## Supporting information

Determination of particle population ratio from experiment 4

## **Notation**

- $V_{tot}$ : Total volume of original particles
- $V_o$ : Volume of one octahedral particle
- $V_a$ : Volume of one amorphous particle
- $V_{tot}$ ': Total volume added to the particles by the regrowth
- $V_o$ ': Volume added to one octahedral particle by the regrowth
- $V_a$ ': Volume added to one amorphous particle by the regrowth
- $n_o$ : Number of octahedral particles
- $n_a$ : Number of amorphous particles

The total volume of  $YF_3$  in the system is known and distributed among both particle populations:

$$V_{_{tot}} = n_{_o}V_{_o} + n_{_a}V_{_a}$$

Since no new particles are formed, all the material is distributed among existing particles:

$$V_{iot} = n_{o}V_{o} + n_{a}V_{a}$$

By solving these simple equations we get:

$$n_{o} = \frac{V_{tot}V_{a} - V_{tot}V_{a}}{V_{o}V_{a} - V_{o}V_{a}}$$

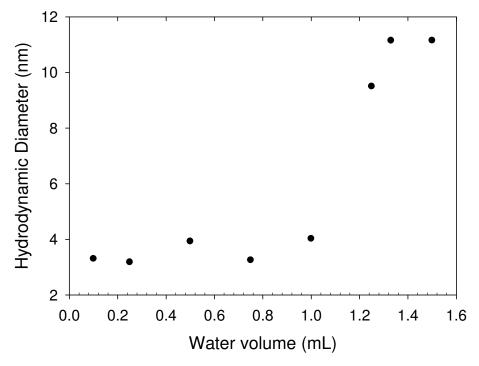
The total volumes of initial and added material are known from the amount of reagent used and the volumes of both particle populations are calculated from TEM size measurements. Therefore, it is possible to calculate the number of particles for both populations. Knowing that, the amount of reagent consumed by each population in the regrowth experiments can also be calculated.

Experiments	Yttrium addition	water content (mL) <sup>a</sup>	Fluoride addition	water content (mL) <sup>a</sup>
Original microemulsion	-	1.00	-	-
1	microemulsion <sup>b</sup>	0.75	microemulsion <sup>b</sup>	0.67
2	aqueous (0.05 mL)	1.05	microemulsion <sup>b</sup>	0.78
3	microemulsion <sup>b</sup>	0.75	aqueous (0.5 mL)	1.00
4	aqueous (0.05 mL)	1.05	aqueous (0.5 mL)	1.55
4b	aqueous (0.1 mL)	1.10	aqueous (0.5 mL)	1.60
4c	aqueous (0.15 mL)	1.15	aqueous (0.5 mL)	1.65

Table 1. Microemulsion water content after each addition for the regrowth experiment<sup>a</sup>.

<sup>a</sup> This is the water volume per 15 mL of cyclohexane. The surfactant content is the same for all experiments.

<sup>b</sup> Added microemulsion contained 0.5 mL water, 2 g surfactant and 15 mL cyclohexane.



**Figure 1.** Hydrodynamic diameter of reverse micelles, measured by DLS, as a function of water volume with 2 g of Igepal CO520 in 15 mL of cyclohexane<sup>1</sup>.

(1) Lemyre, J.-L.; Lamarre, S.; Beaupré, A.; Ritcey, A. M., Langmuir 2010, 26, (13), 10524.