-Supporting Information-

Investigating the Multiple Roles of Polyvinylpyrrolidone for A General Methodology of Oxide Encapsulation

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Preparation of TEM Samples. TEM grids were treated with oxygen plasma in a Harrick plasma cleaner/sterilizer for 45 s to improve the surface hydrophilicity. The hydrophilic face of the TEM grid was then placed in contact with the sample solution. A filter paper was used to wick off the excess solution on the TEM grid, which was then dried in air for 5 min.

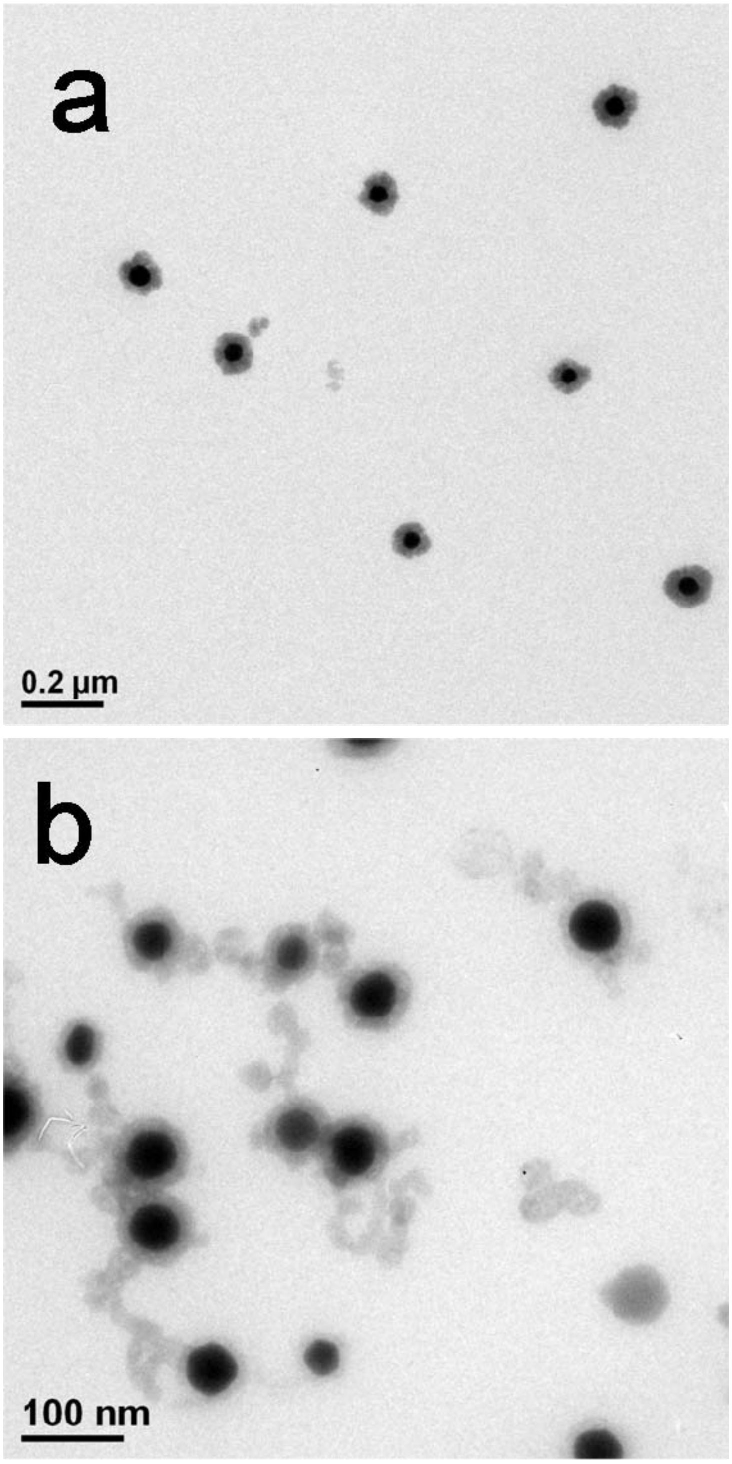

Figure S1. TEM images of the Au@ZnO core-shell NPs prepared from (a) $Zn(CH_3COO)_2$ and HMTA, and (b) $Zn(NO_3)_2$ and Na_2CO_3.

