

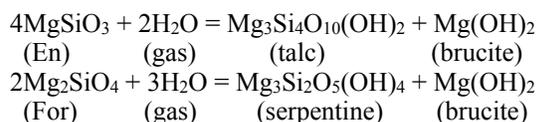
Phyllosilicate Formation and Its Role on the Formation of Organic Materials in the Early Solar Nebula.

H. Nagahara¹ and K. Ozawa¹, ¹Department of Earth and Planetary Science, The University of Tokyo, Hongo, Tokyo 113-0033, JAPAN (hiroko@eps.s.u-tokyo.ac.jp).

Introduction: There are various types of organic matters in molecular clouds, the pre-planetary systems, such as nitrile, aldehyde, alcohol, carbonic acid(s), ketone, refractory organics, and so on. Dusts in molecular clouds are thought to form a core-mantle structure, where amorphous silicate core is mantled by organics and ice at the surface [1]. The dusts are characterized by deuterium enrichment. Chondrites also contain various organic materials and amino acids, but their chemical and isotopic features are affected by thermal metamorphism on the parent bodies, which may be related to the formation of hydrous silicates and carbonates [2, 3], although rare hotspots with extreme isotopic anomalies are thought to be inherited from the molecular cloud [4, 5]. Cometary samples by Stardust contain organics, which is poorer in the aromatic carbon and variety of amino acids compared to chondritic organics [6]. No hydrous silicate was found, but extraterrestrial amino acid is reported [7].

What is unclear is the evolution of the complex dust grains of the molecular cloud origin in the solar nebula. The intimate coexistence of silicate, organics and water ice could cause various chemical reactions depending on the temperature increase in the nebula. We have investigated the chemical interaction of silicate, organics, and water at various temperature conditions to get better understanding of evolution of solid materials in the solar nebula with special interest to the evolution of organic materials on silicate.

Stability of hydrous silicates: The simplest reaction among the three components is hydration of anhydrous silicate. Fegley and Prinn [8, 9] investigated the stability of hydrous silicates and discussed that the reactions:



The crystalline phase of serpentine is chrysotile. By using a simple collision theory (SCT) and the activation energy for MgO hydration at 1 atm ($\sim 70\text{kJ mol}^{-1}$), they concluded that the reaction time scale is much longer the life time of the solar nebula, and therefore, the reaction did not take place, although serpentine plus brucite is thermochemically stable below $\sim 270\text{K}$ at $P_{\text{tot}}=10^{-3}$ bar and $\sim 230\text{K}$ at $P_{\text{tot}}=10^{-5}$ bar. Ganguly [10], on the other hand, extrapolated the experimental data for forward and backward above reactions at 1 and 3kb to the low pressure corresponding to the solar nebula condition (10^{-5} bar) by using the activation volume, which yielded $\sim 32.5\text{ kJmol}^{-1}$, and concluded that

the reactions could proceed within the evolution time scale of the nebula.

Those studies assume crystalline forsterite and enstatite as the reactants, however, as mentioned above, amorphous silicate is the plausible silicate phase as the reactant. We have made thermodynamic calculations to evaluate the equilibrium boundary of amorphous silicate and serpentine+brucite by using the thermochemical data for amorphous silicate [11]. The reaction equation is the same as those described above excepting the amorphous nature of MgSiO_3 and Mg_2SiO_4 . The reaction curves for the crystals are also calculated for comparison using the thermodynamic data by Helgeson et al. [12].

Reaction experiments: Previous workers used kinetic data either obtained at 1 atmosphere [8, 9] or extreme extrapolation of high-pressure data linearly to very low pressure [10]. Therefore, we need reaction experiments at low water vapor pressure and low temperature to obtain the kinetic data to make evaluation whether the hydration reaction proceeds within the evolution time scale of the solar nebula or not.

We have prepared silicate glass as the analog for the amorphous silicate. The glass has a composition of Mg/Si \sim 1 according to the solar abundance. For comparison, we have also prepared single crystals of forsterite, enstatite, and periclase (MgO). Two types of experiments are carried out: one is an open system reaction, where water vapor gas is flowing through a glass tube, which is continuously evacuated. The other is a closed system experiments, in which water vapor was poured into a pre-evacuated capsule with the samples. For the former experiments, samples were kept at the room temperature. For the latter experiments, temperature ranged from the room temperature to 80°C . The weight change was measured and the surface of the samples was observed with SEM-EDS-EBSD.

Dehydration experiments: In order to evaluate the stability of hydrous silicate, dehydration experiments for natural serpentine (chrysotile) and talc were conducted in the temperature range from the room temperature to 1000°C in vacuum chamber.

Results and Discussion: The results of thermodynamic calculation are shown in Fig. 1, where the hydration reaction of amorphous silicates with the composition of MgSiO_3 and Mg_2SiO_4 and crystalline forsterite and enstatite are plotted in the pressure-temperature space. It is worth noting that hydration of the amorphous silicate lies at much higher temperature portion compared to crystalline silicates; the hydration of amorphous MgSiO_3 takes place at higher temperatures by $\sim 300^\circ$ compared to crystalline enstatite, and amorphous Mg_2SiO_4 by $\sim 200^\circ$ than

crystalline forsterite. The time scale for the hydrous mineral formation from amorphous silicate has not yet been evaluated due to the experimental difficulty.

