## **ELECTRONIC SUPPORTING INFORMATION**

## Exploring the transition of polydopamine-shelled perfluorohexane emulsion droplets into microbubbles using small- and ultra-small-angle neutron scattering

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## SUPPLEMENTARY FIGURES



**Figure S1.** Representative transmission electron micrographs. PDA/PFH emulsion droplets with polymerisation times of 2 hours (**A**) and 4 hours (**B**). Emulsion droplets with broken and "flipped" PDA shells (**C**). Collapsed, dried PDA shells of PDA/PFH emulsion droplets with polymerisation times of 1 hour (**D1** and **D2**), 3 hours (**D3** and **D4**), and 5 hours (**D5** and **D6**). Collapsed PDA shells surrounding curved regions that are potentially interfaces of preexistent emulsions or bubbles (**E**). Yellow lines indicate the suspected interface of the colloidal material. Scale bars = 100 nm (**A**, **B**, **C**, **D1–D3**, **D5**, **D6**, and **E**) and 50 nm (**D4** and **D6**).



**Figure S2.** 2D plots of the USANS spectra of samples 2H (**A**) and 4H (**B**) in bubblematched medium (9%  $D_2O/$  91%  $H_2O$ ) and of samples 2H (**C**) and 4H (**D**) in dropletmatched medium (61%  $D_2O/$  39%  $H_2O$ ) at different temperatures. **E**, **F**, **G**, and **H** shows **A**, **B**, **C**, and **D**, respectively, with vertical offset (10× original scattering intensity per temperature increment). Scattering intensities in 61%  $D_2O/$  39%  $H_2O$  and in 9%  $D_2O/$  91%  $H_2O$  arise from microbubbles and emulsion droplets, respectively.



**Figure S3.** 2D plots of the SANS spectra of samples 2H (**A**) and 4H (**B**) in bubble- matched medium (9%  $D_2O/91\%$  H<sub>2</sub>O). **C** and **D** shows **A** and **B**, respectively, with vertical offset (10× original scattering intensity per temperature increment). Scattering intensities in 9%  $D_2O/91\%$  H<sub>2</sub>O arise mainly from emulsion droplets.



**Figure S4.** Scatter plots and model fitting (red line) of the USANS spectra of PDA/PFH emulsions (polymerisation times = 2 hours (**A**) and 4 hours (**B**)) in different contrast-matched dispersing media at 20°C, 90°C, and 20°C (after cooling from 90°C). Scattering intensities in 61%  $D_2O/39\%$  H<sub>2</sub>O and in 9%  $D_2O/91\%$  H<sub>2</sub>O arise from microbubbles and emulsion droplets, respectively.

**Table S1.** Power-law exponents calculated from SANS spectra of emulsion samples in bubble-matched media (9%  $D_2O/$  91%  $H_2O$ ).

Temperature (°C)	Power-law exponent	
	Sample 2H	Sample 4H
20	4.00 ± 0.01	4.00 ± 0.02
30	4.00 ± 0.01	$4.00 \pm 0.02$
40	4.00 ± 0.01	4.00 ± 0.01
50	4.00 ± 0.01	4.00 ± 0.01
60	4.00 ± 0.01	$4.00 \pm 0.01$
70	$4.00 \pm 0.02$	$4.00 \pm 0.01$
80	$4.00 \pm 0.02$	$4.00 \pm 0.03$
90	$4.00 \pm 0.02$	$4.00 \pm 0.02$