

Supporting Information for:

Effect of Equilibration Time on the Structural Gradient in the Vertical Direction for
Bicontinuous Microemulsions in Winsor-III and -IV Systems

Douglas G. Hayes^{*,1}, Brian A. Barth² and S. Venkatesh Pingali^{*,3}

¹ Department of Biosystems Engineering and Soil Science, University of Tennessee, 2506 E.J.
Chapman Drive, Knoxville, TN 37996-4531, dhayes1@utk.edu, 1-865-974-7991

² Department of Chemical and Biomolecular Engineering, University of Tennessee, 1512 Middle
Dr, Knoxville, TN 37996, bbarth@vols.utk.edu

³ Oak Ridge National Laboratory, 1 Bethel Valley Road, Oak Ridge, TN 37831,
pingalis@ornl.gov

1 April 2024

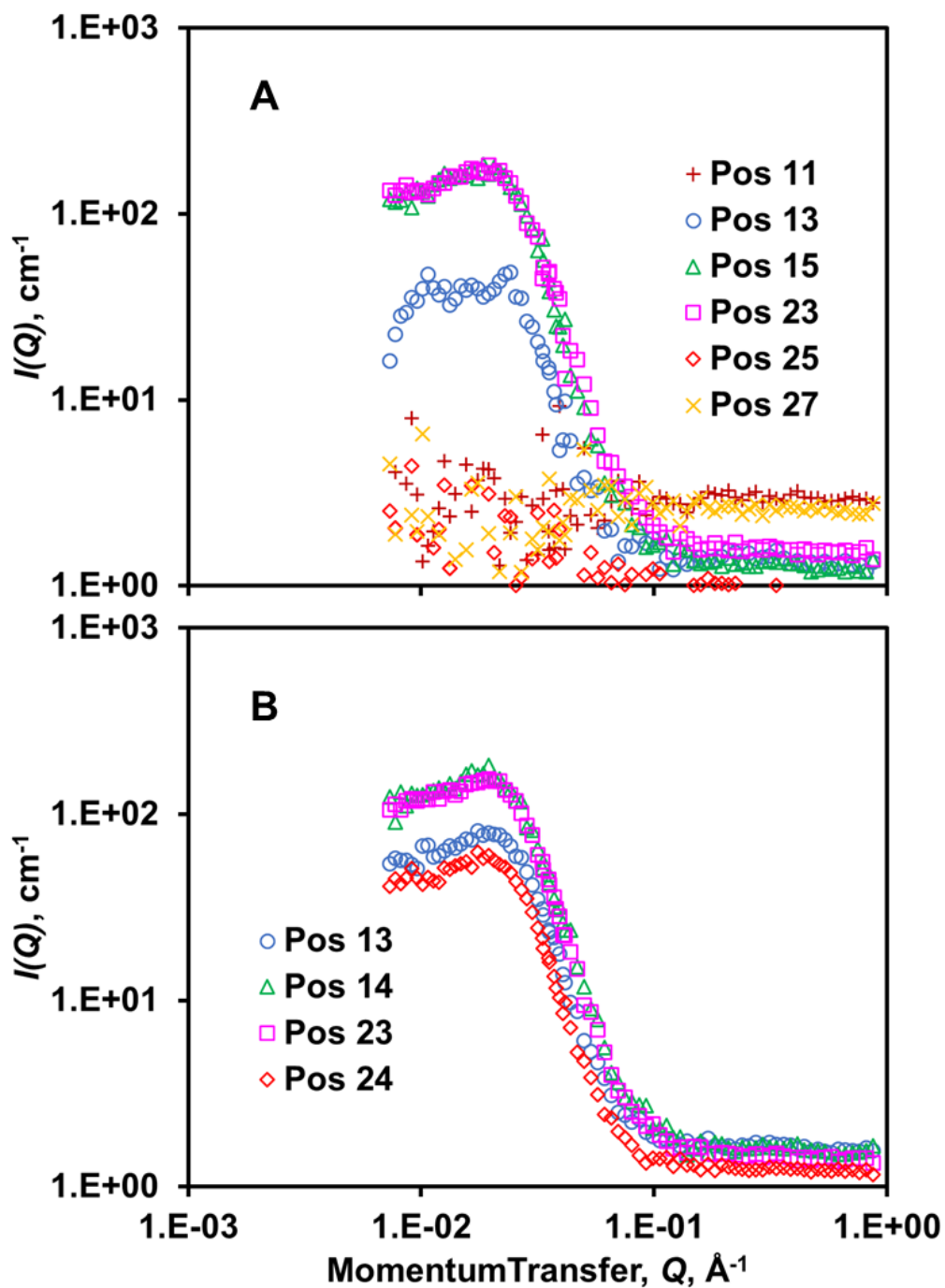


Figure S1. SANS data (Q vs. $I(Q)$) for vertical stage positions 9-15 and 23-27, showing that the bicontinuous phase is located within positions 14 mm to 23 mm, near the top and bottom liquid-liquid interfaces, respectively. Equilibration times: **A** 10 min, cycle 1 (0.5 h) and **B** 1 day, cycle 1 (24.2 h).

