



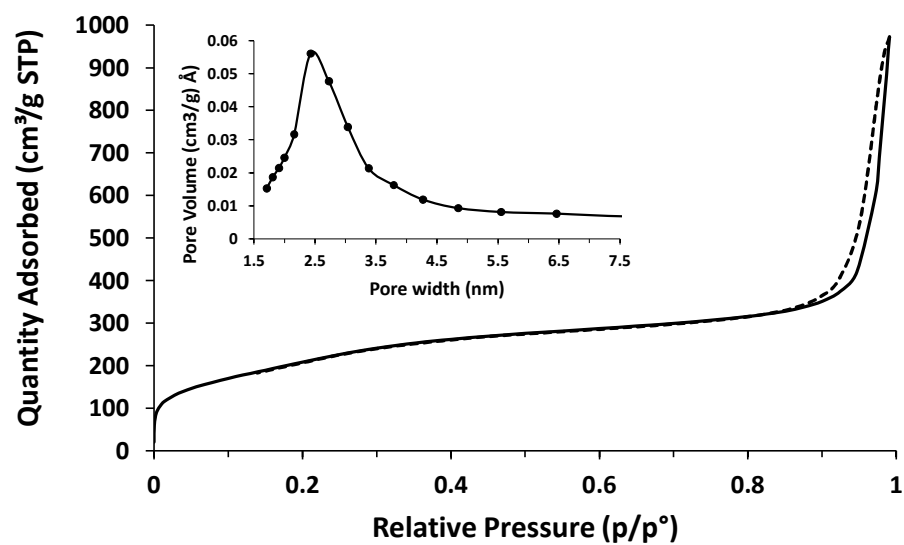
# Supplementary Materials: Mesoporous Silica Nanoparticles Modified inside and out for ON:OFF pH-Modulated Cargo Release

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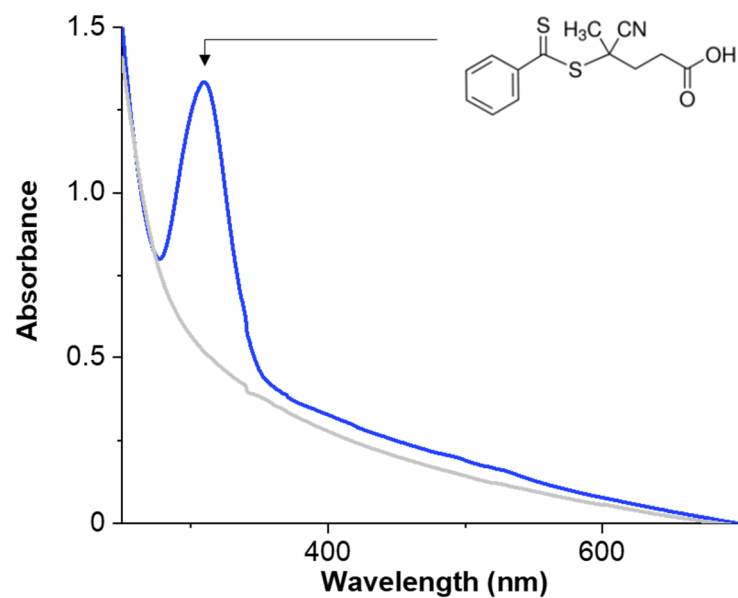
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(A) MSNs Nitrogen Sorption Isotherm



**Figure S1.** Nitrogen adsorption (solid line)-desorption (dotted line) isotherm for the MSNs, and corresponding pore size distribution (inset).

(B) Determination of the amount of RAFT agent on the MSNs



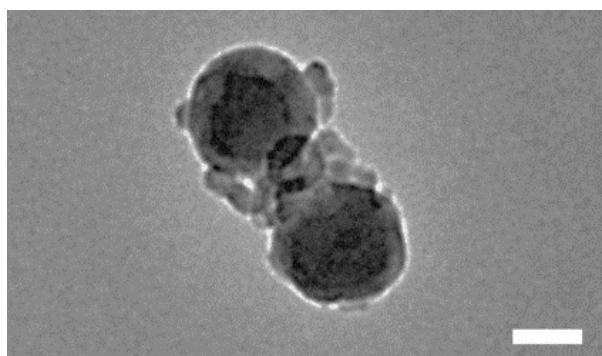
**Figure S2.** The amount of RAFT agent at the MSNs surface was calculated by subtracting the light scattering contribution (measured for bare MSNs, grey curve), from the absorption spectrum of MSN-CTA (blue curve) at the of absorption maximum wavelength (305nm).

