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1 **Reaction between mid-ocean ridge basalt and lower oceanic crust: An experimental**  
2 **study**

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11 **Key Points:**

- 12
- Melt-mush reactions proceed through diffusion-assisted dissolution and reprecipitation.
  - Reaction significantly shifts melt major element compositions, indicating it can limit the application of major-element barometers for MORB.
  - Clinopyroxene Mg#-Na-Ti and olivine Fo-Ni relationships in cumulates provide tracers for melt-mush reaction in nature.
- 16

17

**18 Abstract**

19 Reaction between mid-ocean ridge basalt (MORB) and crystal mush in the lower oceanic crust has  
20 been invoked to explain chemical variations of both MORB and minerals in the lower oceanic  
21 crust. Nonetheless, such reactions have been little studied experimentally. We conducted  
22 experiments to investigate the mechanisms and chemical consequences of melt-mush interaction  
23 by reacting molten MORB with troctolite at 0.5 GPa. Isothermal experiments demonstrate that  
24 melt infiltrates into troctolite with dissolution of plagioclase and olivine. The reacted melts have  
25 higher MgO and Al<sub>2</sub>O<sub>3</sub> and lower TiO<sub>2</sub> and Na<sub>2</sub>O contents, and crystallize more primitive olivine  
26 and plagioclase compared to those crystallized from the unreacted melts, suggesting melt-mush  
27 reaction could result in the formation of high-Al basalt. The melt composition variations induced  
28 by reaction also significantly affects the calculated pressures for MORB fractionation, indicating  
29 that major element-based barometers for MORB fractionation can only be used reliably if reaction  
30 can be ruled out. After reaction, the troctolite contains olivine with plagioclase inclusions and  
31 poikilitic clinopyroxene with partially resorbed olivine and plagioclase chadacrysts, indicating that  
32 melt-mush interaction occurs through dissolution-reprecipitation mechanisms. Clinopyroxene has  
33 high Mg# (>83) and elevated Na<sub>2</sub>O and TiO<sub>2</sub> contents, and olivine has different Fo vs. Ni  
34 correlations from fractional crystallization models, which provide testable parameters for the effect  
35 of melt-mush reaction in the rock record. By comparison with samples from lower oceanic crust  
36 and layered intrusions, we propose that melt-mush reaction plays an important role during magma  
37 transport in the crystal mush in both oceanic and continental magma systems.

**38 Plain Language Summary**

39 Magmas erupted at mid-ocean ridges represent the largest volcanic output from the Earth's  
40 interior, and have long been recognized as a probe to mantle composition and melting history.  
41 During its ascent from the mantle, magma crystallizes minerals in crustal magma reservoirs.  
42 Although crystallization modifies magma compositions, this process is well-understood and can  
43 therefore be corrected for. However, when crystallization proceeds, a network of interconnected  
44 minerals forms with small amounts of magma between them. Recent studies show that magma  
45 compositions might also be modified by reaction with minerals in this so-called crystal mush. Our  
46 study, for the first time, performed experiments to explore how such reactions work, and how they  
47 change the compositions of both magma and minerals. The results show that during reaction,  
48 magma can dissolve some minerals and crystallize others, which changes the magma  
49 compositions. Such variations in magma compositions challenge our understanding on the  
50 histories, depth of magma crystallization derived from them, and eventually affect our  
51 understanding of mantle composition. We also found that minerals in the mush carry distinct  
52 chemical signatures after reaction, which can be used as tracers for melt-mush reaction in nature.  
53 Comparison with natural mineral data suggests that such reaction is common in magma systems.

## 54 **1 Introduction**

55 Mid-ocean ridge basalt (MORB) has long been recognized as a probe to the composition  
56 of the upper mantle. Crustal level modifications to MORB compositions are largely attributed to  
57 fractional crystallization, which can be corrected through parameterizations of experimental data  
58 or thermodynamic calculations (e.g., Grove et al., 1992; Langmuir et al., 1992). However, studies  
59 on both MORB and plutonic rocks from the lower oceanic crust have provided evidence suggesting  
60 that the crustal evolution of MORB is complex, involving more than fractional crystallization  
61 alone (e.g., Lissenberg & MacLeod, 2016 and reference therein). Seismic studies indicate that mid-  
62 ocean ridges are mostly underlain by crystal mush (Canales et al., 2000; Carbotte et al., 2013;  
63 Crawford & Webb, 2002; Crawford et al., 1999; Dunn et al., 2000; Seher et al., 2010; Vera et al.,  
64 1990). Hence, melts and crystals coexist throughout the mid-ocean ridge magma plumbing system,  
65 providing two possible mechanisms for reactions to occur between melts and crystals. The first is  
66 during the emplacement of primitive melts into the mush (Kvassnes & Grove, 2008; Leuthold et  
67 al., 2018). Primitive melts are unlikely to be saturated in (all of) the phases present in the mush in  
68 which they are emplaced, leading to partial melting reactions. The second is during upward flow  
69 of interstitial melts in the mush (Coogan et al., 2000a; Gao et al., 2007; Lissenberg & Dick, 2008;  
70 Lissenberg et al., 2013). Numerical modeling of this process indicates that melt migrating through  
71 such a compacting mush will react with the constituent crystals (Solano et al., 2014), which is  
72 consistent with the rock record (Lissenberg & MacLeod, 2016). Therefore, melt-mush reaction is  
73 a natural consequence of the emplacement of primitive melt into the lower oceanic crust, and the  
74 percolation of (evolved) interstitial melt through the crystal mush.

75 Lower oceanic crustal sections exposed and/or drilled have provided a wealth of  
76 observations on the role of reactions in the magmatic evolution of the lower oceanic crust. This  
77 includes: Hess Deep in the East Pacific Rise (Coogan et al., 2002; Lissenberg et al., 2013); the  
78 Kane area (Coogan et al., 2000; Dick et al., 2008; Lissenberg & Dick, 2008), Atlantis Massif  
79 (Blackman et al., 2011), and 16.5°N (Sanfilippo et al., 2019) along the Mid-Atlantic Ridge;  
80 Atlantis Bank along the Southwest Indian Ridge (Dick et al., 2000; Gao et al., 2007; Lissenberg  
81 & MacLeod, 2016); Uraniwa Hills from the Central Indian Ridge (Sanfilippo et al., 2015a); and  
82 Godzilla Megamullion in the Parece Vela back-arc basin (Sanfilippo et al., 2013). Further evidence  
83 is present in the plutonic sections of ophiolites (Basch et al., 2018, 2019; Bédard, 1991; Bedard et  
84 al., 2000; Rampone et al., 2016; Sanfilippo et al., 2015b). The samples revealed complex zoning  
85 patterns, reaction textures, as well as mineral compositions and modes that could not be explained  
86 by fractional crystallization: instead, the data suggest that melt-mush reaction is a common process  
87 in the lower oceanic crust (Coogan et al., 2000a; Dick et al., 2002; Gao et al., 2007; Leuthold et  
88 al., 2018; Lissenberg & MacLeod, 2016; Lissenberg et al., 2013; Meyer et al., 1989; Ridley et al.,  
89 2006). Such a melt-mush reaction is also proposed to be common in continental magma systems  
90 (e.g., Bédard et al., 1988; Boudreau, 1999; Cashman et al., 2017; Irvine, 1980; Leuthold et al.,  
91 2015; Leuthold et al., 2014; Mathez, 1995; McBirney & Sonnenthal, 1990; Namur et al., 2013).

92 Phenocrysts and xenoliths in MORB samples provide further evidence of magma  
93 replenishment and subsequent reaction. Glassy MORB samples are widely known to host olivine  
94 and plagioclase crystals which are commonly not in equilibrium with their surrounding glass  
95 (Moore et al., 2014; Pan & Batiza, 2002, 2003; Bennett et al., 2019). Some of these samples contain  
96 phenocrysts proportions as high as 50% (Hellevang & Pedersen, 2008; Lange et al., 2013; Bennett  
97 et al., 2019), suggesting physical mixing and/or chemical reaction between a phenocryst-poor  
98 magma with a crystal-bearing mush. Melt-bearing cumulate xenoliths in MORB from the East

99 Pacific Rise and Iceland provide strong evidence for complex melt-mush reaction history in the  
100 lower oceanic crust, including the passage of multiple MORB magmas through the network of  
101 intergranular pores, and disequilibrium-induced dissolution and subsequent growth of crystals in  
102 the mush (Gurenko & Sobolev, 2006; Ridley et al., 2006). High-Al melt inclusion in olivine in  
103 MORB (e.g., Danyushevsky et al., 2003; Koleszar et al., 2009; Laubier et al., 2012) and high-Al  
104 MORB (e.g., Coumans et al., 2016; Yang et al., 2017), mostly with high MgO (>8 wt.%), were all  
105 proposed to form by reaction with a plagioclase-bearing cumulate in the lower oceanic crust.

106 Hence, the available data indicate that disequilibrium induced by both replenishment and  
107 porous flow of melt through a crystal mush is common in the magma plumbing system, and that the  
108 resulting reactions may play an important role in the evolution of the oceanic crust. Experimental  
109 works have been devoted to the reaction of melt with mantle rocks, including peridotite (e.g.,  
110 Daines & Kohlstedt, 1994; Lambart et al., 2009; Morgan & Liang, 2003, 2005; Saper & Liang,  
111 2014; Tursack & Liang, 2012; Van Den Bleeken et al., 2010, 2011; Wang et al., 2013), and dunite  
112 (e.g., Borghini et al., 2018), but very little attention has been paid to reaction of melt with crustal  
113 cumulates. Melt-cumulate interaction experiments have been conducted by melting plagioclase-  
114 olivine and plagioclase-augite mineral pairs at temperatures much higher than the melt-mineral  
115 saturation boundary (Kvassnes & Grove, 2008), which could be applicable during emplacement  
116 of primitive melt in a mush. However, in natural conditions, such interaction might not simply  
117 occur by melting cumulates and subsequent mixing with preexisting melt, especially during flow  
118 of interstitial melts in a primitive cumulate framework. Thus, it remains poorly understood by  
119 which mechanisms melt-mush reactions proceed, how and to what extent such processes would  
120 change MORB compositions and hence, how such interaction will affect reconstructions of mantle  
121 source compositions and melting processes. We conducted a series of melt-mush reaction  
122 experiments to investigate the mechanism and chemical consequences of such process by reacting  
123 both primitive and evolved MORB melts with a primitive cumulate (troctolite). The results have  
124 enabled a reconstruction of the melt-mush reaction mechanisms, and place constraints on the  
125 chemical consequences of melt-mush reaction on both the plutonic rocks and the melt.

## 126 **2 Experimental methods**

127 Reactive crystallization experiments characterizing melt-mush interaction were conducted  
128 at 1180°C and 1200°C (corresponding to the estimated liquidus temperatures for the two starting  
129 melts) and 0.5 GPa for 6-27 hrs using a 19.1-mm piston cylinder apparatus at Brown University.  
130 The reaction couple was formed by juxtaposing a powdered MORB against a troctolite in a  
131 graphite-lined molybdenum capsule (Figure 1).

