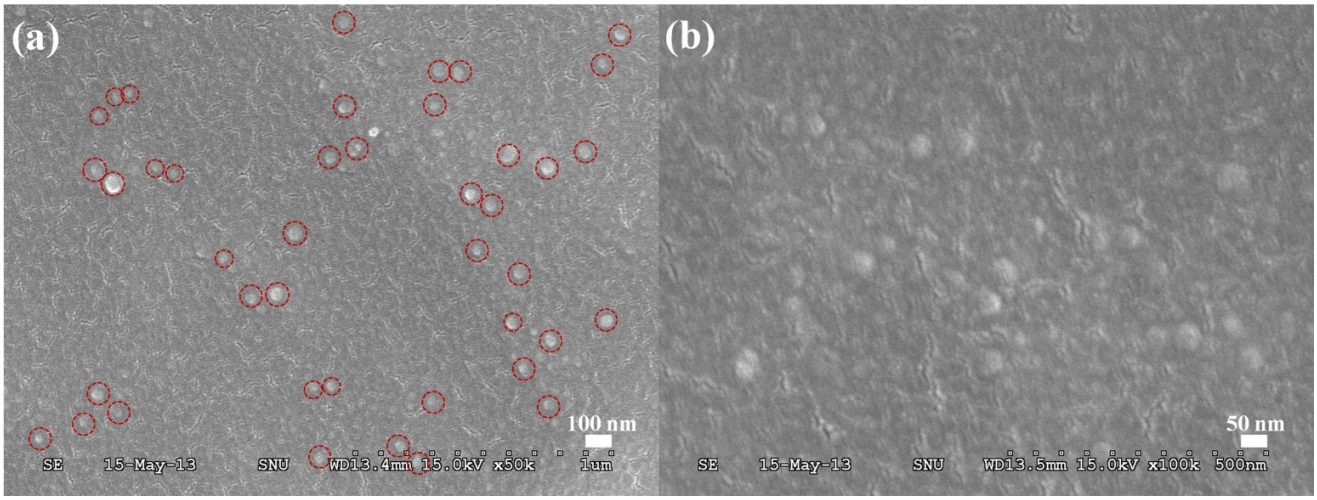

Fig. S4. SEM images of the cross-sectioned area of FSNPs/silicon polymer composite in (a) low and (b) high magnification. The circles are indicating FSNPs.