

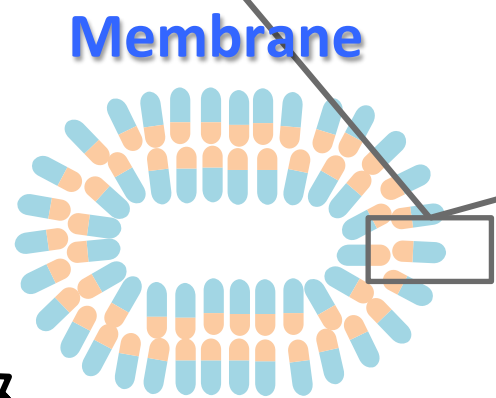
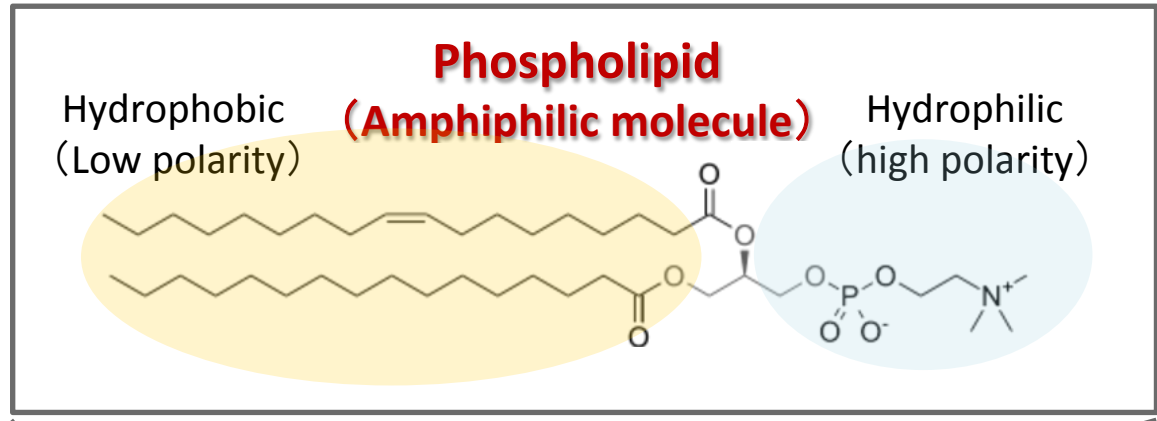
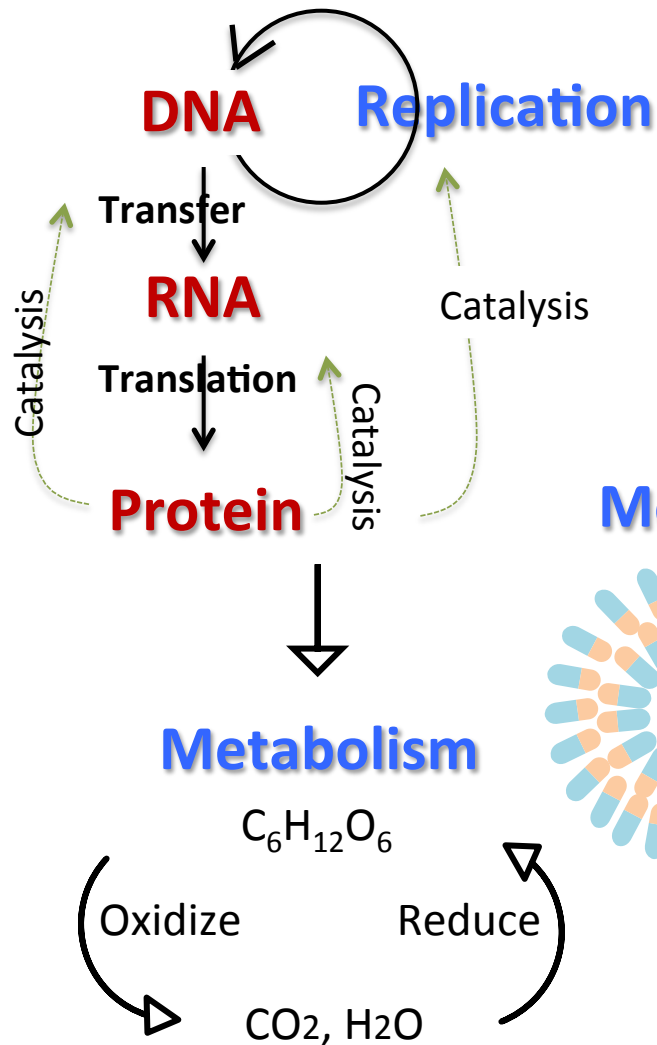
**Dynamics observation of prebiotic microspheres through their chemical and morphological distributions:
Attempt to define "Planetary Life"**

