Electronic Supplementary Material (ESI) for Soft Matter.

Supplementary Information

Alpha Helical Surfactant-Like Peptides Self-Assemble into pH-Dependent Nanostructures

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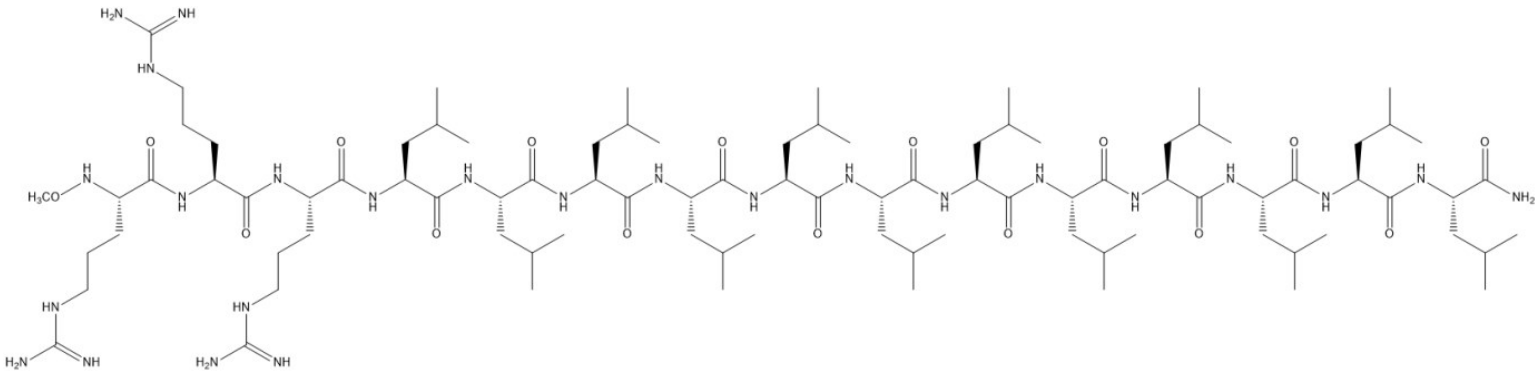

Scheme S1. Molecular structure of peptide R_3L_{12}, capped at both termini.

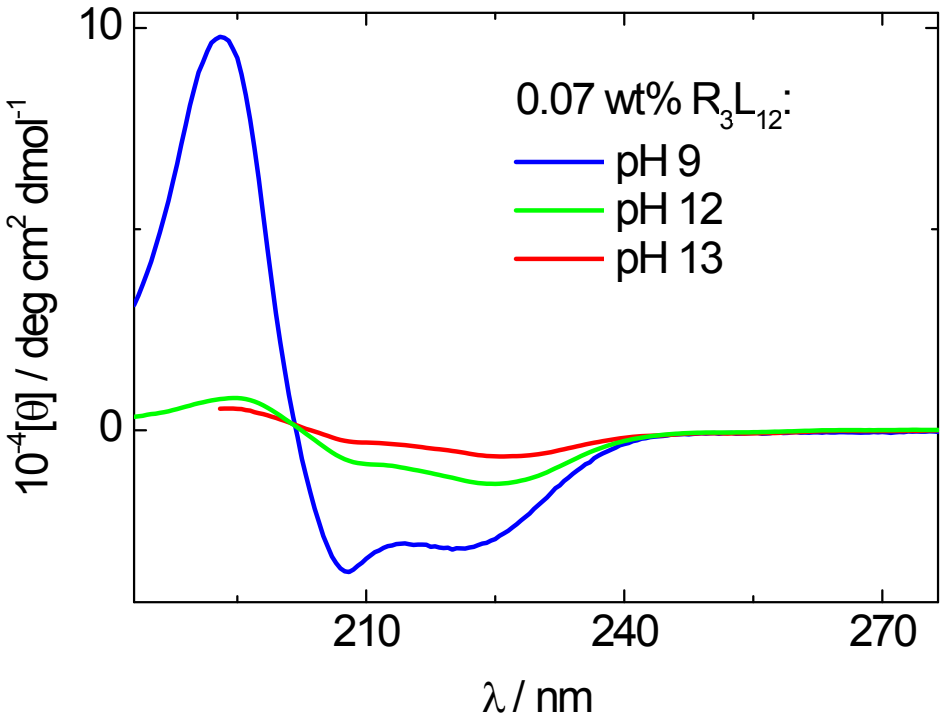

Figure S1. CD spectra for 0.07 wt% R_3L_{12} measured at pH 9, 12 and 13.