### 132 **2.1 Starting materials**

133 The starting materials for the melts are a powdered primitive MORB glass with Mg#  
134 (defined as mole fractions of  $100 \cdot \text{MgO}/(\text{MgO} + \text{FeO}^{\text{T}})$ ) of 64 (KN182-13 D44A, provided by  
135 Alberto Saal) and moderately evolved MORB from the East Pacific Rise (Mg# 54; MOA8712-  
136 095, provided by John Sinton), with the latter representing average East Pacific Rise basalts. A  
137 lower oceanic crust troctolite analog was produced by combining 70 wt.% unzoned plagioclase  
138 (anorthite (An) 87, from Oman ophiolite gabbro sample 97OB1, provided by Benoit Ildefonse)  
139 and 30 wt.% unzoned olivine (forsterite (Fo) 90, from Kilbourne Hole, New Mexico, Sample  
140 KBH-1). In addition, a troctolitic mush analog was produced by combining 45 wt.% plagioclase,  
141 45 wt.% olivine and 10% interstitial primitive MORB glass of the same origin. Optically clean

142 mineral grains of approximately 100-150  $\mu\text{m}$  and glass chips of around 1-3 mm were first hand  
143 separated under a binocular microscope, mixed based on the appropriate weight percentages, and  
144 then ground under ethanol to obtain the desired starting materials. Afterward, the basalts and  
145 troctolite were stored in closed containers in a 110°C oven. Table 1 lists the starting mineral and  
146 melt compositions.

## 147 2.2 Experimental procedure

148 The experimental procedure was similar to that described in Tursack and Liang (2012) and  
149 Saper and Liang (2014). The furnace assembly consists of a molybdenum capsule in an MgO  
150 sleeve sandwiched between two crushable MgO spacers in a graphite, Pyrex and KCl salt sleeve.  
151 The molybdenum capsule (6.5 mm outer diameter and 10 mm long) was lined with a graphite inner  
152 sleeve (4 mm outer diameter, 2 mm inner diameter), which served as an inert buffer between  
153 molybdenum and the silicates. To form a reaction couple, we packed the bottom 2-3 mm of the  
154 graphite inner capsule with troctolite and the top 3-4 mm with the powdered basalt. The top of the  
155 capsule was then sealed with a 1-mm layer of graphite powder, and capped with a molybdenum  
156 lid (0.5-1 mm thick) (Figure 1). The furnace assembly was held in a 110°C oven for at least 12  
157 hrs.

158 The furnace assembly was wrapped with lead foil and loaded into the piston cylinder  
159 apparatus. To run an experiment, the assembly was first cold pressurized to slightly above 0.5 GPa.  
160 The temperature was then ramped to 1180°C (evolved melt) or 1200°C (primitive melt) at a rate  
161 of 75°C/min. After 6 hrs (27 hrs for run TE\_Ls) of reaction at the target temperatures, runs TE1,  
162 TE\_mush and TP1 were quenched, and the others were step-cooled to 1000°C (evolved melt) or  
163 1050°C (primitive melt) and subsequently quenched following the temperature-time history  
164 displayed in Figure 1. Pressure was kept at 0.5 GPa throughout the experiment. Temperature was  
165 measured using a W<sub>97</sub>Re<sub>3</sub>-W<sub>75</sub>Re<sub>25</sub> thermocouple and monitored using a Eurotherm controller with  
166 an uncertainty of 10°C (Morgan & Liang, 2005). After quenching the experiment, the charge was  
167 unloaded and the molybdenum capsule was extracted and sectioned longitudinally. The sectioned  
168 charges were mounted in epoxy, polished, and carbon coated for electron microprobe (EMPA) and  
169 scanning electron microscope (SEM) analyses (detailed methods can be found in the Supporting  
170 Information).

## 171 3 Results

172 Eight melt-mush reaction experiments were conducted in this study, supplemented by two  
173 runs on the melts alone. Figure 1 and Table 2 summarize the temperature-time history of the ten  
174 runs. Five experiments used the troctolite (T, 70% plagioclase, 30% olivine) and evolved MORB  
175 (E) melt (runs TE1, TE2, TE\_FC, TE\_SC, and TE\_Ls) and two the troctolite and primitive MORB  
176 (P; runs TP1 and TP2). The other melt-mush reaction experiment used evolved MORB and the  
177 troctolitic mush (45% olivine, 45% plagioclase and 10% primitive MORB; run TE\_mush).

178 Because we used the liquidus temperatures of the melts during the experiments, the basalt  
179 was liquid and troctolite was mainly solid with varied proportions of interstitial melt. In addition,  
180 two step-cooling experiments with the evolved and primitive MORB melts only (i.e., without  
181 troctolite; runs Es and Ps) were conducted as a reference to compare the crystallization sequence  
182 with and without reaction at the same pressure. Except for TE\_Ls, all runs were reacted at the  
183 respective liquidus temperatures for 6 h. TE1, TP1 and TE\_mush were quenched directly after 6 h  
184 reaction. TE2/Es and TP2/Ps were step-cooled to 1000°C and 1050°C, respectively. TE\_FC and

185 TE\_SC were cooled with constant cooling rates at 1°C/min and 0.1°C/min, respectively, to  
186 compare whether varied cooling rates would affect the results of the reaction experiment. TE\_Ls  
187 is a long duration run, which was reacted at 1180°C for 27 h before step-cooling to 1000°C. Our  
188 reaction experiments produced a variety of reaction textures and mineral compositional variations  
189 depending on the run conditions and the compositions of the reacting melt.

### 190 3.1 Textures of reactive products

#### 191 3.1.1 Isothermal experiments: Melt infiltration into the troctolite with dissolution of olivine and 192 plagioclase

193 Isothermal experiments TE\_mush, TE1, and TP1 all produced basalt at the top and  
194 interstitial melt-bearing troctolite at the bottom (Figure 2 and Figure S1 in Supporting  
195 Information). Euhedral clinopyroxenes crystallized in the upper part of the melt in TE1 (Figure  
196 S1a in Supporting Information). Relatively large olivines (~100 µm in length) crystallized near the  
197 melt-troctolite interface, and small olivine and plagioclase crystals are present in the upper part of  
198 the melt in run TP1 (Figure S1b in Supporting Information).

199 Interstitial melt phase in both TE1 and TP1 runs are volumetrically less than those in  
200 TE\_mush run due to the absence of melt phase in the starting troctolite in the former two runs  
201 (Figure S1 in Supporting Information). Nonetheless, melt infiltrated the troctolite in TE1 and TP1,  
202 with the amount of the interstitial melt gradually decreasing away from the melt-troctolite  
203 interface, forming a melt infiltration zone in the upper part of the troctolite (Figure 2). Plagioclase  
204 grains, much smaller than the starting plagioclase and mostly round in shape, were found to be  
205 included in olivine grains in the melt infiltration zone (Figure 2b). These plagioclase-bearing  
206 olivines are euhedral, in contrast to olivine grains below the zone. Because no plagioclase  
207 inclusions were present in the olivine starting material, this observation indicates that the euhedral  
208 olivine was precipitated either during or after plagioclase was partially dissolved.

#### 209 3.1.2 Step-cooling experiments: clinopyroxene precipitated as reaction product

210 Step-cooling experiments produced olivine-bearing gabbro and clinopyroxene-bearing  
211 troctolite from top to bottom (Figure 3). In the long duration run TE\_Ls, the gabbros contain  
212 relatively large clinopyroxene grains, feldspar laths, olivine, sulfide, and melt. Interstitial melt and  
213 clinopyroxene were present throughout the troctolite (Figure 3). Clinopyroxene shows poikilitic  
214 textures with olivine and plagioclase chadacrysts (Figure 3d), forming darker patches in the  
215 troctolite relative to interstitial melt bearing troctolite with a lighter green color in the element map  
216 (Figure 3a).

### 217 3.2 Chemical consequences of melt-mush reaction

#### 218 3.2.1 Chemical variations of melts in isothermal experiments

219 Overall, melt compositions gradually shift to higher MgO and Al<sub>2</sub>O<sub>3</sub>, lower FeO, and  
220 slightly lower TiO<sub>2</sub>, and Na<sub>2</sub>O contents towards the melt-troctolite interface (Figure 4). Mg# in the  
221 melt increased progressively from the original values for evolved and primitive melts at 54 and  
222 64, to the highest values close to the melt-troctolite interface at 59 and 68 for runs TE1 and TP1,  
223 respectively (Figure 4). Mg# of the melt at the interface of run TE\_mush increased to 68 at the  
224 interface, and further up to 74 for the interstitial melt in the troctolite (Figure 4). Due to the

225 crystallization of clinopyroxene in the upper part of the melt in run TE1, where the temperature is  
226 slightly lower than the center of the capsule (due to presence of a temperature gradient), melt  
227 compositions here shifted to higher Al<sub>2</sub>O<sub>3</sub>, lower MgO and FeO contents (Figure 4).

228 The large olivine crystals in the melt close to the melt-troctolite interface in run TP1 (Figure  
229 S1b in Supporting Information) have similar compositions (Fo 87.3) to those in the troctolite near  
230 the interface (Fo 87.5). Melts and olivine in the troctolite near interface in runs TE1, TE\_mush and  
231 TP1 as well as interstitial melt and olivine in the troctolite in TE\_mush all show Mg-Fe equilibrium  
232 (Figure 5). These observations suggests equilibrium of melt and troctolite near the interface.

### 233 3.2.2 Shifts in melt fractionation sequences following reaction: step-cooling experiments

234 To compare the melt fractionation sequence before and after reaction, step-cooling  
235 experiments of the two starting melts (i.e., without troctolite) were run. The highest Mg# of  
236 clinopyroxene and Fo of olivine in the melt phase in the step-cooling experiments can be used to  
237 discriminate whether clinopyroxene or olivine is the first phase to crystallize in the melt (Figure  
238 6). If the two are in Mg-Fe equilibrium, the melt should be close to olivine and clinopyroxene  
239 cotectic at the onset of crystallization. If the former plots above the equilibrium field (Figure 6),  
240 clinopyroxene is likely to have crystallized earlier than olivine, and vice versa. The mineral  
241 compositions in the step-cooling experiment for evolved melt only (run Es) are clinopyroxene with  
242 Mg# up to 82, and plagioclase with An up to 59 (not in Figure 6 due to the absence of olivine in  
243 run Es). After 6 h reaction with troctolite in TE1, melt compositions at the melt-troctolite interface  
244 have higher MgO, and Al<sub>2</sub>O<sub>3</sub> contents. In the corresponding step-cooling experiments TE2,  
245 TE\_FC, TE\_SC and TE\_Ls, clinopyroxene (Mg# up to 83-86) and olivine (Fo up to 80-85) in the  
246 gabbro sections are roughly in Mg-Fe equilibrium (Figure 6) and are thus close to cotectic.  
247 Plagioclase in these runs also have much higher An than those in run Es (up to 66-76, +17 mol%).  
248 Therefore, reaction with a primitive troctolite caused the melt, which was originally saturated with  
249 clinopyroxene as ferromagnesian phase, to be co-saturated with olivine. Furthermore, reaction led  
250 the melt to crystallize more primitive olivine, clinopyroxene and plagioclase during magma  
251 evolution than it would have done without reaction.

252 The mineral compositions in the step-cooling experiment for primitive melt only (run Ps)  
253 are olivine with Fo up to 84, clinopyroxene with Mg# up to 87, and plagioclase with An up to 68,  
254 with clinopyroxene and olivine in Mg-Fe equilibrium, indicating that magma evolution of the  
255 primitive melt at the run pressure should start with cotectic olivine and clinopyroxene followed by  
256 plagioclase fractionation. After 6 h reaction with troctolite in TP1, melt compositions at the melt-  
257 troctolite interface have higher MgO, and Al<sub>2</sub>O<sub>3</sub> contents. In the corresponding step-cooling run  
258 TP2, spinel crystals are found at the interface, and the reacted melt crystallized more primitive  
259 olivine (Fo up to 87, +3 mol%) and plagioclase (An up to 77, +9 mol%), and less primitive  
260 clinopyroxene (Mg# up to 82, -5 mol%) compared to run Ps (Figure 6). The presence of olivine  
261 crystals in melt phase near the melt-troctolite interface of run TP1 also indicates that olivine was  
262 the first phase to crystallize after reaction.

