

Supporting Information

Generalized preparation of 2D quasi-nanosheets via self-assembly of nanoparticles

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Chemicals

The following chemicals were used as obtained: iron (III) chloride ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, 98%, Sigma-Aldrich), oleic acid (OA, 90%, Sigma-Aldrich), 1-octadecene (ODE, 90%, technical grade, Sigma-Aldrich), 1-eicosene (99%, Sigma-Aldrich), ethylene glycol (EG, 99%, Sigma-Aldrich), octyl ether (99%, TCI), dodecyltrimethylammonium bromide (DTAB, 99%, Sigma-Aldrich), sodium oleate (97%, Sigma-Aldrich), Selenium dioxide (SeO_2 , 99.9%, Sigma-Aldrich), Cadmium nitrate tetrahydrate ($\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, 99.9%, Sigma-Aldrich), cadmium acetate ($\text{Cd}(\text{C}_2\text{H}_3\text{O}_2)_2 \cdot 2\text{H}_2\text{O}$, 99.9%, Sigma-Aldrich), platinum acetylacetonate ($\text{Pt}(\text{acac})_2$, 99.9%, Sigma-Aldrich), iron pentacarbonyl, ($\text{Fe}(\text{CO})_5$, 99.9%, Sigma-Aldrich), oleylamine (OAm, 99.9%, Sigma-Aldrich), hydroxylammonium chloride (99%, Sigma-Aldrich), trioctylphosphine (99.9%, Sigma-Aldrich), cetyltrimethylammonium bromide (99%, Sigma-Aldrich), polyvinylpyrrolidone (PVP, Mw = 55,000, 99.9%, Sigma-Aldrich), hexane (99%, Sigma-Aldrich), dimethyl sulfoxide (DMSO, 99%, Sigma-Aldrich), N,N-dimethylformamide (DMF, 99%, Sigma-Aldrich), toluene (99%, Sigma-Aldrich), chloroform (99%, Sigma-Aldrich), methanol (absolute for analysis, 99%, Sigma-Aldrich), acetone and ethanol (absolute for analysis, ACS, 99.9%, Merck). All materials were used without further purification.

Synthesis of iron-oleate complex: In a typical synthesis of the iron-oleate complex, 10.8 g of iron chloride ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, 98%, Sigma-Aldrich) and 36.5 g of sodium oleate were dissolved in a mixed solvent composed of 40 mL ethanol, 40 mL deionized water and 80 mL hexane. The resulting solution was heated to 60 °C and kept at that temperature for four hours. The dark red-black iron oleate precursors were dissolved in 100 mL hexane, washed 3 times with warm DI water (~50 °C) and then separated in a separatory funnel. After washing, hexane was removed by evaporation, resulting in an iron-oleate complex in a waxy solid form.

Synthesis of iron oxide nanoparticles (NPs):¹ The following represents a typical synthesis of monodisperse 16 nm iron oxide NPs. Iron-oleate complex (3.6 g) synthesized, as described above, and 0.57 g of oleic acid (90%, Sigma-Aldrich) were dissolved in 20 g 1-eicosene at room temperature. The mixture was heated to 110 °C and maintained for 60 min under Ar protection. The reaction mixture was heated to 320 °C with a constant heating rate of 3.3 °C min⁻¹ and then held 30 min. When the reaction

temperature reached 320 °C, an obvious reaction occurred, and the initial transparent solution became turbid and brownish black. The colloidal solution was washed 3 times using ethanol /hexane (1:1 v/v) by sedimentation, and redispersal was accomplished by centrifugation (5000 rpm for 10 min). Finally, the NPs were weighed and redispersed in chloroform at the desired nanoparticle concentration.

Synthesis of Pd NPs (~ 6 nm):² Typically, Pd(acac)₂ (0.1 g) was added to 1 mL of trioctylphosphine to form an orange solution. Then 10 mL of oleylamine was introduced and the mixture was degassed for 10 min. The resulting solution was slowly heated to 250 °C (5 °C/min). After 30 min, the reaction system was cooled quickly, washed with ethanol and hexane, and finally redispersed in hexane.

Synthesis of Pt NPs (~ 3.5 nm):³ Typically, Pt(acac)₂ (0.1 g), octadecene (10 mL), oleic acid (OA) (1 mL), and oleylamine (OAm) (1 mL) were mixed under N₂ and magnetic stirring. The mixture was then heated to 65 °C to dissolve Pt(acac)₂. The temperature was then raised to about 180 °C. A solution of Fe(CO)₅ in hexane (0.1 mL, prepared by adding 0.1 mL Fe(CO)₅ in 1 mL hexane under argon) was quickly injected into the hot solution. The solution was further heated to 200 °C and kept at this temperature for 1 hour before it was cooled down to room temperature. 40 mL of isopropanol was added and then the suspension was centrifuged (8000 rpm, 10 min) to separate the NPs. The particles were dispersed in 10 mL hexane and precipitated out by adding ethanol. The process was repeated one more time to purify the NPs. The final product (50 mg) was dispersed in 10 mL of hexane for further use.

Synthesis of CdSe NPs (~ 15 nm):⁴ 0.1 mmol of CdO, 4 mmol of zinc acetate and 5 ml of oleic acid were placed in a 50 mL flask and heated to 150 °C in flowing high-purity N₂ for 30 min. Then 15 mL of 1-octadecene was added to the flask and the temperature increased to 300 °C. A stock solution containing 2 mL of trioctylphosphine, 0.2 mmol of Se and 3 mmol of S was quickly injected into the flask. The reaction temperature was maintained for 20 min and then cooled to room temperature. The resulting QDs were washed several times (minimum of three times) and finally dispersed in toluene at 10 mg mL⁻¹.

Synthesis of free-standing 2D unary QNS from Pd NPs, Pt NPs and CdSe NPs: In a typical self-assembly experiment, 15 μL ODE was added to a hexane solution of each NPs (1 mg, 500 μL) by a vortex mixer for 30 min. The remaining experimental steps were same as those for the synthesis of 2D Fe₃O₄ QNS.

Synthesis of free-standing 2D binary QNS from co-assembly of Fe₃O₄ NPs and Pd NPs: (I)

Preparation of Fe₃O₄ NP-micelles: 16 μ L ODE were dissolved into a hexane solution of 16-nm Fe₃O₄ NPs (1 mg, 0.45 mL) for 30 min by a vortex mixer. Then, an aqueous solution containing DTAB (100 μ L, 20 mg/mL) was added to the mixture. Afterwards, the mixture was heated and vigorously agitated by vortexing for 15 min. Subsequently, 0.75 mL of mixed solvent (EG and PVP) was added swiftly to the emulsion and vortexed for 90s.

(II) Preparation of Pd NP-micelles: Using the same strategy, 20 μ L ODE were used to stabilize Pd NPs (1 mg, 0.45 mL) to form NP-micelles prior to the self-assembly process, respectively.

(III) Co-assembly of Fe₃O₄ NPs and Pd NPs to form free-standing 2D binary QNS: While vortexing, 1.2 mL of above Pd NP-micelles were added into the Fe₃O₄ NP-micelles drop by drop in 1 min and subjected to further vigorous vortexing for another 2 min. The mixture was then heated to 83 °C, kept at this temperature for 120 min to evaporate the residual hexane phase, and then held for another 1 h before the suspension was cooled to room temperature. Finally, the resulting products were washed with ethanol, centrifuged at 4000 rpm for 8 min, and redispersed in ethanol.

Synthesis of free-standing 2D binary QNS from co-assembly of two types of NPs: Fe₃O₄ NPs - Pt NPs; Fe₃O₄ NPs - CdSe NPs; Pd NPs - Pt NPs; Pd NPs - CdSe NPs; Pt NPs - CdSe NPs: In a typical self-assembly experiment, 20 μ L ODE was added to a hexane solution of each NPs (1 mg, 450 μ L) by a vortex mixer for 30 min. The remaining experimental steps were same as those for the synthesis of 2D binary QNS of Fe₃O₄ - Pd.

