

# Phase Behavior and Aggregate Transition in Aqueous Mixtures of Negatively-Charged Carbon Dots and Cationic Surfactants

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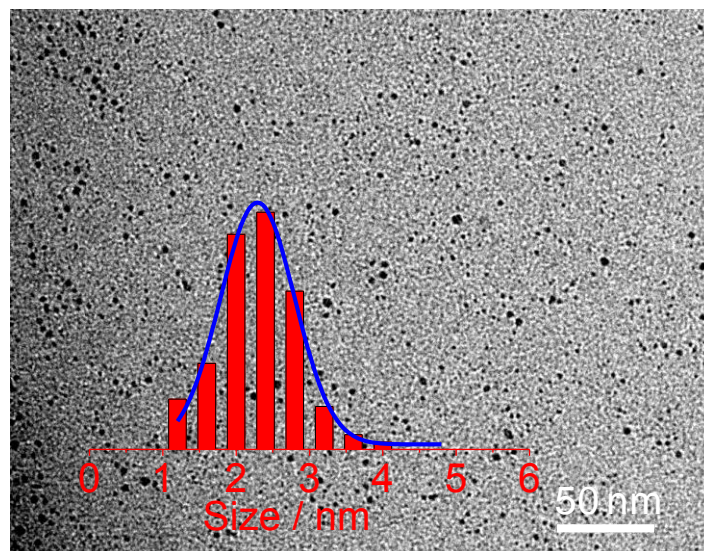
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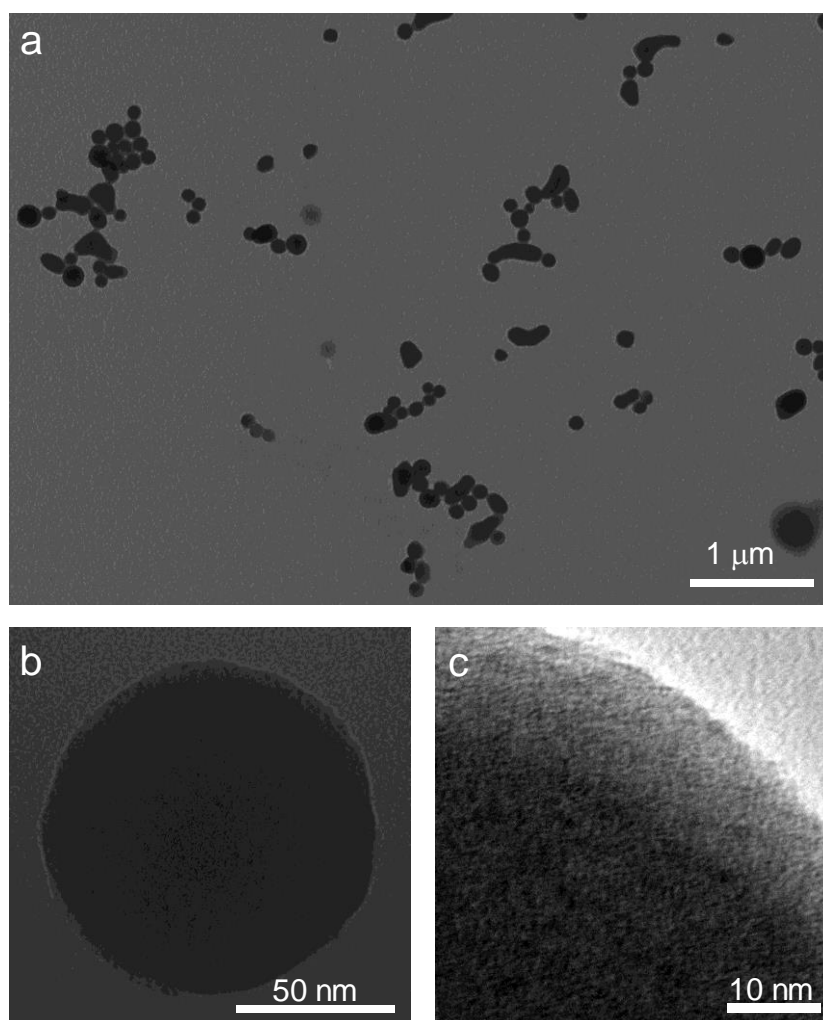
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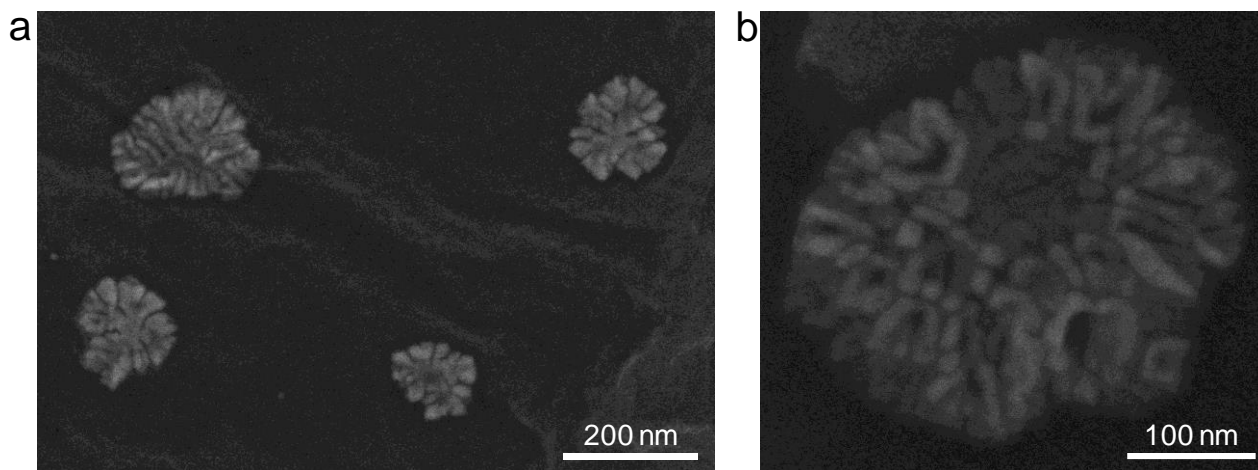
# These authors contribute equally.