263 Although plagioclase crystallization is likely suppressed by the slightly higher pressure of  
264 0.5 GPa of the experiment compared to typical lower oceanic crustal conditions (0.1-0.2 GPa), the  
265 findings of more primitive olivine and plagioclase compositions after reaction with a primitive  
266 troctolite should apply at lower pressures as well. Clinopyroxene crystallized from the reacted  
267 melt, on the other hand, increased in Mg# from 82 to up to 86 in TE runs, but decreased from 87  
268 in run Ps to 82 in run TP2. Extensive olivine crystallization in the melt in run TP2 may have

269 lowered the melt in MgO contents, which thereafter crystallized clinopyroxene with lower Mg#  
270 than run Ps.

### 271 3.2.3 Troctolite compositional variations

272 The starting olivine in the troctolite has Fo of ~90 and NiO contents of ~0.37 wt.%. After  
273 reaction, the olivine compositions show a general decrease in both Fo and NiO contents towards  
274 the melt-troctolite interface in all runs (Figure 7a, b). Variations of olivine compositions in the  
275 troctolite are controlled by both the starting melt compositions and run durations. In isothermal  
276 experiments of TP1 and TE1 with same duration (6 h), olivine cores show significant shifts in the  
277 first 400  $\mu\text{m}$  of the troctolite (Figure 7a, b), corresponding to ~400  $\mu\text{m}$  melt infiltration (Figure 2  
278 and Figure S1 in Supporting Information). The far-field olivine in the troctolite retains same  
279 compositions to the starting olivine within error, and were hence not affected significantly by  
280 reactive melt infiltration. In contrast, the olivine core compositions in the troctolite of long-  
281 duration run TE\_Ls are characterized by more evolved compositions through the entire troctolite  
282 section, and are spatially more homogeneous, indicating that melt infiltrated and reacted to the end  
283 of the troctolite.

284 Olivines show sharper decreases in Fo for TE1 than TP1, as would be predicted by the  
285 differences in melt composition: melt Mg#-olivine Fo relationships (Figure 5) suggest that  
286 chemical equilibrium has been achieved at the interface of both TE and TP runs after 6 h reaction,  
287 indicating Fo in olivine are controlled by the Mg-Fe equilibrium between melt and troctolite.  
288 Hence, lower Mg# of the evolved melt leads to lower Fo in the olivines after reaction. In contrast  
289 to Fo, Ni in olivine compositions are similar between TP1 and TE1 (Figure 7a). Plagioclase, on  
290 the other hand, show clear core-to-rim variations with cores in all runs retaining their original  
291 compositions and rims having lower An numbers towards the melt-troctolite interface (Figure 7c).  
292 Interstitial clinopyroxene in the reacted troctolite in the step-cooling runs have high Na<sub>2</sub>O and TiO<sub>2</sub>  
293 contents (up to 0.81, and 2.51 wt.%, respectively) with high Mg# (>83).

## 294 4 Discussion

295 Melt and mineral compositions vary systematically across experimental charges. Through  
296 reaction with troctolite, melts become more enriched in MgO and Al<sub>2</sub>O<sub>3</sub> and depleted in TiO<sub>2</sub> and  
297 Na<sub>2</sub>O contents, and crystallize more primitive olivine and plagioclase compared to those  
298 crystallized from the starting melts. On the other hand, the troctolite shifted to more evolved  
299 compositions to be equilibrated with the melt near the melt-troctolite interface. These chemical  
300 variations record the progression of melt-mush reaction and provide important clues to the  
301 mechanisms of melt-mush interaction.

### 302 4.1 Mechanism of melt-mush reaction: diffusion-assisted dissolution and reprecipitation

303 Collectively, the observation of interstitial melt in the troctolite in runs TE1 and TP1, the  
304 gradual decrease in melt fraction further into the troctolite, and plagioclase relicts in euhedral  
305 olivines (Figure 2), suggest the incipient melt infiltration into the troctolite and the dissolution of  
306 plagioclase + olivine by melt. Plagioclase relicts in olivines can be explained by different  
307 dissolution and reprecipitation rates of olivine and plagioclase. Experimental study of the  
308 dissolution of olivine and plagioclase in basaltic magma suggest dissolution of plagioclase is ~6  
309 times faster than olivine, whereas the growth rate of plagioclase is at least 10 times slower than  
310 olivine (Donaldson, 1985). Hence, it is likely that melt dissolved plagioclase rapidly with minor

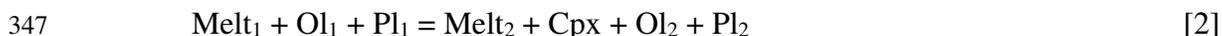
311 plagioclase reprecipitation, whereas olivine was more likely to be dissolved and reprecipitated  
 312 simultaneously. Therefore, during dissolution and precipitation of olivine, plagioclase is partially  
 313 dissolved and the relic grains were incorporated into growing olivine crystals (euhedral in Figure  
 314 2b).

315 The chemical variations of olivine and plagioclase in the troctolite after reaction show  
 316 different characteristics, also suggesting different reaction mechanism for olivine and plagioclase.  
 317 Even the core compositions of olivine crystals have been modified in all the experiments. In  
 318 contrast to olivine, the cores of plagioclase in all runs retained their original compositions, with  
 319 rims having lower An contents towards the melt-troctolite interface (Figure 7c). The difference in  
 320 core-to-rim compositions of olivine and plagioclase appears consistent with the observed fast  
 321 diffusion rates of Mg-Fe-Ni in olivine (e.g., Chakraborty, 1997) and slow diffusion of CaAl-NaSi  
 322 in plagioclase (e.g., Grove et al., 1984; Morse, 1984). To evaluate the control of diffusion on the  
 323 Mg-Fe equilibrium between melt and olivine at the interface, we modeled the effect of diffusion  
 324 on the core-to-rim variations of olivine crystals in the troctolite near the reaction interface in run  
 325 TE1. During the 6 h experimental duration, diffusion should only affect the Fo and Ni contents  
 326 within  $\sim 2 \mu\text{m}$  of the rim (Figure S2 in Supporting Information). However, even the core  
 327 compositions of olivine near the interface are in chemical equilibrium with the interface melt for  
 328 the isothermal runs, suggesting accelerated rate of reequilibration by diffusion-assisted dissolution  
 329 and reprecipitation. This is consistent with the presence of plagioclase inclusions in olivine crystals  
 330 (Figure 2b). Therefore, the reaction occurring in the troctolite during the isothermal experiments  
 331 is as follows:



333 The relative portions of plagioclase and olivine dissolved in the melt are discussed in Section 4.2.

334 The isothermal experiments TE1 and TP1 demonstrate that after 6 h of reaction, melt  
 335 dissolved troctolite around the margins of olivine and plagioclase, forming an interstitial melt  
 336 phase. In most cooling experiments, clinopyroxene formed around some of the olivine and  
 337 plagioclase grains of the troctolite during cooling to form the clinopyroxene-bearing troctolite  
 338 (Figure 3d). In the long duration run TE\_Ls, melt-troctolite reaction for 27 h has produced  
 339 relatively large porosity in the troctolite and interstitial melt phase is present in the entire troctolite  
 340 section (green in the element map in Figure 3a). Upon cooling, clinopyroxene started to form from  
 341 this interstitial melt around some of the olivine and plagioclase grains (Figure 3d), and reduced the  
 342 porosity of the troctolitic mush. The olivine chadacrysts are rounded as a result of earlier  
 343 dissolution, which is in contrast with the larger olivine grains not enclosed in clinopyroxene  
 344 (Figure 3d). These observations indicate that the reaction that occurred in the troctolite during the  
 345 step-cooling experiments included dissolution and reprecipitation of olivine and plagioclase as  
 346 well as the precipitation of clinopyroxene as the new mineral, following the reaction:



#### 348 4.2 Quantifying dissolution and reprecipitation

349 Systematic variations in the reacted melt compositions in runs TE1, TP1 and TE\_mush can  
 350 be used to quantify the dissolution (and reprecipitation) of plagioclase and olivine in the troctolite  
 351 during melt-mush reaction. We used the reaction in Eq. [1], which includes the roles of both  
 352 dissolution and reprecipitation of olivine in modifying melt compositions in the isothermal  
 353 experiments. We neglected plagioclase reprecipitation as a simplification. This is a reasonable

354 assumption since we observed no olivine relics in plagioclase and mostly quench growth of  
355 plagioclase rims. The oxides in the melt that change considerably, such as Al<sub>2</sub>O<sub>3</sub>, MgO, TiO<sub>2</sub>, and  
356 FeO, are used to calculate the net amount of dissolution. Although experimental study suggested  
357 dissolution of plagioclase is ~6 times faster than olivine in basaltic magmas (Donaldson, 1985),  
358 the actual dissolution rate of plagioclase to olivine may be lower considering activation enthalpy  
359 of dissolution and affinity of the minerals in the melt (Lissenberg & Dick, 2008). In our  
360 calculations, we modeled reaction using a range of plagioclase to olivine dissolution ratios (6:1,  
361 3:1, 2:1, and 1:1).

362 The starting evolved and primitive melt compositions were used to model the assimilation  
363 of the starting olivine and plagioclase with varied proportions and the reprecipitation of the olivine  
364 at the melt-troctolite interface for each run. The equilibrium between the core compositions of  
365 olivine near the interface with melt, which could not be explained by diffusion in 6 hours (Figure  
366 S2 in Supporting Information), suggests that the ratio of reprecipitated to dissolved olivine is  
367 relatively high. Our model suggests that high ratios (>0.6) would lead to decreasing MgO in melt,  
368 inconsistent with melt variations. With the reprecipitated to dissolved olivine ratios in the range  
369 0.4-0.6, the interface melt compositions can be accounted for with dissolved plagioclase/olivine  
370 ratios from 2:1 to 1:1. An example is shown in Figure 8 with a reprecipitated to dissolved olivine  
371 ratio of 0.5.

## 372 **5 Geological applications**

### 373 5.1 Comparison with natural observations

374 Previous petrologic studies of lower oceanic crust found that relatively primitive cumulates  
375 (troctolite, olivine gabbro) are commonly crosscut by more evolved gabbro at fast- (e.g., Hess  
376 Deep, ODP Hole 894; Natland & Dick, 1996), slow- (e.g., Kane Megamullion; Dick et al., 2008;  
377 Lissenberg & Dick, 2008; Atlantis Massif, Suhr et al., 2008) and ultraslow-spreading ridges (e.g.,  
378 ODP Hole 735B, Southwest Indian Ridge; Dick et al., 2000). Crystals (both olivine and  
379 plagioclase) in MORB are commonly more primitive than those in equilibrium with the host melt  
380 (Coogan, 2014), indicating that reaction between a more evolved phenocryst-poor magma and a  
381 more primitive crystal-rich mush is common. Microtextures such as poikilitic high-Mg#  
382 clinopyroxene with plagioclase and/or olivine chadacrysts, similar to the high-Mg# clinopyroxene  
383 produced in the experimental charges (Figure 3d), have been found in natural lower oceanic crust  
384 cumulates (e.g., Coogan et al., 2000a; Lissenberg & Dick, 2008; Lissenberg et al., 2013; Sanfilippo  
385 et al., 2013, 2019) and crust sections of ophiolites (e.g. Basch et al., 2019; Sanfilippo et al., 2015b).  
386 All these observations indicate that reactions of MORB with primitive cumulates, as per our  
387 experimental study, are a common process in the lower oceanic crust.