Synthesis of free-standing 2D ternary QNS from co-assembly of three types of NPs (Fe₃O₄ NPs - Pd NPs - Pt NPs): (I) Preparation of Fe₃O₄ NP-micelles: 8 μ L ODE were dissolved into a hexane solution of 16-nm Fe₃O₄ NPs (0.5 mg, 0.40 mL) for 20 min by a vortex mixer. Then, an aqueous solution containing DTAB (50 μ L, 20 mg/mL) was added to the mixture. Afterwards, the mixture was heated and vigorously agitated by vortexing for 10 min. Subsequently, 0.75 mL of mixed solvent (EG and PVP) was added swiftly to the emulsion and vortexed for 90s.

(II) Preparation of Pd NP-micelles: 8 μ L ODE were used to stabilize 6-nm Pd NPs (0.5 mg, 0.40 mL) for 20 min by a vortex mixer. Then, an aqueous solution containing DTAB (50 μ L, 20 mg/mL) was added

to the mixture. Using the same strategy, this mixture was to form NP-micelles.

(III) Preparation of Pt NP-micelles: 10 μL ODE were used to stabilize 3.5-nm Pt NPs (0.5 mg, 0.40 mL) for 20 min by a vortex mixer. Then, an aqueous solution containing DTAB (50 μL , 20 mg/mL) was added to the mixture. Using the same strategy, this mixture was to form NP-micelles.

(IV) Co-assembly of Fe_3O_4 NPs, Pd NPs, and Pt NPs to form 2D ternary QNS: while vortexing, 1.2 mL of above Pd NP-micelles were added into the Fe_3O_4 NP-micelles drop by drop in 1 min and subjected to further vigorous vortexing for another 2 min. The next, 1.2 mL of above Pt NP-micelles were added into as-obtained mixture drop by drop in 1 min and subjected to further vigorous vortexing for another 2 min. The mixture was then heated to 83 $^\circ\text{C}$, kept at this temperature for 120 min to evaporate the residual hexane phase, and then held for another 1 h before the suspension was cooled to room temperature. Finally, the resulting products were washed with ethanol, centrifuged at 4000 rpm for 8 min, and redispersed in ethanol.

The remaining experimental steps were same as those for the synthesis of 2D ternary QNS from Fe_3O_4 NPs - CdSe NPs - Pd NPs; Fe_3O_4 NPs - CdSe NPs - Pt NPs; CdSe NPs - Pd NPs - Pt NPs.

Synthesis of free-standing 2D quaternary QNS from co-assembly of four types of NPs (Fe_3O_4 NPs - CdSe NPs - Pd NPs - Pt NPs): (I) Preparation of Fe_3O_4 NP-micelles: 9.6 μL ODE were dissolved into a hexane solution of 16-nm Fe_3O_4 NPs (0.6 mg, 0.41 mL) for 20 min by a vortex mixer. Then, an aqueous solution containing DTAB (60 μL , 20 mg/mL) was added to the mixture. Afterwards, the mixture was heated and vigorously agitated by vortexing for 10 min. Subsequently, 0.5 mL of mixed solvent (EG and PVP) was added swiftly to the emulsion and vortexed for 90 s.

(II) Preparation of CdSe NP-micelles: 9.6 μL ODE were used to stabilize 6-nm CdSe NPs (0.6 mg, 0.41 mL) for 20 min by a vortex mixer. Then, an aqueous solution containing DTAB (60 μL , 20 mg/mL) was added to the mixture. Using the same strategy, this mixture was to form NP-micelles.

(III) Preparation of Pd NP-micelles: 6.5 μL ODE were used to stabilize 3.5-nm Pd NPs (0.4 mg, 0.40 mL) for 20 min by a vortex mixer. Then, an aqueous solution containing DTAB (60 μL , 20 mg/mL) was added to the mixture. Using the same strategy, this mixture was to form NP-micelles.

(IV) Preparation of Pt NP-micelles: Using the same strategy, 8 μL ODE were used to stabilize 3.5-nm

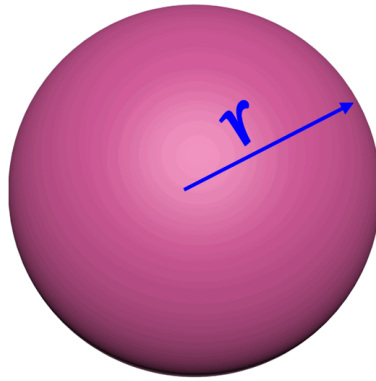
Pt NPs (0.4 mg, 0.40 mL) for 20 min by a vortex mixer. Then, an aqueous solution containing DTAB (60 μ L, 20 mg/mL) was added to the mixture. Using the same strategy, this mixture was to form NP-micelles.

(V) Co-assembly of Fe₃O₄ NPs, CdSe NPs, Pd NPs, and Pt NPs to form 2D quaternary QNS: While vortexing, the above Pd NP-micelles were added into the Fe₃O₄ NP-micelles drop by drop in 1 min and subjected to further vigorous vortexing for another 2 min. Secondly, the above Pt NP-micelles were added into as-obtained mixture drop by drop in 1 min and subjected to further vigorous vortexing for another 2 min. Thirdly, the above CdSe NP-micelles were added into as-obtained mixture drop by drop in 1 min and subjected to further vigorous vortexing for another 2 min. The mixture was then heated to 83 °C, kept at this temperature for 120 min to evaporate the residual hexane phase, and then held for another 1 h before the suspension was cooled to room temperature. Finally, the resulting products were washed with ethanol, centrifuged at 4000 rpm for 8 min, and redispersed in ethanol.

Materials Characterization

Morphology of the samples was characterized with a transmission electron microscope (TEM) system (JEOL Model JEM-2010F) operating at 200 kV, and scanning electron microscopy (SEM) system (JEOL Model 7600F). Atomic force microscopy (AFM) (Digital Instruments) was used to determine the thickness of the QNS. NMR spectra were recorded in CDCl₃ on a Varian Unity spectrometer (400 MHz for ¹H NMR) at 27 °C. All spectra were examined using MestReNova 8.1 (Mnova) software and displayed without the use of the signal suppression function.

Calculation of the number of Fe₃O₄ nanoparticles (NPs) ($N_{particles}$)



For one Fe₃O₄ NP, the radius is r . The density, mass and volume are ρ , m_1 and V_1 . The mass of one Fe₃O₄ NP (m_1) was calculated as

$$m_1 = \rho \cdot V_1 \quad (1)$$

$$V_1 = \frac{4}{3} \pi r^3 \quad (2)$$

$$m_1 = \rho \cdot V_1 = \rho \cdot \frac{4}{3} \pi r^3 \quad (3)$$

The number of Fe₃O₄ NPs ($N_{particles}$) was calculated as

$$N_{particles} = \frac{m_{Total}}{m_1} = \frac{m_{Total}}{\rho \cdot V_1} = \frac{m_{Total}}{\rho \cdot \frac{4}{3} \pi r^3} \quad (4)$$

where $\rho=5.17$ g/cm³, $d=2r=16\pm0.5$ nm, and m_{Total} is the total mass weighted in the experiment: $m_{Total}=1$ mg and $N_{particles}=9.023 \times 10^{13}$ in 1 mg total mass.

Calculation of the number of 1-octadecene (ODE) molecules (N_{ODE})

For ODE molecules, N_A is Avogadro's number ($N_A=6.022 \times 10^{23}$ mol⁻¹), n is the number of mole, M_{ODE} is the molecular weight, and m is total mass. The number of ODE molecules (N_{ODE}) was calculated as

$$N_{ODE} = n \cdot N_A \quad (5)$$

$$n = \frac{m}{M_{ODE}} \quad (6)$$

$$m = \rho \cdot V \quad (7)$$

The number of ODE molecules (N_{ODE}) was calculated as

$$N_{ODE} = n \cdot N_A = \frac{m}{M_{ODE}} \cdot N_A = \frac{\rho \cdot V}{M_{ODE}} \cdot N_A \quad (8)$$

where $\rho=0.789 \text{ g/cm}^3$, $M_{ODE}= 252.48 \text{ g/mol}$, and the ODE content of the purchased material was 90% (ODE, 90%, technical grade, Sigma-Aldrich).

Calculation of the number of ODE molecules per Fe_3O_4 NP (N_0)

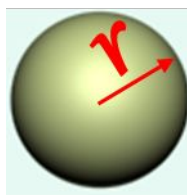
$$N_0 = \frac{N_{ODE}}{N_{particles}}$$

The N_{ODE} and N_0 were calculated as follows:

V_{ODE}	N_{ODE}	N_0
0 μL	0	0
4 μL	6.775×10^{18}	7.509×10^4
8 μL	1.355×10^{19}	1.502×10^5
16 μL	2.710×10^{19}	3.003×10^5
24 μL	4.065×10^{19}	4.505×10^5
32 μL	5.420×10^{19}	6.006×10^5
40 μL	6.775×10^{19}	7.509×10^5
48 μL	8.120×10^{19}	9.010×10^5

These data were used to prepare **Figure 2f**.

