Supplementary Information

Water reorientation dynamics in nano-emulsions

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SI1. Emulsion size distribution

Figures SI1, SI2 and SI3 show exemplary DLS measurements of the full droplet size distributions of emulsions with different SDS concentrations and different processing pressures.
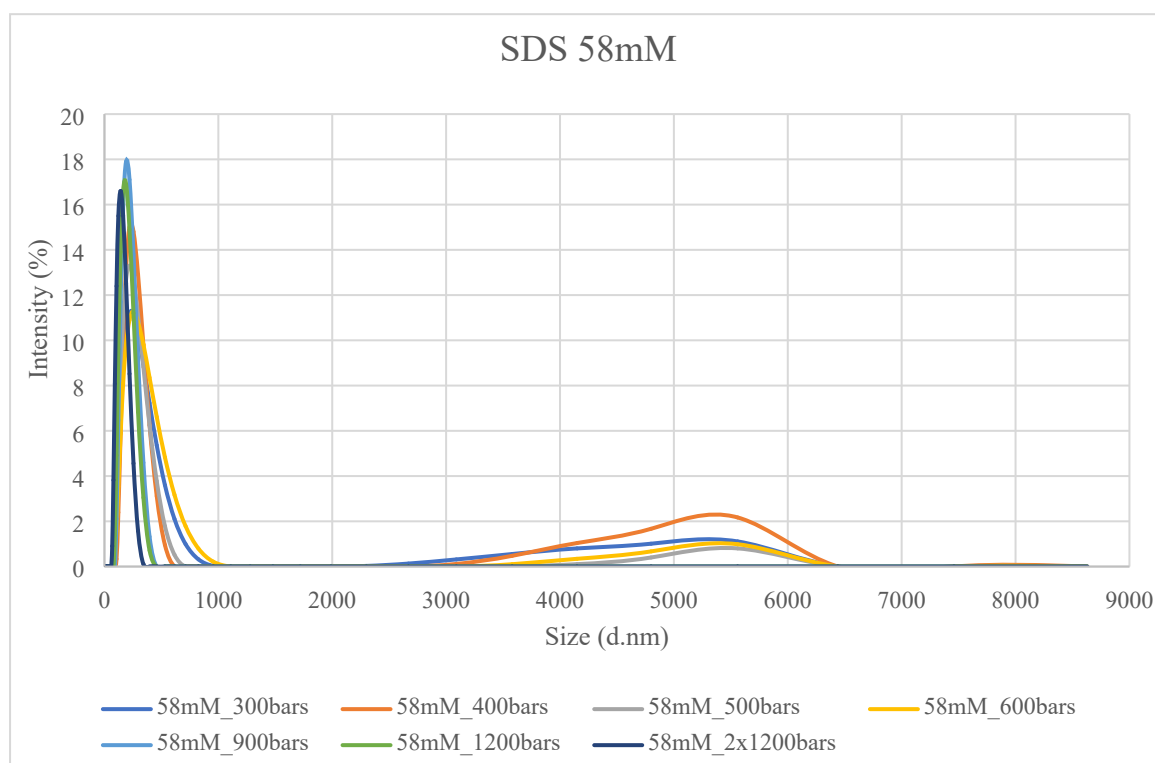