388 In this section, we compare the compositional variations in melts and minerals in our  
389 experimental charges with those observed in samples from lower oceanic crustal cumulates. We  
390 emphasize general mineral compositional trends rather than absolute values and ranges, as the  
391 latter depend specifically on melt composition, temperature, pressure, duration and melt-rock ratio  
392 during melt-mush reaction.

#### 393 5.1.1 Fo-Ni variations in olivine in natural lower oceanic crust cumulates

394 Fo and Ni variations in olivine of cumulates have been used as indicators for magma  
395 chamber processes (e.g., Coogan, 2014; Rampone et al., 2016; Sanfilippo et al., 2013; Suhr et al.,

396 2008). Our experiments suggest that primitive mush reacting with variously evolved magma would  
397 lead to a decrease in both the Fo and Ni contents of the olivine. Ni contents are predominantly  
398 controlled by the change in Ni partition coefficient between evolved and primitive melts. The  
399 higher the Mg# of the melt, the lower partition coefficients of Ni in olivine (Hart & Davis, 1978).  
400 Thus, the primitive melt in TP1 with higher Mg# would have a lower partition coefficient for Ni  
401 in olivine, indicating that reacting with a more primitive melt would decrease the Ni in olivine  
402 more rapidly than an evolved melt (Figure 9). Prolonged interaction with evolved melts (run  
403 TE\_Ls) leads to very low Ni contents (NiO < 0.1%; Figure 9).

404 To compare the relative effects of magma differentiation and melt-mush reaction on the Fo  
405 vs. Ni correlation of olivine in the cumulates, we modeled olivine compositional variations  
406 produced by the fractional crystallization of a primary MORB (ALV0527-001-001, Gale et al.,  
407 2013a) using MELTS (Ghiorso & Sack, 1995). As MORB are known to undergo sulfide-saturated  
408 differentiation before eruption (e.g., Bézous et al., 2005; Yang et al., 2014), and Ni, as a chalcophile  
409 element, is partly controlled by sulfide saturation during magma evolution, we modeled both  
410 sulfide-saturated and sulfide-undersaturated fractionation trends. Ni contents in the olivine were  
411 calculated with composition- and temperature-dependent  $K_D$  for Ni partitioning between olivine  
412 and melt (Matzen et al., 2013), and  $D_{Ni}^{sulf/melt}$  of 500 (Peach et al., 1990) for Ni partitioning between  
413 sulfide and melt. Sulfur-saturated differentiation used a silicate to sulfide fractionation ratio of  
414 1000, equivalent to a sulfide segregation rate of ~10 ppm per % fractional crystallization as  
415 estimated for MORB evolution in Bézous et al. (2005) and Yang et al. (2014). The gentler decreases  
416 in Ni for Fo lower than 84 in the modeled fractionation trends is due to the addition of  
417 clinopyroxene on the liquidus. In Figure 9, we also plot olivine phenocrysts compositions from  
418 MORB, as well as lower oceanic crustal cumulates from fast- (Hess Deep & Pito Deep, East Pacific  
419 Rise), slow- (MARK area, Hole 1309D, Mid-Atlantic Ridge) to ultraslow-spreading (Hole 735B,  
420 Southwest Indian Ridge) ridges. As demonstrated in Figure 9, a large number of olivines in lower  
421 oceanic crustal cumulates and olivine phenocrysts in MORB have both higher and lower NiO at a  
422 given Fo content than olivine from both sulfide-saturated and sulfide-undersaturated fractionation  
423 models, suggesting that olivine compositional variations in both MORB and cumulates cannot be  
424 explained by fractionation alone. Post-crystallization reactions of olivine in cumulates were  
425 suggested to have different effects on Fo and Ni contents, which were proposed to account for  
426 those olivines with both slightly higher Ni (Coogan, 2014; Suhr et al., 2008), and slightly lower  
427 Ni (Sanfilippo et al., 2013) at a given Fo. Our experimental work revealed that Ni variations in  
428 olivine after melt-mush reaction would largely depend on the Mg# of the reacted melt. Compared  
429 to the olivine from normal fractionation trends before clinopyroxene crystallization, higher Mg#  
430 in the primitive melt (run TP1) would result in olivine with lower Ni, while lower Mg# in the  
431 evolved melt (run TE1) produced olivine with higher Ni contents at high Fo (>84) in the reacted  
432 troctolite. Yet, prolonged reaction with troctolite by the evolved melt in run TE\_Ls produced  
433 olivine with much lower Ni than the fractionation trends after clinopyroxene crystallization at low  
434 Fo. Therefore, variably evolved melts reacting with primitive cumulates could be a possible  
435 explanation for the wide range of Ni variations in olivine in both lower oceanic crust cumulates  
436 and MORB globally (Figure 9).

#### 437 5.1.2 Origin of high-Mg# clinopyroxene in the lower oceanic crust

438 During fractional crystallization at crustal depth ( $\leq 2$  kbar), MORB is not predicted to  
439 crystallize clinopyroxene until after significant olivine and plagioclase fractionation (Grove et al.,  
440 1992). Therefore, the Mg# of the magma has already dropped considerably when clinopyroxene

441 saturates, with experiments providing an average Mg# of ~83 for the first clinopyroxene  
442 crystallized (Grove, 1990; Grove & Bryan, 1983; Grove et al., 1992; Tormey et al., 1987; Yang et  
443 al., 1996). Nonetheless, high-Mg# clinopyroxene ( $\text{Mg\#} \geq 85$ ) is not uncommon in the lower  
444 oceanic crust, and has been found in the Atlantic (Kane Megamullion (Lissenberg & Dick, 2008);  
445 IODP Hole 1309D in the Atlantis Massif (Drouin et al., 2009; Suhr et al., 2008)), Pacific (Pito  
446 Deep (Perk et al., 2007)) and Indian (ODP Hole 735B (Dick et al., 2002)) oceans. Both high-  
447 pressure crystallization (Elthon, 1987) and melt-mush reaction (Lissenberg & Dick, 2008) have  
448 been invoked to explain such high-Mg# clinopyroxene. Olivine gabbros collected from within 1  
449 km of the base of the sheeted dike complex at Pito Deep contain clinopyroxene with Mg# up to 89  
450 (Perk et al., 2007), demonstrating that Mg-rich clinopyroxene can also crystallize during MORB  
451 differentiation at low pressures.

452 The Mg# variations of clinopyroxene in the melt portions of TE and TP runs mainly depend  
453 on the extent to which olivine crystallization preceded clinopyroxene crystallization. By reaction  
454 with troctolite in TP1, the olivine-saturated primitive melt increased its Mg#, and crystallized  
455 olivine with high Fo (up to 87) as a result (Figure 6). Such early crystallization of magnesian  
456 olivine would deplete the melt in MgO, which explains the relatively low Mg# of clinopyroxene  
457 (~82; Figure 6) formed upon the step cooling of this reacted melt (experiment TP2). In contrast,  
458 the Fe-Mg systematics indicate that olivine crystallization did not precede clinopyroxene during  
459 the crystallization of the evolved melt, even after reaction (experiments TE2, TE\_SC and TE\_Ls;  
460 Figure 6). The increased Mg# in the melt after reaction, therefore, leads to the formation of high-  
461 Mg# clinopyroxene (Mg# up to 86). Although clinopyroxene crystallization is likely enhanced by  
462 the slightly higher pressure of 0.5 GPa of the experiments compared to typical lower oceanic  
463 crustal conditions (0.1-0.2 GPa), the presence of high-Mg# clinopyroxene in the TE runs but not  
464 in TP runs indicates that the factor controlling high-Mg# clinopyroxene precipitation is not higher  
465 pressure as both runs were at the same pressure. Hence, high-Mg# clinopyroxene is most likely to  
466 form when a relatively evolved, clinopyroxene-saturated melt reacts with primitive cumulates.  
467 This conclusion, which is not sensitive to crystallization pressure, is in keeping with the suggested  
468 formation of high-Mg# clinopyroxene in natural oceanic (Lissenberg & Dick, 2008; Lissenberg &  
469 MacLeod, 2016) and ophiolitic (Basch et al., 2018, 2019; Rampone et al., 2016; Sanfilippo et al.,  
470 2015b) gabbros.

471 The clinopyroxenes crystallized in the (reacted) melt portions of the experiments contain  
472 moderate  $\text{TiO}_2$  and  $\text{Na}_2\text{O}$  contents, as is expected from fractional crystallization models (Figure  
473 10). Interstitial clinopyroxene in the troctolite in the step-cooling runs, on the other hand, extend  
474 to very high  $\text{Na}_2\text{O}$  and  $\text{TiO}_2$  contents (up to 0.81, and 2.51 wt.%, respectively) with high Mg#  
475 (>83), distinct from the clinopyroxene compositions predicted to form by fractional crystallization  
476 (Figure 10). We infer that these titanium and sodium-rich clinopyroxenes form from evolved  
477 interstitial melts (i.e., high  $\text{TiO}_2$ ) that equilibrated with primitive olivine, and thus acquired high  
478 Mg# (cf. Borghini and Rampone, 2007; Borghini et al., 2018; Lissenberg & Dick, 2008). There  
479 are a significant amount of clinopyroxenes in lower oceanic crustal cumulates with compositional  
480 shifts toward higher  $\text{Na}_2\text{O}$  and  $\text{TiO}_2$  at high Mg# relative to the fractionation trend (Figure 10 a,b),  
481 indicating their compositions might have been affected by melt-mush reaction (Lissenberg & Dick,  
482 2008; Lissenberg & MacLeod, 2016; Sanfilippo et al., 2019). Furthermore, clinopyroxene from  
483 some lower crustal sections of ophiolites and layered intrusions have even higher  $\text{Na}_2\text{O}$  and  $\text{TiO}_2$   
484 contents (Figure 10 c,d), suggesting an important role for such melt-mush reaction in the formation  
485 of both continental and oceanic cumulates.

## 5.2 Implications for MORB major element barometry and the origin of high-Al MORB

Major element variations of MORB are commonly used to calculate the pressures at which they underwent (partial) crystallization in the crust prior to eruption (e.g., Danyushevsky et al., 1996; Grove et al., 1992; Herzberg, 2004; Villiger et al., 2007; Wanless & Behn, 2017; Yang et al., 1996). Such calculations depend heavily on the MgO-CaO-Al<sub>2</sub>O<sub>3</sub> systematics of the melt with higher pressures resulting from higher Al<sub>2</sub>O<sub>3</sub> and/or lower CaO, reflecting the more prominent role of clinopyroxene crystallization at higher pressure (Lissenberg & Dick, 2008). Our experimental results demonstrate that interaction of melts with primitive cumulate minerals modifies melt compositions, with higher Al<sub>2</sub>O<sub>3</sub> and MgO after reaction in the isothermal experiments (Figure 4). Such compositional changes of the melt may significantly affect the calculated crystallization pressure. To determine the effect of melt-mush reaction on the calculated crystallization pressure, we applied the parameterization of Herzberg (2004) to the evolved melt compositions in TE1 runs, but note that similar results would be obtained with other MORB major element barometers (Danyushevsky et al., 1996; Grove et al., 1992; Villiger et al., 2007; Yang et al., 1996). We chose the melt compositions which match the criteria proposed in Herzberg (2004) to be saturated in clinopyroxene, olivine and plagioclase (CaO < -0.3MgO+14.5). As shown in Figure 11, melt-mush reaction would result in an increase in the calculated crystallization pressures from ~0.1 GPa to above 0.4 GPa, corresponding to an apparent shift in crystallization depth of >12 km. Such a result is consistent with previous predictions by Lissenberg and Dick (2008), who stated that the MgO-CaO-Al<sub>2</sub>O<sub>3</sub> relationships of MORB could result from melt-mush reaction instead of polybaric fractional crystallization. Hence, melt-mush reaction significantly affect MORB major-element barometers, resulting in erroneous pressures. It follows that such barometers can only be used reliably if reaction can be ruled out.

