Supporting information Zirconia/Phenylsiloxane Nano-composite for LED Encapsulation with High and Stable Light Extraction Efficiency

Ying Lu^{a, b, ‡}, Zhihang Zhao^{a, ‡}, Xianpeng Fan^a, Xinyu Cao^{a,} *, Mingtan Hai^b, Zhou

Yang ^{b,} *, Kun Zheng^a, Jiaxin Lu^a, Jingnan Zhang^a, Yongmei Ma^{a,} *, Rongben Zhang

^a, Shibi Fang ^a

^a Key Laboratory of Green Printing, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China

^b School of Chemistry and Chemical Engineering, University of Chinese Academy of

Sciences, Beijing 100049, China

*Corresponding authors.

‡These authors contributed equally to this work.

E-mail addresses: <u>xinyucao@iccas.ac.cn (X.Y Cao)</u>, <u>yangz@ustb.edu.cn (Z. Yang)</u>, <u>maym@iccas.ac.cn (Y. Ma)</u>.



Fig. S1 ²⁹Si-NMR spectra of PDPh and MDPS with integral data.

The average polymerization degree of PDPh is calculated from Equation 1:

The average polymerization degree of PDPh $= \frac{2(D^0 + D^1 + D^2)}{D^1 + 2D^0}$Equation 1 The average polymerization degree of PDPh $= \frac{2(0.30 + 10.53 + 4.75)}{10.53 + 2 * 0.30} = 2.8.$ The average polymerization degree of MDPS (including end-capping groups) is calculated from the Equation 2:

 $\frac{2(D^1 + D^2 + M^1 + M^1)}{2(D^1 + D^2 + M^1 + M^1)}$

The average polymerization degree of MDPS

$$= \frac{1}{D^1 + M^1 + M^1}$$

......Equation 2 The average polymerization degree of MDPS

$$=\frac{2(0.15+4.18+1.00+2.90)}{(0.15+2.90+1.00)}_{=4.1.}$$

The end-capping ratio of MDPS is calculated from Equation 3.

The end-capping ratio of MDPS
$$= \frac{M^{1} + M^{1'}}{M^{1} + D^{1} + M^{1'}}$$
....Equation 3
The end-capping ratio of MDPS
$$= \frac{2.90 + 1.00}{2.90 + 0.15 + 1.00} = _{96\%}$$

For ²⁹Si-NMR measurement, PDPh and MDPS was dissolved in THF with TMS as standard respectively, and relaxation reagent (Chromium acetylacetonate) was added to promote relaxation. Corresponding information is added in "Characterization" section.

The preparation procedure for TEM samples:

For TEM observation, the samples were first diluted and dispersed in water or THF. Then they were dropped onto copper mesh respectively, and allow the solvents evaporate at rt in a clean area. The "as-synthesized" particles were dispersed in deionized water, and the surface modified ZrO₂ (M-ZrO₂) and the composite resin

(ZCS) were dispersed in THF. The MPDS resin can't be evaporated and should remained. Since it has been diluted, at this stage it can't be observed under TEM.



Fig. S2 Statistical nanoparticle size and distribution (A) The as-synthesized ZrO₂ nanoparticle. (B) The MPTMS modified ZrO₂ nanoparticle (M-ZrO₂).



Fig. S3 XRD patterns of (A) the baseline adopted for peak analysis of the surfaced modified ZrO₂. (B) The peak fitting result of the surface-modified ZrO₂ nanoparticles.
(C) The baseline was adopted for peak analysis of the as-synthesized ZrO₂. (D) The

peak fitting result of the surface as-synthesized ZrO₂ nanoparticles.

In the peak fittings, amorphous peaks were involved to estimate the crystallinity. According to the area ratio of the peaks, the crystallinity of the as prepare ZrO_2 nanoparticles was calculated as 71%. After surface modification and sample purification, it shows the crystallinity of the M- ZrO_2 nanoparticles is ca. 80%.



Figure S4. SEM image of the cross-section of ZSC film and the EDS mapping image



of the marked area.

Fig. S5 (A) TGA of M and ZSC with different ZrO₂ content. (B) The optical transmittance of ZSC films (280μm) with different ZrO₂ content.



Fig. S6 Photos of LED chips before and after red ink test. There is no red ink stain that can be observed for M, M-10/M encapsulation after the red ink test.



Fig. S7 SEM photo of the cross-section of a double-layer encapsulation.