

## \*Electronic Supplementary Information

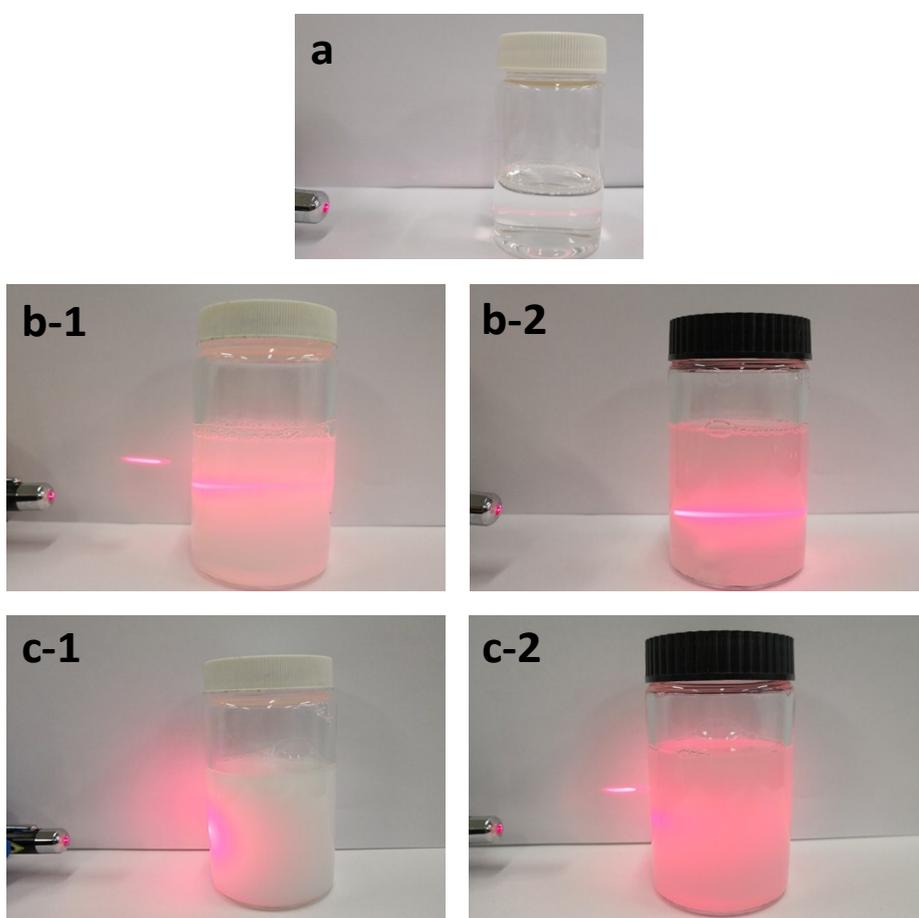
### Onion-like Metal-Organic Colloidosomes from Counterion-induced Self-Assembly of Anionic Surfactant

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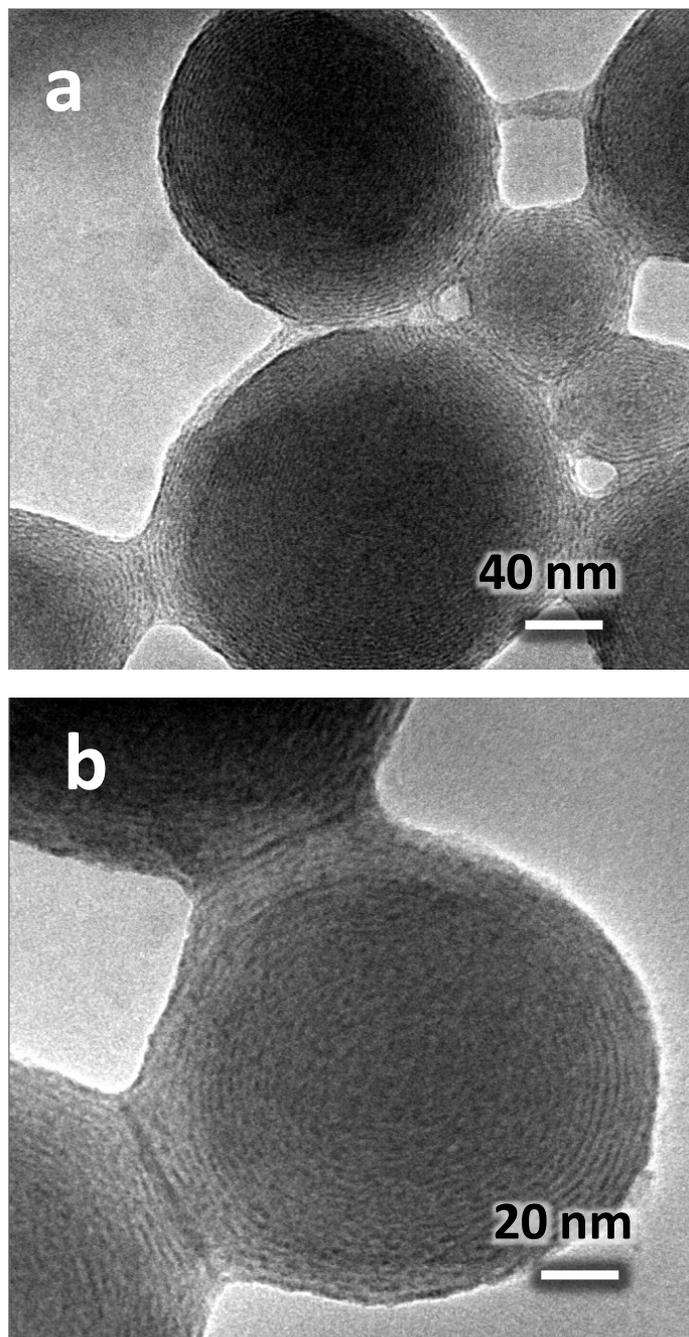