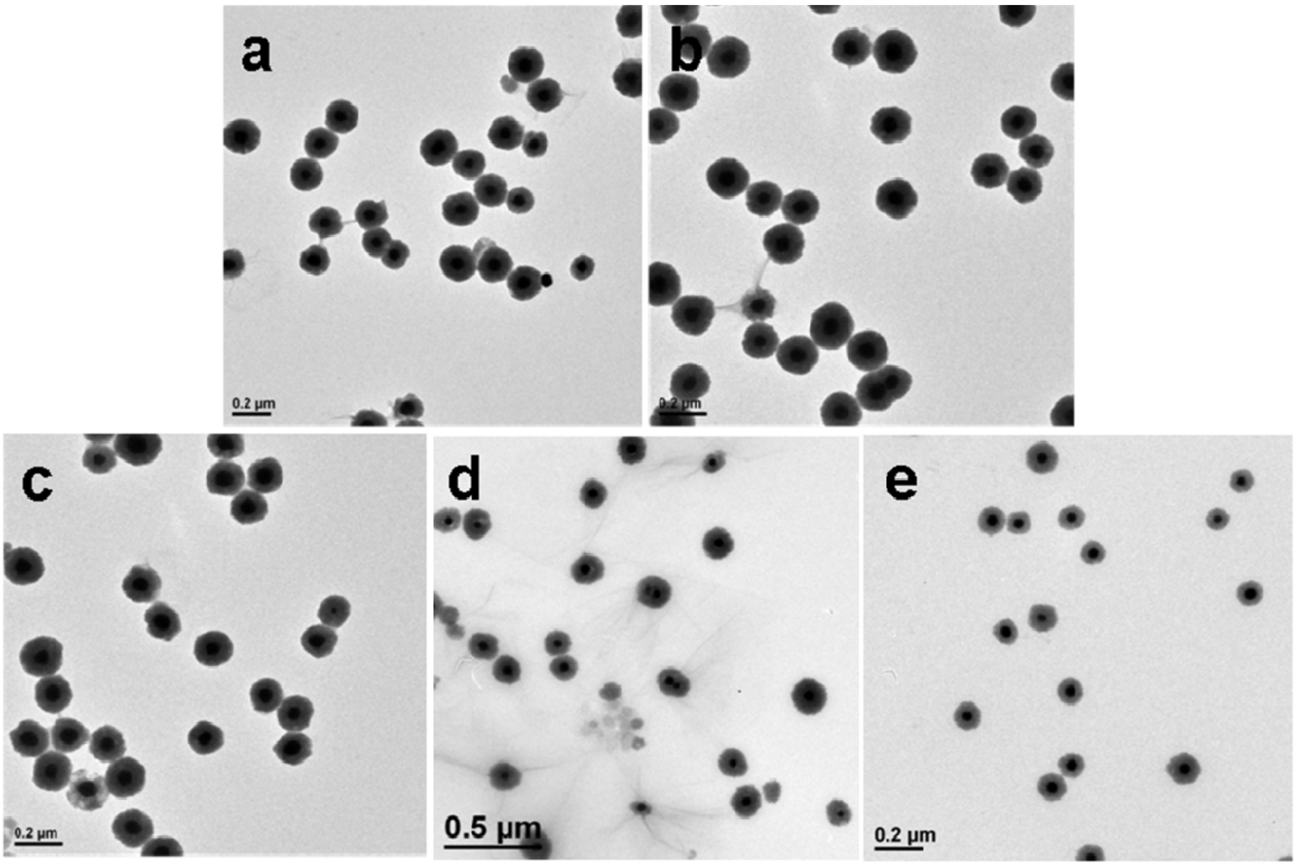

Figure S2. TEM images of the Au@ZnO core-shell NPs prepared in the presence of PVP from Au seeds modified with different ligands: (a) mercaptoacetic acid (ligand **2**), (b) 11-mercaptoundeconoic acid (ligand **3**), (c) 4-ethylthiophenol (ligand **5**) (d) 1-octadecanethiol (ligand **6**), and (e) 2-dipalmitoyl-sn-glycero-3-phosphothioethanol (sodium salt) (ligand **7**).

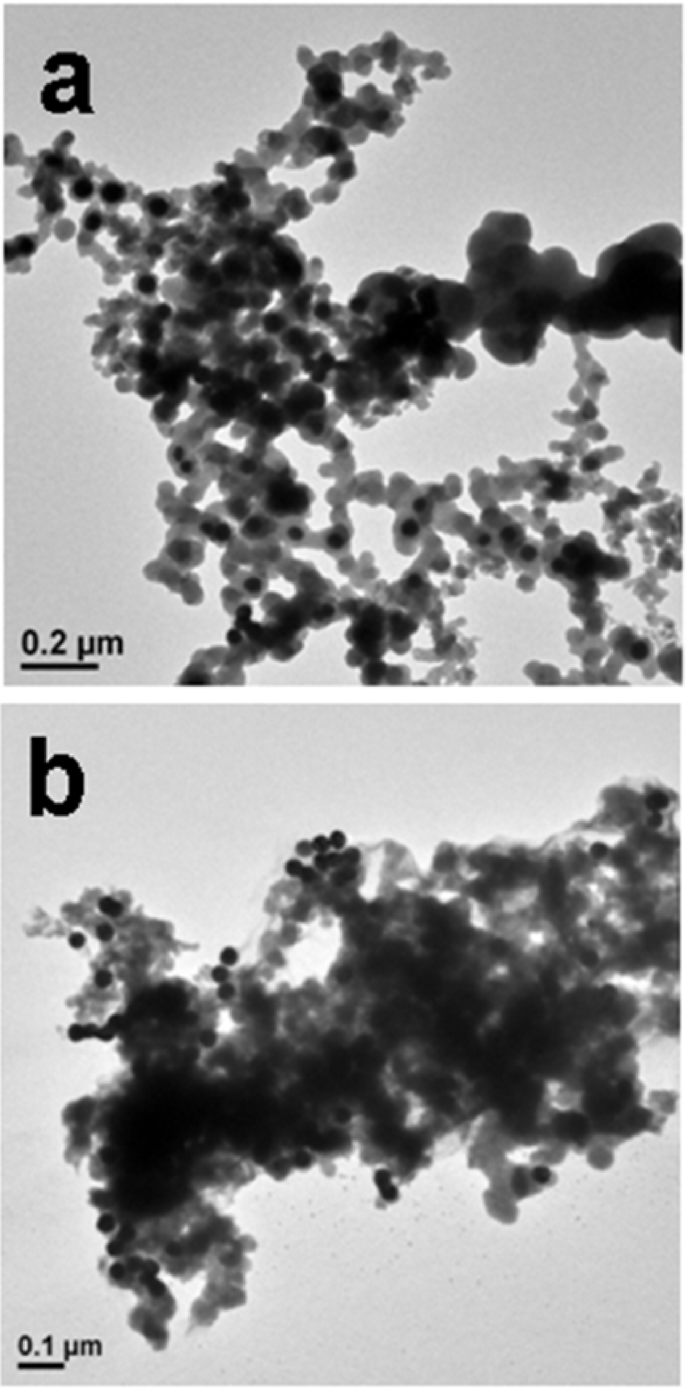

Figure S3. TEM images of the Au/ZnO hybrids prepared in the absence of PVP from Au seeds modified with (a) 11-mercaptoundeconoic acid (ligand **3**) and (b) 2-dipalmitoyl-*sn*-glycero-3-phosphothioethanol (sodium salt) (ligand **7**).