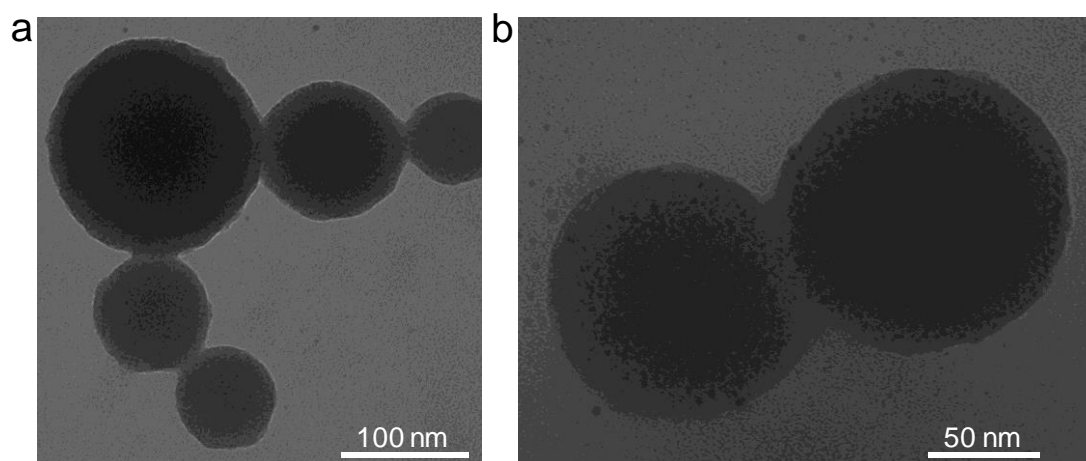
**Figure S1.** HR-TEM image and the size distribution of as-prepared *n*-CDs.



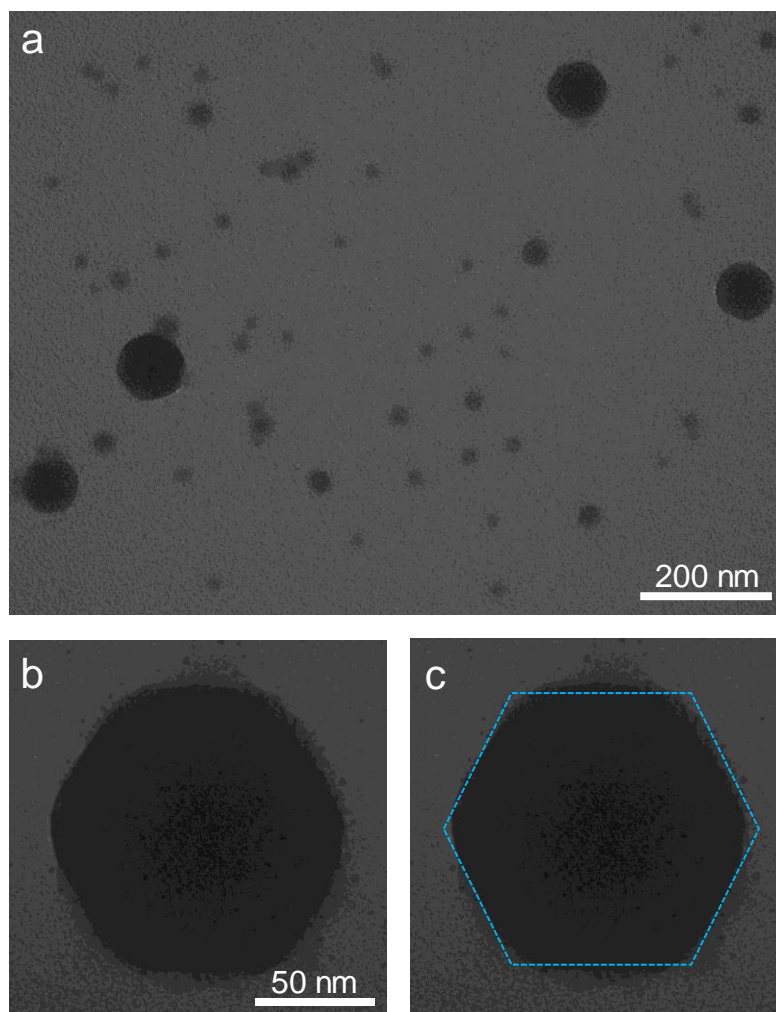
**Figure S2.** Additional TEM and HR-TEM images of the aggregates formed in *n*-CDs/*C*<sub>14</sub>mimB mixture at  $c_s = 0.6 \text{ mmol}\cdot\text{L}^{-1}$  and  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$ . a) TEM image with a low magnification. b) TEM image on a single aggregate. c) HR-TEM image on the edge of a typical aggregate.



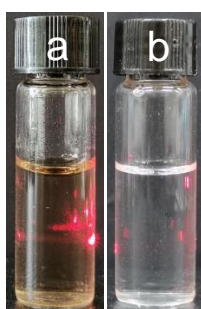
**Figure S3.** Additional SEM images of the aggregates formed in *n*-CDs/C<sub>14</sub>mimB mixture at  $c_s = 0.6$  mmol·L<sup>-1</sup> and  $c_{n\text{-CDs}} = 0.1$  mg·mL<sup>-1</sup>.



