Supporting Information

for

Droplet-based synthesis of homogeneous magnetic iron oxide nanoparticles

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Additional experimental data

Table S1: Droplet reaction conditions for comparing droplet and batch syntheses.

	value	unit
concentration $FeCl_3 \cdot 6H_2O$ (solution A)	0.06	M
concentration $FeCl_2 \cdot 4H_2O$ (solution A)	0.03	M
concentration NH_3 (solution B)	4	M
flow rate (solutions A and B, mineral oil)	5	µL/min
temperature	70	°C
residence time	11.1	min

Table S2: Batch reaction conditions droplet and batch syntheses.

name	value	unit
concentration $FeCl_3 \cdot 6H_2O$ (solution A)	0.06	M
concentration $FeCl_2 \cdot 4H_2O$ (solution A)	0.03	M
concentration NH_3 (solution B)	4	M
amount (solutions A and B), ratio 1:1	100	µL
temperature	70	°C
reaction time	11.1	min

Table S3: Droplet reaction conditions used for analyzing the effect of temperature on the particle size.

name	value	unit
concentration $FeCl_3 \cdot 6H_2O$ (solution A)	0.06	M
concentration $FeCl_2 \cdot 4H_2O$ (solution A)	0.03	M
concentration NH_3 (solution B)	4	M
flow rate (solutions A and B, mineral oil)	20	µL/min
temperature	50/60/70/80/90	°C
residence time	2.8	min

Table S4: Droplet reaction conditions used for analyzing the effect of residence time on the particle size.

name	value	unit
concentration $FeCl_3 \cdot 6H_2O$ (solution A)	0.06	M
concentration $FeCl_2 \cdot 4H_2O$ (solution A)	0.03	M
concentration NH_3 (solution B)	4	M
flow rate (solutions A and B, mineral oil)	3/5/10/20/25	µL/min
temperature	70	°C
residence time	18.5/11.1/5.6/2.8/2.2	min

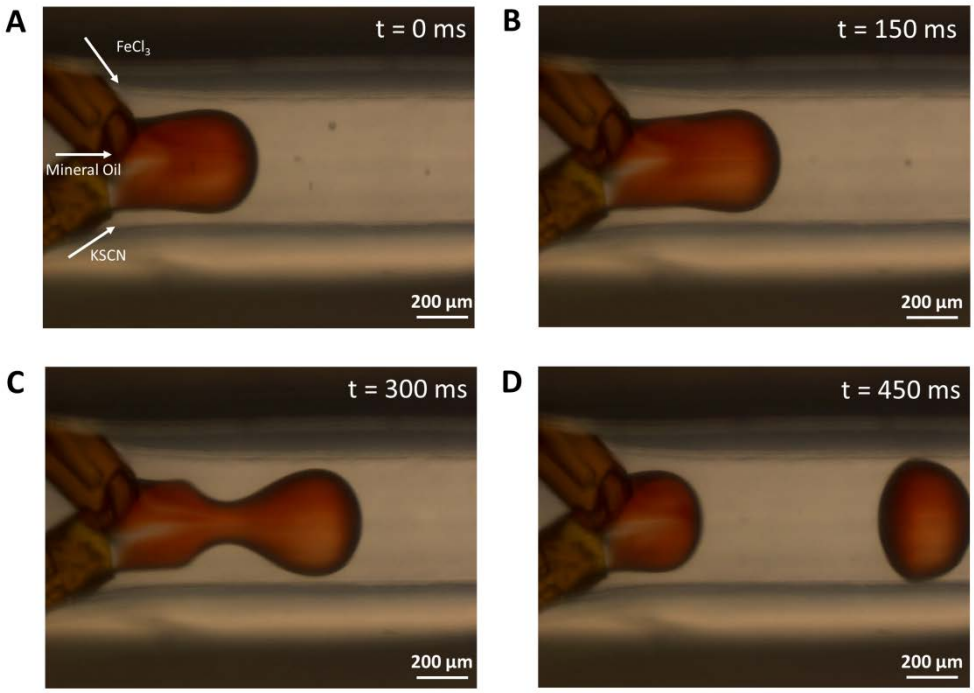
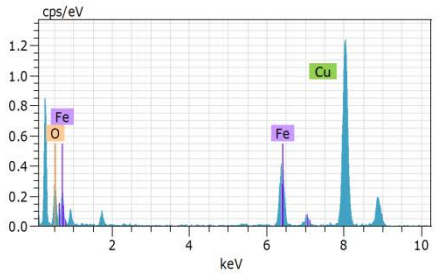


Figure S1: Microscope time-lap images illustrating mixing within the droplets taken at *t* = 0 ms (A), *t* = 150 ms (B), *t* = 300 ms (C), and *t* = 450 ms (D). For this experiment a colorless solution of $FeCl_3$ is flowed through the capillary coming from the top, and another colorless solution of KSCN is flowed through the capillary coming from the bottom. Upon mixing, a red iron complex is formed immediately ($FeCl_3$ + 3KSCN → $Fe(SCN)_3$ + 3 KCl), indicating mixing by the color change. Flow rates are 10 µL/min for the aqueous solution from the capillaries and 10 µL/min for the mineral oil, respectively.

