Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Temperature-Responsive Binary Superlattices Prepared by Selective Solvent Evaporation of O/W Microemulsion Composed of Gold Nanoparticles and Surfactants

Young-Jin Yoon [1,2], Jae-Min Ha [3], Hyuk-Jin Seo[1,2], Jong Dae Jang[4], Changwoo Do[5], Tae-Hwan Kim[1,2,6,7]*

[1]Department of Applied Plasma & Quantum Beam Engineering, Jeonbuk National University, Jeonju 54896, Republic of Korea

[2]Research Center for Advanced Nuclear Interdisciplinary Technology, Jeonbuk National University, Jeonju 54896, Republic of Korea

[3]Samsung Electronics, Hwaseong, Gyeonggi 18448, Republic of Korea

[4]Neutron Science Division, Korea Atomic Energy Research Institute, 1045 Daedeok-daero, Yuseong-gu, Daejeon 34057, Republic of Korea

[5]Neutron Scattering Division, Neutron Sciences Directorate, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, United States

[6]Department of Quantum System Engineering, Jeonbuk National University, Jeonju 54896, Republic of Korea

[7]High-Enthalphy Plasma Research Center, Jeonbuk National University, 546 Bongdong-ro, Bongdong-eup, Wanju-gun, Jeollabuk-do, 55317, Republic of Korea.

- **SAXS intensities of Brij58 surfactant at 35, 40 and 50 wt.% in water**

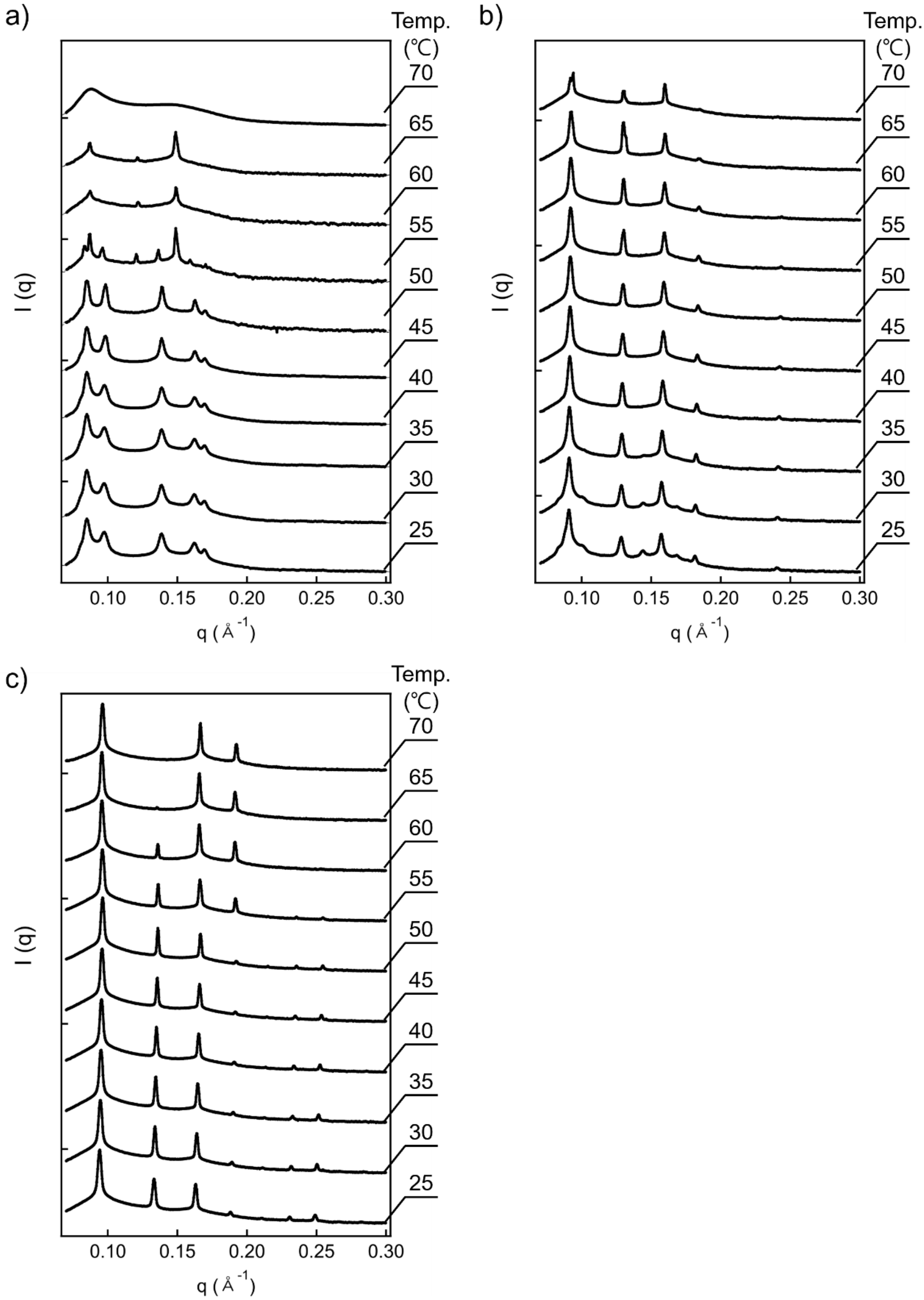

Fig. S1. SAXS intensities of Brij58 in aqueous solution at a different mass fraction of a) 35, b) 40, and c) 50 wt. %. All SAXS intensities are shifted vertically for visual clarity.