High-Al MORB, characterized by high Al<sub>2</sub>O<sub>3</sub> and MgO (mostly >8 wt.%), as well as low SiO<sub>2</sub> (e.g. 48 wt. %) and low TiO<sub>2</sub> contents, mostly occur at slow-spreading ridges or close to fracture zones and ridge terminations (e.g., Eason & Sinton, 2006; Gale et al., 2014; Gale et al., 2013b; Langmuir & Bender, 1984; Laubier et al., 2012; Melson & O'Hearn, 1979; Meyzen et al., 2003; Standish et al., 2008; Yang et al., 2017). They are proposed to result from either high-pressure clinopyroxene crystallization (Eason & Sinton, 2006) or melt-cumulate reaction (Danyushevsky et al., 2004; Danyushevsky et al., 2003; Gale et al., 2013b; Kamenetsky et al., 1998; Laubier et al., 2012; Lissenberg & Dick, 2008; Slater et al., 2001; Yang et al., 2017). Our experiments indicate that plagioclase is dissolved more rapidly than olivine, and the dissolution of plagioclase is much faster than its reprecipitation during melt-mush reaction. The preferential dissolution relative to reprecipitation of plagioclase would also result in MORB melts with higher Al<sub>2</sub>O<sub>3</sub> contents (Figure 4), which would precipitate plagioclase with higher-An (up to 17 mol% higher as in step-cooling TE runs) upon cooling, indicating melt-mush reaction is a possible mechanism for the formation of high-Al MORB.

## 6 Conclusions

To investigate the mechanisms and chemical consequences of reaction between basaltic magma and primitive cumulates, we conducted melt-mush reaction experiments by juxtaposing troctolite with either a primitive or an evolved MORB. From our results and comparison to natural data, we draw the following conclusions:

1. Melt-mush reactions proceed through diffusion-assisted dissolution and reprecipitation. The troctolite in the isothermal experiments shows interstitial melt phase after reaction, indicating

530 melt infiltration and dissolution of plagioclase and olivine with plagioclase/olivine ratios of 2 to  
531 1. Even the cores of olivine crystals in the troctolite at the interface have achieved Mg-Fe  
532 equilibrium with the melt, and they contain plagioclase inclusions, neither of which can be  
533 accounted for by diffusion on the time scale of the experiments, indicating reprecipitation of  
534 olivine during reaction. Furthermore, the troctolite in the step-cooling experiments show both  
535 interstitial melt and poikilitic clinopyroxene with rounded olivine and plagioclase chadacrysts,  
536 indicating clinopyroxene precipitated upon cooling as reaction product.

537 2. Through reaction with troctolite, melts became more enriched in MgO and Al<sub>2</sub>O<sub>3</sub> and  
538 depleted in TiO<sub>2</sub> and Na<sub>2</sub>O contents, and crystallized more primitive olivine and plagioclase  
539 compared to those crystallized from the starting melts. On the other hand, the troctolite shifted to  
540 more evolved compositions to be equilibrated with the melt near the melt-troctolite interface.  
541 Olivines in the troctolite have different Fo vs. Ni correlations for different starting melt  
542 compositions, resulting from varied Ni partition coefficients with different MgO contents of the  
543 melt. Clinopyroxene precipitated in the troctolite has high Mg# (>83) with enriched Na<sub>2</sub>O and  
544 TiO<sub>2</sub> contents, distinct from those crystallized in the melts. Therefore, high-Mg# clinopyroxene  
545 can result from melt-mush reaction, and cannot simply be used as evidence for high-pressure  
546 crystallization.

547 3. The higher MgO and Al<sub>2</sub>O<sub>3</sub> contents of reacted melts significantly affect the pressures  
548 inferred from MORB major element barometers. The reacted melt yielded higher calculated  
549 pressures (~ 0.4 – 0.5 GPa) than that calculated from the starting compositions (~ 0.1 GPa),  
550 indicating that major element-based barometers for MORB fractionation can only be used reliably  
551 if reaction can be ruled out.

552 4. The Fo-Ni correlations of olivine, as well as Mg# vs. TiO<sub>2</sub> and Na<sub>2</sub>O correlations of  
553 clinopyroxene in cumulate rocks can be used to identify the role of melt-mush reaction in the  
554 chemical variations of cumulates. By comparison with cumulates from lower oceanic crust,  
555 ophiolite and layered intrusion, we propose that melt-mush reaction plays an important role during  
556 magma emplacement and transport in crystal mush in both oceanic and continental magma  
557 systems.

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871 augite saturated melts from 0.001 to 10 kbar. *Contributions to Mineralogy and Petrology*, 124(1), 1-18.

873 **Figure Captions**

874 **Figure 1.** Setup and cooling history of all the experimental charges in this study. Time = 0 marks  
 875 the beginning of the experiment, when the target temperatures of 1180°C and 1200 °C were  
 876 reached. See the text in Section 3 for detailed description of each run.

877 **Figure 2.** Texture of isothermal run TE1. a) Backscattered electron (BSE) image for run TE1  
 878 showing interstitial melt phases in the troctolite (~5% in panel 2b, and almost none in 2d),  
 879 forming a melting-infiltration zone in the troctolite. b) shows plagioclase (pl) included in  
 880 euhedral olivine (ol) grains, indicating that the troctolite underwent dissolution-  
 881 reprecipitation reactions. c) interstitial melt phases occur mainly as thin film surrounding  
 882 plagioclase, whereas d) at the bottom of the troctolite section without interstitial melt phase,  
 883 plagioclase boundaries are obscure. e) Ti element map of the 400 µm melt infiltration zone  
 884 in run TE1 highlights the melt phase in green, showing more interstitial melt in the  
 885 troctolite closer to the melt-troctolite interface. The locations of a) and e) are shown in the  
 886 full BSE image of TE1 in Figure S1a in Supporting Information

887 **Figure 3.** Texture of long duration step-cooling run TE\_Ls. a) Element map showing the texture  
 888 of upper gabbro and lower troctolite. Troctolite section is composed of plagioclase (pl, dark  
 889 grey color representing low-Na plagioclase in the troctolite) and olivine (ol, bright red  
 890 color) with interstitial clinopyroxene (cpx, darker red color surrounding olivine and  
 891 plagioclase) in b), and interstitial melt (green color) in c). Plagioclase in the gabbro has a  
 892 lighter blue color due to higher Na contents in the plagioclase crystallizing from the melt  
 893 compared to those in the troctolite. d) backscattered electron image for poikilitic  
 894 clinopyroxene (outlined with yellow dashed curve) enclosing plagioclase and olivine  
 895 chadacrysts (outlined with white curves and labeled with white fonts) in the troctolite  
 896 section. Plagioclase relict hosted in olivine suggests reprecipitated olivine grew over  
 897 resorbed plagioclase. The olivine chadacrysts in the poikilitic clinopyroxene are rounded as  
 898 a result of dissolution, in contrast with the larger olivine grains (labeled with yellow fonts)  
 899 not enclosed in clinopyroxene

900 **Figure 4.** Plots of Mg# (defined as mole fractions of  $100 \cdot \text{MgO}/(\text{MgO} + \text{FeO}^{\text{T}})$ ) and oxides in  
 901 melt as a function of distance away from the melt-troctolite interface in runs TE1, TP1, and  
 902 TE\_mush. Logarithmic scales were used to plot the  $\text{TiO}_2$  variations in run TE1 and TP1.  
 903 Solid lines correspond to the starting melt compositions. Clinopyroxene is present in the  
 904 upper part of melt phase in run TE1, represented by the area to the right of the dashed line  
 905 in a) and b). Interstitial melt compositions in the troctolite were also plotted for TE\_mush,  
 906 shown to the left of the dashed line in e) and f)

907 **Figure 5.** Plot of Mg# in melt and fosterite content of olivine (Fo) in the troctolite near interface  
 908 for runs TE1, TE\_mush and TP1. Grey and black dashed curves indicate equilibrated melt  
 909 and olivine compositions with  $K_D$  of  $0.30 \pm 0.03$  (e.g., Roeder & Emslie, 1970). Grey circle  
 910 and triangle represent the starting melt and olivine compositions of run TE1, TE\_mush and  
 911 TP1, respectively. Blue circle, purple diamond and orange triangles represent melt and  
 912 olivine compositions near the melt-troctolite interface for the three runs after reaction, and  
 913 they are plotted within the field defined by  $K_D$  of  $0.30 \pm 0.03$ , suggesting chemical  
 914 equilibration was achieved at the interface after reaction. Olivine in both the troctolite (tr)  
 915 and melt (m) are plotted for run TP1.

916 **Figure 6.** Plot of the highest Mg# of clinopyroxene and Fo of olivine in the gabbro sections of  
 917 each step-cooling run. Grey shaded area represents clinopyroxene and olivine compositions  
 918 in equilibrium, defined by the Fe-Mg exchange equilibrium constant  $K_D^{\text{cpx/melt}}$  from 0.23-  
 919 0.26 (Grove & Bryan, 1983; Sisson & Grove, 1993) and  $K_D^{\text{ol/melt}}$  from 0.27-0.33 (e.g.,  
 920 Roeder & Emslie, 1970). Experimental charges plotted below the equilibrium field indicate  
 921 olivine crystallized early than clinopyroxene and vice versa, demonstrating the first phase to  
 922 crystallize in the gabbro sections changed from cotectic clinopyroxene and olivine to olivine  
 923 in TP runs (shown by the orange line with an arrow), and from clinopyroxene to cotectic  
 924 olivine and clinopyroxene in TE runs

925 **Figure 7.** Plots of a) Fo and b) NiO contents in the olivine cores for runs TP1, TE1 and TE\_Ls  
 926 and c) Anorthite (An) in the cores and rims of plagioclase for run TE1 in the troctolite as a  
 927 function of distance away from the melt-troctolite interface. Rim analyses were not plotted  
 928 as most of the olivine are too small to yield good-quality data on the rims due to the  
 929 possible effect of secondary fluorescence contamination (e.g., Llovet & Galan, 2003). Grey  
 930 shaded fields represent the starting olivine and plagioclase compositions

931 **Figure 8.** Modeling of melt compositional variations during melt-troctolite reaction in runs TE1  
 932 (a-c), TP1 (d-f), and TE\_mush (g-i). Grey circle and triangle represent the starting  
 933 compositions of evolved and primitive melts, respectively. Black lines are for reaction  
 934 models with the actual mineral proportions in the troctolite, and lines with other colors  
 935 show different dissolved plagioclase to olivine ratios as noted. Marks on the modeled lines  
 936 denote the relative percentage of assimilants (in 5% increments). Different symbols indicate  
 937 varied distances from the analyzed melt spots to the melt-troctolite interface.