(C) Polymer incorporation values

**Table S1.** Values of polymer incorporated per mass of nanoparticles in the different polymerization reactions.

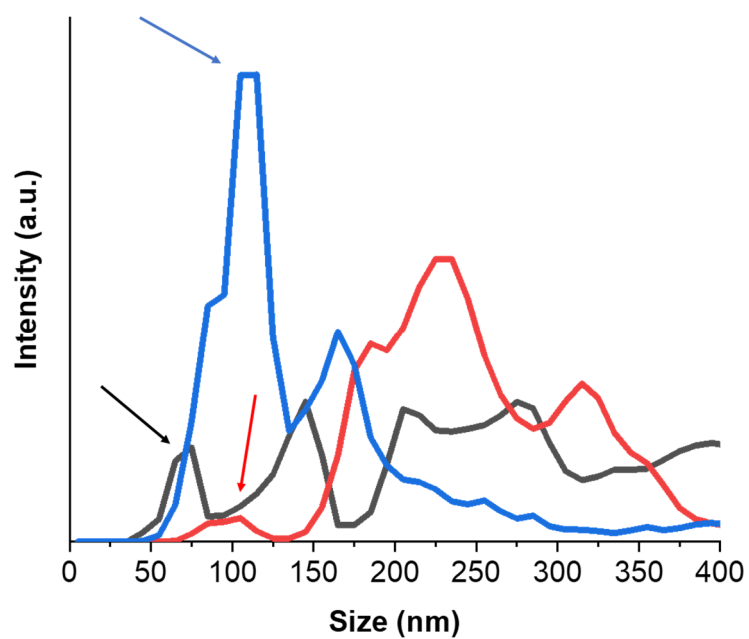
<b>[DAEM] (M)</b> \ <b>[CTA]/[AIBN]</b>	<b>5/1</b>	<b>10/1</b>
<b>0.5</b>	0.9 wt%	2.1 wt%
<b>2</b>	1.9 wt%	5.1 wt%

(D) TEM image of Hybrid MSN



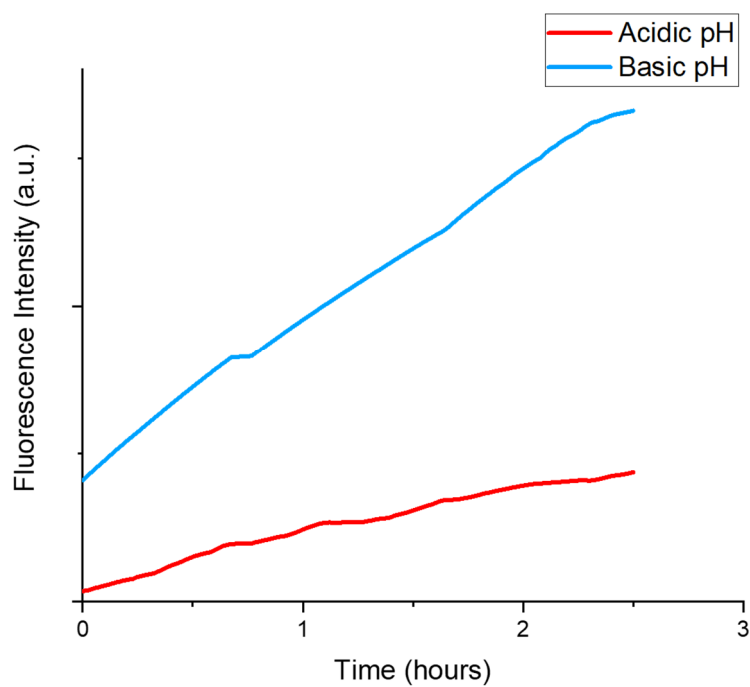
**Figure S3.** TEM image of the MSN-pDAEM15 showing the polymer shell. Scale bar = 50 nm.

(E) Size distribution data from NTA analysis



**Figure S4.** Hydrodynamic diameter distribution of MSN-pDAEM2 (black curve), MSN-pDAEM5 (red curve) and MSN-pDAEM15 (blue curve) at low pH, obtained by NTA. The lower value peak of each distribution corresponds to the diameter of the individual hybrid nanoparticles. The peaks at larger values are due to particle flocculation during the measurement.

(F) Release from SRB-loaded MSN-pDAEM2 at pH 5 and pH 9



**Figure S5.** Fluorescence intensity of SRB released from SRB-loaded MSN-pDAEM2 in PBS at pH 5 (red line) and pH 9 (blue line). The released SRB diffuses across the dialysis membrane and is measured in the bottom compartment of the cuvette ( $\lambda_{exc} = 566$  nm,  $\lambda_{em} = 589$  nm). The release rate ratio is 4.5 and the release amount ratio is 4.0 after 2.5 hours.

(G) Comparison with nanocarriers reported in the literature

**Table S2.** Comparison of ON:OFF release ratios at 2h30 between our system MSN-pDAEM15 and recently reported controlled release nanocarriers using pH-responsive polymers.

System	Core diameter (nm)	pH (ON)	pH (OFF)	ON:OFF release ratio (at 2h30)	Ref
<b>Core-Shell</b> Core: MSNs Shell: pDAEM	45 ± 9	9	5	6	MSN-pDAEM15
<b>Core-Shell</b> Core: MSNs-CAT Shell: pDAEM	45 ± 9	9	5	5	MSN-CAT-pDAEM
<b>Core-shell</b> Core: silica nanoparticles Shell: poly(acrylic acid)	130	3	7.4	2	[1]
<b>Core-shell</b> Core: MSNs Shell: poly(vinyl pyridine)	40	5.8	7.4	5	[2]
<b>Core-shell</b> Core: MSNs Shell: poly( <i>N,N</i> -dimethylaminoethyl methacrylate)	80 ± 16	2	7.4	3	[3]
<b>Core-shell</b> Core: silica nanoparticles Shell: poly(2-(diethylamino)ethyl methacrylate)	22-25	5.5	7.4	1	[4]
<b>Hybrid hollow nanogel</b> Poly( <i>N,N</i> -dimethylaminoethyl methacrylate)	10-15	3.5	7.5	2	[5]

## References

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