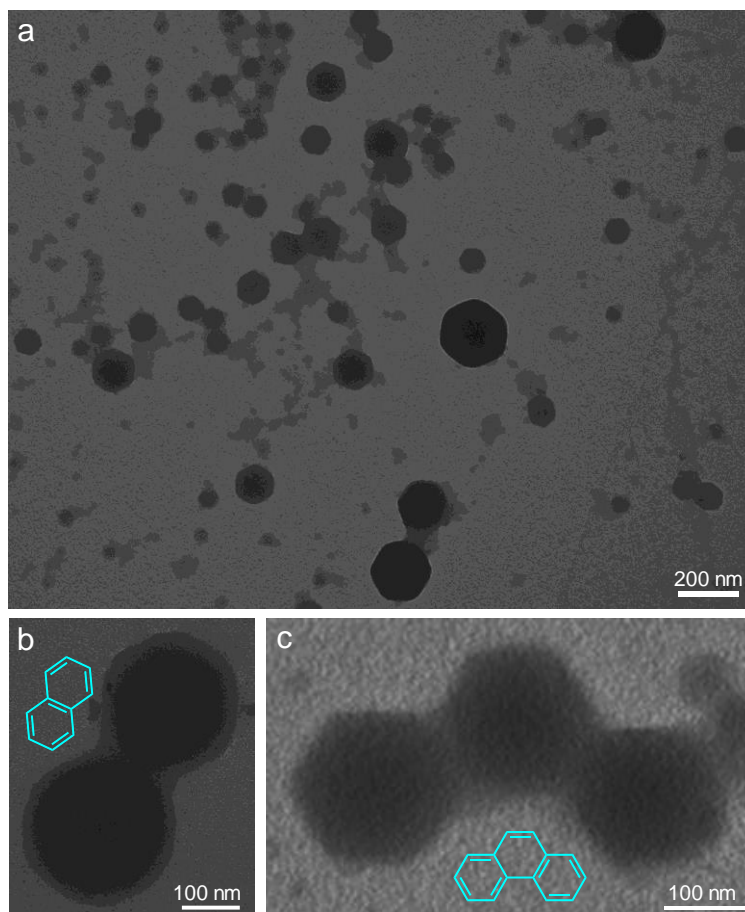
**Figure S4.** Additional TEM images of the aggregates formed in *n*-CDs/C<sub>14</sub>mimB mixture at  $c_s = 1.4$  mmol·L<sup>-1</sup> and  $c_{n\text{-CDs}} = 0.1$  mg·mL<sup>-1</sup>. a) Image of a cluster of the aggregates. b) Image of a dimer of the aggregates.



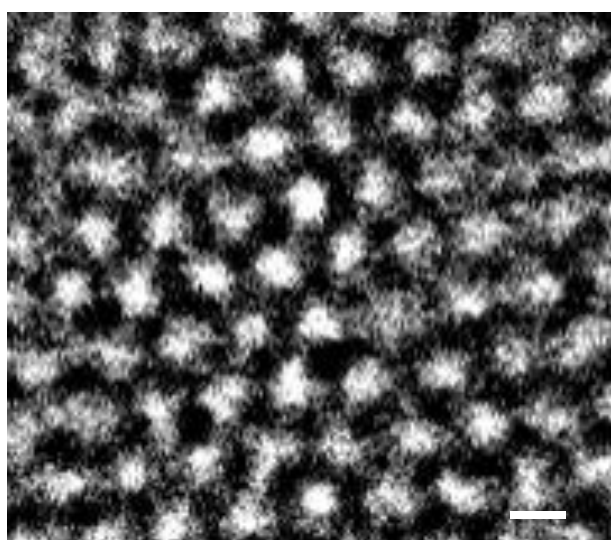
**Figure S5.** Additional TEM images of the aggregates formed in *n*-CDs/ $C_{14}$ mimB mixture at  $c_s = 2.8 \text{ mmol}\cdot\text{L}^{-1}$  and  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$ . a) Image with a low magnification. b) Image on a single aggregate. c) The same aggregate in image b together with a regular hexagon.



**Figure S6.** The Tyndall effect of pure *n*-CDs ( $0.1 \text{ mg}\cdot\text{mL}^{-1}$ , a) and  $C_{14}$ mimB ( $3 \text{ mmol}\cdot\text{L}^{-1}$ , b).

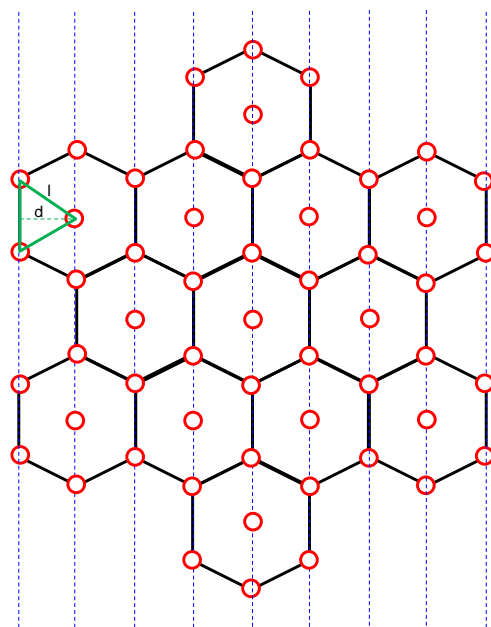


**Figure S7.** Additional TEM images of the aggregates formed in *n*-CDs/C<sub>14</sub>mimB mixture at  $c_s = 3.0 \text{ mmol}\cdot\text{L}^{-1}$  and  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$ . a) Image with a low magnification. b) Image of a dimer of the hexosomes. Inset is the chemical structure of naphthalene. c) Image of a trimer of the hexosomes. Inset is the chemical structure of anthracene.