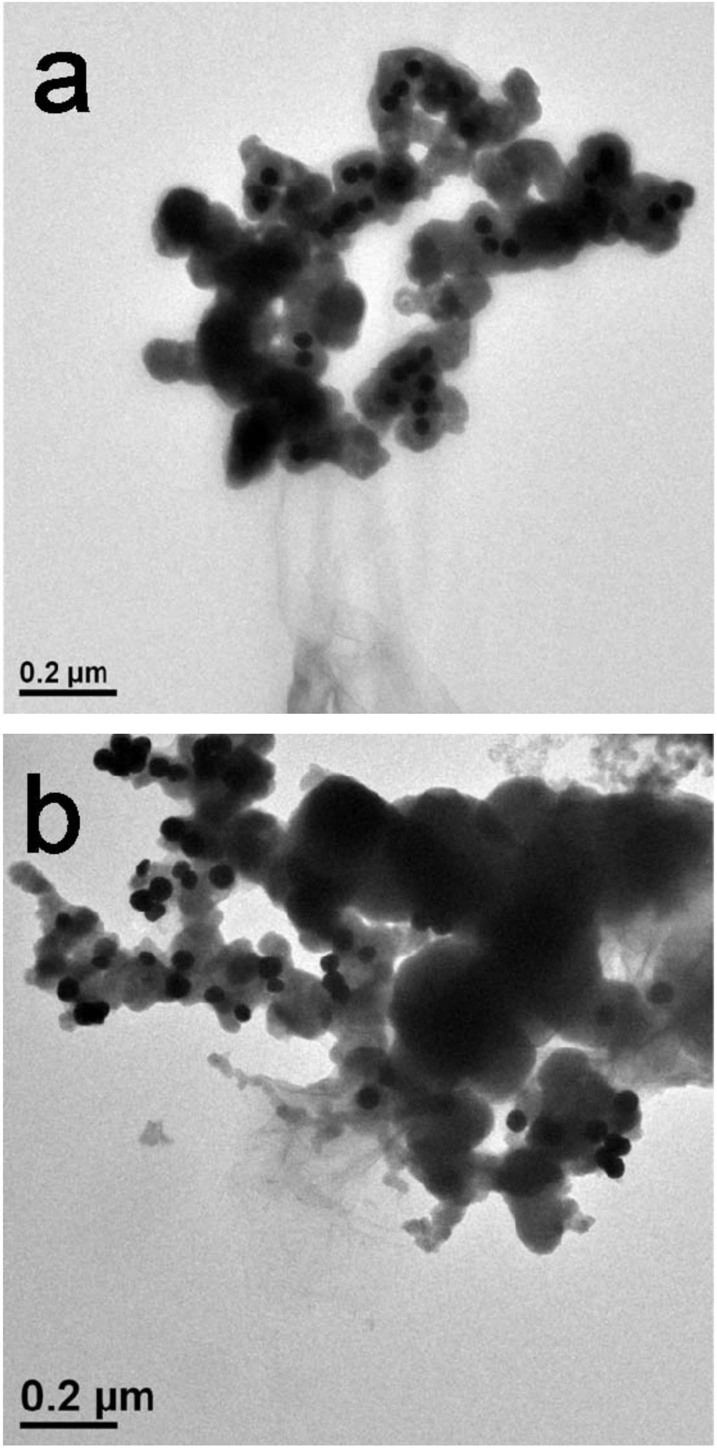

Figure S4. TEM images of the Au/ZnO hybrids prepared in the presence of PEG from Au seeds modified with different ligands: (a) 4-mercaptobenzoic acid (ligand **1**) and (b) 2-naphtalenethiol (ligand **4**).