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Approach to Origins of Life

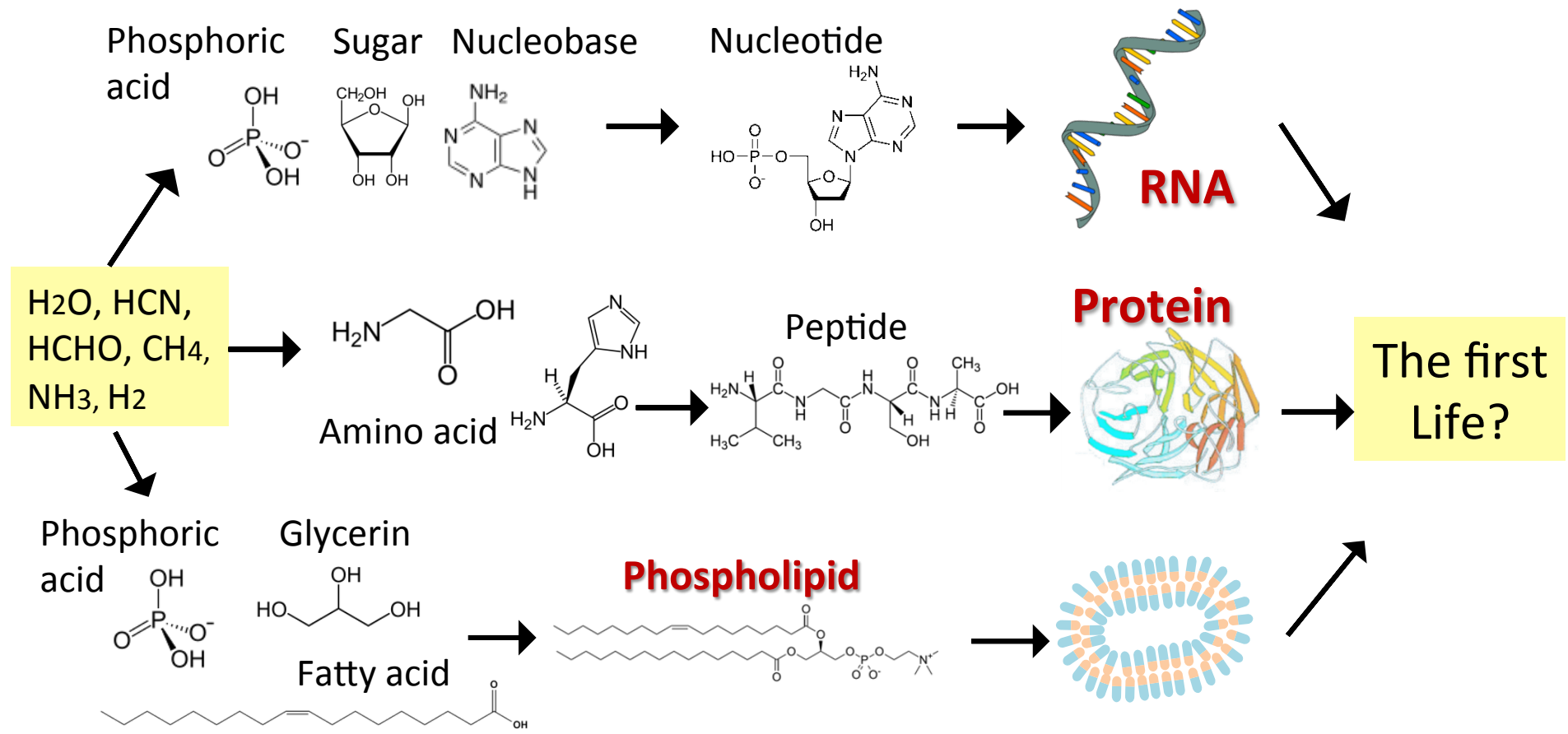
based on the definition of "the present Earth-Life"



- Protect and concentrate the inner molecules within for **facilitating their reactions**
- Maintain concentration gradients of ions and **provide a source of free energy** (Deamer et al., 2002)

The Earth Life
(Szostak et al., 2001)

