Supporting Information

Versatile assembly of metal sulfide supraparticles in aqueous phase

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Section 1. Calculation of Photothermal Conversion Efficiency.

1□ The photothermal conversion efficiency (η) of different size Cu_2S SPs was calculated as the following equation:

$\eta = hS\,(T_{max,\,SPs} - T_{max,\,solvent})/[I\,(1 - 10^{-A808})]$
$\quad = mc\,(T_{max,\,SPs} - T_{max,\,solvent})/[I\,(1 - 10^{-A808})\,\tau_s]$
$t = -\tau_s\,\ln\theta$
$\theta = (T - T_{surr})/(T_{max,\,SPs} - T_{surr})$

where h is the heat transfer coefficient, S is the irradiated area, m is the mass (1.0 g) and C is the heat capacity (4.2 J g⁻¹) of the solvent, A_{808} is the absorption of Cu_2S SPs solutions (110 μg mL⁻¹), τ_s is calculated and shown in Figure S10. $T_{max,\,SPs}$ and $T_{max,\,solvent}$ are the highest temperature of the system and the surrounding, respectively. The detailed calculations were shown as follows:

$\eta_{20\,nm} = 1 \times 4.2 \times (64.3 - 23.6)/[0.75 \times (1 - 10^{-0.488}) \times 380.69] \times 100\% = 88.7\%$

$\eta_{40\,nm} = 1 \times 4.2 \times (67.1 - 22)/[0.75 \times (1 - 10^{-0.915}) \times 428.29] \times 100\% = 67.1\%$

$\eta_{60\,nm} = 1 \times 4.2 \times (75.4 - 23.5)/[0.75 \times (1 - 10^{-1.185}) \times 430.57] \times 100\% = 72.2\%$

2□ The photothermal conversion efficiency (η) of Cu_2S/Fe_2O_3 SPs was calculated as follow equation:

$\eta = hS\,(T_{max,\,SPs} - T_{max,\,solvent})/[I\,(1 - 10^{-A1064})]$
$\quad = mc\,(T_{max,\,SPs} - T_{max,\,solvent})/[I\,(1 - 10^{-A1064})\,\tau_s]$
$t = -\tau_s\,\ln\theta$
$\theta = (T - T_{surr})/(T_{max,\,SPs} - T_{surr})$

where h is the heat transfer coefficient, S is the irradiated area, m is the mass (1.0 g) and C is the heat capacity (4.2 J g⁻¹) of the solvent, A_{1064} is the absorption of Cu_2S/Fe_2O_3 SPs solutions (50 μg mL⁻¹), τ_s is calculated and shown in Figure S11. $T_{max,\,SPs}$ and $T_{max,\,solvent}$ are the highest temperature of the system and the surrounding, respectively. The detailed calculations were shown as follows:

$\eta_{sp} = 1 \times 4.2 \times (55 - 30)/[1 \times (1 - 10^{-0.75}) \times 282.417] \times 100\% = 45.2\%$

Section 2. Calculation of crystal domain size.

Table 1 Calculation of crystal domain size

pattern	2θ (°)	FWHM (°)
CdS, 4.82 nm	43.28	1.86
Cu_2S, 6.56 nm	48.84	1.39
ZnS, 3.67 nm	43.60	2.43

