Supporting Information - Additional Model Fits

Structural information (i.e. size and shape) about the micelles in solution was determined from the SANS profiles by fitting data to established models for spheres and ellipsoids. Model fits to the scattering profiles were performed using SasView (www.sasview.org), an open source, collaboratively developed software for the analysis of small angle scattering data. This process employed a Levenberg-Marquardt, a.k.a. damped least-squares, algorithm for optimization of the fit. The Q-range used for fitting was $0.008 < Q < 0.5$ Å^{-1}. Data were well-described by oblate spheroid shapes, in agreement with previous theory,[1] simulations,[2] and experimental results.[3-5] Simultaneous fits of the scattering data to spherical models were included for comparison (Figure S2).

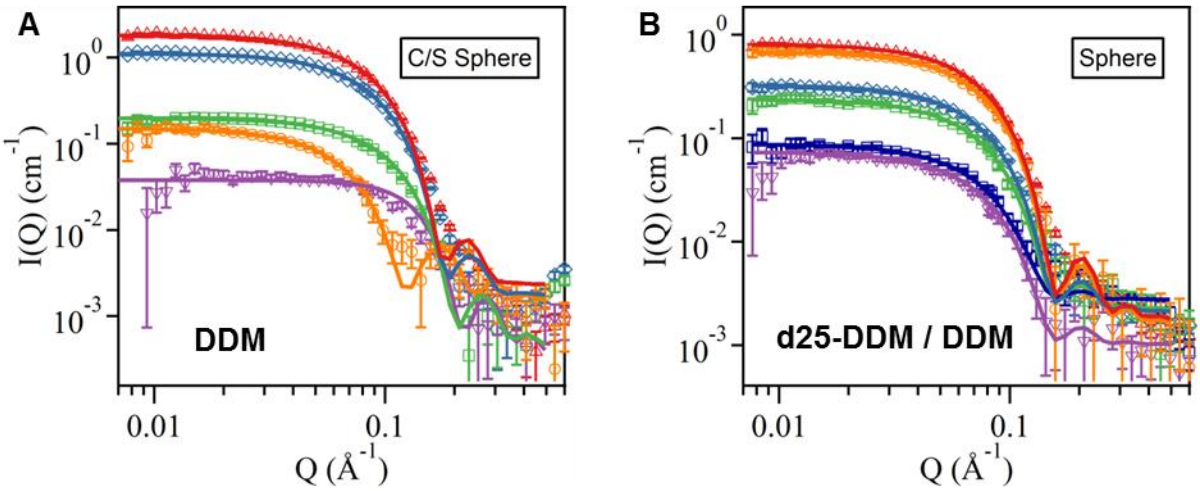

Figure S2. Simultaneous model fits from SasView A) to the DDM contrast series data shown as solid lines using a core-shell (C/S) sphere model, and B) to the d25-DDM / DDM contrast series data using a solid sphere model. C/S sphere fit: R = 22.2 Å, T_s = 7.4 Å; χ^2 = 13.5. Solid sphere fit: R = 28.2 Å, χ^2 =: 4.1. Same color coding used as in Figure 2.

The shape of an oblate ellipsoid is defined by rotating an ellipse about its minor axis, such that it contains an equatorial radius (R_{equat}) greater than the polar radius (R_{polar}). A measure of the shape's ellipticity (ϵ) is given by $\sqrt{(1-(R_{po}^2 / R_{eq}^2))}$.[6] For the case of DDM micelles with an inherent contrast between the detergent head and tail, a core-shell ellipsoid model was used with an outer layer of uniform thickness (T_{shell}) corresponding to the shell formed by detergent head groups and core composed of alkyl chains. A solid ellipsoid model was employed for the mixed micelles engineered with equal SLDs between the core and shell. This method permits a straightforward comparison of the resulting size (total dimensions) and shape (ellipticity) for each micelle condition.

In order to obtain the model parameters (polar and equatorial radii for the solid ellipsoid model, with an added shell thickness for the core-shell ellipsoid model), calculated values from MULCh for the SLDs of all micelle components and buffer were held fixed. As the parameter x_{core} describing the ratio of the polar and equatorial radii was directly obtained from the model fitting process, the product of this factor and the equatorial radius yielded the polar radius. This fitting process was performed independently for each SANS measurement in the contrast series, and also as a simultaneous fit to each full contrast series (DDM and d25-DDM / DDM). The simultaneous fitting process involved fitting to the whole contrast series as opposed to a single contrast condition. Uncertainties in the model fits were determined using a two-tails confidence test with degrees of freedom equal to the sum of the total data points and model fitting parameters minus one, and a significance level corresponding to 1σ (68% confidence).

