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Thermodynamic modeling of melt addition to peridotite: Implications for the refertilization of the non-cratonic continental mantle lithosphere

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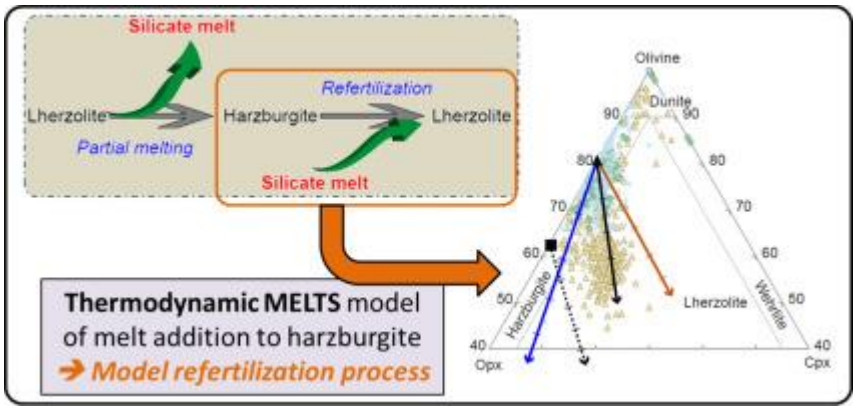
Abstract :

In a classic model of evolution of the non-cratonic continental mantle lithosphere, harzburgites represent the refractory (< 5% clinopyroxene) residues of high degrees of partial melting of fertile mantle, while lherzolites (> 5% clinopyroxene) represent residues of lesser degrees of partial melting. However, partial melting is not the only process that could explain the peridotite compositional variability that ranges from fertile (> 2 wt% Al2O3, < 45 wt% MgO) to refractory (< 2 wt% Al2O3, > 45 wt% MgO). In the refertilization process, harzburgite is a refractory protolith (potentially previously formed by partial melting of a fertile mantle) that undergoes reactive percolation of silicate melts derived from the underlying asthenosphere, resulting in the crystallization of a new generation of minerals (mostly clinopyroxene). A simple but critical first step towards understanding the refertilization process is to examine how modal and major element compositions evolve as melts are added to peridotites. Here we use a thermodynamically-constrained two-component mixing model to independently evaluate the roles of five different parameters: pressure, temperature, redox conditions, and compositions of the initial peridotite and the added basaltic melt (hereafter referred to P-T-fO2-X pi-Xmelt), during melt addition. We compare the results with observed suites of peridotites. The main observations are as follows: (1) the produced model is consistent with the global peridotite database, and (2) T, fO(2) and small variations of pressure have almost no impact on the evolution of the system. In contrast, the mineralogy of the percolated harzburgite has a substantial effect on the variation of the modal proportions. The parameter with the most significant impact is Xmelt, which is directly linked to the geodynamic context and melting conditions. This parameter directly controls the refertilization reaction and so, the phase proportions and the bulk-rock composition. Elements that partition preferentially in the melt phase (e.g., Na) display depletions in natural assemblages that are stronger than those predicted from the simple mixing model, consistent with the fact that the natural process occurs in an open system, and that reactive percolation likely results in incompatible element enrichment in the associated melt. Our results corroborate the suggestion that most of the spectrum of compositional variability observed in lithospheric mantle peridotites can be explained by the impregnation of primitive silicate melt in refractory harzburgites.

Graphical abstract



Keywords : Continental mantle lithosphere, Refertilization, Mixing model, Peridotites, pHMELTS, Harzburgites, Lherzolites

1. Introduction

The subcontinental lithospheric mantle (SCLM) is petrologically and geochemically heterogeneous (Allègre & Turcotte, 1986; Hofmann, 1997; Stracke et al., 2005; Griffin et al., 2009; Bodinier & Godard, 2014; Pearson et al., 2014). This lithospheric domain is documented by xenoliths in alkali basalts and kimberlites and by orogenic peridotite massifs. In non-cratonic regions, the SCLM has on average a lherzolitic bulk composition (> 5% clinopyroxene; Griffin et al., 1999). Lherzolites are fertile peridotites, and are aluminum-rich (> 2 wt.% Al_2O_3) and magnesium-poor (< 45 wt.% MgO). Pyroxenites, dunites and, to a greater extent, harzburgites (< 5% clinopyroxene) are also common SCLM lithologies. Harzburgites are aluminum-poor (< 2 wt.% Al_2O_3) and magnesium-rich (> 45 wt.% MgO). The genetic relationships between these rock types have been the subject of much debate in the last decades (e.g., Frey and Prinz, 1978; Kelemen et al., 1992; Le Roux et al., 2007). In addition to potential preexisting compositional variability, the spectrum of natural peridotite compositions can be mostly explained by the combination of two end-member processes (**Fig. 1**): partial melting and refertilization (see reviews by Simon et al., 2008; Bodinier and Godard, 2014; Pearson et al., 2014; Warren, 2016). The partial melting model implies that harzburgites and clinopyroxene-poor lherzolites are formed by variable degrees of partial melting of fertile lherzolites (e.g., Dick et al., 1984; Frey et al., 1985; McKenzie and Bickle, 1988; McKenzie and O'Nions, 1991; Hellebrand et al., 2001). The refertilization model implies that lherzolites are secondary (refertilized) lithologies formed from the interaction between a refractory, lithospheric mantle (mostly harzburgitic) with silicate melts derived from the underlying asthenosphere (e.g., Van der Wal and Bodinier, 1996; Garrido and Bodinier, 1999; Muntener and Piccardo, 2003; Pilet et al, 2005, 2008, 2011) or, more locally, from a secondary melting event of the lithosphere (e.g., Chazot et al., 1996; Rampone et al., 2020). As used in this contribution, the term "refertilization" will refer to the process of modal transformation of harzburgite from the addition of, and/or interaction with silicate melt in the non-cratonic mantle. This can produce clinopyroxene-enriched harzburgite, lherzolite, or other types of peridotite depending on