The relationship between domain size and FWHM is based on the Debye-Scherrer expression according to which: domain size in nm = $0.94 * \lambda_X / (\cos\theta * FWHM$ in radian) where λ_X is Cu Kα X-ray radiation wavelength of 0.154 nm.
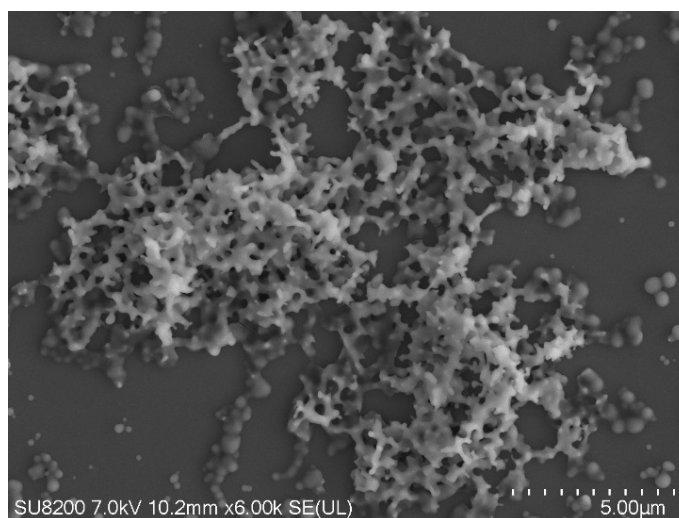
Figure S1. SEM image of the precursor compounds cysteine-Cd^{2+} before NaOH was added.
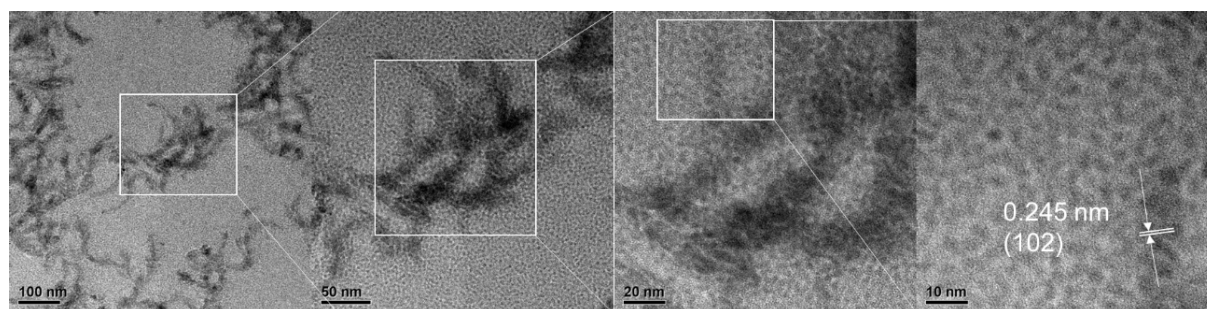
Figure S2. TEM image of the precursor compounds cysteine-Cd^{2+} after NaOH was added.
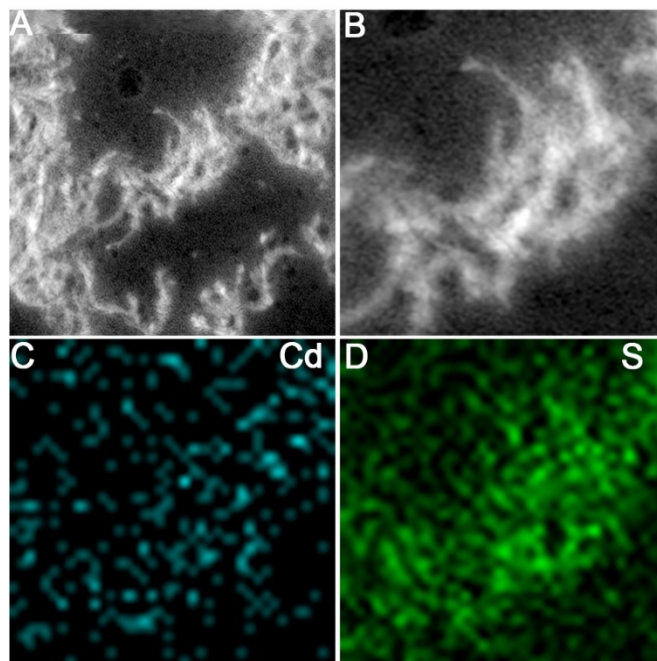
Figure S3. The elemental distribution mapping images of the precursors compound cysteine-Cd^{2+} after NaOH was added.
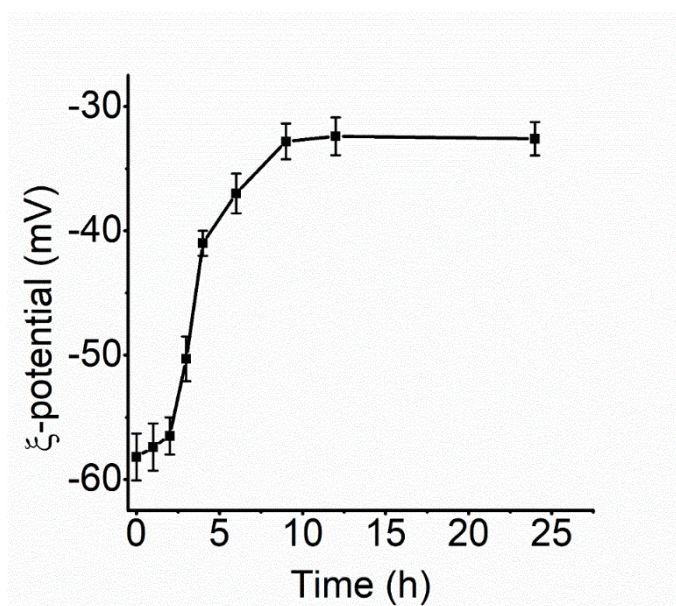