- **Transmission Electron Microscopy (TEM) image of the Birj58/AuNP4.1/water after sonication.**

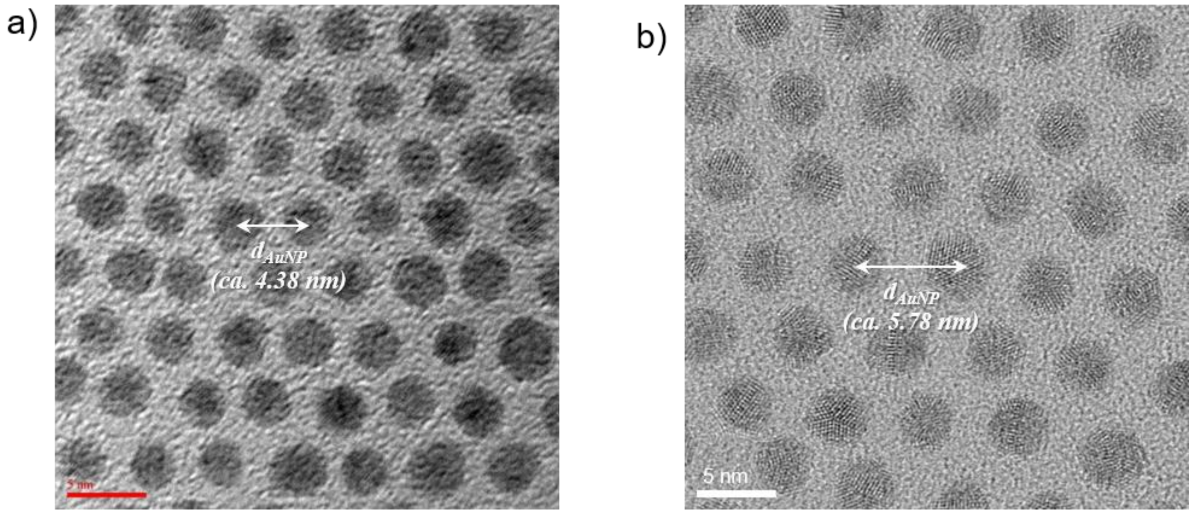

Fig. S2. (a) TEM image of AuNP4.1 only and (b) a Birj58/AuNP4.1/water complex before secondary evaporation (where the sample for TEM measurement was collected by centrifugation in pallet form and then redispersed in water). The center-to-center distances of the AuNPs (d_{AuNP}) in the AuNP4.1 and the Birj58/AuNP4.1/water complex were estimated to be 4.38 nm and 5.78 nm, respectively, indicating the increase of d_{AuNP} upon the successful encapsulation of AuNP surface.

- **SAXS intensities of the Brij58/AuNP4.1/water complexes at the different concentrations.**

