Supplementary Information

Protocrystallinity of Monodispersed Ultrasmall Templated Mesoporous Silica Nanoparticles

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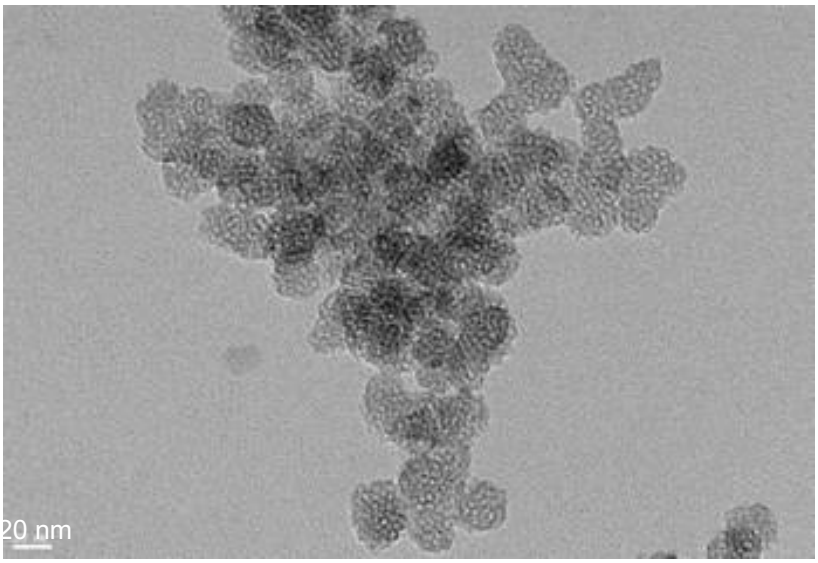
Table S1. Zeta potential of the reaction solution for ultra small MCM-41nanoparticles, **MSN-20-A**.

Reaction time	Zeta (mV)
(pH 5.5) 5 min	+ 24
(pH 5.5) 4 h	+ 32
(pH 5.5) 3 d	+ 24
(pH 5.5) 4 h → (pH 7) 5 min	+ 42
(pH 5.5) 4 h → (pH 7) 3 d	+ 59



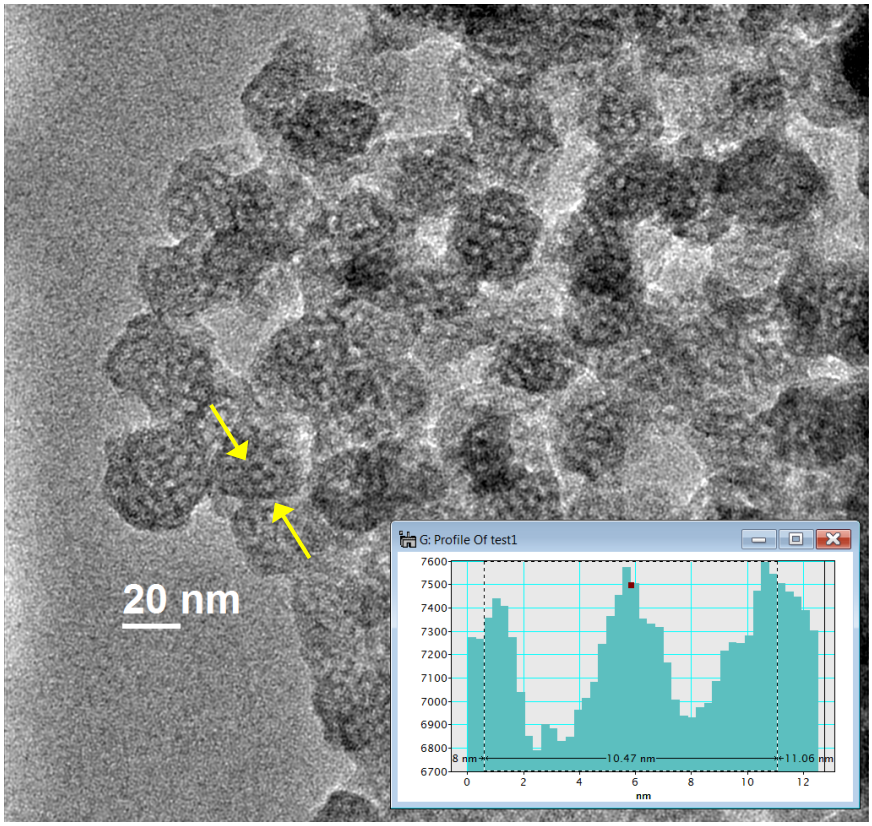


Figure S1. HRTEM image of calcined **MSN-23-A** typical intensity profile analysis showing a distance 5.4 ± 0.2 nm between two pores and an average size diameter of 23 ± 3 nm prepared as calcined MSN-22-A shown above about with an average size of 22 nm ± 3 nm.

