Supporting information

Controllable spherical aggregation of monodisperse carbon nanodots

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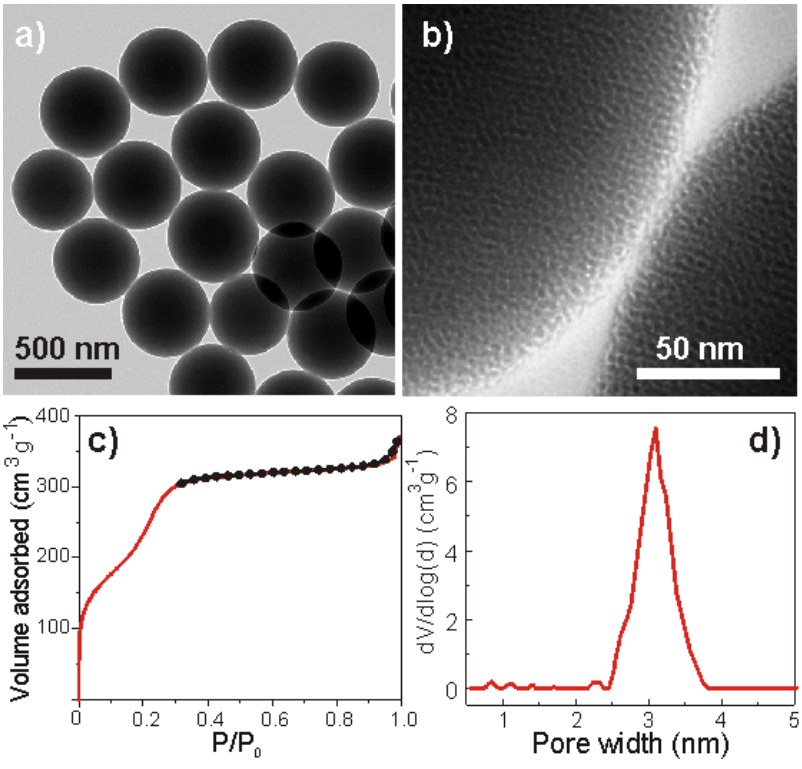

Fig. S1. Characterization of the monodisperse spherical mesoporous silica particles used as a template for MCND synthesis. (a,b) TEM images, (c) N_2 adsorption and desorption isotherms at 77 K, (d) pore size distribution calculated by the Non-Local Density Functional Theory (the oxide surface model assuming a cylindrical pore shape).

