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Denis Andrault, Nathalie Bolfan-Casanova, Mohamed Ali Bouhifd, Asmaa Boujibar, Gaston Garbarino, et al.. Toward a coherent model for the melting behavior of the deep Earth's mantle. *Physics of the Earth and Planetary Interiors*, 2017, 265, pp.67-81. 10.1016/j.pepi.2017.02.009 . hal-01503000

HAL Id: hal-01503000

<https://hal.science/hal-01503000v1>

Submitted on 20 Feb 2023

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Toward a coherent model for the melting behavior of the deep Earth's mantle

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41 1. ABSTRACT

42 Knowledge of melting properties is critical to predict the nature and the fate of melts produced
43 in the deep mantle. Early in the Earth's history, melting properties controlled the magma ocean
44 crystallization, which potentially induced chemical segregation in distinct reservoirs. Today, partial
45 melting most probably occurs in the lowermost mantle as well as at mid upper-mantle depths, which
46 control important aspects of mantle dynamics, including some types of volcanism. Unfortunately,
47 despite major experimental and theoretical efforts, major controversies remain about several aspects
48 of mantle melting. For example, the liquidus of the mantle was reported (for peridotitic or chondritic-
49 type composition) with a temperature difference of ~ 1000 K at high mantle depths. Also, the Fe
50 partitioning coefficient ($D_{\text{Fe}}^{\text{Bg/melt}}$) between bridgmanite (Bg, the major lower mantle mineral) and a
51 melt was reported between ~ 0.1 and ~ 0.5 , for a mantle depth of ~ 2000 km. Until now, these
52 uncertainties had prevented the construction of a coherent picture of the melting behavior of the deep
53 mantle.

54 In this article, we perform a critical review of previous works and develop a coherent, semi-
55 quantitative, model. We first address the melting curve of Bg with the help of original experimental
56 measurements, which yields a constraint on the volume change upon melting (ΔV_m). Secondly, we
57 apply a basic thermodynamical approach to discuss the melting behavior of mineralogical
58 assemblages made of fractions of Bg, CaSiO_3 -perovskite and $(\text{Mg,Fe})\text{O}$ -ferropericlasite. Our analysis
59 yields quantitative constraints on the SiO_2 -content in the pseudo-eutectic melt and the degree of
60 partial melting (F) as a function of pressure, temperature and mantle composition; For examples, we
61 find that F could be more than 40% at the solidus temperature, except if the presence of volatile
62 elements induces incipient melting. We then discuss the melt buoyancy in a partial molten lower
63 mantle as a function of pressure, F and $D_{\text{Fe}}^{\text{Bg/melt}}$. In the lower mantle, density inversions (i.e. sinking
64 melts) appear to be restricted to low F values and highest mantle pressures.

65 The coherent melting model has direct geophysical implications: (i) In the early Earth, the magma
66 ocean crystallization could not occur for a core temperature higher than ~ 5400 K at the core-mantle
67 boundary (CMB). This temperature corresponds to the melting of pure Bg at 135 GPa. For a mantle
68 composition more realistic than pure Bg, the right CMB temperature for magma ocean crystallization
69 could have been as low as ~ 4400 K. (ii) There are converging arguments for the formation of a
70 relatively homogeneous mantle after magma ocean crystallization. In particular, we predict the bulk
71 crystallization of a relatively large mantle fraction, when the temperature becomes lower than the
72 pseudo-eutectic temperature. Some chemical segregation could still be possible as a result of some
73 Bg segregation in the lowermost mantle during the first stage of the magma ocean crystallization, and
74 due to a much later descent of very low F , Fe-enriched, melts toward the CMB. (iii) The descent of
75 such melts could still take place today. Their formation should be related to incipient mantle
76 melting due to the presence of volatile elements. Even though, these melts can only be denser than
77 the mantle (at high mantle depths) if the controversial value of $D_{\text{Fe}}^{\text{Bg/melt}}$ is indeed as low as suggested
78 by some experimental studies. This type of melts could contribute to produce ultra-low seismic
79 velocity anomalies in the lowermost mantle.

80 **2. INTRODUCTION**

81 **2.1. MELTING IN THE EARLY EARTH'S MANTLE**

82 Within the first 100 million years of the Earth's history, the giant Moon forming impact (MFI)
83 melted the Earth almost entirely (Nakajima and Stevenson, 2015). The release of energy induced by
84 the gravitational segregation of the impactor's core could have potentially heated the Earth's core by
85 3500–4000 degrees above the temperature reached prior to the giant impact (Herzberg et al., 2010;
86 Nakajima and Stevenson, 2015; Rubie et al., 2015). It could have led to a core-mantle boundary
87 (CMB) temperature of the order of 6000 K (Davies et al., 2015; Labrosse, 2015; Nakagawa and
88 Tackley, 2010), well above the mantle solidus of ~4150 K (Andrault et al., 2011; Fiquet et al., 2010).
89 Therefore, the MFI could have caused intensive melting of the mantle at all depths. However,
90 geochemical evidences point out to large scale isotopic heterogeneities that seem to have survived
91 the episode of MFI. For example, the excess in ^{182}W (produced by the decay of ^{182}Hf) measured in
92 2.7 Ga-old komatiites requires a large-scale magmatic differentiation during the first 30 Ma of the
93 Solar system's history (Touboul et al., 2012). The difference in Xenon isotopic composition between
94 MORBs and the Icelandic plume (Mukhopadhyay, 2012), also indicates two sources with distinct
95 isotopic composition. Finally, geodynamical arguments suggest the presence of an unmixed and
96 primordial material in the deep mantle (Davaille, 1999; Kellogg et al., 1999). Therefore, it is unlikely
97 that primordial mantle temperatures exceeded the liquidus throughout the mantle.

98 A low viscosity of the magma-ocean is expected to induce vigorous and turbulent convective
99 flow, favoring a homogeneous mixing. In the meantime, heat flux at the magma ocean surface could
100 have been as high as $\sim 10^6 \text{ W/m}^2$, which suggests a very fast crystallization, within $\sim 10^3$ years after
101 the MFI (Solomatov, 2007). Longer time scales for cooling are also possible, due to the possible
102 formation of an opaque atmosphere at the Earth's surface preventing heat loss, or other geodynamical
103 complications, such as physical or chemical mantle layering, etc. (Lebrun et al., 2013; Sleep et al.,
104 2014). On the other hand, petrological analyses of Archean and Proterozoic basalts preserved on the
105 Earth's surface show primary magma compositions compatible with mantle potential temperatures
106 (T_p) of only 200–300 degrees higher than today (Herzberg et al., 2010). A similar temperature change
107 is reported between Archean tonalite-trondhjemite-granodiorite associations of 4.0 to 2.5 Ga old
108 (Martin and Moyen, 2002). Such a modest difference of mantle temperature compared to the present
109 situation suggests a relatively rapid mantle cooling just after the MFI.

110 Finally, it has been suggested that a basal magma ocean (BMO) could have lasted in the
111 lowermost mantle for very long times, billions of years, in correlation with an outer-core temperature
112 significantly higher than the mantle solidus (e.g. (Labrosse, 2015)). This issue remains largely
113 debated, however, because geodynamical models do not explain what could prevent such a hot core
114 from rapid cooling (Monteux et al., 2016; Nakagawa and Tackley, 2010)).

115 **2.2. EVIDENCES FOR MANTLE MELTING TODAY**

116 In the upper mantle, seismic and magneto-telluric profiles present prominent anomalies, in
117 particular at depths between 80 and 200 km (e.g. (Romanowicz, 1995)). Low velocity anomalies have
118 also been reported atop the 410-km mantle discontinuity (Song et al., 2004; Tauzin et al., 2010). Low
119 degree partial melting has often been advocated to explain these anomalies (Revenaugh and Sipkin,
120 1994), as thermochemical effects alone cannot explain the magnitude of the geophysical anomalies.
121 The possible mechanisms for mantle melting, however, are many. As for the temperature profile,
122 anchor points of the geotherm are provided by phase transformations in major mantle minerals,

123 typically olivine, the constituent responsible for the seismic discontinuities between 410 and 660 km
124 depths. It yields a potential temperature (T_p) of ~ 1600 K, which corresponds to the extrapolation to
125 the Earth's surface of the adiabatic temperature profile passing through these anchor points (Katsura
126 et al., 2010). Such a mantle geotherm plots at significantly lower temperatures compared to the
127 melting curve of the peridotitic mantle determined experimentally (Litasov and Ohtani, 2002; Zhang
128 and Herzberg, 1994). Noticeable exceptions exist, for example at mantle ridges, where the adiabatic
129 decompression of the mantle leads to melting in the asthenosphere. On the other hand, in order to
130 invoke melting at higher depths one has to invoke either higher temperatures (such as for OIB,
131 Oceanic Island Basalts), or volatiles that depress the solidus (e.g. (Hirschmann et al., 1999)).

132 Temperatures in the lowermost mantle depart from the upper mantle geotherm as they are
133 influenced by the heat flux coming from the core at the CMB. A large decrease ($>10\%$) of shear wave
134 velocities is reported in the D"-layer sitting just above the CMB. Even if alternative interpretations
135 are possible, such as a solid-state origin (e.g. Fe-enriched (Mg,Fe)O (Wicks et al., 2010)), it strongly
136 suggests occurrence of partial melting (e.g. (Herzberg et al., 2013; Rost et al., 2005)). Furthermore,
137 the non-ubiquitous character of the seismic features in the D"-layer implies a CMB temperature lower
138 than the mantle solidus. Otherwise, there would be a continuous melting line above the CMB. At the
139 CMB pressure of 135 GPa, the solidus temperature of chondritic and peridotitic mantle was reported
140 to be ~ 4150 K (Andrault et al., 2011; Fiquet et al., 2010). There, where seismic anomalies appear,
141 mantle partial melting could occur for different reasons: (i) presence of volatile elements, such as in
142 the study of (Nomura et al., 2014) where their pyrolitic composition included ~ 400 ppm H_2O , which
143 lowered the solidus temperature to ~ 3570 K. However, we note that a high water content is unlikely
144 in the lower mantle, due to the limited capability of lower mantle minerals to store water below the
145 660 km discontinuity (Bolfan-Casanova et al., 2003; Panero et al., 2015); (ii) the presence of an
146 excess of silica has been invoked; Indeed, the solidus temperature at the CMB of a mid-ocean ridge
147 basalt, was reported to be $3800 (\pm 150)$ K, thus significantly lower than for the melting of the average
148 mantle (Andrault et al., 2014). Presence of basalt in the lowermost mantle is supported by seismic
149 tomography imaging deep descent of slabs toward the CMB (e.g. (van der Hilst and Karason, 1999));
150 (iii) A very large amount of FeO could also lower the melting point of the mantle (Mao et al., 2005).
151 Altogether, the non-ubiquitous seismic features in the D"-layer could very well be correlated to the
152 presence of volatiles, basalt and/or high FeO concentration, together with a CMB temperature of 4100
153 (± 200) K (e.g. (Andrault et al., 2016)).

154 3. MELTING OF PURE BRIDGMANITE

155 3.1. IMPORTANT BRIDGMANITE PROPERTIES

156 MgO becomes extremely refractory with increasing pressure, with a melting point estimated at
157 ~8000 K for a pressure of 135 GPa (Du and Lee, 2014). This contributes in making ferropericlase
158 (Fp) the liquidus phase above 25 GPa, but only up to ~33 GPa, where Bg becomes the liquidus phase
159 for mantle compositions (Ito et al., 2004). The melting curve of Bg dominates the topology of melting
160 diagrams at high mantle depths. The role of Bg is further emphasized by a possible increase of Bg-
161 content with increasing mantle depth, as the comparison between geophysical observations and
162 experiments (e.g. (Murakami et al., 2012; Samuel et al., 2005)) suggest a mantle (Mg+Fe)/Si ratio
163 closer to unity.

164 Mantle Bg can adopt chemical compositions ranging from (Mg,Fe)SiO₃, produced after
165 decomposition of olivine, to Al-bearing (Mg,Fe)SiO₃, after the high pressure transformation of
166 majoritic garnet below the 660 km discontinuity. A higher compositional variability could also come
167 from the intrinsic high compliance of the Bg crystal structure, which composition can be decomposed
168 in a number of (theoretical) end-members (MgSiO₃, FeSiO₃, FeAlO₃, Al₂O₃, MgAlO_{2.5}). It can adopt
169 variable Mg/Si, Fe³⁺/ΣFe, Al/Fe ratios and Fe-content as a function of pressure, temperature, oxygen
170 fugacity, and exchange with ferropericlase. (e.g. (Boujibar et al., 2016; Lauterbach et al., 2000)). The
171 Bg crystal chemistry can thus vary with mantle depth (Andrault et al., 2007) and also laterally if the
172 mantle presents significant heterogeneities. We also remind that the transition pressure from majoritic
173 garnet to Bg increases significantly with the Al-content: compared to pure MgSiO₃, an additional 3-
174 4 GPa is required to produce Bg with ~10 wt% Al₂O₃ (Akaogi and Ito, 1999; Irifune et al., 1996).

175 3.2. PREVIOUS STUDIES OF BRIDGMANITE MELTING

176 In early LH-DAC experiments, visual observations of the sample behavior were used as a major
177 criterion to address the melting of Bg (Figure 1). Two studies performed on MgSiO₃ and (Mg,Fe)SiO₃
178 compositions agree for a steep melting curve above the transition zone pressure, at ~24 GPa (Shen
179 and Lazor, 1995; Zerr and Boehler, 1993). On the other hand, comparison between the melting curves
180 of MgSiO₃ and Mg₃Al₂Si₃O₁₂ evidences a significant melting temperature depletion, by ~300 K at 40
181 GPa, due to the presence of Al (Shen and Lazor, 1995). Early works were limited to 60-70 GPa,
182 compared to the 135 GPa prevailing at the CMB. Concerning melting of MgSiO₃ in the upper mantle,
183 we note an important curvature of the melting line typical of the lower-density polymorphs (Katsura
184 and Ito, 1990; Presnall and Gasparik, 1990). It can be related to a rapid decrease of the volume of
185 melting (ΔV_m) with increasing pressure and, thus, a relatively high melt compressibility.

186 Several theoretical works address the equation of state and the melting curve of Bg up to more
187 than 135 GPa, using *ab initio* calculations (Figure 1). The MgSiO₃ end-member is generally studied,
188 because such calculations encounter difficulties in addressing the role of transition elements and point
189 defects (such as Al³⁺, Fe³⁺, etc.). In all calculations, the initial slope of the melting curve (above ~24
190 GPa) is relatively steep. This points out to a relatively large volume of MgSiO₃-Bg melting, which is
191 dominated by the ~10% volume change at the transition from majoritic garnet to Bg at 660 km-depth
192 (Yu et al., 2011). Based on the propagation of shock waves, MgSiO₃ melting was identified at 5000-
193 5500 K for a pressure above 100 GPa (Akins et al., 2004; Mosenfelder et al., 2009). It was also
194 suggested that the volume of Bg melting becomes very small at pressures typical of the lowermost
195 mantle (Mosenfelder et al., 2009; Petitgirard et al., 2015).

