

## Supplementary data

# Optimized local synthetic conditions induce size reduction and phase purification in $\{[\text{Fe}(\text{Htrz})_2(\text{trz})](\text{BF}_4)\}_n$ spin crossover particles

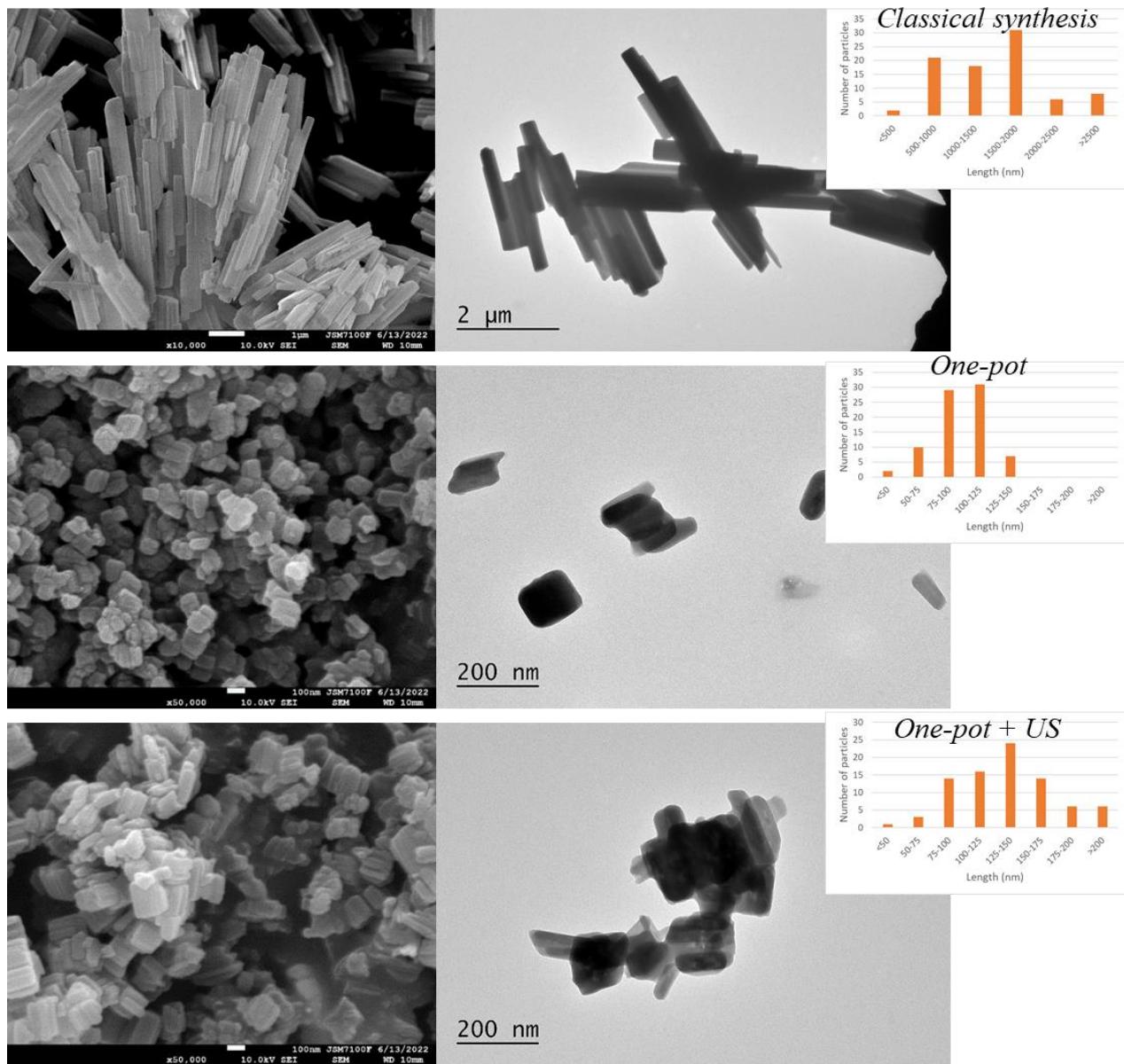
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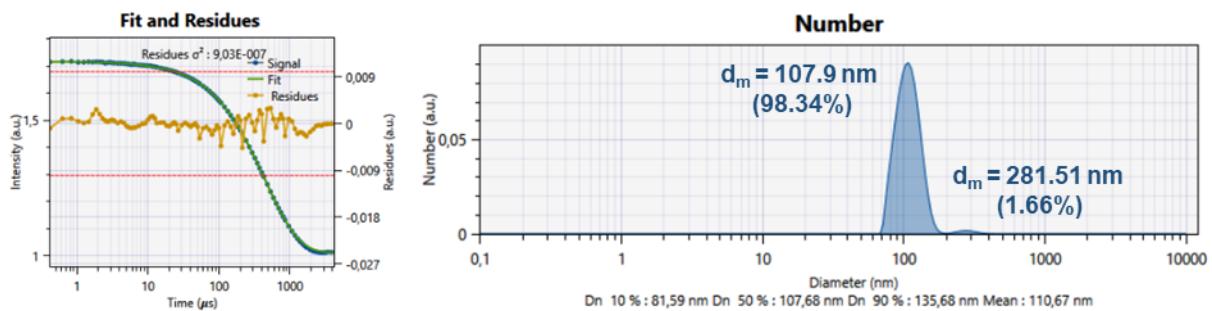
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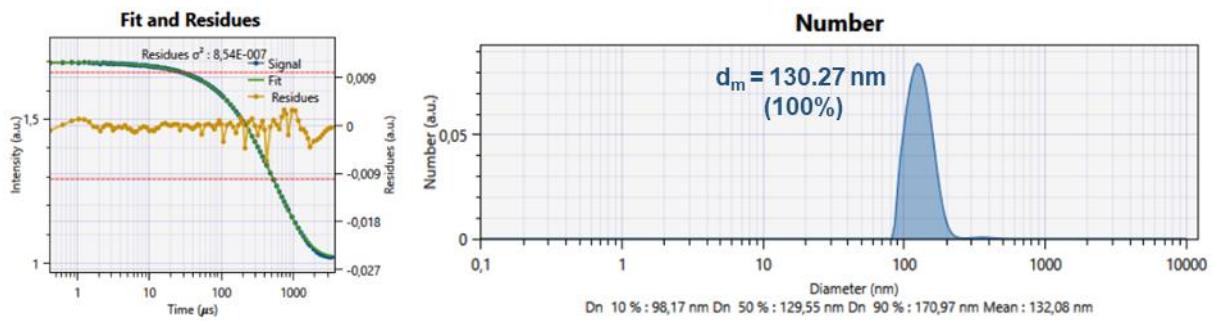
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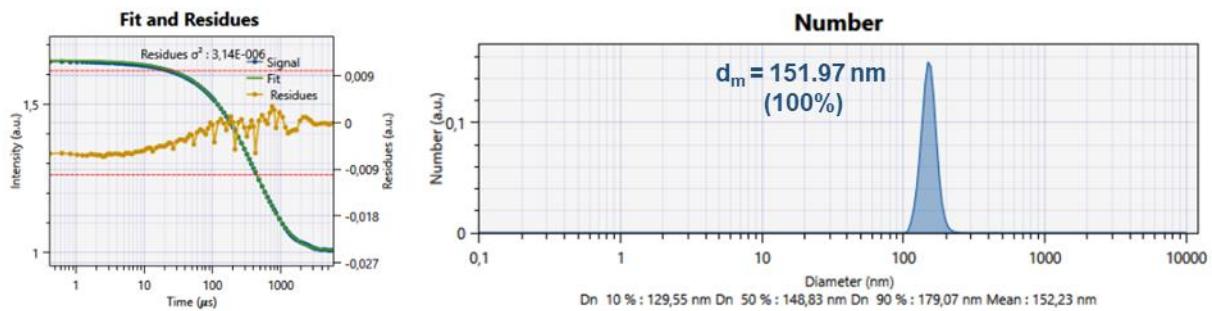
**Figure SI1.** SEM (left, x 5k or 50k) and TEM (right) images of samples **2a** (top), **2b** (center), **2c** (bottom) with their respective length size distributions obtained from TEM analyses over more than 60 particles (inset).