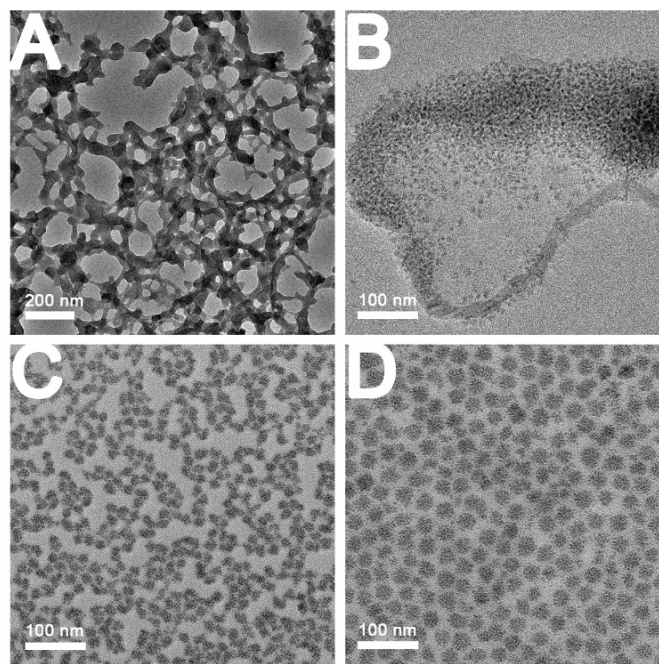**Figure S4. The plot of the ζ-potential values of the CdS SPs vs. reflux time.**

Figure S5. The self-assembly process of Cu₂S SPs. (A) The TEM images of the precursor compound cysteine-Cu⁺ before NaOH was added (the inset is the photo picture of the corresponding reaction solution). (B) The TEM images of the products fabricated at 1 h. (C) The TEM images of the products fabricated at 5 h. (D) The TEM images of the products fabricated at 12 h.
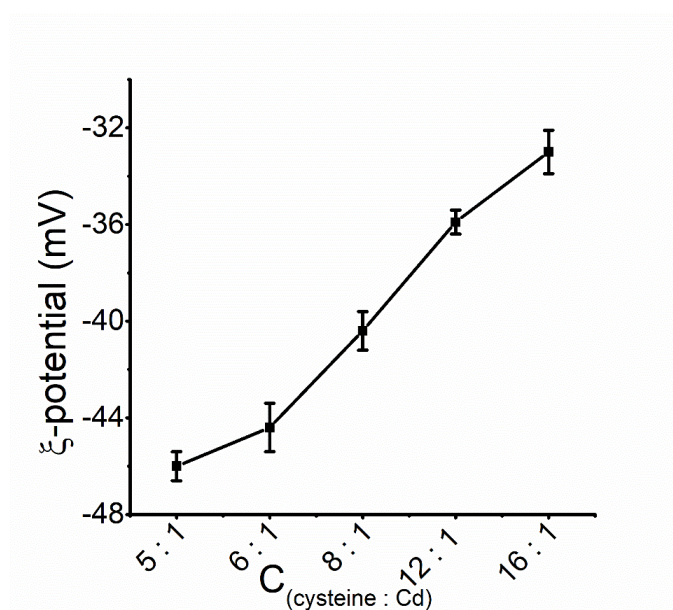

