Time-development morphological changes of self-aggregates of amphiphiles

Tadashi Sugawara,[1,2] Taro Toyota,[3] Kensuke Kurihara,[2] Kentaro Suzuki[1,2]
[1]Research Center of Life Science as Complex Systems, [2]Dept of Basic Science, The Univ. of Tokyo, Komaba 3-8-1, Meguro, Tokyo 153-8902, Japan, [3]Dept. of Engineering, Chiba Univ. Yayoi 1-33, Inage, Chiba, Chiba 263-8522, Japan.
e-mail for presenting author: suga@pentacle.c.u-tokyo.ac.jp

Morphological transformations of multi-component vesicles can be understood by the change in the average curvature of a membrane. The curvature of vesicular membranes depends on the average packing parameters of the individual components as rationalized by Tanford's theory. We constructed such a dynamic system using amphiphile **1** [1]. Since the hydrophilic site of **1** is connected with a hydrophobic chain by the imine bond that is located in proximity to the interfacial site, it is hydrolyzed to afford electrolyte **2** and lipophile **3**, even though **1** forms a self-aggregate in water. Whereas electrolyte **2** is dissolved in an aqueous phase, lipophile **3** remains in lipid layer of the aggregate, increasing the average packing parameter as the hydrolysis of **1** proceeds. As a result, it turned out that the morphology of the bi-component membranes changes from a spherical micelle, a string micelle, a needle-like structure, a giant vesicle, a tubular giant vesicle, and a nested multi-lamella vesicle (Figure 1a).

A defined morphology of the self-aggregate can also be constructed by arranging a dispersion containing an appropriate composition of **1**, **2** and **3**. In fact, when two kinds of dispersions of **1**, **2** and **3** with compositions of (10 : 0 : 0) and (5 : 5 : 5), respectively, are added to a mixing chamber through two opposite ends, corresponding morphologies appear along the linear chamber (Figure 1b).

This finding is, in particular, interesting from the aspect that the morphologies appearing in the phase diagram of self-aggregates of amphiphiles emerge successively along the time period and that the morphological transformation from micelles to giant vesicles with bilayer membranes takes place sequentially in the same dispersion.

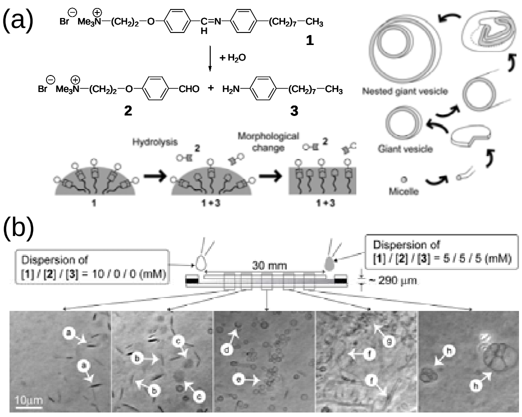

Figure 1 Morphological evolution induced by hydrolysis of an amphiphile.

Another topic is self-reproduction of giant vesicles containing catalyst caused by the addition of a membrane precursor. We have constructed a robust self-reproducing giant vesicular system in which giant multi-lamella vesicles (**GV**s) containing acid catalysts **C** in their membranes [2]. Membrane precursor **V*** is a bola-type amphiphile in which a cationic head group is connected with a membrane unit through the imine bond. When **V*** is hydrolyzed to afford electrophile **E** and membrane molecule **V**, **GV** becomes corpulent since **V** dissolves into the membranes. It dived into two **GV**s with almost the same size by the effect of **E**. As a result the number of **GV**s increases (Figure 2).

In order to analyze a massive amount of self-reproducing giant vesicles statistically, we carried out a population analysis on a self-reproducing system containing fluorescent catalyst **C** in terms of flow cytometry [3]. We found that a new population of GVs appeared after addition of membrane precursor **V*** to the dispersion of **GV**s. The size-distribution of the reproduced vesicles was similar to that of the original, whereas the content of the catalyst was decreased. The result indicated that the self-reproduction of GVs underwent repeatedly in a mass scale.

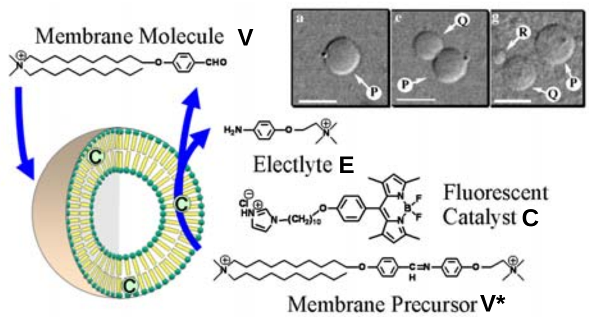

Figure 2 Self-reproducing GV system.

[1] T. Toyota, K. Takakura, J. Kose, T. Sugawara, *ChemPhysChem* ***7***, 1425 (2006).
[2] K. Takakura, T. Sugawara, *Langmuir* ***20***, 3832 (2004).
[3] T. Toyota, K. Takakura, Y. Kageyama, K. Kurihara, N. Maru, K. Ohnuma, K. Kaneko, T. Sugawara *Langmuir, in press*.