Traditional idea of chemical evolution toward Origin of Life



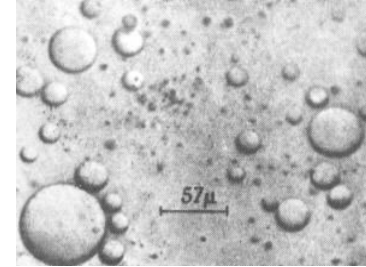
- Any prebiotic synthesis has *not* been able to demonstrate the formation pathways above (for more than 60 years since Millar’s experiment!)
- Which came first, DNA or protein? (“Chicken or the egg” problem) is still unknown

Difficult to model “the present” biomolecules

Prebiotic formation of organic microspheres

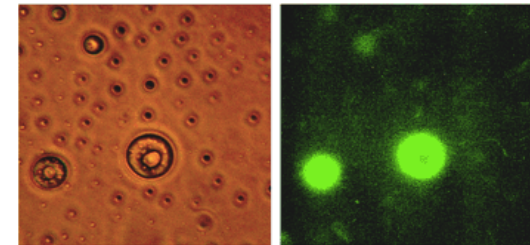
✓ **Coacervate** formed from gelatin and gum arabic (Oparin et al.,1976)

✓ **Protenoid microspheres** produced by thermal copolymerization of glutamic acid or aspartic acid (Fox and Harada ,1958)



(Fox and Harada ,1958)

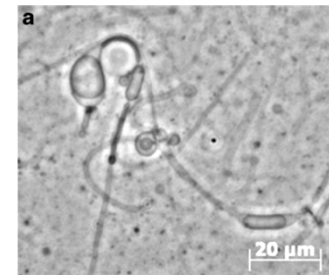
✓ **Microspheres** formed from organic extracts of meteorites and interstellar ice analogue (Deamer, 1985; Deamer and Pashley, 1989; Dworkin et al., 2001)



30 μ m
(Dworkin et al., 2001)

✓ **Acylglycerols** synthesized by condensation of fatty acid and glycerol under hydrothermal condition (Simoneit et al.,2007)

✓ **Microspheres** formed from a mixture of polycyclic aromatic hydrocarbon (PAHs) and fatty acids (Groen et al.,2012)



(Groen et al.,2012)

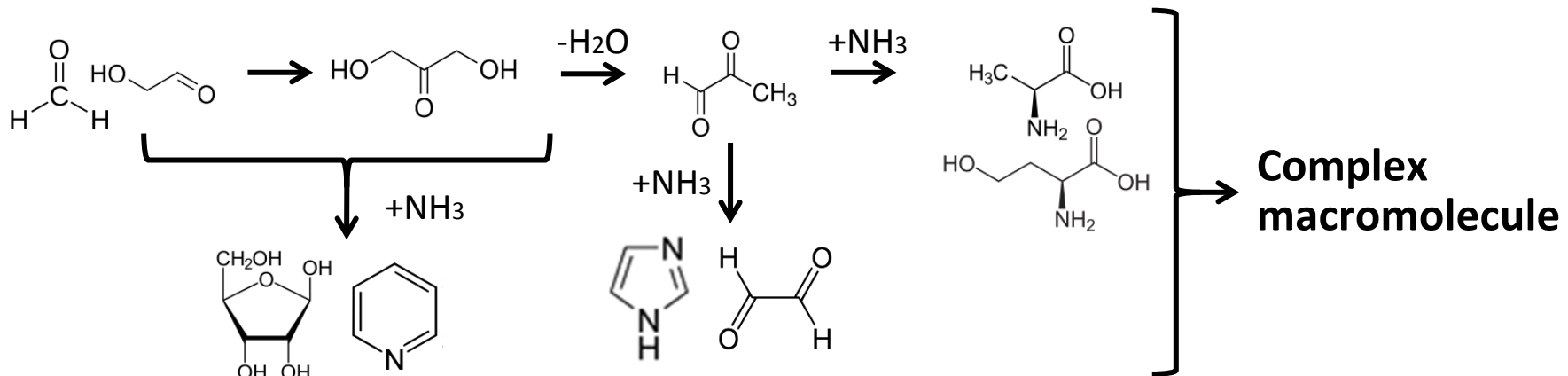
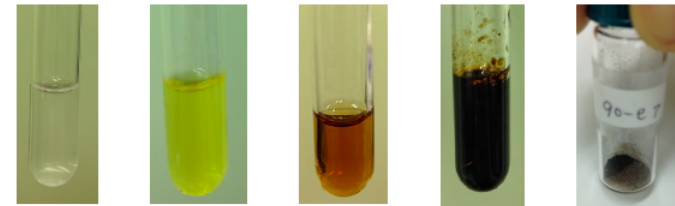
However, the cell-like functions are not observed from the materials