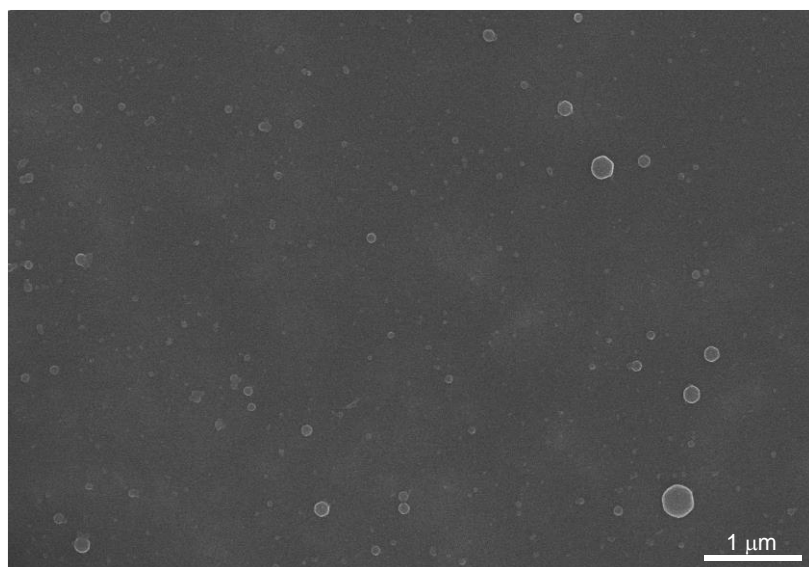


**Figure S8.** HR-TEM image of the hexagonal organization inside the aggregate. The scale bar corresponds to 5 nm.

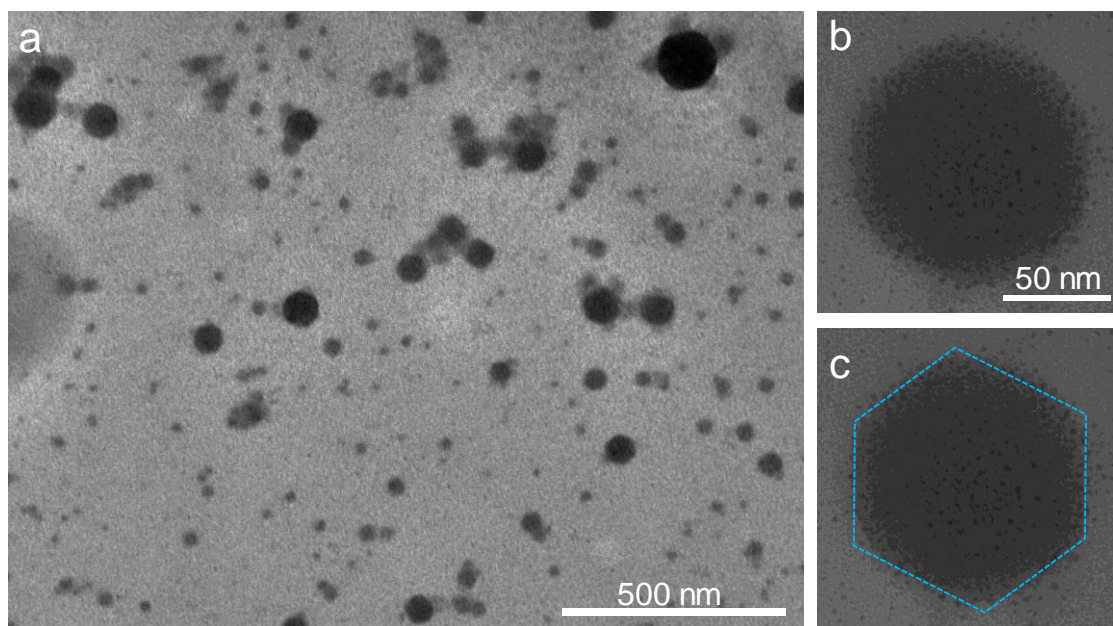




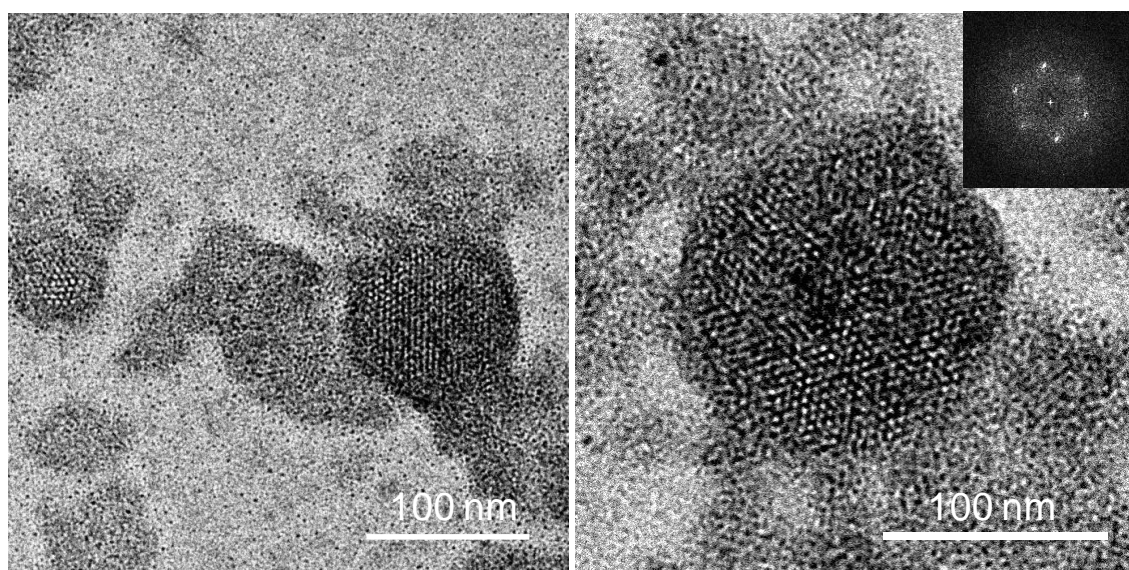
**Figure S9.** Schematic illustration of structures for the honeycomb structure formed in  $n$ -CDs/C<sub>14</sub>mimB mixture at  $c_s = 3.0 \text{ mmol}\cdot\text{L}^{-1}$  and  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$ .