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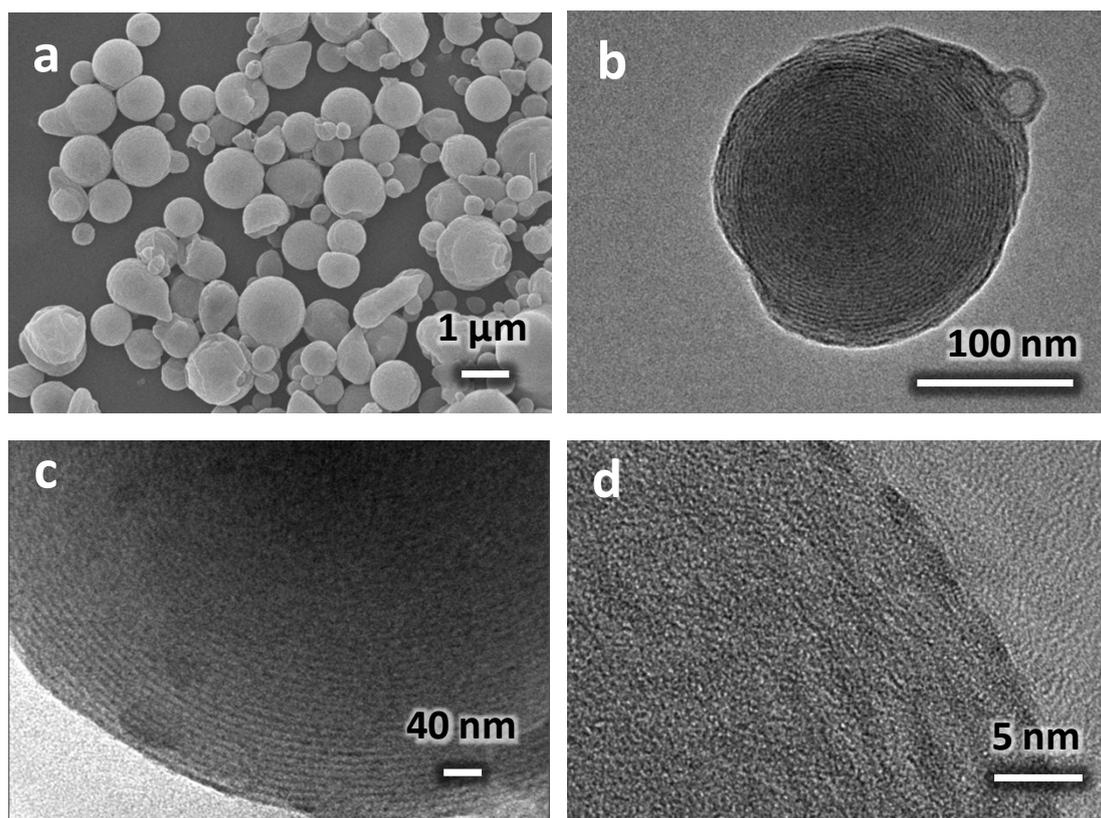
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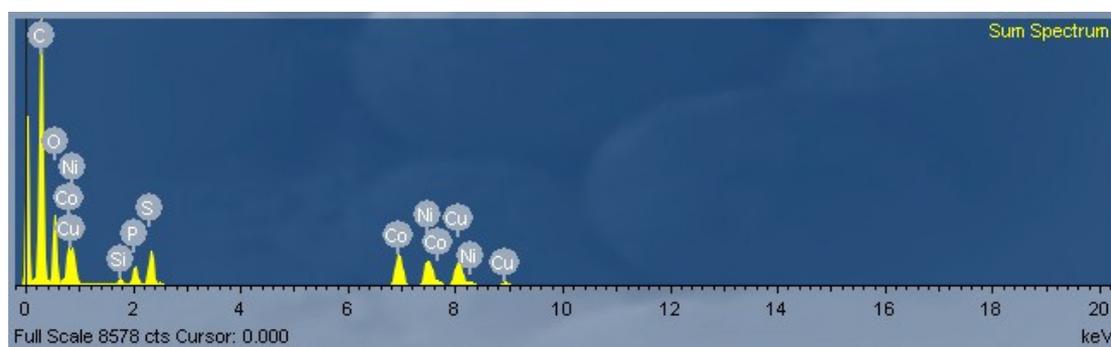
**Figure S1.