

Supplementary Information

for

Structure and growth behavior of centimeter-sized helical oleate assemblies formed with assistance of medium-length carboxylic acids

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Figure S1 and Figure S2

Micrographs of oleate helical assemblies in the presence of a small amount of *N*-decanoyl-L-alanine.

Figure S3

HPLC analysis results for assemblies prepared in the presence of *N*-decanoyl-L-alanine.

Figure S4

Micrographs of oleate helical assemblies in the presence of several fatty acids.

Figure S5

Site-selective SAXS for DOPC myelin figures using synchrotron microbeam X-ray diffraction.

Figure S6

pH titration curve of **ONa** (6.6 mM) and **1** (0.66 mM)

AND

Movie (available from <http://pubs.rsc.org/>)

Microscopic movie for growth dynamics of oleate helical assembly from terminal scaffold
(compressed to 1024-fold speed).

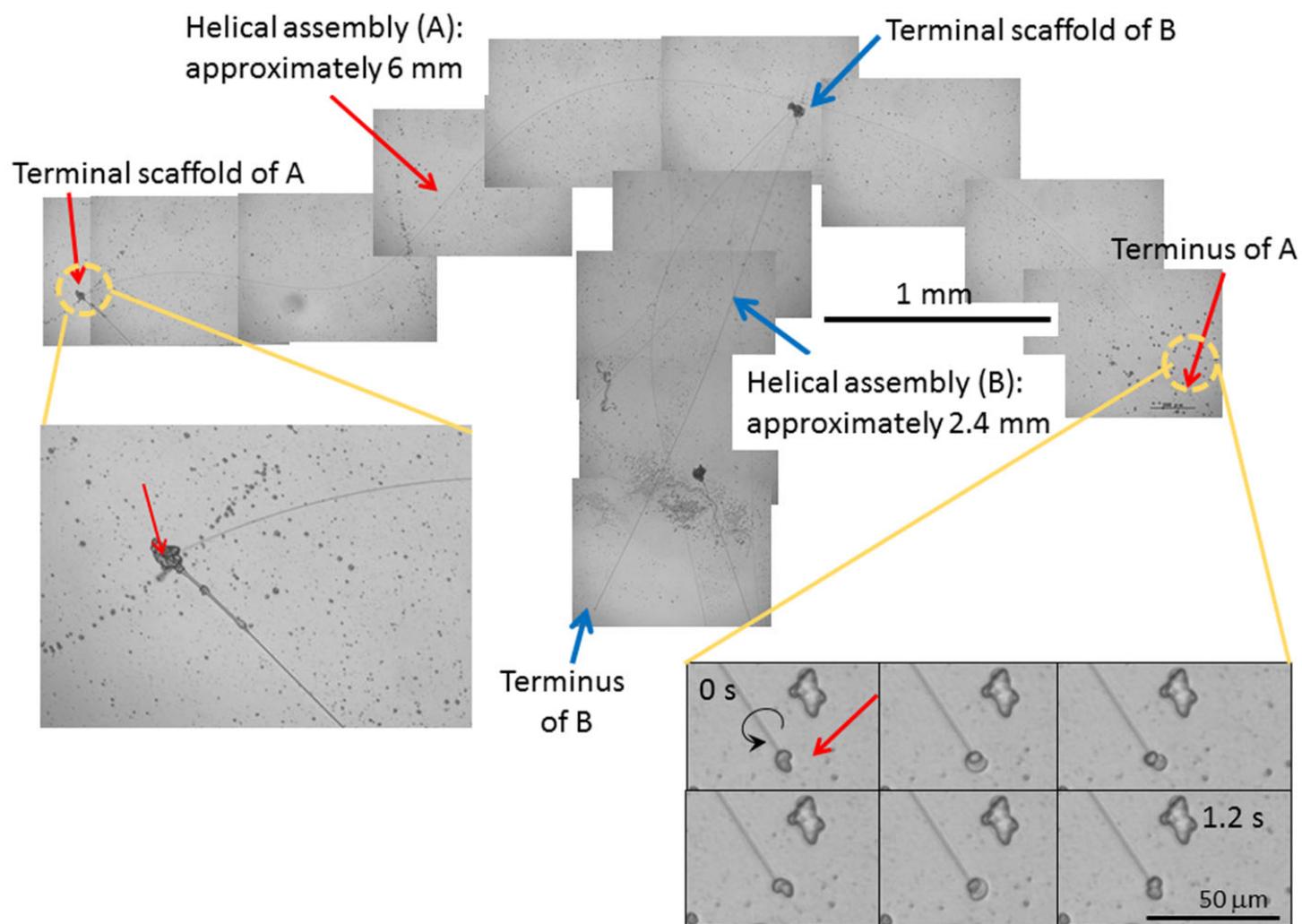


Figure S1. Several helical assemblies of oleate in several millimeter-long in the presence of **1** (1% (w/w)) in bicine-buffered water observed after incubation for 1 day. The winding frequency of helical assembly A, indicated by red arrows, was approximately 0.8 Hz.