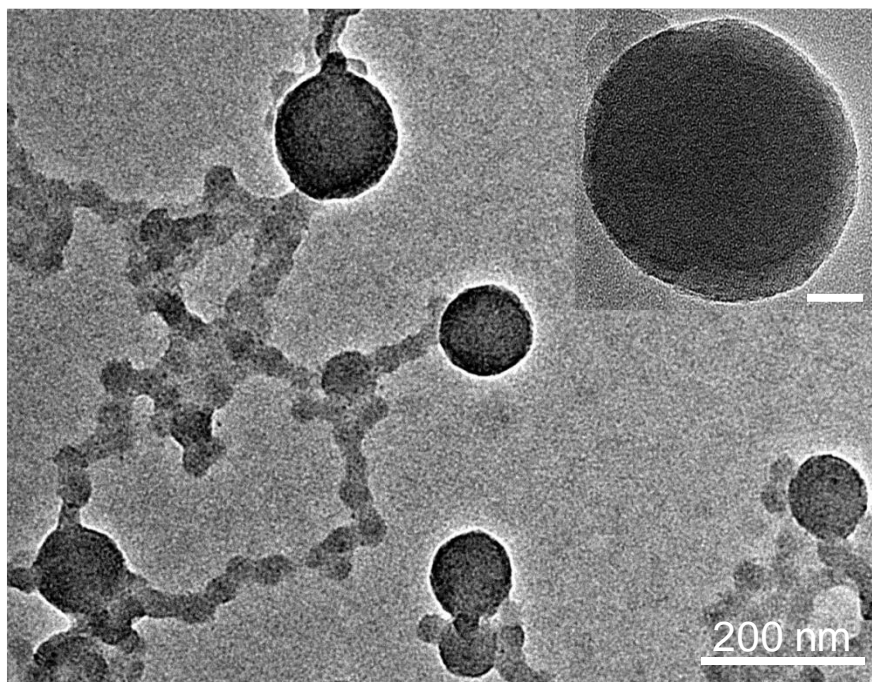
**Figure S10.** Additional SEM image with a low magnification of the aggregates formed in  $n$ -CDs/C<sub>14</sub>mimB mixture at  $c_s = 3.0 \text{ mmol}\cdot\text{L}^{-1}$  and  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$ .



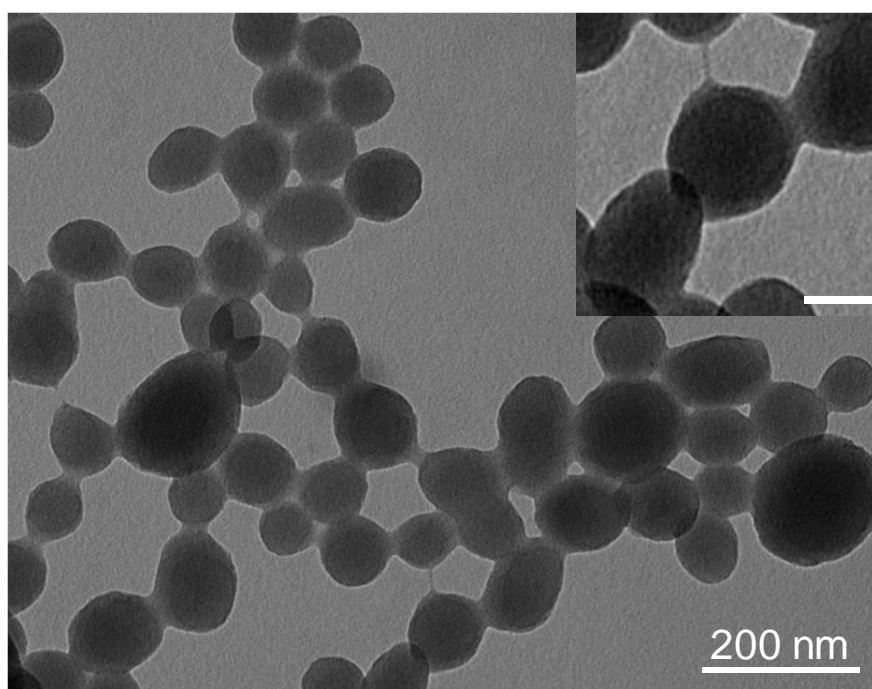
**Figure S11.** Additional TEM images of the aggregates formed in *n*-CDs/*C*<sub>14</sub>mimB mixture at  $c_s = 4.0 \text{ mmol}\cdot\text{L}^{-1}$  and  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$ . a) Image with a low magnification. b) Image on a single aggregate. c) The same aggregate in image b together with a regular hexagon.



**Figure S12.** TEM image of the aggregates formed in *n*-CDs/*C*<sub>14</sub>mimB mixture at  $c_s = 5.0 \text{ mmol}\cdot\text{L}^{-1}$ .  $c_{n\text{-CDs}}$  is fixed at  $0.1 \text{ mg}\cdot\text{mL}^{-1}$ .