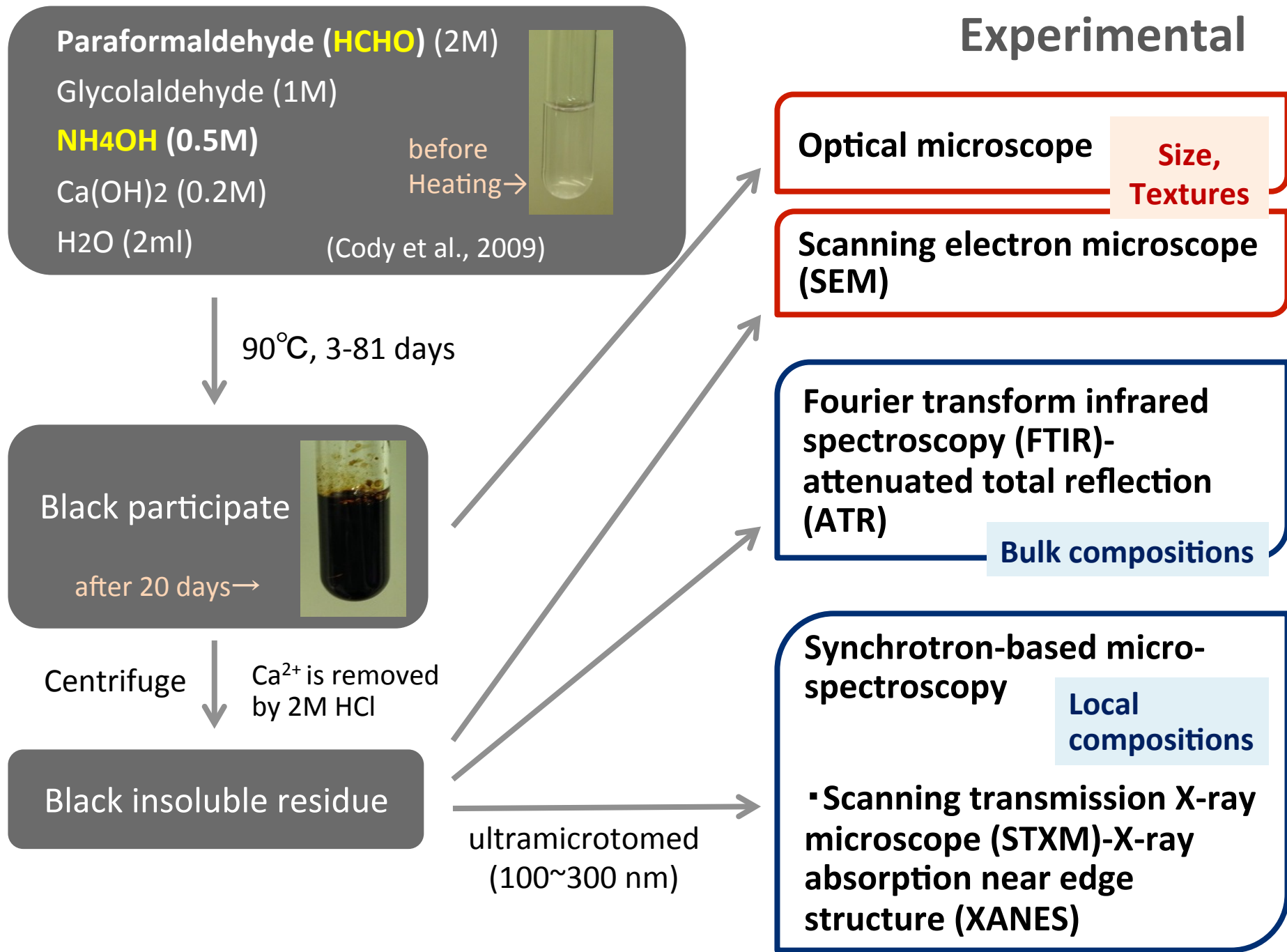
Trying to identify a boundary between abiotic materials and bio-functions

Goal in this study

In order to seek the protocell-like functions, we investigated the formation mechanisms of prebiotic organic microspheres produced by Maillard-type reaction of formaldehyde (HCHO) and ammonia (NH₃) which could have been the common molecules in the early Earth.

◆ Maillard type reaction (Weber, 2001 modified) (Browning reaction, Aminocarbonyl reaction)





Observation of solid by optical microscope: **Size distribution**

5 days (liquid)

5 days (solid)

10 days (solid)

Sorry , the results are unpublished data.