196 **3.3. UPDATED MELTING CURVE OF BRIDGMANITE**

197 In this article, we report original measurements of the melting curve of a (Fe,Al)-bearing Bg
 198 phase with $\text{Fe}/(\text{Mg}+\text{Fe})=0.12$ and $\text{Al}/\text{Fe}=0.9$. We acknowledge that this composition corresponds to
 199 relatively high Al and Fe contents compared to the expected composition for natural Bg. Experiments
 200 were performed using LH-DAC coupled with *in situ* X-ray diffraction on the ID-27 beamline (ESRF,
 201 France). All experimental methods are detailed in *Supplementary Materials*. At relatively low
 202 pressures, the melting curve of this Bg appears significantly more flat than those reported for pure
 203 MgSiO_3 (Figure 1). This observation tends to confirm a significant effect of Al on the melting slope,
 204 as reported previously between Al-free and Al-bearing starting materials (Shen and Lazor, 1995). We
 205 also find that the discontinuous increase of the melting curve slope, related to the majorite to Bg phase
 206 transition, is not observed at ~ 24 GPa, but instead at ~ 30 - 35 GPa for this (Fe,Al)-bearing composition.
 207 This behavior can be logically related to extension to higher pressures of the garnet stability field in
 208 the presence of Al, as described in previous works for Fe-free starting materials (Akaogi and Ito,
 209 1999; Irifune et al., 1996). The relatively lower melting temperature of (Fe,Al)-bearing Bg in the 25-
 210 35 GPa pressure range, compared to pure- MgSiO_3 and, most importantly, to other lower mantle
 211 minerals such as MgO, explains why Bg is not the liquidus phase in this pressure range for typical
 212 mantle compositions (Ito et al., 2004).

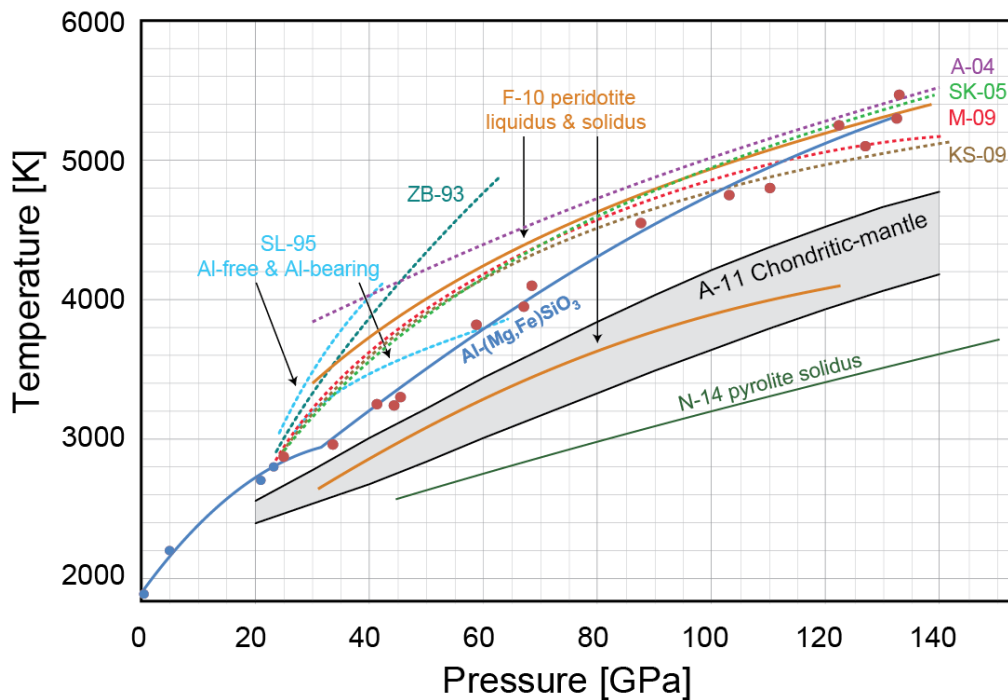


Figure 1: Melting curve of Bg inferred from different experimental and theoretical studies determined using LH-DAC for $(\text{Mg}_{0.88}\text{Fe}_{0.12}\text{SiO}_3)$ (Zerr and Boehler, 1993), MgSiO_3 and $\text{Mg}_3\text{Al}_2\text{Si}_3\text{O}_{12}$ (Shen and Lazor, 1995), shock measurements for MgSiO_3 (Akins et al., 2004; Mosenfelder et al., 2009) and *ab initio* calculations for MgSiO_3 (e.g. (de Koker and Stixrude, 2009; Stixrude and Karki, 2005)). We also include experimental reports of solidus and liquidus of peridotite (Fiquet et al., 2010), chondritic-type mantle (Andrault et al., 2011) and pyrolite with ~ 400 ppm H_2O (Nomura et al., 2014). Our new LH-DAC measurements performed from ~ 24 to ~ 135 GPa on a (Fe,Al)-rich Bg (red dots and blue guide for the eyes) plot in continuity with previous determinations of mantle melting performed at lower pressures using the large volume press (Blue dots, (Katsura and Ito, 1990; Presnall and Gasparik, 1990)).

214 At higher pressures, the slope of the (Fe,Al)-bearing Bg melting curve remains fairly constant up
215 to the CMB pressure of 135 GPa, which contrasts with the flattening reported for MgSiO₃. This makes
216 the different available results to converge to a temperature of 5100-5500 K for the melting of Bg at
217 the CMB. We remind that the melting curve of MgSiO₃-Bg should always plot at higher temperature
218 than that of the (Fe,Al)-bearing composition, due to the incompatible character of Fe and Al. Thus,
219 our experimental determination of the (Fe,Al)-bearing Bg melting curve implies that the melting
220 temperature of MgSiO₃-Bg at very high mantle pressures was underestimated by a couple hundred
221 degrees in some previous studies (de Koker and Stixrude, 2009; Mosenfelder et al., 2009). Thus, for
222 further discussions, we will choose the higher melting curve of MgSiO₃ Bg as determined from *ab*
223 *initio* calculations (Stixrude and Karki, 2005). It can be modeled using a modified Simon and Glatzel
224 equation [$T=T_0 (P/a + 1)^{1/c}$] with parameters $T_0=91$ K, $a=0.00125$ GPa and $c=2.83$ (Simon and
225 Glatzel, 1929).

226 We note that the liquidus temperature profile reported previously for peridotite (Fiquet et al.,
227 2010) appears superimposed with the melting curve of MgSiO₃-Bg derived from *ab initio* calculations
228 and shock experiments (Figure 1). This is unexpected because (i) the more complex chemical
229 composition of the Bg phase itself, compared to MgSiO₃, and (ii) additional Fp and CaSiO₃-
230 perovskite (CaPv) phases should both lower the peridotite liquidus below the melting temperature of
231 pure MgSiO₃. In their study, (Fiquet et al., 2010) observed large crystals of Bg at the center of the
232 heated spot, where the melting behavior was monitored using *in situ* X-ray diffraction. Chemical
233 composition of Bg grains measured at different locations on a sample synthesized at ~61 GPa show
234 Fe/(Mg+Fe) and Al/Si ratios of ~2.5% and ~8%, respectively. It is possible that their reported
235 measurements of the peridotite liquidus correspond in fact to the melting curve of these large grains
236 of MgSiO₃-bearing Bg.

237 3.4. VOLUME OF BRIDGMANITE MELTING

238 The dT/dP slope of the Bg melting curve is directly related to the volume (ΔV_m) and entropy
239 (ΔS_m) of Bg melting through the classical Clapeyron relation $dT/dP = \Delta V_m/\Delta S_m$. ΔS_m was reported
240 to be 42.2 J/molK for MgSiO₃ at room pressure, based on calorimetric measurements (Stebbins et al.,
241 1984). For melting of MgSiO₃ in the stability field of Bg, *ab initio* simulations suggest a ΔS_m value
242 1.5 times higher (Stixrude et al., 2009). Assuming then a constant ΔS_m value of 63.3 J/molK for
243 melting of (Fe,Al)-bearing Bg at all lower mantle conditions, we now calculate ΔV_m values from the
244 clapeyron slopes of the different melting curves available for Bg (Figure 2a). For the (Fe,Al)-bearing
245 composition, we observe a progressive decrease of $\Delta V_m/V_{Bg}$ (V_{Bg} being the Bg volume) from ~9%
246 to ~5%, when pressure increases from 30 to 140 GPa, respectively. For comparison, earlier studies
247 of the melting behavior of pure MgSiO₃ suggest a significantly larger effect of pressure, with
248 $\Delta V_m/V_{Bg}$ varying from 13% at low pressures for some *ab initio* calculations (Stixrude et al., 2009) or
249 even 19% for a shock-wave study (Mosenfelder et al., 2009), to 3-4% at pressures of 135 GPa. The
250 flatter ΔV_m found for the (Fe,Al)-bearing implied by our results is due to a more linear melting curve
251 measured from 30-35 GPa to 135 GPa, compared to pure MgSiO₃ (Figure 1). We also show recent
252 measurements of the MgSiO₃-glass density performed in the DAC at 300 K up to the CMB pressure
253 (Petitgirard et al., 2015). The $\Delta V_m/V_{Bg}$ recalculated from the later study is significantly lower than
254 all other calculations at high pressures, which should translate into a very flat melting curve. Based
255 on the shadowed region in Figure 2a, we define a most probable range of $\Delta V_m/V_{Bg}$ values decreasing
256 from 11(± 2)% to 4(± 1)% when pressure is increased from 24 to 140 GPa, respectively.

257 We also calculated ΔV_m at pressures below 24 GPa using $\Delta S_m=42.2$ J/molK (Stebbins et al.,
 258 1984) and using the Clapeyron of melting curves measured in large volume press (LVP) experiments.
 259 We note that the final interpretation of ΔV_m can be complicated by the occurrence of four different
 260 $MgSiO_3$ polymorphs between 1 bar and 24 GPa (Presnall, 1995). In particular, the Clapeyron slope
 261 of majorite melting remains largely uncertain, due to a limited pressure range available. For this
 262 reason, we modeled the $MgSiO_3$ melting line using a 2nd degree polynomial regression across the
 263 whole pressure range from 0 to 24 GPa. The calculated $\Delta V_m/V_{Bg}$ decreases strongly with increasing
 264 pressure converging to 1% at ~24 GPa (Figure 2a). It makes a volume change of ~8% at the majorite
 265 to Bg transition for the (Fe,Al)-bearing composition of our study, or ~10% in average for the most
 266 probable range of $\Delta V_m/V_{Bg}$ that we have defined above, both values agreeing well with the ~10%
 267 suggested previously for $MgSiO_3$ (Yu et al., 2011).

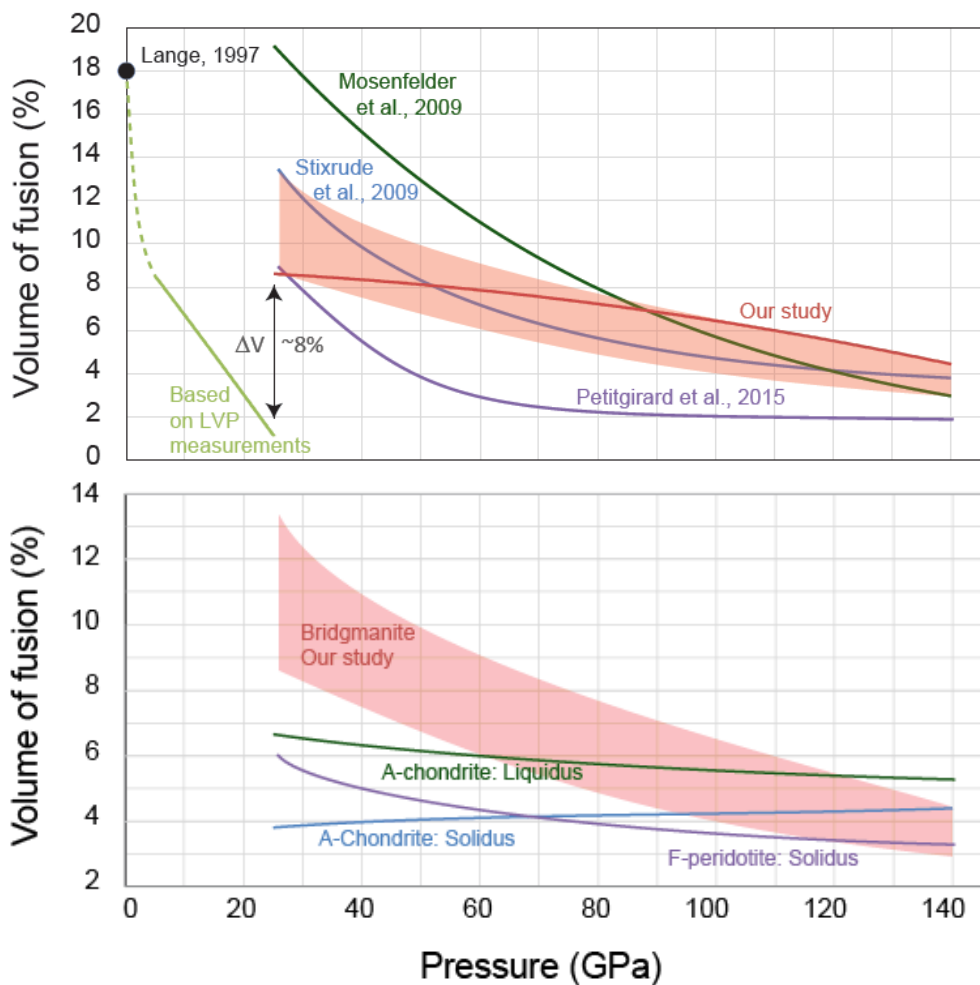


Figure 2: (a) Volume of melting ($\Delta V_m = V_{melt} - V_{Solid} / V_{Bg}$, where V_{Bg} is volume of Bridgmanite) derived from the melting curve of Bg (Figure 1) with various (Mg,Fe)(Si,Al) O_3 compositions. We also report the value of 18% measured for pure $MgSiO_3$ at 1 bar (Lange, 1997) and $\Delta V_m/V_{Bg}$ recalculated from density measurements of $MgSiO_3$ -glass in a DAC (Petitgirard et al., 2015). Shaded pink region is the most probable range of mantle $\Delta V_m/V_{Bg}$ values. (b) ΔV_m derived from the solidus and/or liquidus profiles of the A-chondritic (Andraut et al., 2011) and F-peridotitic (Fiquet et al., 2010) models of mantle melting. ΔV_m is calculated using the Clapeyron relation. Calculation of $\Delta V_m/V_{Bg}$ along solidus and liquidus profiles assume an ideal mixing model to estimate the melt entropy (see text).