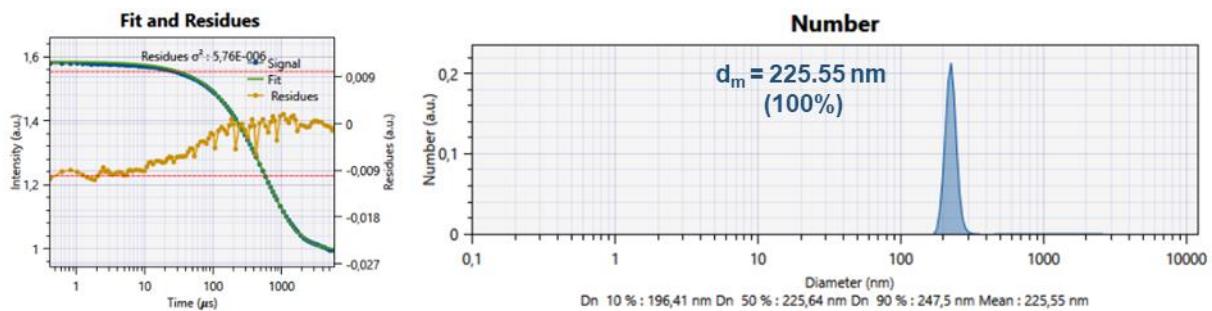
**Figure SI2.** Left: Correlogram of **1b** (dotted blue curve) with best fit (green curve) and corresponding residues (dotted orange curve). Right: Corresponding distribution size in number.