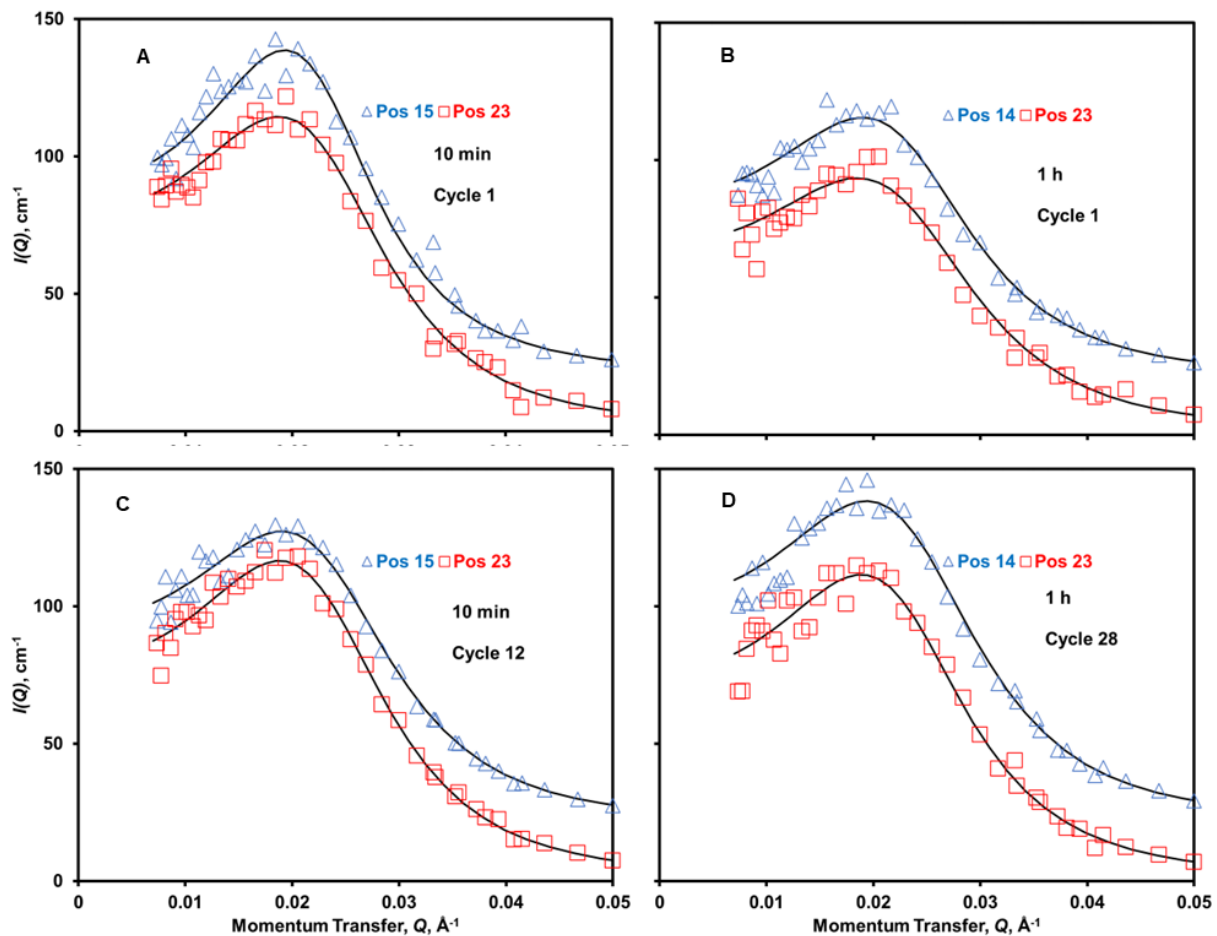


Figure S2. Teubner-Strey (TS) model fit to SANS data (Q vs. $I(Q)$) for vertical stage positions 14 mm or 15 mm (near the upper liquid-liquid interface) and 23 mm (near the lower interface) and the indicated equilibration time (10 min or 1 h) and cycle number. A constant, 20 cm^{-1} , was added to $I(Q)$ for position 14 mm or 15 mm to increase clarity. Total times for 10 min equilibration: cycle 1 = 0.5 h, cycle 12 = 1.6 h; m 1 h equilibration: cycle 1 = 1.1 h, cycle 28 = 10.1 h

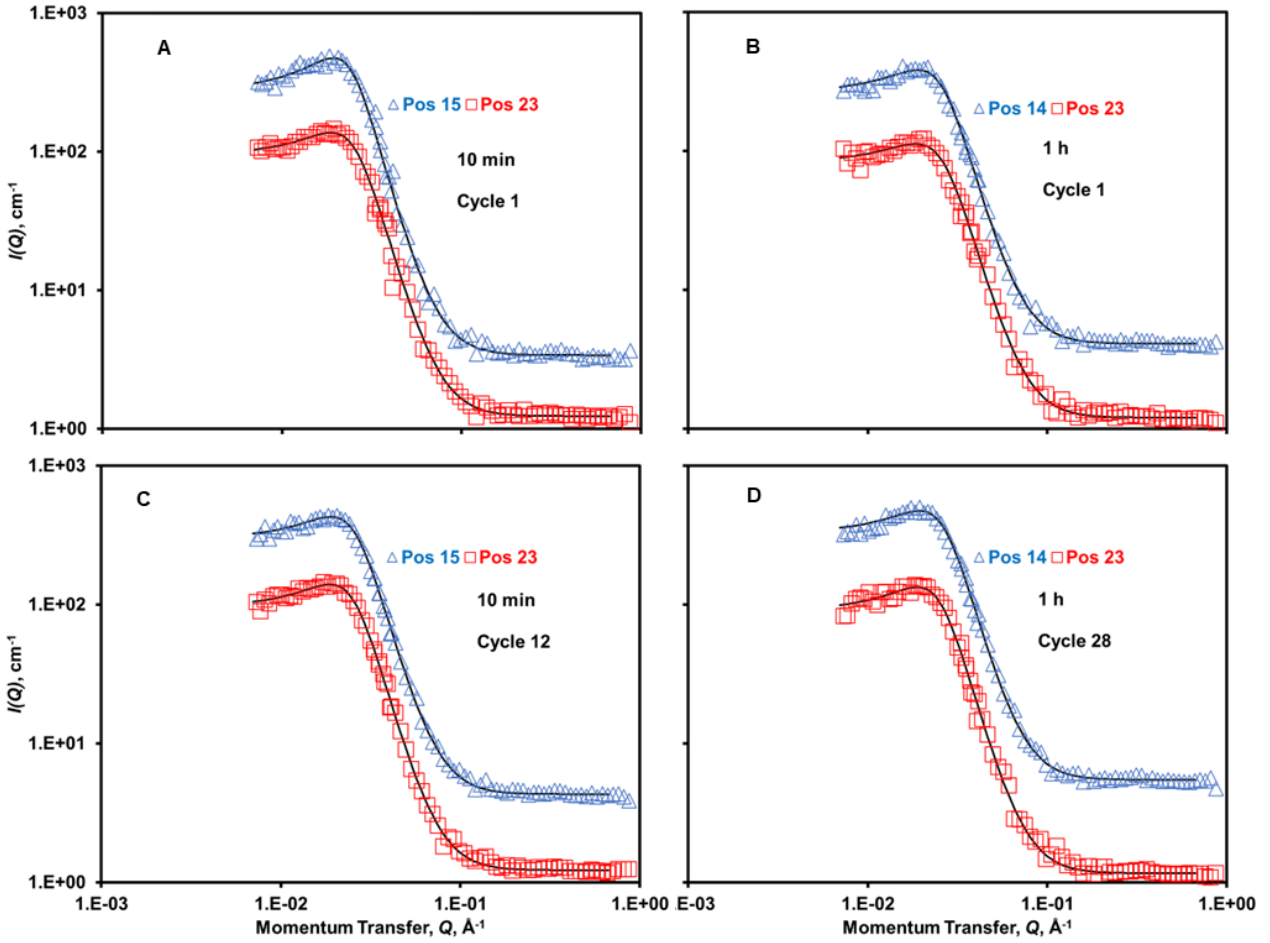


Figure S3. Teubner-Strey (TS) model fit to SANS data ($\log Q$ vs. $\log I(Q)$, log-log coordinates) for vertical stage positions 14 mm or 15 mm (near the upper liquid-liquid interface) and 23 mm (near the lower interface) and the indicated equilibration time (10 min or 1 h) and cycle number. A constant, 4 and 1.2, was multiplied to $I(Q)$ for position 14 mm or 15 mm and 23 mm, respectively, to increase clarity. This is a replotting of Figure S2 data on log-log coordinates. Total times for 10 min equilibration: cycle 1 = 0.5 h, cycle 12 = 1.6 h; 1 h equilibration: cycle 1 = 1.1 h, cycle 28 = 10.1 h

