

Supporting Information for the manuscript

## The Pivotal Step of Nanoparticle Functionalization for the Preparation of Functional and Magnetic Hybrid Opal Films

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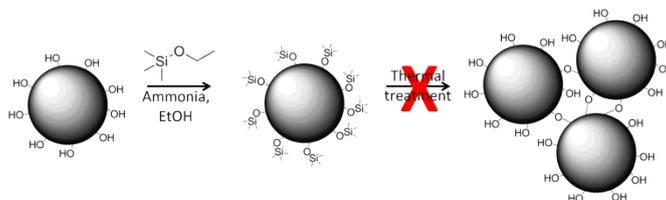


Fig. S1 Modification of silica surface with TMES avoiding inter-particle condensation reactions.

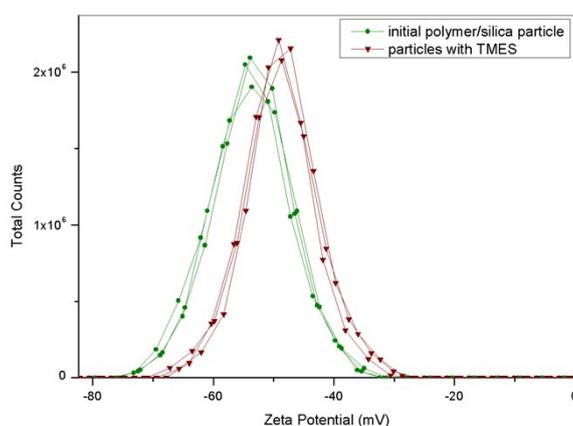


Fig. S2. Zeta potential of initial hybrid particles and TMES-modified particles.

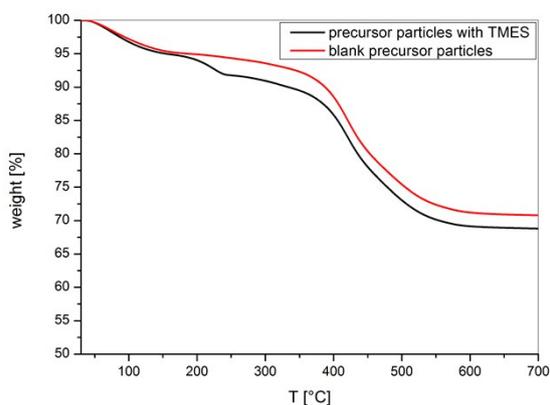
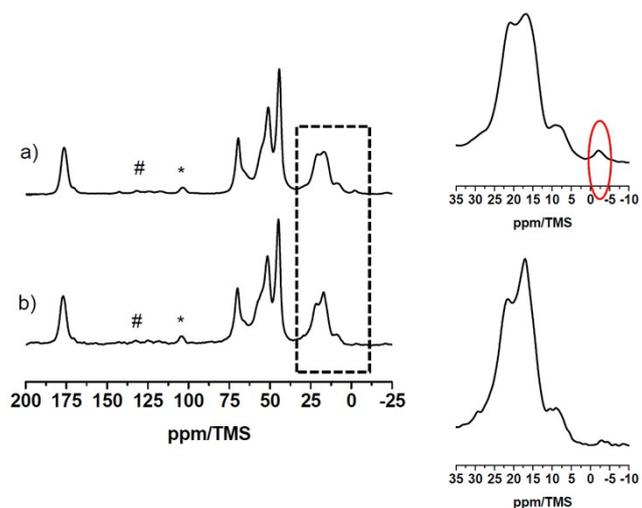
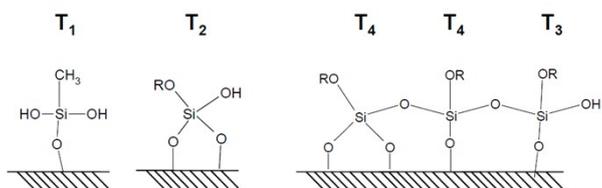


Fig. S3. TGA measurement of bare polymer/silica particles and particles modified with TMES.