4. MELTING DIAGRAMS FOR TYPICAL LOWER MANTLE MATERIALS

4.1. EXPERIMENTAL DETERMINATION OF THE MELTING BEHAVIOR

270 Studying experimentally phase relations in a partially molten geological material under P-T
271 conditions of the Earth's interior remains a challenging task. The use of LVP facilitates largely the
272 chemical analysis of coexisting phases in recovered samples, compared to LH-DAC. For example,
273 melting behavior of fertile peridotite and CI chondritic mantle were documented up to ~35 GPa (Ito
274 et al., 2004). It was demonstrated that the liquidus phase changes from Fp to Bg at about 33 GPa, a
275 situation that was confirmed later by LH-DAC experiments (Andraut et al., 2011; Fiquet et al., 2010).
276 However, LVP samples (and *a fortiori* LH-DAC samples) synthesized at lower mantle conditions
277 always encounter relatively large temperature gradients, which yields heterogeneous microstructure
278 and chemical zoning. In such cases, the sequence of phase disappearance from solidus to liquidus
279 temperatures is usually retrieved from the position of phases relative to the temperature gradient. Still,
280 this procedure prevents the precise determination of the equilibrium solid-liquid partition coefficients.

281 When coupled with *in situ* X-ray diffraction, LH-DAC is well suited to determine melting curves
282 of various geological materials, because the available temperature range extends up to more than
283 6000 K (Suppl. Figure 1). Melting criteria are generally based on the disappearance of diffraction
284 peaks and phase(s), which corresponds in fact to the determination of a pseudo-eutectic temperature
285 (PET). Recently, PET of peridotite (Fiquet et al., 2010), chondritic-mantle (Andraut et al., 2011) and
286 pyrolite (Nomura et al., 2014) was reported with substantially different values. Disagreements cannot
287 be reconciled based on the moderate compositional variations between starting materials, among
288 which the major difference lies in the Bg/(Bg+CaPv+Fp) ratio from 0.59, 0.63 and 0.75 for peridotite,
289 pyrolite and chondritic mantle, respectively. Studies using peridotitic (F-peridotitic model) and
290 chondritic (A-chondritic model) starting materials show similar solidus evolution as a function of
291 pressure, pointing out to a PET of 4150 ± 150 K at the CMB. The lower value reported in the study
292 using pyrolite could be due to a more precise determination of the solidus, thanks to the use of 3D X-
293 ray tomography, but more likely because of the presence of 400 ppm H₂O in their sample that triggers
294 melting (Nomura et al., 2014).

4.2. PSEUDO-EUTECTIC TYPE MELTING IN THE DEEP MANTLE?

296 The melting diagram between the three lower mantle phases (Bg, CaPv and Fp) presents an
297 eutectic behavior (e.g. (de Koker et al., 2013; Liebske et al., 2005; Liebske and Frost, 2012)), which
298 implies (i) a PET and (ii) a melt composition at the PET located within the ternary diagram. The PET
299 and the melt composition should be independent of the Bg, CaPv and Fp molar fractions in the bulk
300 material, as long as the chemical compositions of these three phases do not vary significantly. A major
301 issue for geological materials is that the onset of melting (the solidus) can possibly occur at a
302 temperature lower than the PET. In such a case, a melt appears without disappearance of a phase from
303 the mineral assemblage. This can happen when the solubility limit of an incompatible element is
304 overpassed at a given condition of pressure and temperature. This behavior has not been reported yet
305 for any mantle compositions under the P-T conditions typical of the lower mantle. In contrast,
306 available LH-DAC experiments report disappearance of one phase at the onset of melting (see above).
307 Also, chemical analyses of melts obtained after partial melting of peridotite at ~24 GPa show
308 compositions close to the starting material (e.g. with a relatively lower MgO/SiO₂ ratio (Tronnes and
309 Frost, 2002)), which is incompatible with incipient mantle melting.

311 Above the solidus temperature, partition coefficients between Bg or CaPv and the melt
 312 ($D_i^{\text{Solid/melt}}$) were investigated experimentally for many elements. Except for a very few of them (such
 313 as Li and Ba), $D_i^{\text{Solid/melt}}$ values are found to lie above $3 \cdot 10^{-1}$, which corresponds to a moderately
 314 incompatible behavior (Corgne et al., 2005). These results suggests that the minor elements may not
 315 reach their solubility limit easily. Of course, volatiles such as H₂O (but also CO₂) are well known to
 316 depress the solidus and potentially favor incipient mantle melting. We note, however, that (i)
 317 carbonate phases may be relatively refractory in the lower mantle (Thomson et al., 2014) and (ii)
 318 a high water content in the lower mantle is unlikely, due to limited capability of the lower mantle
 319 minerals to store water below the 660 km discontinuity (Bolfan-Casanova et al., 2003). Thus, even if
 320 water has an effect of the solidus of the lower mantle it does not mean that water is actually present.
 321 In summary, experimental evidences strongly suggest a moderate temperature difference between
 322 solidus and PET, if any.

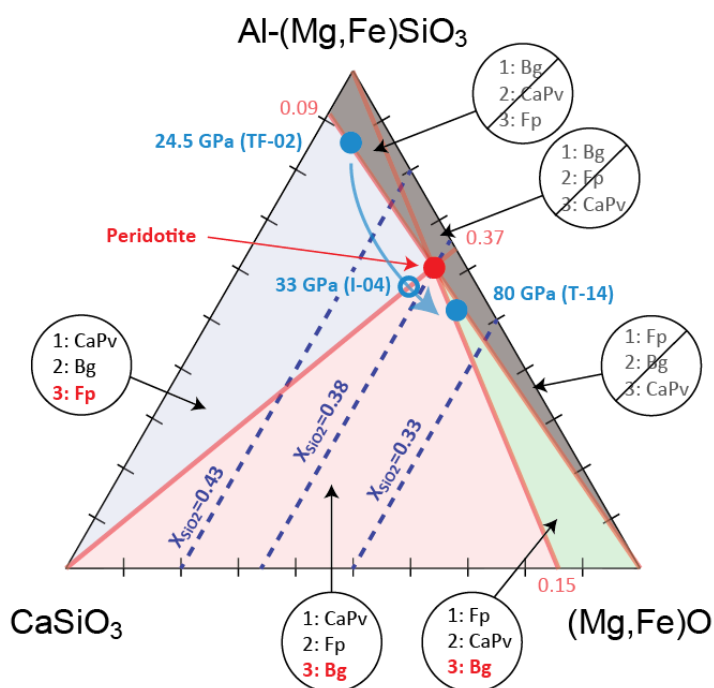


Figure 3: Ternary diagram between Bg, Fp and CaPv, the three major phases in the lower mantle. Due to its eutectic shape, melt compositions should always plot within this diagram. The Red dot and lines correspond to the peridotitic composition and its associated Fp/Bg, CaPv/Bg and CaPv/Fp ratios. Blue dots are melt compositions determined experimentally (Ito et al., 2004; Tateno et al., 2014; Tronnes and Frost, 2002). The blue arrow shows the change in melt composition with pressure. In each colored region of the ternary diagram, the expected sequence of phase disappearance upon melting are reported inside circles: 1, 2 and 3 are solidus, intermediate and liquidus phases, respectively.

323 4.3. SEQUENCE OF PHASE DISAPPEARANCE UPON MELTING

324 In the ternary diagram between Bg, CaPv and Fp, the difference in composition between the bulk
 325 composition and the melt defines the sequence of phase disappearance upon melting. For example,
 326 melting of peridotite would likely induce a melting sequence from CaPv (at solidus), Bg to Fp (at
 327 liquidus), if the PE-melt would present significantly higher CaSiO₃-content and lower (Mg,Fe)O-
 328 content, compared to peridotite. Thus, the ternary diagram can be divided into six different regions
 329 where the melting sequence should be different, depending on the PE-melt composition (Figure 3).

330 Composition of melts relevant to deep mantle partial melting were reported after experiments
 331 performed at 24.5 GPa using LVP (Tronnes and Frost, 2002) and 80 GPa using LH-DAC (Tateno et
 332 al., 2014). Despite the fact that these melts may differ from PE-melts, if experimental temperatures
 333 have been higher than PET, they provide precious anchors for melt compositions in the ternary
 334 diagram. Composition of these two melts (corresponding to 24.5 and 80 GPa) plot on each side of the
 335 Fp/Bg ratio of 0.37, which corresponds to the peridotite composition. This observation is compatible
 336 with a third experimental anchor point where the liquidus phase has changed from Fp to Bg at about

337 ~33 GPa for a fertile peridotite (Ito et al., 2004). Thus, the melt composition evolves toward higher
338 (Mg,Fe)O-contents with increasing pressure.

339 Another important observation is the melt composition at 80 GPa, which presents a CaPv/Fp
340 ratio very close to the peridotitic value of 0.15. It suggests that both phases, CaPv and Fp, disappear
341 almost simultaneously at the PET upon partial melting of peridotite in the deep lower mantle. In this
342 work, we make the assumption that CaPv and Fp disappear simultaneously at the PET at lower mantle
343 pressures. This assumption is not severe. If Fp would disappear first, negligible amount of CaPv
344 would remain. Indeed, the total amount of CaPv is below ~6 mol% for compositions relevant to the
345 deep mantle. In the case where CaPv would disappear first, higher Fp content could theoretically
346 remain after melting at the PET. Indeed, Fp is found at a level of 19, 32 or 35 mol%, for chondritic,
347 pyrolitic or peridotitic compositions, respectively. However, the molar volume of Fp is twice less
348 than those of Bg and CaPv. Also, it is likely that the melt dissolves a significant part of Fp at the PET
349 (Tateno et al., 2014; Tronnes and Frost, 2002), which should yield to a low Fp-content in the residual
350 solid.

351 **4.4. LIQUIDUS TEMPERATURES AND BG-CONTENT IN MELTS**

352 The melting behavior can be modeled at various mantle pressures (for example 40 GPa, 80 GPa
353 and 135 GPa) using (pseudo) binary phase diagrams, where one end-member is Bg, the liquidus phase,
354 and the other is the sum of all the other chemical components (Figure 4). We prefer to use Fe-free Bg
355 as an end-member, instead of (Fe,Al)-bearing Bg, due to the preferential partition of Fe to the melt
356 (this matter is discussed later in the article). Still, we consider an Al-bearing Bg end-member, because
357 the Al-behavior upon partial melting remains poorly constrained. Compositional axis of such binary
358 diagram is thus the molar fraction of Fe-free Bg. Melts include Fp, CaPv, (virtual) FeSiO₃-Bg and a
359 fraction of Fe-free Bg end-members. Peridotitic and chondritic mantle, as well as our (Fe,Al)-bearing
360 Bg composition plot at 59, 75 and 88 mol% of Fe-free Bg, respectively. For the PET, the A-chondritic
361 model indicate “solidus” temperatures of 2690, 3340 and 4130 K at pressures of 40, 80 and 135 GPa,
362 respectively (Andrault et al., 2011). For the Fe-free Bg end-member, we use ab initio calculations for
363 MgSiO₃-Bg (Stixrude and Karki, 2005), as discussed above. Two LH-DAC studies report liquidus
364 temperatures of A-chondritic and F-peridotitic models (Andrault et al., 2011; Fiquet et al., 2010).
365 Finally, the independent reports of the Bg-content in melts (i) measured after partial melting of
366 pyrolite (Tateno et al., 2014) and (ii) calculated in the MgSiO₃-MgO system (de Koker et al., 2013;
367 Liebske and Frost, 2012) are also shown. We note that the two later studies did not included the CaPv
368 component, which could numerically yield an overestimation of the Bg-content in the melt.

369 At each selected mantle pressure, we draw lines from the melting temperature of the Bg end-
370 member, through the liquidus temperature of the A-chondritic model, toward the PET (Figure 4).
371 These lines provide estimates of liquidus temperatures as a function of the bulk mantle composition.
372 In addition, intersections between these lines and PET provide estimates of the Bg-content in PE-
373 melts, as a function of pressure. Uncertainties arise from (i) controversial reports of the Bg melting
374 temperature as a function of pressure (Figure 1) and (ii) unknown curvature of the liquidus profile in
375 these diagrams. Consequently, we colored in pink regions where the presence of liquidus is the most
376 probable. We excluded from colored regions (i) PE-melts with a fraction of Fe-free Bg higher than
377 peridotite, because it would be contradictory with Bg becoming the liquidus phase above ~33 GPa
378 (Ito et al., 2004), (ii) a previous study reporting inaccurate Bg melting curve at low pressures (Akins
379 et al., 2004) and (iii) trends extrapolated from (de Koker and Stixrude, 2009; Mosenfelder et al., 2009)
380 as they may have underestimated the Bg melting temperature by a couple hundred degrees in the

381 lowermost mantle (see text associated to [Figure 1](#)). We note that our method is not appropriate to
 382 pressures lower than ~33 GPa, when Bg is not the liquidus phase.

383 Based on the width of the regions of confidence, the liquidus temperatures appears to be
 384 constrained within ± 150 K at all mantle pressure, temperature and compositions. An uncertainty to
 385 which we should still add a similar value associated to the experimental error in the determination of
 386 PET and Bg melting temperature. On the other hand, uncertainty in Bg-content in PE-melt and along
 387 liquidus lines is about $\pm 5\%$. The Bg-content in the PE-melt appears almost independent of lower
 388 mantle pressure at $55(\pm 5)$ mol%. With increasing temperature above PET, the Bg-content in melt
 389 should logically increase until it reaches the bulk mantle composition. Altogether, these binary
 390 melting diagrams can be numerically constrained by three parameters: (i) a constant Bg-content of
 391 $55(\pm 5)$ mol% in PE-melt, and two modified Simon and Glatzel (SG) equations $[T(P)=T_0 (P/a+1)^{1/c}]$
 392 with parameters (ii) $T_0=2045$ K, $a=92$ GPa and $c=1.3$ for the PET (Andrault et al., 2011) and (iii)
 393 $T_0=91$ K, $a=0.00125$ GPa and $c=2.83$ for the Bg end-member ((Stixrude and Karki, 2005), see above).
 394 Then, liquidus temperatures between PE-melt and Bg can be defined as a function of pressure and
 395 mantle composition using:

$$396 T_{\text{Mantle}}^{\text{Liq}}(P, X_{\text{Bg}}^{\text{Mantle}}) = \text{SG}(\text{Bg}) - [(\text{SG}(\text{Bg}) - \text{SG}(\text{PEM})) * (1 - X_{\text{Bg}}^{\text{Mantle}}) / (1 - X_{\text{Bg}}^{\text{PEM}})]$$

397 where $\text{SG}(\text{PEM})$ and $\text{SG}(\text{Bg})$ are PE-melt and Bg melting temperatures, respectively, calculated at a
 398 target mantle pressure (P) using SG equations. $X_{\text{Bg}}^{\text{PEM}}$ and $X_{\text{Bg}}^{\text{Mantle}}$ are Bg mole fractions in the PE-
 399 melt (fixed at 55 mol%) and the bulk mantle, respectively.