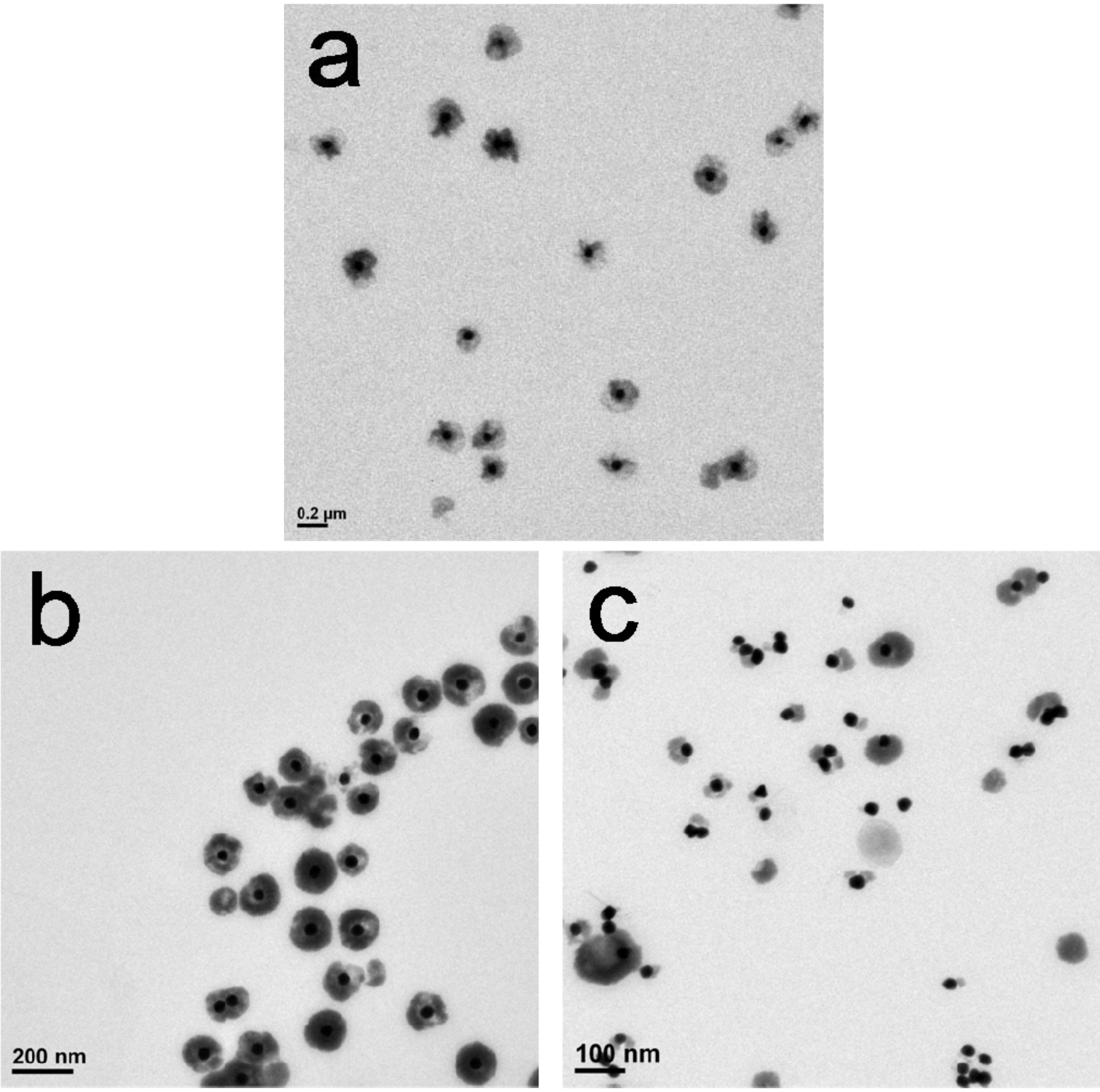

Figure S5. PVP incorporation in ZnO. Using high molecular weight PVP (360,000), the segregated PVP domains in the Au@ZnO NPs can be directly observed in the TEM images: (a) 4-mercaptobenzoic acid (ligand **1**) and (b) 2-naphtalenethiol (ligand **4**), and (c) no ligand was used.

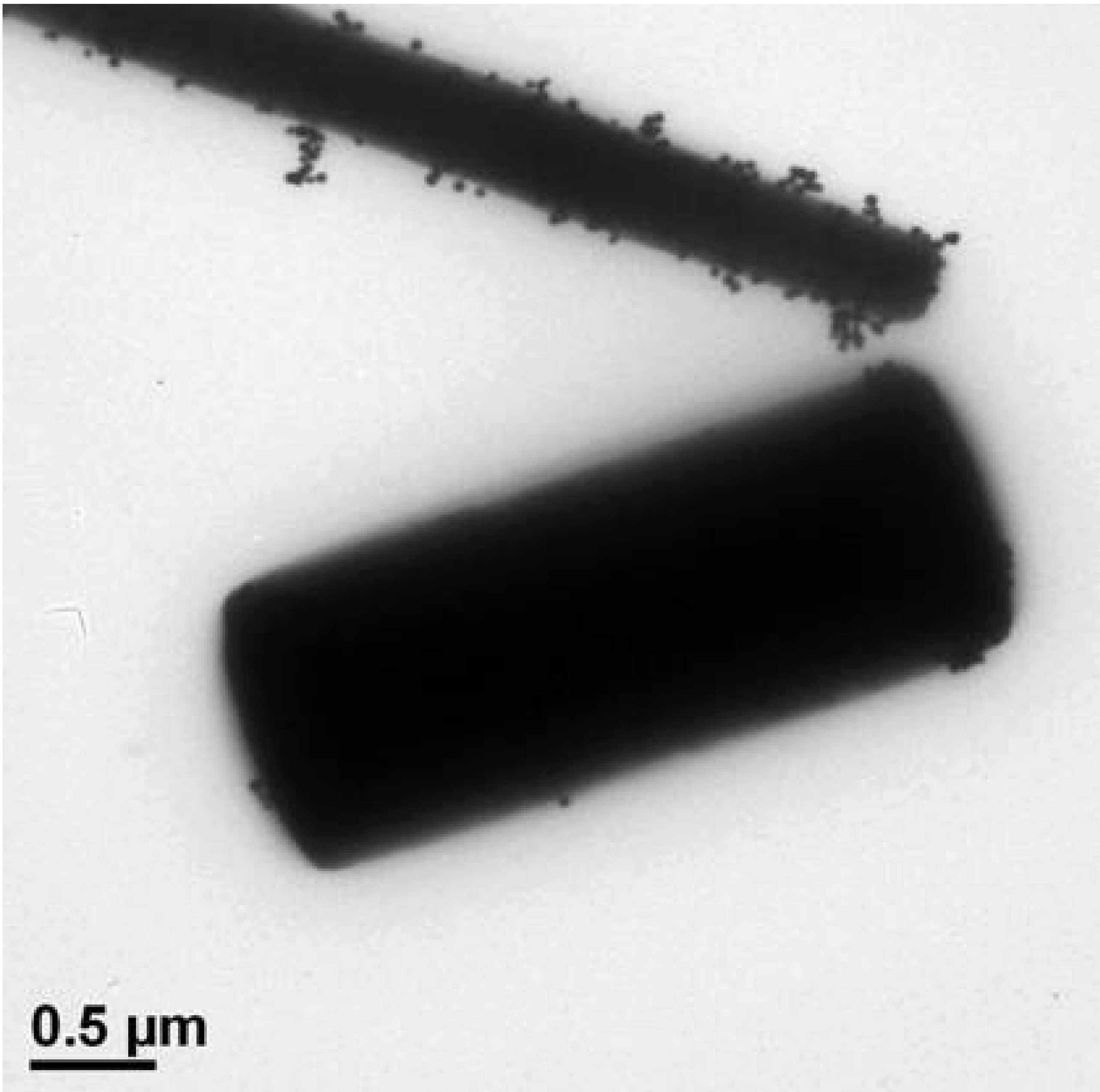

Figure S6. TEM image of Au/ZnO nanohybrid prepared in the absence of PVP and ligand.