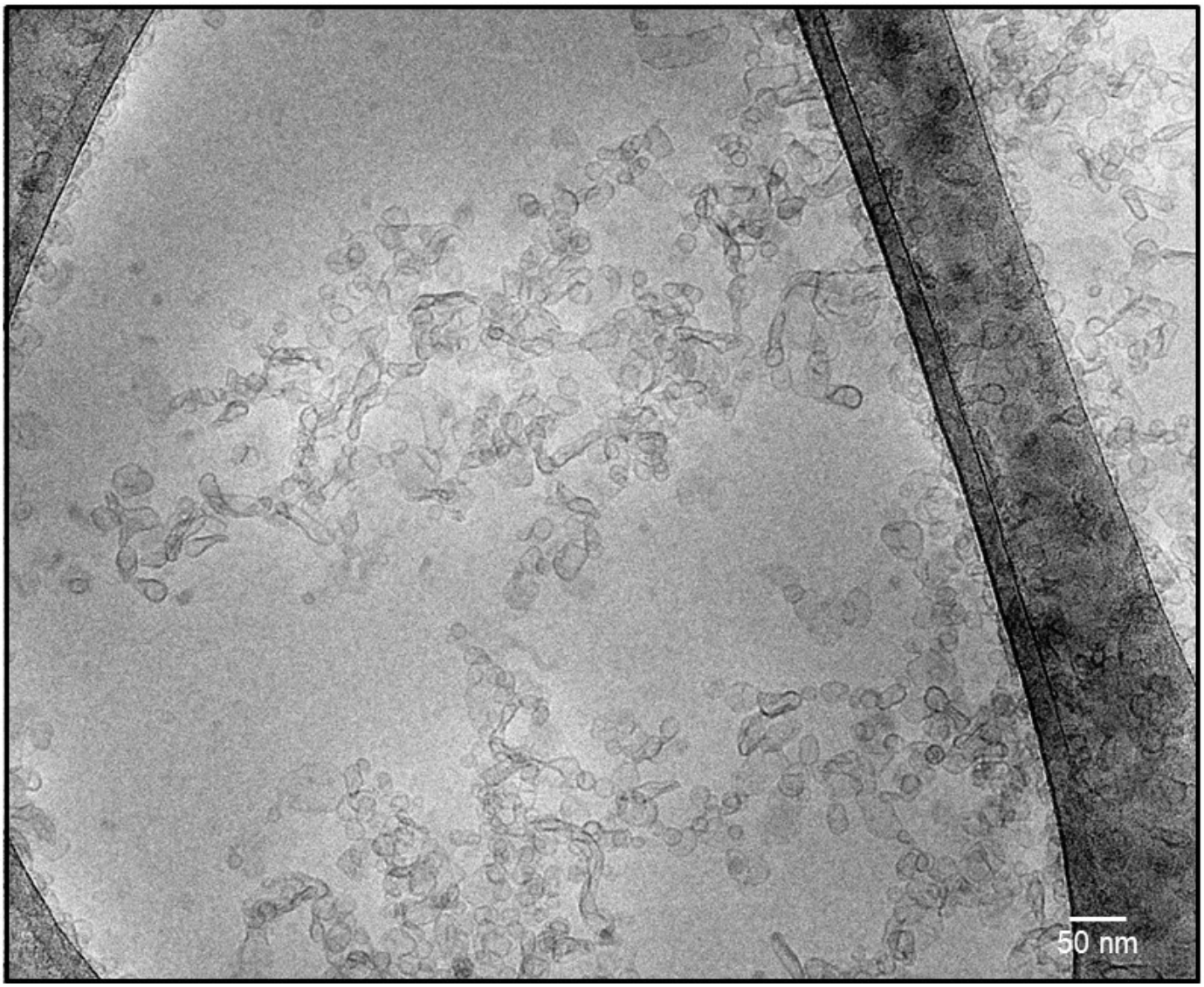

Figure S2. Cryo-TEM image for 0.07 wt% R_3L_{12} at pH 9 showing branched nanotubes.