77 the percolating melt composition. We do not consider the full range of metasomatic processes and
78 agents that often result in the enrichment of incompatible trace elements and modification of isotopic
79 compositions in peridotites. Nevertheless, we recognize that refertilization and metasomatism
80 represent a continuum of processes and the distinction between them can be ambiguous. We also do
81 not consider processes of metamorphic segregation (e.g., Tilhac et al., 2021) that can contribute to
82 local sharp changes of modal proportions.

83 Support for the refertilization model is provided by petrographic, geochemical and structural
84 observations. Geochemical evidence comprises variations of trace and major elements in bulk
85 compositions and minerals (e.g., Elthon, 1992; Hellebrand et al., 2002; Le Roux et al., 2007; Soustelle
86 et al., 2009; Mundl et al., 2015; Gu et al., 2016), while structural observations include lherzolite
87 foliations and websterite layering that cross-cut harzburgite deformation (Le Roux et al., 2008).
88 Additionally, the presence of intergranular pyroxenes and spinels, often associated with sulfides
89 (Bodinier & Godard, 2014; Lorand and Luguet, 2016) has been interpreted as secondary phase
90 crystallization from liquid percolation. Al_2O_3 content in the bulk-rock is very frequently used as a
91 proxy for peridotite fertility, though MgO and CaO contents, Yb and Lu concentrations, olivine Fo#
92 (=MgO/(MgO+FeO)*100, in mol.) and spinel Cr# (=Cr_2O_3/(Cr2O3+Al_2O_3)*100, in mol.) are also
93 often used (e.g., Gao et al., 2002; Handler et al., 2003; Wu et al., 2006; Armytage et al., 2014; Byerly
94 and Lassiter, 2012). Most fertility proxy correlations can be interpreted by both partial melting and
95 refertilization processes, but there are exceptions. Elthon (1992) showed that the linear relationship
96 between Na_2O and MgO observed in abyssal peridotites was inconsistent with partial melting, but
97 could be explained by refertilization. In addition, variations of major, minor and trace elements
98 through the harzburgite-lherzolite contacts in the Lherz massif indicate that the lherzolites were
99 formed through a refertilization process (Le Roux et al., 2007). The refertilization mechanism also
100 provides a key to understanding the paradoxical association of light rare earth element (LREE)
101 enrichment with depleted harzburgite often observed in mantle xenoliths and in tectonically-emplaced
102 peridotites (e.g., Frey and Prinz, 1978; McDonough and Frey, 1989; Godard et al., 1995; Le Roux et
103 al., 2007; Tilhac et al., 2021). Finally, radiogenic isotope signatures, such as those of Hf, Nd, Sr and

Os (e.g., Saal et al., 2001; Müntener et al., 2004; Le Roux et al., 2009, 2016; Lawley et al., 2020; Borghini et al., 2021; Reisberg, 2021), have often been attributed to the effects of refertilization, though this effect may be indirect, reflecting radiogenic ingrowth following ancient enrichment. For instance, the negative correlation of $^{87}Sr/^{86}Sr$ with Al_2O_3 and the positive correlation of $^{143}Nd/^{144}Nd$ with Al_2O_3 observed in Pyrenean peridotites (Downes et al., 1991; Bodinier & Godard, 2003) probably result from refertilization, with the most likely scenario being radiogenic decay following ancient enrichment in incompatible elements by a percolating melt with a depleted (MORB-like) isotopic composition (Le Roux et al., 2007). Similarly, frequently observed positive correlations between $^{187}Os/^{188}Os$ and Al_2O_3 in lithospheric peridotites may reflect radiogenic ingrowth after refertilization occurring soon after ancient melting events (Reisberg, 2021).