The result suggests that hydrous silicates are easily formed in the solar nebula when the amorphous dusts inherited from the molecular cloud are heated.

Dehydration rate of serpentine is rapid in the initial stage and slowed with time. Complete dehydration to attain ~12% weight loss was achieved at high temperatures above 500°C. From the time dependent mass loss rate, dehydration time scale was estimated as a function of temperature. The experimental results are used to evaluate whether the phyllosilicates formed from amorphous silicate survives in the solar nebula or not. Extrapolation of the experimental results to lower temperatures shows that serpentine can survive for the life time of the solar nebula if it is retained at temperatures as low as ~300K (Fig. 2). If we assume the total pressure of the solar nebula to be 10^{-4} bar, amorphous silicate is hydrated to be talc plus brucite at ~600K, they will further dehydrate at that temperature in an order of days. In order to retain hydrous silicates, they should be transported to cooler region of the nebula rapidly.

The hydrous phyllosilicates act as the catalyst for the organic compound formation as is well known in the terrestrial material science. The organic components in the core-mantle type dust grains would be changed into more complex form if the core amorphous silicate is converted to phyllosilicate, suggesting that organic materials can evolve in the early solar nebula before incorporation into parent bodies.

References: [1] Greenberg, J. M. (1989) *IAUS* 135, 345-355. [2] Cody, G. D. et al. (2008) *EPSL* 272, 446-455. [3] Alexander, C. M. O'D et al. (2007) *GCA* 71, 4380-4403. [4] Busemann, H. et al. (2006) *Science* 279, 727-730. [5] Cody, G. D. et al. (2008) *MAPS* 43, 353-366. [6] Sandford, S. A. (2008) *Proc. IAU Symp.* 251, 299-307. [7] Elsil, J. E. et al. (2009) *MAPS* 44, 1323-1330. [8] Fegley, B., Jr. and Prinn, R. G. (1989) in *The Formation and Evolution of Planetary Systems*, 171-211. [9] Prinn, R. G. and Fegley, B., Jr. (1989) in *Origin of the Solar Systems*, 78-136. [10] Ganguly, J. (1995) *LPS XXVI*, 441-442. [11] Golczewski, J. A. et al. (1998) *Calphad* 22, 381-396. [12] Helgeson, H. C. et al. (1978) *Amer. J. Sci.* 278A, 638-686.

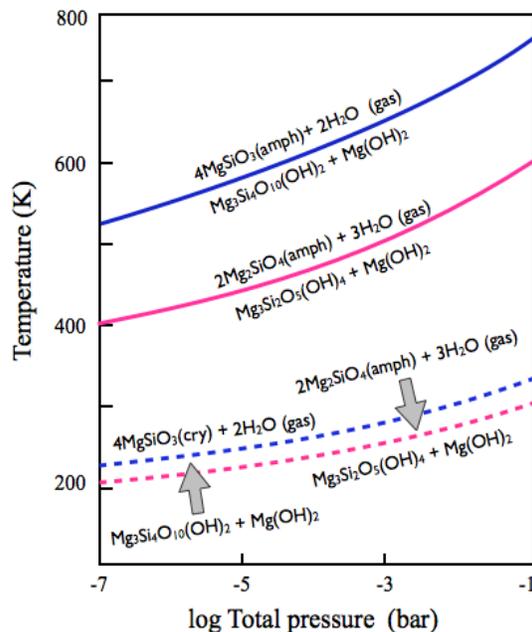


Fig. 1: Stability of hydrous silicate coexisting with amorphous or crystalline silicates in the solar nebula. The upper two curves are for the hydration of amorphous silicate (amph) with the composition of $MgSiO_3$ and Mg_2SiO_4 , and the lower two are for crystalline enstatite and forsterite (cry). Thermodynamic data by [11] and [12] were used.

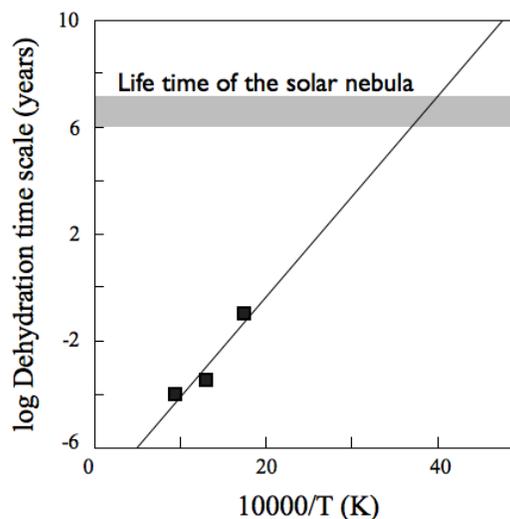


Fig. 2: Dehydration time of serpentine (chrysotile) obtained by the experiments in vacuum.