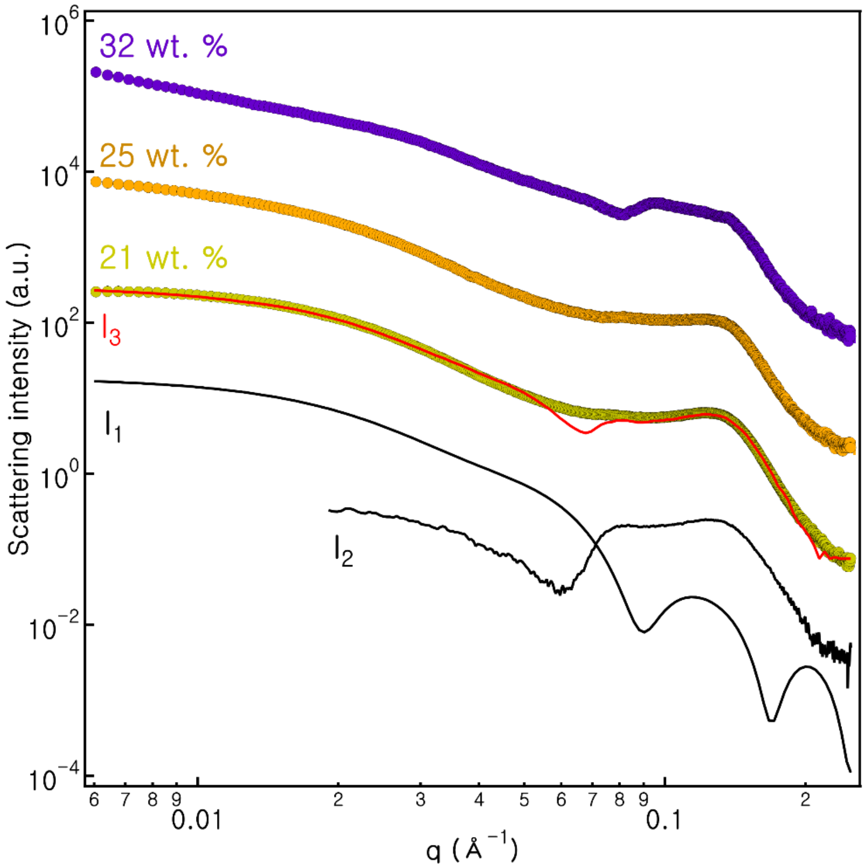

Fig. S3. SAXS intensities of the Brij58/AuNP4.1/water complexes at the different concentrations of 21 (yellow), 25 (orange), and 32 (violet) wt. % and the Brij58 at 20 wt. % (I_2) in water. I_1 is the simulated intensity for the core-shell particle consisted of the core of 4 nm and the shell thickness of 7.6 nm (which describes the surfactant-encapsulated AuNPs), and I_3 is a sum of the I_1 and I_2. Herein, we did not consider the interparticle interference between the surfactant micelle to core-shell AuNP because it could not be independently measured in the complex system. The SAXS intensities of the Brij58/AuNP4.1/water complexes (21 wt. %) was successfully reproduced by a sum of the I_1 and I_2, indicating that the complex is a mixture of the surfactant micelles and the surfactant-encapsulated AuNPs. Since all the SAXS intensities are not quite different, we expect that the structure of the Brij58/AuNP4.1/water complexes still remains even though the concentration increases.

- **SAXS intensities of the Brij58/AuNP2.1/water complexes at the different m_p**