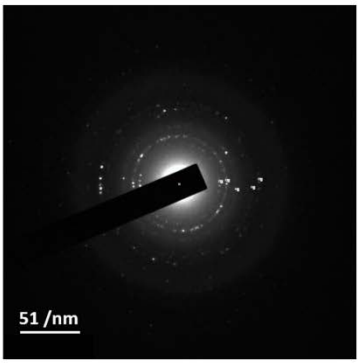
A



B

Element	At%
Oxygen	59.28
Fe	40.72

C



D

Spot No.	D-Spacing [Å]	Spinel crystalline plane
1	2.9	2 2 0
2	2.5	3 1 1
3	2.1	4 0 0
4	1.6	5 1 1
5	1.5	4 4 0

Figure S2: Energy-dispersive X-ray spectroscopy (EDS) of nanoparticles synthesized in droplets, recorded EDS spectrum of the particles (A). The large copper peak originates from the copper grid used for sample preparation. Elemental composition of the nanoparticles extracted from EDS spectrum (B), confirming that the particles consist of Fe_3O_4. The electron diffraction pattern of the particles confirms the crystallinity of the nanoparticles (C). Table of *d*-spacing values extracted from electron diffraction pattern with the corresponding spinel crystalline planes indicated (D).

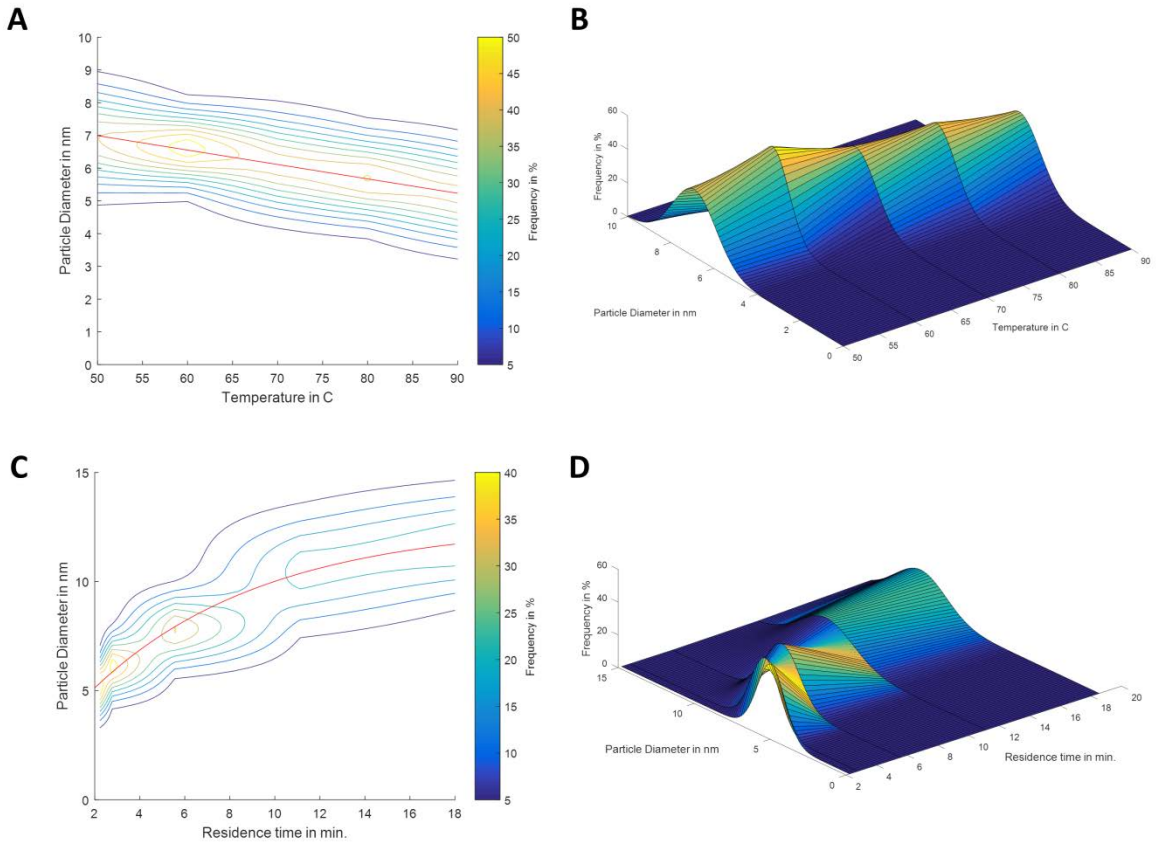


Figure S3: Contour (A) and surface plot (B) of the particle size distributions dependence on temperature, and contour (C) and surface plot (D) of the particle size distributions dependence on droplet residence time. In both contour plots, the fitted liner/exponential function is indicated by a red line.