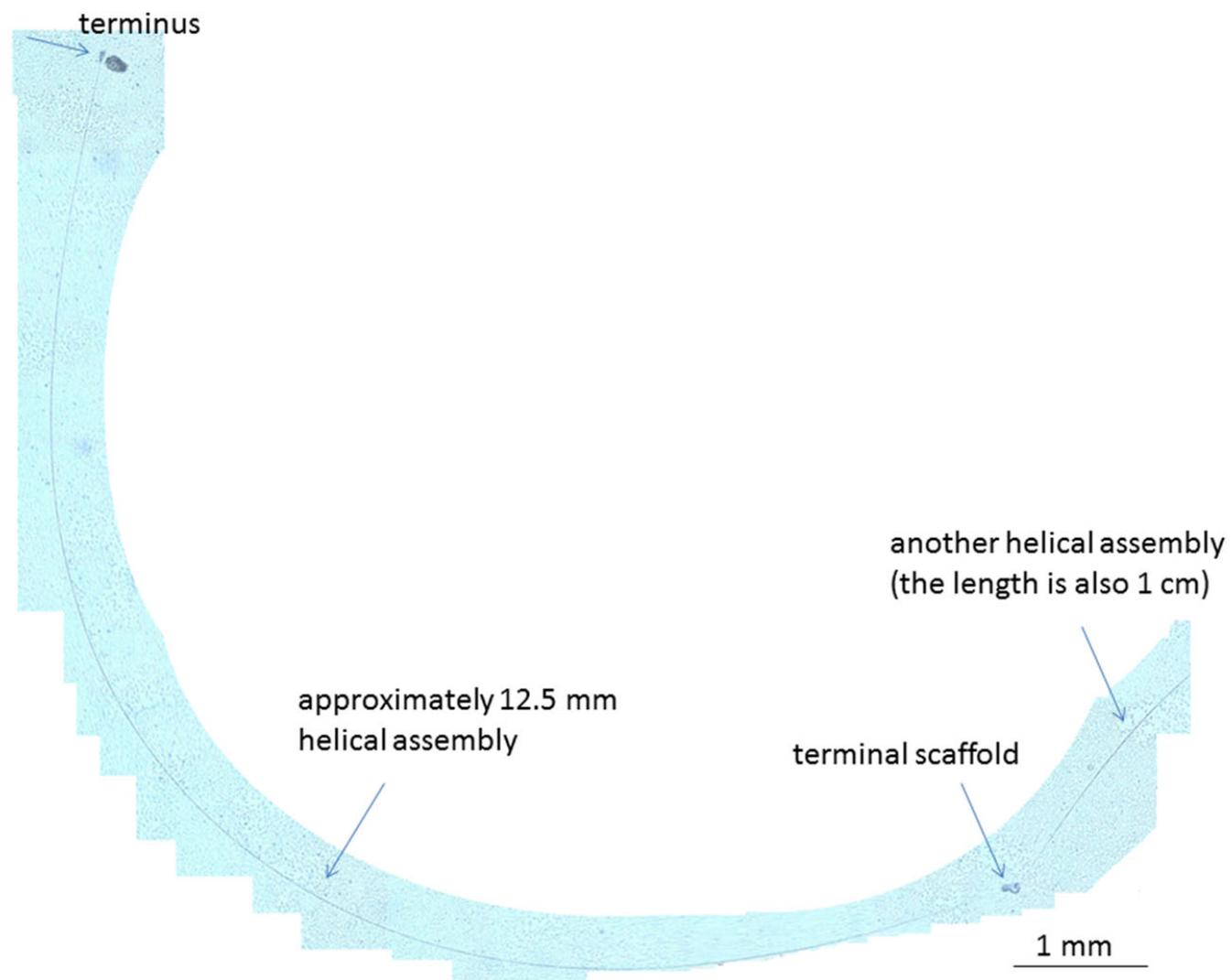


Figure S2. Approximately 12.5 millimeter-long helical assembly of oleate in the presence of 0.1% (w/w) **1** in bicine-buffered water, observed after incubation for 2 days.