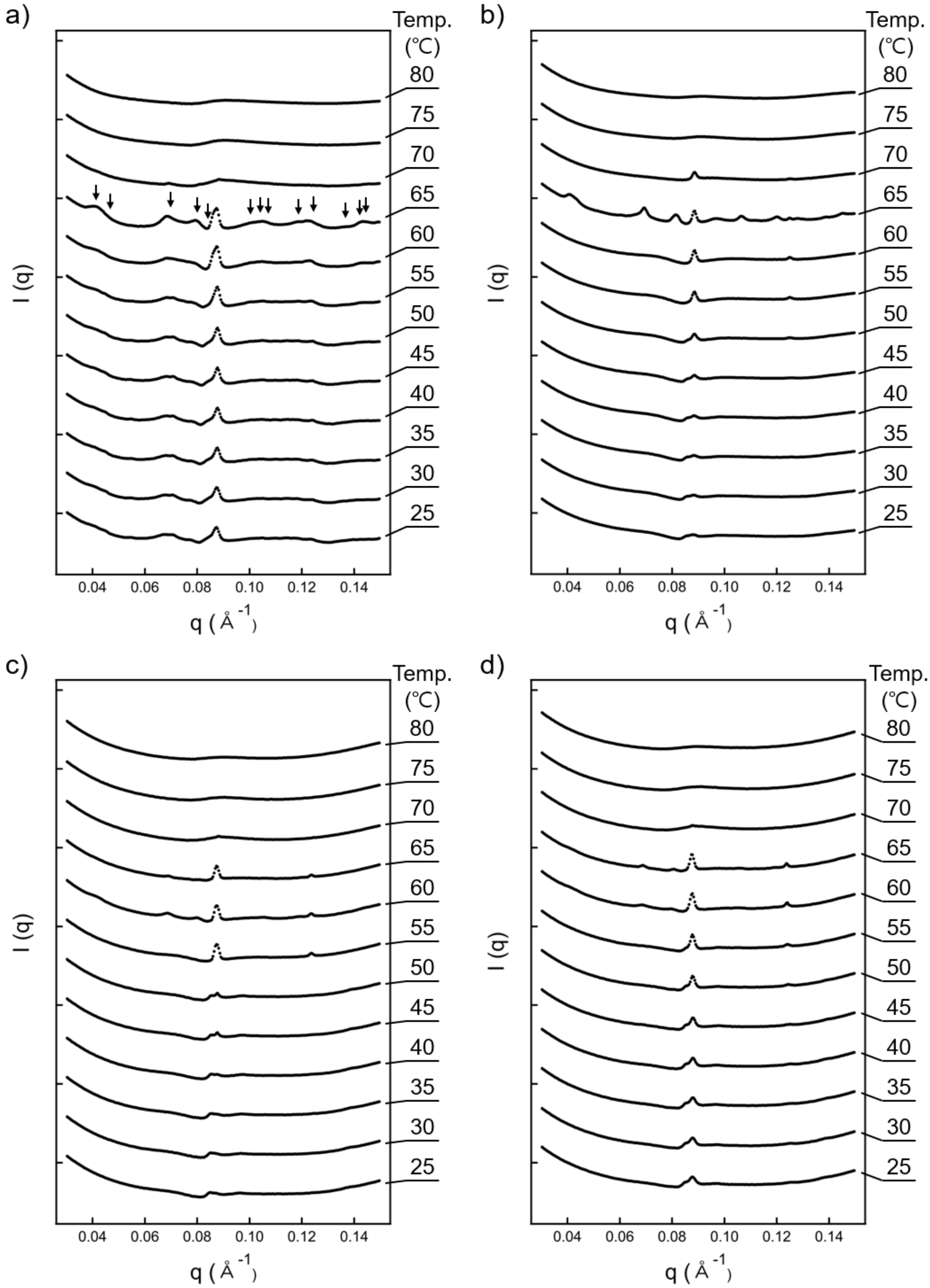

Fig. S4. SAXS intensities of the Brij58/AuNP2.1/water complexes at the different m_p of a) 0.6, b) 1.3, c) 1.9, and d) 3.1. All SAXS intensities are shifted vertically for visual clarity. The black arrows indicate the Bragg reflection peaks corresponding to Li_3Bi-type BNSLs structure.

- **SAXS intensities of the Brij58/AuNP4.1/water complexes at the different m_p**

