Electronic Supplementary Material (ESI) for Soft Matter. This journal is © The Royal Society of Chemistry 2015

Electric Supplementary Information

Micrometer-sized network structure of novel DNA-lipid conjugates induced by heat stimulation

K. Takahashi,^{a,b} M. Matsuo,^a T. Banno,^{a,†} and T. Toyota*,a,c

^a Department of Basic Science, Graduate School of Arts and Sciences, The University of Tokyo,

3-8-1 Komaba, Meguro, Tokyo 153-8902, Japan. E-mail: cttoyota@mail.ecc.u-tokyo.ac.jp

^b Department of Lipid Signaling, Research Institute, National Center for Global Health and Medicine, Shinjuku, Tokyo 162-8655, Japan

E-mail: ktakahashi@ri.ncgm.go.jp

^c Research Center for Complex Systems Biology, Graduate School of Arts and Sciences, The University of Tokyo, 3-8-1 Komaba, Meguro, Tokyo 153-8902, Japan.

[†] Present address: Department of Applied Chemistry, Faculty of Science and Technology, Keio University, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama, Kanagawa 223-8522, Japan.



Fig. S1. Phase contrast microscopy images of Mg^{2+} -containing HEPES buffers including 32 μ M DNA-RNA chimera lipid **1** after incubation under different temperature. (A) The solution of DNA-RNA chimera lipid **1** in the Mg^{2+} -containing buffer exhibited no constitution of the micrometer-sized structure 20 minutes after preparation of the chamber under room temperature. Bar = 50 μ m. (B) The solution of DNA-RNA chimera lipid **1** incubated at 60°C for 20 minutes in the Mg^{2+} -containing buffer afforded similar structure observed in the case of heat stimulation of 40°C (see main text). Bar = 20 μ m. (C) In the case of incubation of the solution at 95°C for 20 minutes, the micrometer-sized network structure were constituted in more condensed manner than the case of 40°C. Bar = 20 μ m.



Fig. S2. Collapse of the micrometer-sized structures composed of each DNA-lipid conjugate after TBE buffer injection. (A) Phase contrast microscopy images of the Mg²⁺-containing buffer solutions (20 μ L of 50 mM HEPES buffer including 10 mM MgCl₂) containing the structure of DNA-RNA chimera lipid **1** incubated at 40°C for 15 minutes before and after 5 μ L 1 × TBE buffer injection. (B) Phase contrast microscopy images of the Mg²⁺-containing buffer solutions (20 μ L of 50 mM HEPES buffer including 10 mM MgCl₂) containing buffer solutions (20 μ L of 50 mM HEPES buffer including 10 mM MgCl₂) containing the granules of 1:1 mixture of DNA-lipid conjugate **2** and **3** incubated at room temperature before and after 5 μ L 1 × TBE buffer injection. Bar = 20 μ m. The collapse of the structures by the chelating for the Mg²⁺ ions by TBE buffer (including EDTA) implied that micrometer-sized structures of DNA-lipid conjugates were constituted in the presence of Mg²⁺.



Fig. S3. Phase contrast microscopy image of the sample dispersion of 1 in the Mg²⁺-containing buffer incubated at 23 ± 1 °C for 5 days.



Fig. S4. Phase contrast microscopy images of the sample dispersion of **1** in the buffer without Mg^{2+} incubated at 40 °C for 30 min and cooled down at room temperature for 30 min. The sample dispersion was observed before (A) and after (B) the injection of a small amount of the $MgCl_2$ solution (the final concentration in the sample was 10 mM). Bar = 20 μ m.



Fig. S5. Phase contrast microscopy images of Mg²⁺-containing HEPES buffers including 1:1 mixtures of each oligo-A and oligo-T based DNA-lipid conjugate with oleoyl chains. The total concentration in each mixture was 32 μ M. (A) A8 and T11 (B) T8 and A11 (C) A8 and A11 (D) T8 and T11 (E) A8 and T8 (F) T11 and A11. Bar = 50 μ m. Oligo-A based DNA-lipid conjugate A8 and A11 had the following sequences; 5'-AAAAAAAA-3' with the 5' terminus modified by amide bonds linked to oleic acid, and 5'-AAAAAAAAA-3' with the 3' terminus modified by amide bonds linked to oleic acid. Oligo-T based DNA-lipid conjugate T8 and T11 had the following sequences; 5'-TTTTTTTT-3' with 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, and 5' terminus modified by amide bonds linked to oleic acid, respectively.

Description of movie clip

Movie S1. Movie of the induction of a micrometer-sized three-dimensional network structure following heat stimulation (40°C) of DNA-RNA chimera lipid **1** in the Mg²⁺-containing buffer (Figure 4A in main text). Movie was generated from phase contrast images taken with a time interval of 1 frame every 30 s. The granular structures appeared in the specimen 10 minutes after start of heat stimulation. The size of the observation space is 296 μ m × 223 μ m.