938 **Figure 9.** Variation in olivine Fo and Ni contents in both melt/gabbro and troctolite (tr.) sections  
 939 in the experimental charges. Light and dark grey dots represent olivine in global lower  
 940 oceanic cumulates (data are from Dick et al., 2002; Hébert et al., 1991; Lissenberg et al.,  
 941 2013; Miller et al., 2009; Natland & Dick, 1996; Perk et al., 2007; Ross & Elthon, 1997;  
 942 Suhr et al., 2008; Yamazaki et al., 2009) and olivine phenocrysts in MORB (from PetDB  
 943 database), respectively. Orange dots represent sulfur-undersaturated fractionation trend of a  
 944 primary MORB (ALV0527-001-001, Gale et al., 2013a). From this melt olivine, followed  
 945 by plagioclase and clinopyroxene is removed by fractional crystallization, and each dot  
 946 represents a 1°C temperature step, modeled using MELTS (Ghiorso & Sack, 1995). Blue  
 947 dots represent the sulfur-saturated fractionation trend from the same melt with a  
 948 silicate/sulfide fractionation ratio of 1000 based on the sulfide segregation rates during  
 949 MORB differentiation from Bézou et al. (2005) and Yang et al. (2014).

950 **Figure 10.** Variations in TiO<sub>2</sub> and Na<sub>2</sub>O contents versus Mg# of clinopyroxene in both  
 951 melt/gabbro (m) and troctolite (tr) sections in runs TE and TP. Grey dots represent  
 952 clinopyroxene in a&b) lower oceanic cumulates (data are from Coogan et al., 2002; Coogan  
 953 et al., 2000b; Dick et al., 2002; Hébert et al., 1991; Lissenberg et al., 2013; Miller et al.,  
 954 2009; Natland & Dick, 1996; Perk et al., 2007; Ross & Elthon, 1997; Sanfilippo et al.,  
 955 2019; Suhr et al., 2008; Yamazaki et al., 2009), and c&d) Erro-Tobbio ophiolite (Basch et  
 956 al., 2019) and Panzhihua layered intrusion (Pang et al., 2009). Black dots represent  
 957 fractional crystallization trend of the same MORB magma as described in Figure 9. Arrows  
 958 show the effect of melt-mush reaction on the clinopyroxene compositions

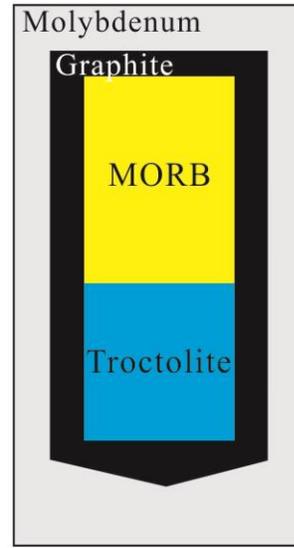
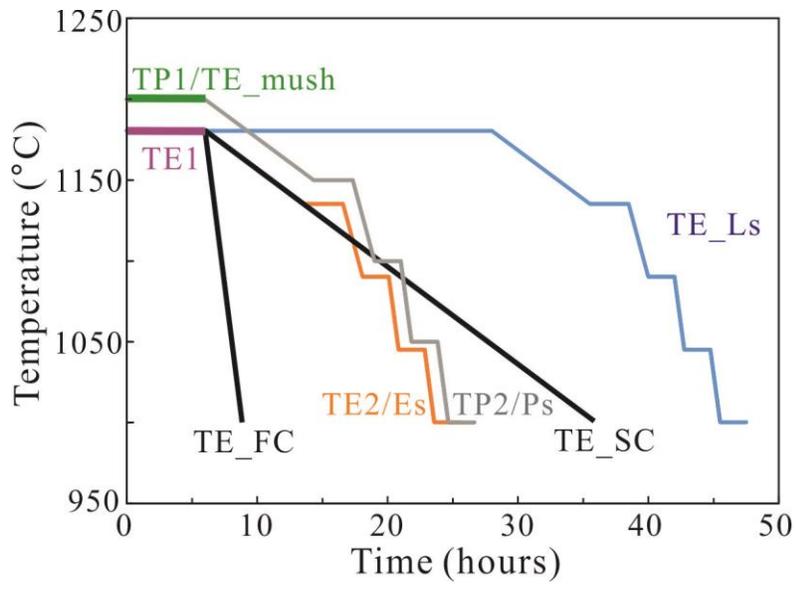
959 **Figure 11.** Plot of fractionation pressures calculated using the method in Herzberg (2004) as a  
960 function of distance away from the melt-troctolite interface in run TE1. Melt compositions  
961 that match the criteria of  $\text{CaO} < -0.3\text{MgO} + 14.5$  as proposed in Herzberg (2004) were  
962 chosen for calculation. Orange arrow shows the trend for the effect of melt-mush reaction  
963 on the calculated pressures. Grey line indicates the calculated pressure for the starting  
964 evolved melt composition as listed in Table 1

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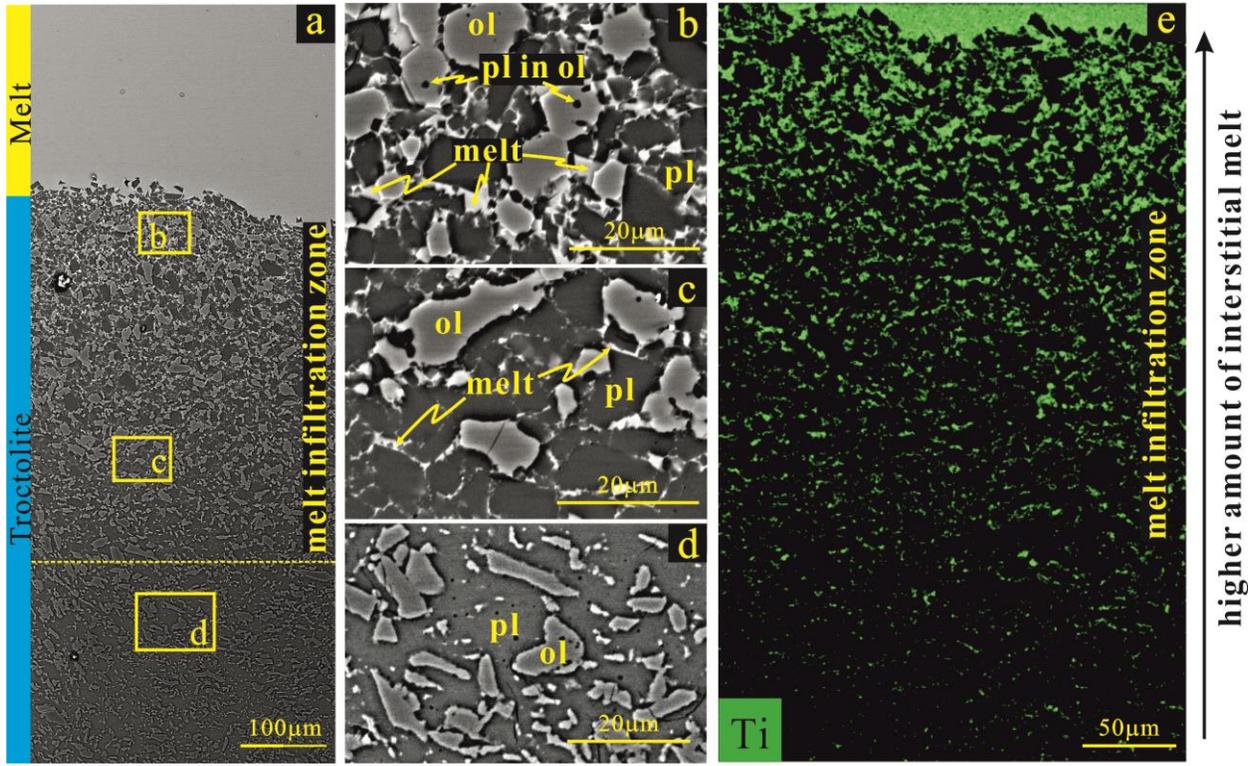
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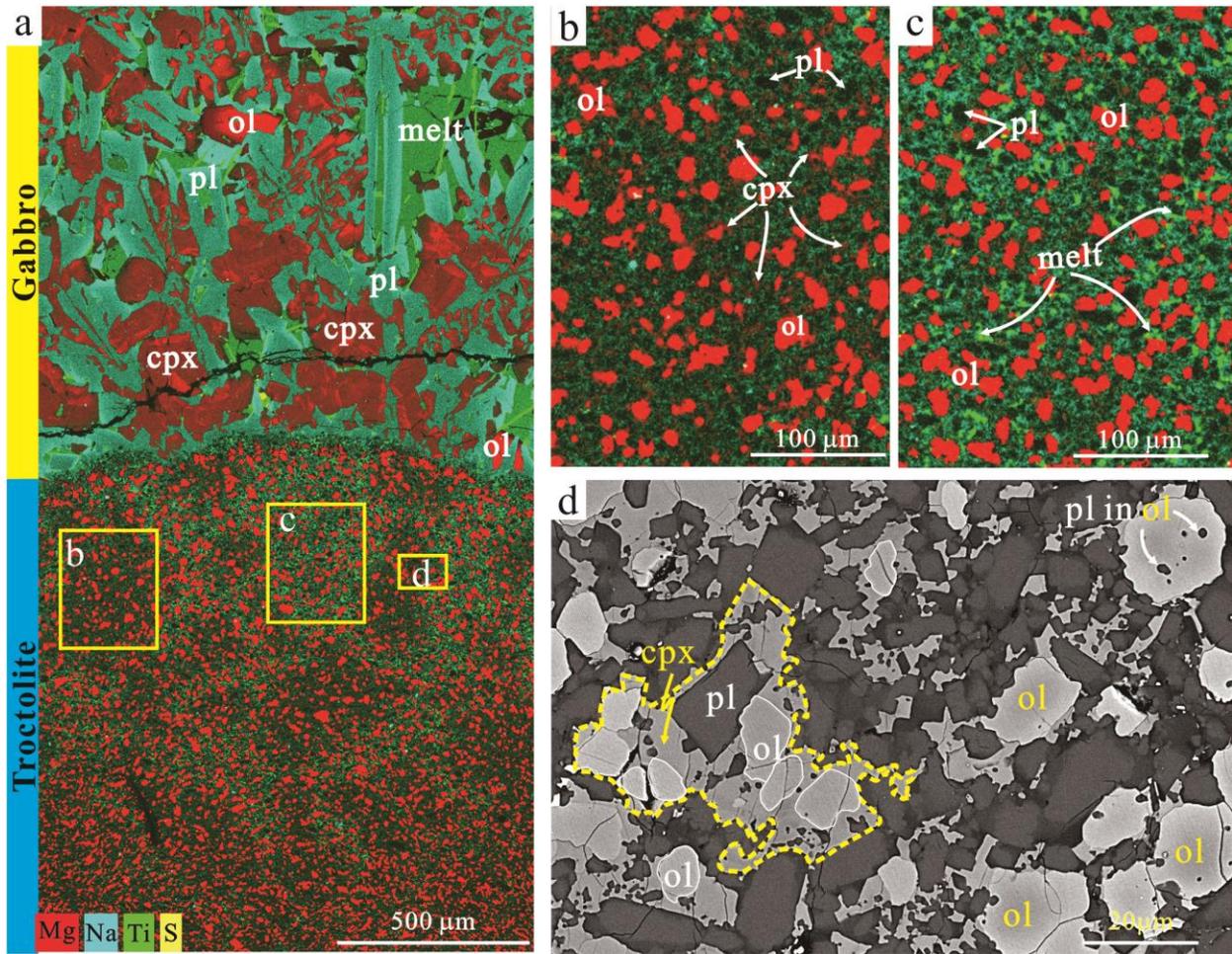
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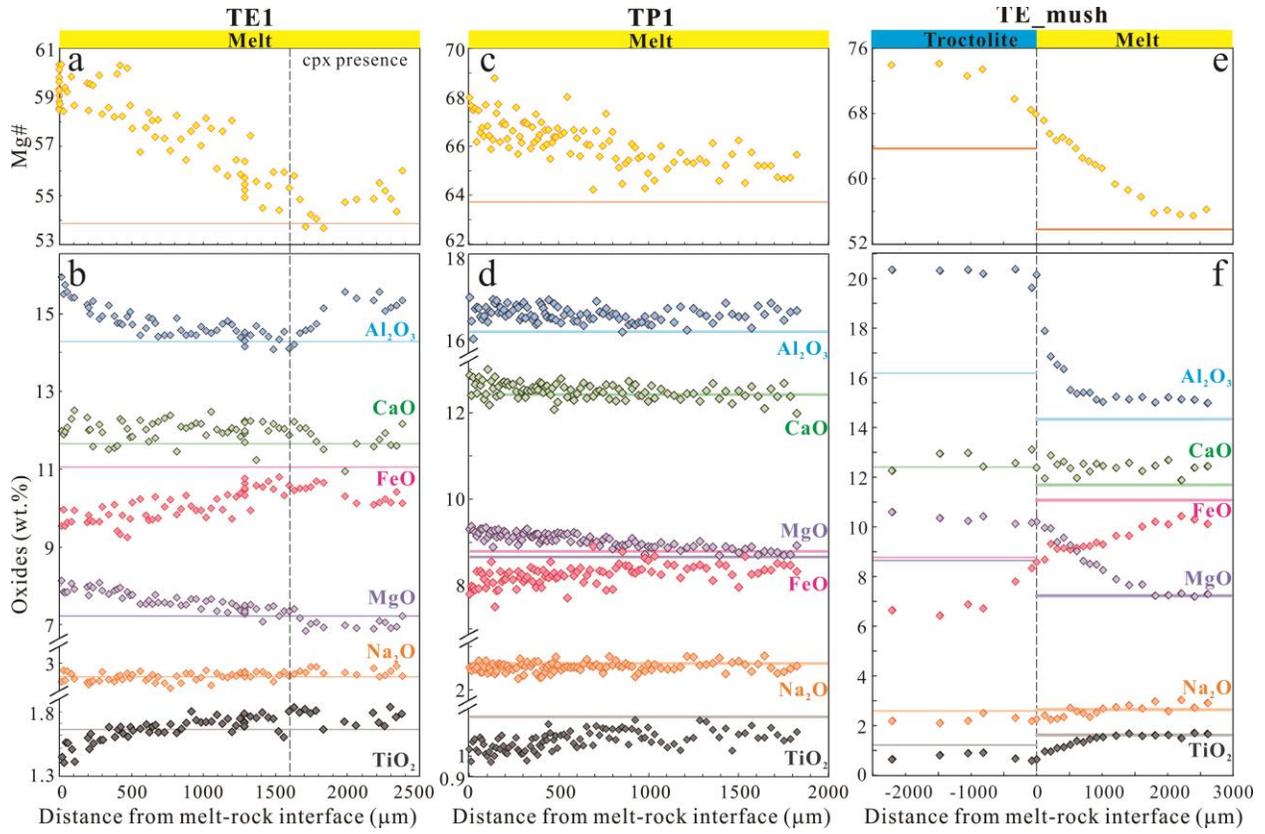