Individual model fits serve to highlight that the mixed micelle system was represented by a solid object with consistent dimensions at all contrast points, as well as the effect of contrast on resolving different micelle components when either core or shell are near solvent match points in

the DDM micelles. Individual fit parameters are summarized by Tables S2 and S3 for direct comparison to the simultaneous fit results (Table S4).

Table S2. Table of ellipsoid model fit results to individual curves of the d25-DDM / DDM mixed micelle contrast series.

D_2O (%)	SLD (10^{-6} Å^{-2})	Scale (10^{-3} cm^{-1})	Bckgrd (10^{-3} cm^{-1})	R_{polar} (Å)	R_{equat} (Å)	Chi-squared
0	2.33	9.1	1.3	20.6 ±0.5	33.1 ±0.4	0.70
22	2.55	11.1	2.0	18.3 ±1.2	34.1 ±0.7	1.00
35	2.68	11.7	1.0	21.8 ±1.6	32.2 ±1.3	0.93
65	2.98	12.2	2.6	15.6 ±0.5	33.5 ±0.7	0.94
80	3.13	10.8	1.8	18.1 ±0.2	33.3 ±0.2	1.24
100	3.32	9.8	1.3	18.8 ±0.1	33.6 ±0.1	0.92

The SLD of the solvent was determined for each contrast point using the relationship given from MULCh analysis: $SLD_{solvent}$ (10^{-6} Å^{-2}) = -0.532 + 6.893·f_{D2O}.

Table S3. Table of core-shell ellipsoid model fit results to individual curves of the DDM micelle contrast series.

D_2O (%)	SLD_{shell} (10^{-6} Å^{-2})	Scale (10^{-3} cm^{-1})	Bckgrd (10^{-3} cm^{-1})	R_{polar} (core) (Å)	R_{equat} (core) (Å)	Chi-squared
0	1.87	9.7	1.3	14.0 ±6.4	29.3 ±8.7	0.97
22	2.31	12.8	1.7	13.8 ±2.0	26.0 ±2.0	0.88
35	2.57	9.0	0.6	16.0 ±2.3	29.1 ±2.2	0.65
49	2.84	9.4	0.7	16.5 ±0.8	26.2 ±0.4	1.54
80	3.46	15.8	0.9	11.5 ±0.6	25.5 ±1.3	1.35
100	3.86	11.4	1.0	12.3 ±0.3	26.2 ±0.5	2.74

The SLD of the solvent was determined for each contrast point using the relationship: $SLD_{solvent}$ (10^{-6} Å^{-2}) = -0.532 + 6.894·f_{D2O}. The SLD_{core} contains no exchangeable hydrogen, and remains constant over the range of D_2O in the solvent. SLD_{core} (10^{-6} Å^{-2}) = -0.394.

Table S4. Summary of the simultaneous model fit results for each contrast series.

contrast series	R_{polar} (Å)	R_{equat} (Å)	T_{shell} (Å)
d25-DDM/DDM	19.7 ±0.6	33.1 ±0.5	

DDM	14.4 ±0.9	27.4 ±1.0	6.7 ±1.0

(1) Tanford, C., *The Hydrophobic Effect: Formation of Micelles and Biological Membranes 2d Ed*. J. Wiley.: 1980.
(2) Abel, S.; Dupradeau, F.-Y.; Raman, E. P.; MacKerell, A. D.; Marchi, M., Molecular Simulations of Dodecyl-β-maltoside Micelles in Water: Influence of the Headgroup Conformation and Force field Parameters. *J. Phys. Chem. B* **2011,** *115* (3), 487-499. DOI: 10.1021/jp109545v.
(3) Oliver, R. C.; Lipfert, J.; Fox, D. A.; Lo, R. H.; Doniach, S.; Columbus, L., Dependence of Micelle Size and Shape on Detergent Alkyl Chain Length and Head Group. *Plos One* **2013,** *8* (5). DOI: 10.1371/journal.pone.0062488.
(4) Timmins, P. A.; Leonhard, M.; Weltzien, H. U.; Wacker, T.; Welte, W., A Physical Characterization of Some Detergents of Potential Use for Membrane-Protein Crystallization. *FEBS Lett.* **1988,** *238* (2), 361-368. DOI: 10.1016/0014-5793(88)80513-1.
(5) Dupuy, C.; Auvray, X.; Petipas, C., Anomeric effects on the structure of micelles of alkyl maltosides in water. *Langmuir* **1997,** *13* (15), 3965-3967. DOI: 10.1021/la9604285.
(6) Weisstein, E. W. "Oblate Spheroid". From *MathWorld*--A Wolfram Web Resource. http://mathworld.wolfram.com/OblateSpheroid.html (accessed June 9, 2017).