Figure S1. Full droplet size distribution for emulsions with SDS concentration 58 mM, at different processing pressures.
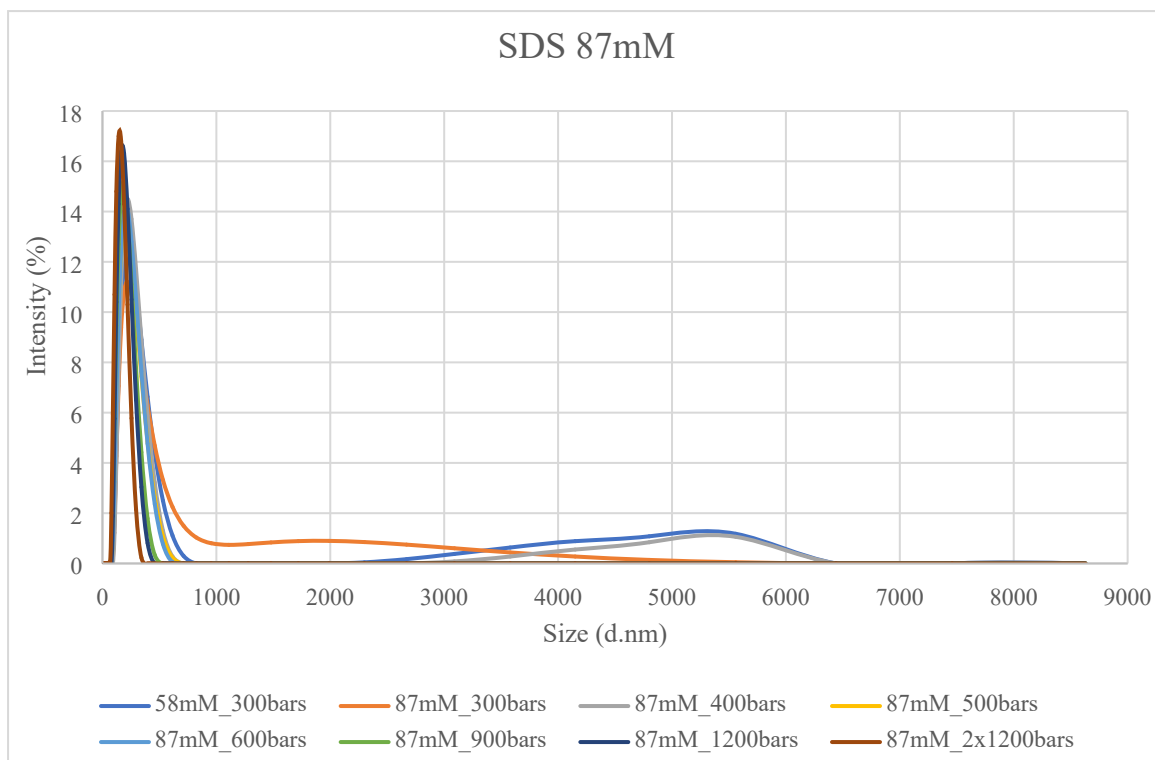
Figure S2. Full droplet size distribution for emulsions with SDS concentration 87 mM, at different processing pressures.
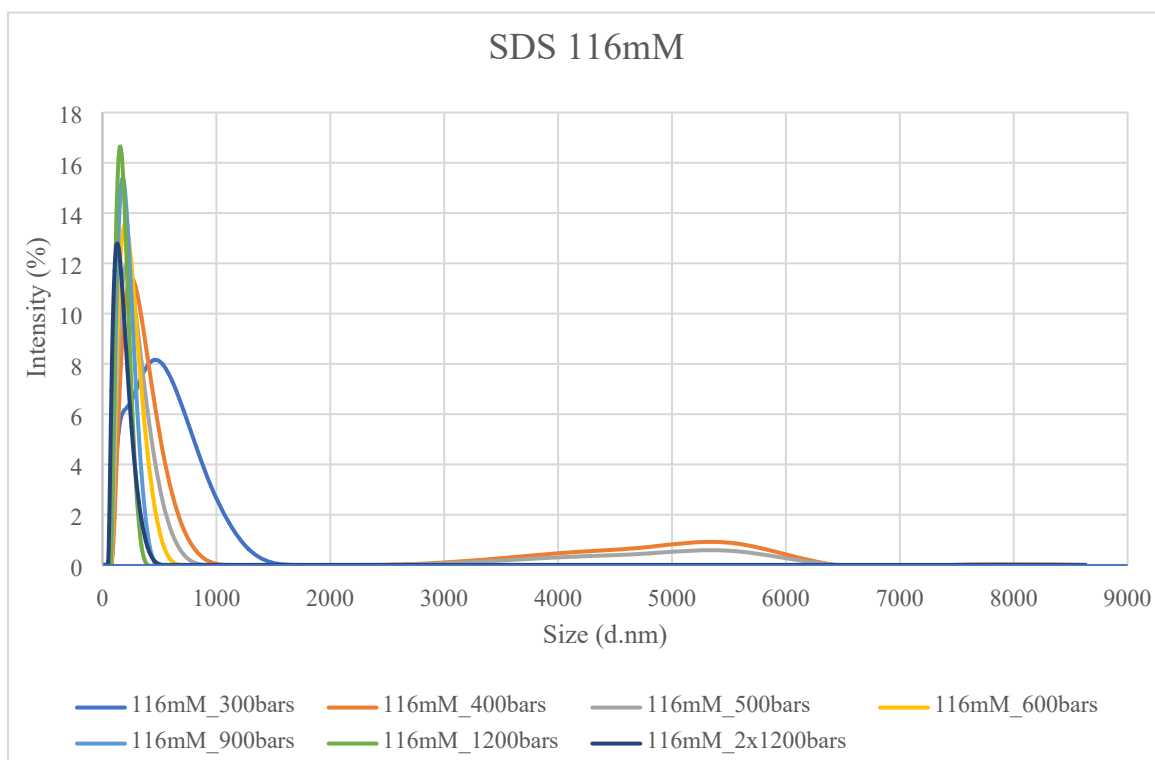