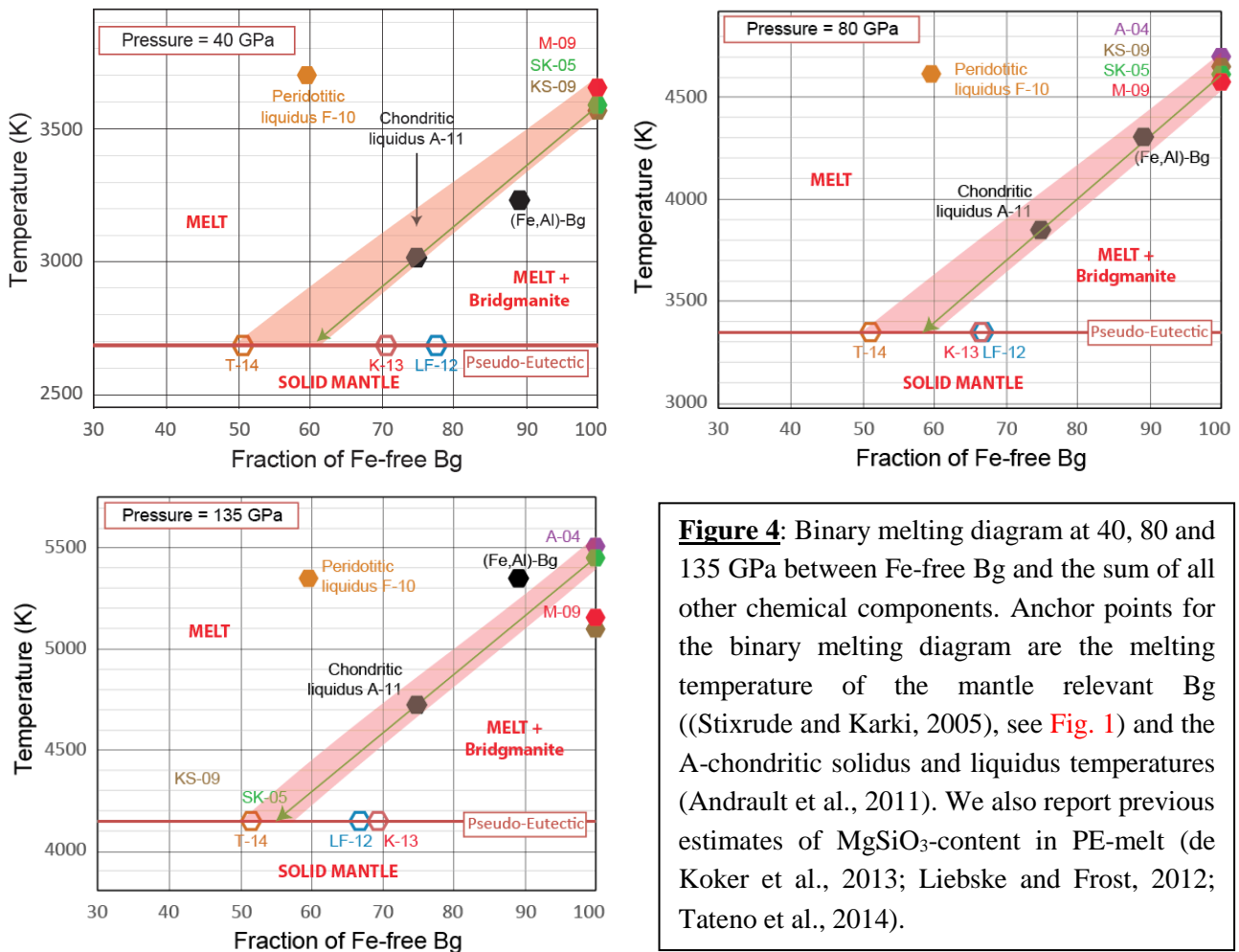


Figure 4: Binary melting diagram at 40, 80 and 135 GPa between Fe-free Bg and the sum of all other chemical components. Anchor points for the binary melting diagram are the melting temperature of the mantle relevant Bg ((Stixrude and Karki, 2005), see [Fig. 1](#)) and the A-chondritic solidus and liquidus temperatures (Andrault et al., 2011). We also report previous estimates of MgSiO_3 -content in PE-melt (de Koker et al., 2013; Liebske and Frost, 2012; Tateno et al., 2014).

400 For a peridotitic mantle composition, binary phase diagrams suggest a difference of less than 300
 401 K between PET and liquidus at all lower mantle pressures. This is in very good agreement with the
 402 value of 250 K proposed based on a thermodynamic melting model (Liebske and Frost, 2012). The

403 difference between the PET and the liquidus increases for larger fractions of Bg in the bulk material,
 404 and in a primitive chondritic mantle it can reach up to 500 degrees. This confirms the wide range of
 405 mantle depths where partial melting should have occurred in the framework of an early magma ocean
 406 (e.g. (Kojitani and Akaogi, 1997; Miller et al., 1991)).

407 4.5. SiO₂ CONTENT IN PSEUDO-EUTECTIC MELTS

408 Another parameter controlling the melt properties is its SiO₂-content (X(SiO₂)). It can be derived
 409 directly from Figure 4, by converting the amount of Fe-free Bg in the melt into its SiO₂ contribution.
 410 Based on our primary assumption that Fp and CaPv dissolve concomitantly in the melt at the PET,
 411 the SiO₂-content of the CaPv component is also added to the melt composition. This calculation can
 412 be performed for all types of melt along liquidii, as long as pressure is above ~33 GPa. Still, we only
 413 discuss here the composition of PE-melts. We actually translate into X(SiO₂) the regions of
 414 confidence defined above for Bg-contents in PE-melts (Figure 5). Independent values of X(SiO₂) in
 415 lower mantle melts were reported previously based on *ab initio* calculations (de Koker et al., 2013),
 416 thermodynamic modeling of LVP experiments (Liebske and Frost, 2012) and chemical analyses of
 417 LH-DAC samples (Nomura et al., 2011; Tateno et al., 2014). At about ~33 GPa, the change of liquidus
 418 phase from Fp to Bg reported upon peridotite melting suggests that X(SiO₂) in the melt crosses the
 419 peridotitic value of 38.3% (Ito et al., 2004).

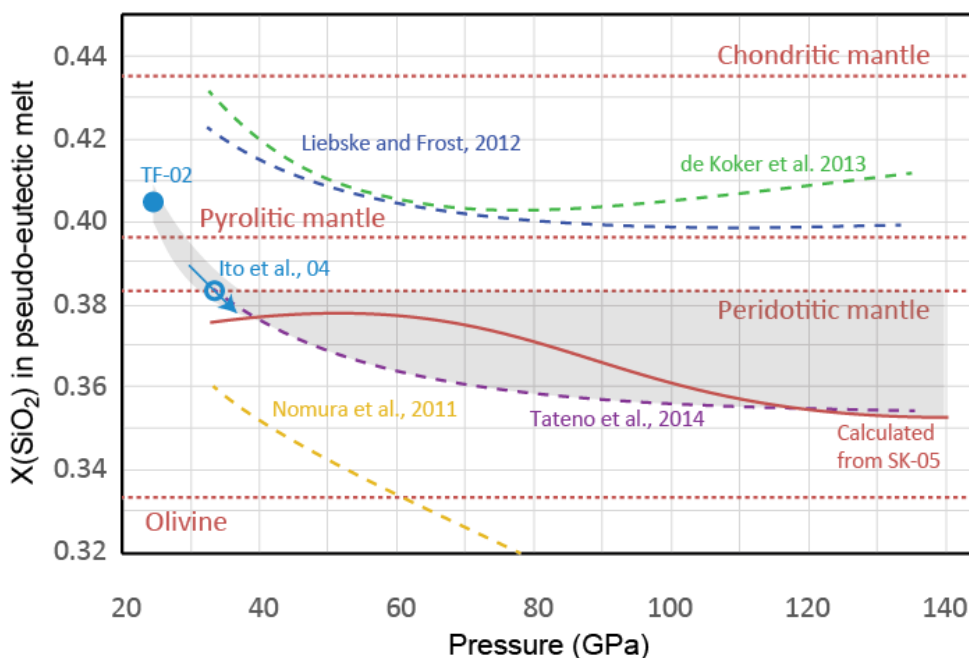


Figure 5: Pressure change of the SiO₂-content in pseudo-eutectic melt. Horizontal dotted lines are X(SiO₂) typical of olivine, peridotite, pyrolite and chondritic-mantle. The red line is derived from the Bg melting curve (Stixrude and Karki, 2005) associated with the thermodynamical treatment elaborated for the discussion of Figure 4. Dashed lines represent previous calculations (de Koker et al., 2013; Liebske and Frost, 2012) or the experimental determination (Tateno et al., 2014) of X(SiO₂) in melts. The shaded gray area corresponds to the most likely X(SiO₂) of lower mantle melts.

420 The melt X(SiO₂) likely remains significantly lower than 0.40 at higher pressures in the lower
 421 mantle, because Bg was broadly reported to be at the liquidus for pyrolite (Nomura et al., 2014) and
 422 peridotite (Fiquet et al., 2010). The fact that two models (de Koker et al., 2013; Liebske and Frost,
 423 2012) propose higher X(SiO₂) could be related to calculations restricted to the MgSiO₃-MgO system.
 424 The difference between the simple MgO-SiO₂ system and the mantle-related compositions suggests
 425 that Ca and Fe should enter the melt preferentially as CaO and FeO, rather than CaSiO₃ and FeSiO₃,

426 thus lowering $X(\text{SiO}_2)$. Such behavior translates into a significantly stronger incompatible character
 427 of Ca and Fe, compared to Mg and Si (Liebske et al., 2005), which is already well known for low-
 428 pressure melting. The range of possible melt $X(\text{SiO}_2)$ appears to extend between the peridotite and
 429 chemical analyses performed on LH-DAC samples (Tateno et al., 2014). It corresponds to an almost
 430 fixed $X(\text{SiO}_2)$ value of 37.0% ($\pm 1.5\%$). This most probable range of $X(\text{SiO}_2)$ plots in good continuity
 431 with data reported at lower pressure based on LVP experiments (Ito et al., 2004; Tronnes and Frost,
 432 2002). In summary, deep PE-melts appear to adopt a composition only slightly enriched in Fp
 433 compared to peridotite, if any.

434 4.6. DEGREE OF PARTIAL MELTING AT PSEUDO-EUTECTIC TEMPERATURES

435 We can now calculate the expected degree of partial melting (F) at the PET for peridotitic,

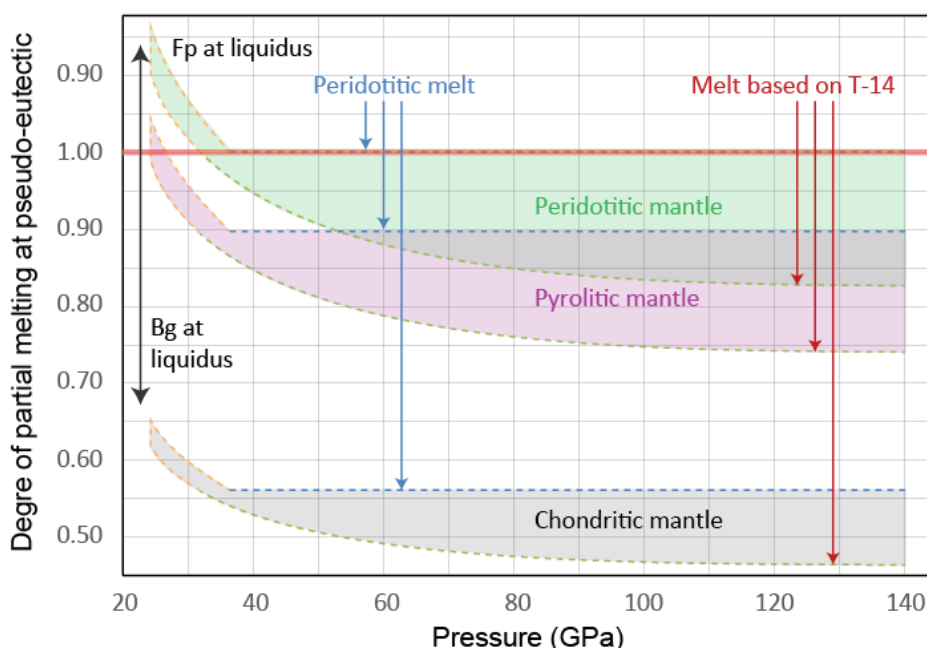


Figure 6: Degrees of mantle partial melting (vol%) expected at pseudo-eutectic temperature (PET) in the lower mantle under the assumptions that CaPv and Fp phases disappear jointly upon melting. Degrees of partial melting are calculated for typical chondritic, pyrolitic and peridotitic mantle compositions, based on the possible range of PE-melt composition refined in Figure 5. T-14 stands for (Tateno et al., 2014).

436 pyrolitic and chondritic mantle compositions, based on our estimation of deep-mantle melt
 437 compositions (Figures 4 and 5). F (in vol%) at PET can be estimated using the molar volumes for Bg,
 438 Fp and CaPv (Equations of state needed to take into account the effect of pressure and temperature on
 439 volumes are described below). Logically, the calculated F value at PET varies with the difference in
 440 composition between the PE-melt and the bulk material. The closest the mantle composition is from
 441 the melt, the highest F value is expected at the PET. Calculated ranges of possible F values at PET
 442 appear to be more than 83, 74 and 46 vol%, for peridotitic, pyrolitic and chondritic mantle
 443 compositions, respectively (Figure 6).

444 We note that a similar conclusion was reached previously (e.g. (Herzberg and Zhang, 1998))
 445 based on the residual harzburgite signature of most komatiites with Cretaceous and late-Archaeon
 446 ages, which can be interpreted to have formed by about 25 to 60% anhydrous melting of mantle
 447 peridotite in hot plumes.