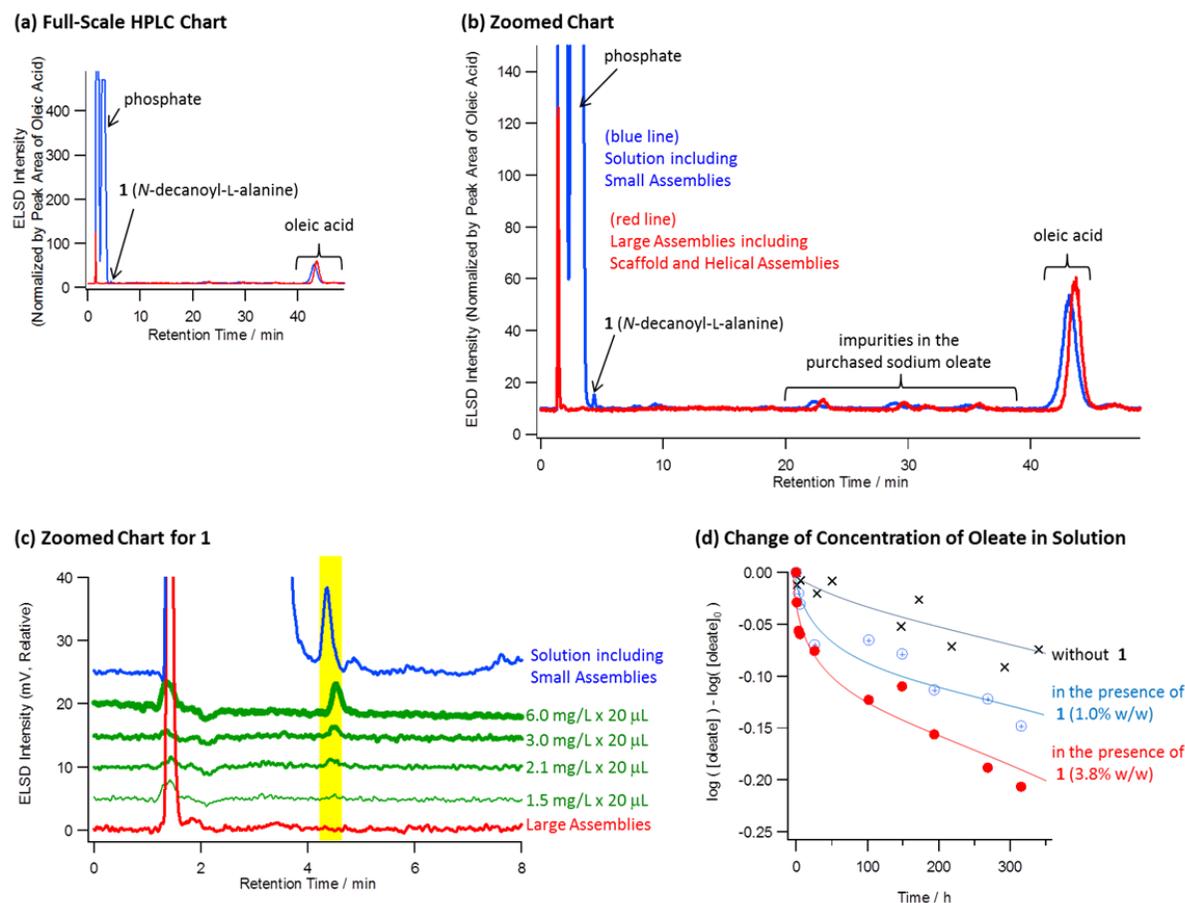


Figure S3. (a-c) HPLC charts (MeOH:0.05%TFA water = 78:22) for the aqueous phase, including small assemblies and monodispersed molecules (blue), and large assemblies (red). The large assemblies were separated from the dispersion and dissolved in MeOH. The sample dispersion was prepared by mixing ONa (2.4 mg) and **1** (0.031 mg) in 1.2 mL of phosphate buffer. Green lines in (c) are HPLC charts for variable concentrations of a MeOH/water solution of **1**. The peak of **1** was not detected in the large assemblies. (d) Change of the concentration of oleate in the aqueous solution phase including small assemblies in the absence of **1** and in the presence of 1.0%(w/w) and 3.8%(w/w) of **1**. The sample dispersions were prepared using a pH 7.6 phosphate buffer. The concentration values were calculated by HPLC (MeOH:0.05%TFA water = 90:10). The lines were drawn freehand. This result indicates **1** promotes the assembling of oleate to form large assemblies.