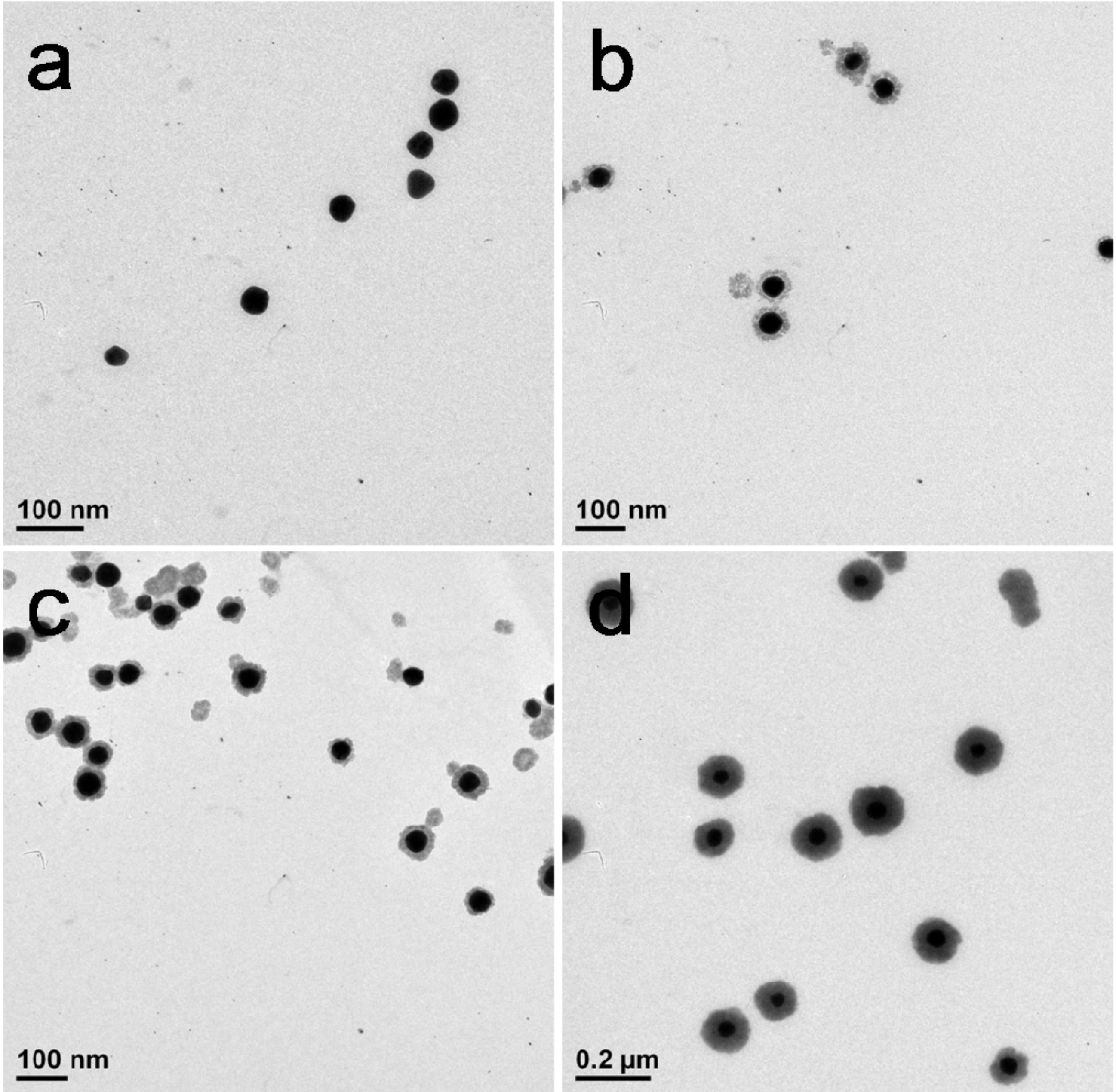

Figure S7. TEM images of temporal evolution of Au@ZnO core-shell NPs: (a) t = 10 min, (b) t = 20 min, (c) t = 30 min, and (d) t = 1 h.