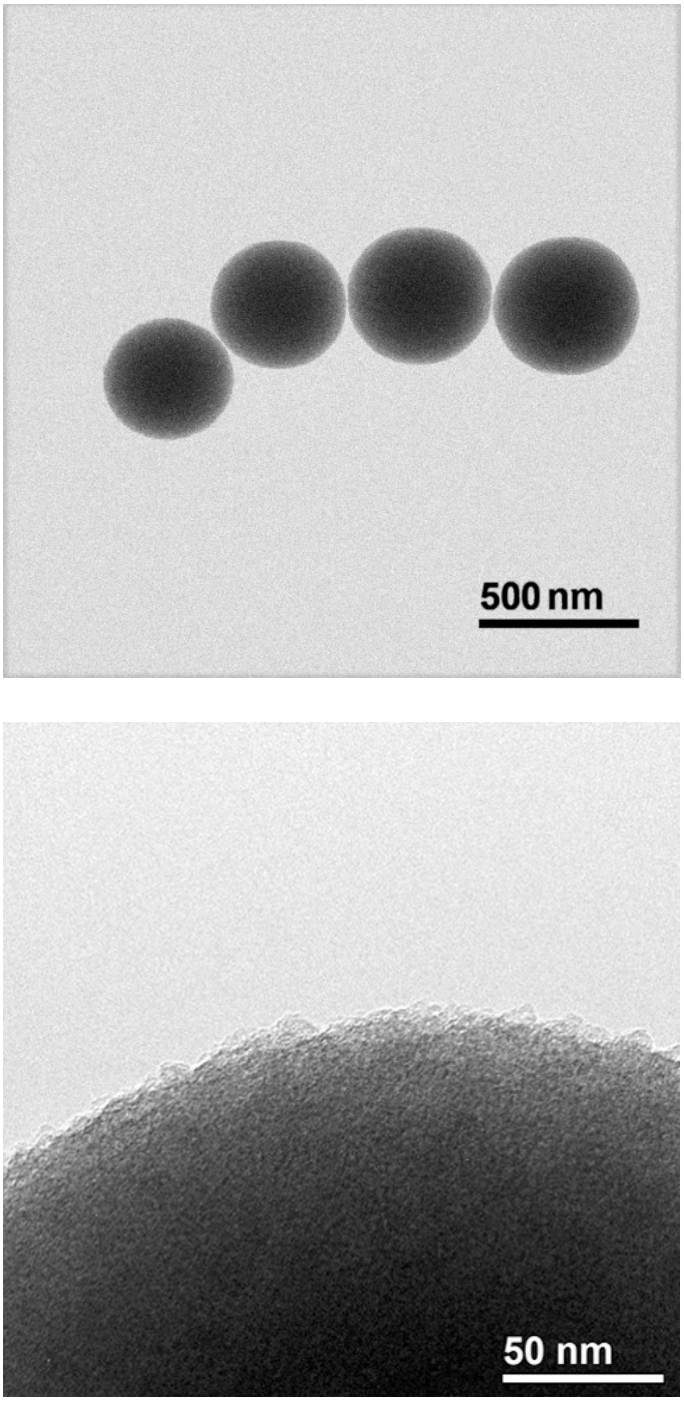

Fig. S2. TEM images of monodisperse spherical mesoporous silica particles filled with carbon nanodots.

AFM measurements

Particle size analysis using AFM was performed by a microscope Dimension 3100, Veeco, USA. A cantilever was used with a needle radius of 2 nm TESP-SS, Bruker, Germany. Carbon nanodots were applied on the surface according to the technology of Pelco (USA) recommended by the manufacturer for similar nanoparticles of colloidal gold [Ref.1]. The substrate of mica was refreshed by the method of flaking the atomic layers by means of an adhesive tape to obtain an atomically smooth surface. Then the surface was treated with Poly-L-Lysine: A drop of Poly-L-Lysine, 20 µl in volume, was applied for 60 seconds then it was washed off with distilled water. The drop of the MCND hydrosol had a volume of about 20 µl. The concentration of MCNDs in hydrosol was $5 \cdot 10^{-5}$ wt. %. The drop was kept on the substrate for 10 minutes then it was pumped out with a pipette after which the sample was dried at 40 °C for 5 minutes. The substrate imperfection and the insufficient resolution of closely located nanoparticles because of the finiteness of the rounding radius of the AFM probe required using of a special topographic image processing technique that eliminates particle loss in image analysis [Ref2].

[Ref1] PELCO AFM Gold Calibration Kit: http://www.tedpella.com/technote_html/16200%20TN.pdf

[Ref2] V.A. Sevriuk, P.N. Brunkov, I.V. Shalnev, A.A. Gutkin, G.V. Klimko, S.V. Gronin, S.V. Sorokin, S.G. Konnikov. Statistical analysis of AFM topographic images of self-assembled quantum dots, *Semiconductors*, 2013, **47**, 930-934.