Formation of lherzolites by refertilization of depleted harzburgite during percolation of a silicate melt is frequently proposed to explain the composition of peridotite suites, notably in the Lherz massif (Pyrénées, France), type-locality of lherzolites (Le Roux et al., 2007), but also in numerous other worldwide occurrences including the Western Gneiss Region of Norway (Beyer, 2006), abyssal peridotites (Elthon, 1992; Seyler et al., 2007), or peridotite suites from the Kaapvaal craton (Simon et al., 2007) among other examples. Given the Os isotope evidence that large-scale refertilization can be temporally associated with the melting event that led to harzburgite depletion, refertilization is sometimes viewed as an integral part of the melting process, and has been described as "autometasomatism" (Rudnick and Walker, 2008). When melt circulation in the mantle is more channelized, cumulative pyroxenites may form (Bodinier et al., 2008; France et al., 2015; Tilhac et al., 2016; Borghini et al., 2020; Dai et al., 2021); such pyroxenites are among the most fertile mantle lithologies, and could thus be important components in any potential subsequent partial melting episode (e.g., Lambart et al., 2013; France et al., 2015).

An improved knowledge of refertilization of lithospheric peridotites is critical to understanding how the continental lithosphere developed its current composition. In this contribution, we investigate the influence of various parameters such as pressure, temperature, and redox conditions, as well as the chemical composition of the initial peridotite and percolating melt on the chemical and modal

composition of the resulting solid phase, using a thermodynamically based mixing model. Our model considers lithospheric processes related to the percolation of melts formed in the major melting regime, and does not explore the related processes with strongly silica-undersaturated liquid that form near the solidus under oxidized conditions (e.g., on the carbonated peridotite solidus). A similar approach (thermodynamic model of peridotite impregnation by a silicate melt) has been adopted by Lambart et al. (2012) and Shaw et al. (2018). Lambart et al. (2012) produced a simplified model of impregnation of a peridotite by a pyroxenite-derived melt at constant P and T, resulting in the generation of various peridotitic and pyroxenitic lithologies with systematic production of clinopyroxene. Shaw et al. (2018) were able to produce wehrlites by addition of orthopyroxene-undersaturated mafic alkaline magma to orthopyroxene bearing peridotite, followed by isobaric equilibration. While our approach is not entirely new, the goal of this paper is to determine the minimal number of parameters required to explain the SCLM compositional variability, as well as to confirm if the current understanding of the refertilization process is thermodynamically viable. The main goal of this paper is to determine whether a simple mixing model can reproduce both the bulk composition and the modal proportions of non-cratonic SCLM peridotites. Although simple (and therefore incomplete), our model is consistent with a global database of orogenic peridotites (Bodinier and Godard, 2014).

2. Approach

In this section, we reproduce the two end-member models: (1) mixing at constant P-T-fO_2 conditions during which a fixed mass of melt is progressively added to a refractory peridotite, and (2) partial melting of a fertile lherzolite.

2.1. pHMELTS and Adiabat-1ph

In this study, we aim to test if the refertilization of a refractory peridotite by a silicate melt can explain the range of modal proportions observed in global non-cratonic lithospheric peridotites. We used the thermodynamic model pHMELTS (Asimow et al., 2004) and the adiabat-1ph front end (Smith & Asimow, 2005) to model the effects of magma impregnation in peridotites at constant pressure,

157 temperature and oxygen fugacity. The pHMELTS model calculates equilibrium assemblages in
158 peridotitic systems by minimizing the Gibbs energy of the system from constraints on bulk
159 composition, temperature, pressure or volume, enthalpy or entropy, and/or oxygen fugacity. The
160 pHMELTS routine is part of the MELTS group of algorithms (Ghiorso & Sack, 1995). These
161 algorithms are used to model mantle melting and magma crystallization, and also allow for melt-rock
162 impregnation models. We use the pHMELTS software as it is suitable for pressures comprised
163 between 1 and 3 GPa (Hirschmann et al., 1998), relevant for the non-cratonic SCLM.

164 The interface adiabat-1ph (Smith & Asimow, 2005) allows the automation of pHMELTS calculations.
165 It is used to calculate equilibrium assemblages along a thermodynamic path. In particular, we used the
166 'Adiabat assimilate' option of the front-end to simulate the impregnation process as it allows us to add
167 a fixed mass of melt after each calculation stage at constant P and T conditions. The output
168 parameters (e.g., phase proportions and compositions) are calculated at each step along the
169 thermodynamic path after equilibrium conditions are achieved.

170 The use of pHMELTS is subject to certain constraints due to the proven limits of the software:

171 – The subsolidus routines do not handle K_2O well if feldspar is not present (Asimow &
172 Ghiorso, 1998). We therefore did not consider potassium.
173 – The incorporation of chromium is oversimplified. It does not enter into the modeled
174 composition of pyroxenes and garnets (Asimow et al., 1995), which leads to an
175 overestimation of the range of conditions for which spinel can crystallize (e.g., Lambart et al.,
176 2009). In our calculations, we did include chromium due to the importance of spinel in the
177 reaction but we considered the overestimation of spinel in the interpretation of the results.
178 – pHMELTS offers a choice between two thermodynamic models for garnet. The older model
179 tends to overestimate the proportion of the grossular component (Berman & Koziol, 1991),
180 while the new one tends to overestimate the stability range for garnet (e.g, Elkins et al., 2019).
181 In our calculations, we used the older model of garnet but we performed calculations at a
182 maximum pressure of 2 GPa to avoid the garnet stability field.

183 – The solidus temperatures of peridotites are overestimated (Ghiorso et al., 2002).