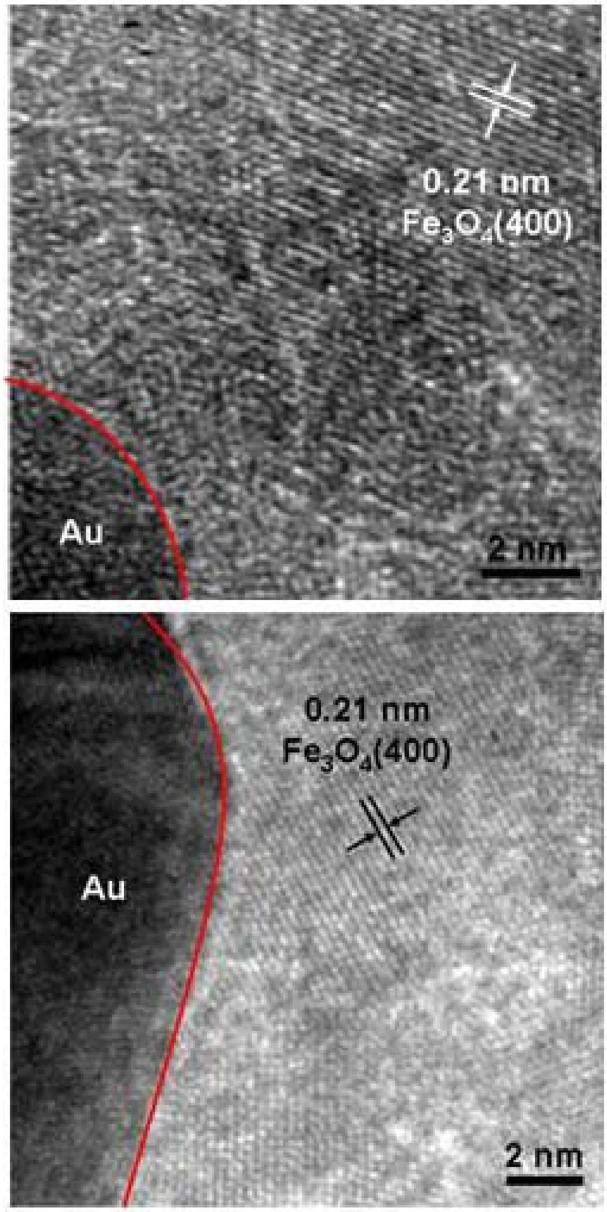

Figure S8. HRTEM images of the Au@Fe_3O_4 core-shell NPs. The lattice spacing was measured as 0.21 nm, consistent with the (400) plane of Fe_3O_4 lattice.

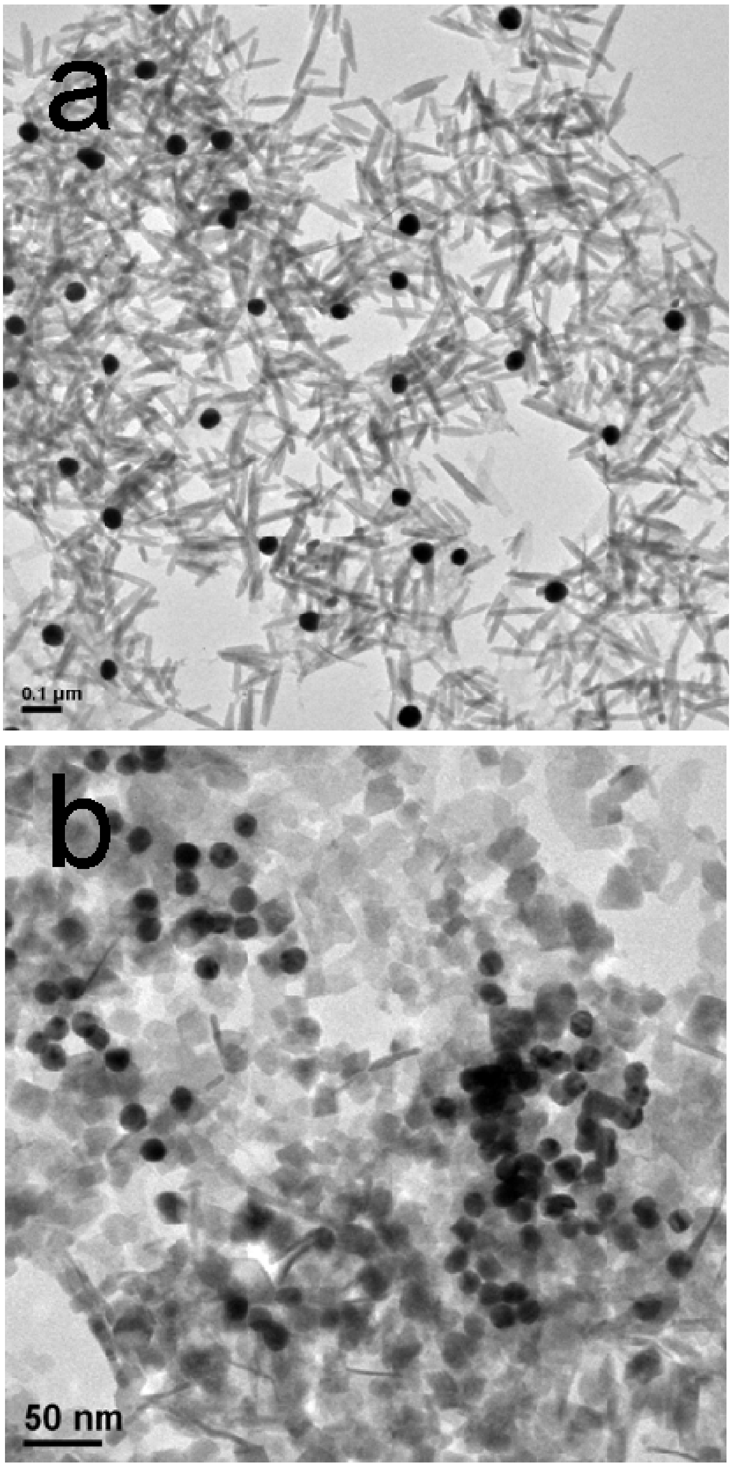

Figure S9. TEM images of (a) Au/Fe_2O_3 hybrid prepared from $FeCl_3$ and HMTA and (b) Au/MnO hybrid prepared from $MnCl_2$ and HMTA.