23 days (solid)

51 days (solid)

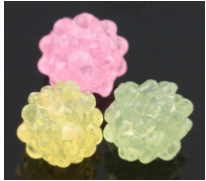
81 days (solid)

Most of the microspheres were distributed inside of the organic solid.

The size of the microspheres increased until 51 days, but after that, smaller microspheres increased (81 days).

Observation of solid by optical microscope: Morphology

5 days



10 days

51 days

Sorry , the results are unpublished data.

51 days



63 days



81 days

Observation of solid by scanning electron microscope (SEM)

10days (irregular shape)

20days (smooth surface)

51days (hollow)

Sorry , the results are unpublished data.



60days (constricted)

Size distributions of microspheres

Relative abundances (the total numbers are set as 100%)

5 days

10 days

Sorry , the results are unpublished data.

51 days

63 days

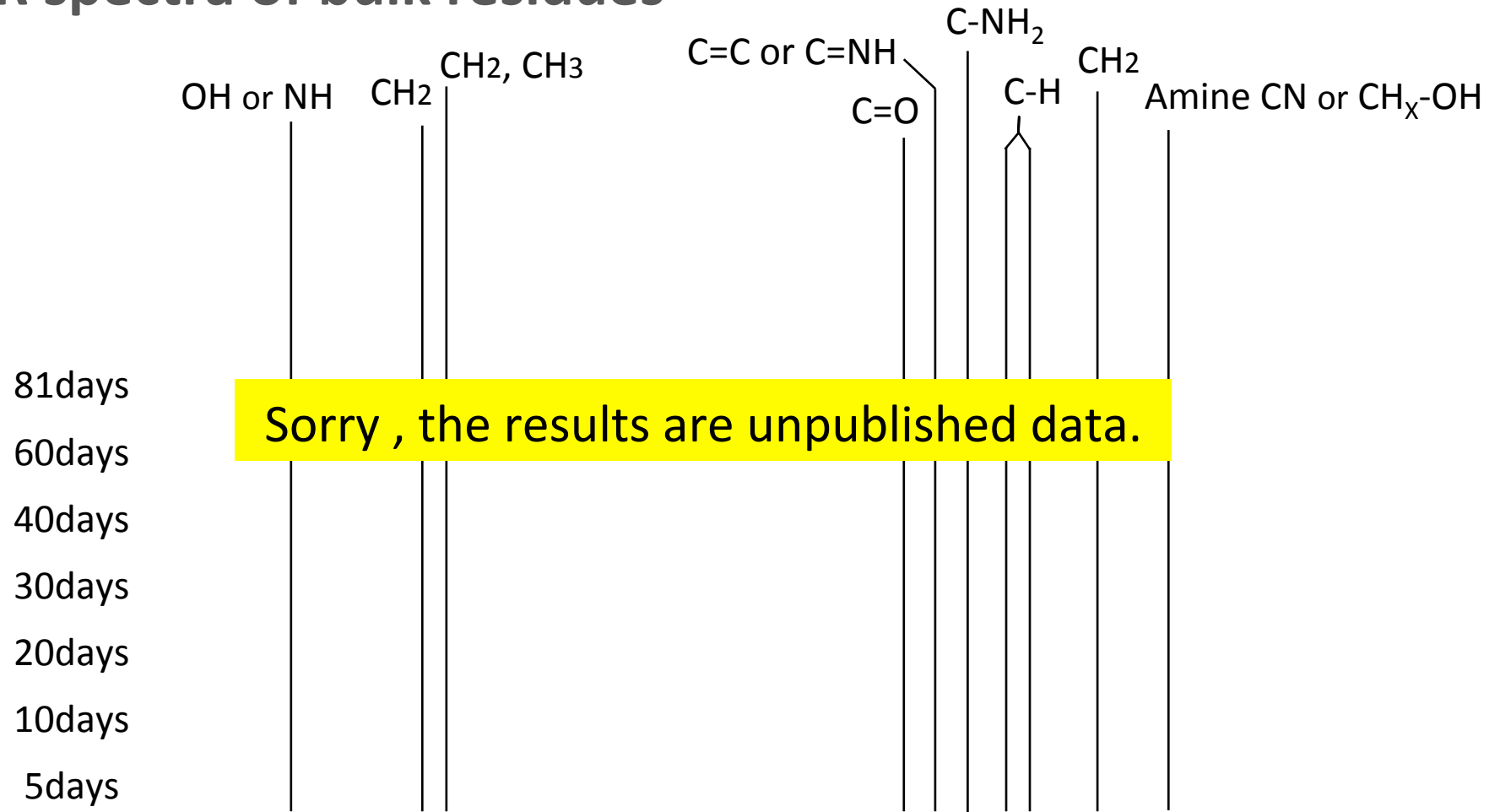
81 days

▪ Production of large microspheres is resulted from **combination** of small microspheres

