Voronoi-like Structure Consisting of Continuous Phospholipid Membranes

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Voronoi structure is a mathematically defined geometric pattern (Figure 1) and similar structures are widely found in nature. Due to the characteristic physical properties derived from the irregularity of the shape of the constituent compartments, the structure has been artificially constructed for various applications such as artificial bone tissue[1] and building skin.[2] However, materials and methods that can be used are very limited, and the bottom-up approach of molecular self-assembly has never been applied to the construction of Voronoi structure. Therefore, only static Voronoi structures have been obtained, hindering full exploration and utilization of this unique structure.

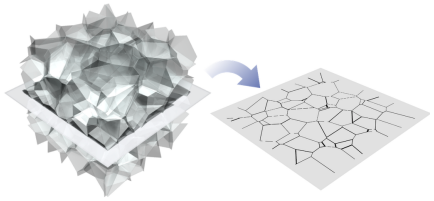

Figure 1. Schematic illustration of 3D Voronoi structure with closed-cell foams and its cross section.

In the present research, we successfully constructed three-dimensional (3D) Voronoi-like structure (Figure 2b and 2c) consisting of a phospholipid DOPEGu, which carries a guanidinium (Gu^+) ion as a head group (Figure 2a). We found that the Voronoi-like structure comprises continuous phospholipid membranes, where the phospholipids are intermolecularly salt-bridged between its Gu^+ ion and phosphate ester. Furthermore, the aqueous phase containing 3D Voronoi-like structure gelates even at a low concentration of the phospholipid (~0.1 wt%). Of interest, the continuous phospholipid membrane allows for the diffusion of lipophilic molecules over the Voronoi-like structure (Figure 2c).

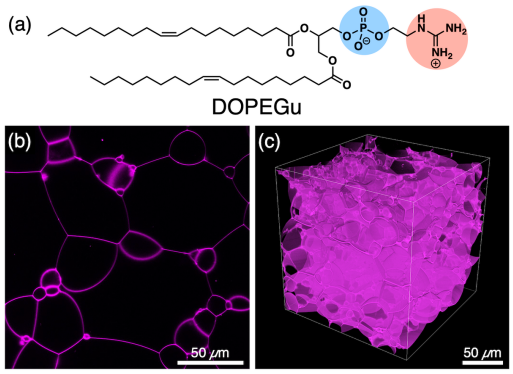

Figure 2. (a) Molecular structure of DOPEGu. (b, c) (b) 2D image and (c) 3D image (side view) of confocal laser scanning microscopy (λ_{ex} = 552 nm) of DOPEGu hydrogel after addition of a fluorescent dye.

1) S. Gómez et al., *Acta Biomater.* **2016**, *42*, 341.
2) G. Angelucci et al., *Front. Built. Environ*. **2018**, *4*, 78.