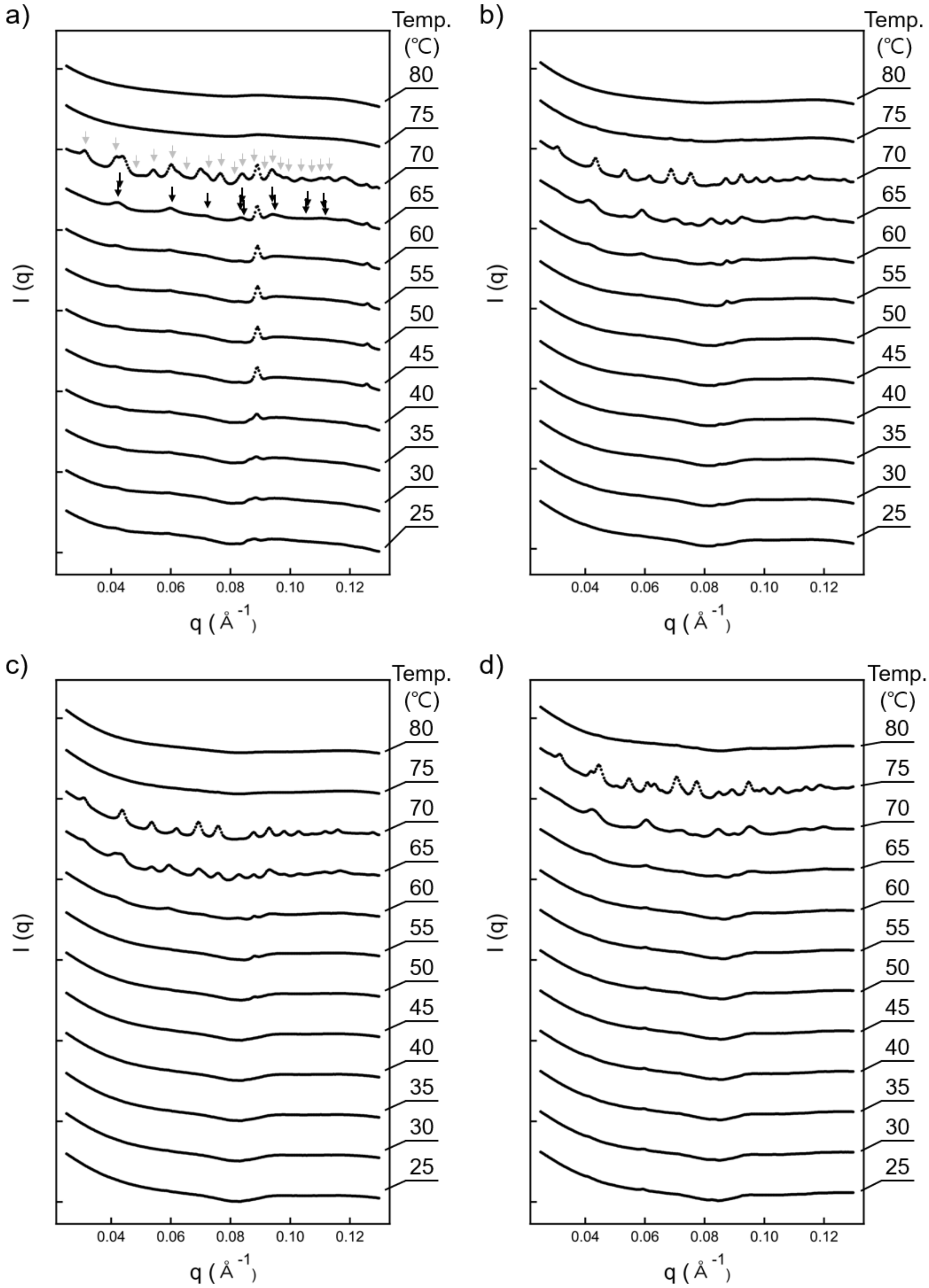

Fig. S5. SAXS intensities of the Brij58/AuNP4.1/water complexes at the different m_p of a) 1.3, b) 1.9, c) 3.1, and d) 5.0. All SAXS intensities are shifted vertically for visual clarity. The black and gray arrows indicate the Bragg reflection peaks corresponding to AlB_2- and $NaZn_{13}$-type BNSLs structure, respectively.

- **SAXS intensities of the Brij58/AuN4.6/water complexes at the different m_p**

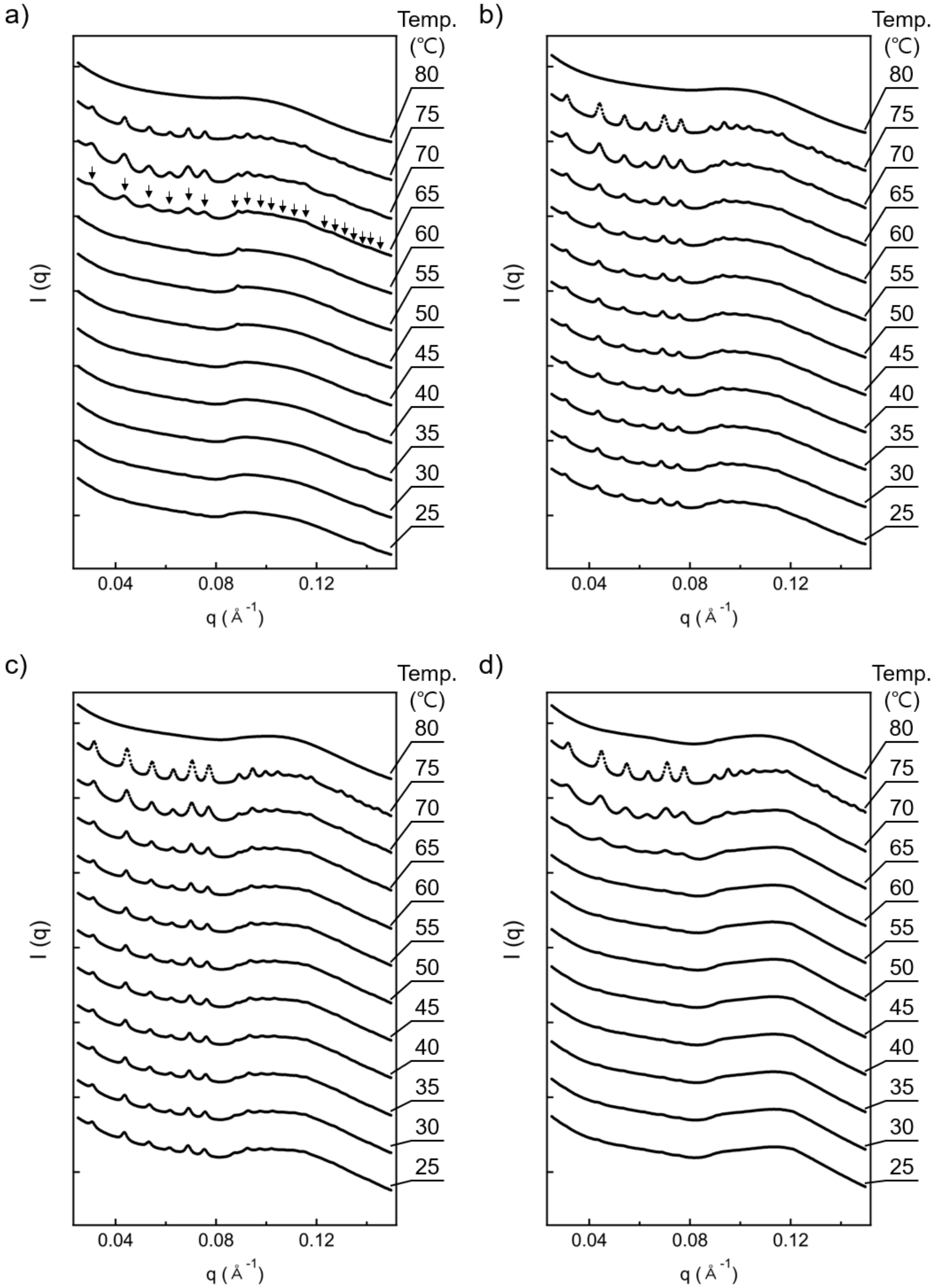

Fig. S6. SAXS intensities of the Brij58/AuNP4.6/water complexes at the different m_p of a) 1.9, b) 3.1, c) 5.0, and d) 7.5. All SAXS intensities are shifted vertically for visual clarity. The black arrows indicate the Bragg reflection peaks corresponding to $NaZn_{13}$-type BNSLs structure.