**Figure SI3.** Left: Correlogram of **1c** (dotted blue curve) with best fit (green curve) and corresponding residues (dotted orange curve). Right: Corresponding distribution size in number.



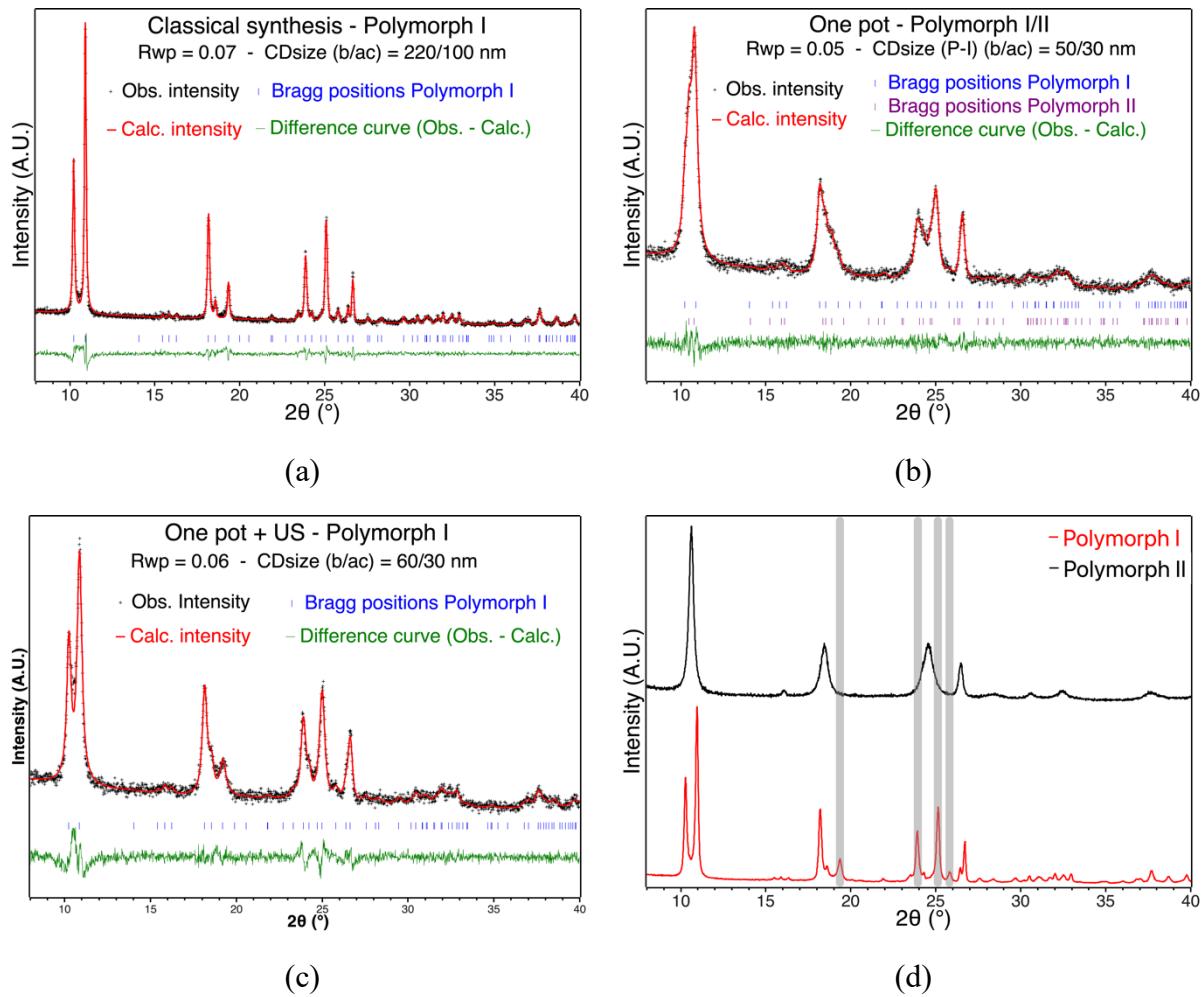
**Figure SI4.** Left: Correlogram of **2b** (dotted blue curve) with best fit (green curve) and corresponding residues (dotted orange curve). Right: Corresponding distribution size in number.