Calculation of the number of Pd NPs (N_{Pd})



For one Pd NP, the radius is r . The density, mass and volume are ρ , m_2 and V_2 . The mass of one Pd NP (m_2) was calculated as

$$m_2 = \rho \cdot V_2 \quad (9)$$

$$V_2 = \frac{4}{3} \pi r^3 \quad (10)$$

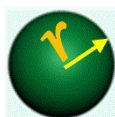
$$m_2 = \rho \cdot V_2 = \rho \cdot \frac{4}{3} \pi r^3 \quad (11)$$

The number of Pd NPs (N_{Pd}) was calculated as

$$N_{Pd} = \frac{m_{Total}}{m_2} = \frac{m_{Total}}{\rho \cdot V_2} = \frac{m_{Total}}{\rho \cdot \frac{4}{3} \pi r^3} \quad (12)$$

where $\rho=11.9 \text{ g/cm}^3$, $d=2r=6\pm0.5 \text{ nm}$, and m_{Total} is the total mass weighted in the experiment: $m_{Total}=1 \text{ mg}$ and $N_{Pd}=7.433 \times 10^{14}$ in 1 mg total mass.

Calculation of the number of Pt NPs (N_{Pt})



For one Pt NP, the radius is r . The density, mass and volume are ρ , m_3 and V_3 . The mass of one Pt NP (m_3) was calculated as

$$m_3 = \rho \cdot V_3 \quad (13)$$

$$V_3 = \frac{4}{3} \pi r^3 \quad (14)$$

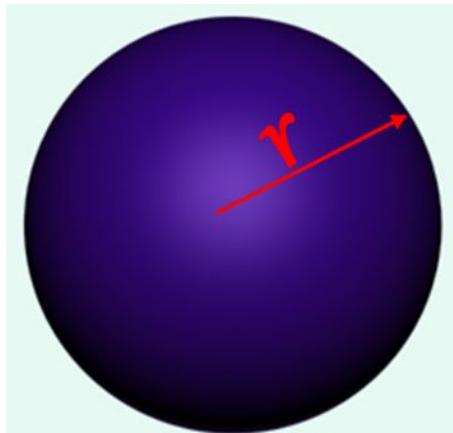
$$m_3 = \rho \cdot V_3 = \rho \cdot \frac{4}{3} \pi r^3 \quad (15)$$

The number of Pt NPs (N_{Pt}) was calculated as

$$N_{Pt} = \frac{m_{Total}}{m_3} = \frac{m_{Total}}{\rho \bullet V_3} = \frac{m_{Total}}{\rho \bullet \frac{4}{3} \pi r^3} \quad (16)$$

where $\rho=21.45$ g/cm³, $d=2r=3.5\pm0.5$ nm, and m_{Total} is the total mass weighted in the experiment: $m_{Total}=1$ mg and $N_{Pt}=2.078 \times 10^{15}$ in 1 mg total mass.

Calculation of the number of CdSe NPs (N_{CdSe})



For one CdSe NP, the radius is r . The density, mass and volume are ρ , m_4 and V_4 . The mass of one CdSe NP (m_4) was calculated as

$$m_4 = \rho \bullet V_4 \quad (17)$$

$$V_4 = \frac{4}{3} \pi r^3 \quad (18)$$

$$m_4 = \rho \bullet V_4 = \rho \bullet \frac{4}{3} \pi r^3 \quad (19)$$

The number of CdSe NPs (N_{CdSe}) was calculated as

$$N_{CdSe} = \frac{m_{Total}}{m_4} = \frac{m_{Total}}{\rho \bullet V_4} = \frac{m_{Total}}{\rho \bullet \frac{4}{3} \pi r^3} \quad (20)$$

where $\rho=5.82$ g/cm³, $d=2r=15\pm0.5$ nm, and m_{Total} is the total mass weighted in the experiment: $m_{Total}=1$ mg and $N_{CdSe}=9.728 \times 10^{13}$ in 1 mg total mass.

Calculation of the mole ratio of NPs (N_0) and mass ratio of NPs (M_0) for 2D binary QNS from co-assembly of two types of NPs

<i>Binary QNS</i>	<i>ratio</i>	N_0	M_0
Fe ₃ O ₄ - Pd	$N_{Fe_3O_4} : N_{Pd}$	1 : 8.23	1 : 1
Fe ₃ O ₄ - Pt	$N_{Fe_3O_4} : N_{Pt}$	1 : 23.03	1 : 1
Fe ₃ O ₄ - CdSe	$N_{Fe_3O_4} : N_{CdSe}$	1 : 1.08	1 : 1
Pd - Pt	$N_{Pd} : N_{Pt}$	1 : 2.80	1 : 1
CdSe - Pd	$N_{CdSe} : N_{Pd}$	1 : 7.64	1 : 1
CdSe - Pt	$N_{CdSe} : N_{Pt}$	1 : 21.36	1 : 1

Calculation of the mole ratio of NPs (N_0) and mass ratio of NPs (M_0) for 2D ternary QNS from co-assembly of three types of NPs

<i>Ternary QNS</i>	<i>ratio</i>	N_0	M_0
Fe ₃ O ₄ - Pd - Pt	$N_{Fe_3O_4} : N_{Pd} : N_{Pt}$	1 : 8.23 : 23.03	1 : 1 : 1
Fe ₃ O ₄ - CdSe - Pd	$N_{Fe_3O_4} : N_{CdSe} : N_{Pd}$	1 : 1.08 : 8.23	1 : 1 : 1
Fe ₃ O ₄ - CdSe - Pt	$N_{Fe_3O_4} : N_{CdSe} : N_{Pt}$	1 : 1.08 : 23.03	1 : 1 : 1
CdSe - Pd - Pt	$N_{CdSe} : N_{Pd} : N_{Pt}$	1 : 7.64 : 21.36	1 : 1 : 1

Calculation of the mole ratio of NPs (N_0) and mass ratio of NPs (M_0) for 2D quaternary QNS from co-assembly of four types of NPs

<i>Quaternary QNS</i>	<i>ratio</i>	N_0	M_0
Fe ₃ O ₄ - CdSe - Pd - Pt	$N_{Fe_3O_4} : N_{CdSe} : N_{Pd} : N_{Pt}$	1 : 1.08 : 5.49 : 15.35	1 : 1 : 0.67 : 0.67