448 The melting relations discussed above have implications for our understanding of the melting in
 449 the deep mantle today. For example, in the ultra-low velocity zones (ULVZ) just above the CMB, the

450 high F values determined for PET imply that the ULVZ cannot correspond to partial melting of the
451 average mantle, because the degree of partial melting in the ULVZ as inferred by seismology is much
452 lower than the threshold value of 46%. But a lower F-value could be achieved (i) for a locally high
453 concentration in volatile elements, which would favor incipient mantle melting below the PET or (ii)
454 for another type of geological material, such as subducted basalts (Andrault et al., 2014).
455

5. MELT BUOYANCY IN THE DEEP MANTLE

5.1. MAJOR PARAMETERS CONTROLLING THE MELT BUOYANCY

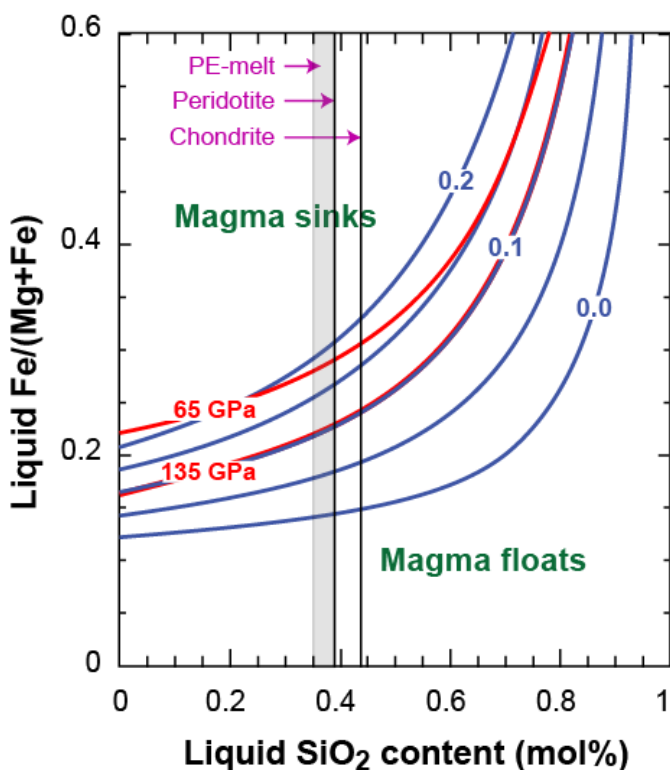
Dynamical behavior of a partially molten mantle is largely affected by the density contrast between the melt and the solid fractions ($\Delta\rho_{m-s}=(\rho_m-\rho_s)/\rho_s$). While melt buoyancy plays a major role in chemical segregation and planetary differentiation, the fact that its sign is positive or negative produces two different geodynamical situations. Melt buoyancy can also vary with mantle depth, due to (i) a possible change of the melt composition and (ii) different PVT equations of state for the melt and the mantle residue. In the present-day mantle, ascent, or descent, of some melts could contribute to surface volcanism, or accumulation of melts in the lowermost mantle, respectively. In a crystallizing magma ocean, it is classically accepted that crystals would settle, compact and reject the melt toward the Earth's surface (Boukare et al., 2015; Solomatov, 2007). However, this question became much debated after it was proposed that an early basal magma ocean could be produced by dense melts stored above the CMB (Labrosse et al., 2007; Nomura et al., 2011). Solid-liquid density inversions are not unusual. Many years ago, it was already proposed that basaltic and komatiitic magmas would become denser than olivine and garnet at depths of 245-500 km (Agee, 1998).

Major parameters controlling the buoyancy of deep-mantle melts are (i) changes in atomic packing between solid and liquid phases, which most generally produces a positive volume of melting (ΔV_m), (ii) Fe partition coefficient between the solid residue and the melt ($D_{Fe}^{Bg/melt}$), because Fe is the heaviest major element, and (iii) MgO/SiO₂ ratio in the melt, because SiO₂-based atomic packing presents a relatively high bulk modulus. We chose to use $D_{Fe}^{Bg/melt}$ ($D=X_{Fe}^{Bg}/X_{Fe}^{melt}$, with X being the molar fraction), rather than the distribution coefficient $K_{Fe}^{Bg/melt}$ ($K=(X_{Fe}/X_{Mg})^{Bg}/(X_{Fe}/X_{Mg})^{melt}$), because X_{Mg}^{melt} can vary with pressure, temperature and bulk composition independently from X_{Fe}^{melt} . Nothing prevents X_{Mg}^{melt} and X_{Fe}^{melt} to vary in a correlated way, which would contradict the proper definition of K.

Concerning the volume difference between solids and melts, both phases undergo (i) volume changes upon P and T according to their respective equation of state and (ii) a number of structural transformations with increasing mantle depth. In the solid state, equation of states of major mantle minerals are relatively well constrained (Matas et al., 2007). Structural transformations in the melt are much more progressive than in the solid, but can still contribute to a net increase of the melt density with increasing mantle depths. Due to the disordered and time-dependent nature of the melt structure, each structural entity has a finite lifetime. And when pressure is applied, structural entities can progressively evolve. In polymerized liquids, polyhedral chains can bend, eventually break and recombine differently (Stolper and Ahrens, 1987; Wang et al., 2014). All major cations progressively change their coordination shell (Bajgain et al., 2015) to allow compaction of the oxygen sub-lattice (Zeidler et al., 2014). The major change is probably Si, which progressively changes coordination number from four-fold to six-fold between ~20 to ~40 GPa (Benmore et al., 2010; Guillot and Sator, 2007; Sanloup et al., 2013; Sato and Funamori, 2010). These progressive transformations yield a room pressure bulk modulus significantly lower for the melt than for the crystalline counterpart. However, the pressure derivative of the bulk modulus is generally much higher, due to the progressive loss of some compaction mechanisms, after major voids have disappeared from the melt structure at moderate pressures.

497 At shallow mantle depths, while the melt structure has already undergone significant compaction,
 498 major mantle minerals, in particular olivine, have not yet transformed to a denser polymorph. For this
 499 reason, there could be a neutral melt buoyancy, or even a positive $\Delta\rho_{m-s}$ (i.e. a sinking melt), at mantle
 500 depths just above the 410 km discontinuity (Matsukage et al., 2005; Sakamaki et al., 2006). It could
 501 produce a kind of density trap, where melts produced above and below would accumulate and remain
 502 gravitationally stable in this mantle region (e.g. (Lee et al., 2010)). At lower mantle pressures, a
 503 previous study tentatively addresses $\Delta\rho_{m-s}$ for a number of melt and solid compositions (Figure 7;
 504 (Funamori and Sato, 2010), see also (Thomas et al., 2012)). Reporting in this diagram our estimates
 505 of $X(\text{SiO}_2)$ in PE-melt retrieved from Figure 5 narrows down the range of possible silica contents
 506 between ~ 35.2 to ~ 38.3 mol%. We note that the SiO_2 -content in the melt should increase with
 507 increasing temperature above the PET, because the PE-melt is relatively depleted in SiO_2 compared
 508 to the mantle composition. Ultimately, the melt and the bulk mantle become similar at the liquidus
 509 temperature, when the solid residue disappears completely. Altogether, the rather limited range of
 510 possible SiO_2 -content in mantle melts points out to the fact that the less constrained parameters needed
 511 to address melt buoyancy at mantle depths are ΔV_m , the volume of melting, and $D_{\text{Fe}}^{\text{Bg/melt}}$, the Fe solid-
 512 liquid partitioning.

Figure 7: Relative buoyancy between mantle melts of various compositions and (i) the $(\text{Mg,Fe})\text{SiO}_3$ Bg with Fe-contents of 0, 10, or 20% (blue lines) and (ii) the lower mantle at pressures of 65 and 135 GPa (red lines). Along each blue or red lines, the melt and the given solid phase (Bg or lower mantle) have the same density. Higher SiO_2 -content or lower Fe/(Mg+Fe) ratio, compared to a given line, yield melts buoyant compared to the solid phase considered. The gray region corresponds to the likely SiO_2 -content in PET-melts in the mantle (see Figure 5). SiO_2 -content values for chondritic and peridotitic mantle are also reported. This Figure is modified from (Funamori and Sato, 2010).



513 5.2. VOLUME OF MANTLE MELTING

514 While the ΔV_m corresponding to the melting of a pure phase, such as Bg, can be estimated based
 515 on its melting curve (Figure 2a), the volume of melting becomes more difficult to address in the case
 516 of the partial melting of the mantle between its solidus and liquidus. In addition, the equation of state
 517 of the melt changes significantly with its composition (de Koker et al., 2013; Thomas and Asimow,
 518 2013). For example, Mg_2SiO_4 -liquid becomes denser than lower mantle phases at pressures above
 519 ~ 40 GPa, while MgSiO_3 -liquid remains buoyant at all mantle pressures (Thomas and Asimow, 2013).
 520 Thus, assuming a simplified melt composition, such as MgSiO_3 , may not be much relevant for
 521 discussing the buoyancy of natural melts in the deep mantle. A more realistic exercise is to estimate
 522 the real density difference between the solid residue and a melt of relevant composition in the Bg-
 523 CaPv-Fp system. The solid residue can be a simple phase (Bg, or Fp below ~ 33 GPa), or a typical

524 mantle composition (e.g. pyrolite), which would be relevant to model melt buoyancy in the context
525 of an early crystallizing magma ocean (high F), or partial melting in the present-day lower mantle
526 (low F), respectively. The melt composition at various P and T can be derived from melting diagrams
527 (Figure 4).

528 In Figures 2b, we intend to use the slopes of solidus and liquidus reported for mantle melting
529 from LH-DAC experiments (Andraut et al., 2011; Fiquet et al., 2010) to retrieve thermodynamical
530 information relevant to melts. We compare the solidus, or liquidus, profile to a Clapeyron slope which
531 would record melting of the first fraction of solid mantle, or the last fraction of Bg, respectively. At
532 the solidus temperature, the entropy of melting (ΔS_m) can be estimated from the sum of melting
533 entropies of each phase present in the melt, plus a mixing entropy. We used ΔS_m^i values of 63.3, 63.3
534 and 13.0 J/molK for the melting entropy of Bg (Stixrude et al., 2009), CaPv (similar to Bg) and Fp,
535 respectively. We note that the room pressure value of ΔS_m^{MgO} was reported to be significantly larger
536 than that of Fp, at ~ 35 J/molK, however, it decreases rapidly with increasing pressure to 25 GPa
537 (Vocadlo and Price, 1996). Composition of the PE-melt is considered to be 60% Bg, 30% Fp and
538 10% CaPv, in agreement with Figures 3 and 4. However, changing the phase contents does not affect
539 greatly the calculated ΔS_m value of ~ 55 J/molK. At the liquidus temperature, we assume that the ΔS_m
540 of the bulk rock is similar to that of Bg, neglecting the fact that the melt in which last grains of Bg
541 get dissolved has a composition slightly different from Bg. Based on these assumptions, we calculate
542 volumes of melting at solidus and liquidus, using the Clapeyron equation (Figure 2b).

543 For the A-chondritic model, the values of $\Delta V_m/V_{Bg}$ at the solidus and at the liquidus appear
544 almost independent of mantle pressure, at values of $4(\pm 1)\%$ and $6(\pm 1)\%$, respectively. The $\Delta V_m/V_{Bg}$
545 at the solidus of the F-peridotitic model plots at similar values, however, due to a larger curvature in
546 the melting curve, its associated $\Delta V_m/V_{Bg}$ varies more with pressure. We note that if we would have
547 included a supplementary non-ideal mixing contribution to ΔS_m (de Koker et al., 2013), the $\Delta V_m/V_{Bg}$
548 values would have been $\sim 0.5\%$ higher. Calculations appear compatible with a small variation of the
549 volume of melting with temperature increasing from the solidus to the liquidus and a small decrease
550 with pressure from $\sim 6\%$ to $\sim 4\%$ from 30 to 130 GPa, respectively. This significantly positive value
551 of $\Delta V_m/V_{Bg}$ should contribute to a positive melt buoyancy for all types of melts.

552 5.3. LIQUID-SOLID FE PARTITIONING

553 Based on LVP experiments performed up to ~ 35 GPa, Fe is recognized as an incompatible
554 element, with a concentration ~ 1.5 to ~ 3 times higher in the melt compared to the solid mantle (e.g.
555 (Ito et al., 2004; Liebske et al., 2005)). For samples recovered from LH-DAC experiments, Fe was
556 reported to become extremely incompatible with the Bg structure at pressures above ~ 70 GPa, with
557 a Fe concentration more than 10 times higher in the melt, compared to Bg (Nomura et al., 2011)
558 (Figure 8). Such an enrichment of iron in melts (associated to a very low Fe-concentration in the
559 residual Bg) favors their higher density relative to the solid residue, facilitating their sinking towards
560 the CMB. In this previous study, olivine was used as a starting material and the absence of Al could
561 have favored a relatively low $D_{Fe}^{Bg/melt}$ value (Wood and Rubie, 1996). Still, another study performed
562 on an Al-bearing peridotitic composition using the same experimental methodology confirms the very
563 low Fe-content in the residual Bg (Tateno et al., 2014). We note that the Al partition coefficient
564 between Bg and melt ($D_{Al}^{Bg/melt}$), which was also reported to decrease significantly from 1.8 to 0.64
565 with increasing pressure from 34 to 88 GPa in the same study. On the other hand, much higher
566 $D_{Fe}^{Bg/melt}$ values of 0.5-0.6 were reported for a chondritic-mantle starting material using *in situ* X-ray
567 fluorescence spectroscopy (Andraut et al., 2012). These last results favor denser Bg grains and

568 buoyant melts. The controversy cannot arise from slight compositional differences between starting
 569 materials, especially between peridotitic and chondritic mantle compositions used in (Tateno et al.,
 570 2014) and (Andraut et al., 2012), respectively. It should be related to differences in experimental
 571 and/or analytical techniques.

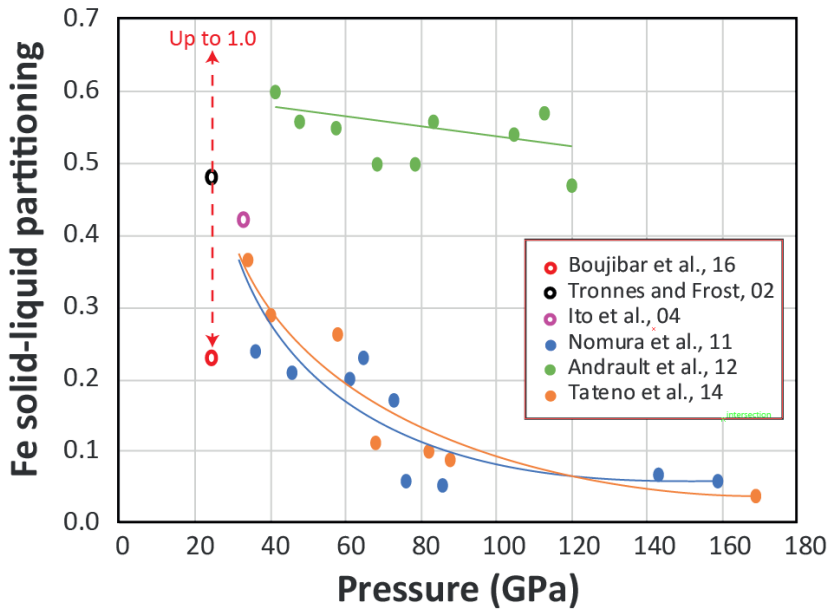


Figure 8: Change in Fe partitioning ($D_{\text{Fe}}^{\text{Bg/melt}}$) between the Bg and a coexisting melt. Starting materials for LH-DAC experiments were olivine (bleu, (Nomura et al., 2011)), chondritic-type mantle (green, (Andraut et al., 2012)) and peridotite (red, (Tateno et al., 2014)). Open circles correspond to previous multi-anvil experiments (Boujibar et al., 2016; Ito et al., 2004; Tronnes and Frost, 2002). The red arrow corresponds to the range of $D_{\text{Fe}}^{\text{Bg/melt}}$ measured in a same study at varying F values.