- **Lattice parameters obtained from SAXS analysis of Brij58 surfactant**

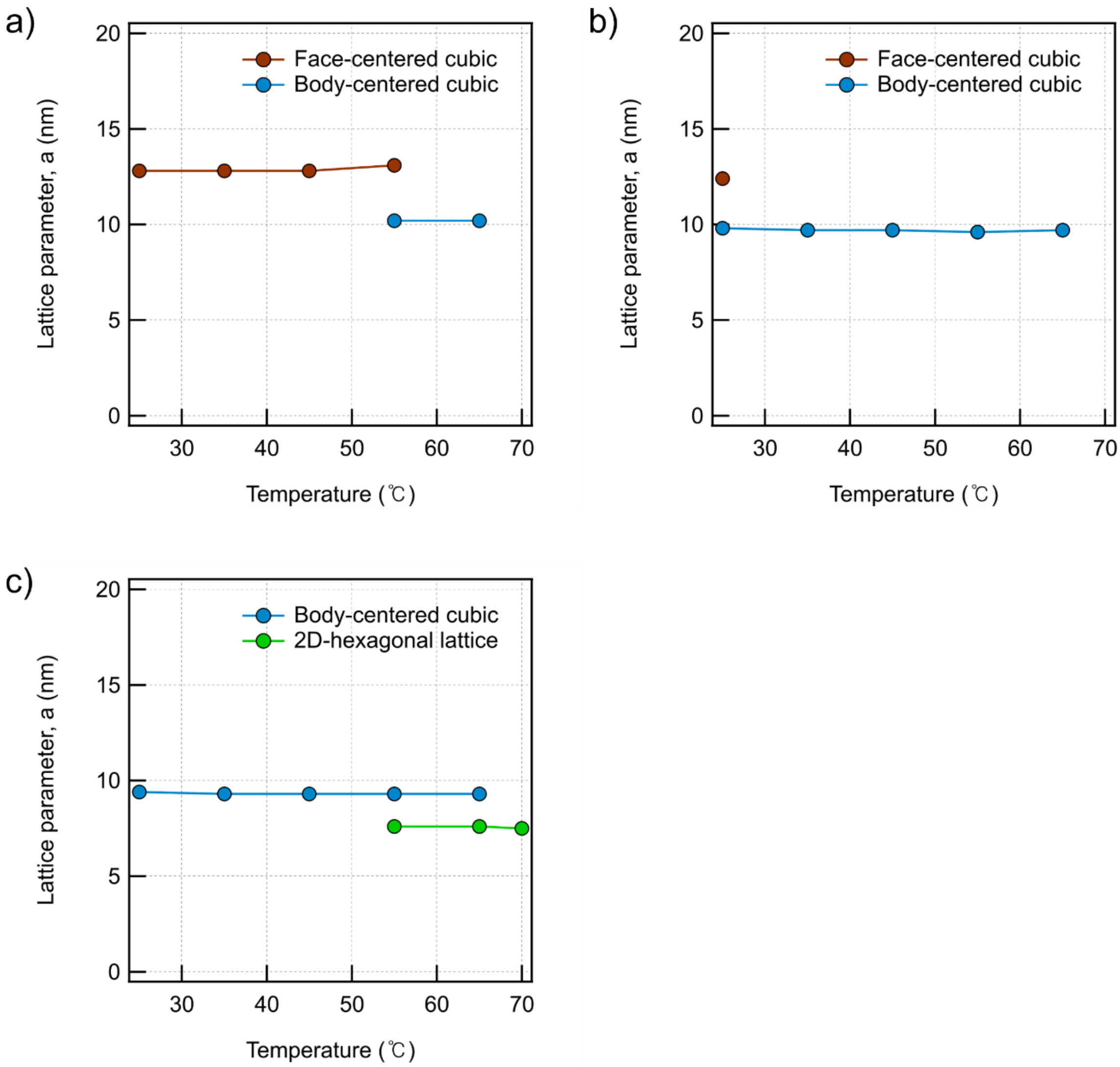

Fig. S7. Lattice parameters of a) Brij58 35 wt.%, b) Brij58 40 wt.%, and c) Brij58 50 wt.% in aqeuous solution (where lattice parameters were calculated by the aquation of $a = \frac{2\pi}{q_{111}} \times \sqrt{3}$, $\frac{2\pi}{q_{110}} \times \sqrt{2}$, and $\frac{2\pi}{q_{100}} \times \frac{2}{\sqrt{3}}$ for the face-centered cubic, the body-centered cubic, and the 2D-hexagonal lattice, respectively).