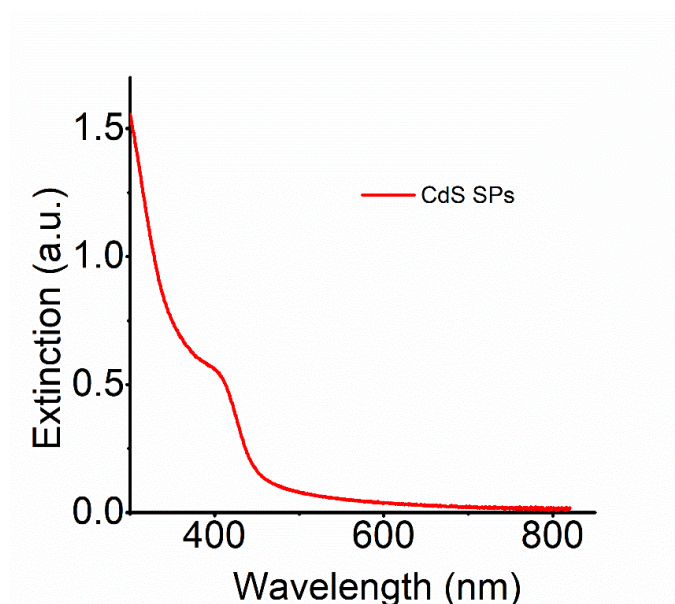**Figure S6. The plot of the ζ-potential values of the different ratios of cysteine and Cd⁺.**

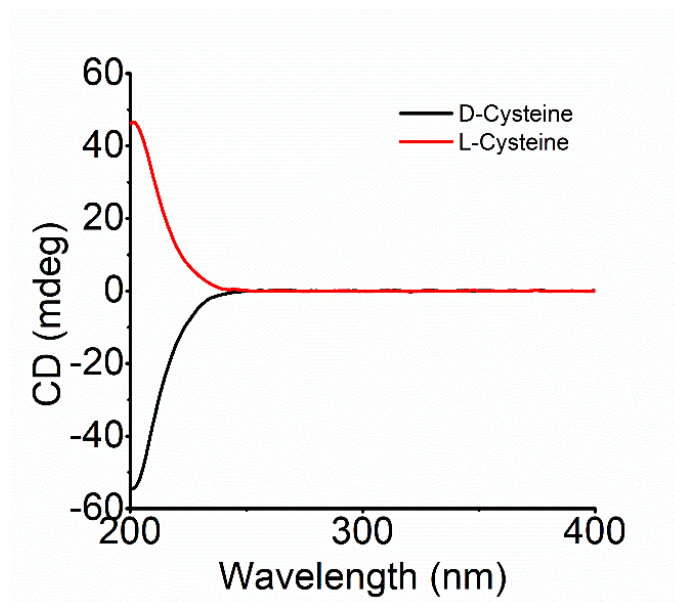**Figure S7. Extinction spectra of CdS SPs.**

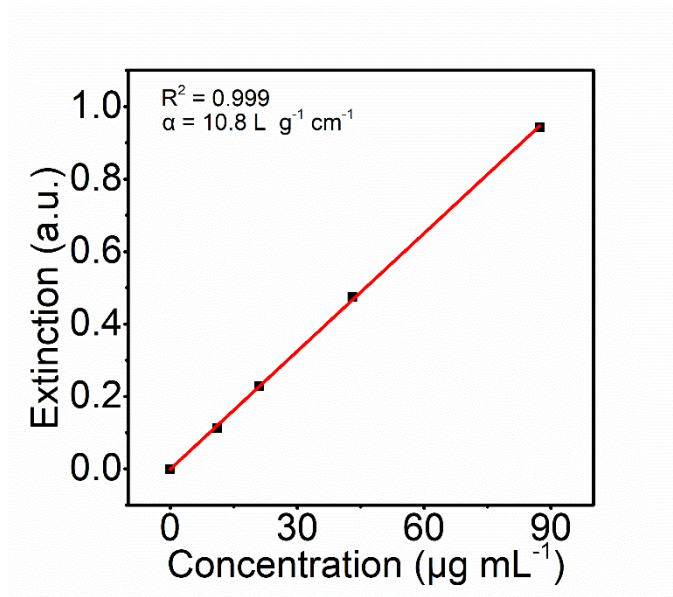**Figure S8. CD spectra of D-cysteine and L-cysteine.**