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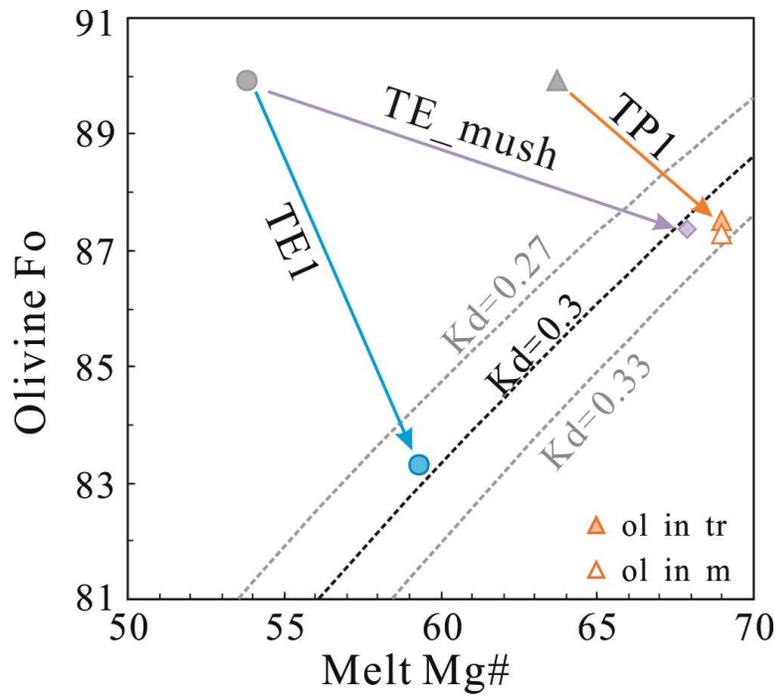


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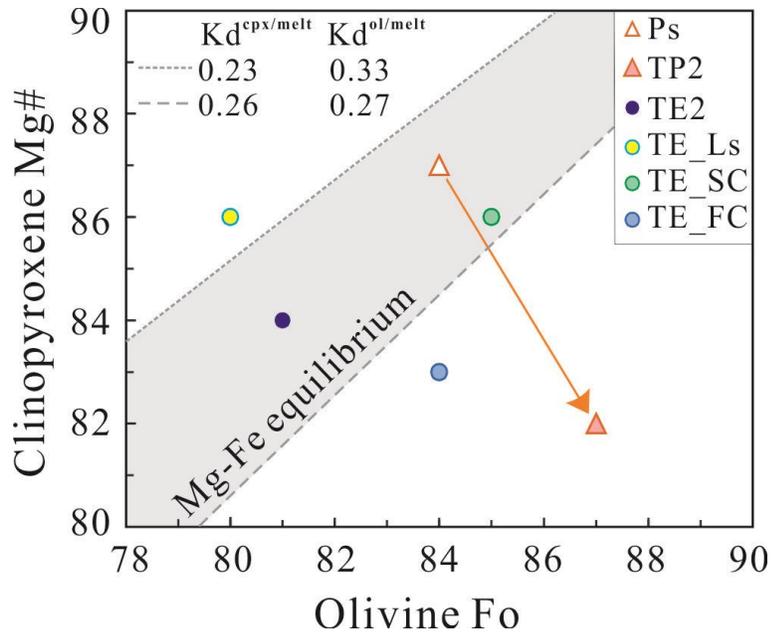


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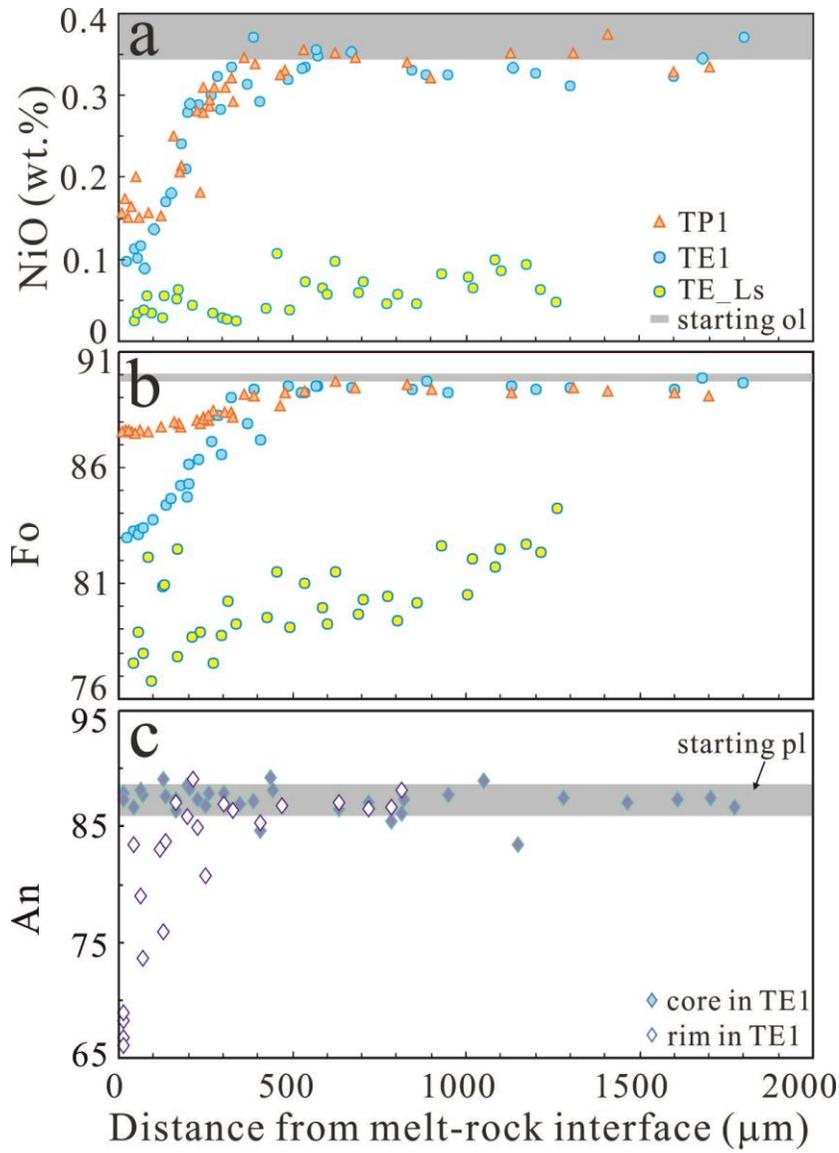
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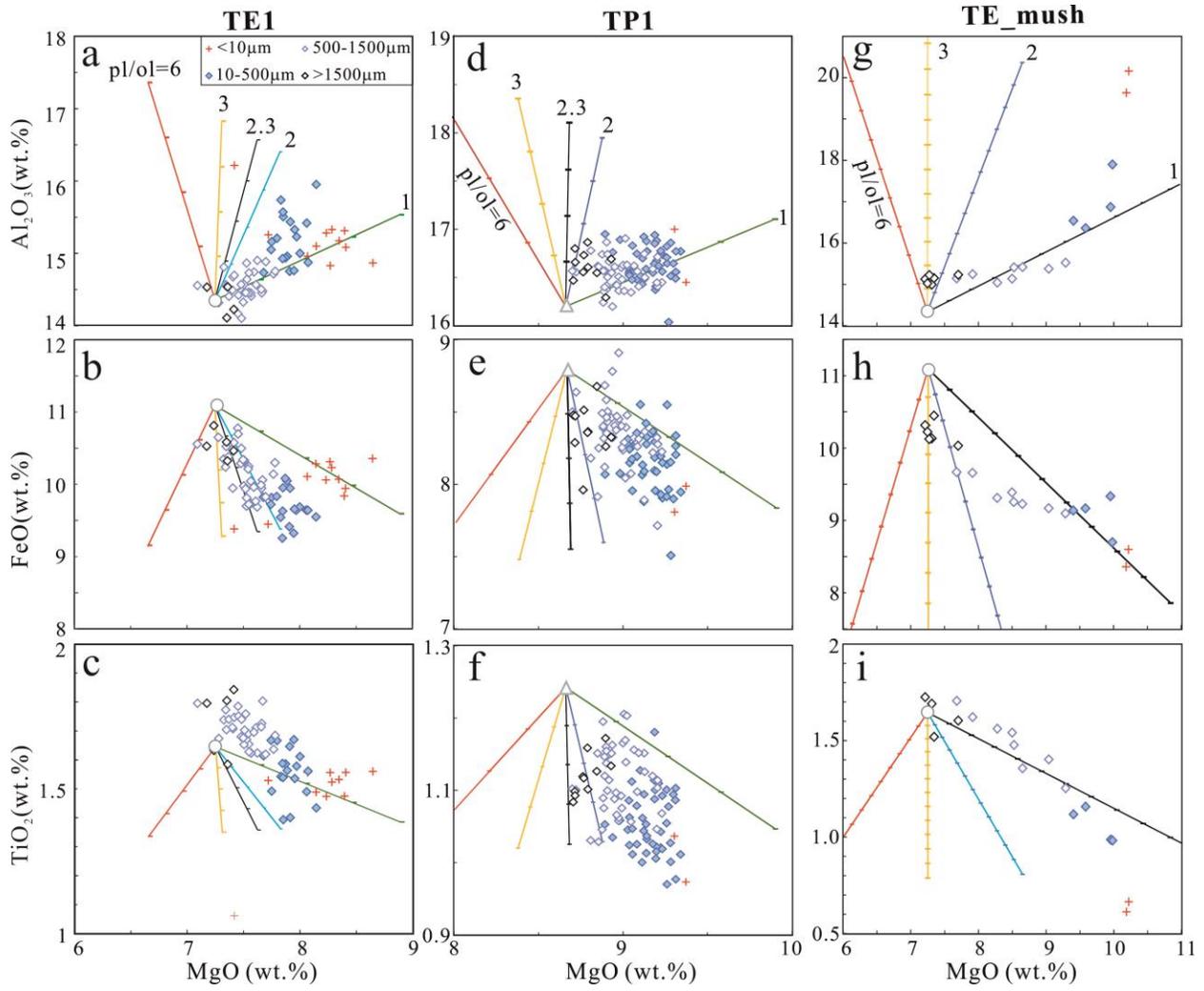