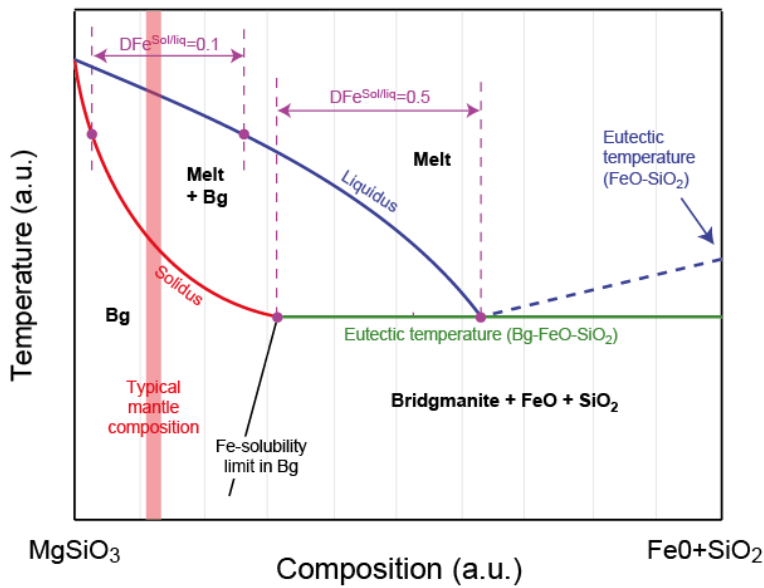


Figure 9: A possible, schematic, melting diagram between MgSiO_3 and FeSiO_3 in the lower mantle. We plot $\text{FeO}+\text{SiO}_2$ as the Fe-bearing end-member because $\text{FeSiO}_3\text{-Bg}$ is unstable, except maybe at very high pressures. Vertical purple dashed lines are compositions of coexisting Bg and melt. Strong curvatures of solidus and liquidus lines can yield a decrease of $D_{\text{Fe}}^{\text{Bg/melt}}$ with increasing temperature between the eutectic and the liquidus temperature, in agreement with a previous study (Boujibar et al., 2016).

572 A recent study addresses the value of $D_{\text{Fe}}^{\text{Bg/melt}}$ as a function of F, the degree of partial melting,
 573 based on LVP experiments performed at ~25 GPa (Boujibar et al., 2016). $D_{\text{Fe}}^{\text{Bg/melt}}$ was found to vary
 574 from ~1 to a constant value of ~0.3, for F values below ~5 wt% or above 10 wt%, respectively. It
 575 suggests a $D_{\text{Fe}}^{\text{Bg/melt}}$ negatively correlated with temperature between the solidus and the liquidus, in
 576 agreement with a typical binary melting diagram (Figure 9). Thus, controversial results between
 577 previous LH-DAC experiments could arise from different heating conditions. Experiments reporting
 578 very low $D_{\text{Fe}}^{\text{Bg/melt}}$ values were performed using a very thin insulating material (hardly detectable
 579 using X-ray diffraction). A clear advantage is to facilitate the recovery of relatively thick samples,
 580 which can be cut after the experiment in order to obtain relatively large and homogeneous sample
 581 regions for chemical analyses using the electron microprobe. However, such experimental procedure
 582 implies strong axial temperature gradient between the two diamonds. The center part of the recovered
 583 samples is likely to correspond to relatively high temperatures, potentially close to the liquidus (high
 584 F possibly inducing low $D_{\text{Fe}}^{\text{Bg/melt}}$, Figure 9). Alternatively, experiments reporting higher $D_{\text{Fe}}^{\text{Bg/melt}}$

585 values used very thin samples well embedded in the insulating material (producing major diffraction
586 peaks). While this type of loading minimizes axial temperature gradients in the sample, it makes the
587 sample recovery much more difficult. In a previous study, X-ray diffraction was used to monitor
588 carefully the onset of melting at the solidus temperature (Andrault et al., 2011) (low F possibly
589 inducing high $D_{\text{Fe}}^{\text{Bg/melt}}$). Differences in heating procedure and thus in melting temperature between
590 previous works could have induce significantly different values of $D_{\text{Fe}}^{\text{Bg/melt}}$. We note that the Fe
591 behavior could be complicated by other factors: (i) the role of Al, which is not well constrained at
592 high pressures; Due to the strong Al-Fe coupling in the Bg structure, an Al-depletion from the Bg
593 structure would contribute to lower $D_{\text{Fe}}^{\text{Bg/melt}}$; (ii) the variable Fe^{2+} and Fe^{3+} fractions, because each
594 one could partition differently between Bg and the melt, etc. Needless to say that a full quantitative
595 model of $D_{\text{Fe}}^{\text{Bg/melt}}$ at lower mantle conditions requires additional experimental and theoretical studies.

596 **5.4. SOLID-LIQUID DENSITY CONTRAST**

597 The $\Delta\rho_{\text{m-s}}$ can be evaluated based on the density of both phases. All calculations were performed
598 for a bulk mantle of pyrolitic composition. We consider a bulk Fe/(Mg+Fe) ratio (Fe#) of 0.1. We
599 adjust the Fe-contents in the solid and the melt in order to reproduce the bulk mantle Fe-content, for
600 various values of $D_{\text{Fe}}^{\text{Bg/melt}}$, which remains an adjustable parameter in the calculation. For the
601 coexisting solid and melt compositions, we first calculate the densities in the solid state, before we
602 apply a correction to the melt density of 6% to 4% for pressures between 30 and 130 GPa,
603 respectively, to take into account the volume of melting (Figure 2). Composition of the solid can
604 correspond either to (i) pure Bg, as expected for the residual solid from solidus to liquidus
605 temperatures in our pseudo-binary melting diagrams (Figure 4), or to (ii) the mineral assemblage
606 typical of pyrolite, which would correspond to the mantle composition in case of incipient melting.
607 On the other hand, composition of the melt is approximated by a mixture of 55% Bg, 6.2 % CaPv and
608 38.8 % Fp, which represents well the PE-melt at all lower mantle pressures (Figure 4). We neglect
609 the P-T dependence of melt composition, because the $X(\text{SiO}_2)$ value of the PET is not very different
610 from the bulk pyrolitic mantle. Densities at high P and T of each phases are calculated based on their
611 respective molar fractions of the following five different end-members: Al-bearing MgSiO_3 , FeSiO_3 ,
612 MgO , FeO , CaPv , using the PVT equation of state of each of them. We used the Mie-Grüneisen
613 formalism with an auto-coherent set of elastic parameters (Matas et al., 2007).

614 Our calculation shows that the degree of partial melting has a major effect on melting properties
615 (Figure 10). It affects (i) the Bg-content (and thus $X(\text{SiO}_2)$) in the melt, which increases moderately
616 with temperature from PET to the liquidus (Figure 4), (ii) potentially the $D_{\text{Fe}}^{\text{Bg/melt}}$ value ((Boujibar
617 et al., 2016); Figure 9) and (iii) Fe-contents in coexisting solid and melt, which vary with F
618 independently from $D_{\text{Fe}}^{\text{Bg/melt}}$. In the context of mantle partial melting, two end-member situations
619 arise:

620 A) During the first stages of magma ocean crystallization, F values are expected to be large. They
621 could range between ~0.6 to 1, which corresponds to temperatures between the PET and the liquidus
622 (Figure 6). We also note the threshold F value of ~0.4 corresponding to point when the magma ocean
623 becomes more viscous (Abe, 1997). The situation corresponds to a variable fraction of Bg grains
624 entrained in a major amount of melt. Close to the liquidus (F~1), Fe# in Bg could vary from ~0.05 to
625 ~0.01, depending on the controversial values of $D_{\text{Fe}}^{\text{Bg/Melt}}$ from 0.5 to 0.1, respectively, because the
626 Fe# in the melt is ~0.1, similar to the bulk mantle. Using the procedure described above, we calculate
627 $\Delta\rho_{\text{m-s}}$ between coexisting Bg and pyrolitic melt along melting curves and obtain mostly negative
628 values, meaning a relatively denser Bg grains (Figure 10). The value of $\Delta\rho_{\text{m-s}}$ increases with pressure,

629 due to a smaller volume of fusion at higher pressures but remains negative at all pressures from 30 to
 630 135 GPa. $\Delta\rho_{m-s}$ also increases with decreasing the melt fraction, because the melt Fe# increases faster
 631 than the solid Fe#. We find no solid-melt density crossover for $D_{Fe}^{Bg/Melt} = 0.5$, at least for F values
 632 higher than 0.4. For $D_{Fe}^{Bg/Melt} = 0.1$, Bg can become less dense than the melt for F values below ~ 0.7 .
 633 B) At the latest stages of magma ocean crystallization, as well as in the context of partial melting
 634 in the present-day lower mantle, the situation corresponds to a minor amount of melt trapped in the

Figure 10: Density contrast for various couples of coexisting melt and Bg, in the context of a crystallizing magma ocean (F higher than 0.4). All values are normalized to the density of a same Bg having an Fe# of 5% (red). Negative values correspond to buoyant melt. We report relative densities of Bg with Fe# of 1% (green), fully molten pyrolite (yellow) and partial melts calculated for F values from 0.9 to 0.4. We perform the calculation for $D_{Fe}^{Bg/melt}$ values of 0.1 (blue area, corresponding to (Nomura et al., 2011; Tateno et al., 2014)) and 0.5 (brown area, corresponding to

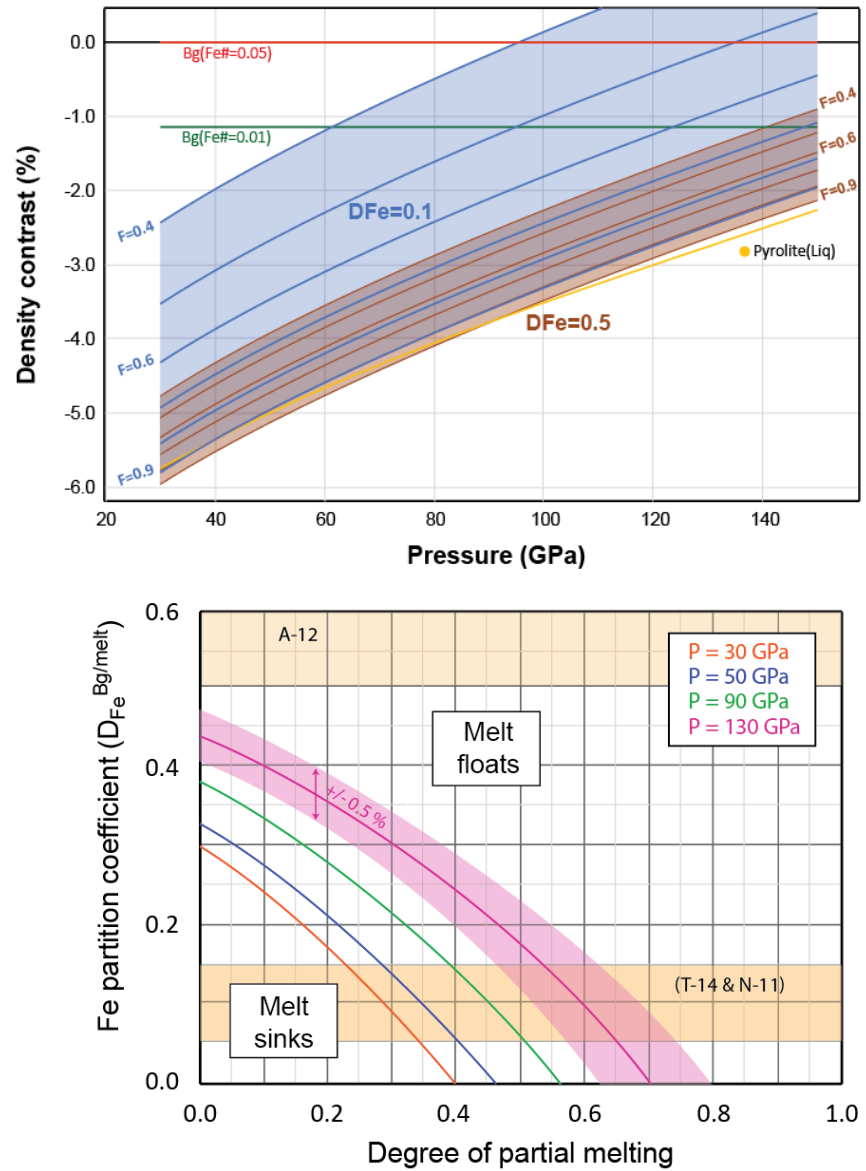


Figure 11: Domains of positive or negative melt buoyancy for varying degrees of mantle partial melting and various possible values of the solid-liquid Fe partition coefficient. We report boundaries between “sink” and “float” domains at pressures of 30, 50, 90 and 130 GPa. The pink shaded region corresponds to a density contrast within 0.5%. Horizontal orange shaded regions correspond to ranges of $D_{Fe}^{Bg/melt}$ reported around 0.1 (T-14 & N-11; (Nomura et al., 2011; Tateno et al., 2014)) and 0.5 (A-12; (Andrault et al., 2012)) at high mantle depths (Figure 8).

635 solid mantle. The solid phase can be Bg or pyrolite, depending on the melting degree, F. Here, we
 636 report $\Delta\rho_{m-s}$ values corresponding to a solid phase made of pure Bg, however, results are not very
 637 different for pyrolite. Due to the lack of constraints, we assume a melt with $X(SiO_2)$ equal to that of
 638 the PE-melt, even if the situation could correspond to incipient mantle melting below the PET (low

639 F). Under these assumptions, the major change in melt composition with decreasing F is a higher Fe#.
640 Calculations show that a denser melt, compared to a bridgmanitic mantle, is favored for (i) lower
641 $D_{\text{Fe}}^{\text{Bg/Melt}}$, (ii) lower F and (iii) higher pressures (Figure 11). There is a significant range of parameters,
642 for values of F below 0.7 and $D_{\text{Fe}}^{\text{Bg/Melt}}$ below 0.45, where melts could sink in the solid mantle toward
643 the CMB. We remind here that the possible increase of $D_{\text{Fe}}^{\text{Bg/Melt}}$ at low F values (Boujibar et al.,
644 2016) would favor buoyant melts.