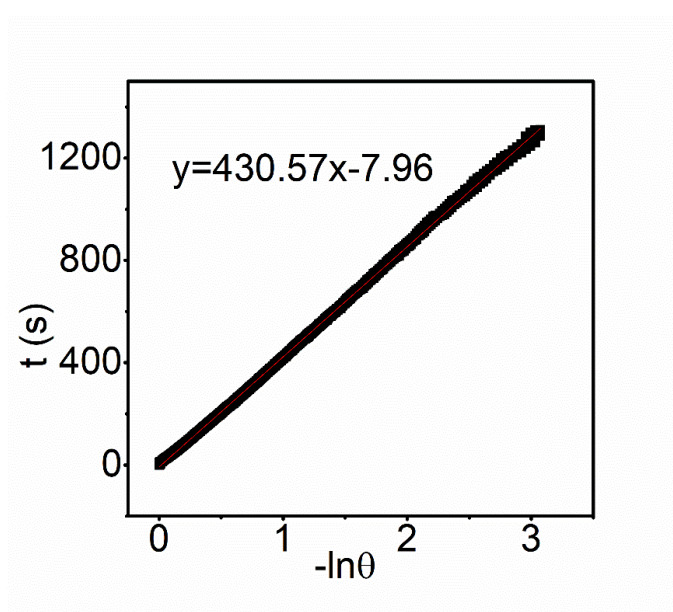**Figure S9. The linear relationship between concentration and extinction intensity of Cu$_{2-x}$S SPs.**

Figure S10. The linear diagrams of dimensionless driving factors and time for Cu$_{2-X}$S SPs in the cooling process respectively.
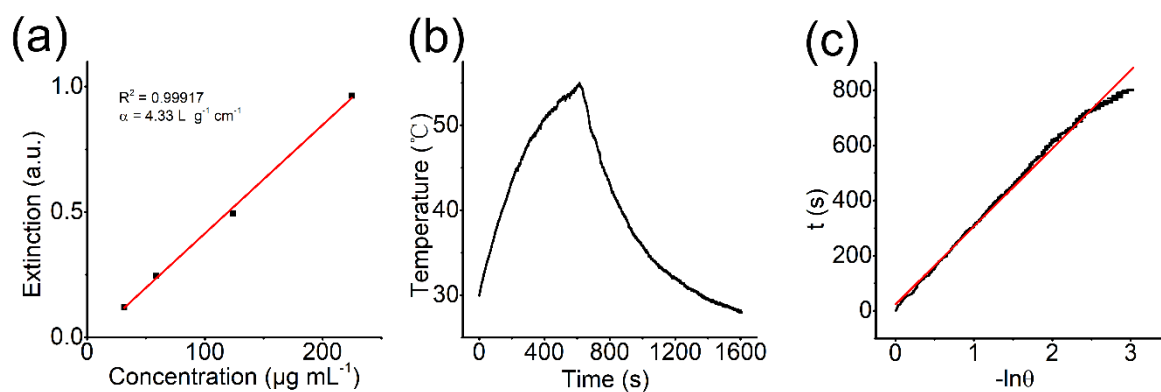
Figure S11. (A) The linear relationship between material concentration and extinction intensity at 1064 nm of $Cu_{2-x}S/Fe_2O_3$ SPs. **(B)** The temperature changes of $Cu_{2-x}S/Fe_2O_3$ SPs before and after laser irradiation at 1064 nm and 1.0 W cm^{-2} followed by natural cooling after the laser was turned off. **(C)** Linear diagrams of dimensionless driving factors and time for materials in the cooling process respectively.
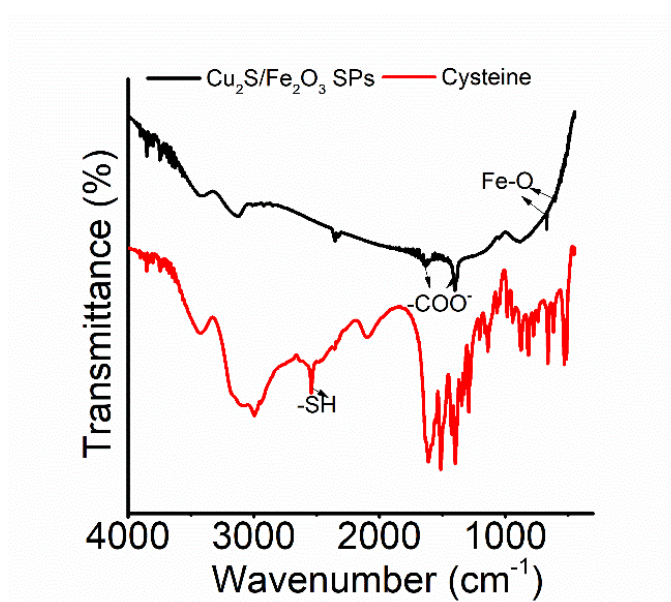

Figure S12. FT-IR spectra of Cu_2S/Fe_2O_3 SPs SPs