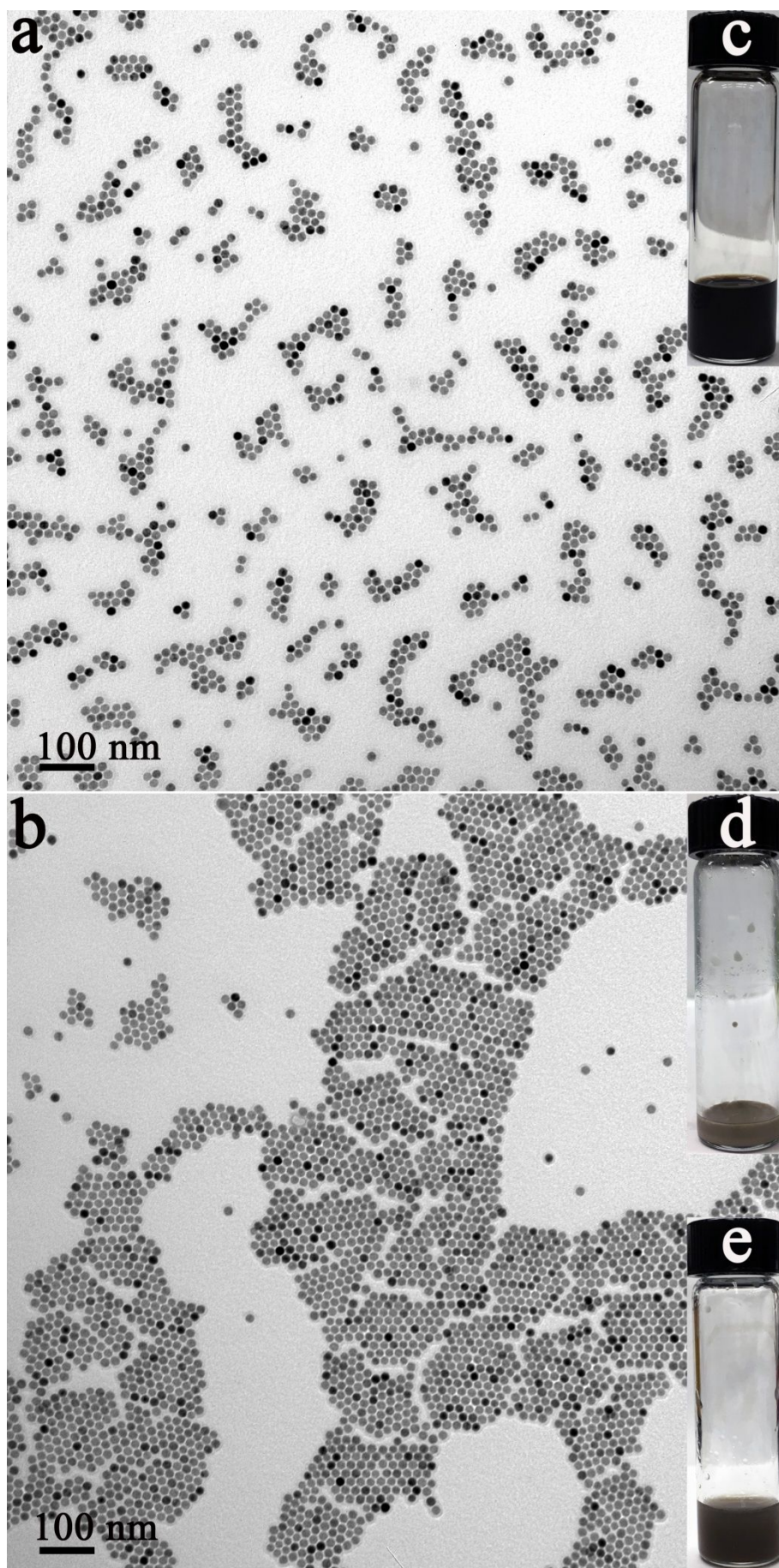


Figure S1 TEM images of (a) Fe₃O₄ NPs (~16 nm) and (b) Fe₃O₄ NP micelles (contained ODE and DTAB); photograph of (c) Fe₃O₄ NPs (~16 nm), (d) Fe₃O₄ NP micelles and (e) mixed solution (EG and PVP) added into NP micelles (before thermal annealing).

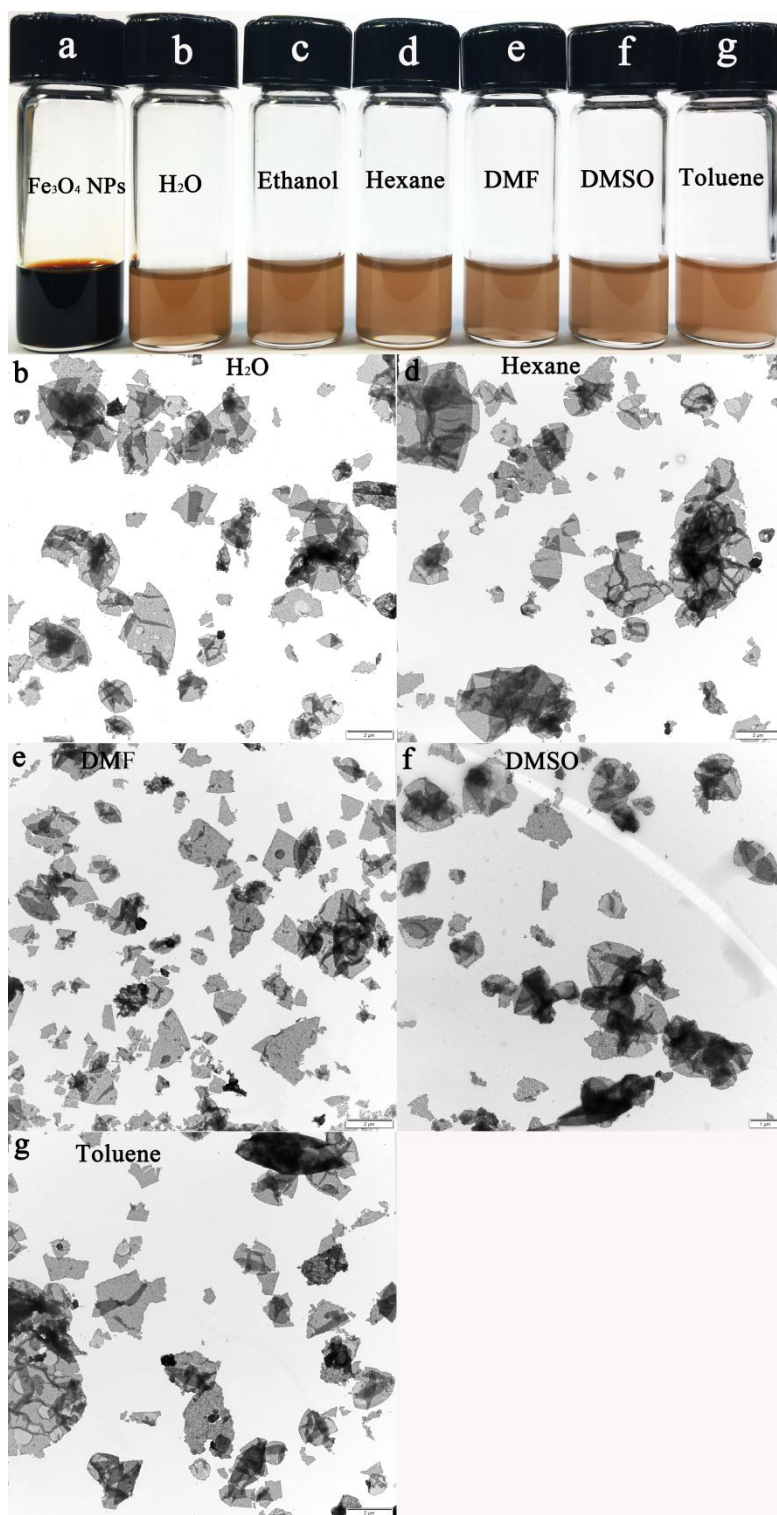


Figure S2 The TEM images of free-standing 2D QNS in different solvents after keeping 1 day: a) Fe₃O₄ NP building blocks (**Figure S1**); b) DI-water; c) ethanol (**Figure 1**); d) hexane; e) DMF; f) DMSO; g) toluene.

Note: To investigate the stability, we dispersed 2D QNS in different solvents and kept them for 7 days at room temperature. From **Figure S2b-g**, the color of all solutions is yellow, which is totally

different from that of their NP building blocks (black, **Figure S2a**). Our TEM results confirmed that the samples were stable without disassembly (**Figure S2** and **Figure 1**). Therefore, as-assembled 2D QNS exhibited excellent stability in different solvents (DI-water, ethanol, hexane, DMF, DMSO, toluene) for long time rather than disassembly into their NP building blocks.