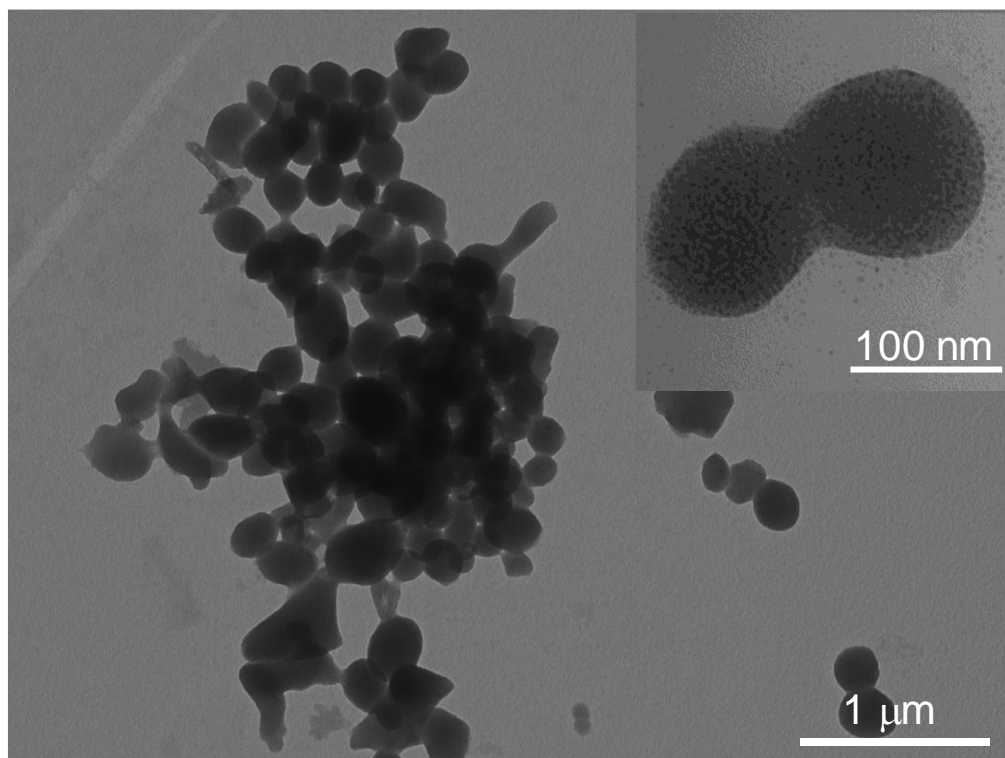


**Figure S13.** A typical HR-TEM image of the aggregates formed in *n*-CDs/TTAB mixture at  $c_s = 0.4$  mmol·L<sup>-1</sup> and  $c_{n\text{-CDs}} = 0.1$  mg·mL<sup>-1</sup>. The bar in the inset image corresponds to 20 nm.

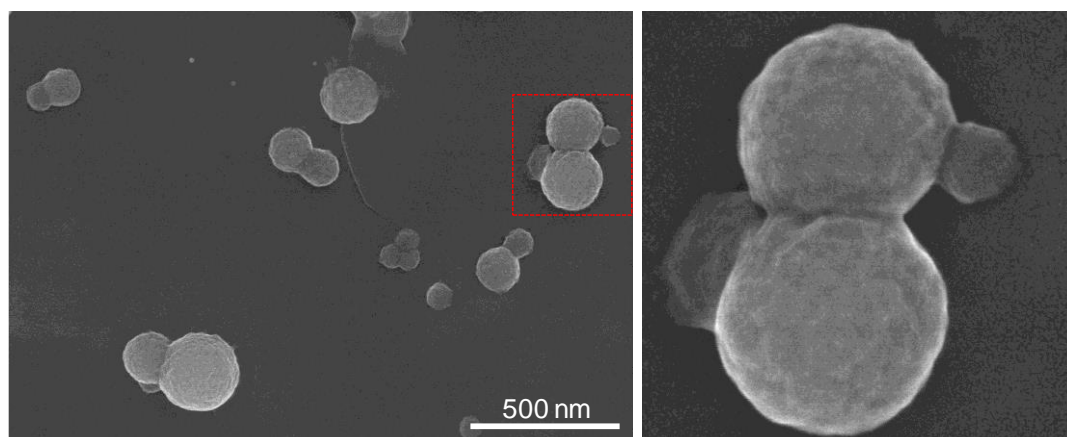


**Figure S14.** A typical TEM image of the aggregates formed in *n*-CDs/TTAB mixture at  $c_s = 0.7$  mmol·L<sup>-1</sup> and  $c_{n\text{-CDs}} = 0.1$  mg·mL<sup>-1</sup>. The bar in the inset image corresponds to 50 nm.