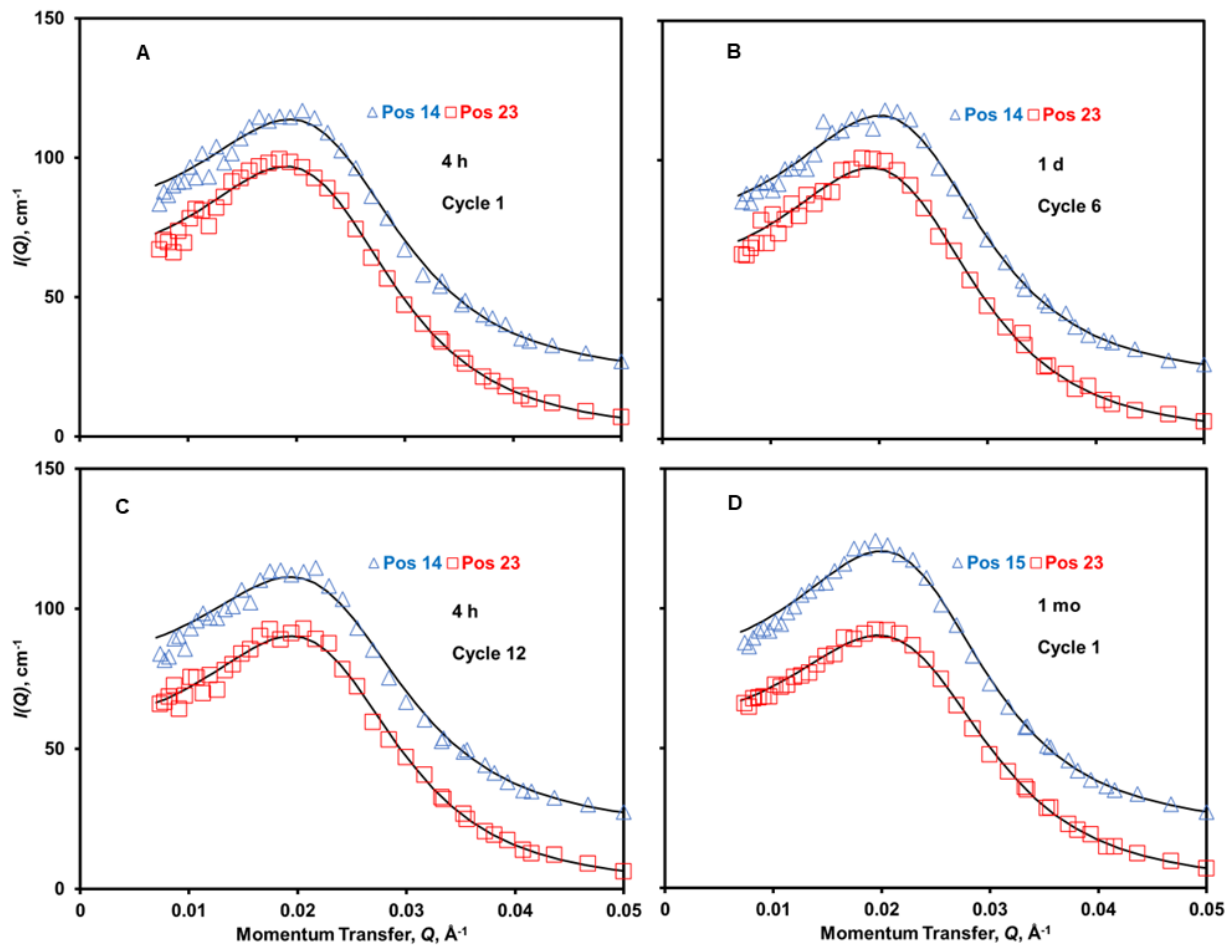


Figure S4. Teubner-Strey (TS) model fit to SANS data (Q vs. $I(Q)$) for vertical stage positions 14 mm (near the upper liquid-liquid interface) and 23 mm (near the lower interface) and the indicated equilibration time (4 h, 1 d, or 1 mo) and cycle number. A constant, 20 cm^{-1} , was added to $I(Q)$ for position 14 mm to increase clarity. Total times for 4 h equilibration: cycle 1 = 4.4 h, cycle 12 = 15.1 h; 1 d: cycle 6 = 28.8 h; 1 mo = 745 h.