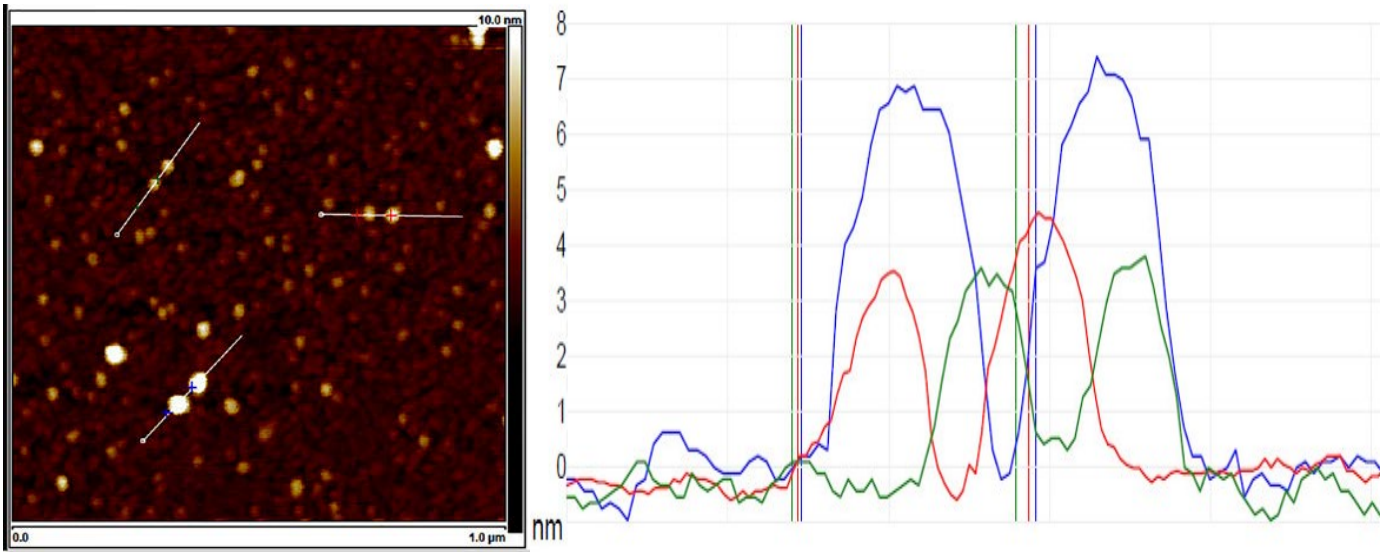

Fig. S3. Left – AFM image of MCNDs on mica substrate. Right – profiles of the selected particles. Larger particles present on the image are presumably aggregates since they are twice as high as individual MCNDs.