184 **2.2. The mixing model**

185 We consider a simple model in which a subsolidus homogenous mantle domain composed of depleted
186 peridotite (harzburgite) is impregnated by a finite amount of melt at constant pressure and
187 temperature. The impregnation process is simulated by adding up to 50 g of melt by increments of
188 0.01 g, to 100 g of depleted peridotite. Each incremental addition of melt is followed by chemical re-
189 equilibration of the impregnated peridotite. Lambart et al. (2012) used a similar approach to
190 investigate the fate of pyroxenite-derived melt during interaction with the surrounding peridotite, with
191 larger increments (5g). We do not model melt percolation, but simply melt-peridotite interaction
192 during melt addition; the implications of this limitation are discussed below. Our model also does not
193 aim to reproduce the physical aspects of the refertilization process. The actual amount of melt
194 required during the refertilization process is controlled by factors such as the degree of disequilibrium
195 (Oliviera et al., 2020), the mobility of the melt (Watson et al., 1990), the pressure gradient (Pec et al.,
196 2015), or the grain size of the protolith (Turner et al., 2015). It is beyond the scope of this paper to test
197 the effects of such parameters. Here we focus on the major element and modal composition of the
198 solid phases after interaction.

199 **2.2.1. Selected input parameters**

200 *Pressure, temperature and oxygen fugacity:* We selected a range of oxygen fugacity (fO_2) between
201 FMQ-3 and FMQ +3 (with FMQ standing for the fayalite-magnetite-quartz oxygen buffer) to look at
202 the effects of oxygen fugacity variations around the average value for the oceanic lithosphere (Cottrell
203 et al., 2021; Foley, 2011). Calculations were performed at 1.5 and 2 GPa, for temperatures (T) varying
204 between 1150 and 1300°C, conditions corresponding to the subsolidus lithospheric mantle
205 (Hirschmann, 2000). Under these conditions, silicate melts are expected to be strongly consumed
206 during the reaction, for any realistic melt composition (Lambart et al., 2012). Details of the conditions
207 for each test are given in **Table 1** (runs #1 to #12).

208 *Harzburgite compositions*: We selected three natural samples of depleted peridotites from the Lherz
209 massif (Roux et al. 2007) as starting compositions (**Table 2**). H1 corresponds to an average
210 harzburgite, H2 is slightly enriched in orthopyroxene, and H3 is orthopyroxene-rich.

211 *Melt compositions*: Several experimental studies have shown that a large part of the mineralogical and
212 compositional diversity in the mantle can be explained by melt-rock reactions (e.g., Kelemen et al.,
213 1990; Yaxley and Green, 1998; Morgan and Liang, 2003, 2005; Herzberg 2011; Lambart et al., 2012;
214 Mallik and Dasgupta, 2013; Soustelle et al., 2014; Mitchell & Grove, 2016). Le Roux et al. (2007)
215 suggested a refertilization process as a melt-rock reaction resulting in the dissolution of olivine and
216 crystallization of pyroxenes and spinel:

217 $$\text{Olivine} + \text{melt}_1 \rightarrow \text{Clinopyroxene} + \text{Orthopyroxene} + \text{Spinel} + \text{melt}_2 \qquad (1)$$

218 Not every melt, however, can trigger reaction (1) as the silica activity of the interacting melt must be
219 higher than the silica activity of the melt in equilibrium with the harzburgite protolith (Lambart et al.,
220 2012). If the silica activity is lower, clinopyroxene and olivine are produced at the expense of
221 orthopyroxene. For instance, Shaw et al. (2018) interpreted the formation of wehrlite as the result of
222 infiltration of orthopyroxene-undersaturated alkaline melt into the lithospheric mantle. The silica
223 activity calculated (Ghiorso et al., 1995) for the melt used in their model is low (e.g., $a_{liq}^{SiO_2} \sim 0.2$ at P
224 = 1.25 GPa and T =1350°C, and FMQ). In comparison, under similar conditions, the silica activity of
225 a melt in equilibrium with orthopyroxene and olivine is ~0.35 (Lambart et al., 2012).

226 In our calculations, we used three different basaltic melts (**Table 3**). The MOR melt is a basaltic
227 composition with a silica activity of 0.372, obtained in a partial melting experiment of a fertile
228 peridotite (Lambart et al., 2009) and corresponds to a spreading-ridge context. HS and HS2 are
229 picritic basalt compositions from Hawaii (Kilauea; Norman & Garcia, 1999) and Iceland (Reykjanes
230 Peninsula; Jakobsson, 1978), respectively. In our calculations, they represent primitive magmas from
231 hot spot contexts, with HS being more iron-rich than HS2. Unlike HS (which is olivine-rich), HS2
232 contains almost no phenocrysts and thus more closely represents a melt composition.

The silica activity of HS and HS2 melts are 0.485 and 0.551, respectively. Both of these melts have higher silica activity than an experimental melt produced from partial melting of a fertile spinel lherzolite at 2 GPa and 1375 °C, which has a value (i.e., 0.370; see KLB-1 run #21; Hirose & Kushiro, 1993) very similar to that of the MOR melt.