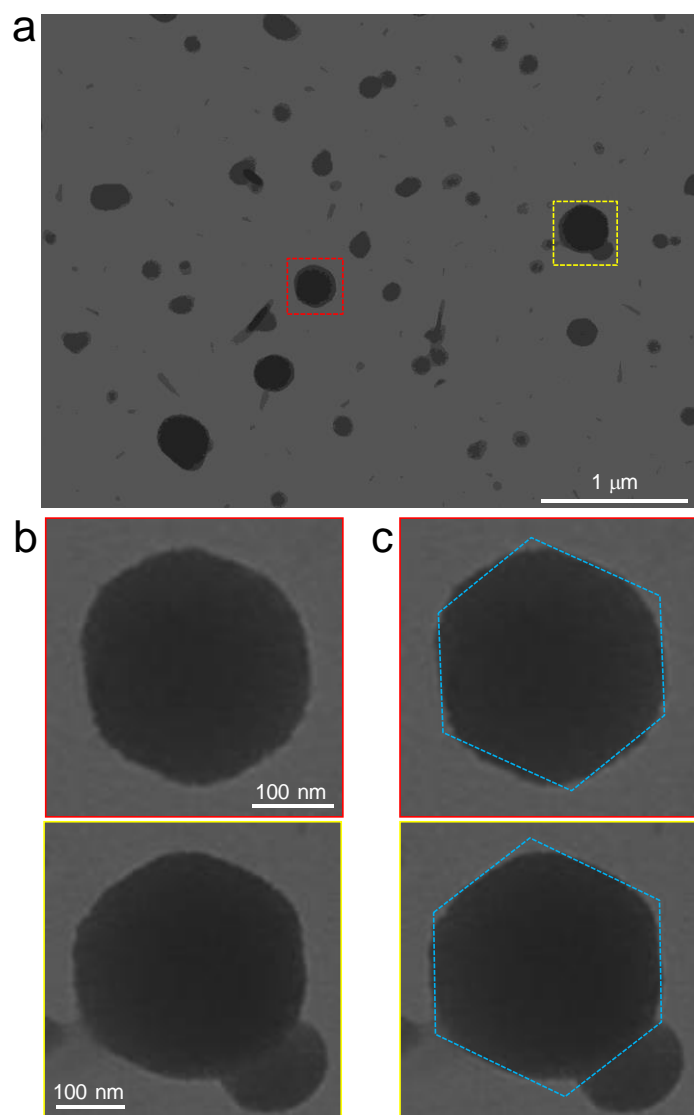




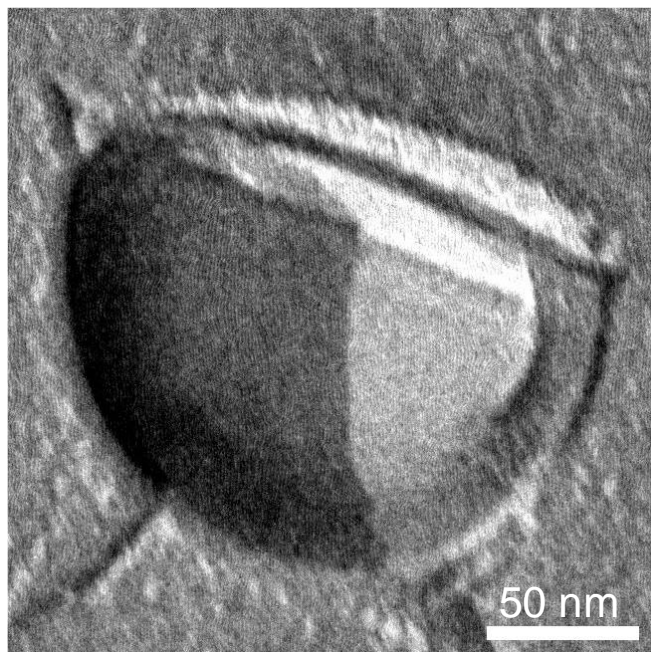
**Figure S15.** A typical HR-TEM image of the aggregates formed in *n*-CDs/TTAB mixture at  $c_s = 2.4 \text{ mmol}\cdot\text{L}^{-1}$  and  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$ .



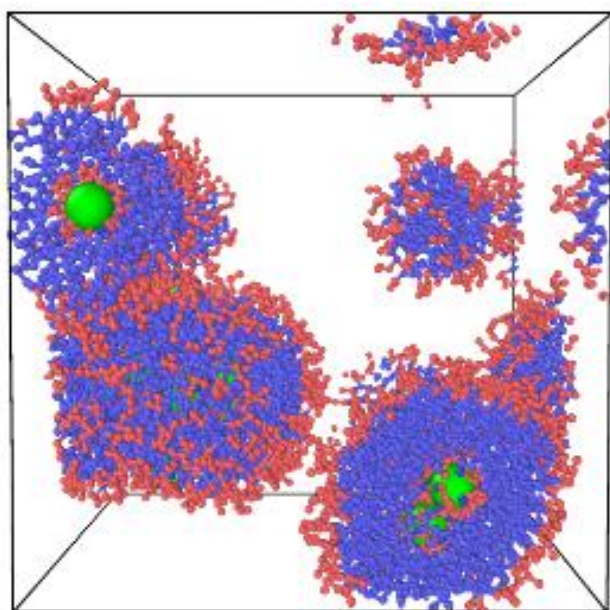
**Figure S16.** A typical SEM image and the magnified graph on the marked region of the aggregates formed in *n*-CDs/TTAB mixture at  $c_s = 3.0 \text{ mmol}\cdot\text{L}^{-1}$  and  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$ .



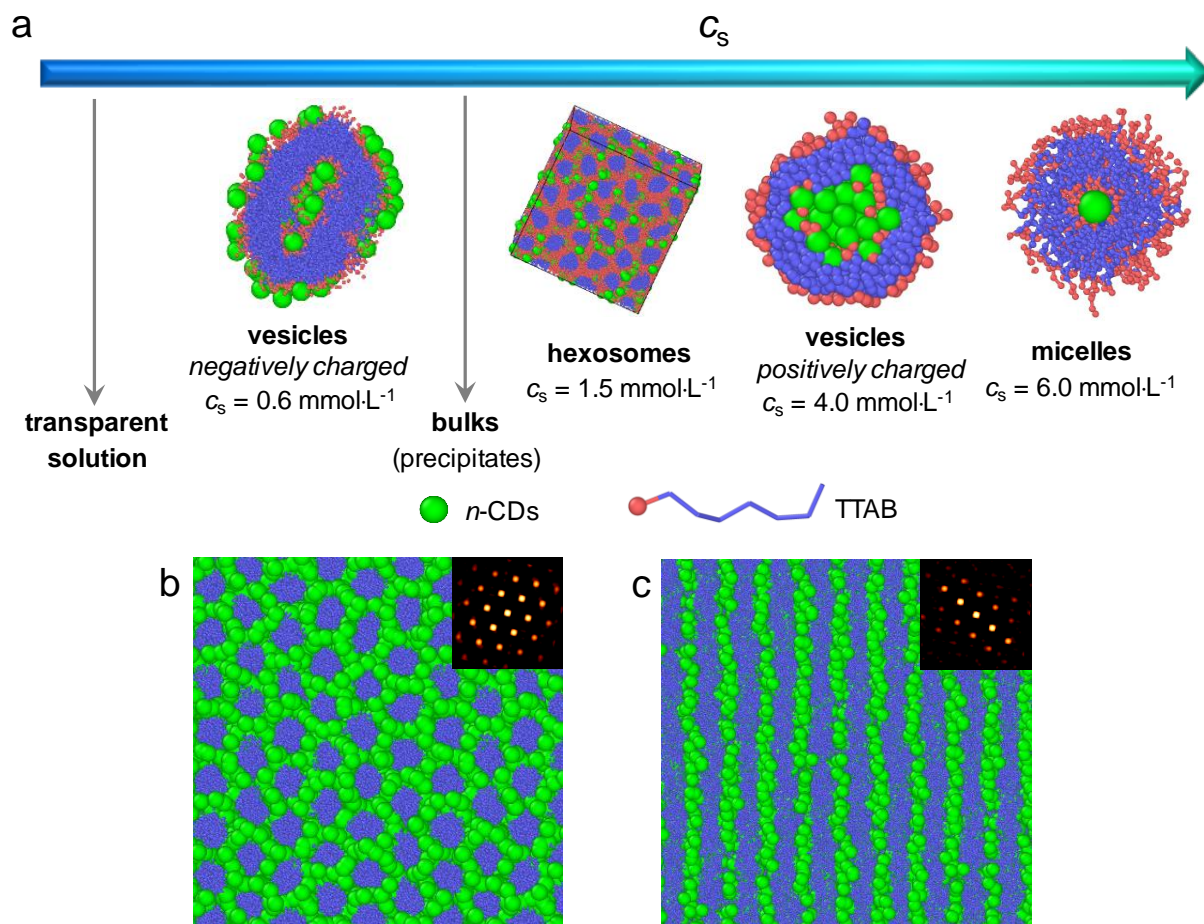
**Figure S17.** TEM image and the magnified graphs on the marked regions of the aggregates formed in *n*-CDs/TTAB mixture at  $c_s = 4.0 \text{ mmol}\cdot\text{L}^{-1}$  and  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$ . a) Image with a low magnification. b) Magnifications of the two marked regions in image a. c) The same aggregates in image b together with regular hexagons.