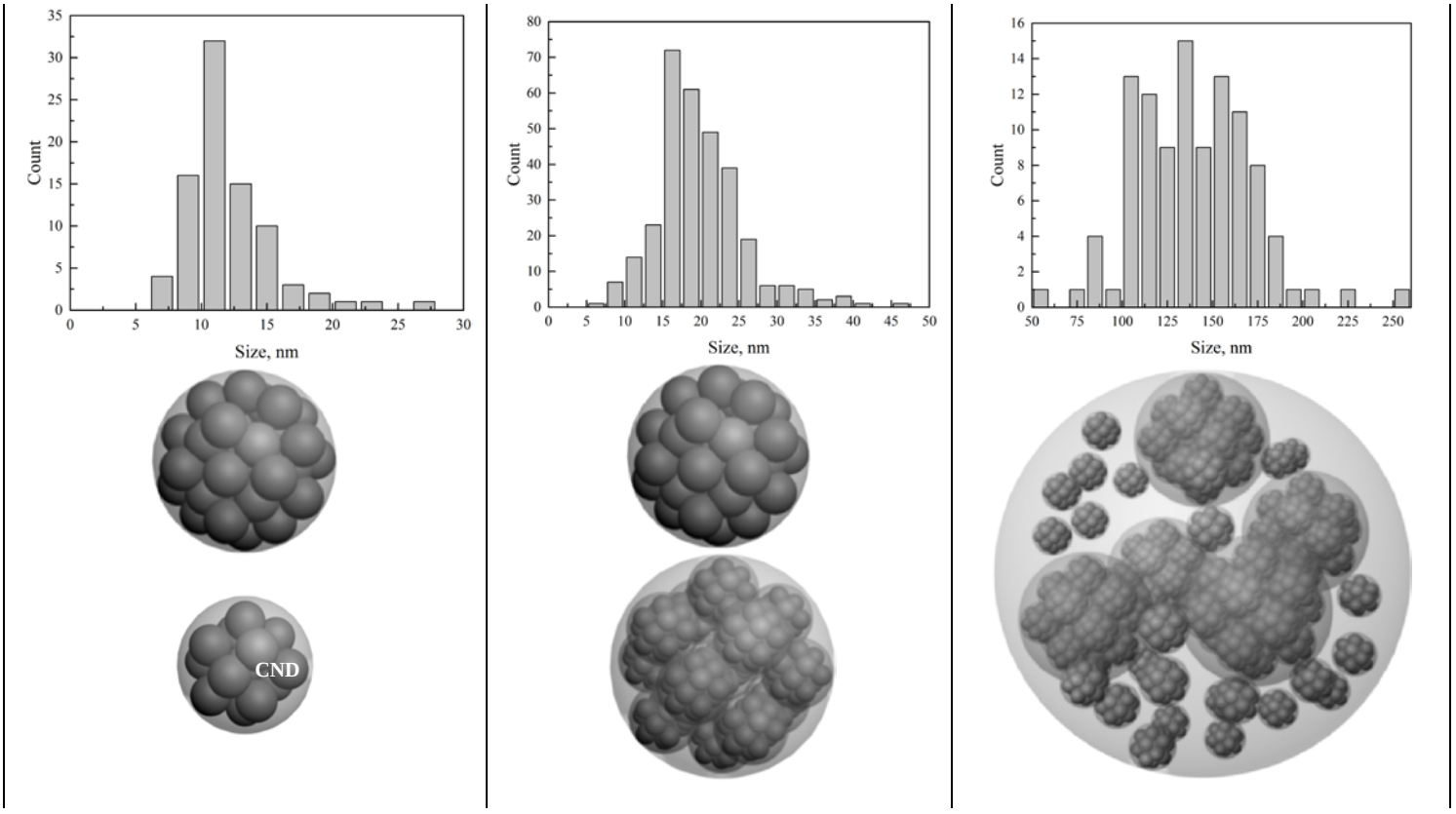

Fig. S4. Size distribution of the primary (a,b) and secondary hierarchical (b,c) MCND aggregates, determined from the obtained TEM images.