▪ Re-production of smaller microspheres is due to **division** of larger microspheres

サイズ (μm)

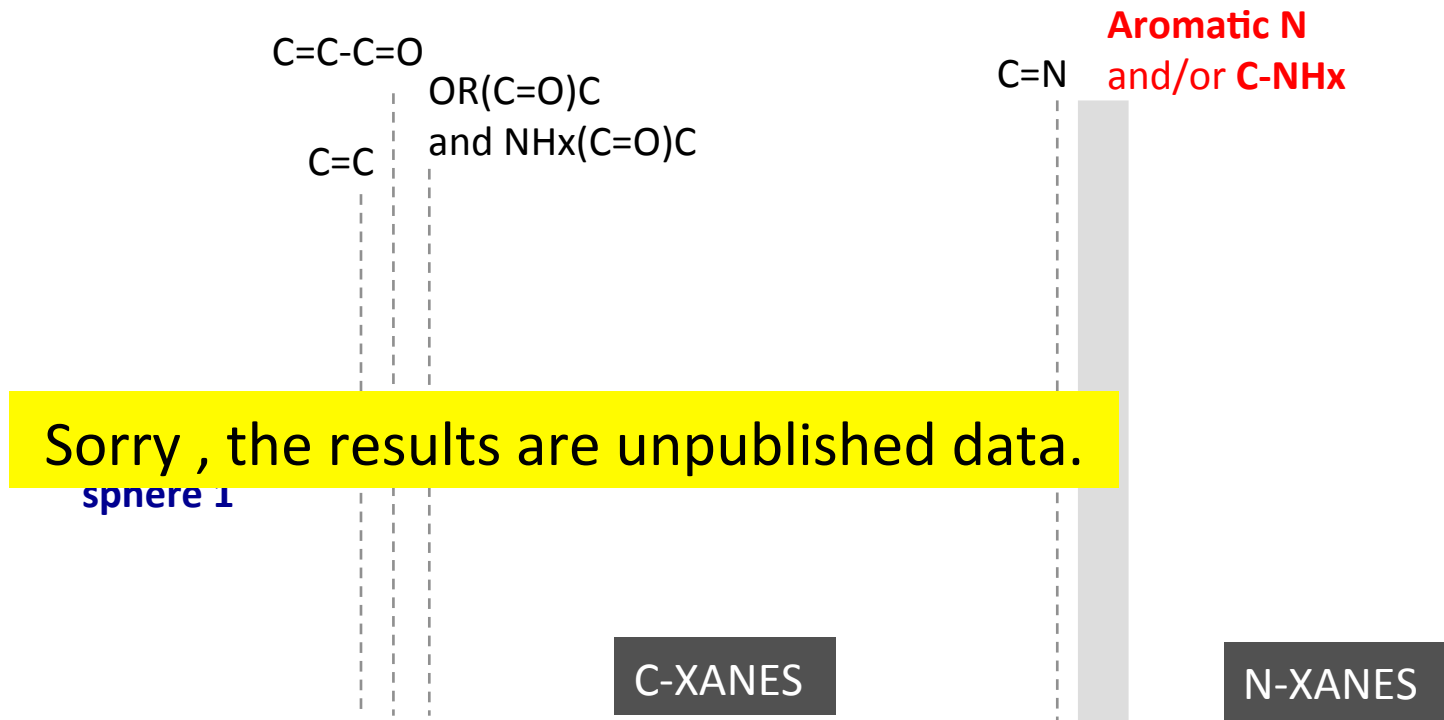
FTIR spectra of bulk residues



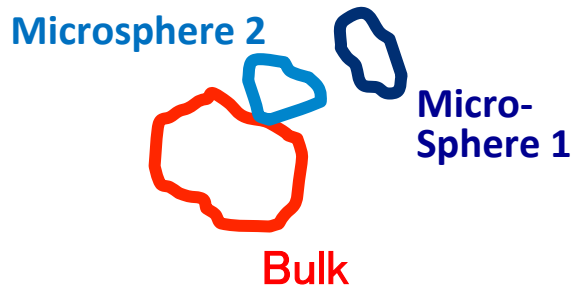
The functional group compositions were largely constant over heating time. The microspheres were formed **by self-assembly of molecules due to different polarities.**