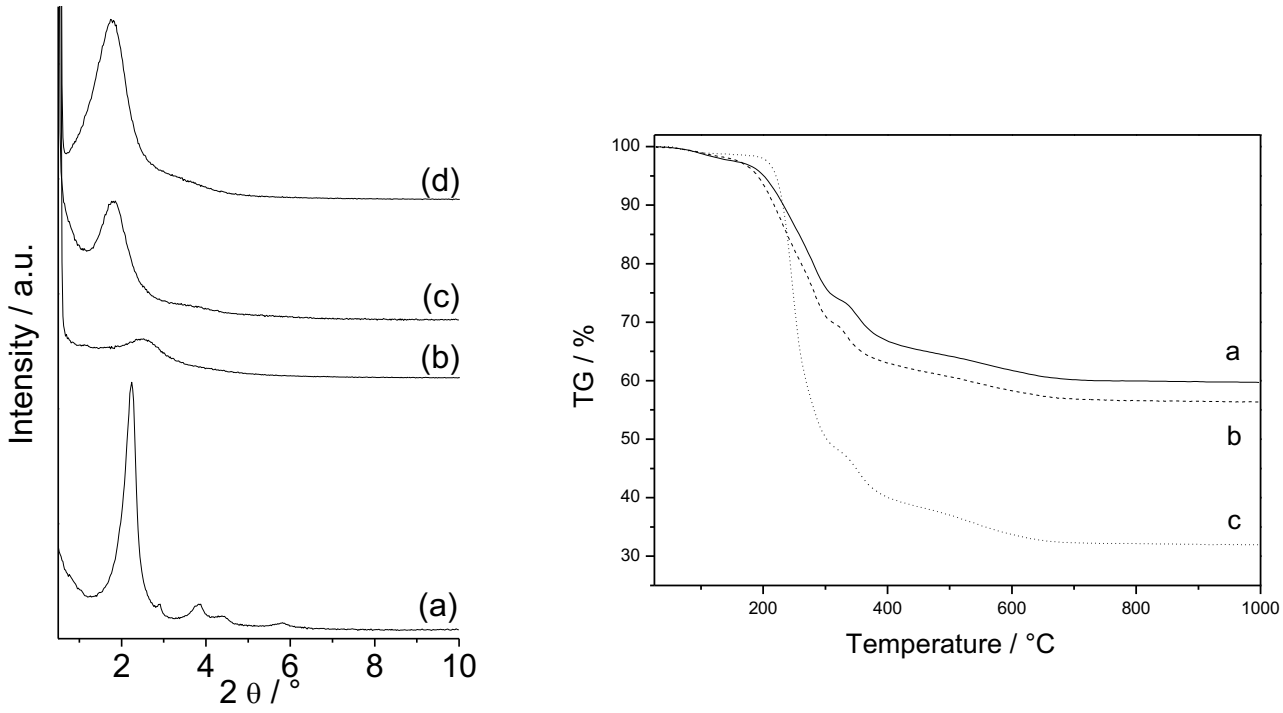


Figure S2. (left) Comparison of XRD patterns of MSM-100 (a,b) and MSN-20 (c, d) as-synthesized (a, c) and calcined then treated in distilled water (b, d), i. e., i) calcined mesoporous silica powder 0.1 g + distilled water 10 g, stirred at RT overnight, ii) centrifugated and iii) dried at 50°C; (right) TGA of the as-synthesized nanoparticles of (a) **MSN-20-A**, (b) **MSN-100-A** acid treated (c) **MSN-100** (washed with deionized water).