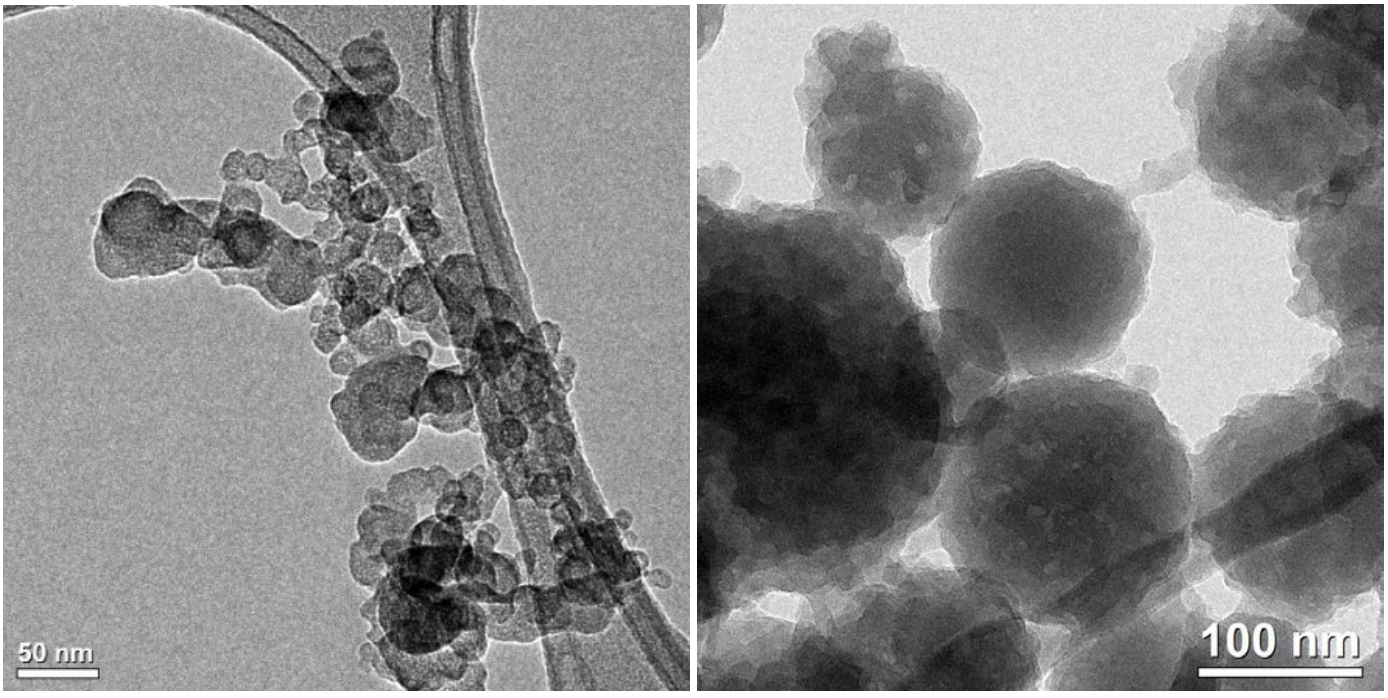

Fig. S5. TEM images of MCND aggregates obtained by drying diluted (10^{-3} mg mL^{-1}, left panel) and concentrated (10^{-1} mg mL^{-1}, right panel) MCND hydrosols on lacey carbon films.

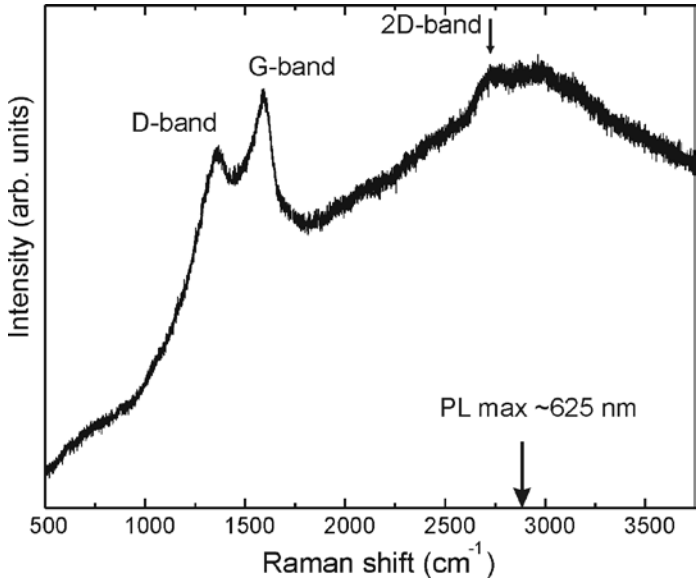

Fig. S6. Raman spectrum of MCND aggregates (λ_{ex}= 532nm).