645

646 **6. IMPLICATIONS FOR CRYSTALLIZATION OF THE MAGMA OCEAN**

647 **6.1. DYNAMICS OF THE MAGMA OCEAN CRYSTALLIZATION**

648 The crystallization of the early magma ocean is the primary mechanism invoked to produce a
649 chemical stratification of the mantle (Boyet and Carlson, 2005; Caro et al., 2005). Concerning the
650 mantle depth at which the crystallization began, our model suggests a mantle liquidus increasing
651 smoothly with pressure up to the CMB pressure of 135 GPa (i.e. the A-chondritic model in **Figure 1**).
652 Thus, the adiabatic temperature profile of the cooling magma ocean, which also presents a relatively
653 constant slope, would likely cross the liquidus starting from the CMB upwards (e.g. **Figure 7a** in
654 (Thomas and Asimow, 2013)). Magma ocean solidification should therefore occur from bottom up.
655 We note that major changes in the dynamics of the magma ocean should occur at F values lower than
656 $\sim 40\%$, the critical melt fraction when the mantle becomes more viscous (Abe, 1997), rather than when
657 the adiabatic profile crosses the liquidus (Monteux et al., 2016).

658 It has been proposed that Bg grains could settle in the magma ocean, inducing rejection of melts
659 toward the Earth's surface (Boukare et al., 2015; Solomatov, 2007). Our calculations confirm a Bg
660 denser than the coexisting melt at F values higher than 0.7 (**Figure 11**). Still, the effective gravitational
661 segregation of Bg grains in a crystallizing magma ocean is not obvious. Highly turbulent flows have
662 certainly dominated in the molten mantle. $\Delta\rho_{m-s}$ may not have been sufficiently high to
663 counterbalance mixing forces, especially in the lowermost mantle where $\Delta\rho_{m-s}$ decreases. This could
664 have prevented major chemical segregation until late stages of the magma ocean crystallization. On
665 the other hand, while an abrupt change in rheological behavior at $F\sim 40\%$ should disable turbulent
666 mixing flows, it should also slow down chemical segregation by inhibiting the atomic diffusion. Thus,
667 the two side-effects of crystallization, turbulent flow followed by high mantle viscosity, are not in
668 favor of large scale chemical heterogeneities early in the crystallizing Earth's mantle.

669 Following the assumption that Bg grains have efficiently segregated from the crystallizing
670 magma ocean, the melt composition should drift toward higher Fe#, because Fe enters preferentially
671 into the melt (e.g. (Boukare et al., 2015)) and also to a slightly higher SiO₂-content (due to the
672 composition of the PET melt, **Figure 5**). Increasing the Fe# in melt favors higher $\Delta\rho_{m-s}$ and sinking
673 melts. This effect could have contributed to produce an early BMO (Labrosse et al., 2007) and/or
674 overturns (Elkins-Tanton, 2008). However, our melting model suggests that the Fe-enrichment of the
675 melt could remain moderate. If we consider the most extreme case of (i) full gravitational segregation
676 of the crystallizing Bg grains, (ii) a $D_{Fe}^{Sol/liq}$ value close to 0 and (iii) a bulk fractional crystallization
677 of 60% (thus $F=0.4$), we find that the melt would reach Fe# of ~ 0.25 (for a bulk pyrolitic mantle with
678 Fe# of ~ 0.1). For more reasonable sets of parameters, a high degree of Fe-enrichment can only occur
679 at the very final stages of the magma ocean crystallization.

680 **6.2. CHEMICAL SIMILARITIES BETWEEN THE PSEUDO EUTECTIC-MELT AND PERIDOTITE**

681 Major consequences arise from the very high F values found at the PET (**Figure 6**). Brutal
682 solidification of the magma ocean could occur when the degree of partial melting becomes lower than
683 83% (for peridotite), 74% (for pyrolite) or 56% (for chondritic mantle) at a given mantle depth. Such
684 F values are significantly higher than the threshold value of 40% necessary to produce a relatively
685 viscous mantle (Abe, 1997). Therefore, final mantle crystallization could be achieved at the same time
686 as the mantle became viscous. It would disable chemical segregation by melt gravitation in a mushy
687 mantle and favor instead a homogeneous mantle after bulk magma ocean crystallization. Mantle

688 overturn would also be unlikely, because the magma ocean would crystallize before a too significant
689 Fe-enrichment of the melt.

690 Our model shows that the peridotite composition is close to the PE-melt at high pressure (Figures
691 3 to 6). This compositional similarity was already discussed a long time ago (e.g. (Zhang and
692 Herzberg, 1994)). This situation could result from a large scale chemical differentiation during magma
693 ocean crystallization, where the upper part of the mantle would correspond to the last melt that
694 solidified, and the residual solid, Bg, would be concentrated in the deep mantle. A floating layer of
695 peridotitic melt of several hundred kilometers thickness (possibly even more than 1000 km) would
696 have crystallized as a bulk, in agreement with our high F values at the PET. A highly bridgmanitic
697 deep lower mantle would agree with previous studies (Matas et al., 2007; Murakami et al., 2012;
698 Samuel et al., 2005).

699 Today, presence of volatile elements could induce incipient partial melting in the deep mantle.
700 The melt could be denser than the mantle and sink, in the case the value of $D_{\text{Fe}}^{\text{Bg/melt}}$ would indeed be
701 as low as 0.4, as suggested by some previous studies (Nomura et al., 2011; Tateno et al., 2014).
702 Sinking melts could produce the ULVZ in the lowermost mantle, independently from the temperature
703 prevailing in the lowermost mantle early in Earth's history. This scenario contrasts with another one
704 suggesting that the ULVZ are the relic of an ancient BMO (Labrosse et al., 2007).

705 **6.3. MAXIMUM CORE TEMPERATURE AFTER MAGMA OCEAN CRYSTALLIZATION**

706 After the Moon forming giant impact, the Earth has been largely molten. Subsequent cooling of
707 the magma ocean could have been relatively fast, within 100 ky (Abe, 1997; Monteux et al., 2016;
708 Solomatov, 2007). It was proposed that a BMO could have persisted in the lowermost mantle for a
709 much longer time, due to a large amount of heat stored in the core (Labrosse et al., 2007). Still, the
710 early temperature of the CMB (a good proxy for the core temperature), as well as the lifetime of the
711 BMO remain highly debated. To keep a hot BMO thermally stable, there must be a layer of solid
712 material just above it producing an efficient thermal blanketing between the molten BMO in contact
713 with the hot core and the solid mantle. This solid shell would hamper escape of the core heat. This
714 solid material could only be Bg, the liquidus phase in the deep mantle. Consequently, the Bg melting
715 point of ~5400 K at a pressure of 135 GPa (Figure 1) corresponds formally to the highest possible
716 CMB temperature after crystallization of the magma ocean. It is ~1300 K above the current CMB
717 temperature.

718 However, a temperature of ~5400 K represents an improbable upper limit for the early CMB
719 temperature. Indeed, the physical contact between the BMO and the overlaying Bg layer can react
720 chemically. The further away from Bg is the chemical composition of the BMO, the lower should be
721 the equilibrium temperature, in agreement with binary phase diagrams (Figure 4). And the more likely
722 the CMB temperature after magma ocean crystallization could correspond to the first occurrence of
723 a more viscous mantle at the CMB (Monteux et al., 2016). This temperature is estimated to ~4400 K,
724 only ~300 K higher than the CMB temperature today (Andraut et al., 2016).

725 **Acknowledgments:** This work has been supported by the ANR contract “OxyDeep”. This research
726 was financed by the French Government Laboratory of Excellence initiative n°ANR-10-LABX-
727 0006, the Région Auvergne and the European Regional Development Fund. This is Laboratory of
728 Excellence ClerVolc contribution number XX.

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731

References:

732

Abe, Y., 1997. Thermal and chemical evolution of the terrestrial magma ocean. *Phys. Earth Planet. Inter.* 100,
733 27-39.

734

Agee, C.B., 1998. Crystal-liquid density inversions in terrestrial and lunar magmas. *Phys. Earth Planet. Inter.*
735 107, 63-74.

736

Akaogi, M., Ito, E., 1999. Calorimetric study on majorite-perovskite transition in the system $Mg_4Si_4O_{12}$ -
737 $Mg_3Al_2Si_3O_{12}$: transition boundaries with positive pressure-temperature slopes. *Phys. Earth Planet. Inter.*
738 114, 129-140.

739

Akins, J.A., Luo, S.N., Asimow, P.D., Ahrens, T.J., 2004. Shock-induced melting of $MgSiO_3$ perovskite and
740 implications for melts in Earth's lowermost mantle. *Geophys. Res. Lett.* 31, L14612.

741

Andraut, D., Bolfan-Casanova, N., Bouhifd, M.A., Guignot, N., Kawamoto, T., 2007. The role of Al-defects
742 on the equation of state of Al-(Mg,Fe)SiO₃ perovskite. *Earth Planet. Sci. Lett.* 263, 167-179.

743

Andraut, D., Bolfan-Casanova, N., Lo Nigro, G., Bouhifd, M.A., Garbarino, G., Mezouar, M., 2011. Melting
744 curve of the deep mantle applied to properties of early magma ocean and actual core-mantle boundary.
745 *Earth Planet. Sci. Lett.* 304, 251-259.

746

Andraut, D., Monteux, J., Le Bars, M., Samuel, H., 2016. The deep Earth may not be cooling down. *Earth*
747 *Planet. Sci. Lett.* 443, 195-203.

748

Andraut, D., Pesce, G., Bouhifd, M.A., Bolfan-Casanova, N., Henot, J.M., Mezouar, M., 2014. Melting of
749 subducted basalt at the core-mantle boundary. *Science* 344, 892-895.

750

Andraut, D., Petitgirard, S., Lo Nigro, G., Devidal, J.L., Veronesi, G., Garbarino, G., Mezouar, M., 2012.
751 Solid-liquid iron partitioning in Earth's deep mantle. *Nature* 487, 354-357.

752

Bajgain, S., Ghosh, D.B., Karki, B.B., 2015. Structure and density of basaltic melts at mantle conditions from
753 first-principles simulations. *Nature Communications* 6.

754

Benmore, C.J., Soignard, E., Amin, S.A., Guthrie, M., Shastri, S.D., Lee, P.L., Yarger, J.L., 2010. Structural
755 and topological changes in silica glass at pressure. *Phys. Rev. B* 81.

756

Bolfan-Casanova, N., Keppler, H., Rubie, D.C., 2003. Water partitioning at 660 km depth and evidence for
757 very low water solubility in magnesium silicate perovskite. *Geophys. Res. Lett.* 30, L017182.

758

Boujibar, A., Bolfan-Casanova, N., Andraut, D., Bouhifd, M.A., Trcera, N., 2016. Incorporation of Fe^{2+} and
759 Fe^{3+} in bridgmanite during magma ocean crystallization. *Am. Miner.* 101, 1560-1570.

760

Boukare, C.E., Ricard, Y., Fiquet, G., 2015. Thermodynamics of the MgO-FeO-SiO₂ system up to 140GPa:
761 Application to the crystallization of Earth's magma ocean. *J. Geophys. Res.: Solid Earth* 120, 6085-6101.

762

Boyet, M., Carlson, R.W., 2005. Nd-142 evidence for early (> 4.53 Ga) global differentiation of the silicate
763 Earth. *Science* 309, 576-581.

764

Caro, G., Bourdon, B., Wood, B.J., Corgne, A., 2005. Trace-element fractionation in Hadean mantle generated
765 by melt segregation from a magma ocean. *Nature* 436, 246-249.

766

Corgne, A., Liebske, C., Wood, B.J., Rubie, D.C., Frost, D.J., 2005. Silicate perovskite-melt partitioning of
767 trace elements and geochemical signature of a deep perovskitic reservoir. *Geochim. Cosmochim. Acta*
768 69, 485-496.

769

Davaille, A., 1999. A simultaneous generation of hotspots and superswells by convection in a heterogeneous
770 planetary mantle. *Nature* 402, 756-760.

771

Davies, C., Pozzo, M., Gubbins, D., Alfe, D., 2015. Constraints from material properties on the dynamics and
772 evolution of Earth's core. *Nat. Geosci.* 8, 678-+.

773

de Koker, N., Karki, B.B., Stixrude, L., 2013. Thermodynamics of the MgO-SiO₂ liquid system in Earth's
774 lowermost mantle from first principles. *Earth Planet. Sci. Lett.* 361, 58-63.

775

de Koker, N., Stixrude, L., 2009. Self-consistent thermodynamic description of silicate liquids, with
776 application to shock melting of MgO periclase and $MgSiO_3$ perovskite. *Geophys. J. Int.* 178, 162-179.

777

Du, Z., Lee, K.K.M., 2014. High-pressure melting of MgO from (Mg,Fe)O solid solutions. *Geophys. Res. Lett.*
778 41, 8061-8066.

779

Elkins-Tanton, L.T., 2008. Linked magma ocean solidification and atmospheric growth for Earth and Mars.
780 *Earth Planet. Sci. Lett.* 271, 181-191.

781 Fiquet, G., Auzende, A.L., Siebert, J., Corgne, A., Bureau, H., Ozawa, H., Garbarino, G., 2010. Melting of
782 Peridotite to 140 Gigapascals. *Science* 329, 1516-1518.

783 Funamori, N., Sato, T., 2010. Density contrast between silicate melts and crystals in the deep mantle: An
784 integrated view based on static-compression data. *Earth Planet. Sci. Lett.* 295, 435-440.

785 Guillot, B., Sator, N., 2007. A computer simulation study of natural silicate melts. Part II: High pressure
786 properties. *Geochim. Cosmochim. Acta* 71, 4538-4556.

787 Herzberg, C., Asimow, P.D., Ionov, D.A., Vidito, C., Jackson, M.G., Geist, D., 2013. Nickel and helium
788 evidence for melt above the core-mantle boundary. *Nature* 493, 393-U134.

789 Herzberg, C., Condie, K., Korenaga, J., 2010. Thermal history of the Earth and its petrological expression.
790 *Earth Planet. Sci. Lett.* 292, 79-88.

791 Herzberg, C., Zhang, J.Z., 1998. Melting experiments in the systems CaO-MgO-Al₂O₃-SiO₂ and MgO-SiO₂
792 at 3 to 15 GPa. *Am. Miner.* 83, 491-500.

793 Hirschmann, M.M., Asimow, P.D., Ghiorso, M.S., Stolper, E.M., 1999. Calculation of peridotite partial
794 melting from thermodynamic models of minerals and melts. III. Controls on isobaric melt production and
795 the effect of water on melt production. *Journal of Petrology* 40, 831-851.

796 Irifune, T., Koizumi, T., Ando, J., 1996. An experimental study of the garnet-perovskite transformation in the
797 system MgSiO₃-Mg₃Al₂Si₃O₁₂. *Phys. Earth Planet. Inter.* 96, 147-157.

798 Ito, E., Kubo, A., Katsura, T., Walter, M.J., 2004. Melting experiments of mantle materials under lower mantle
799 conditions with implications for magma ocean differentiation. *Phys. Earth Planet. Inter.* 143-144, 397-
800 406.

801 Katsura, T., Ito, E., 1990. Melting and subsolidus phase-relations in the MgSiO₃-MgCO₃- system at high-
802 pressures - Implications to the evolution of the Earth's atmosphere. *Earth Planet. Sci. Lett.* 99, 110-117.