- **Lattice parameters obtained from SAXS analysis of Brij58/AuNP/water complexes**

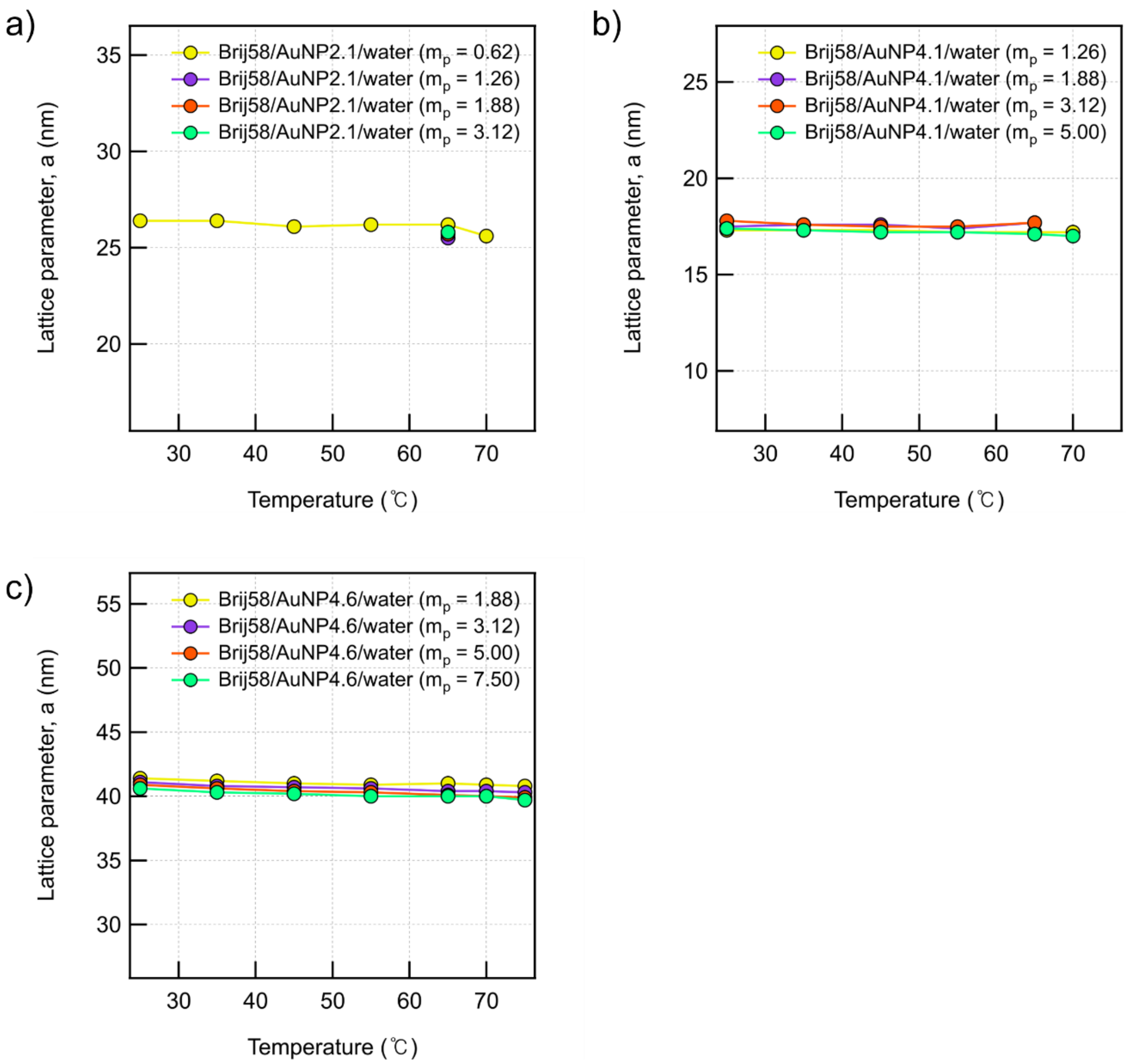

Fig. S8. Lattice parameter of the a) Brij58/AuNP2.1/water, b) Brij58/AuNP4.1/water, and c) Brij58/AuNP4.6/water complexes obtained from SAXS analysis. The lattice parameters were calculated by the aquation of $a = \frac{2\pi}{q_{111}} \times \sqrt{3}, = \frac{2\pi}{q_{100}} \times \frac{2}{\sqrt{3}},$ and $= \frac{2\pi}{q_{200}} \times 2$ for the Li_3Bi-type, the AlB_2-type, and the $NaZn_{13}$-type structures, respectively.