Figure S3. Full droplet size distribution for emulsions with SDS concentration 116 mM, at different processing pressures.
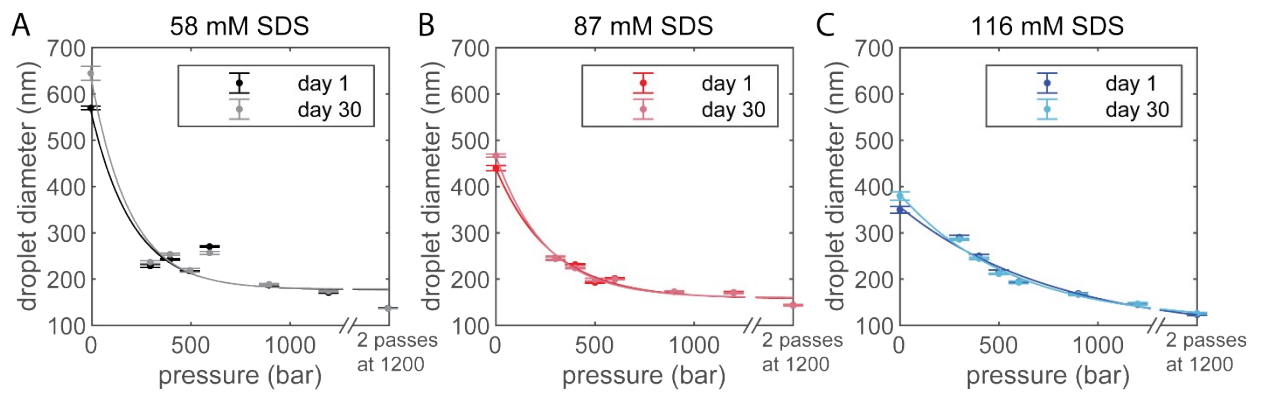
SI2. Emulsion stability

Figure SI4 Droplet diameter as a function of processing pressure of the ultra high-pressure impinging jet mixer, for surfactant concentrations of (A) 58 mM (B) 87 mM and (C) 116 mM SDS. We show the stability of the emulsions by reporting the droplet diameter measured on the day of emulsions preparation (day 1) and 30 days later (day 30). The solid lines are guides to the eye.

To examine the stability of the prepared nano-emulsions, we measured their size again, 30 days after preparation. Figure SI4 shows the droplet size as a function of processing pressure for all surfactant concentrations, at the day of preparation (day 1) and 30 days later. We observe that within this time window, the emulsions prepared using the ultra high-pressure impinging jet mixer, do not change significantly in size.

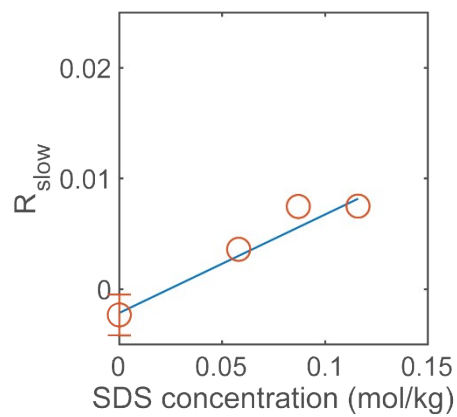## SI3. Slow reorienting OH groups per SDS molecule

Figure SI5. Offset of the anisotropic decay for as a function of SDS concentration. The solid line is a linear fit with a slope of 0.089.