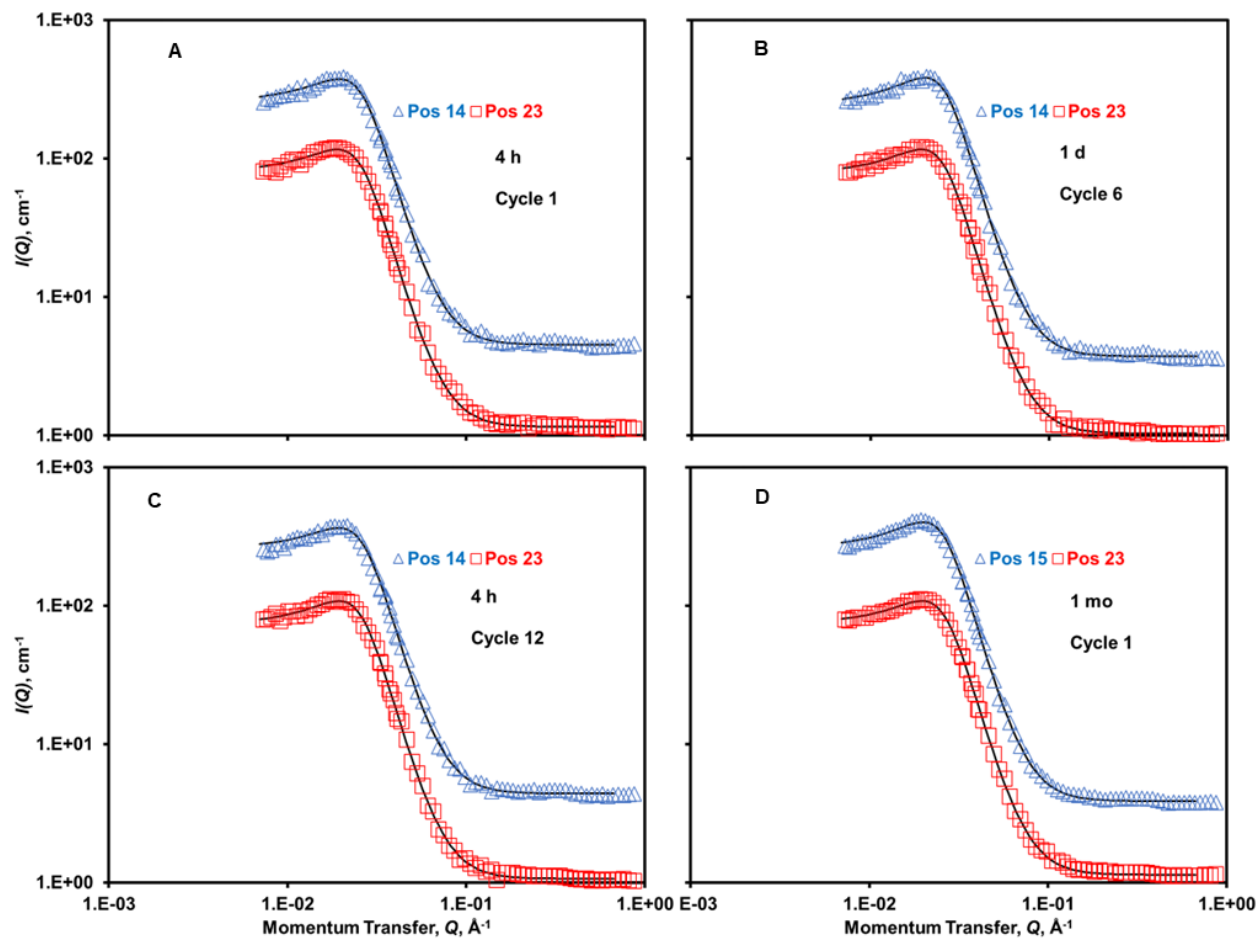


Figure S5. Teubner-Strey (TS) model fit to SANS data ($\log Q$ vs. $\log I(Q)$, log-log coordinates) for vertical stage positions 14 mm (near the upper liquid-liquid interface) and 23 mm (near the lower interface) and the indicated equilibration time (4 h, 1 d, or 1 mo) and cycle number. A constant, 4 and 1.2, was multiplied to $I(Q)$ for position 14 mm and 23 mm, respectively, to increase clarity. This is a replotting of Figure S4 data on log-log coordinates. Total times for 4 h equilibration: cycle 1 = 4.4 h, cycle 12 = 15.1 h; 1 d: cycle 6 = 28.8 h; 1 mo: 745 h.

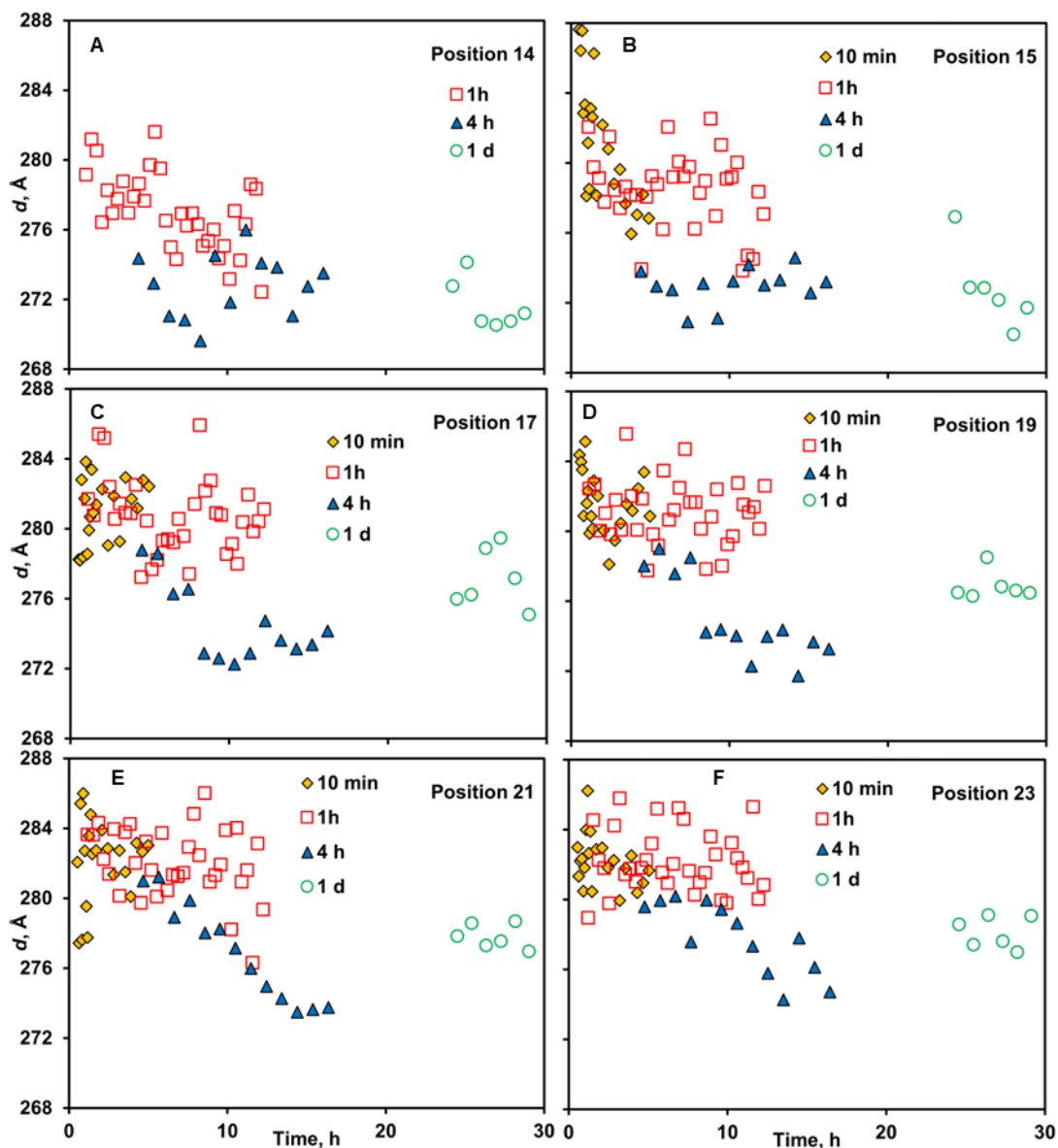


Figure S6. Effect of equilibration (legend) and relaxation times on the quasi-periodic repeat distance, d , for the middle, bicontinuous phase, of W_{III} systems formed by the Water/SDS/1-pentanol/dodecane system at 22°C for different vertical stage positions. Abscissa represents the total (equilibration + relaxation) time. Positions 14 mm and 23 mm are near the upper and lower liquid-liquid interfaces, respectively.

