Supporting Information

Controlled Synthesis of Fluorescent Silica Nanoparticles inside Microfluidic Droplets

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Experimental setup of the bleaching experiment



Figure 1: For the bleaching experiment, two capillaries were filled with fluorescent solution (containing fluorescent nanoparticles or pure FITC). The sample capillary was constantly illuminated with light from a mercury lamp. Every 30 seconds, an image of the sample capillary and the reference capillary was taken.

Mixing in microdroplets

To demonstrate the efficient mixing of reagents in droplets which are transported through the meandering outlet channel, we injected ink-dyed ethanol and pure ethanol in the HM inlet and the SA inlet, respectively. As shown in Fig. 2, after only two U-turns, the intensity profile of a bicolored droplet flattened dramatically, indicating fast mixing.



Figure 2: Microscope image showing mixing of ink-dyed and pure ethanol inside moving droplets.

Estimation of the production rate of nanoparticles

From the flow rate of SA (0.003 μ L/s = 0.018 μ L/min) and the concentration of APTES and TEOS in SA (0.025 M and 0.25 M, respectively), we calculate molar flow rates: q_{APTES} =4.5 nmole/min and q_{TEOS} =45 nmole/min. We make the following assumptions:

- APTES is completely integrated in the silica network (100% reaction efficiency). This assumption is reasonable since APTES shows high reactivity towards even small amounts of water.
- 90% of TEOS is converted into silica. This assumption is based on the observation that in the Stöber process only a few ethoxy side chains may not be hydrolyzed ¹.

• The number of colloidally stable particles does not change during the growth of the particles, i.e. between 50 nm and 400 nm.

We therefore find a total silica conversion rate of $4.5 \mu g/min$.

From the curves shown in figure 2a) of the main manuscript, we may safely assume that the maximum attainable diameter of the nanoparticles is 400 nm ($3.35*10^{-14}$ cm³). According to Plumeré *et al.*², we expect a range of nanoparticle densities 1.6 g/cm³ $\leq \rho_{SiO2} \leq 2.0$ g/cm³ and therefore obtain a production rate between 67*10⁶ min⁻¹ and 84*10⁶ min⁻¹.

References

- 1. G. H. Bogush and C. F. Zukoski IV, J. Colloid Interface Sci., 1991, 142, 1-18.
- 2. N. Plumeré, A. Ruff, B. Speiser, V. Feldmann and H. A. Mayer, *J. Colloid Interface Sci.*, 2012, **368**, 208-219.