Supporting information for

## High-Tap Density Flower-like LiFePO<sub>4</sub> microsphere developed by combined computational and experimental approaches

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Fig. S1 SEM images of samples prepared with glycol solvent at 180°C for 10 h

As shown in Fig. S1, it is clear that the sample consists of microspheres with three diameters distribute in 14 $\mu$ m, 7  $\mu$ m and 2  $\mu$ m. The tap density of the ternary unequal spheres is about 1.515 g cm<sup>-3</sup>.



Fig. S2 SEM images of samples prepared with glycol solvent at 200°C for 24h

(Reference: YuanJin, XincunTang, RSC.Adv, 2016, 6, 75602-75608)

As shown in Fig. S2, the sample consists of microspheres with uniform and monodisperse microsphere morphology assembled by nanosheets, whose average diameter is about  $15\mu$ m. Note that the LiFePO<sub>4</sub> microspheres with unique morphology here promise an ultrahigh powder tap density of 1.4 g/cm<sup>3</sup>, higher than those reported values in references.



Fig. S3 SEM images of samples prepared with water/diethylene glycol (DEG)

mixed solvent (1:7, v/v) at 200°C for 24h

As shown in Fig. S3, at the radius ratio r/R of 0.302, the tap density of the binary unequal spheres is about 1.436 g cm<sup>-3</sup>.



Fig. S4 SEM images of samples with diethylene glycol (DEG) solvent at 200°C for 24h

As shown in Fig. S4, at the radius ratio r/R of 0.812, the tap density of the binary unequal spheres is about 1.188 g cm<sup>-3</sup>.