Heterogeneity of local compositions (STXM-XANES)

90°C, 40 days



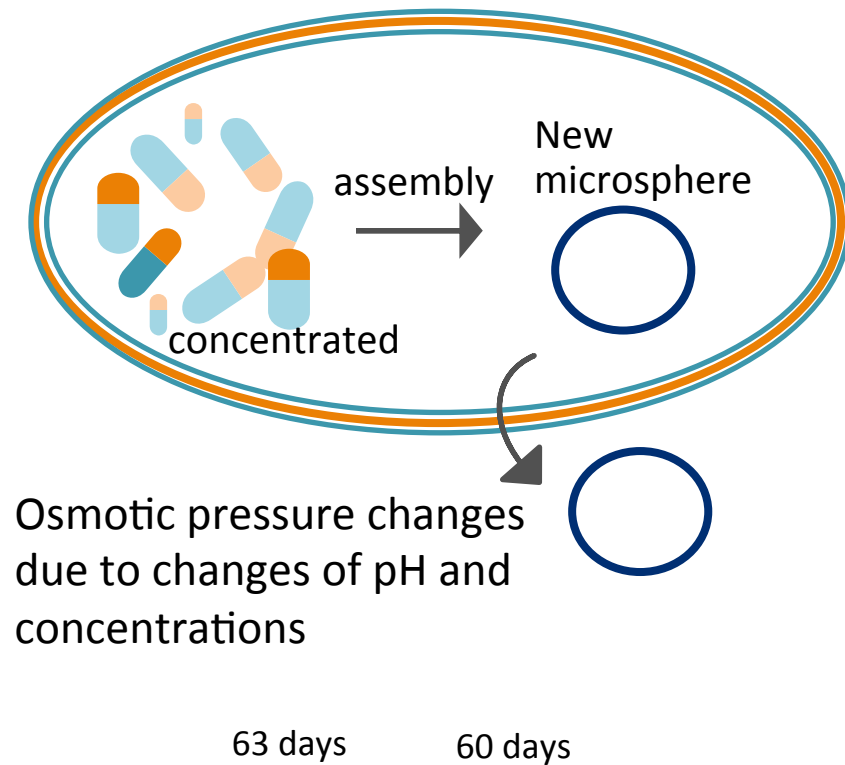
Optical image



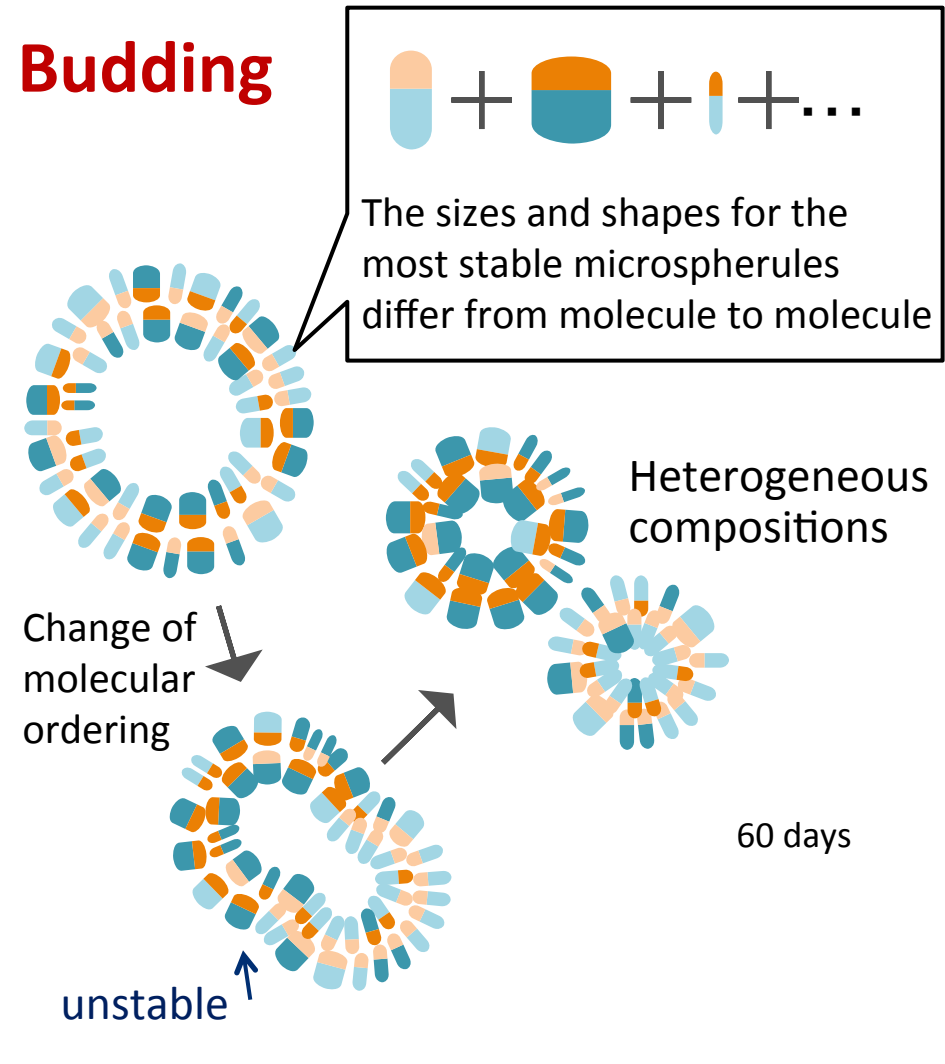
- ✓ Microspheres and host organics consist of the similar chemical compositions in most cases
- ✓ Individual microspheres have a slight extent of chemical heterogeneities