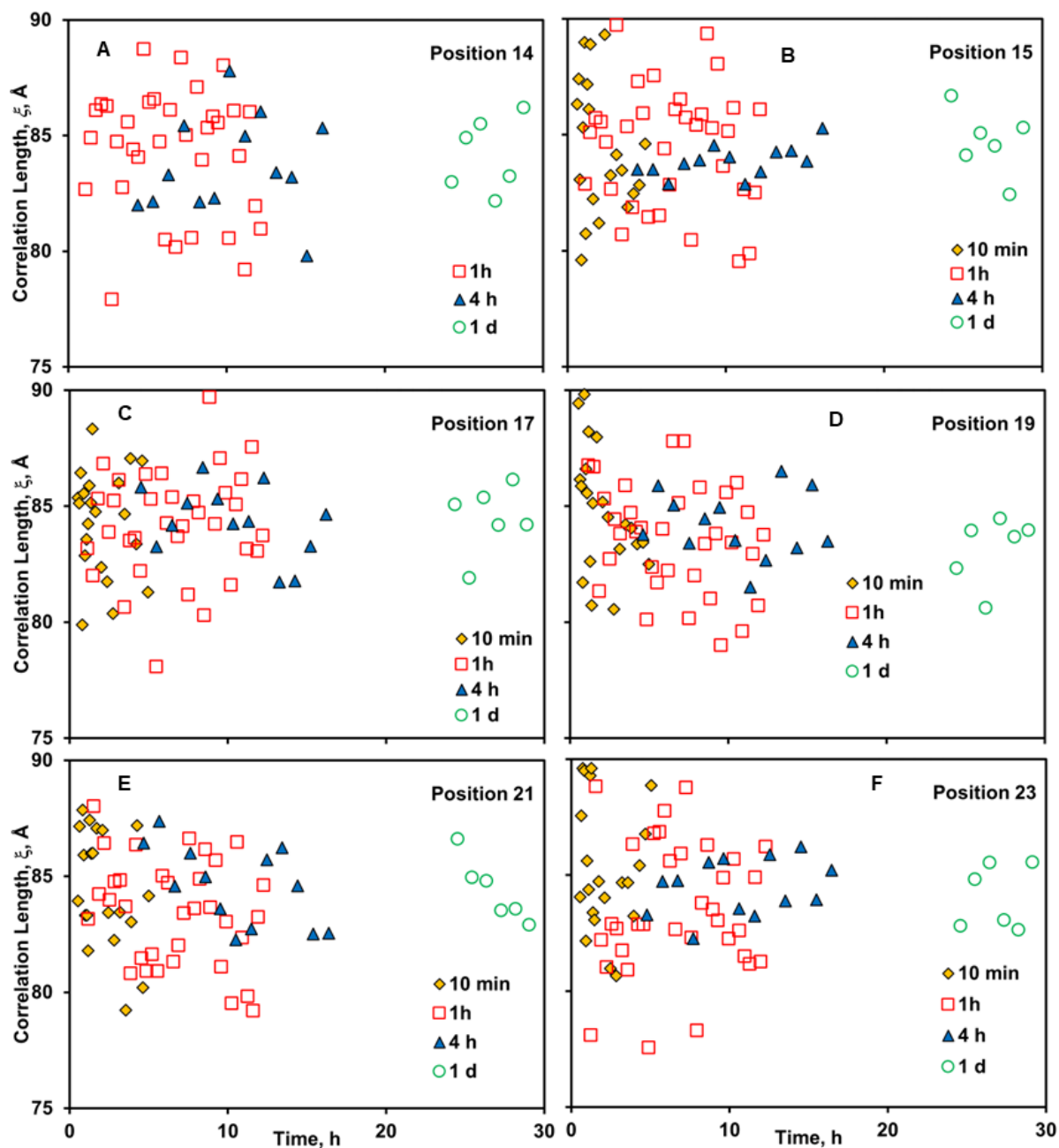


Figure S7. Effect of equilibration (legend) and relaxation times on the correlation coefficient, ξ , for the middle, bicontinuous phase, of W_{III} systems formed by the Water/SDS/1-pentanol/dodecane system at 22°C for different vertical stage positions. Abscissa represents the total (equilibration + relaxation) time. Positions 14 mm and 23 mm are near the upper and lower liquid-liquid interfaces, respectively.

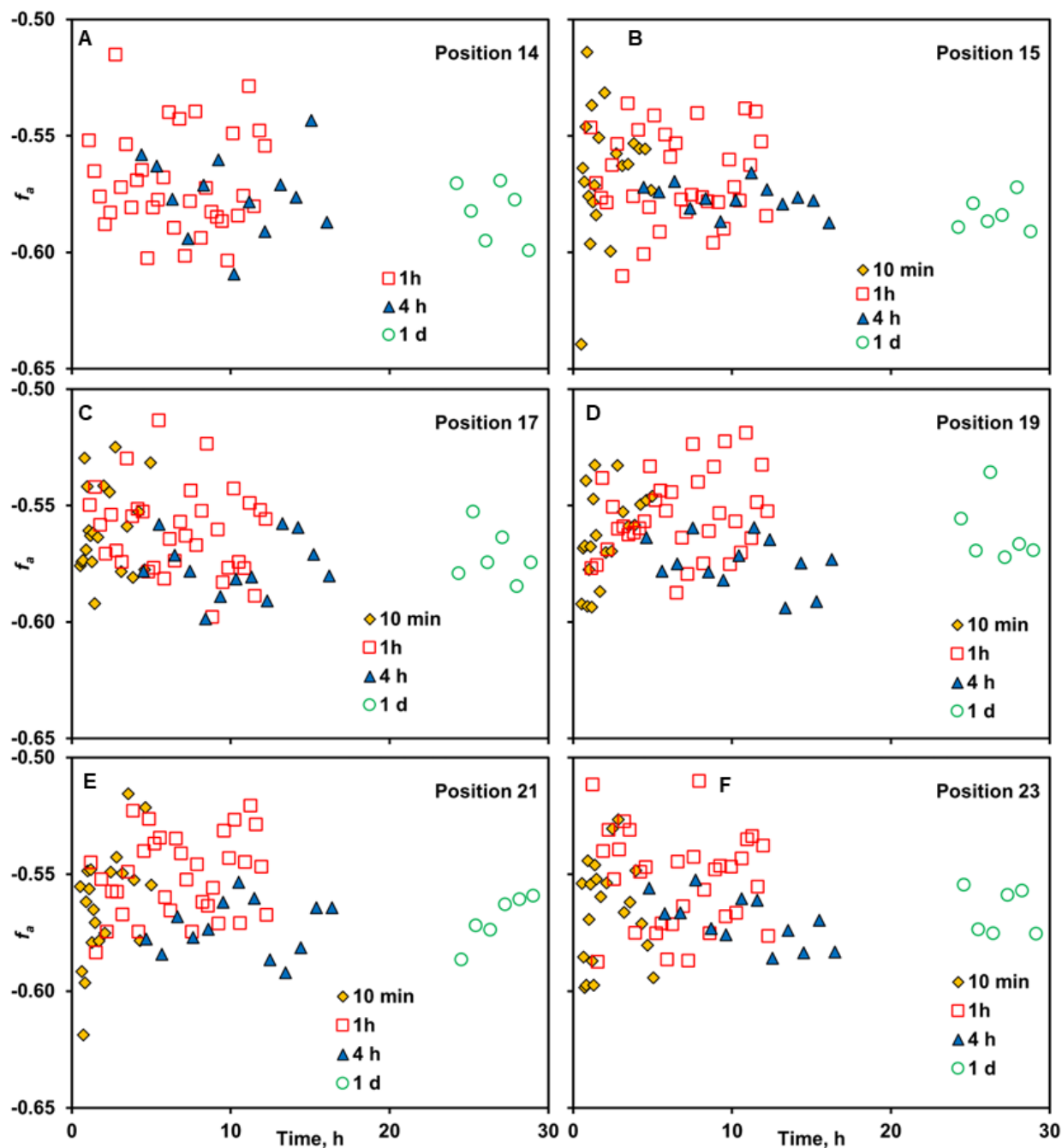


Figure S8. Effect of equilibration (legend) and relaxation times on the amphiphilicity factor, f_a for the middle, bicontinuous phase, of W_{III} systems formed by the Water/SDS/1-pentanol/dodecane system at 22°C for different vertical stage positions. Abscissa represents the total (equilibration + relaxation) time. Positions 14 mm and 23 mm are near the upper and lower liquid-liquid interfaces, respectively.