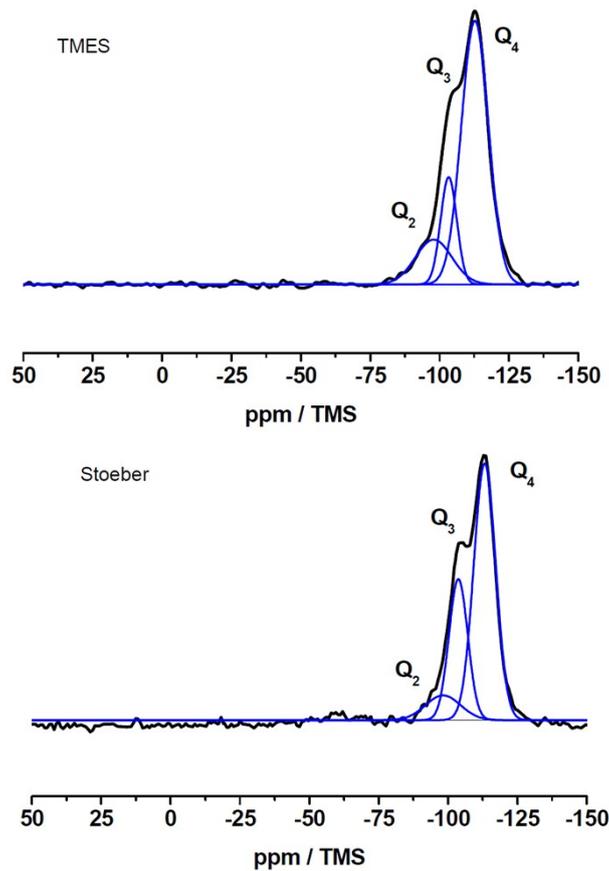
**Fig. S4.**  $^{13}\text{C}$  CP MAS spectra measured at 11 kHz spinning with a contact time of 2 ms of a) sample TMES, and b) sample Stöber. Note: Signals marked with # and \* are spinning sidebands.



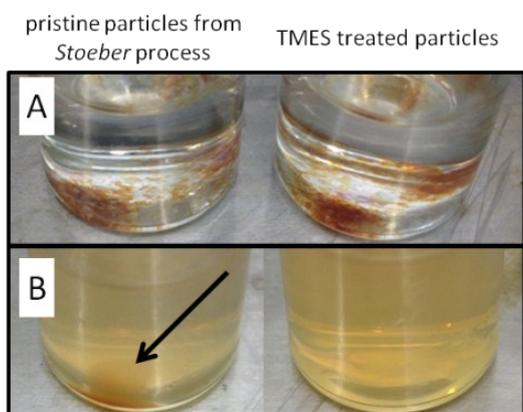
**Fig. S5.** Structural assignment of  $T_n$  groups (derived from Albert *et al.*).<sup>25</sup>

**Table S1.**  $T_1$  relaxation times for  $^{29}\text{Si}$  extracted by biexponential fit of the data from saturation recovery experiments.

