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Magnesium Tolerance and Preferential Selectivity of A Lipid in Binary Lipid Systems: An Evolutionary Approach to Modern Membranes

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Introduction: Early compartmentalization of prebiotic molecules to form protocell membranes is considered an important process for the emergence of life. These protocell membranes are assumed to be composed of single chain amphiphiles (SCAs) due to their prebiotic availability [1]. However, SCA membranes are known to be unstable in the presence of dissolved divalent cations [2, 3], which would have been prevalent on young Earth. Eventually, membranes evolved to more robust phospholipid membranes. Here, we investigated the transition of SCAs to mixed SCA-phospholipid membranes and to phospholipid membranes in the presence of Mg^{2+}. Mg-tolerance of vesicles composed of a SCA, oleic acid (OA) and a phospholipid, palmitoyl-2-oleoylphosphatidylcholine (POPC) was examined. The concentration of magnesium needed to disprupt ~ 100 % of the intact bilayers was defined as the fatal magnesium concentration ($[Mg^{2+}]_{fatal}$). We found that Mg^{2+} acts as an environmental selection pressure for membrane evolution from SCAs to pure phospholipid membranes and the mechansims were identified.

Methods: $[Mg^{2+}]_{fatal}$ for pure OA, pure POPC, and mixed OA-POPC at various stoichiometric ratios (10:1, 5:1, 3:1, and 1:1) in the prescence of Mg^{2+} was determined by different analytical methods namely; fluorescence assay, dynamic light scattering, zeta potential (ζ) measurements, HPLC, and optical microscopy. Quantitative estimation of OA and POPC in the vesicle was achieved by HPLC after the removal of Mg^{2+}-lipid aggregates by filtration through 0.22 μm pore filters.

Results and Discussion: Interestingly, the $[Mg^{2+}]_{fatal}$ increased dramatically with an increase in the POPC content of the vesicle. For instance, $[Mg^{2+}]_{fatal}$ was 3-5 mM for pure OA, ~ 22.5 mM for OA-POPC (10:1), ~ 30 mM for OA-POPC (5:1), and > 40 mM for OA-POPC (1:1) and pure POPC, at 2 mM total lipid concentration. Increasing POPC (zwitterionic) content decreased the relative negative charge density of the vesicles as indicated by the zeta potential measurements, therefore more Mg^{2+} was required to destabilize the vesicles. Further, the relative distribution of OA/POPC in vesicles in the presence of magnesium had significantly decreased in comparison to the original vesicles. Thus, it could be inferred that magnesium preferentially binds to and abstracts OA from the mixed OA-POPC vesicles which results in lower [OA]/[POPC] ratio as compared to the starting bulk ratio. This is the first report on the evolution of fatty acid membranes towards phospholipid-enriched membranes driven by a divalent ion. Increased robustness and greater immunity of SCA-phospholipid vesicles against magnesium may hold implications in assisting Mg^{2+}-poromted processes such as RNA polymerization.

References:

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