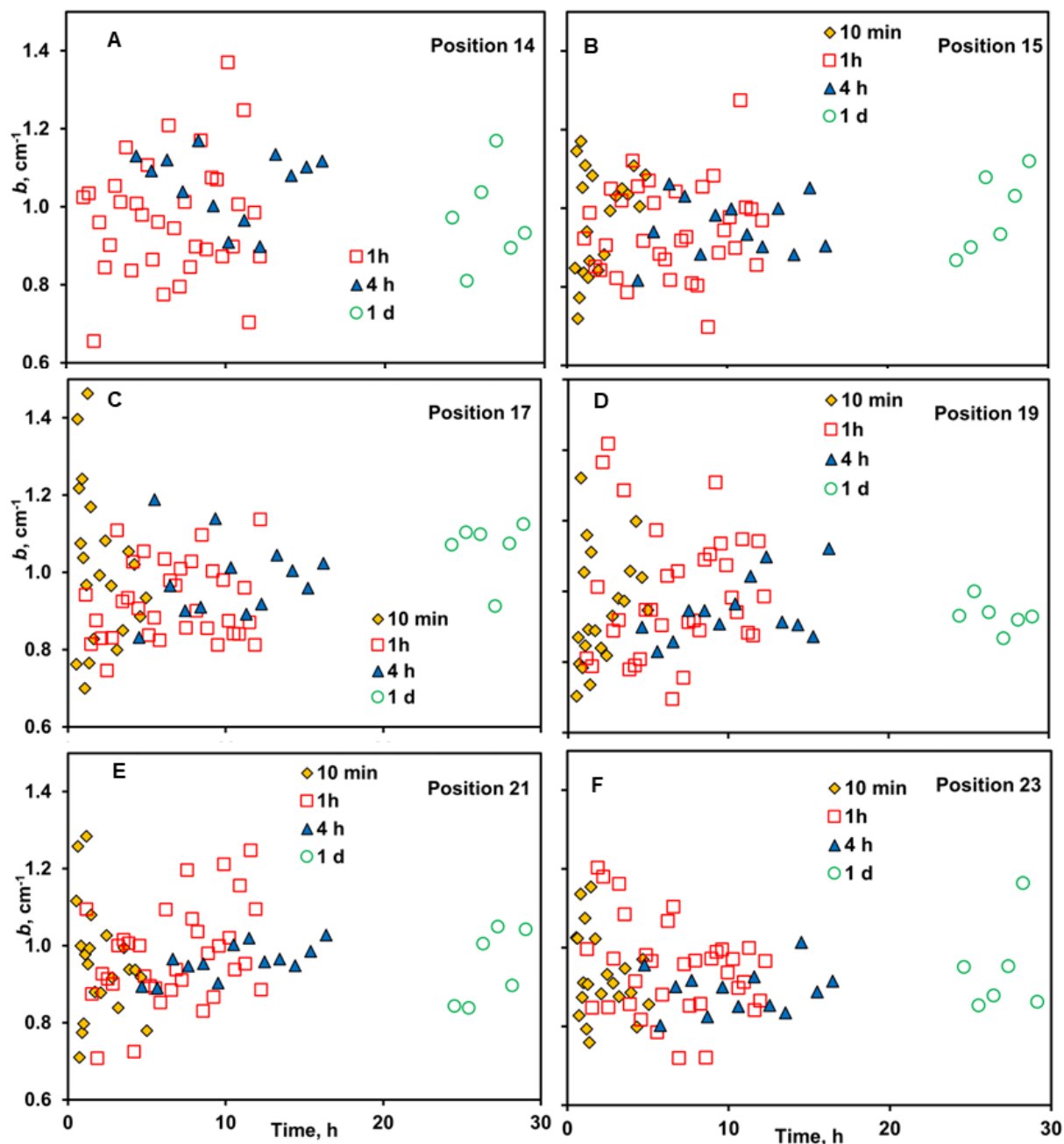


Figure S9. Effect of equilibration (legend) and relaxation times on the background, b , for the middle, bicontinuous phase, of W_{III} systems formed by the Water/SDS/1-pentanol/dodecane system at 22°C for different vertical stage positions. Abscissa represents the total (equilibration + relaxation) time. Positions 14 mm and 23 mm are near the upper and lower liquid-liquid interfaces, respectively.