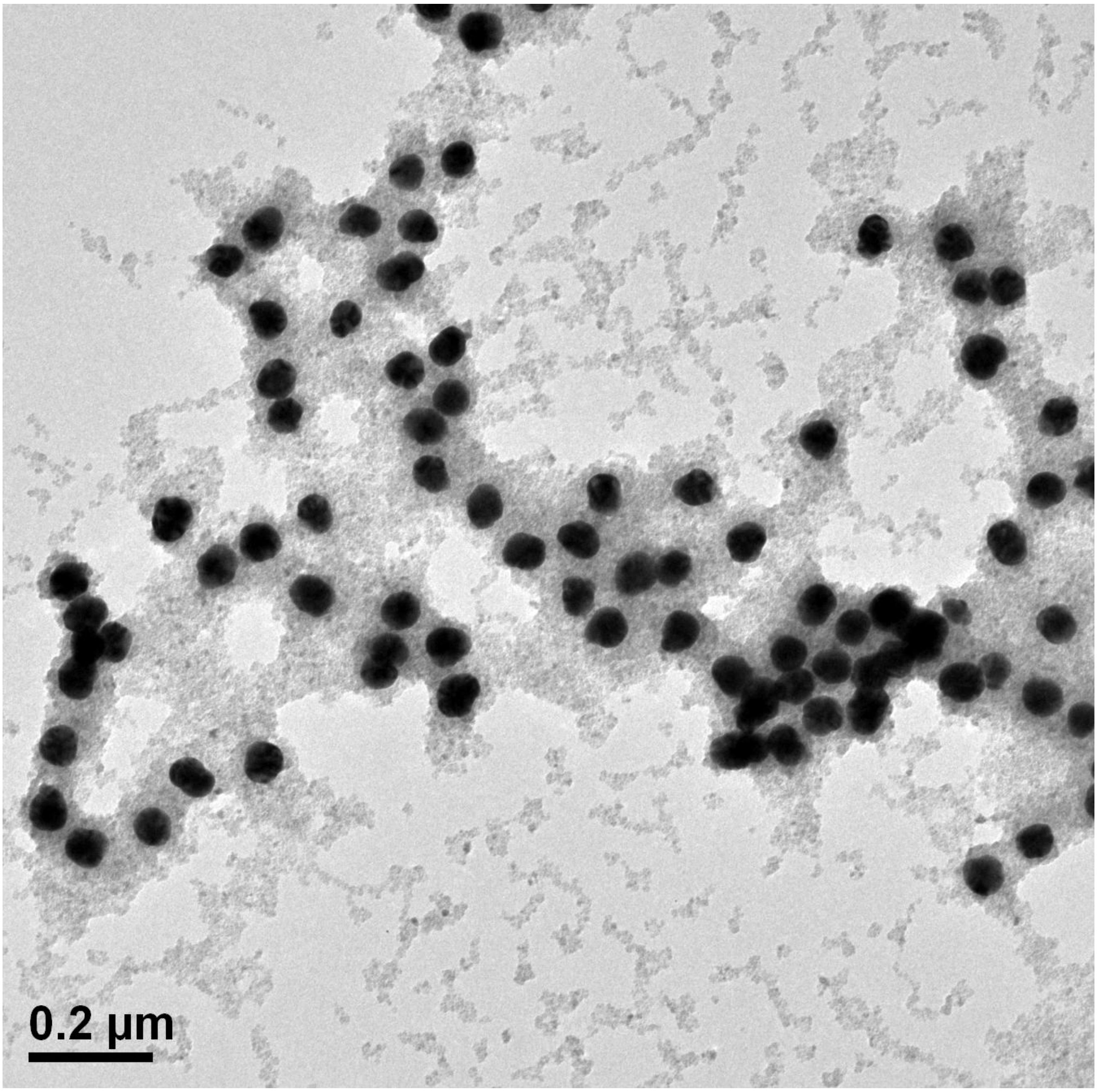

Figure S10. TEM image of Au/TiO2 hybrid prepared from TiF_4 and HCl in the aqueous solution.

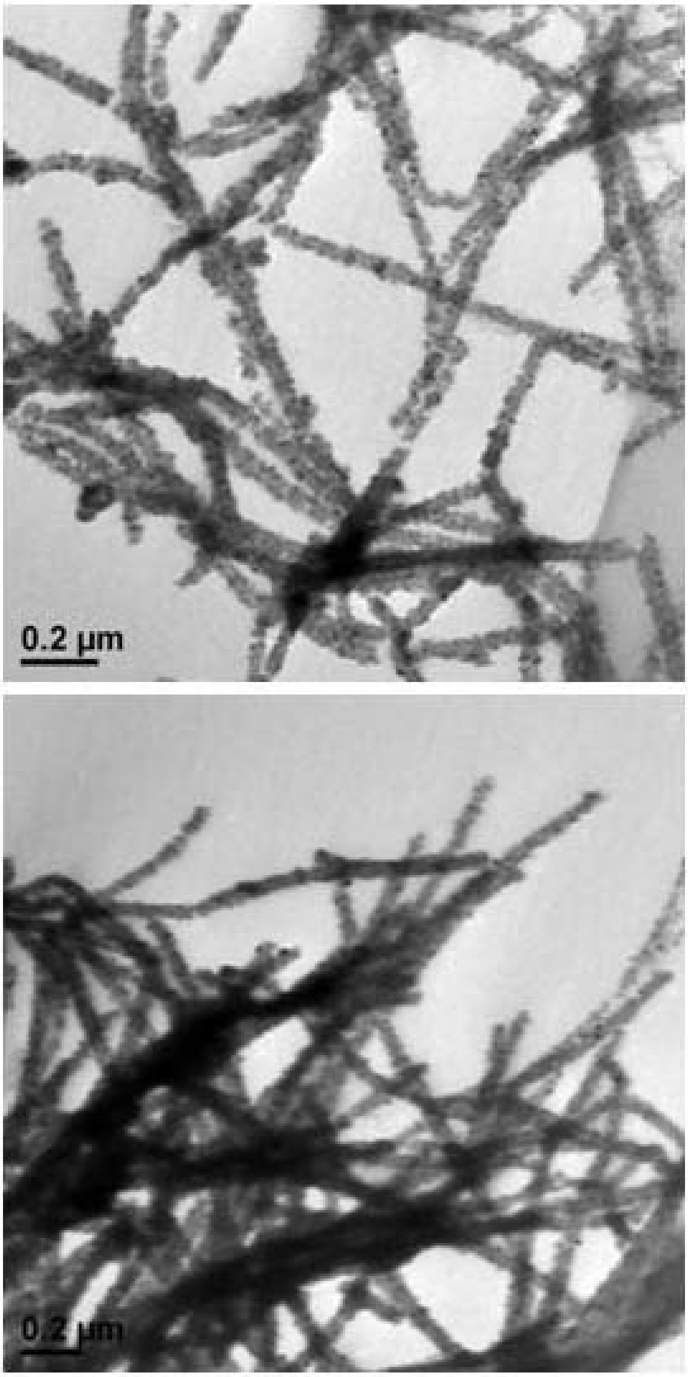

Figure S11. Two TEM images of CNT@CdS nanocomposites.