** Tydall effects for: (a) pure SDBS solution; (b-1) SDBS solution with Ni(II) or (b-2) Co(II) added; (c-1) solutions in (b) added with  $N_2H_4$  for the (c-1) Ni(II) and (c-2) Co(II) cases.



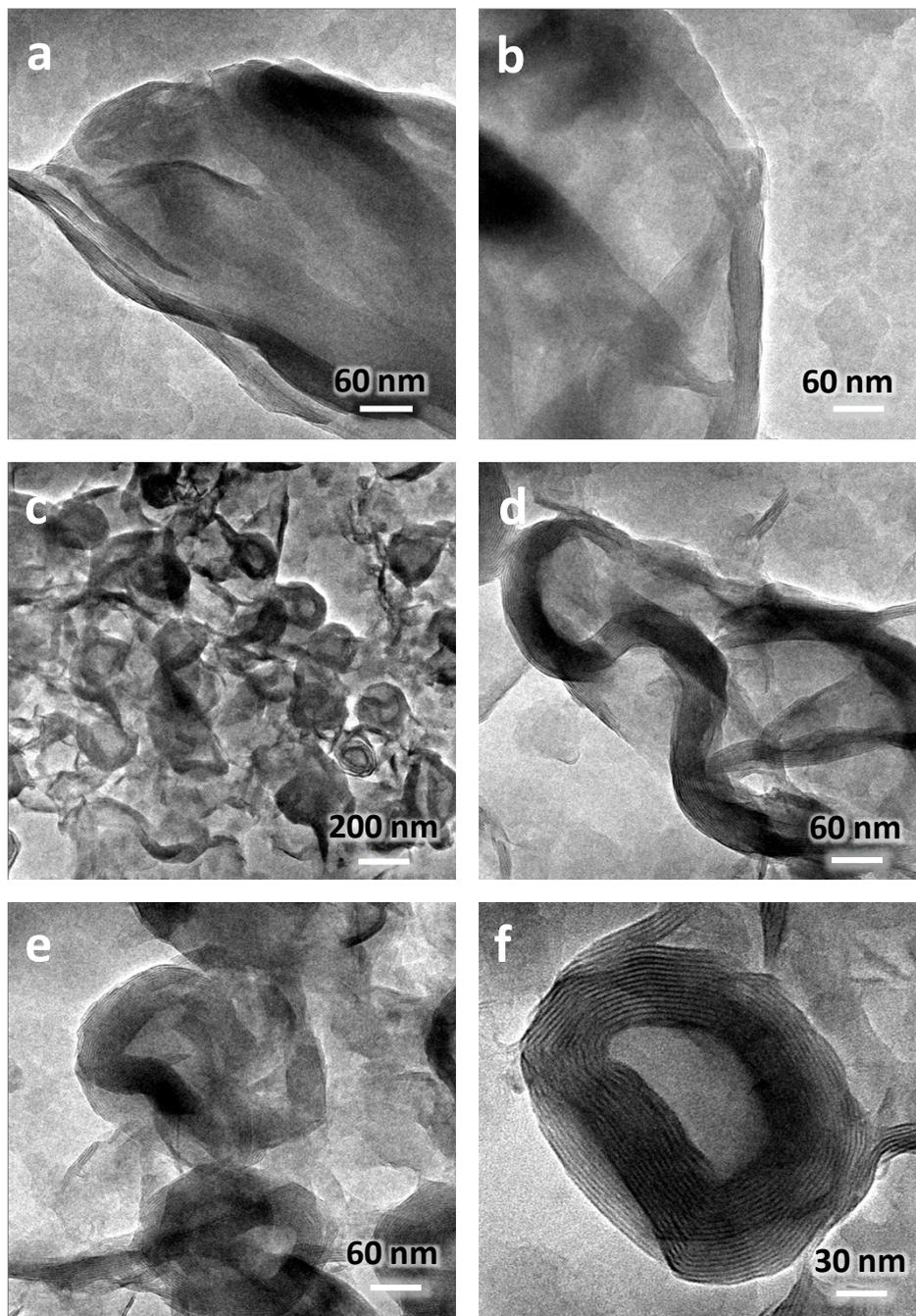
**Figure S2.** TEM images of the onion-like colloidosomes for the Co(II) case.



