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Free Energy Change of Micelle Formation for Sodium Dodecyl Sulfate from a Dispersed State in Solution to Complete Micelles along Its Aggregation Pathways Evaluated by Chemical Species Model Combined with Molecular Dynamics Calculations

YOSHII Noriyuki [1,2,*], KOMORI Mika [2], KAWADA Shinji [2], TAKABAYASHI Hiroaki [2], FUJIMOTO Kazushi [2], OKAZAKI Susumu [1,2,*]

[1] Center for Computational Science, Graduate School of Engineering, Nagoya University, Nagoya 464-8603, Japan.

[2] Department of Applied Chemistry, Nagoya University, Nagoya 464-8603, Japan.

[*]Corresponding authors. Email: yoshii@ccs.engg.nagoya-u.ac.jp; Tel./Fax: +81-52-788-6213 (N.Y.).

Email: okazaki@apchem.nagoya-u.ac.jp; Tel./Fax: +81-52-789-5829 (S.O.).

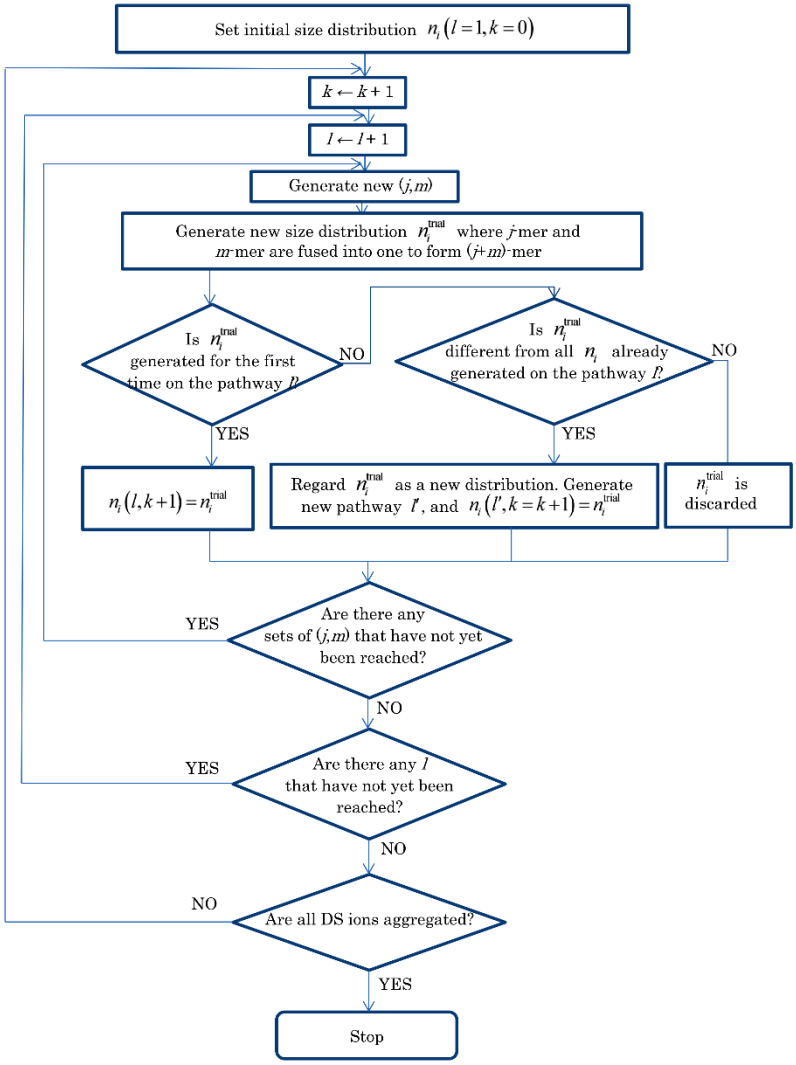

Fig. S1 Generation procedure of the aggregation paths shown by the flowchart.

S2 Molecular dynamics calculations

The details of the calculation are the same as that in Ref. 23. Here, the calculation method is described briefly. First, random conformations were prepared by MD calculations of single SDS molecule at high temperature. Using the obtained conformations, 60 DS ions were randomly placed in the cubic unit cell without any overlap, because the average aggregation number of SDS micelles is about 60 (MD calculations [7] and experiments [S1–S3]). The remaining space of the unit cell contained 8488 water molecules and 60 sodium ions. Six independent initial configurations were prepared.

After the energy optimization, constant-volume MD calculations were performed where only the solvent water molecules and sodium ions are allowed to move. During the calculation the velocity was scaled at T = 300 K. Then, the *NPT* ensemble MD calculation was performed at T = 300 K by the Nosé-Hoover chain method [S4] and at P = 0.1 MPa by the Andersen method [S5]. We adopted the CHARMM27 [S6] force field for SDS and TIP4P [S7] model for water. TIP4P can correctly reproduce the three-body correlation of liquid water. Combination of these potentials [7] can reproduce experimental mean aggregation number of the micelles. The particle mesh Ewald method [S8] was used for the electrostatic interaction calculation. The r-RESPA method [S9] was used for numerical integration of the equation of motion. The time step was 1 fs. The production run of the MD calculation was 100 ns. The MD calculation was performed using highly parallelized general purpose MD software, MODYLAS [S10].

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