803 Katsura, T., Yoneda, A., Yamazaki, D., Yoshino, T., Ito, E., 2010. Adiabatic temperature profile in the mantle.
804 *Phys. Earth Planet. Inter.* 183, 212-218.

805 Kellogg, L.H., Hager, B.H., van der Hilst, R.D., 1999. Compositional Stratification in the Deep Mantle.
806 *Science* 283, 1881-1884.

807 Kojitani, H., Akaogi, M., 1997. Melting enthalpies of mantle peridotite: calorimetric determinations in the
808 system CaO-MgO-Al₂O₃-SiO₂ and application to magma generation. *Earth Planet. Sci. Lett.* 153, 209-
809 222.

810 Labrosse, S., 2015. Thermal evolution of the core with a high thermal conductivity. *Phys. Earth Planet. Inter.*
811 247, 36-55.

812 Labrosse, S., Hernlund, J.W., Coltice, N., 2007. A crystallizing dense magma ocean at the base of the Earth's
813 mantle. *Nature* 450, 866-869.

814 Lange, R.A., 1997. A revised model for the density and thermal expansivity of K₂O-Na₂O-CaO-MgO-Al₂O₃-
815 SiO₂ liquids from 700 to 1900 K: extension to crustal magmatic temperatures. *Contributions to*
816 *Mineralogy and Petrology* 130, 1-11.

817 Lauterbach, S., McCammon, C.A., van Aken, P., Langenhorst, F., Seifert, F., 2000. Mössbauer and ELNES
818 spectroscopy of (Mg,Fe)(Si,Al)O₃ perovskite: a highly oxidized component of the lower mantle.
819 *Contribution in Mineralogy and Petrology* 138, 17-26.

820 Lebrun, T., Massol, H., Chassefiere, E., Davaille, A., Marcq, E., Sarda, P., Leblanc, F., Brandeis, G., 2013.
821 Thermal evolution of an early magma ocean in interaction with the atmosphere. *J. Geophys. Res.: Planets*
822 118, 1155-1176.

823 Lee, C.-T.A., Luffi, P., Hoink, T., Li, J., Dasgupta, R., Hernlund, J.W., 2010. Upside-down differentiation and
824 generation of a "primordial" lower mantle. *Nature* 463, 930-933.

825 Liebske, C., Corgne, A., Frost, D.J., Rubie, D.C., Wood, B.J., 2005. Compositional effects on element
826 partitioning between Mg-silicate perovskite and silicate melts. *Contrib. Mineral. Petrol.* 149.

827 Liebske, C., Frost, D.J., 2012. Melting phase relations in the MgO-MgSiO₃ system between 16 and 26 GPa:
828 Implications for melting in Earth's deep interior. *Earth Planet. Sci. Lett.* 345, 159-170.

829 Litasov, K., Ohtani, E., 2002. Phase relations and melt compositions in CMAS-pyrolite-H₂O system up to 25
830 GPa. *Phys. Earth Planet. Inter.* 134, 105-127.

831 Mao, W.L., Meng, Y., Shen, G., Prakapenka, V.B., Campbell, A.J., Heinz, D.L., Shu, J., Caracas, R., Cohen,
832 R.E., Fei, Y., Hemley, R.J., Mao, H.K., 2005. Iron-rich silicate in the Earth's D" layer. *Proc. Natl. Acad.*
833 *Sci. U. S. A.* 102, 9751-9753.

834 Martin, H., Moyen, J.F., 2002. Secular changes in tonalite-trondhjemite-granodiorite composition as markers
835 of the progressive cooling of Earth. *Geology* 30, 319-322.

836 Matas, J., Bass, J.D., Ricard, Y., Mattern, E., Bukowinsky, M.S., 2007. On the bulk composition of the lower
837 mantle: predictions and limitations from generalized inversion of radial seismic profiles. *Geophys. J. Int.*
838 170, 764-780.

- 839 Matsukage, K.N., Jing, Z.C., Karato, S., 2005. Density of hydrous silicate melt at the conditions of Earth's
840 deep upper mantle. *Nature* 438, 488-491.
- 841 Miller, G.H., Stolper, E.M., Ahrens, T.J., 1991. The Equation of State of a Molten Komatiite 2. Application to
842 Komatiite Petrogenesis and the Hadean Mantle. *J. Geophys. Res.* 96, 11,849-811,864.
- 843 Monteux, J., Andraut, D., Samuel, H., 2016. On the cooling of a deep terrestrial magma ocean. *Earth Planet.*
844 *Sci. Lett.* 448, 140-149.
- 845 Mosenfelder, J.L., Asimow, P.D., Frost, D.J., Rubie, D.C., Ahrens, T.J., 2009. The MgSiO₃ system at high
846 pressure: Thermodynamic properties of perovskite, postperovskite, and melt from global inversion of
847 shock and static compression data. *J. Geophys. Res.: Solid Earth* 114, B01203.
- 848 Mukhopadhyay, S., 2012. Early differentiation and volatile accretion recorded in deep-mantle neon and xenon.
849 *Nature* 486, 101-U124.
- 850 Murakami, M., Ohishi, Y., Hirao, N., Hirose, K., 2012. A perovskitic lower mantle inferred from high-
851 pressure, high-temperature sound velocity data. *Nature* 485, 90-U118.
- 852 Nakagawa, T., Tackley, P.J., 2010. Influence of initial CMB temperature and other parameters on the thermal
853 evolution of Earth's core resulting from thermochemical spherical mantle convection. *Geochem.,*
854 *Geophys., Geosyst.* 11, Q06001.
- 855 Nakajima, M., Stevenson, D.J., 2015. Melting and Mixing States of the Earth's Mantle after the Moon-Forming
856 Impact. *Earth Planet. Sci. Lett.*, in press.
- 857 Nomura, R., Hirose, K., Uesugi, K., Ohishi, Y., Tsuchiyama, A., Miyake, A., Ueno, Y., 2014. Low Core-
858 Mantle Boundary Temperature Inferred from the Solidus of Pyrolite. *Science* 343, 522-525.
- 859 Nomura, R., Ozawa, H., Tateno, S., Hirose, K., Hernlund, J.W., Muto, S., Ishii, H., Hiraoka, N., 2011. Spin
860 crossover and iron-rich silicate melt in the Earth's deep mantle. *Nature* 473, 199-202.
- 861 Panero, W.R., Pigott, J.S., Reaman, D.M., Kabbes, J.E., Liu, Z.X., 2015. Dry (Mg,Fe)SiO₃ perovskite in the
862 Earth's lower mantle. *J. Geophys. Res.: Solid Earth* 120, 894-908.
- 863 Petitgirard, S., Malfait, W.J., Sinmyo, R., Kuppenko, I., Hennem, L., Harries, D., Dane, T., Burghammer, M.,
864 Rubie, D.C., 2015. Fate of MgSiO₃ melts at core-mantle boundary conditions. *Proceedings of the*
865 *National Academy of Sciences* 112, 14186-14190.
- 866 Presnall, D.C., 1995. Phase diagrams of Earth-forming minerals, in: Ahrens, T. (Ed.), *Mineral Physics and*
867 *Crystallography*. AGU, Washington, D.C, pp. 248-268.
- 868 Presnall, D.C., Gasparik, T., 1990. Melting of enstatite (MgSiO₃) from 10 to 16.5 GPa and the forsterite
869 (Mg₂SiO₄) - majorite (MgSiO₃) eutectic at 16.5 GPa - Implications for the origin of the mantle. *J.*
870 *Geophys. Res.* 95, 15771-15777.
- 871 Revenaugh, J., Sipkin, S.A., 1994. Seismic evidence for silicate melt atop the 410 km mantle discontinuity.
872 *Nature* 369, 474-476.
- 873 Romanowicz, B., 1995. A global tomographic model of the shear attenuation in the upper mantle. *J. Geophys.*
874 *Res.: Solid Earth* 100, 12375-12394.
- 875 Rost, S., Garnero, E.J., Williams, Q., Manga, M., 2005. Seismological constraints on a possible plume root at
876 the core-mantle boundary. *Nature* 435, 666-669.
- 877 Rubie, D.C., Nimmo, H.J., Melosh, H.J., 2015. Formation of the Earth's core, in: Schubert, G. (Ed.), *Treatise*
878 *on Geophysics*, 2 ed. Elsevier, Amsterdam, pp. 43-79.
- 879 Sakamaki, T., Suzuki, A., Ohtani, E., 2006. Stability of hydrous melt at the base of the Earth's upper mantle.
880 *Nature* 439, 192-194.
- 881 Samuel, H., Farnetani, C.G., Andraut, D., 2005. Heterogeneous lowermost mantle: Compositional constraints
882 and seismological observables, in: VanDerHilst, R.D., Bass, J.D., Matas, J., Trampert, J. (Eds.), *Earth's*
883 *Deep Mantle: Structure, Composition, and Evolution*, pp. 101-116.
- 884 Sanloup, C., Drewitt, J.W.E., Konopkova, Z., Dalladay-Simpson, P., Morton, D.M., Rai, N., van Westrenen,
885 W., Morgenroth, W., 2013. Structural change in molten basalt at deep mantle conditions. *Nature* 503,
886 104-+.
- 887 Sato, T., Funamori, N., 2010. High-pressure structural transformation of SiO₂ glass up to 100 GPa. *Phys. Rev.*
888 *B* 82.
- 889 Shen, G., Lazor, P., 1995. Measurement of melting temperatures of some minerals under lower mantle
890 conditions. *J. Geophys. Res.* 100, 17699-17713.
- 891 Simon, F., Glatzel, G., 1929. Fusion-pressure curve. *Zeitschrift für anorganische und allgemeine Chemie* 178,
892 309-316.
- 893 Sleep, N.H., Zahnle, K.J., Lupu, R.E., 2014. Terrestrial aftermath of the Moon-forming impact. *Philos. Trans.*
894 *R. Soc., A* 372.
- 895 Solomatov, V.S., 2007. Magma Oceans and Primordial Mantle Differentiation, in: Schubert, G. (Ed.), *Treatise*
896 *on Geophysics*. Elsevier, Amsterdam, pp. 91-119.

897 Song, T.R.A., Helmberger, D.V., Grand, S.P., 2004. Low-velocity zone atop the 410-km seismic discontinuity
898 in the northwestern United States. *Nature* 427, 530-533.

899 Stebbins, J.F., Carmichael, I.S.E., Moret, L.K., 1984. Heat-capacities and entropies of silicate liquids and
900 glasses. *Contributions to Mineralogy and Petrology* 86, 131-148.

901 Stixrude, L., de Koker, N., Sun, N., Mookherjee, M., Karki, B.B., 2009. Thermodynamics of silicate liquids
902 in the deep Earth. *Earth Planet. Sci. Lett.* 278, 226-232.

903 Stixrude, L., Karki, B.B., 2005. Structure and freezing of MgSiO₃ liquid in the Earth's lower mantle. *Science*
904 310, 297-299.

905 Stolper, E.M., Ahrens, T.J., 1987. On the nature of pressure induced coordination changes in silicate melts or
906 glasses. *Geophys. Res. Lett.* 14, 1231-1233.

907 Tateno, S., Hirose, K., Ohishi, Y., 2014. Melting experiments on peridotite to lowermost mantle conditions. *J.*
908 *Geophys. Res.: Solid Earth* 119, 4684-4694.

909 Tauzin, B., Debayle, E., Wittlinger, G., 2010. Seismic evidence for a global low-velocity layer within the
910 Earth's upper mantle. *Nat. Geosci.* 3, 718-721.

911 Thomas, C.W., Asimow, P.D., 2013. Direct shock compression experiments on premolten forsterite and
912 progress toward a consistent high-pressure equation of state for CaO-MgO-Al₂O₃-SiO₂-FeO liquids. *J.*
913 *Geophys. Res.: Solid Earth* 118, 5738-5752.

914 Thomas, C.W., Liu, Q., Agee, C.B., Asimow, P.D., Lange, R.A., 2012. Multi-technique equation of state for
915 Fe₂SiO₄ melt and the density of Fe-bearing silicate melts from 0 to 161 GPa. *J. Geophys. Res.: Solid*
916 *Earth* 117.

917 Thomson, A.R., Walter, M.J., Lord, O.T., Kohn, S.C., 2014. Experimental determination of melting in the
918 systems enstatite-magnesite and magnesite-calcite from 15 to 80 GPa. *Am. Miner.* 99, 1544-1554.

919 Touboul, M., Puchtel, I.S., Walker, R.J., 2012. W-182 Evidence for Long-Term Preservation of Early Mantle
920 Differentiation Products. *Science* 335, 1065-1069.

921 Tronnes, R.G., Frost, D.J., 2002. Peridotite melting and mineral–melt partitioning of major and minor elements
922 at 22–24.5 GPa. *Earth Planet. Sci. Lett.* 197, 117-131.

923 van der Hilst, R.D., Karason, H., 1999. Compositional heterogeneity in the bottom 1000 kilometers of Earth's
924 mantle: Toward a hybrid convection model. *Science* 283, 1885-1888.

925 Vocadlo, L., Price, G.D., 1996. The melting of MgO - Computer calculations via molecular dynamics. *Physics*
926 *and Chemistry of Minerals* 23, 42-49.

927 Wang, Y.B., Sakamaki, T., Skinner, L.B., Jing, Z.C., Yu, T., Kono, Y., Park, C., Shen, G.Y., Rivers, M.L.,
928 Sutton, S.R., 2014. Atomistic insight into viscosity and density of silicate melts under pressure. *Nature*
929 *Communications* 5.

930 Wicks, J.K., Jackson, J.M., Sturhahn, W., 2010. Very low sound velocities in iron-rich (Mg,Fe)O: Implications
931 for the core-mantle boundary region. *Geophys. Res. Lett.* 37.

932 Wood, B.J., Rubie, D.C., 1996. The effect of alumina on phase transformations at the 660-kilometer
933 discontinuity from Fe-Mg partitioning experiments. *Science* 273, 1522-1524.

934 Yu, Y.G., Wentzcovitch, R.M., Vinograd, V.L., Angel, R.J., 2011. Thermodynamic properties of MgSiO₃
935 majorite and phase transitions near 660 km depth in MgSiO₃ and Mg₂SiO₄: A first principles study. *J.*
936 *Geophys. Res.: Solid Earth* 116.

937 Zeidler, A., Salmon, P.S., Skinner, L.B., 2014. Packing and the structural transformations in liquid and
938 amorphous oxides from ambient to extreme conditions. *Proc. Natl. Acad. Sci. U. S. A.* 111, 10045-10048.

939 Zerr, A., Bohler, R., 1993. Melting of (Mg,Fe)SiO₃-perovskite to 625 kbar: Indication of a high melting
940 temperature in the lower mantle. *Science* 262, 553–555.

941 Zhang, J., Herzberg, C., 1994. Melting experiments on anhydrous peridotite KLB-1 from 5.0 to 22.5 GPa. *J.*
942 *Geophys. Res.* 99, 17729-17742.

943

944