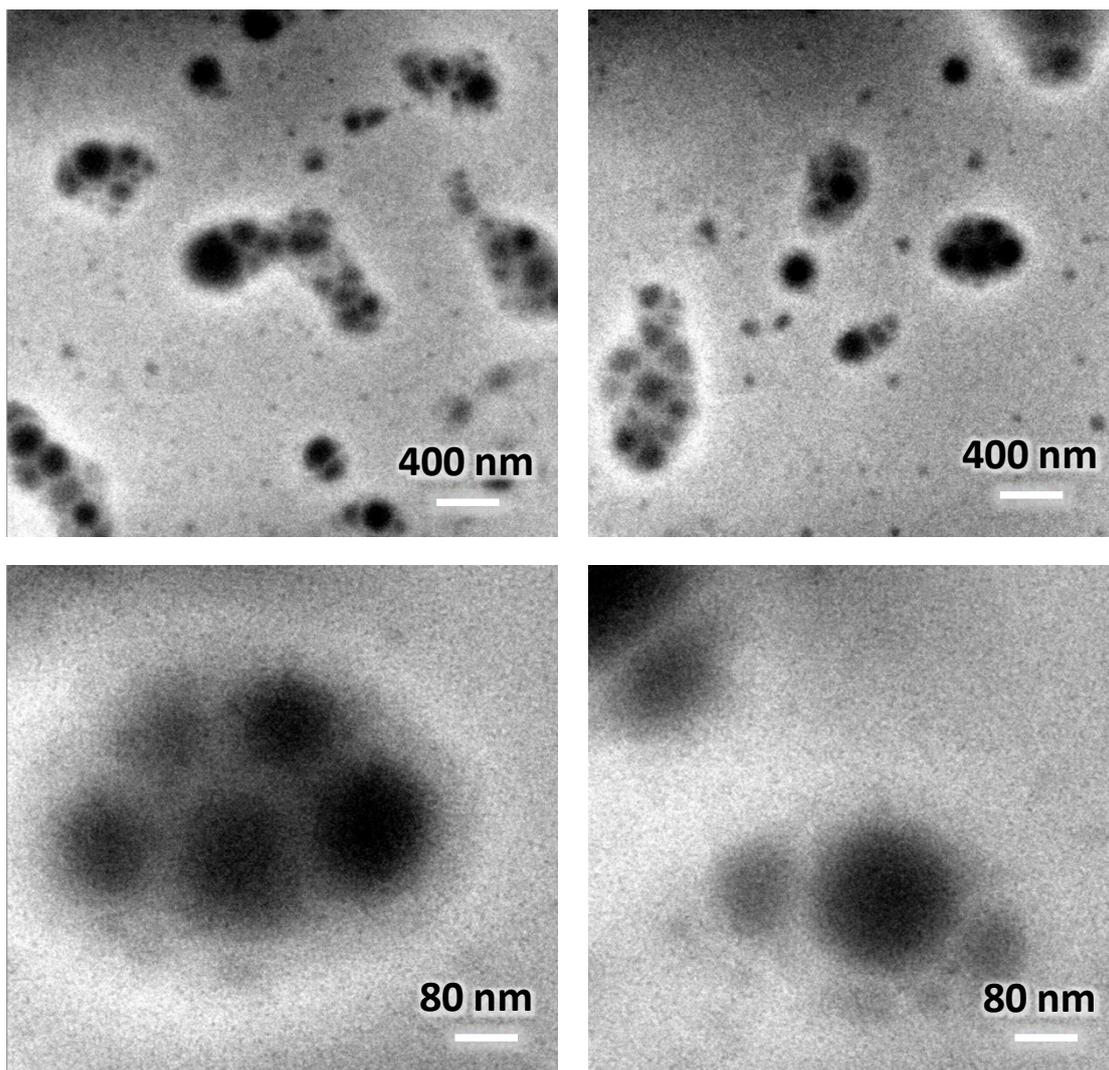
**Figure S3.** (a) FESEM image, (b,d) TEM images and (c) HRTEM image of onion-like colloidosomes in the Ni(II) case.



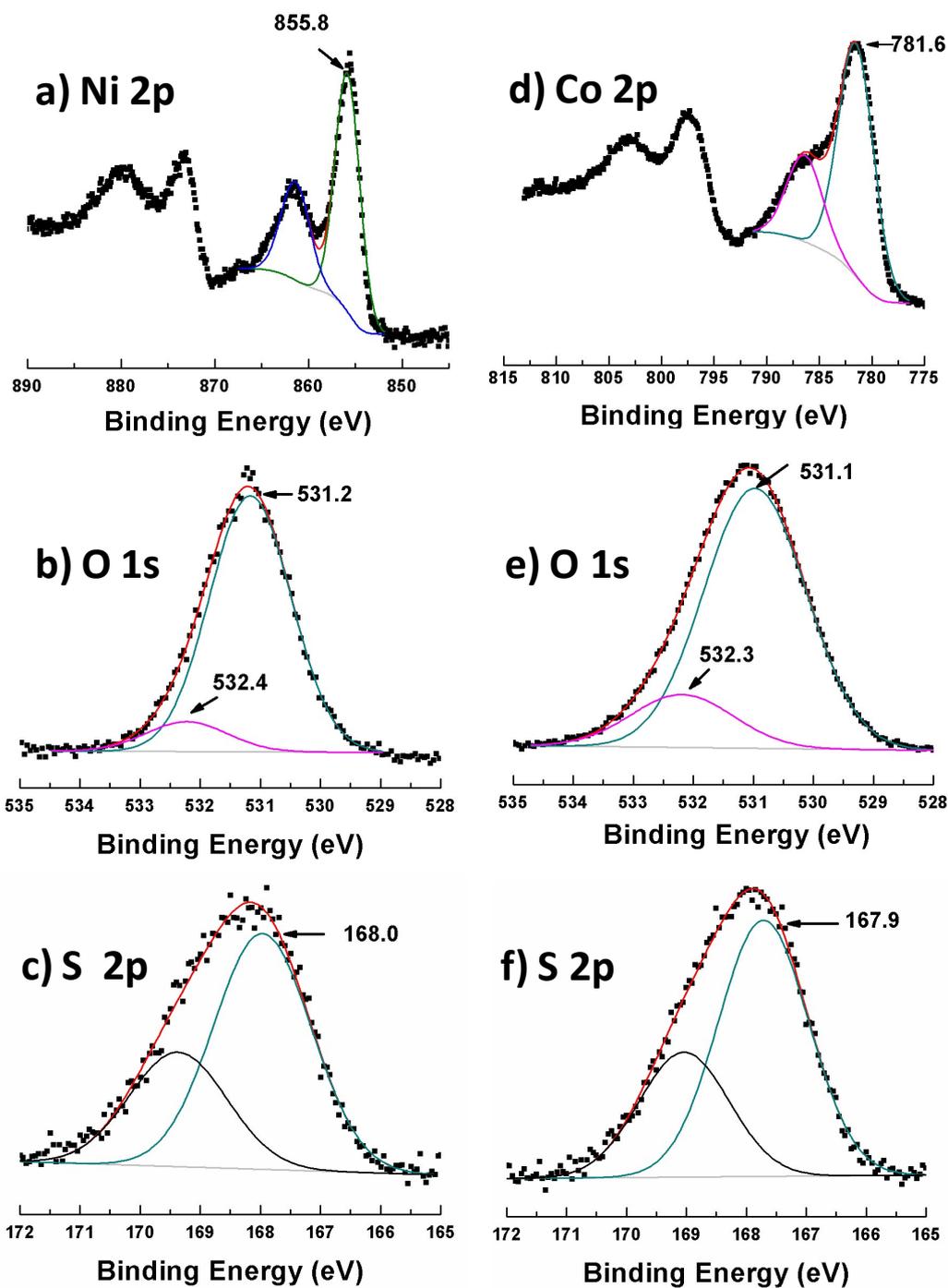
**Figure S4.** EDX spectrum of the binary onion-like colloidosome consisting of both Ni(II) and Co(II) cations. Note that the elements Co, Ni, O and S are all found within the sample, and the signals of Cu are from the copper grid and the signals of Si, and P are noise signals.