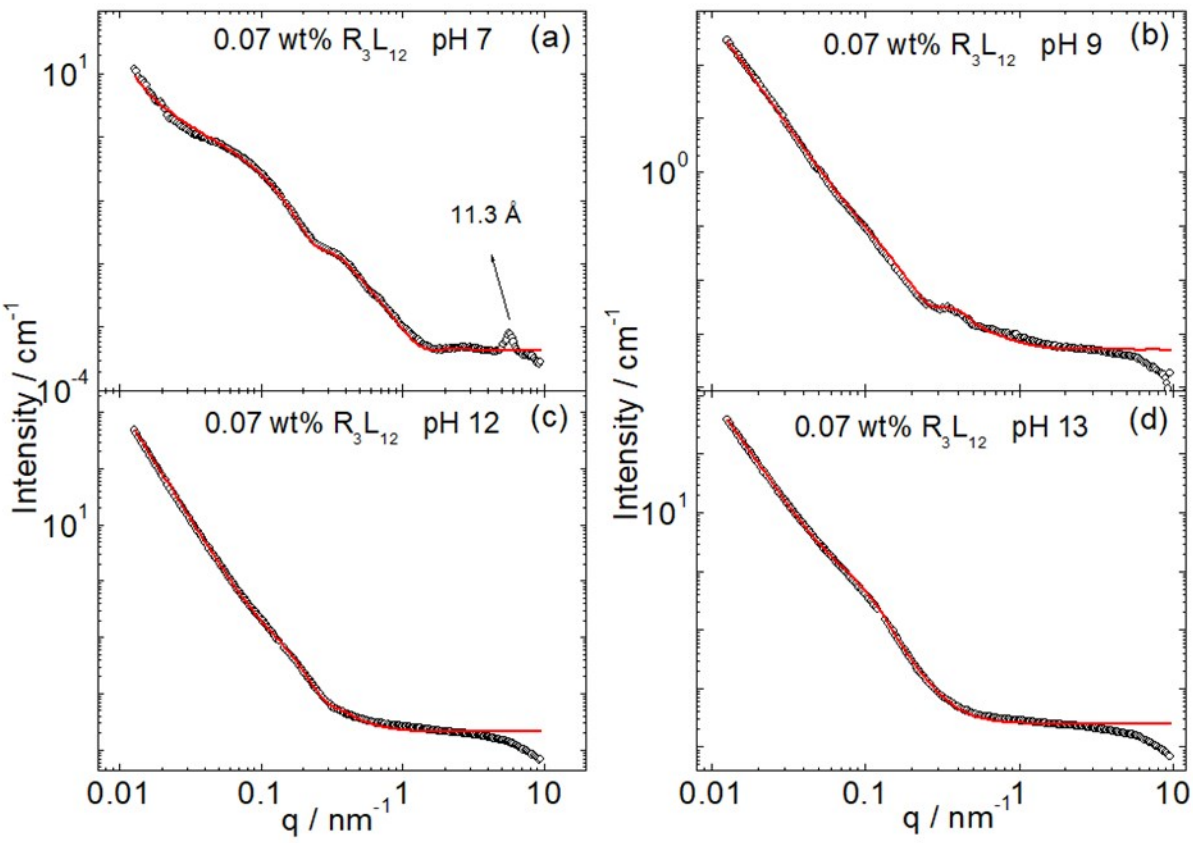

Figure S3. Measured SAXS intensity profiles (open symbols) fitted with form factors (solid lines) corresponding to nanotubes at pH 7 and pH 9, and spherical shells at pH 12 and pH 13.