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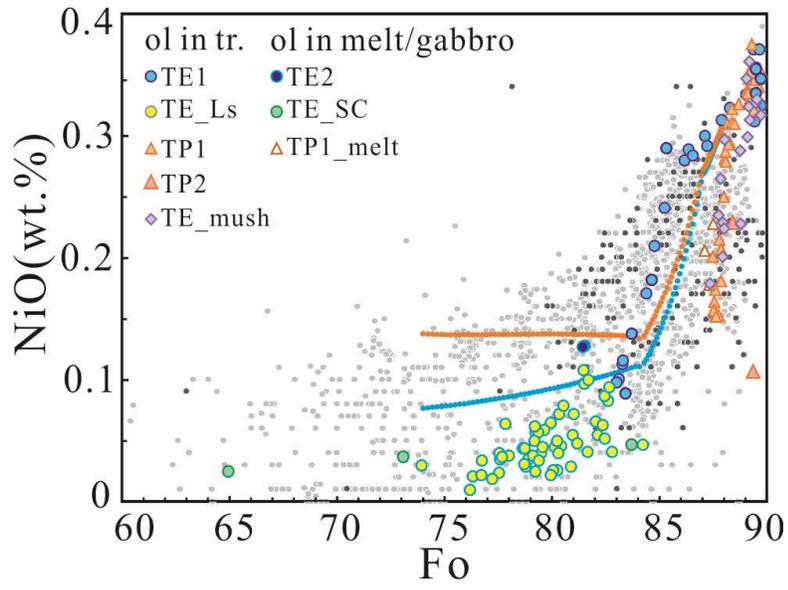
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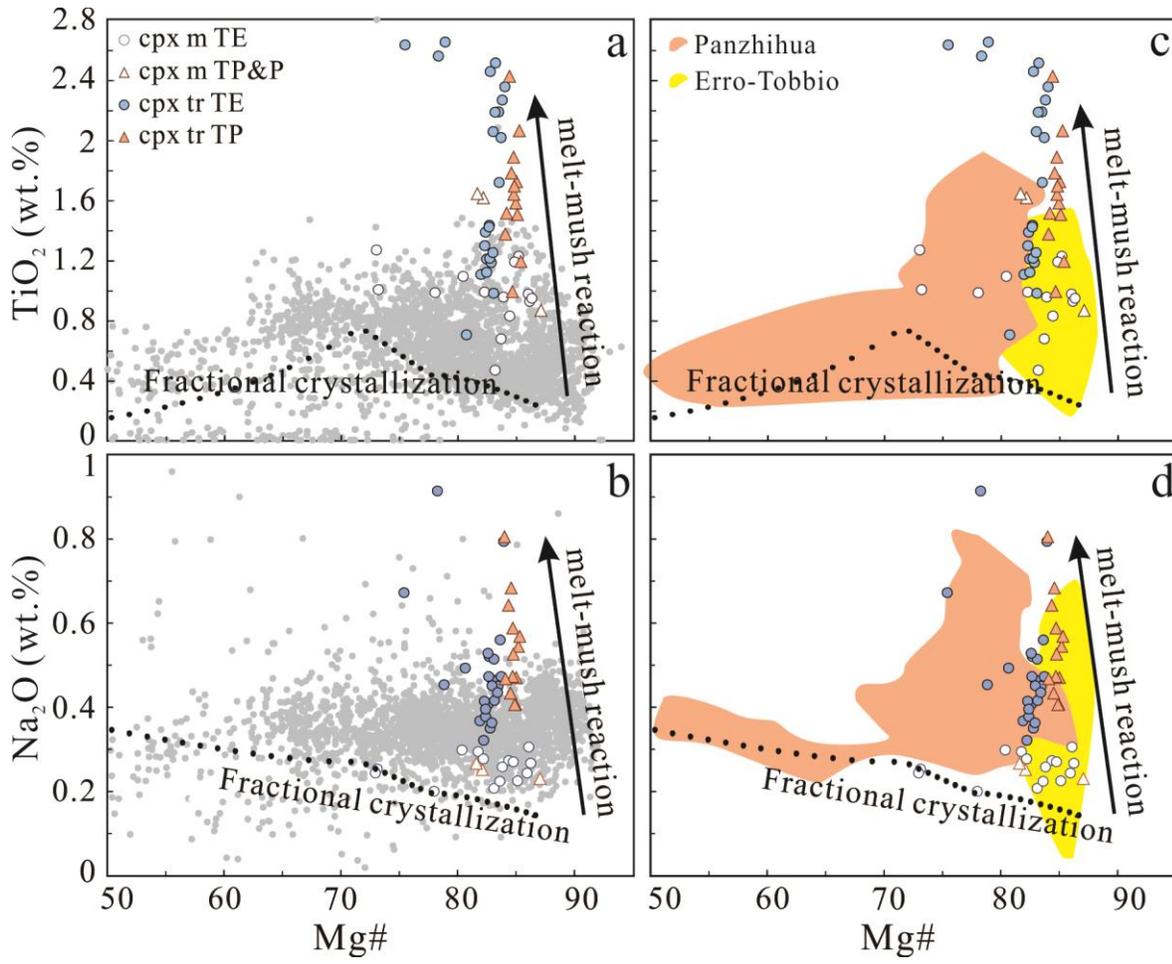
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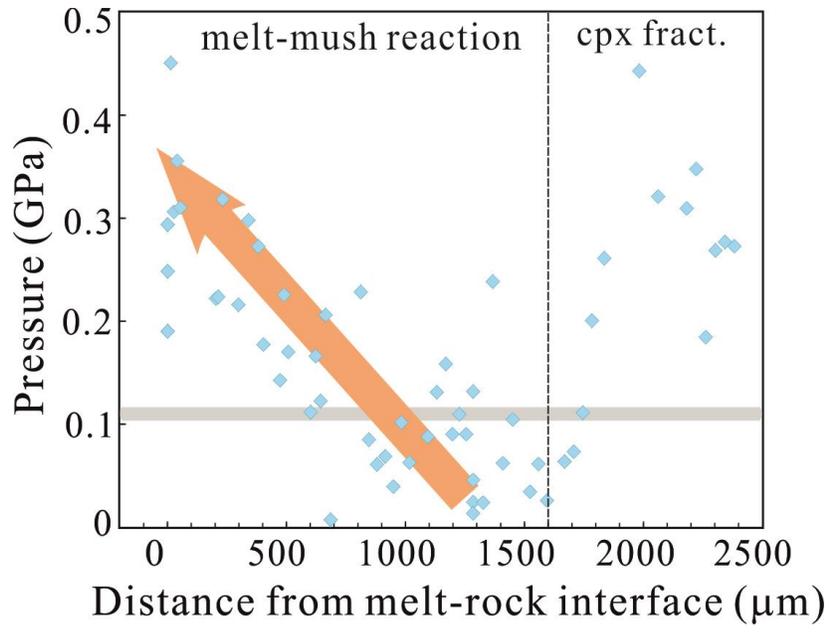
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992 **Table 1.** Starting compositions (in wt.%)

Oxide	Primitive MORB <sup>a</sup>	Evolved MORB <sup>b</sup>	Plagioclase	Olivine
SiO <sub>2</sub>	50.13	50.49	47.57	41.54
TiO <sub>2</sub>	1.25	1.64	0.01	0.00
Al <sub>2</sub> O <sub>3</sub>	16.31	14.29	32.88	0.03
Cr <sub>2</sub> O <sub>3</sub>				0.01
FeO <sup>T</sup>	8.85	11.05	0.56	9.60
NiO				0.37
MnO	0.17	0.3	0.01	0.07
MgO	8.72	7.22	0.03	47.97
CaO	12.5	11.66	17.52	0.07
Na <sub>2</sub> O	2.63	2.66	1.40	
K <sub>2</sub> O	0.02	0.12	0.00	
P <sub>2</sub> O <sub>5</sub>	0.08	0.18		
Total	100.66	99.61	99.98	99.65
Mg <sup>#c</sup> /An/Fo	63.7	53.8	87.3	89.9

<sup>a</sup> Major element composition of MORB sample KN182-13 D44A from Dr. Alberto Saal

<sup>b</sup> Major element composition of MORB MOA9812-095 from Dr. John Sinton

<sup>c</sup> All Fe as ferrous Fe to calculate Mg#

993

994 **Table 2.** Summary of experimental conditions

	TE1	TE2/Es	TE_FC	TE_SC	TE_Ls	TE_mush <sup>a</sup>	TP1	TP2/Ps
Melt	Evolved					Evolved&PrimitivePrimitive		
Pressure (kbar)	5	5	5	5	5	5	5	5
Initial temp (°C)	1180 (6h) <sup>b</sup>	1180 (6h)	1180 (6h)	1180 (6h)	1180 (27h)	1200 (6h)	1200 (6h)	1200 (6h)
Final temp (°C)	1180	1000	1000	1000	1000	1200	1200	1050
Cooling process	Quench	Step <sup>c</sup>	1°C/min	0.1°C/min	Step	Quench	Quench	Step
Total duration	6	26	9	35	48	6	6	26

995 <sup>a</sup>TE\_mush run used evolved melt to react with a troctolitic mush with 10% interstitial primitive melt

996 <sup>b</sup>Number in parenthesis is the run duration at the specific temperature

997 <sup>c</sup>Step-cooling is at an average rate of ~ 0.15°C/min to the final temperatures

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