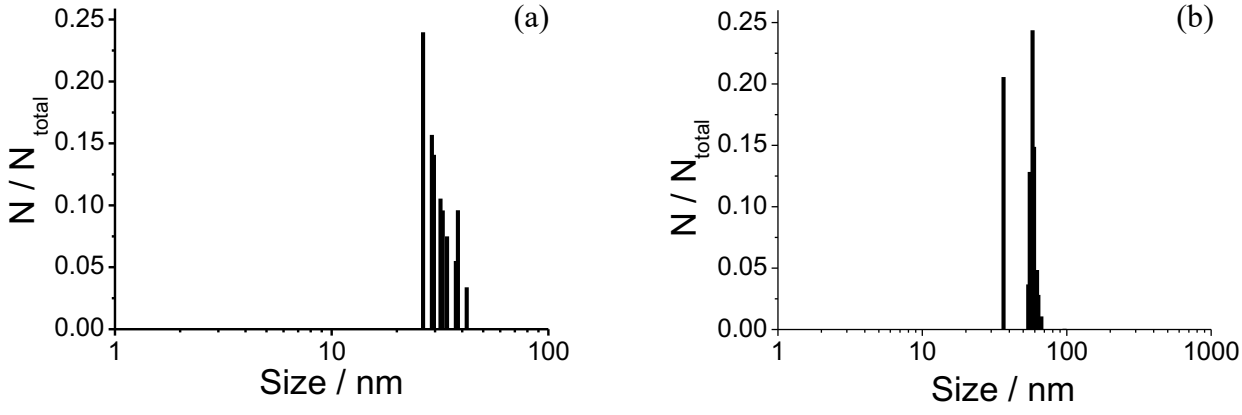


Figure S3. DLS size distribution of **MSN-20-A**, measured after the synthesis with flocculate comprised between 24 and 41 nm (a) and 3 months later with a single peak at 37 and a distribution in the range 52-69 nm (b).

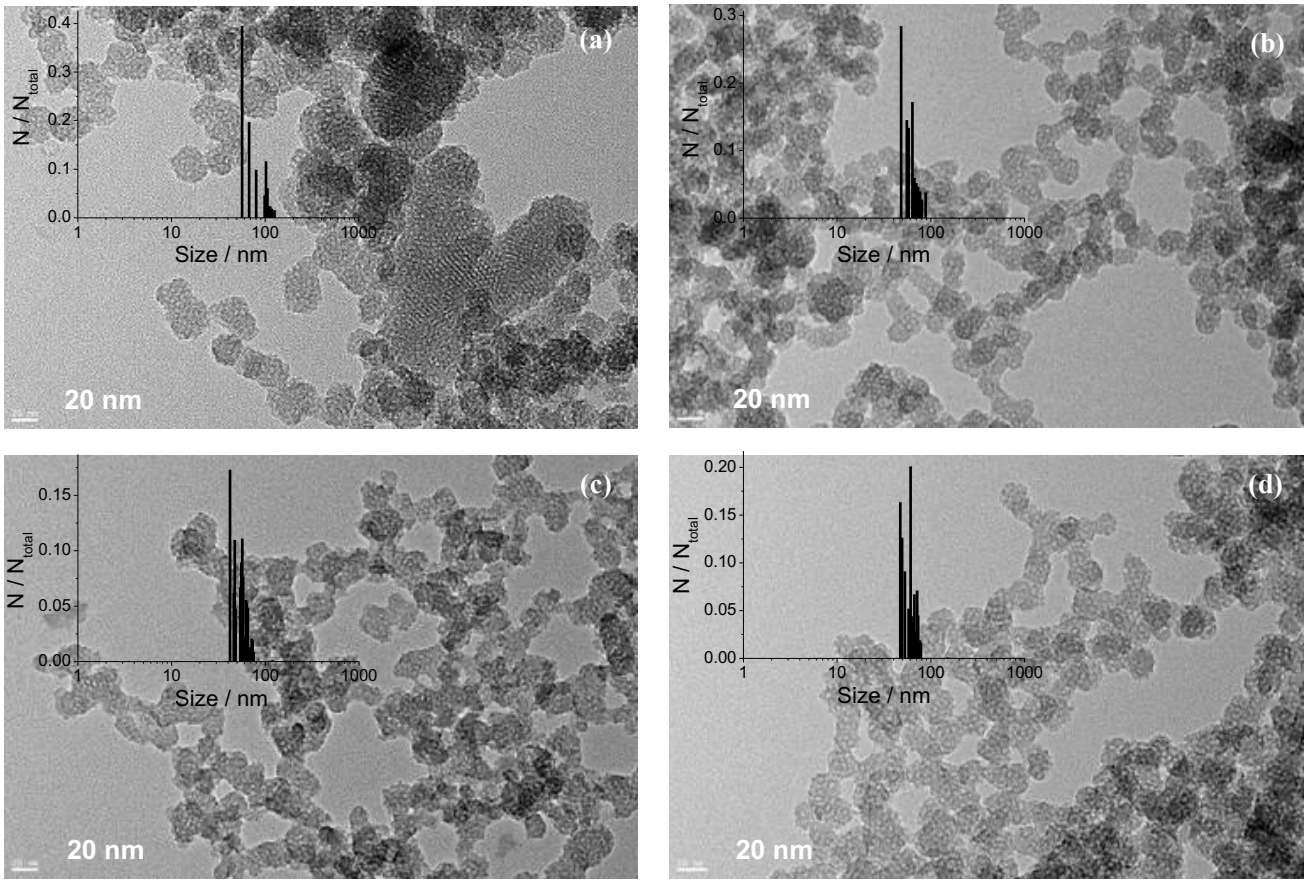


Figure S4. TEM images of US-MSN obtained from the reaction solution at the molar ration of 1 SiO_2 : 0.5 NaOH : x CTAB : y F127 : 130 H_2O. (a) x = 0.12, y = 0.000104 (twice more F127), (b) x = 0.12, y = 0.000026 (twice less F127), (c) x = 0.24 (twice more CTA), y = 0.000052, and (d) x = 0.06 (twice less CTA), y = 0.000052. Same nucleation quenching using dilution by 6 times. Inserts : corresponding DLS size distribution of the nanoparticle aggregates in the as-synthesized solution (a, b, c, and d).