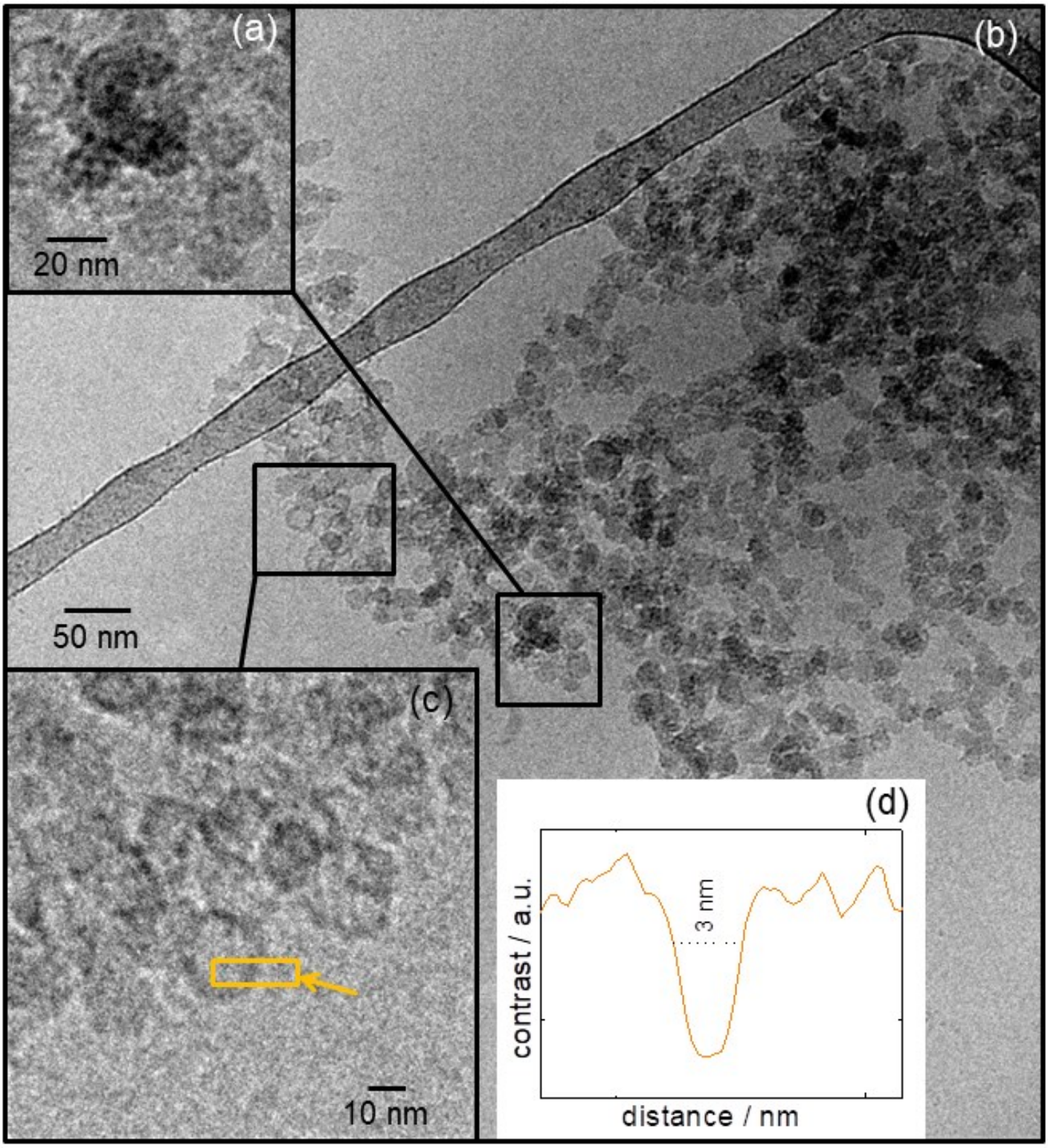

Figure S4. (a-c) Cryo-TEM images for 0.04 wt% R_3L_{12} at pH 12; (a, b) magnification of the area inside the squares in (b); (d) linear relative contrast profile of the rectangular mask across the raspberry particle wall, as indicated by an arrow (c). The relative contrast scale in (d) extends from dark (lower) to light (upper).