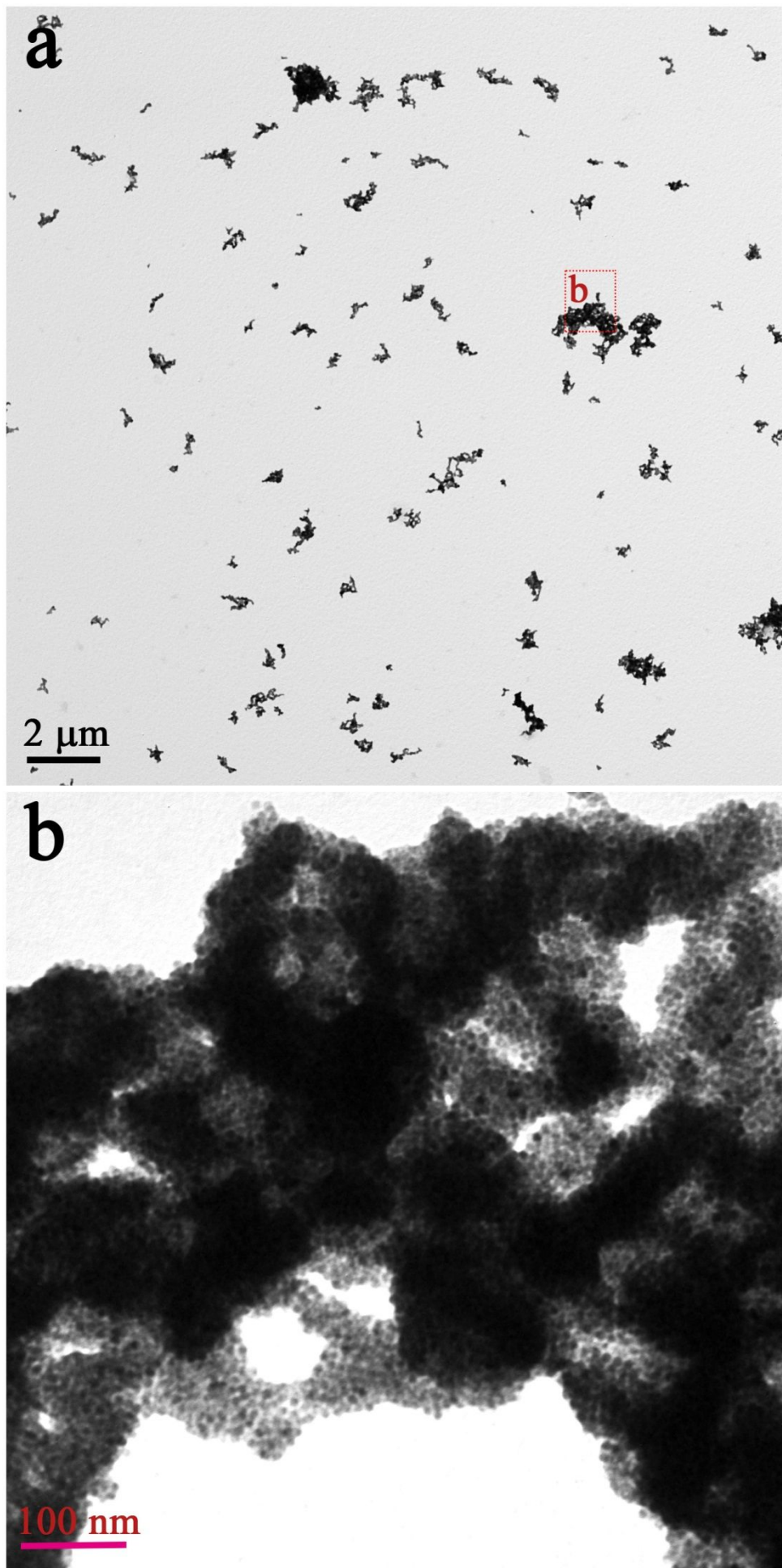


Figure S3 TEM images of samples prepared from Fe₃O₄ NPs assembly without DTAB.

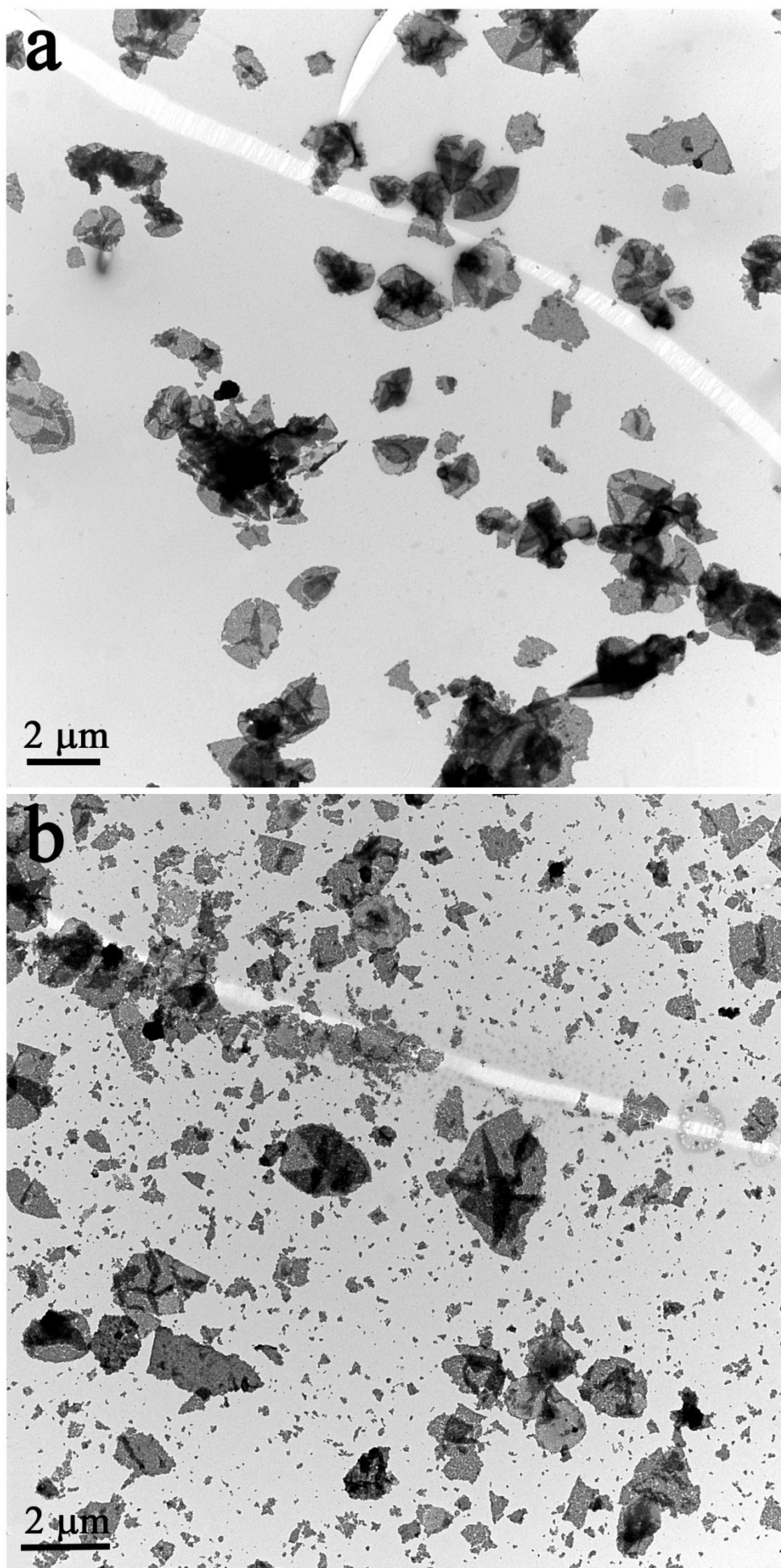


Figure S4 TEM images of samples prepared from Fe_3O_4 NPs assembly using different amounts of ODE: a) 16 μL and b) 40 μL .