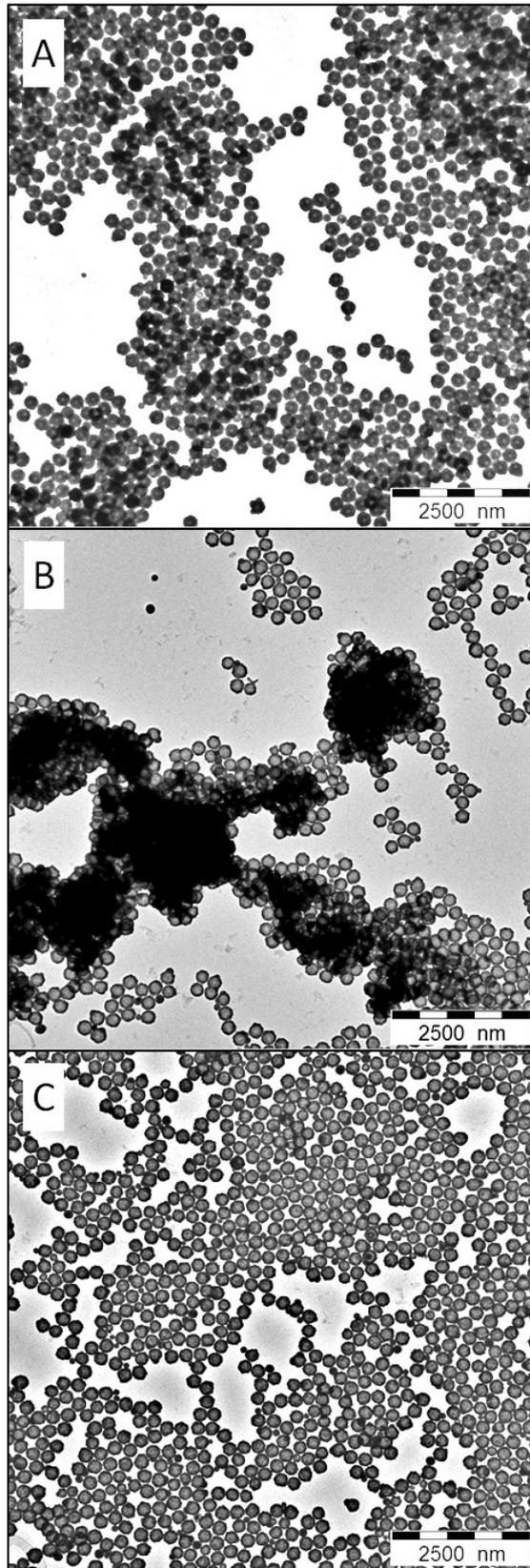
Sample Name	T1 short	T1 long
Sample TMES	15.4 s	134.4 s
Sample Stöber	1.9 s	62.1 s
Sample Nanorattle 500°C	1.5 s	31.5 s



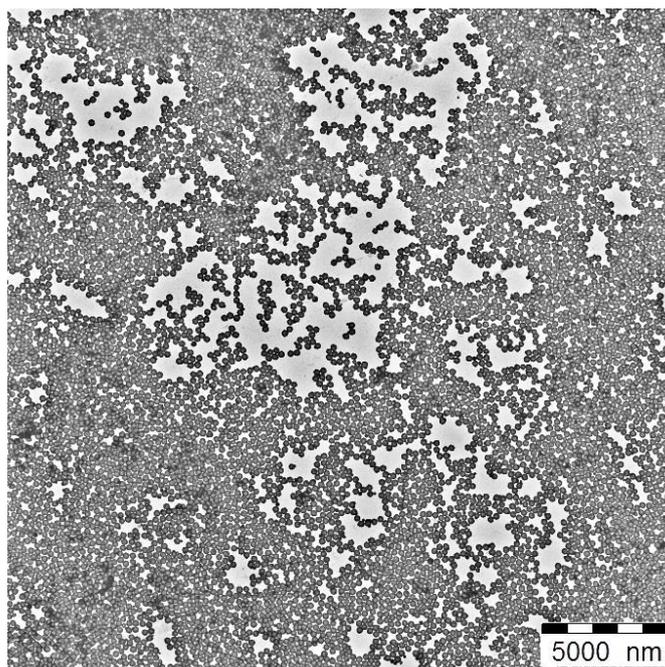
**Fig. S6.**  $^{29}\text{Si}$  MAS spectra measured at 8 kHz spinning for quantification of  $Q_n$  groups for sample TMES measured with  $d_1=412\text{s}$  and sample Stöber with  $d_1=300\text{s}$