For the P-T conditions chosen for the calculations, the liquid is completely, or very nearly (runs #3 and #5), consumed during the reaction. The strong consumption of silicate melt is partly due to the overestimation of the solidus temperatures in pHMELTS (Ghiorso et al., 2002), regardless of the composition (or silicate activity) of the melt and is also supported by experimental studies (e.g., Yaxley and Green, 1998; Lambart et al., 2012). It results in an enrichment of the incompatible element concentration in the produced solid. When the liquid is absent after equilibration, the system can be viewed as a two-component mixture between the depleted peridotite and the added melt.

As the melt consumption is usually accompanied by the precipitation of clinopyroxene (+orthopyroxene/olivine), it results in a local decrease in the permeability of the system. Lambart et al. (2012) suggested that such permeability decrease might result in the affected lithology evolving as a near-closed system, similar to what is modeled here. Nevertheless, a silicate melt in mineralogical assemblages dominated by olivine can however become interconnected at a very low melt fraction (e.g., Waff & Faul, 1992; Zhu et al., 2011; Laumonier et al., 2017), and a real closed system is unlikely. The escape of a small melt fraction has however very little impact on the behavior of elements with an affinity for the solid phase (i.e., the compatible elements), but can significantly affect elements preferentially partitioning into the melt phase (i.e., incompatible elements). The latter should thus be preferred if we are to characterize the open/closed character of the process. In order to test the impact of the presence of a melt phase on the incompatible element concentrations of the coexisting solid phase, we also performed a run with a liquid phase present (run #12, **Table 1**). Calculations were performed at T = 1400°C, an unlikely temperature for the continental lithosphere, which forced the resulting system to have a melt phase. In this melt-present run, the incompatible elements are partitioned into the melt phase during each step of the modeled process. While the closed system approximation underscores the simplicity of our model and its limited applicability to

incompatible element concentrations, in many aspects our calculations may be representative of a process of simple impregnation of a refractory peridotite by a silicate melt in the continental mantle lithosphere.

2.3. The partial melting model

In order to compare with the impregnation model, we also performed 3 partial melting simulations (runs PM1, PM2 and PM3, **Table 1**). We used the Primitive Upper Mantle composition from McDonough (1995) as a starting composition (PUM in **Table 2**). Run PM1 was performed at constant pressure (2 GPa), with a temperature rising from 1200 to 1520°C. Run PM2 was performed at constant temperature (1230°C), with pressure decreasing from 2 to 0.6 GPa. Finally, run PM3 was performed at constant entropy, with pressure decreasing from 2 to 0.6 GPa, and starting temperature of 1396°C (PUM solidus temperature at 2 GPa), in order to model more geodynamically realistic conditions. As the melt remains in contact with the residue throughout the simulations, all three of these cases can be viewed as thermodynamically constrained batch melting models.

3. Results

We focus on the solid bulk composition and modal proportions of the solid phase and do not consider mineral phase compositions. In fact, inter-mineral partitioning of major element components such as Fe-Mg in the lithospheric mantle is strongly dependent on the equilibration temperature (Brey and Köhler, 1990). As thermal reequilibration is the rule in lithospheric domains, comparing the elemental compositions of the individual phases obtained from our thermodynamic model outputs with natural data would not produce meaningful results. Bulk residual assemblage compositions and modal proportions are less significantly affected by the temperature of reequilibration, and we present the corresponding results hereafter.

3.1. Reference simulation (run #1)

The main results of the mixing models are the evolutions of modal proportions (**Figs. 2, 3, Table 4, and Supplementary Fig. S1**), and of bulk solid assemblage compositions (**Fig. 4 and 5**). We will use

285 the outputs of run#1 as a reference for comparison with other tests. Results for this run show
286 production of spinel (+2.5), orthopyroxene (+0.2), and clinopyroxene (+28.6), with the values
287 indicating the absolute change in the modal proportion of each phase (normalized to 100). In contrast,
288 the liquid is totally consumed and olivine proportion also diminishes (-31.3) (**Fig. 2**). This decrease in
289 olivine proportion is not due solely to the precipitation of the other phases but also to olivine
290 dissolution (Fig. 2b). Initial harzburgite is hence progressively modified to clinopyroxene-rich
291 harzburgite, and eventually to a lherzolite assemblage (**Fig. 3**). As no liquid is present after
292 equilibration, the bulk chemical composition of the solid assemblage (**Fig. 4**) evolves along mixing
293 lines between the composition of the initial peridotite and the composition of the melt. Therefore, for
294 run#1, after addition of 50% liquid, the bulk chemical composition shows an enrichment in Al_2O_3
295 (+5.4 wt.%), CaO (+3.8 wt.%), SiO_2 (+1.6 wt.%), and Na_2O (+1 wt.%), a decrease in MgO (-11.7
296 wt.%), and a slight decrease in FeO_t (-0.3 wt.%) (FeO_t: total iron content expressed as FeO).