Division mechanisms of microspheres: **Self-reproducing ?**

Birthing



Budding



(Sakuma & Imai, 2011; Sakuma, et al., 2010 modified)

We suggest that this cell-like behavior of prebiotic microspheres linked to self-replication, as a definition of “Planetary Life” (which is beyond that of the Earth-life) (wishful thinking)

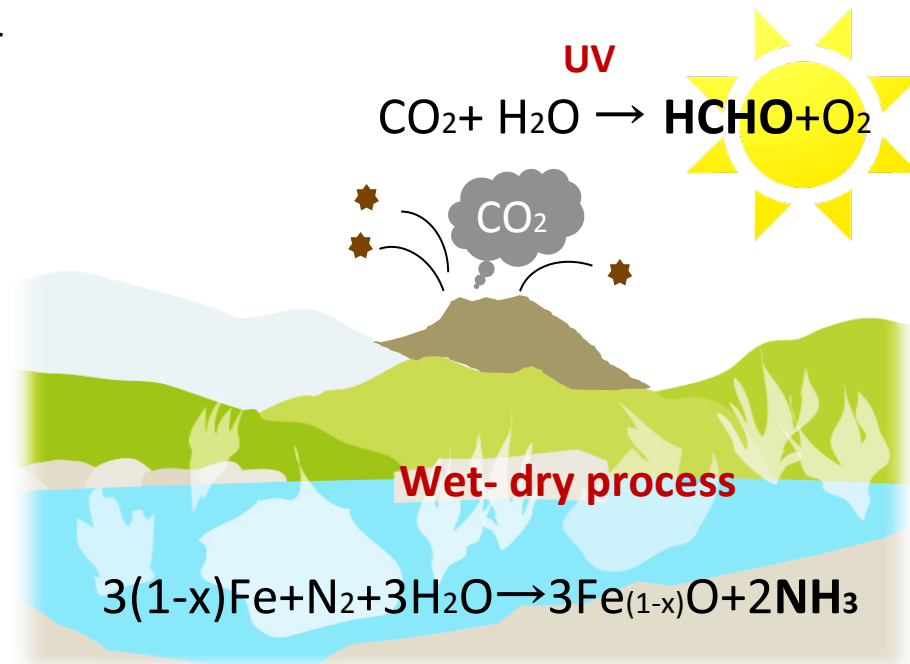
Plausible conditions for the growth and division of microspheres:

◆ Volcanic or Geothermal fields

- Concentration of materials by wet-dry cycle
- Continuous supply of materials via photochemical reaction in atmosphere and mineral catalyzed synthesis (Washington, 1999)

◆ Subduction zone

- Concentration and reactions of dissolved materials by dehydration (Holm and Neubeck, 2009)



These environments could exist not only the Earth but other planets such as Mars and Icy moons

Summary

- (1) Origins of Life is **NOT** necessarily **the so-called biomolecules or high-order functional molecules of the present Earth-life.**
- (2) Prebiotic organic microspheres synthesized in this study were formed by **self-assembly of the complex molecules.** They demonstrated a **self-reproducing-like behavior** by continuous growth and **division.**
- (3) The starting materials for formation of microspheres **do not need to be ammonia (NH₃) and formaldehyde (HCHO),** but their reaction products need to be **amphiphilic (different polarity)**
- (4) Plausible conditions for the growth and division of microspheres could be volcanic or geothermal field, or subduction zone, where **materials are continuously supplied** and **concentrated.**