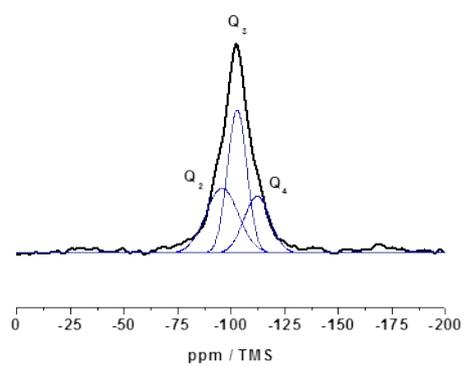
**Fig. S7.** Dispersions of the pristine particles obtained from the Stöber process (left) and particles treated with TMES (right) in water (after thermal treatment for 5h at 500 °C) are compared before (A) and after



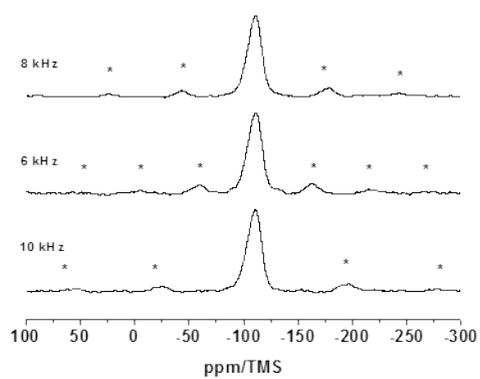
**Fig. S8.** TEM image of the particle dispersions in water on carbon-coated copper grids after drying using the drop-cast method for the initial polymer/silica particle sample (A), after thermal treatment 5h, 500°C (B) and TMES modified particles after thermal treatment (C).



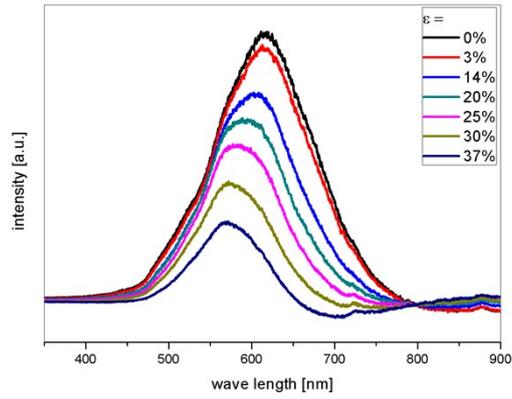
**Fig. S9.** TEM image of TMES-modified silica particles after thermal treatment and dispersing in water on carbon-coated copper grid after drying using the drop-cast method.



**Fig. S10.**  $^{29}\text{Si}$  CP-MAS spectra of nanorattles after heat treatment at 500 °C.



**Fig. S11.**  $^{29}\text{Si}$  MAS spectra measured at different spinning rates employing a recycle delay of 2.5s and 5400 scans for the 8 kHz respectively 1536 scans for the 6 and the 10 kHz spectra.



**Fig. S12.** Reflection spectra of elastomeric opal film dependent on strain  $\epsilon$ .

**Table S2.** Calculation of reflected wave lengths  $\lambda_{111}$  and relative reflectivity  $R$  for various opal film materials and particle diameter.

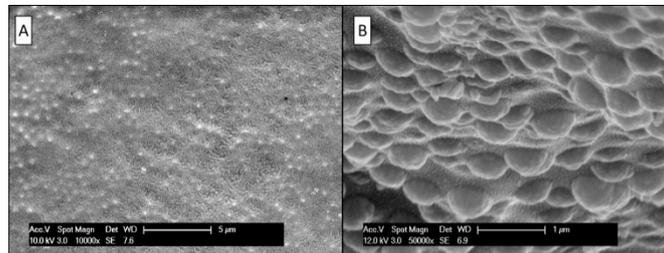
material	particle diameter	reflected wave length	relative reflectivity
Opal film PE/SiO <sub>2</sub>	670 nm	1430 nm	0.005
Opal film PE/SiO <sub>2</sub>	350 nm	745 nm	0.005
Opal film PEcoPBzMA/SiO <sub>2</sub>	350 nm	755 nm	0.071
Inversopal SiO <sub>2</sub> /air	350 nm	628 nm	0.998

The reflected wave length  $\lambda_{111}$  was calculated according to the following equation<sup>1</sup>:

$$\lambda_{111} = 2d \sqrt{\frac{3}{2} n_{eff} \sin \delta}$$

The relative reflectivity  $R$  is calculated according to the following equation<sup>2</sup>:

$$R = \left[ \frac{(n_0 - n_s \left(\frac{n_1}{n_2}\right)^{2N})^2}{(n_0 + n_s \left(\frac{n_1}{n_2}\right)^{2N})} \right]^2$$



**Fig. S13.** SEM image of polymer film surface (A) and cross section (B) of elastomeric polymer film containing yolk/shell particles.