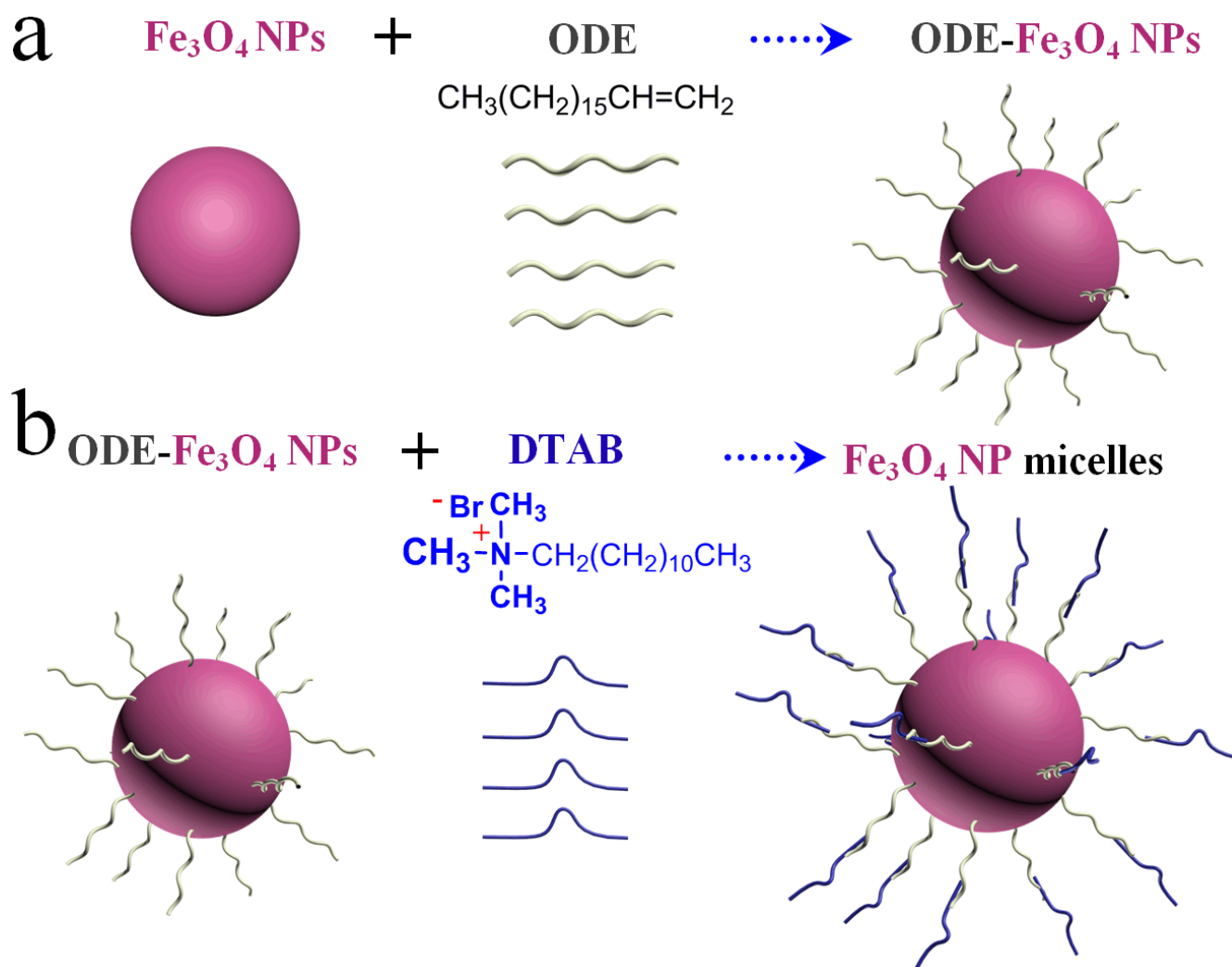


Figure S5 Scheme of Fe_3O_4 NP micelles: a) ODE-capped NPs; b) NP micelles.

Note: These micelles were synthesized as follows: A DTAB solution was added to ODE-capped Fe_3O_4 NPs ($\text{ODE-Fe}_3\text{O}_4$, **Figure S5a**) in chloroform and heated to create a water-in-oil microemulsion (**Figure S5b**). After injection, some NP micelles were quickly encapsulated by mixed solvent (EG and PVP), which is a protective layer to stabilize these capping micelles through repulsive steric interactions (**Figure 3a**).⁵ During the thermal-annealing process (83 °C, 4 h), chloroform evaporated from the system, and the DTAB layer (secondary layer, **Figure S5b**) detached from the surface of NP micelles, because of the good solubility of DTAB in mixed solvent.⁶ This led to micelle decomposition to reform $\text{ODE-Fe}_3\text{O}_4$ NPs in polar solvent (EG) (**Figure 3b**). Finally, the distance between NPs gradually increases because of the increase of ligand elastic repulsion,⁷ weakening the attractive forces between NPs and facilitating the spread of NPs (**Figure 3c**),⁸ finally leading to the formation of 2D lamellar structures (**Figure 2c-d**, **Figure 3d** and **Figure 1**).⁹

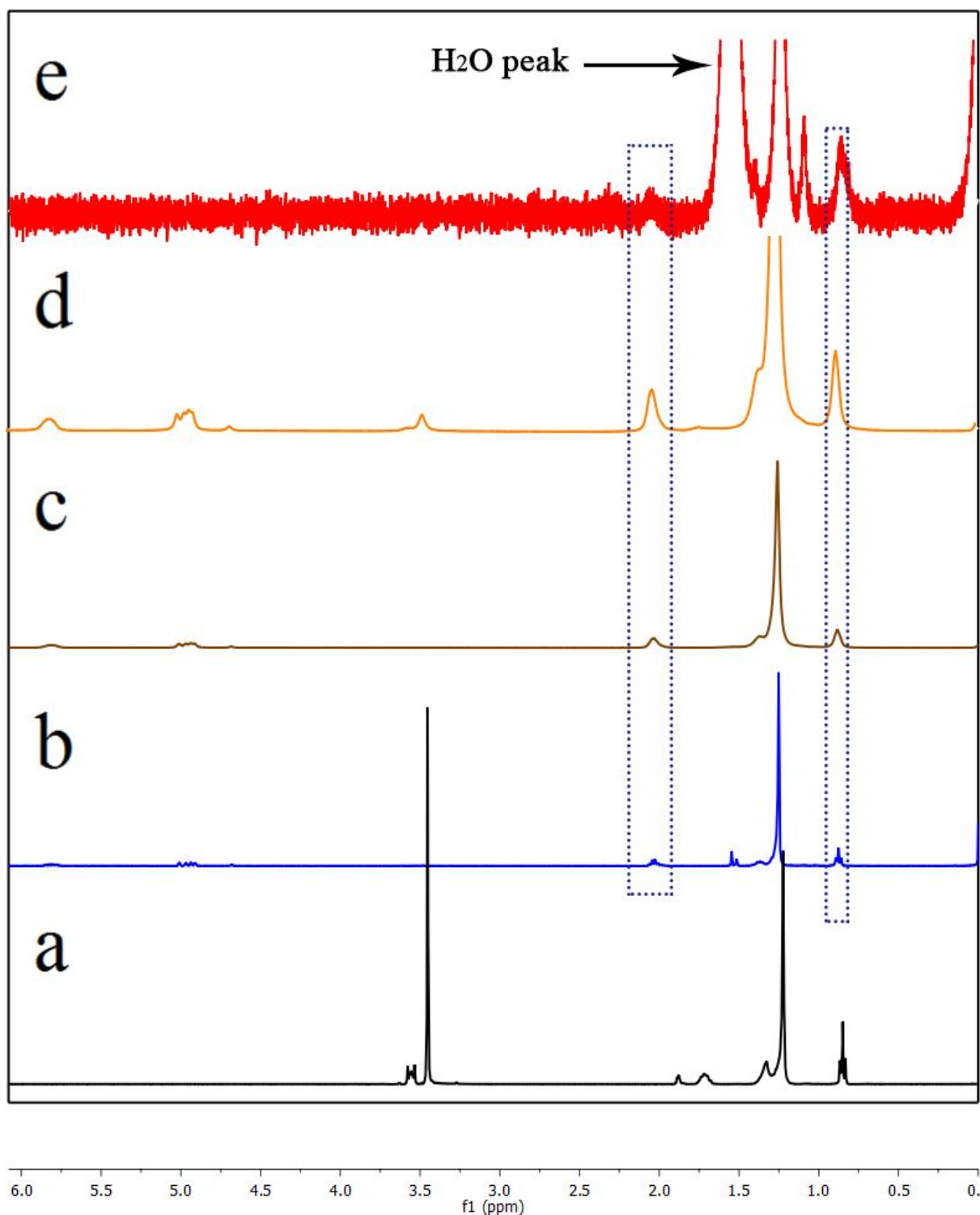


Figure S6 ^1H NMR spectra of a) DTAB; b) ODE; c) ODE-capped Fe_3O_4 NPs; d) Fe_3O_4 NP micelles (DTAB and ODE capped on the Fe_3O_4 NPs); and e) the residual organic ligands (ODE) on the Fe_3O_4 QNS. (^1H NMR of Fe_3O_4 QNS was performed after HCl was used to dissolve them in a vial, then, dried it at 80°C oven for overnight. ^1H NMR spectra were recorded in CDCl_3 on a Varian Unity spectrometer (400 MHz for ^1H NMR) at 27°C).