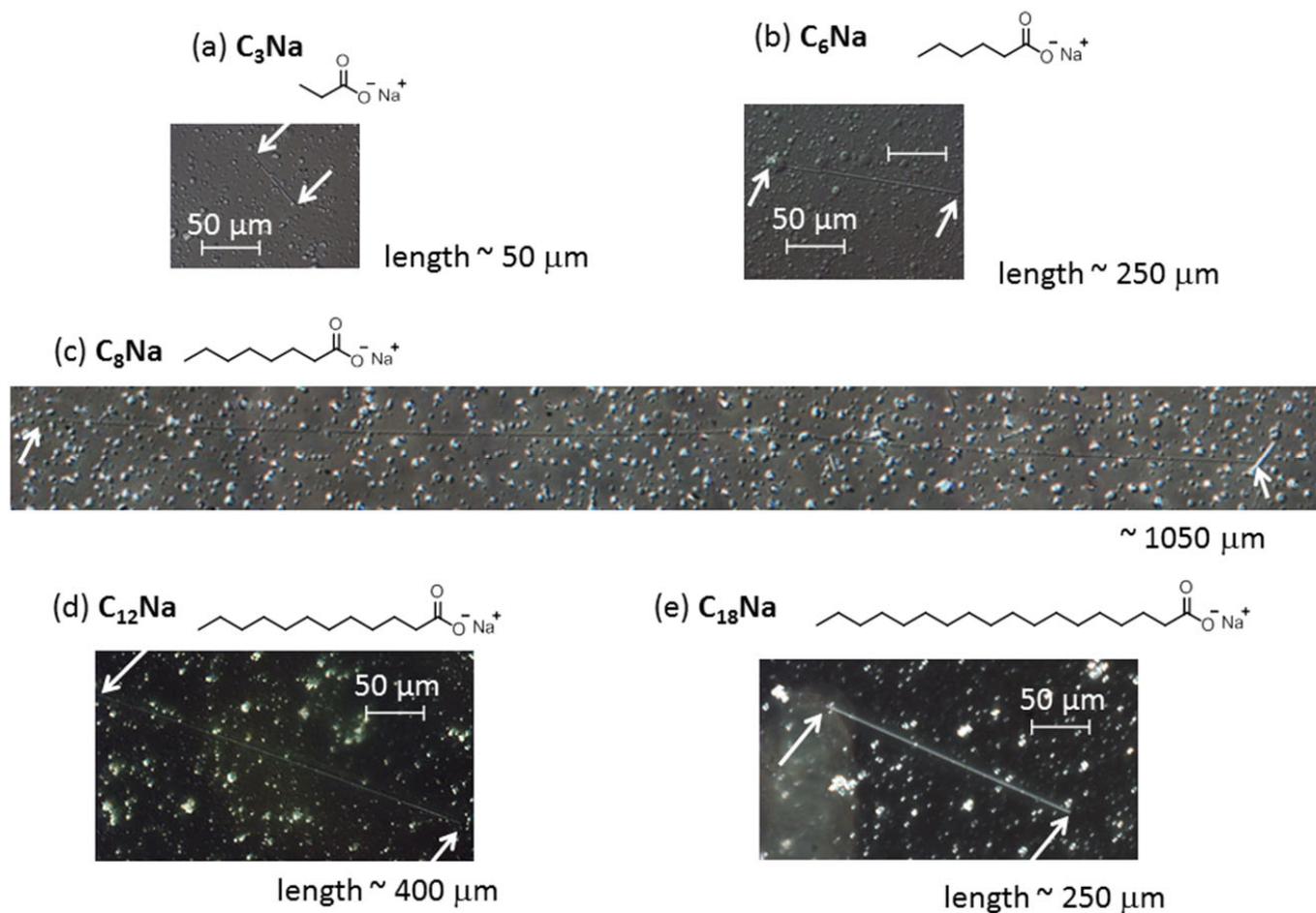


Figure S4. Helical assemblies of oleates in the presence of fatty acids. (a) 50 micrometer-long assembly in the presence of C_3Na (1% (w/w)); (b) 200 micrometer-long assembly in the presence of C_6Na (1% (w/w)); (c) 1 millimeter-long assembly in the presence of C_8Na (1% (w/w)); (d) 400 micrometer-long assembly in the presence of $C_{12}Na$ (1% (w/w)); (e) 250 micrometer-long assembly in the presence of $C_{18}Na$ (1% (w/w)). Millimeter-length assemblies were not observed.