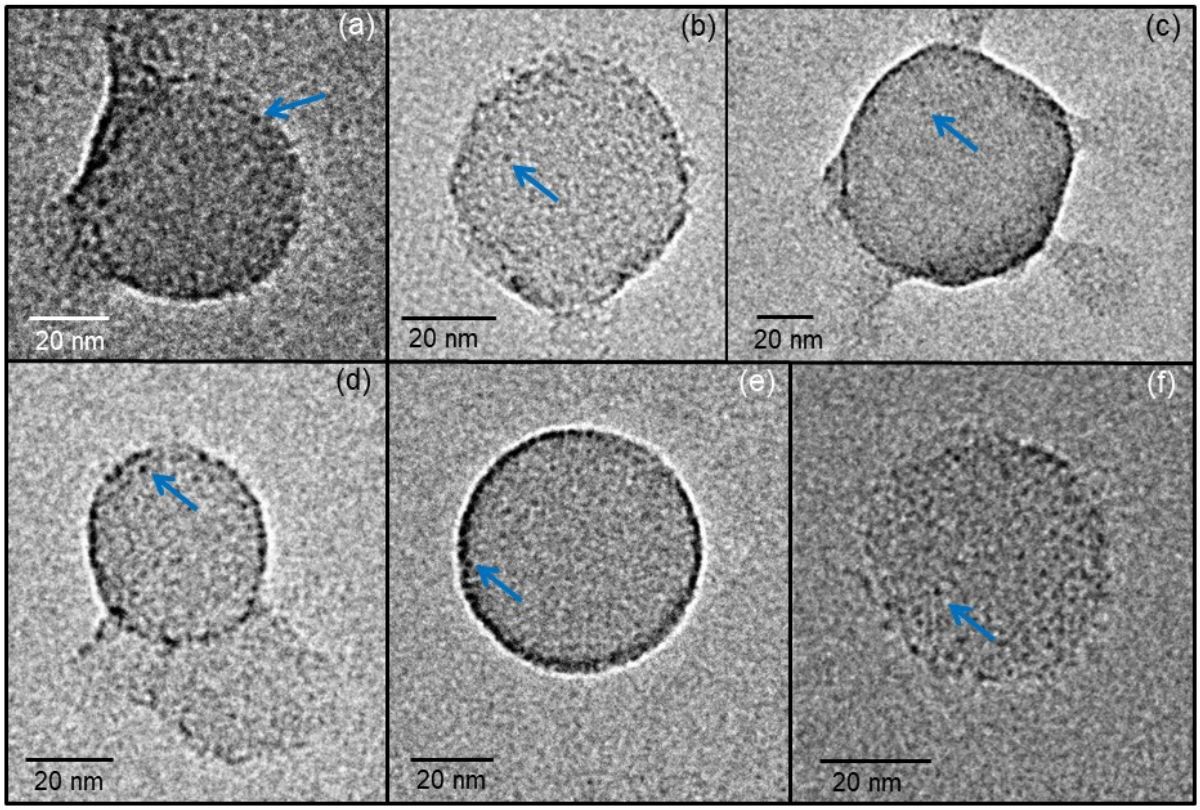

Figure S5. (a-f) Representative examples of spherical aggregates imaged by TEM for 0.04 wt% R_3L_{12} measured at pH 12. The blue arrows point to the 3-nm dotted motif decoration on the surface of the raspberry particle.