Note: ^1H NMR spectra show that the organic ligands on the surface of NP micelles were both ODE and DTAB, as shown in **Figure S6d**. In contrast, the residual organic ligands on the surface of Fe_3O_4 QNS were only ODE, while DTAB was not measurable in **Figure S6e**. ^1H NMR results confirmed the loss of DTAB and presence of ODE during the assembly process.

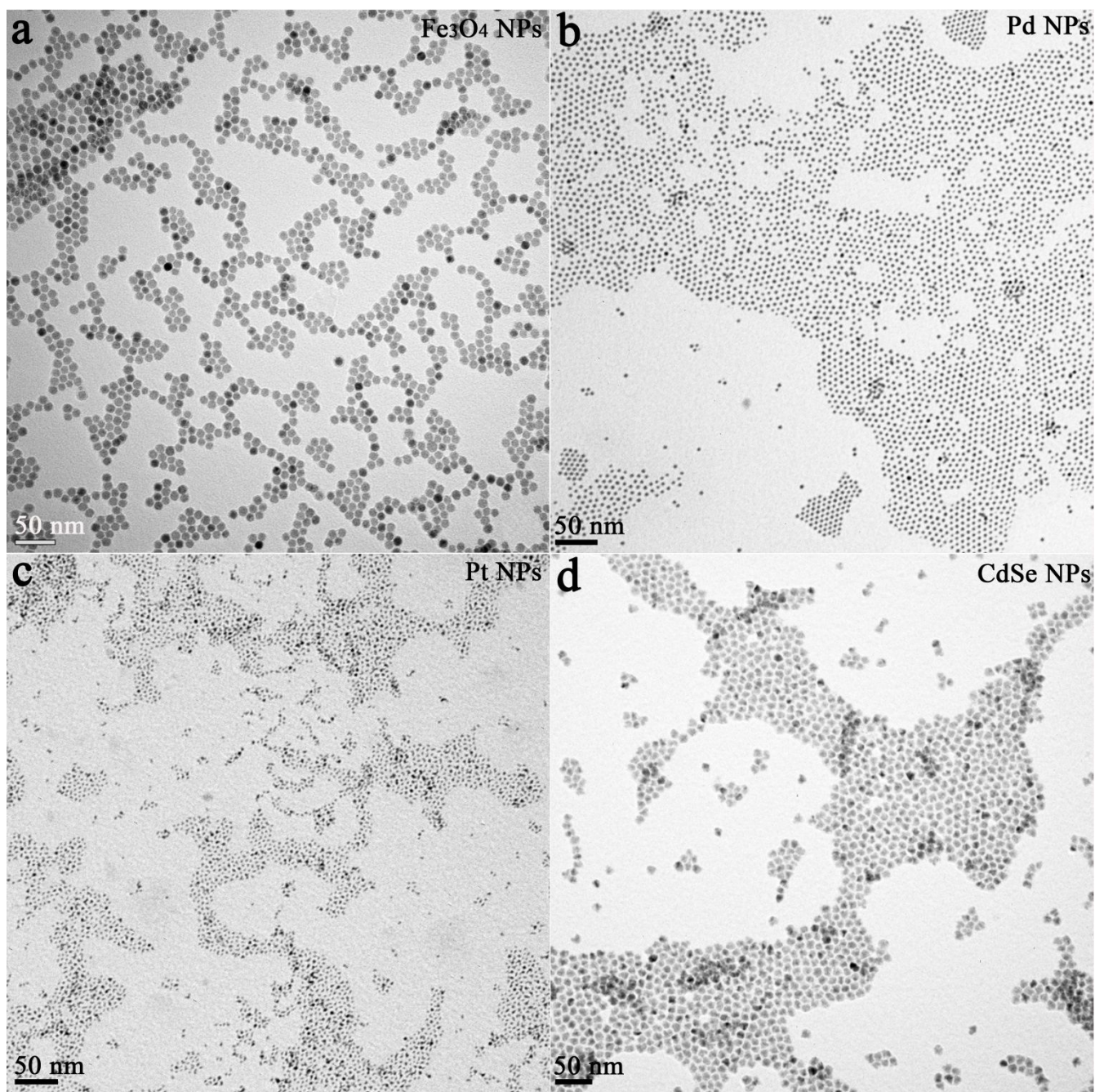


Figure S7 TEM images of a) Fe₃O₄ NPs (~16 nm); b) Pd NPs (~6 nm); c) Pt NPs (~ 3.5 nm); d) CdSe NPs (~15 nm).

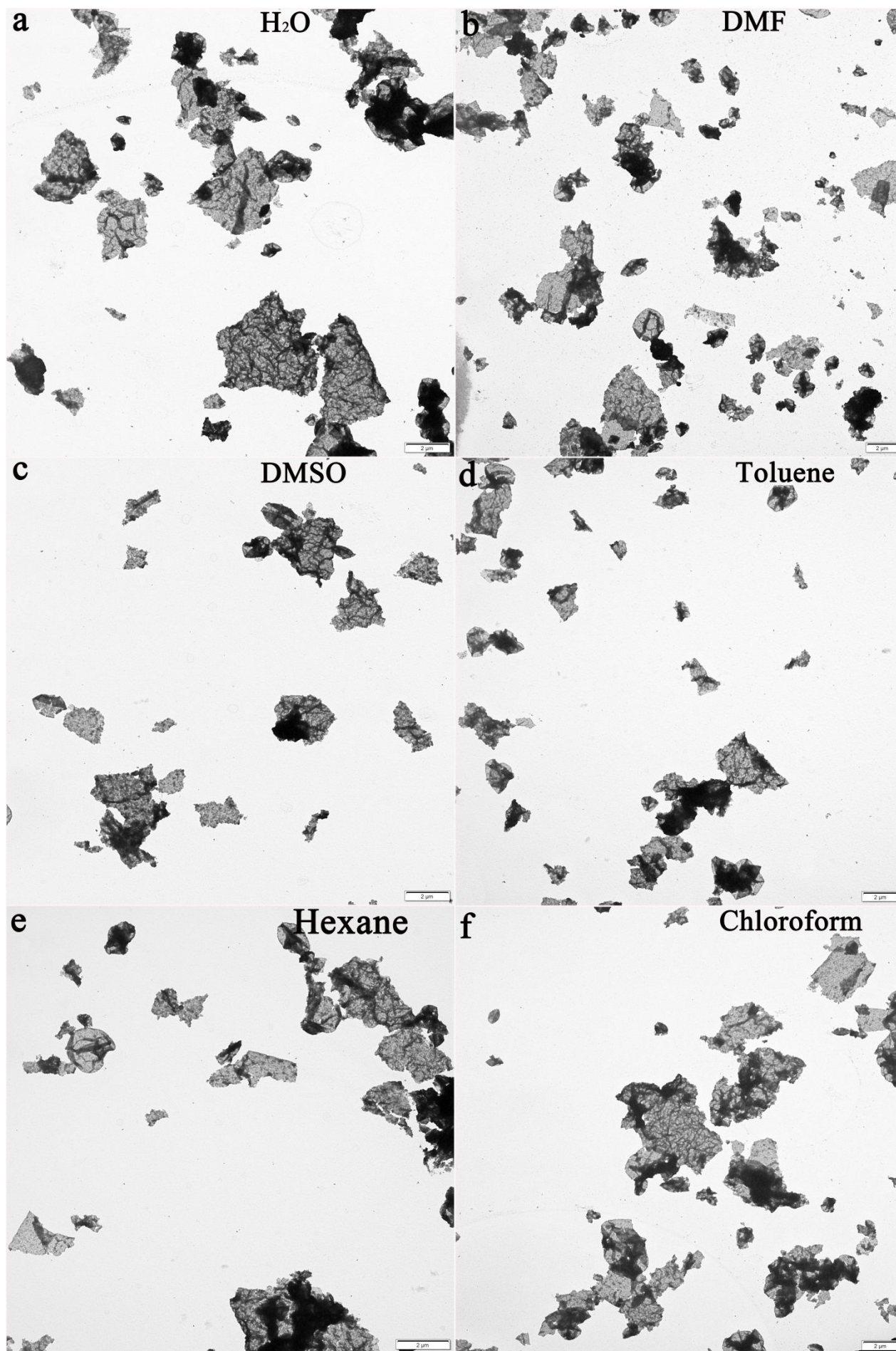


Figure S8 The TEM images of free-standing 2D binary QNS (Fe₃O₄ NPs and Pd NPs) in different solvents after keeping day: a) DI-water; b) DMF; c) DMSO; d) toluene; e) hexane; d) chloroform; **Figure 1** for sample in ethanol.