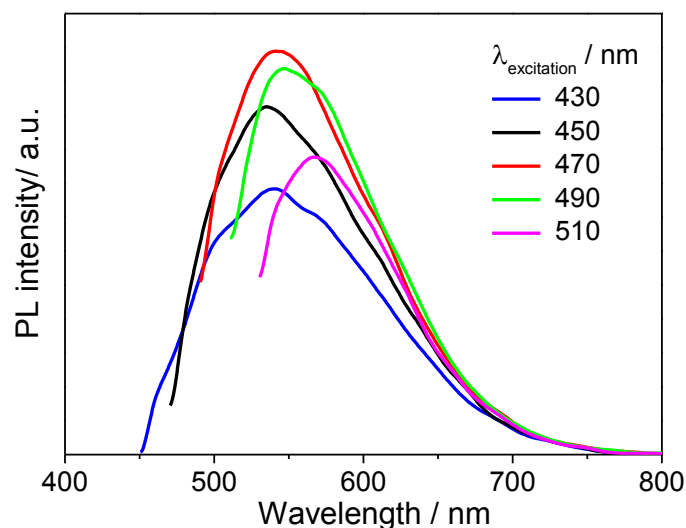
**Figure S18.** A typical FF-TEM image showing the presence of facted aggregate in *n*-CDs/TTAB mixture at  $c_s = 4.0 \text{ mmol}\cdot\text{L}^{-1}$  and  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$ .



**Figure S19.** Snapshot showing the coexistence of micelles and vesicles in *n*-CDs/ $\text{C}_{14}\text{mimB}$  mixture at  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$  and  $c_s = 6.0 \text{ mmol}\cdot\text{L}^{-1}$ .

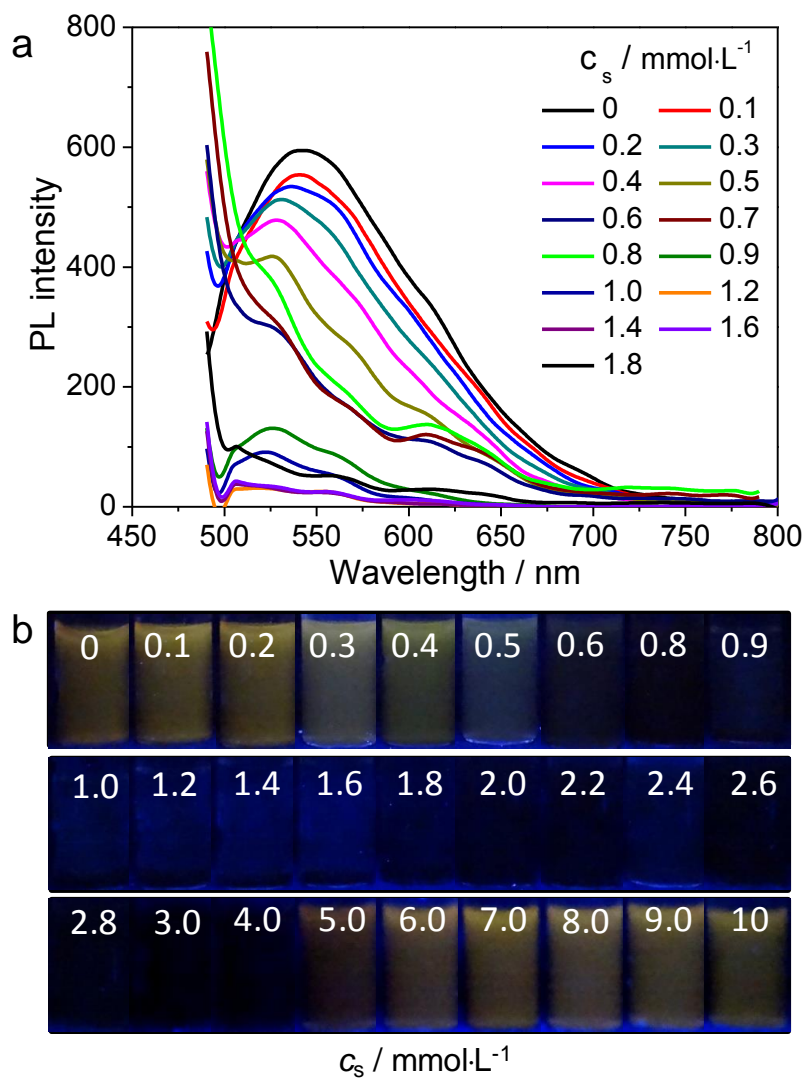


**Figure S20.** a) The self-assemblies formed in  $n\text{-CDs/TTAB}$  mixture with increasing  $c_s$  obtained by DPD method. For vesicles, only the intact cross sectional structures are shown. b,c) Structures of the hexosomes viewed from transverse and longitudinal surfaces, respectively. Insets are corresponding diffraction patterns.  $c_{n\text{-CDs}}$  is fixed at  $0.1 \text{ mg}\cdot\text{mL}^{-1}$ .



**Figure S21.** Emission curves recorded at varying excitation wavelength for the aqueous solution of  $n\text{-CDs}$  ( $0.1 \text{ mg}\cdot\text{mL}^{-1}$ ).





**Figure S22.** Emission at 470 nm excitation (a) and photos (b) of typical samples from *n*-CDs/TTAB mixture at  $c_{n\text{-CDs}} = 0.1 \text{ mg}\cdot\text{mL}^{-1}$  and varying  $c_s$  as indicated.