

Viscosity measurement of silicate melts under mantle conditions infers the primordial structure of Earth's silicate mantle

Owing to massive energy released by a giant Moon-forming impact, the early Earth is believed to experience large-scale melting, which forms a deep or even whole-mantle magma ocean (MO) [1]. The present-day mantle is evolved from the primordial mantle after the solidification of MO. Whether the primordial mantle is chemical-layered or not depends on the solidification type (fractional or equilibrium), which in turn depends on the suspension of crystals in a convecting MO. If the crystals can be suspended, equilibrium solidification should occur, resulting in a homogeneous primordial mantle. If the crystals can't be suspended, fractional solidification should occur, resulting in a layered primordial mantle. The solidification type of MO is therefore vital to constrain the evolutionary scheme of the mantle. Although the solidification type was investigated by geodynamic modeling, poorly constrained physical parameters prevent the geodynamic modeling from giving a definitive conclusion.

Besides the heat flux (a flow of energy per unit of area per unit of time) of MO surface, viscosity is a key parameter for geodynamic investigations [2]. The viscosity of a fluid is a measure of its resistance to deformation at a given rate. For example, honey has higher viscosity than water. *In situ* falling sphere

viscometry is the best method to directly measure viscosity under high-pressure conditions. Based on the Stoke's law (Eq. 1), the viscosity of melt is evaluated from the vertical velocity of the sphere, which is monitored through sequential radiographic images (Fig. 1).

$$\eta = \frac{2gr_s^2(\rho_s - \rho_m)}{9v_s} \quad (1)$$

where v_s , r_s , ρ_s , ρ_m and g are the terminal velocity, sphere radius, sphere density, melt density, and gravity acceleration, respectively.

Unfortunately, the viscosity measurement of silicate melts was limited to ~13 GPa (upper mantle) for more than 10 years due to technique difficulties (Fig. 1(b)). Major limitations encountered in previous works of the same type were (i) lack of X-ray transparent heating element to generate ultrahigh temperatures (more than ~2500 K) required to melt the silicate phases entirely and (ii) the extremely low viscosity of silicate melts at high pressure, requiring very fast radiographic measurements. Being refractory and highly X-ray transparent, recently developed boron-doped diamond heater [3] is a perfect heater to extend the pressure range of viscosity data. By combining

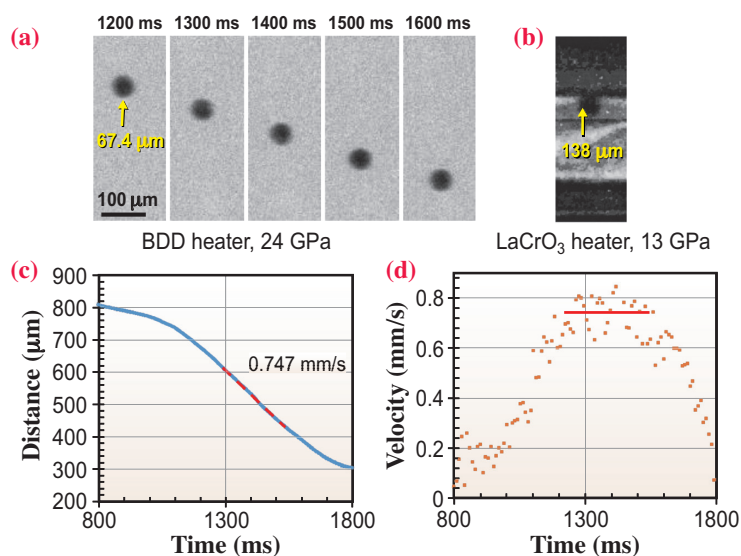


Fig. 1. Example falling path and terminal velocity of a probe sphere. (a) Selected radiographic image of a probe sphere falling in liquid forsterite taken with a frame rate of 1000 frame/s. (b) Radiographic image of a probe sphere in LaCrO₃ heater for comparison [5]. (c) Position of the sphere as a function of time according to radiographic images. The sphere position was fitted by a Gaussian function in each X-ray radiographic image (blue symbol). The melt viscosity can be calculated from the terminal velocity (red dashed line). (d) Corresponding velocity/time plot of the sphere, using a sampling time of 10 ms. The red dashed line is a best fit through the data points located on the "velocity plateau" corresponding to the terminal velocity.