**Figure S5.** TEM images of the multishelled-structures in the Ni(II) case with a molar ratio of SDBS to Ni<sup>2+</sup> as (a,b) 3:1 and (c,d,e,f) 4:1. Note that the samples were prepared by mixing the Ni<sup>2+</sup> precursor (2.0 ml, 0.1 M) and SDBS (2.0-8.0 ml, 0.1 M) into 20.0 ml H<sub>2</sub>O, followed by the addition of N<sub>2</sub>H<sub>4</sub> (1.2 ml, 7%), which was then treated with urea (22 mg) at 120°C for 15 h.



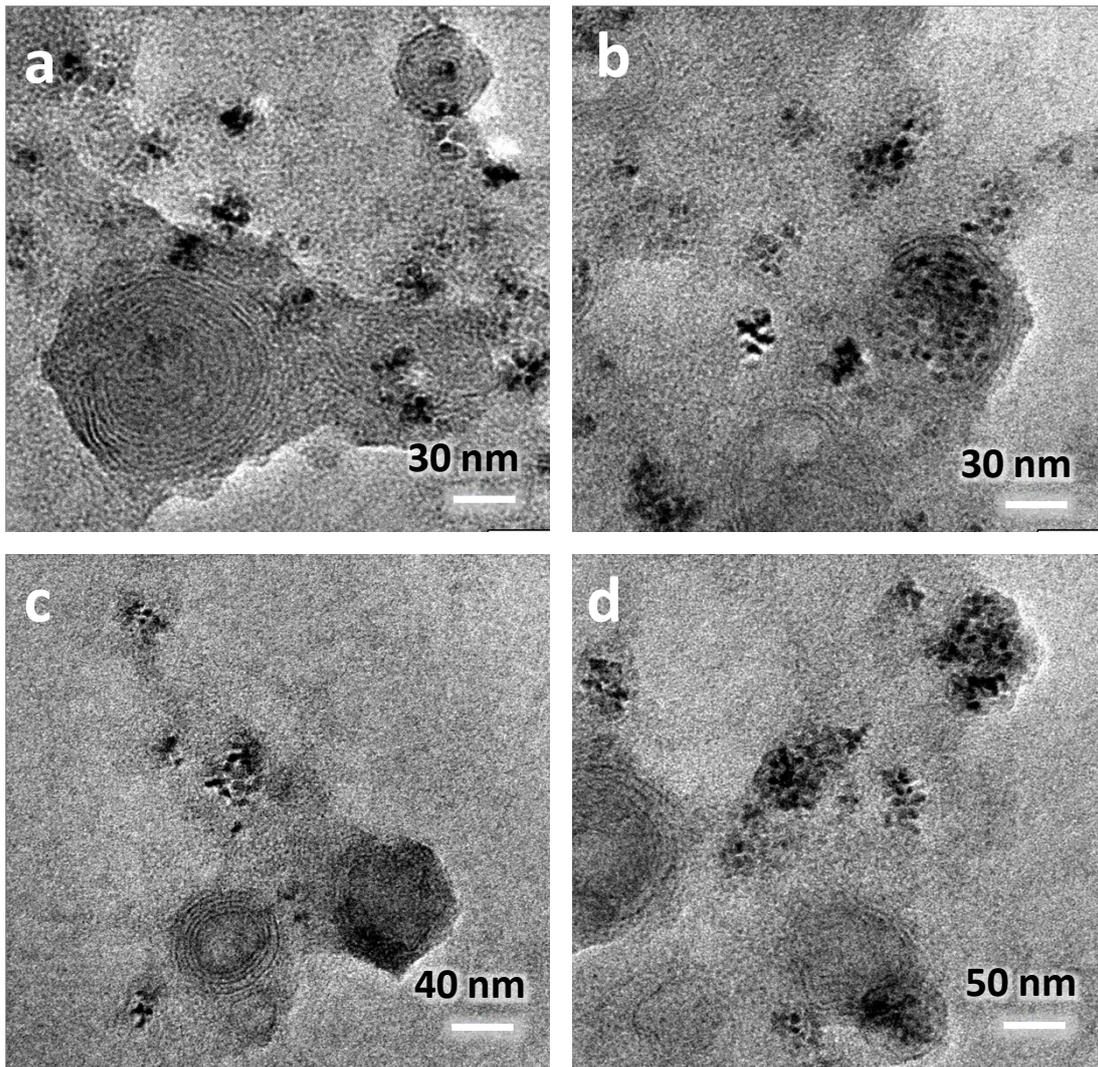
**Figure S6.** TEM images (with different magnifications) of the pristine sample (i.e., those in Figure 1b in the main text) in the Co(II) case. Note that the TEM sample for observation was prepared directly using the as-formed pristine solution, therefore the sample was covered with a thick layer of surfactant.



**Figure S7.** XPS spectra: (a) Ni 2p, (b) O 1s, (c) S2p in the pristine sample of the Ni(II) case; (d) Co 2p, (e) O 1s and (c) S 2p in the pristine sample of the Co(II) case. Note that the dots are the experimental data and the curves are calculated, and for the Ni, Co and S elements, only the BE energies of their relevant 2p<sub>3/2</sub> were shown.

**Table S1.** Specific conditions to prepare the samples in Figure 4 in the main text.

Samples	Specific experimental conditions