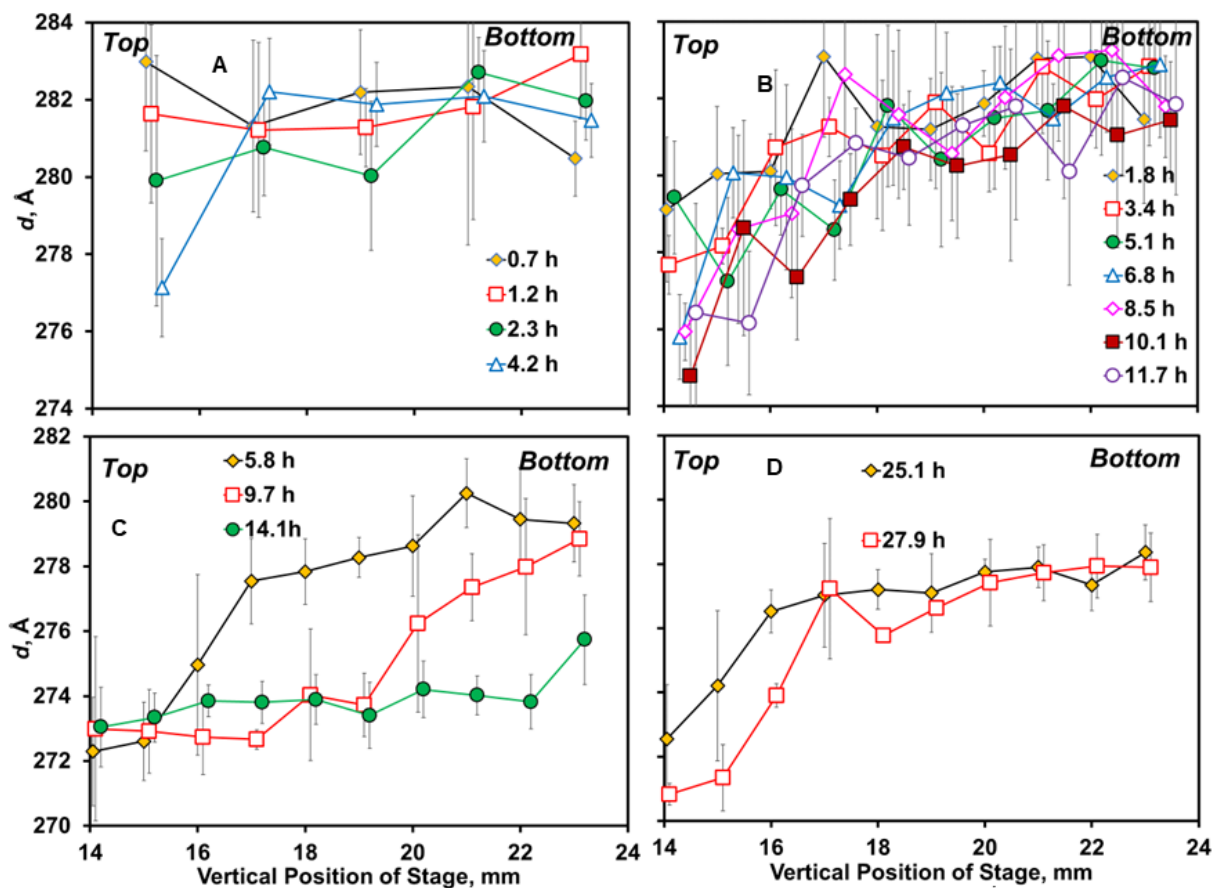


Figure S10. Effect of time for equilibration and relaxation on the change of the quasi-periodic repeat distance, d , for the middle, bicontinuous phase, of W_{III} systems formed by the Water/SDS/1-pentanol/dodecane system at 22°C vs. vertical stage position. Positions 14 mm and 23 mm are near the top and bottom liquid-liquid interfaces, respectively. Figs A-D represent equilibration times of 10 min, 1 h, 4 h, and 1 d, respectively, while legends represent total (equilibration + relaxation) time. Each data series represents the average of 5 consecutive relaxation cycles for 10 min and 1 h, 4 cycles for 4 h, and 3 cycles for 1 d. Replotting of Fig. 2, but with error bars (standard deviation of the means) provided. Vertical stage positions were increased by 0.1 mm to improve visualization.

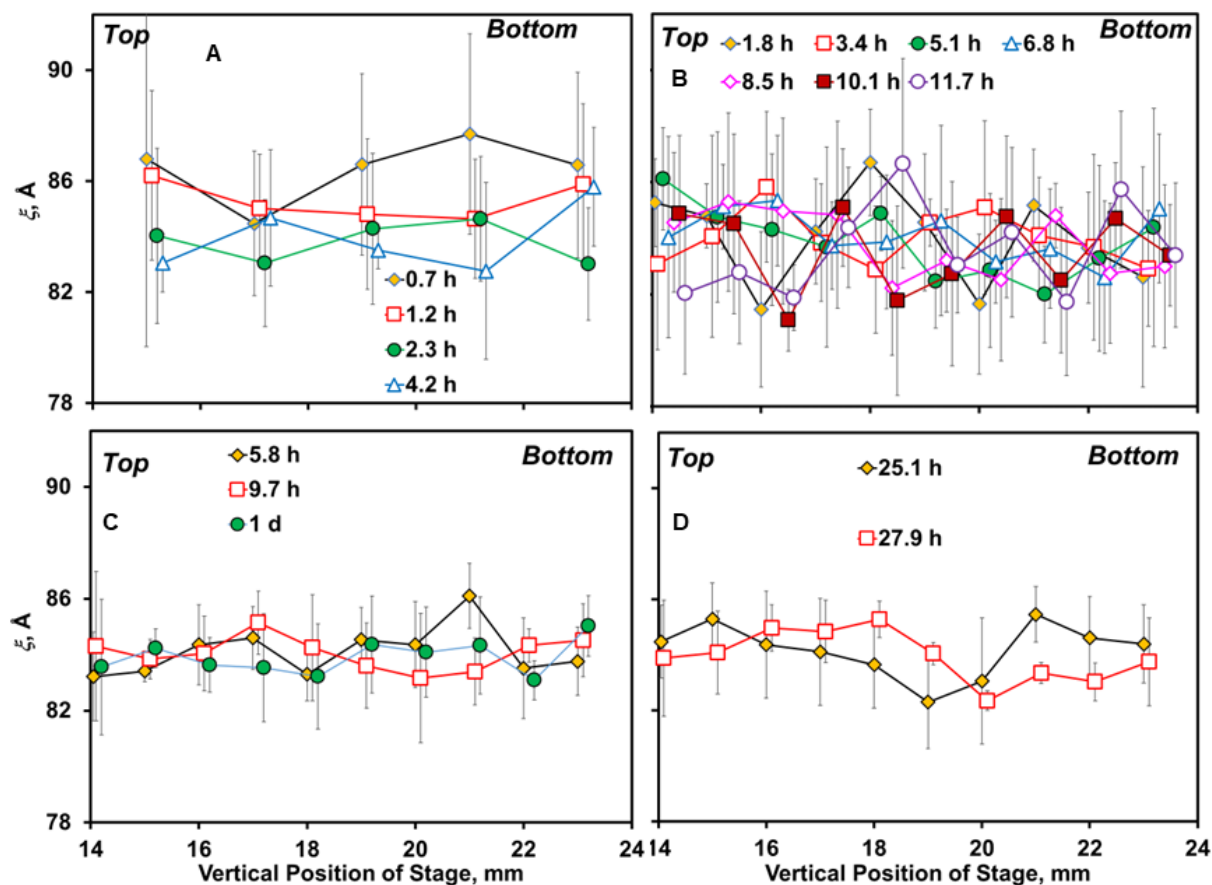


Figure S11. Effect of equilibration and relaxation times on the change of the correlation coefficient, ξ , for the middle, bicontinuous phase, of W_{III} systems formed by the Water/SDS/1-pentanol/dodecane system at 22°C vs. vertical stage position. Positions 14 mm and 23 mm are near the top and bottom liquid-liquid interfaces, respectively. Figs A-D represent equilibration times of 10 min, 1 h, 4 h, and 1 d, respectively, while legends represent total (equilibration + relaxation) time. Each data series represents the average of 5 consecutive relaxation cycles for 10 min and 1 h, 4 cycles for 4 h, and 3 cycles for 1 d. Replotting of Fig. 3, but with error bars (standard deviation of the means) provided. Vertical stage positions were increased by 0.1 mm to improve visualization.