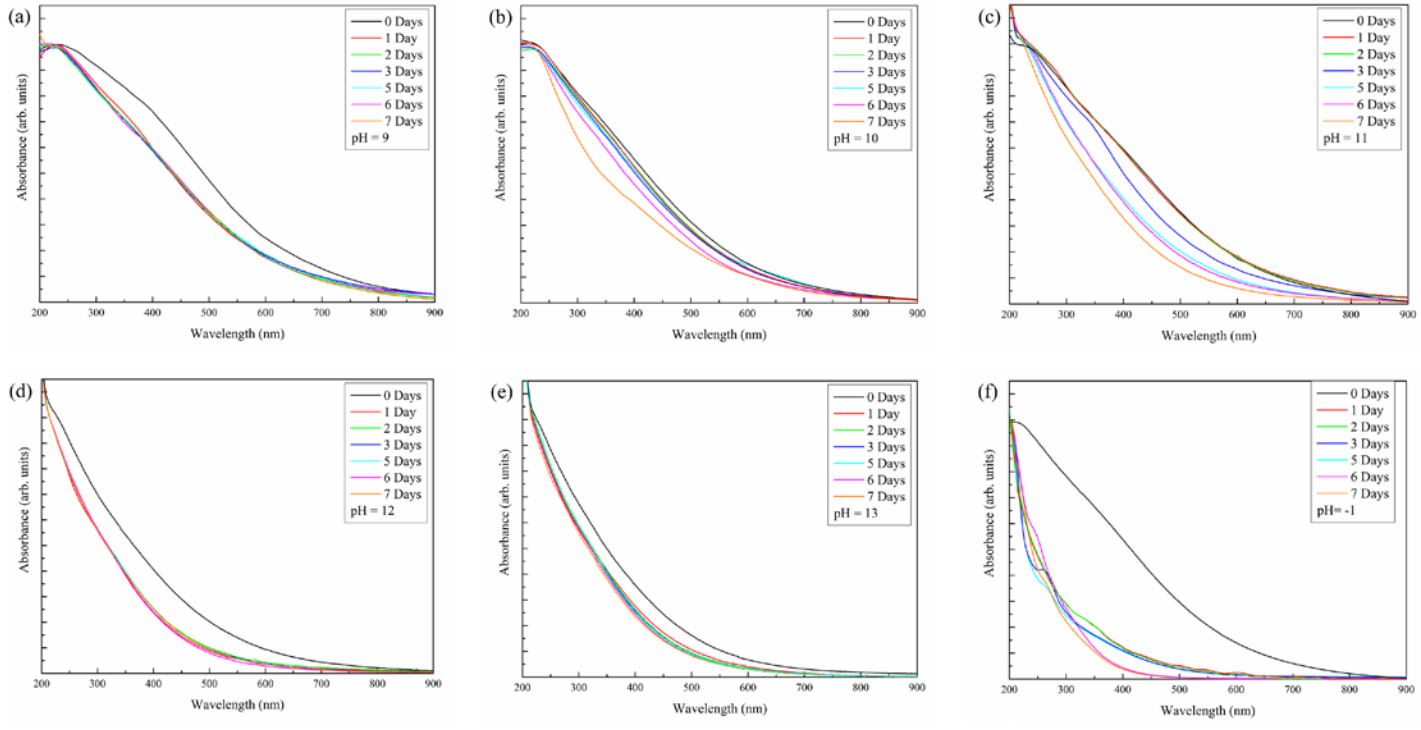

Fig. S7. Modification of the UV-Vis spectra of MCND suspensions with different pH as a function of time. Decrease in scattering is observed due to progressive peptization of the MCND hierarchical aggregates to individual 3-nm nanodots.

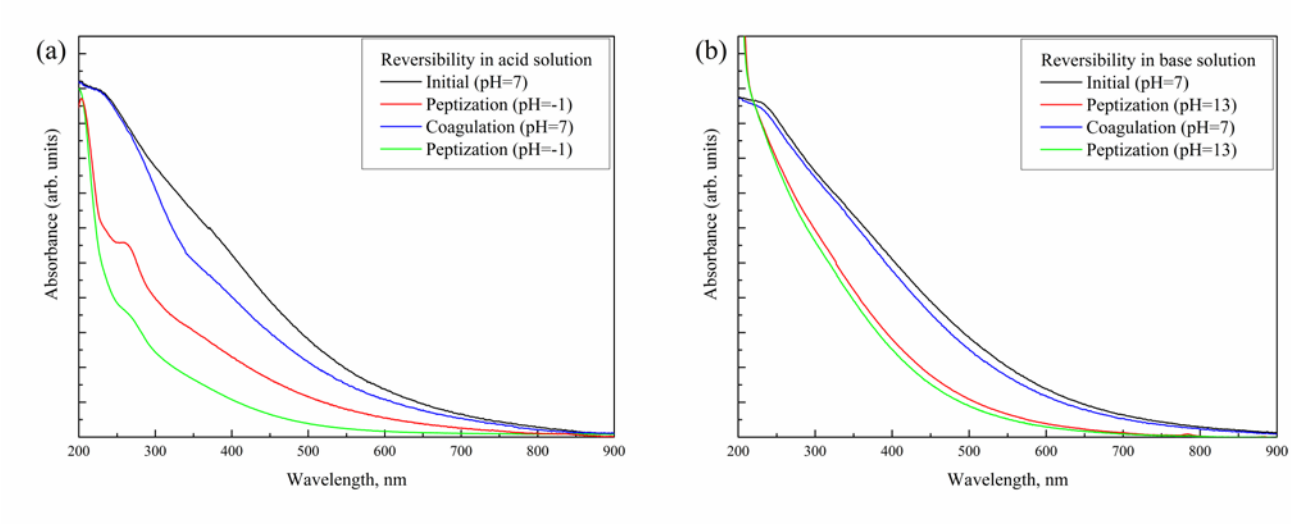

Fig. S8. Modification of the UV-Vis spectra of MCND hydrosols during the reversible peptization and coagulation processes.