- **Simulation of structure factor for FCC-based BNSLs**

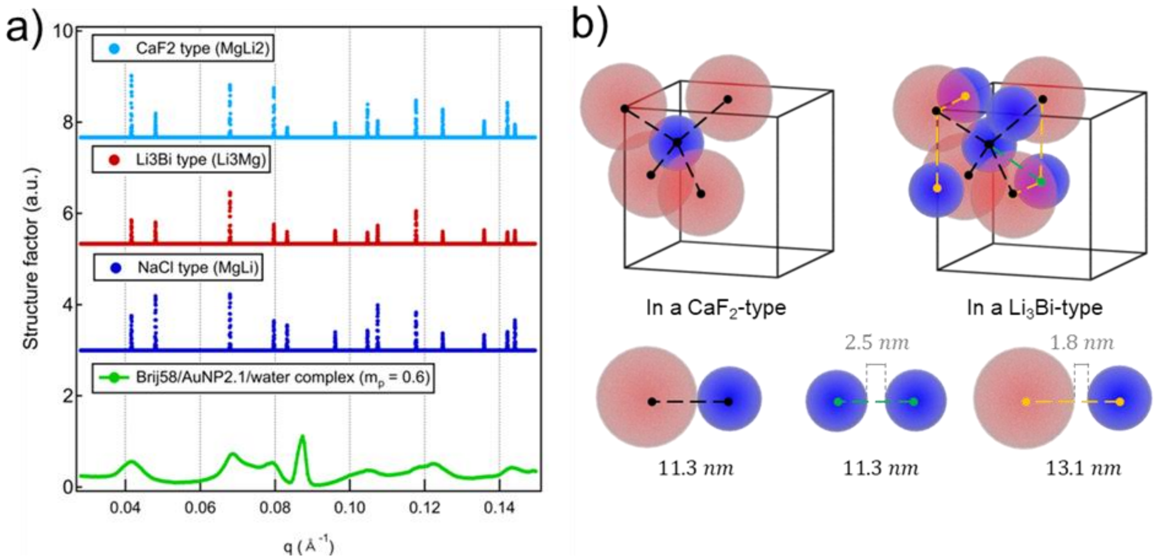

Fig. S9. a) The calculation of structure factors for the CaF_2- and Li_3Bi-type structures. b) Consideration of structural stability of Ca_F2- and Li_3Bi-type structures.

- **Calculation of packing efficiency of AlB_2-, CaF_2-, Li_3Bi-, $NaZn_{13}$-, and ZnS-type BNSLs**

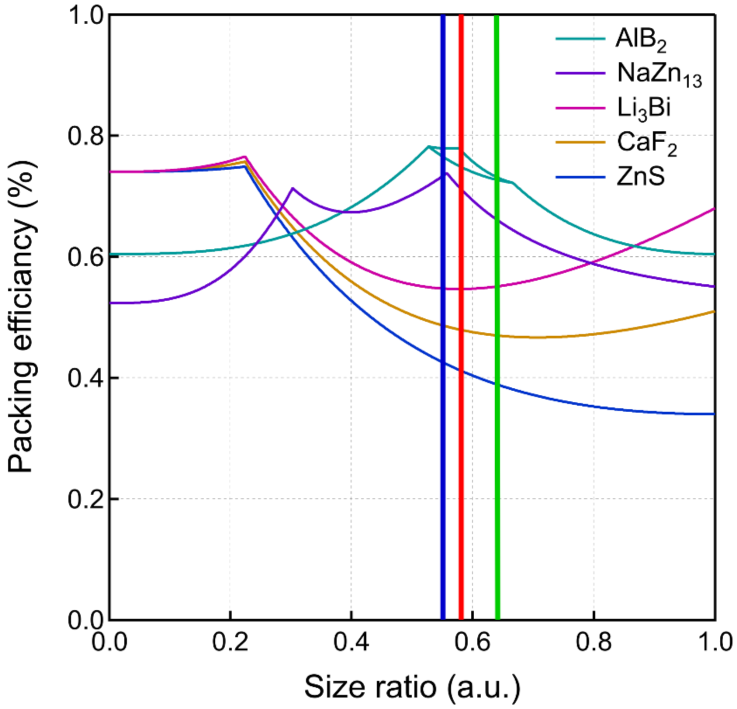

Fig. S10. Packing efficiancy of the AlB_2-, $NaZn_{13}$-, Li_3Bi-, CaF_2-, and ZnS-type BNSLs, where vertical lines indicate a size rato of 0.55, 0.58, and 0.64 for blue, red and green, respectively.

Table S1. Relative ratio of AuNPs (Brij58/AuNP/water = 20/ m_p /80 by weight) before evaporation and the calculated number ratio of two particles (surfactant micelles (n_{mic}):encapsulated AuNPs (n_{AuNP})).

Label	m_p	$n_{mic} : n_{AuNP}$
Brij58/AuNP2.1/water complex	0.6	20 : 1
	1.3	9 : 1
	1.9	5 : 1
	3.1	3 : 1
Brij58/AuNP4.1/water complex	1.3	79 : 1
	1.9	52 : 1
	3.1	30 : 1
	5.0	17 : 1
Brij58/AuNP4.6/water complex	1.9	74 : 1
	3.1	43 : 1
	5.0	26 : 1
	7.5	16 : 1