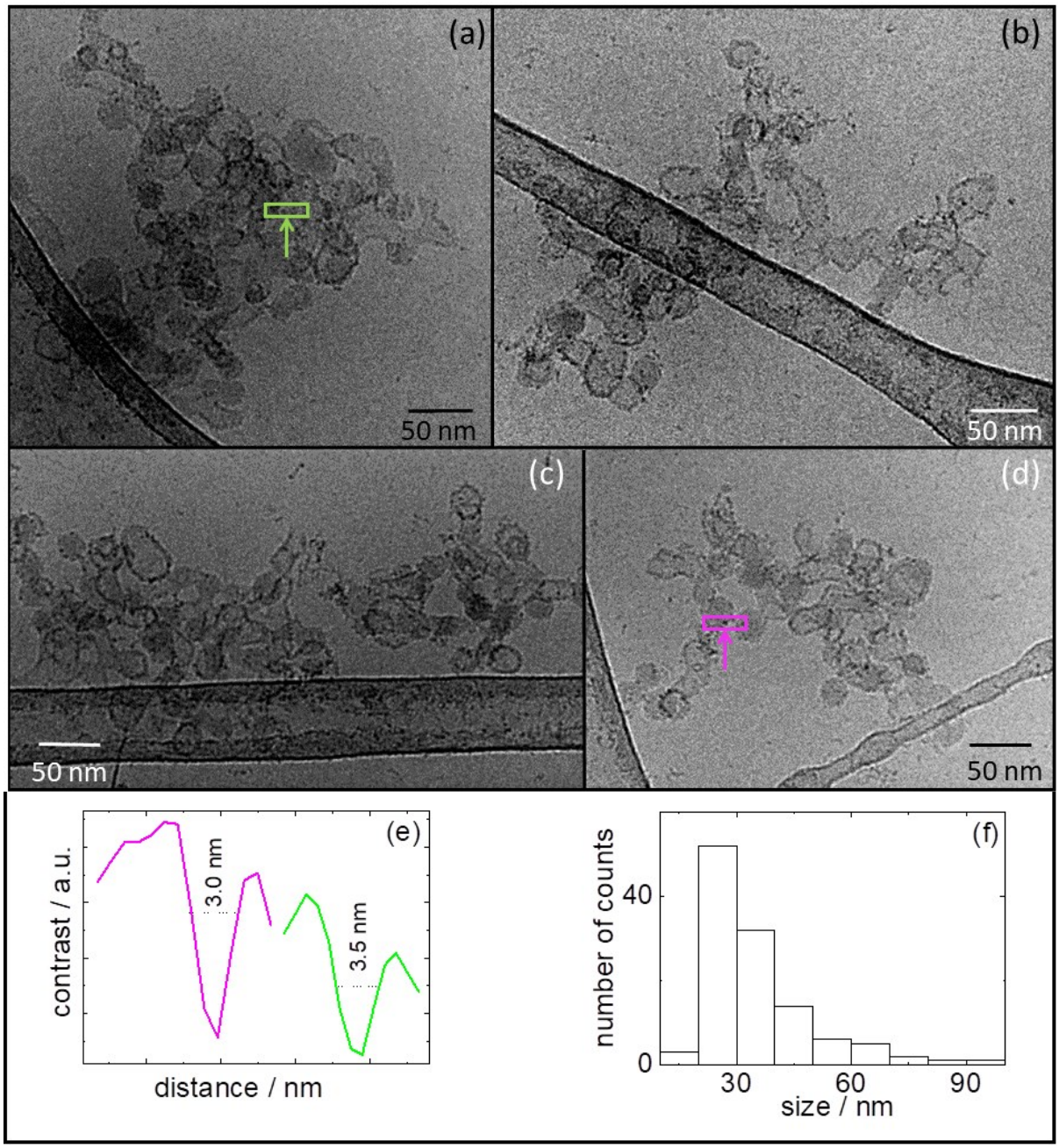

Figure S6. (a-d) Cryo-TEM images for 0.07 wt% R_3L_{12} at pH 12; (e) linear relative contrast profile of the rectangular masks enclosing particles, as indicated by arrows in (a, b). The lines in (e) correspond to the same colour boxes in (a, d). (f) Distribution of aggregate sizes, measured from the whole set of cryo-TEM images. The relative contrast scale in (e) extends from dark (lower) to light (upper).