We calculate the number of slowly reorienting water OH groups per SDS surfactant molecule from the concentration dependence of the offset of the anisotropy decay. Figure SI5 shows the offset, Rslow, as a function of concentration. We fit Rslow with a linear function with a slope of 0.089±0.009, meaning that each surfactant molecule slows down 25±2 water OH groups.
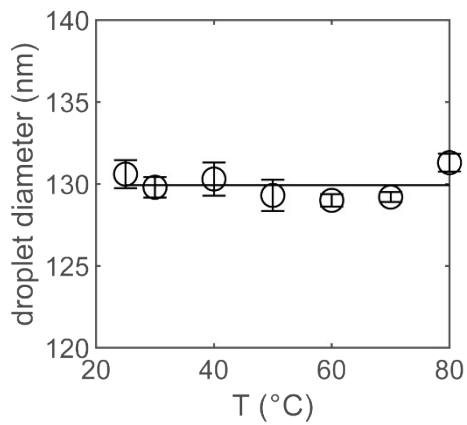
SI4. Temperature size dependence

Figure SI6 Droplet diameter as a function of temperature, for a a nano-emulsion composed of 50% econa oil and 116 mM SDS in isotopically diluted water.

Figure SI6 shows the droplet size of a nano-emulsion composed of 50% econa oil and 116 mM SDS in isotopically diluted water as a function of measurement temperature. The droplet size does not change significantly upon changing the measurement temperature.

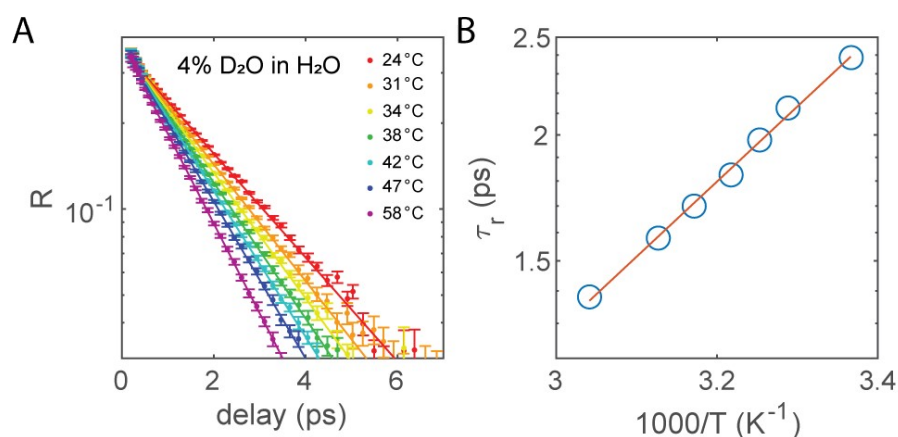## SI5. Temperature dependent pump-probe measurements of isotopically diluted water

Figure SI7. (A) Anisotropy decay of the OD stretch vibration as a function of delay time for a solution of 4% D_2O in H_2O, at different temperatures. The solid lines are fits to an exponential function. (B) Reorientation time τ_r as a function of inverse temperature, obtained from the fit to the anisotropy decay. The solid line represents an Arrhenius-type fit. Reproduced from the Supporting Information of reference [1].

We fit the anisotropy decays of the nano-emulsion at different temperatures with an exponential decay with an offset. The reorientation time τ_r is determined from independent measurements of isotopically diluted water at different temperatures, and the reorientation time obtained is fixed in the anisotropy decay fit of the nano-emulsion.

Figure SI7A shows the anisotropy decay of isotopically diluted water as a function of delay time, for different temperatures. Since the anisotropy of isotopically diluted water does not contain a slow component, we fit the anisotropy with a single exponential decay. The obtained exponential reorientation time constant is shown in figure SI7B, plotted on a logarithmic scale as a function of the inverse temperature. The reorientation time decreases with increasing temperature, and this relation can be described with an Arrhenius-type fit using the following the equation:

$$1/\tau_r = Ae^{-E_{act}/RT}$$

Where A is the pre-exponential factor, E_{act} is the activation energy (in J/mol), and R is the gas constant. For isotopically diluted water we find the activation energy to be 14±1 kJ/mol, in agreement with previous findings .[2–4] For the fit of the anisotropic decay of the nano-emulsion, we use the reorientation time determined by the Arrhenius fit for isotopically diluted water, while keeping the anisotropy offset as a free fitting parameter.

References

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(3)   Tielrooij, K. J.; Hunger, J.; Buchner, R.; Bonn, M.; Bakker, H. J. Influence of Concentration and Temperature on the Dynamics of Water in the Hydrophobic Hydration Shell of Tetramethylurea. *J. Am. Chem. Soc.* **2010**, *132* (44), 15671–15678.

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