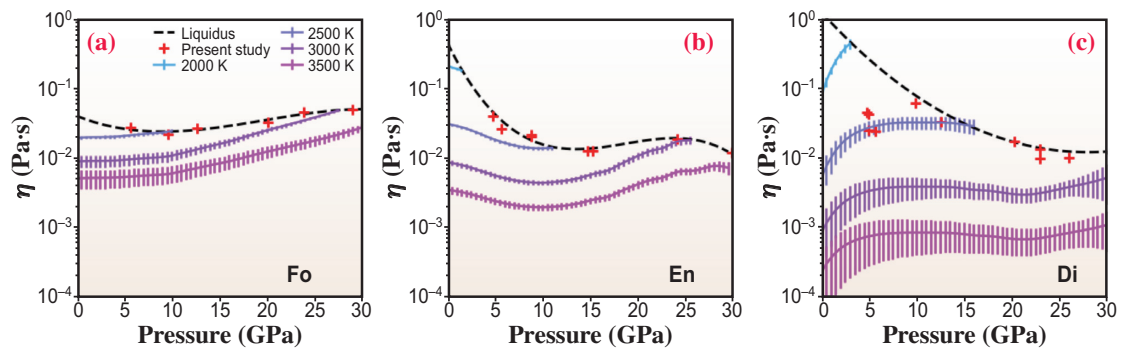


Fig. 2. Viscosities of silicate melts under pressure. (a, b, c) Fo, En and Di composition, respectively. Dashed black lines are viscosities along liquidus. Colored lines are viscosities recalculated along isotherms with one standard deviation using an Arrhenius equation.

the boron-doped diamond heater and an ultra-fast camera (frame rate reaches 1000 f/s), we succeeded to extend the pressure range of *in situ* falling sphere viscometry to 30 GPa at SPring-8 BL04B1 (Japan) and PSICHE, SOLEIL (France) [4].

We investigated the viscosity of melts with compositions similar to major mantle minerals, namely forsterite (Mg_2SiO_4 , Fo), enstatite (MgSiO_3 , En) and

diopside ($\text{CaMgSi}_2\text{O}_6$, Di). The viscosity of silicate melts in mantle conditions is found very low, in the order of 0.01 Pa·s, which is comparable to water (Fig. 2). Then, we applied the new viscosity data set to constrain the crystallization processes of the MO in the early Earth. We concluded that a bridgmanite-enriched layer forms at the top lower-mantle during the cooling of magma ocean (Fig. 3).

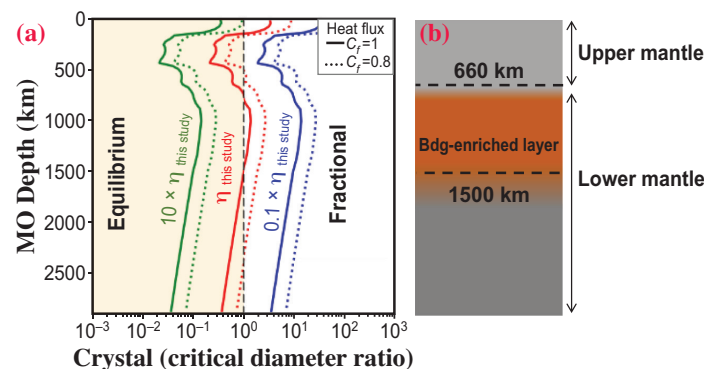


Fig. 3. Crystal/critical diameter ratio and solidification type during MO cooling. (a) Crystal/critical diameter ratio (R_{cc}) as a function of MO depth. Critical diameter is the largest grain size of crystal that can be suspended in a convecting MO. Crystal diameter is the grain size in the MO. Fractional or equilibrium solidification should occur when R_{cc} is large or smaller than 1, respectively. The heat flux of MO is affected by the atmosphere. When we ignore the blanketing effect of atmosphere ($C_f = 1$), MO has the highest heat flux, resulting in the smallest R_{cc} value (solid lines). When we consider the blanketing effect (e.g., 20%, i.e., $C_f = 0.8$), MO has a lower heat flux, resulting in a larger R_{cc} value (dotted lines). In order to show the effect of viscosity on R_{cc} value, R_{cc} calculated with 10 and 1/10 times viscosity are also plotted. Even with the highest heat flux, fractional solidification ($R_{cc} > 1$) should occur at depth of ~1000 km. (b) A conceptual drawing of mantle after magma ocean solidification. Bdg: bridgmanite.

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