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Unveiling the Self-Assembly Behavior and Salt-Induced Morphological Transition of Double PEG-Tailed Unconventional Amphiphiles

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Abstract : PEG-based amphiphiles are of tremendous importance for its widespread applications in pharmaceutics, household purposes, and drug delivery. Previously, a number of single PEG-tailed amphiphiles having significant applications have been reported from our group. Therefore, it was of immense interest to explore the properties and application potential of PEG-based double tailed amphiphiles. Herein, for the first time, two novel double PEG-tailed amphiphiles having different PEG chain lengths have been developed. The self-assembly behavior of the newly developed amphiphiles in aqueous buffer (pH 7.0) was thoroughly investigated at 25 oC by a number of techniques including, 1H-NMR, and steady-state and time-dependent fluorescence spectroscopy, dynamic light scattering, transmission electron microscopy, atomic force microscopy, and isothermal titration calorimetry. Despite having two polar PEG chains both molecules were found to have strong tendency to self-assemble in aqueous buffered solution above a very low concentration. Surprisingly, the amphiphiles were shown to form stable vesicles spontaneously at room temperature without any external stimuli. The results of calorimetric measurements showed that the vesicle formation is driven by the hydrophobic effect (positive entropy change) of the system, which is associated with the helix-to-random coil transition of the PEG chain. The spectroscopic data confirmed that the bilayer membrane of the vesicles is constituted by the PEG chains of the amphiphilic molecule. Interestingly, the vesicles were also found to exhibit structural transitions upon addition of salts in solution. These properties of the vesicles enable them as potential candidate for drug delivery.

Keywords: double-tailed amphiphiles, fluorescence, microscopy, PEG, vesicles

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