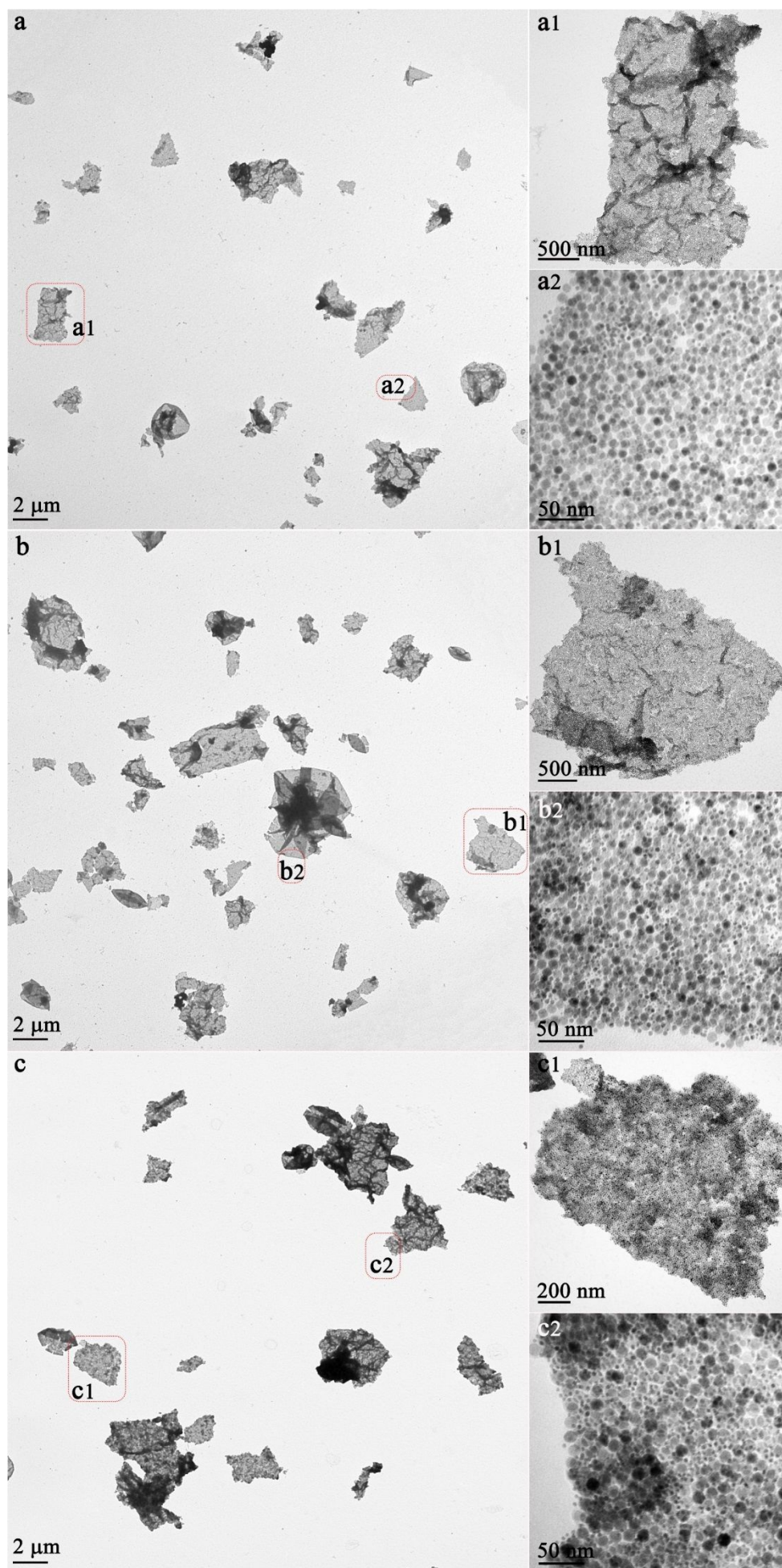


Figure S9 TEM images of 2D binary QNS from co-assembly of Fe₃O₄ NPs and Pd NPs with different Fe₃O₄/Pd particle ratios: a-a2) 1:1; b-b2) 1:2; c-c2) 1:8.23.

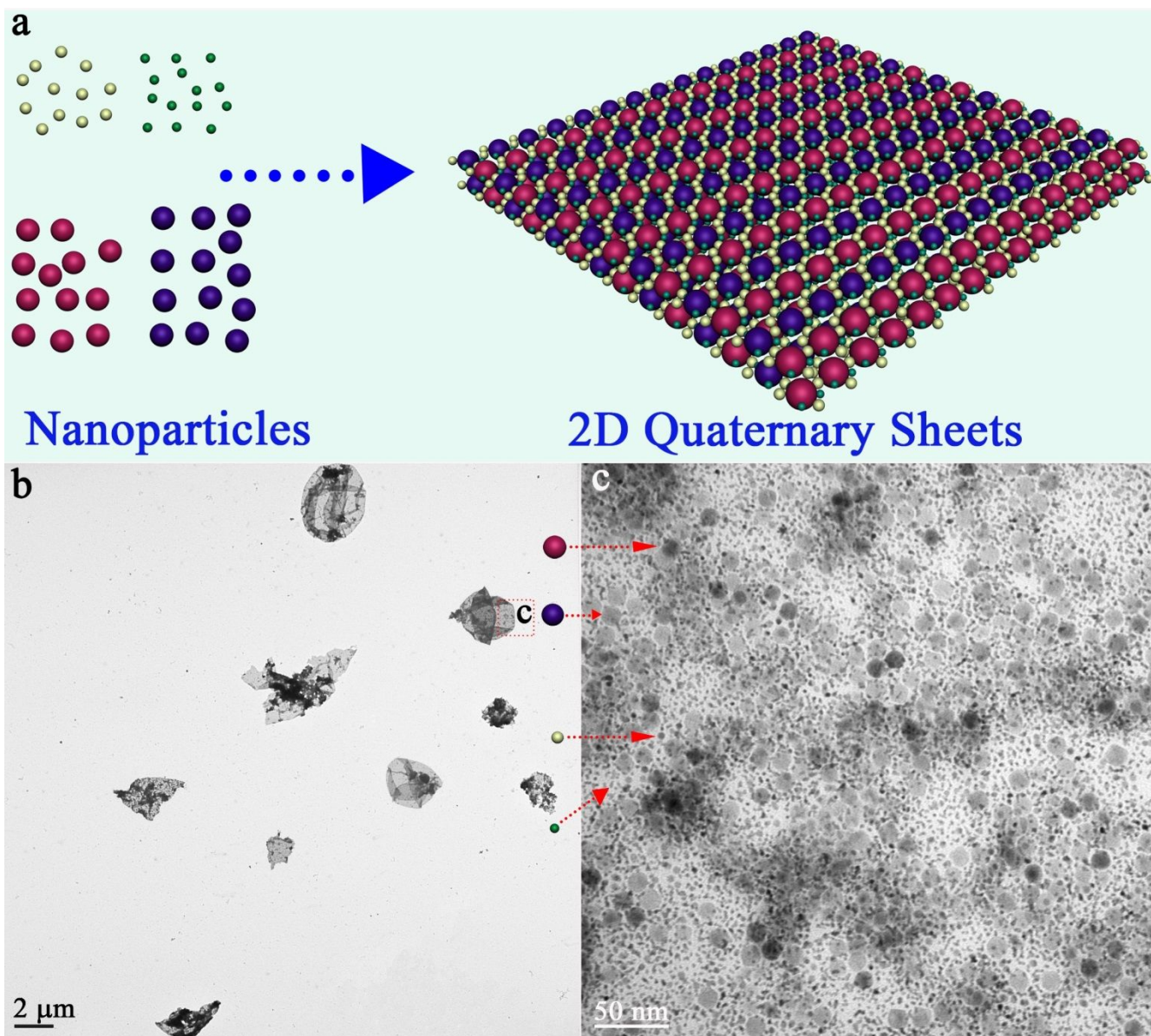


Figure S10 a) Scheme for 2D quaternary QNS from assembly of four types of NPs; b-c) TEM images of 2D quaternary QNS of Fe_3O_4 - CdSe - Pd - Pt.

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