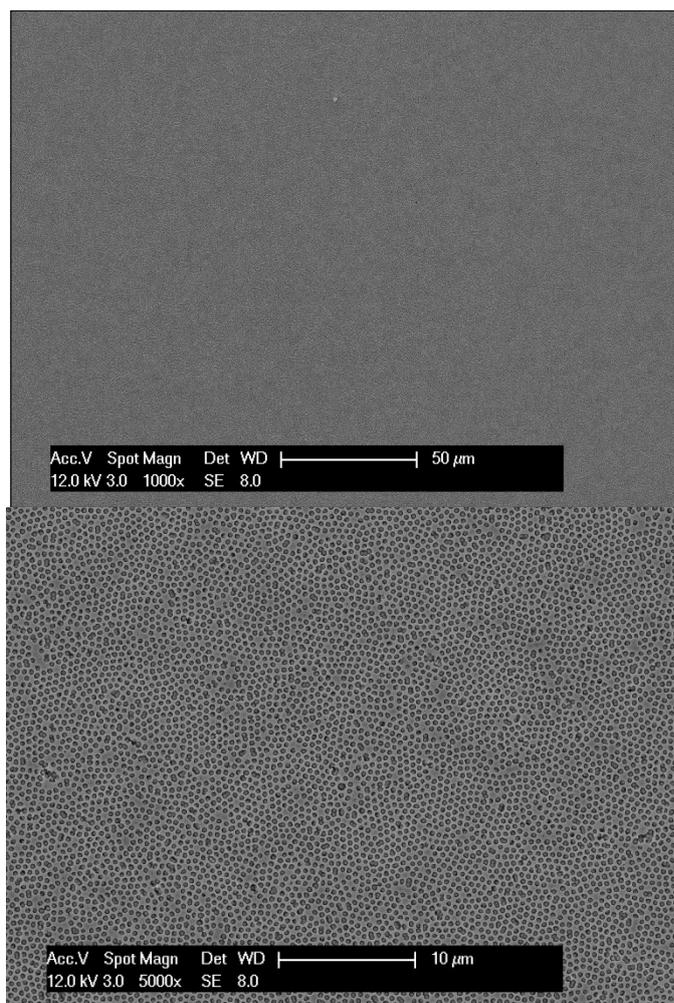


Fig. S14. SEM topography image of double inverse opal film.

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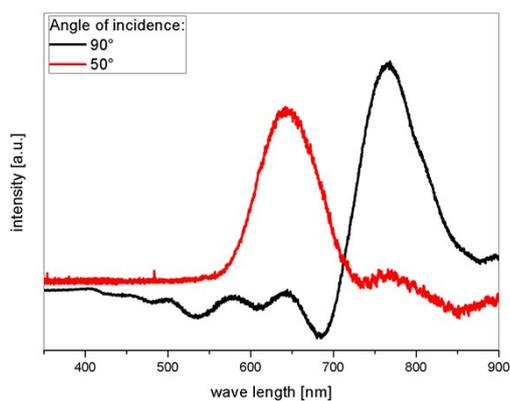


Fig. S15. UV/Vis reflection spectra of double-inverse opal film prepared by vertical deposition at 90° and 50° angle of incidence.

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## References

1. L. Gonzalez-Urbina, K. Baert, B. Kolaric, J. Perez-Moreno and K. Clays, *Chemical reviews*, 2012, **112**, 2268-2285.
2. J. Ge and Y. Yin, *Angew. Chem. Int. Ed. Engl.*, 2011, **50**, 1492-1522.