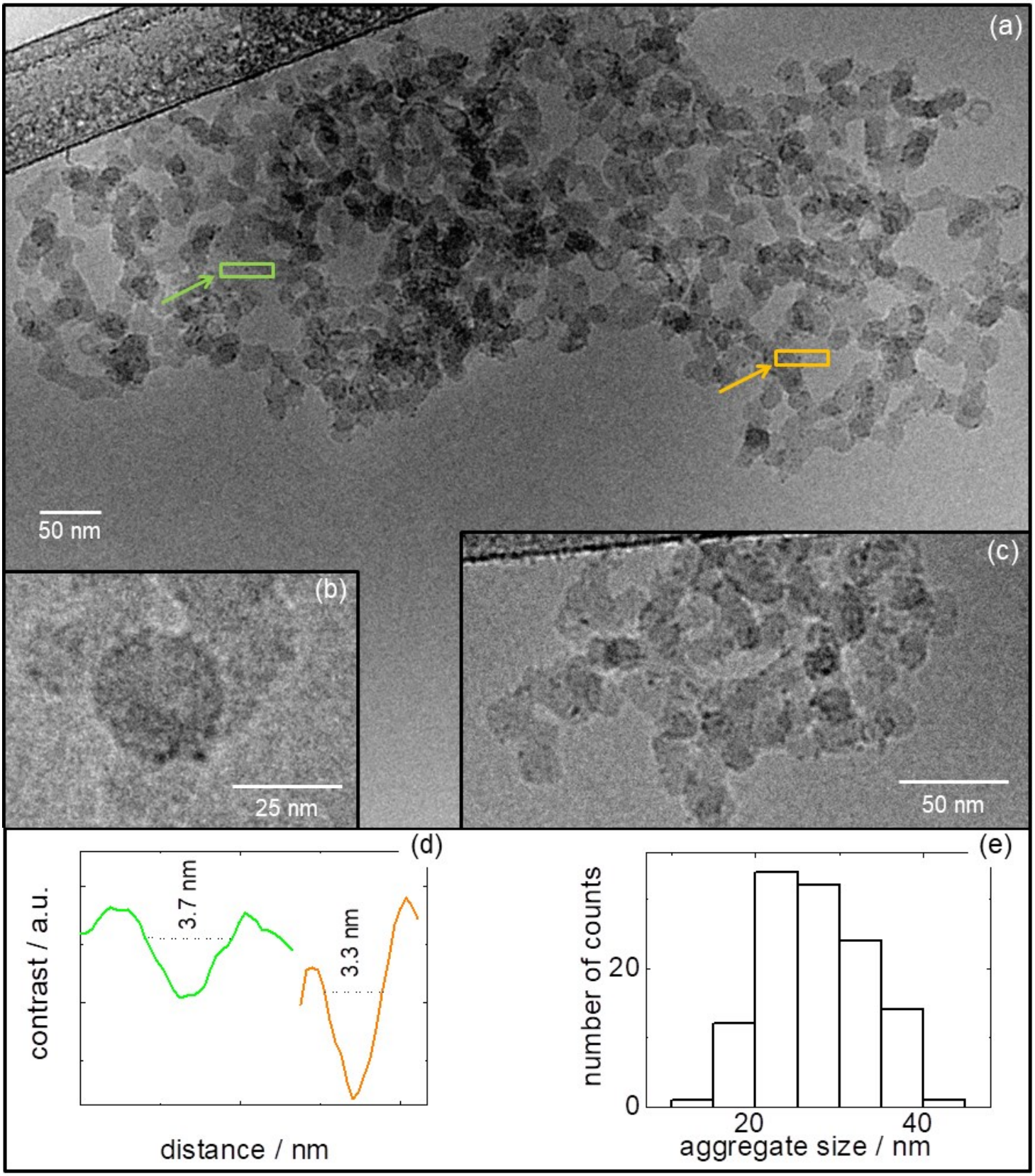

Figure S7. (a-c) Cryo-TEM images for 0.07 wt% R_3L_{12} at pH 13; (d) linear relative contrast profile of the rectangular masks enclosing particles, as indicated by arrows in (a). The lines in (d) correspond to the same colour boxes in (a). (e) Distribution of aggregate sizes, measured from the whole set of cryo-TEM images. The relative contrast scale in (d) extends from dark (lower) to light (upper).

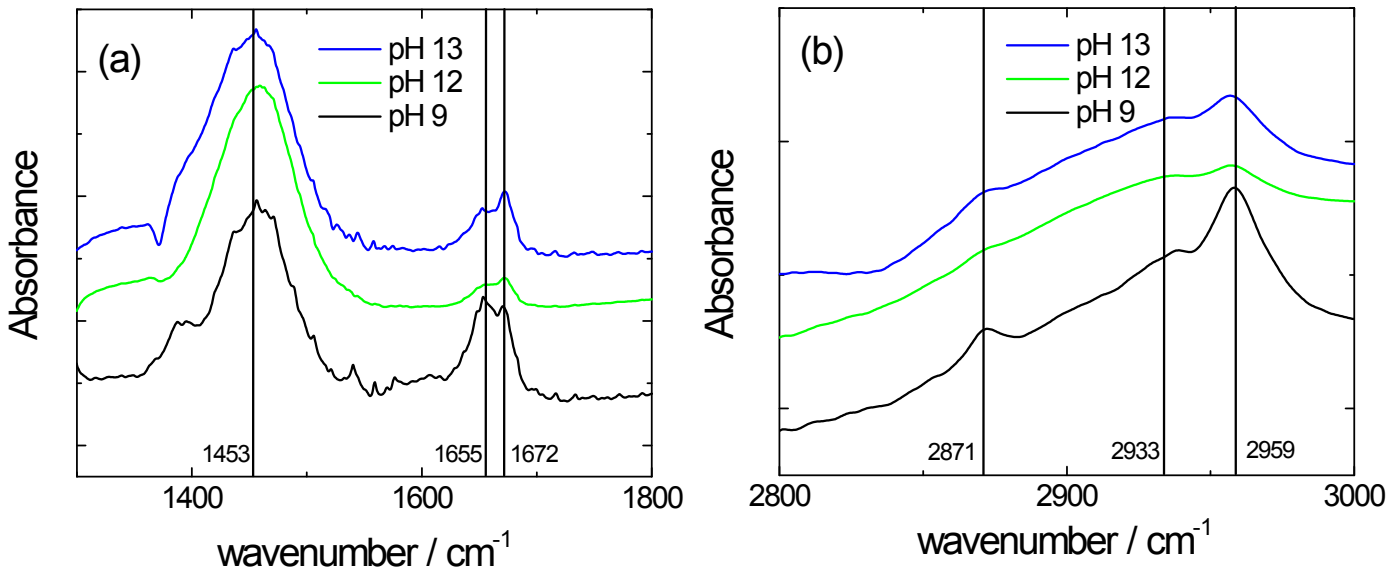

Figure S8. FTIR spectra for 0.07 wt% solutions of R_3L_{12} at the pH values indicated, in regions of the spectra corresponding to (a) amide I, (b) $CH/CH_2/CH_3$ deformation modes. The positions of main peaks are delineated. Spectra are offset vertically for clarity.