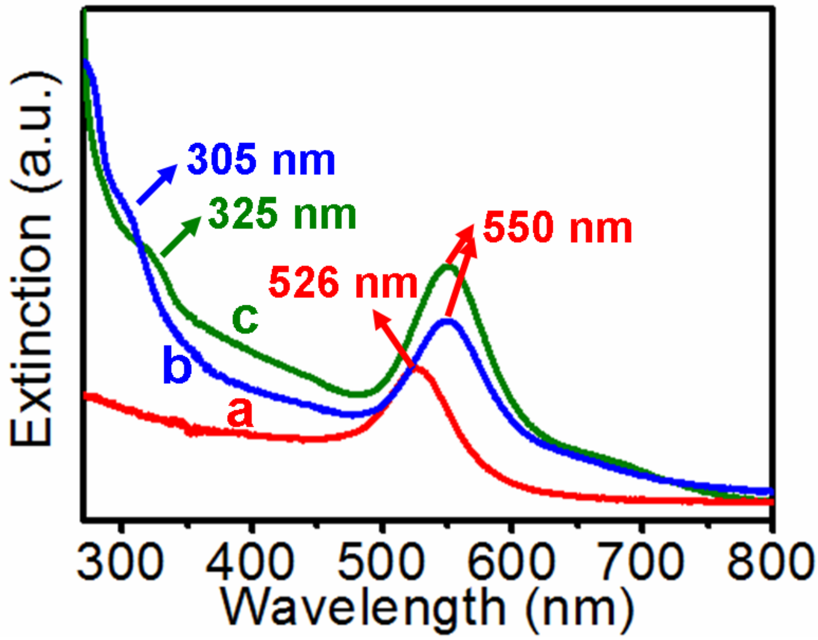

Figure S12. Optical-extinction spectral of (a) citrate-stabilized AuNPs (40 nm), (b) as-prepared Au@ZnO core-shell NPs, (c) Au@ZnO core-shell NPs after incubating in DMF (140 ^{o}C, 12 h).

The localized-surface-plasmon-resonance (LSPR) peak of Au@ZnO core-shell NPs shifts to ca. 550 nm (***Figure S12b,c***) from the original 526 nm of citrate-stabilized AuNPs (***Figure S12a***). These red-shifts arose due to an overall increase in the refractive index of the dielectric enviroment surrounding the AuNPs upon ZnO coating. Moreover, the as-prepared Au@ZnO core-shell NPs exhibit a ZnO excitonic peak at 305 nm (***Figue S12b***), which is blue shifted compared to bulk ZnO at 373 nm. It is noted that the ZnO shell in our case is included PVP modifier, and this blue shift in the ZnO/PVP samples have already been reported by other researcher.[1] Furthermore, the quantum confinement and/or deformed lattice of ZnO shell due to its polycrystalline nature may also contribute to this blue shift.[2] This excitonic peak red shifts from 305 nm to 325 nm after incubating Au@ZnO core-shell NPs in DMF (140 ^{o}C) for 12 h (***Figue S12c***), which improves the crystal quality of the ZnO shell.

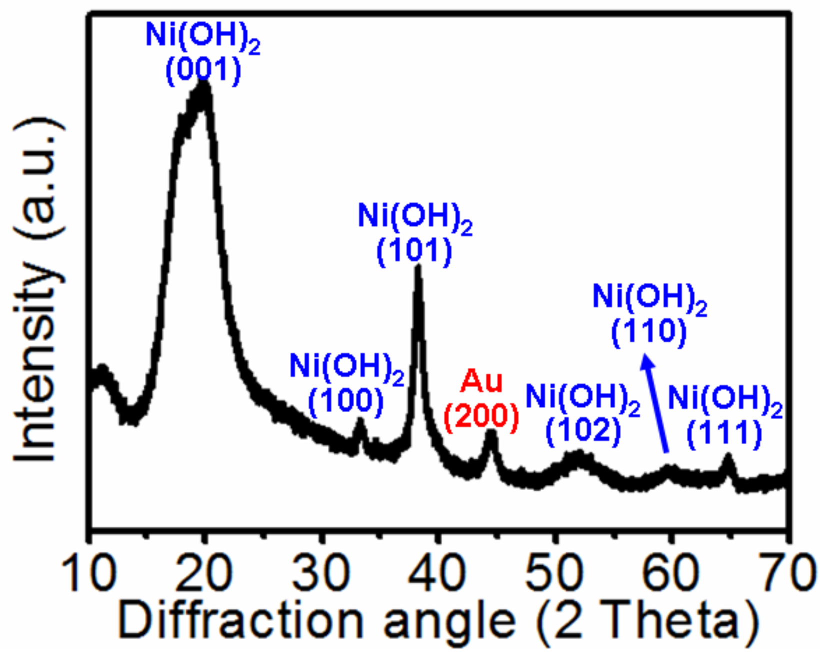

Figure S13. XRD pattern of Au@ β-Ni(OH)$_2$ core-shell NPs.

XRD pattern agreed with the (001), (100), (101), (102), (110), (111) planes of hexagonal β-Ni(OH)$_2$ structure (a = 0.3126 nm, c = 0.4605 nm, JCPDS file No. 14-0117).[3] The remaining peak could be indexed to the face-centered-cubic Au (200) plane. Thus, the composition of the shell is hexagonal structured β-Ni(OH)$_2$.

(1) Yao, K. X.; Zeng, H. C. *J. Phys. Chem. C* **2007**, *111*, 13301.

(2) Zhou, T.; Lu, M.; Zhang, Z.; Gong, H.; Chin, W. S.; Liu, B. *Adv. Mater.* **2010**, *22*, 403.

(3) Wang, Y.; Zhu, Q.; Zhang, H. *Chem. Commun.* **2005**, 5231.