Size-controlled Wurtzite Zinc Oxide Sphere with the Characteristics of Visible Absorption and Mie Scattering

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| | Zn (at %) | O (at%) | C (at %) |
|------|-----------|---------|----------|
| S4 | 14.86 | 50.37 | 34.77 |
| S12 | 16.00 | 49.16 | 34.84 |
| S14 | 14.92 | 50.69 | 34.68 |
| S4a | 48.30 | 51.70 | - |
| S12a | 48.67 | 51.33 | - |
| S14a | 49.19 | 50.81 | - |

Table S1 EDS analysis of samples



Fig. S1 SEM image of the spheres synthesized with various concentration of HMTA: (a) S4, 40 mM, (b) S6, 60 mM, (c) S8, 80 mM, (d) S10, 100 mM, (e) S12, 120 mM, (f) S13, 130 mM, and (g) S14, 140 mM.



Fig. S2 XRD patterns of the spheres synthesized with various concentration of HMTA (a) S4, (b) S6, (c) S8, (d) S10, (e) S12, (f) S13, and (g) S14. The additional diffraction peaks located at 33.3° and 59.0°. These very weak peaks correspond to the brucite-like structure indexed as (100) and (110), implying the presence of layered basic zinc salts (LBZ).



Fig. S3 IR spectra of as-synthesized samples. The spectra were very similar but it was found some differences in the range of 1450-1620 cm⁻¹ and 2850-3000 cm⁻¹. Please see Fig. 2 in the manuscript.



Fig. S4 (a) SEM image and (b) XRD pattern of the sample which was synthesized with 15 mM zinc acetate and 40 mM HMTA as reactants in a reflux reaction at 115 $^{\circ}$ C for 1h.



Fig. S5 (a) SEM image and (b) XRD pattern of the sample which was synthesized with 15 mM zinc acetate, 40 mM HMTA and 6 mM citrate as reactants in a reflux reaction at 115 $^{\circ}$ C for 30 min. It was noted that HMTA solution was pre-heated at 90 $^{\circ}$ C for 30min.



Fig. S6 (a)(b) SEM images and (c) XRD patterns of samples synthesized with 40 mM HMTA and the Zn^{2+} /citrate molar ratio of 5:1 in a reflux reaction at 115 °C for 1h. (a) 15 mM zinc acetate dehydrate and 3 mM trisodium citrate. (b) 30 mM zinc acetate dehydrate and 6 mM trisodium citrate.



Fig. S7 SEM image of (a) initial spheres, and products for different aging time (b) 15 min, (c)(d) 30 min, and (e)(f) 60 min, respectively. The S10 powder as precursor (70 mg) was suspended in 6 mM sodium acetate (50 ml) and the suspension was refluxed at 90 $^{\circ}$ C for different time (15, 30, 60 min). After the reflux reaction, the white powder in the solution was collected by centrifugation.



Fig. S8 (a) TGA profiles of amorphous/LBZ spheres, (b) TEM image (inset: SAED) of S4a, (c) Raman and (d) PL spectra of ZnO sphere which have been normalized by the intensity of the peak at $\lambda = 382$ nm.

$\begin{array}{c} \begin{array}{c} \theta = 0^{\circ} \\ \theta = 5.7^{\circ} \end{array} \end{array}$ $\theta = 0^{\circ}$ $\theta = 5.7^{\circ}$ а b ABS. (A. U.) ABS. (A. U.) 400 400 500 600 700 800 500 600 700 800 wavelength (nm) wavelength (nm) $\begin{array}{c} \begin{array}{c} \begin{array}{c} \theta = 0^{\circ} \\ \theta = 5.7^{\circ} \end{array} \end{array}$ $\underbrace{\begin{array}{c} \begin{array}{c} \begin{array}{c} \theta = \\ \theta = \\ \theta = \\ \end{array}}_{\theta = 5.7^{\circ}} \end{array}$ С d ABS. (A. U.) ABS. (A. U.) 700 400 400 500 600 800 500 600 700 800 wavelength (nm) wavelength (nm)

Fig. S9 UV-vis absorption spectra of spheres with different scattering angles: (a) S4, (b) S8, (c) S12, and (d) S14.



Fig. S10 The simulated Mie scattering spectra by using MiePlot ($\theta = 0^{\circ}$).