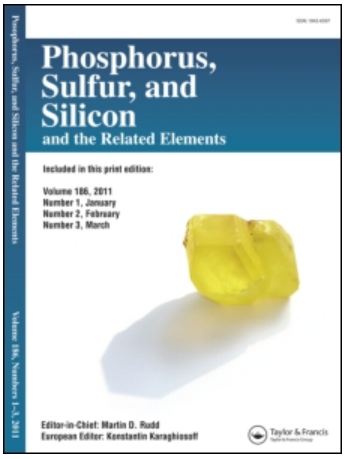

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Polyprenyl Phosphates: Do they Form Vesicles, Like Natural Phospholipids?

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POLYPRENYL PHOSPHATES: DO THEY FORM VESICLES, LIKE NATURAL PHOSPHOLIPIDS ?

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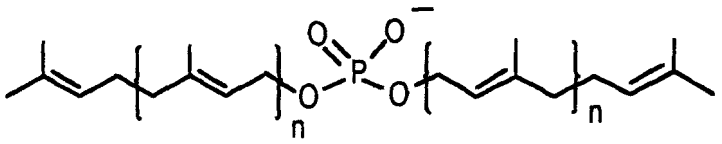
Abstract By sonication of sodium di-polyprenyl phosphates, spherical self-organized structures have been obtained. They are plausible models of primitive biomembranes.

INTRODUCTION

Discussions of molecular aspects of prebiotic chemistry tend to be focussed mostly on the origin of proteins and nucleic acids. They are remarkably superficial, or even totally silent, on the prebiotic origin of membranes.

One of us[1] and Wächtershäuser[2] have independently proposed a theory concerning prebiotic steps in biomembrane formation: the prenyl phosphate - prenyl phosphate condensation (isoprenyl unit elongation) might occur on the surface of a clay or other minerals and thus di-polyprenyl phosphates might form spontaneously and self-organize into vesicles.

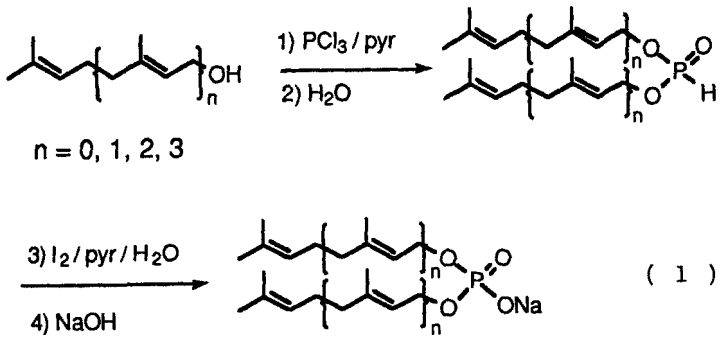


n=0, 1, 2, 3

1

SYNTHESIS

We have synthesized di-monoprenyl (n=0) or di-polyprenyl (n=1, 2, 3) phosphates from their phosphites with iodine as oxidant, according to Eq.(1).



(1)

1

Total yield is 50 -70 % (except for DDMAPNa : 10%). The free acid is not stable. Its sodium salt was prepared and found quite stable, as such or in water solution. The structures of five di-isoprenyl phosphates (DIPPNa: sodium di-isopentenyl phosphate, DDMAPNa: sodium di-dimethylallyl phosphate, **1**, n = 0, DGPNa: sodium di-geranyl phosphate, **1**, n = 1, DFPNa: sodium di-farnesyl phosphate, **1**, n = 2, DGGPNa: sodium di-geranylgeranyl phosphate, **1**, n = 3) were confirmed by ^{1}H-NMR, FAB-MS, ^{31}P-NMR and microanalysis (Ca salt).

PHYSICAL PROPERTIES

Differential scanning calorimetry showed no endothermic phase transitions (5-70°C) for these phosphates. Isothermal compression study of monolayer films obtained on a water surface from the compounds (n = 2 and 3) showed no fluid-condensed transition. The ^{1}H-NMR spectra of aqueous suspensions of the phosphates presented fairly sharp peaks. These facts show that their assemblies are fluid. The C_5 phosphates give optically homogeneous solution. The other ones gives milky suspensions.

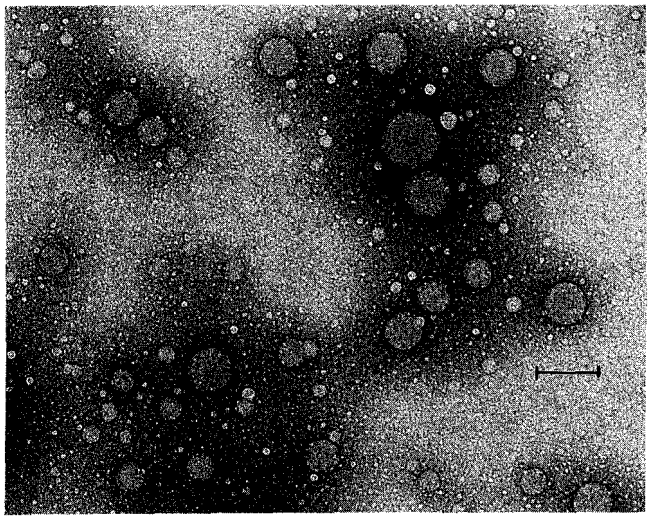
FORMATION OF SELF-ORGANIZED STRUCTURES

We have sonicated an aqueous suspension of each phosphate (Ar, 1 h). After centrifugation and extrusion through polycarbonate filters, the sample was subjected to the observation by electron microscope after staining with uranyl acetate or phosphotungstic acid. Sodium di-geranyl phosphate (C_{10}), di-farnesyl phosphate (C_{15}) and di-geranylgeranyl phosphate (C_{20}) gave almost exclusively spherical vesicular type structures with the average diameter is about 200 nm, which was confirmed by dynamic light scattering measurements.

The presence of closed vesicular systems was also demonstrated by a trapped water soluble fluorescence probe, 5-carboxyfluorescein.

Di-dimethylallyl phosphate (C_5) and di-isopentenylphosphate (C_5) are soluble in water: their lipophilic chains are too short to self-organize.

The formation of organized systems already with C_{10} chains (bilayer thickness: about 16Å) was unexpected.



Electron micrograph of the sonicated sample of sodium di-geranylgeranyl phosphate, stained by uranyl acetate. Scale : 250 nm.

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1. G. Ourisson, Pure Appl.Chem., 62, 1401-1404 (1990).
2. G. Wächtershäuser, Microbiol. Rev., 52, 452-484.(1988).