Table S1. Parameters extracted from the fitting of the SAXS in Figure 3 and Figure S2.

	0.04 wt% R_3L_{12} pH 7	0.07 wt% R_3L_{12} pH 7	0.04 wt% R_3L_{12} pH 9	0.07 wt% R_3L_{12} pH 9	0.04 wt% R_3L_{12} pH 12	0.07 wt% R_3L_{12} pH 12	0.04 wt% R_3L_{12} pH 13	0.07 wt% R_3L_{12} pH 13
N_c [cm^{-3}]	1	1	0.5	1	--	--	--	--
$R_c \pm \Delta R_c$ [nm]	5.3±2.8	6.0±3.0	9.0±2.0	7.0±2.0	--	--	--	--
D_c [nm]	3.7	3.7	2.6	2.1	--	--	--	--
η_c [cm^2]	$1.8x10^{-6}$	$3x10^{-6}$	$2.8x10^{-5}$	$1x10^{-5}$	--	--	--	--
η_s [cm^2]	$2.6x10^{-5}$	$3.1x10^{-5}$	$7.5x10^{-5}$	$3.1x10^{-5}$	--	--	--	--
η_{solv}[cm^2]	$2.2x10^{-6}$	$4x10^{-6}$	$3.2x10^{-5}$	$1.2x10^{-5}$	--	--	--	--
N_b [cm^{-3}]	1	1	--	--	--	--	--	--
2zH [nm]	3.0	2.0	--	--	--	--	--	--
σ_H [nm]	0.3	0.3	--	--	--	--	--	--
η_H [cm^2]	$3x10^{-6}$	$8x10^{-6}$	--	--	--	--	--	--
σ_C [nm]	0.2	0.3	--	--	--	--	--	--
η_C [cm^2]	$-2.4x10^{-5}$	$2.2x10^{-6}$	--	--	--	--	--	--
N_s [cm^{-3}]	--	--	--	--	1	1	1	1
$R_s \pm \Delta R_s$ [nm]	--	--	--	--	8.0 ± 3.0	7.0 ± 3.0	12.3 ± 3.0	10.0 ± 7.0
D_s [nm]	--	--	--	--	3.8	3.4	4.0	3.8
η_c [cm^2]	--	--	--	--	$1.7x10^{-5}$	$9.0x10^{-6}$	$1.7x10^{-5}$	$3.1x10^{-5}$
η_s [cm^2]	--	--	--	--	$5x10^{-5}$	$7x10^{-5}$	$7x10^{-7}$	$4x10^{-5}$
a_o	$2.1x10^{-4}$	$4.3x10^{-4}$	$5.5x10^{-4}$	$5x10^{-4}$	$2x10^{-3}$	$2.1x10^{-3}$	$2.5x10^{-3}$	$2.5x10^{-3}$
a_1	--	$1.5x10^{-8}$	$4.1x10^{-5}$	$7x10^{-6}$	$3x10^{-5}$	1.3x10-5	$6x10^{-6}$	$2.5x10^{-5}$
c_0	--	4.5	3.6	4	4	4	4	3.8

Key. Cylindrical core-shell: scale factor, N_c, core radius, R_c, shell thickness, D_c, scattering length density of the core, η_c, shell, η_s, and solvent, $\eta_{solv.}$ **Nanotube wall Gaussian density profile form factor:** scale factor, N_b, Gaussian half-width at half-maximum for polydispersity, $\Delta_{2zH,}$ inter-head group thicknesses, $2z_H$, Gaussian half-width for outer layer surface, σ_H, electron density for headgroup, η_H, Gaussian half-width for inner layer, σ_C, relative electron density for inner layer, η_C. BG is the background. **Spherical shell:** scale factor, N_s, external shell radius, R_s, shell thickness, D_s, scattering length density of inner core, η_c, shell, η_s, and inner shell, $\eta_{sc.}$. a_1, a_2 and c_0 are the constants for the background $a_1+a_2q^{-c0}$.