297 **3.2. Influence of the impregnation conditions**

298 The effect of variations of P, T, and redox parameters on the model results are presented in **Fig. 3a**.
299 Runs #1, #3 and #4 illustrate the effect of temperature on the reaction (T = 1230, 1300, and 1150°C,
300 respectively), runs #1 and #5 are used to investigate the effect of a small variation of pressure (P = 2
301 and 1.5 GPa, respectively). Runs #1, #6a, #6b, #7a and #7b are used to track the effect of redox
302 conditions on the reaction (fO_2 = FMQ, FMQ+1, FMQ+3, FMQ-1, and FMQ-3, respectively). In all of
303 these cases, the same initial peridotite composition and the same melt composition were used (H1;
304 MOR). The effect of pressure and temperature is insignificant for the ranges considered in this study
305 (**Fig. 3a**). Additionally, for most of the fO_2 conditions tested here, the modal proportions of the
306 impregnated peridotite strongly overlap with each other. Under highly oxidized conditions (i.e., run
307 #6b: FQM+3) however, 4.9 wt.% of orthopyroxene is produced at the expense of olivine (-35.3 wt.%).
308 This is because Fe^{3+} is not easily incorporated in olivine and a higher Fe^{3+}/Fe^{2+} ratio will favor the
309 dissolution of olivine and precipitation of pyroxene.

310 **3.3. Influence of the composition**

The influence of the melt composition on the refertilization process was tested in runs #1, #2 and #11 (MOR, HS, and HS2, respectively). The influence of the initial peridotite composition was tested in runs #1, #8 and #9 (H1, H2, and H3, respectively). Run #10 also investigates a variation of both starting material composition (H3) and melt composition (HS). These six runs are displayed in **Figs. 3b, 3c and 4**. Because the melt is consumed during the reaction, bulk compositions follow mixing lines between the composition of the initial peridotite and the composition of the melt (**Fig. 4**). In the major element diagrams (**Fig. 4**), this results in solid assemblages with greater sodium and calcium enrichments for models performed with MOR addition, and slightly greater iron enrichment for models performed with HS addition, relative to that performed with HS2 addition, directly reflecting the differences in melt compositions (**Table 3**). In the Streckeisen projections (**Fig 3b**), the addition of the HS2 melt (run #11) results in a strong orthopyroxene enrichment and the final lithology (after 50% melt addition) is an orthopyroxene-rich olivine websterite (35% olivine. 52% orthopyroxene, 9% clinopyroxene). On the contrary, the reaction with the MOR melt (run #1) results in both clinopyroxene and orthopyroxene enrichments, but the final lithology is in the field of lherzolite (49% olivine. 20% orthopyroxene, 29% clinopyroxene). The addition of HS melt (run #2) results in a near-vertical trend in the Streckeisen diagram with a final lithology in the field of lherzolite with a similar proportion of olivine but a higher orthopyroxene/clinopyroxene ratio (49% olivine. 30% orthopyroxene, 20% clinopyroxene). Finally, the main effect of varying the initial harzburgite composition is to change the initial location of the mixing trend, that is the orthopyroxene/olivine ratio in the Streckeisen projection (**Fig. 3c**). Runs #1 and #8 evolve from the domain of harzburgites to lherzolites, and run #9 to websterites.

3.4. Other runs

3.4.1 Partial melting runs

Partial melting simulations PM1, PM2 and PM3 are presented in **Fig. 3d** and **Fig. 5**. The end of run PM1 corresponds to an amount of liquid produced of 30 wt.% (**Table 4, Supplementary Fig. S1n**), while the ends of PM2 and PM3 correspond to 7 wt.% and 5 wt.% of liquid produced, respectively

(**Supplementary Figs. S1o and S1p**). The low amount of melt produced in PM2 and PM3 is due to the overestimation of the solidus temperature of the peridotite in pHMELTS (Ghiorso et al., 2002). In the partial melting run at constant pressure (PM1), as the temperature rises above the PUM (Primitive Upper Mantle; McDonough, 1995) solidus temperature at 2 GPa (1296°C), clinopyroxene is strongly consumed (-22 modal %) corresponding to a change in lithology from lherzolite to harzburgite (**Fig. 3d**). The chemical composition of the residual solid shows a final enrichment in MgO (+6 wt.%), and SiO_2 (+0.8 wt.%), associated with a decrease in Al_2O_3 (-2.5 wt.%), CaO (-2.5 wt.%), FeO_t (-1.4 wt.%), and Na_2O (-0.3 wt.%), with values representing absolute changes in the proportions of these species in the solid phase. In the plot of Na_2O vs. Al_2O_3 a curved trend is observed, which starts out steeply at Al_2O_3 contents > 4 wt.%, and Na_2O contents > 0.1 wt.% and flattens out with continued melting (**Fig. 5**).

The partial melting run under conditions of isentropic decompression (PM3) also starts at 1296°C. pHMELTS adapts the temperature during the simulation in order to maintain constant entropy (down to 1214°C). By the end of the run, olivine has been produced (+2.4%) at the expense of clinopyroxene (-8.6 %) (**Fig. S1p**). However, in detail some complexity is observed as pressure decreases. At first the formation of orthopyroxene (+1.6 %) is slightly favored over that of olivine (+1.1%), but starting at around 0.8 GPa, the orthopyroxene/olivine ratio shows a progressive decrease, as olivine production is favored in comparison to orthopyroxene (**Fig. 3d**), consistent with the increasing stability of olivine with decreasing pressure (Stolper, 1980). The chemical composition of the residual solid shows an enrichment in MgO (+2 wt.%) and FeO_t (+0.2 wt.%), and a decrease in Al_2O_3 (-1 wt.%), SiO_2 (-0.6 wt.%), CaO (-0.3 wt.%), and Na_2O (-0.3 wt.%) (**Fig. 5**).