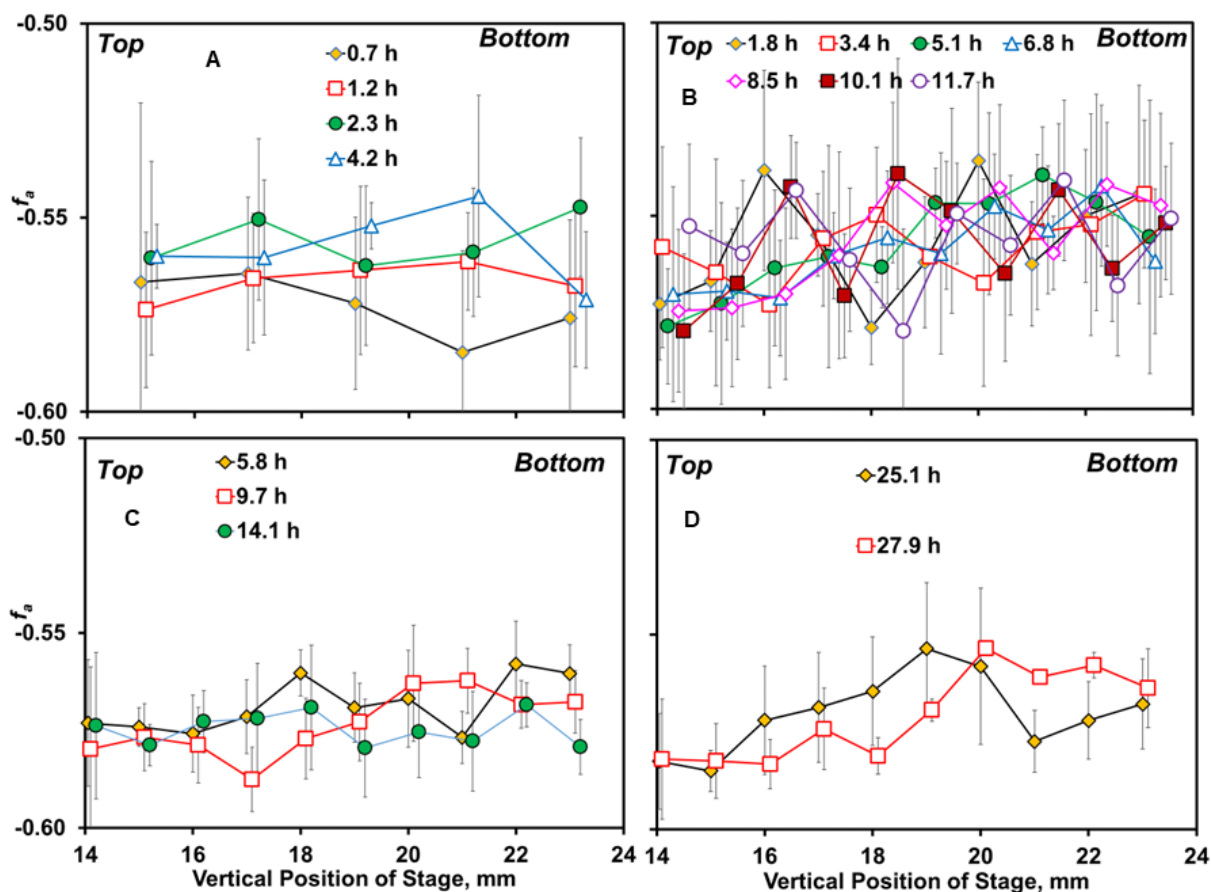


Figure S12. Effect of equilibration and relaxation times on the change of the amphiphilicity factor, f_a , for the middle, bicontinuous phase, of W_{III} systems formed by the Water/SDS/1-pentanol/dodecane system at 22°C vs. vertical stage position. Positions 14 mm and 23 mm are near the top and bottom liquid-liquid interfaces, respectively. Figs A-D represent equilibration times of 10 min, 1 h, 4 h, and 1 d, respectively, while legends represent total (equilibration + relaxation) time. Each data series represents the average of 5 consecutive relaxation cycles for 10 min and 1 h, 4 cycles for 4 h, and 3 cycles for 1 d. Replotting of Fig. 4, but with error bars (standard deviation of the means) provided. Vertical stage positions were increased by 0.1 mm to improve visualization.

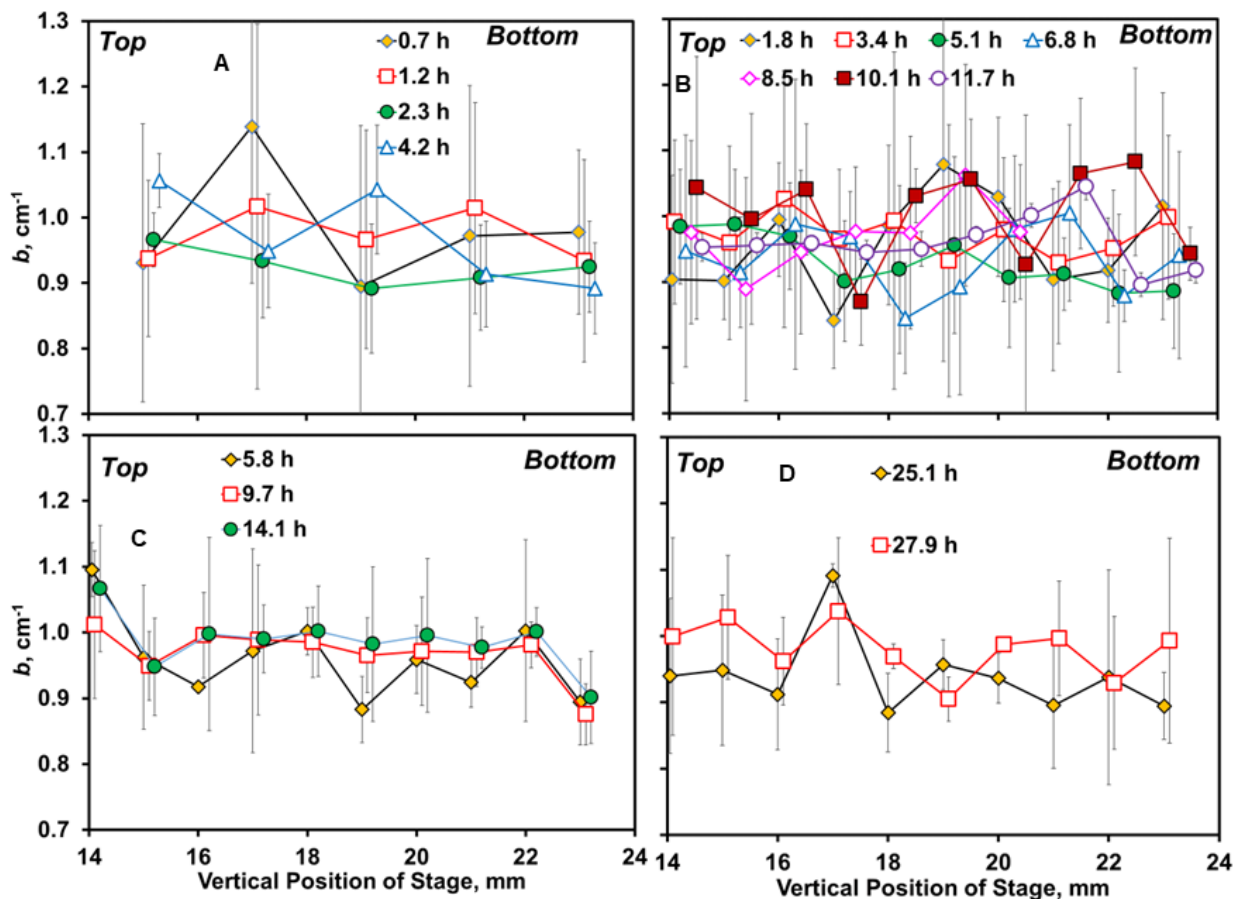


Figure S13. Effect of equilibration and relaxation times on the change of the background, b , for the middle, bicontinuous phase, of W_{III} systems formed by the Water/SDS/1-pentanol/dodecane system at 22°C vs. vertical stage position. Positions 14 mm and 23 mm are near the top and bottom liquid-liquid interfaces, respectively. Figs A-D represent equilibration times of 10 min, 1 h, 4 h, and 1 d, respectively, while legends represent total (equilibration + relaxation) time. Each data series represents the average of 5 consecutive relaxation cycles for 10 min and 1 h, 4 cycles for 4 h, and 3 cycles for 1 d. Replotting of Fig. 5, but with error bars (standard deviation of the means) provided. Vertical stage positions were increased by 0.1 mm to improve visualization.