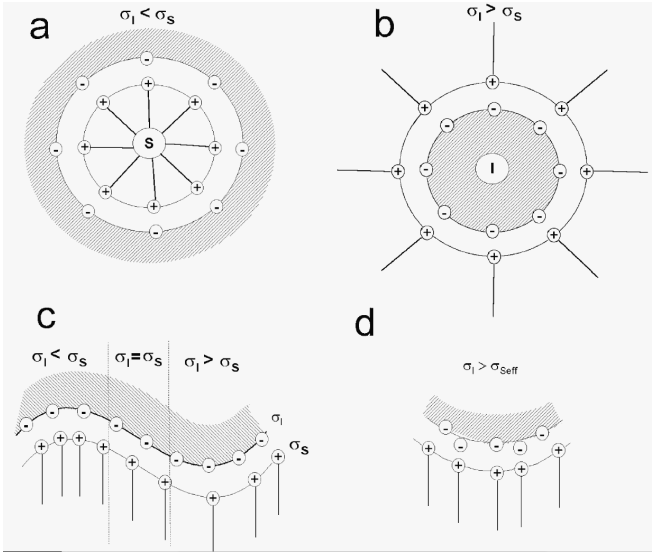


Figure S5. Charge density mismatch and generation of surface curvature in an electrical interface where there is charge separation between the Helmholtz plan according to ref. [51] where σ_i and σ_s stands for the charge density on the inorganic phase (Si-O^-) and on the surfactant micelles entrapped in the inorganic matrix (positively charged ammonium heads).

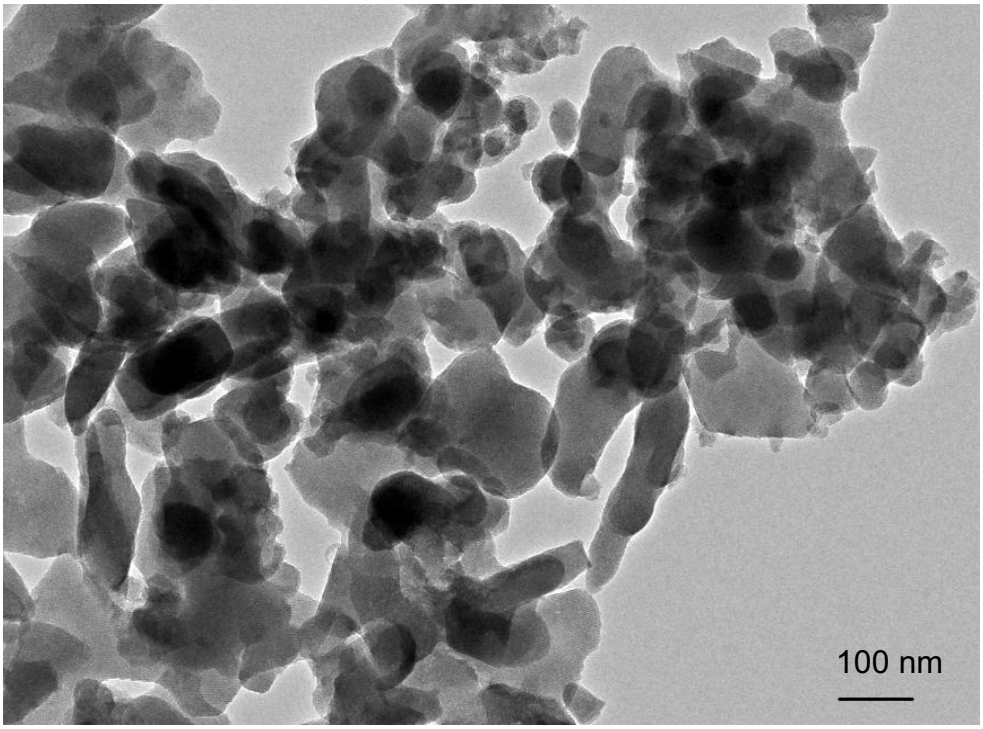


Figure S6. TEM image of **MSN-100** synthesized in the presence of a large quantity of F127 as reported in ref. [42] by Imai *et al.;* average particle size of 100 nm.