The partial melting run by decompression at constant T (PM2) shows similar evolution to that at constant entropy (PM3). Olivine is produced (+3%) at the expense of clinopyroxene (-5.5%) and orthopyroxene (-4.3%) (**Fig. 3d** and **Supplementary Fig. S1o**). The residual solid is enriched in MgO (+2.36 wt.%) and FeO_t (+0.25 wt.%), and depleted in Al_2O_3 (-1.2 wt.%), SiO_2 (-0.7 wt.%), CaO (-0.4 wt.%), and Na_2O (-0.28 wt.%) (**Fig. 5**). In runs PM2 and PM3, the relatively limited change in bulk

363 composition, which does not attain the harzburgite field, is likely due to the overestimation of the
364 solidus temperature in pHMELTS noted above.

365 **3.4.2. Liquid-present run**

366 In most respects, the liquid-present run#12 (**Figs. 3d and 5**) shows a similar trend to the runs
367 performed at lower temperature. A notable exception is Na content, which is less enriched in the
368 refertilized assemblage than in the melt-free models (+0.4 wt.% for run #12, compared to +1 wt.% for
369 the reference run). This is because Na is strongly partitioned into the liquid phase. The final
370 percentage of melt in the assemblage is 7.5 wt.%. Minor amounts of liquid are also present in runs #3
371 (1300°C) and #5 (1.5GPa) and these also show a slightly lower Na enrichment in the solid assemblage
372 than in the melt-free runs. Nevertheless, the final melt fractions being 0.9 wt.% and 1.6 wt.%
373 (respectively) in these runs, the variation of the Na content compared to the reference simulation is
374 not significant (+0.8 wt.% and +0.7 wt.% for runs #3 and #5 respectively, compared to +1 wt.% for
375 the reference run). For the other chemical elements, these two runs show similar trends to those of the
376 reference run.

377 **4. Discussion**

378 Refertilization of the SCLM is a complex phenomenon that likely involves numerous agents and
379 mechanisms. Nevertheless, our model confirms that the reaction: Olivine + $melt_1$ → Clinopyroxene +
380 Orthopyroxene + Spinel + $melt_2$ suggested based on petrological grounds (e.g., Le Roux et al., 2007)
381 is thermodynamically possible, and thus supports its key participation in the refertilization process.
382 The composition of the infiltrating melt has a first-order control on the range of chemical and
383 mineralogical compositions observed in the SCLM.

384 **4.1. Natural data**

385 Most ophiolitic and abyssal peridotites are represented by harzburgitic lithologies (e.g., Bodinier and
386 Godard, 2014). On the contrary, peridotites from the sub-continental lithosphere, particularly orogenic
387 peridotites and ultramafic xenoliths from non-cratonic terrains, encompass many lherzolitic

lithologies (**Fig. 3**). There is substantial scatter in natural peridotite chemical compositions (**Fig. 4 and 5**). However, most natural compositions define a broad trend, with increasing SiO_2, CaO, Al_2O_3 and Na_2O contents, coupled with decreasing MgO content and relatively constant FeO content. Some of the scatter in modal and chemical composition in lithospheric peridotites could be due to inherited heterogeneity from the convecting mantle, related to crustal recycling, convection and melting, and melt-solid interactions beneath the ridges and at hot spots (e.g., Sobolev et al., 2005), or various degrees of partial melting under various conditions (e.g., melting at different pressures; Olivera et al., 2020; Tomlinson & Kamber, 2021). Also temperature reequilibration could trigger noticeable modal variations is some specific samples (e.g., two-pyroxene exsolutions, pyroxene exsolution from garnet, spinel exsolution from clinopyroxene; Kornprobst et al., 1990; France et al., 2015), which could also trigger scatter in the database, and that would eventually hamper direct comparison with our model outputs for those specific samples. However, the presence of a trend in major element compositions (**Figs. 4 and 5**) strongly suggests a genetic link between refractory (i.e., harzburgite) and fertile (i.e., lherzolite) lithologies. The goal of the following exercise is not to reproduce the full spectrum of natural compositions but to test whether our thermodynamic mixing model is consistent with this potential genetic link, and discuss which parameters in our model are the most likely to explain the spectrum of compositions observed in natural peridotites.