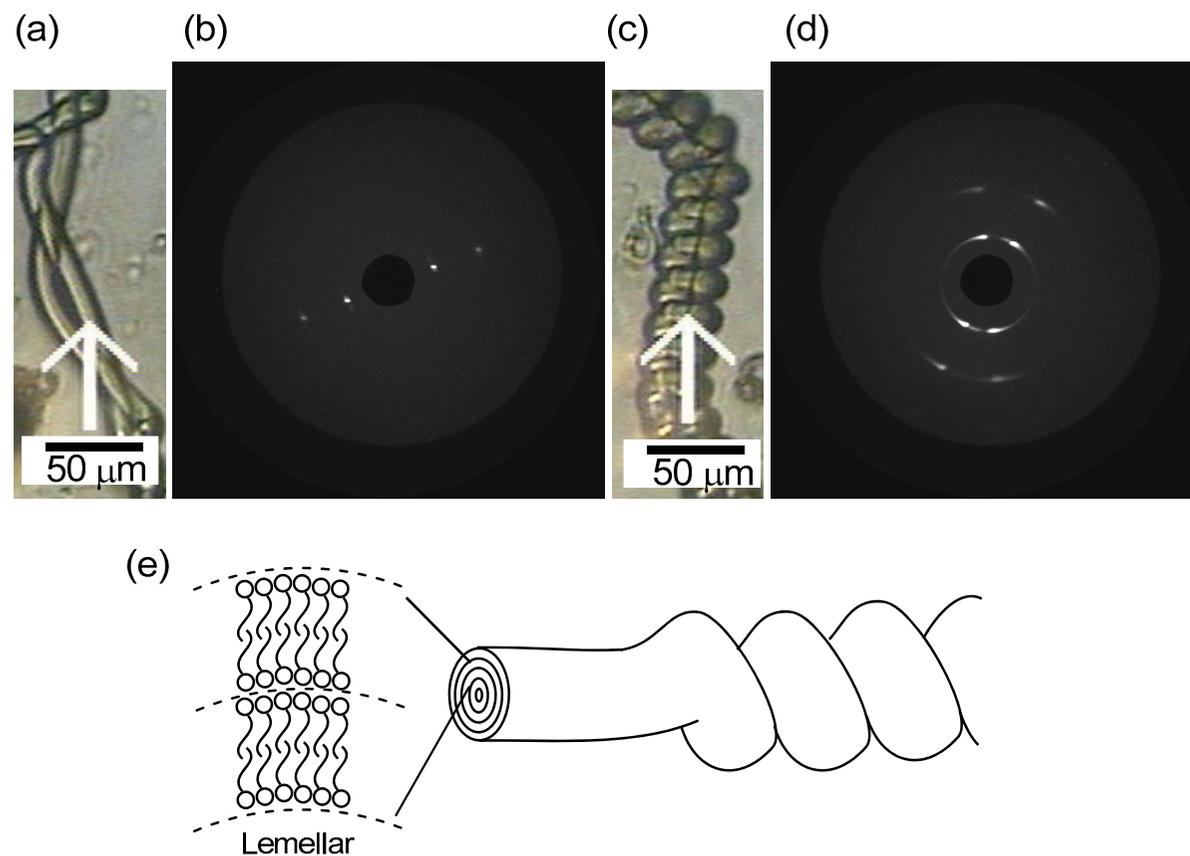


Figure S5. (a)-(d) Site-selective SAXS for DOPC myelin figures, obtained by using synchrotron microbeam X-ray diffraction (KEK PF BL-4A). (a) Optical micrograph of a straight myelin figure and (b) its SAXS pattern image at the site indicated by the arrow in (a). (c) Optical micrograph of a helical myelin figure and (d) its SAXS pattern image at the site indicated by the arrow in (c). The ratio of the radius of the first and second fringe circles was 1:2 ($d = 6.1 \pm 