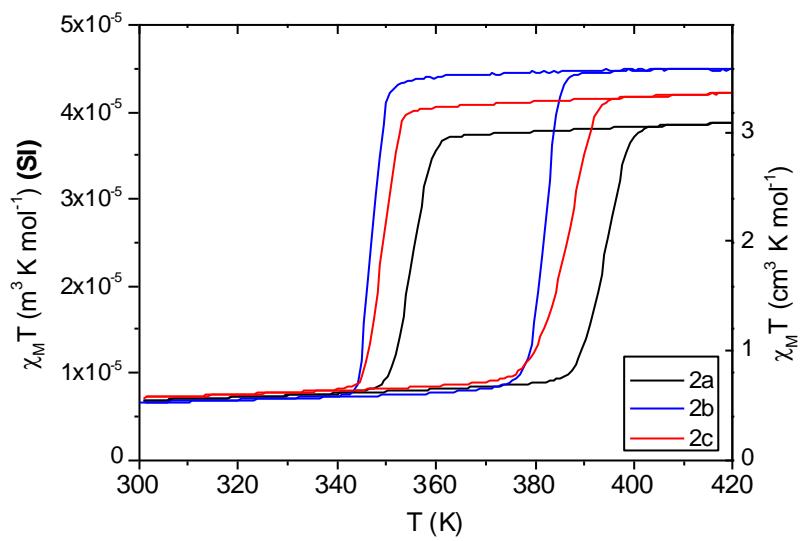
**Figure SI5.** Left: Correlogram of **2c** (dotted blue curve) with best fit (green curve) and corresponding residues (dotted orange curve). Right: Corresponding distribution size in number.

**Table SI1.** Characteristic parameters extracted from the above correlograms extracted with continuous multimodal Sparse Bayesian Learning (SBL) algorithm (in number) provided by the Nano Kin™ software (Cordouan Technologies, Pessac, France).

	<b>Mode (nm)</b>	<b>Mean (nm)</b>	<b>Std Dev (%)</b>	<b>Number (%)</b>	<b>Decay Rate (s<sup>-1</sup>)</b>	<b>Diffusion Coeff. (m<sup>2</sup>/s)</b>
<b>1b</b>	107.68	107.9	18.91	98.34	2741.05	38.36525 E-13
	271.48	281.51	17.38	1.66	1087.22	15.21709 E-13
<b>1c</b>	123.7	130.27	20.84	100	2388.03	33.3962 E-13
<b>2b</b>	148.83	151.97	13.04	100	1983.15	27.75721 E-13
<b>2c</b>	225.64	225.55	9.68	100	138.07	18.3085 E-13



**Figure S6.** PXRD pattern for samples (a) **2a**, (b) **2b**, (c) **2c** with profile matching refinement (Le Bail) and theoretical peak positions of phases **I** and **II**. Peak broadening can be unambiguously assigned to a size reduction and a phase impurity (“one-pot”) or a size reduction only “one-pot + US” on the basis of the refinement. Figure (d) shows the PDRX diagrams of pure polymorphs **I** and **II** showing some specific peaks of polymorph **I** (grey bands).



**Figure SI7.** Thermal evolution of the molar magnetic susceptibility  $\chi_M$  times the temperature for **2a** (black curve), **2b** (blue curve) and **2c** (red curve). These curves correspond to the 4<sup>th</sup> cycle.