Sample in Figure 4a,b	20 ml H <sub>2</sub> O + 4.0 ml SDBS (0.1 M) + Co <sup>2+</sup> (2.0 ml, 0.1 M) + N <sub>2</sub> H <sub>4</sub> (1.2 ml, 7%) + urea (22 mg) ; hydrothermal treat at 120°C, 10 h.
Sample in Figure 4c,d	20 ml H <sub>2</sub> O + 4.0 ml SDBS (0.1 M) + Co <sup>2+</sup> (2.0 ml, 0.1 M) + N <sub>2</sub> H <sub>4</sub> (1.2 ml, 7%) + urea (22 mg) , followed by hydrothermal treat at 120°C, 15 h.
Sample in Figure 4e,f	20 ml H <sub>2</sub> O + 4.0 ml SDBS (0.1 M) + Ni <sup>2+</sup> (2.0 ml, 0.1 M) + N <sub>2</sub> H <sub>4</sub> (1.8 ml, 7%) + urea (70 mg), followed by hydrothermal treat at 120°C, 5 h.
Sample in Figure 4g,h	20 ml H <sub>2</sub> O + 4.0 ml SDBS (0.1 M) + Co <sup>2+</sup> (2.0 ml, 0.1 M) + N <sub>2</sub> H <sub>4</sub> (1.8 ml, 7%) + NaOH solution (0.15 M, 5.7 ml, dropwise addition ).
Sample in Figure 4i,j	20 ml H <sub>2</sub> O + 4.0 ml SDBS (0.1 M) + Co <sup>2+</sup> (2.0 ml, 0.1 M) + N <sub>2</sub> H <sub>4</sub> (1.8 ml, 7%) + ammonia solution (10%, 5.5 ml) at room temperature.



**Figure S8.** TEM image of the  $\text{Co(OH)}_x(\text{DBS})_y$  calcined in Ar at 300 °C for 3 h. Note that some of the particles after calcination still maintain its multishelled structure, and the nanoclusters are mostly embedded within the carbon framework.