0.1$ nm), and the spots were located in a pattern of rotational symmetry, typical for a multilamellar tubular liquid crystal. (e) Schematic illustration of the DOPC myelin figure.

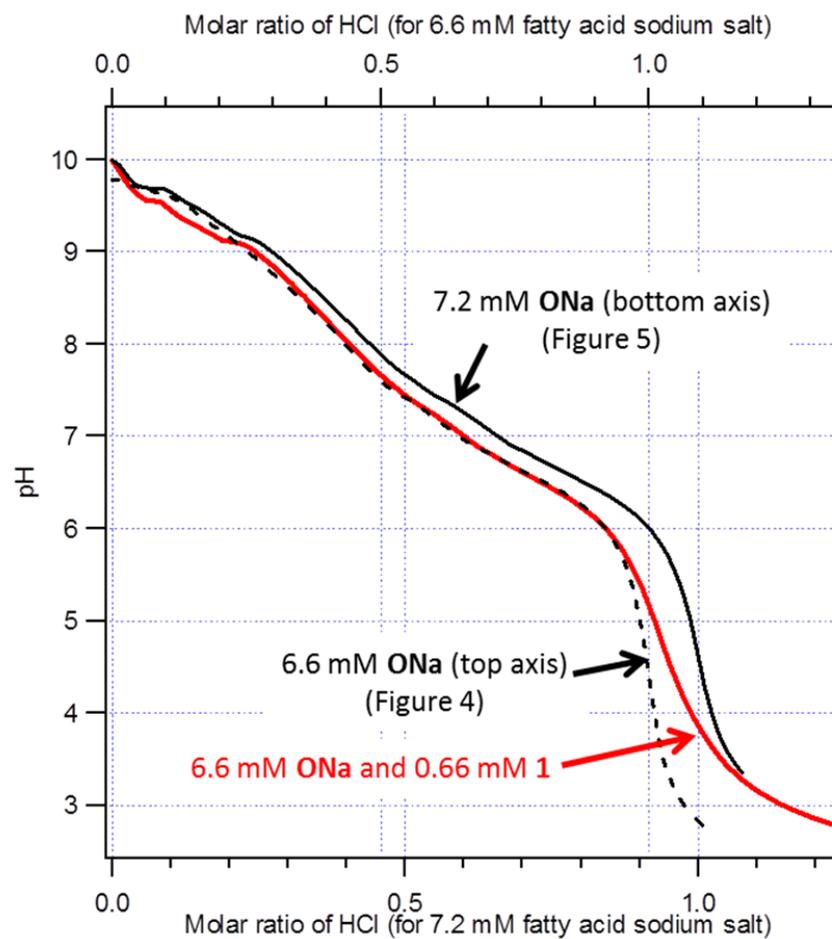


Figure S6. (red —) pH titration curve of **ONa** (6.6 mM) and **1** (0.66 mM) in 0.1 M NaCl(aq) with a titrant of 0.5 M HCl aq at 25 °C. (black —) pH titration curve of **ONa** (7.2 mM), which is shown in Figure 5. (black - -) pH titration curve of **ONa** (6.6 mM), which is shown in Figure 4.