In **Figures 3, 4 and 5**, we compare the results of our thermodynamic mixing models with the global database for tectonically emplaced peridotites, including orogenic, ophiolitic and abyssal peridotites (Bodinier and Godard, 2014). Our results show that most of the wide range of modal proportions in lherzolite can be reproduced by our model of impregnation of harzburgitic lithologies by silicate melts. In addition, it is clear from **Fig. 3** that further impregnation would make the impregnated peridotite enter the domain of websterites, rocks composed essentially of orthopyroxene and clinopyroxene, and less than 40% olivine. This suggests that very extensive refertilization could be at the origin of some websterite and pyroxenites, and that harzburgites, lherzolites and pyroxenites may form a continuum rather than distinct groups. Such a continuum is commonly reported in mantle series and mainly explained by melt-rock reactions (e.g., from clinopyroxenite to orthopyroxenite in

Kornprobst, 1969; from dunite to lherzolite in Morgan and Liang, 2005; from dunite to websterite in Denis et al., 2018, or from harzburgite to websterite in Lambart et al., 2022). The simple impregnation model presented here however, is unlikely to explain the scatter in the most refractory lithologies (i.e., olivine-rich harzburgite and dunite). Indeed, scatter in natural peridotite compositions (**Figs. 4 and 5**) exceeds the range of variation in our impregnation model with some rare peridotites showing very high CaO/Al_2O_3 ratio or high FeO (>10 wt.%) concentrations. However, most of the natural compositions and the impregnation model results follow similar chemical trends. The effects of each input parameter of the model are discussed below.

4.2. Influence of varying parameters on the refertilization products

The impregnation models presented here highlight that variations of temperature and redox conditions, as well as small pressure variations have almost no impact on the evolution of the system in terms of modal composition (**Fig. 3a**). Hereafter we first discuss the two main parameters that influence the evolution of the system in our calculations: the composition of the percolating melt, and to a lesser extent the composition of the percolated harzburgite.

4.2.1. Percolated harzburgite composition

Although significant scatter is present in the natural data when compared to our models, our results show that a refertilization process has the potential to reproduce the harzburgite - lherzolite suites. The scatter in the orthopyroxene/olivine ratio in natural lherzolite series could be explained by a variation of the orthopyroxene/olivine ratio in the initial harzburgite (**Fig. 3c**). This scenario would require a large preexisting compositional variability of the refractory mantle, *before* impregnation. Harzburgite H3, for instance, is an orthopyroxene-rich harzburgite with a very low calcium content compared to aluminum that can be viewed as an extreme orthopyroxene-rich end-member.

Several processes could explain the origin of such compositional variability (variations of the orthopyroxene/olivine ratio). For instance, in the decompression runs (PM2 and PM3), the stabilities of olivine and orthopyroxene during the melting reaction change with pressure. The coexistence of

various residual harzburgites formed by partial melting at different pressures could result in different orthopyroxene/olivine ratios. The preexisting heterogeneity could also be due to the delamination of the cratonic lithospheric mantle into the convecting mantle, be a natural consequence of oceanic crust subduction into the convecting mantle, or result from the interaction of peridotite with an eclogite or pyroxenite-derived melt (e.g., Yaxley and Green, 1998; Tomlinson & Kamber, 2021).

4.2.2. Percolating melt composition

For the following discussion, we used H1, a typical harzburgite (**Table 2**) as the starting composition to examine the effect of melt composition on the output of the impregnation modeling (**Figs. 3b and 4**). Because all three percolating melts have high silica activities (**Table 3**), in all cases the proportion of pyroxenes increases during melt impregnation. However, the clinopyroxene/orthopyroxene ratio is strongly affected by the Ca content of the melt with more clinopyroxene produced for Ca-rich melts (**Table 3**, **Fig. 3b**). In addition to being Ca-poor, HS2 is also more Si-rich than the two other basalt compositions, resulting in more efficient crystallization of pyroxene, particularly orthopyroxene, at the expense of olivine (**Fig. 3b and Supplementary Fig. S1l**). This results in a final mineralogical assemblage for run #11 in the field of orthopyroxene-rich olivine websterite.

Except for the liquid-present run (run #12), the melt phase is entirely, or almost entirely (runs #3 and #5; **Supplementary Fig. S1b and S1d**), consumed during the impregnation process (**Table 4**). This implies that the bulk composition of the percolated peridotite evolves along a mixing trend between the starting harzburgite and the melt composition (**Fig. 4**). We note that melt HS2 is too Ca-poor to reproduce the trend of natural compositions. Additionally, part of the vertical scatter for all oxides in **Fig. 4** cannot be explained by our impregnation model (especially for ophiolitic and abyssal peridotites; e.g., variation of SiO_2 content for a given Al_2O_3 content). Finally, for the three melt compositions tested here, the modeled Na_2O vs. Al_2O_3 enrichment trend is much steeper than the correlation observed in the natural dataset (**Fig. 4**). As described in sections 2.2.1 and 3.4.2, this is due to the full consumption of liquid during the reaction, which likely differs from olivine-dominated systems in which a small fraction of liquid is expected to leave the system during melt percolation

(see reaction *(1)*). Without this liquid phase, elements that partition preferentially into the melt phase such as Na remain in the solid phases (**Fig. 4**). On the contrary, in the presence of a liquid phase (run #12), Na preferentially partitions into the melt resulting in a much lower increase of the Na_2O content in the solid phase during the impregnation process (**Fig. 5**), similar to the trend observed in natural data. In the liquid-present run, the final melt fraction in the system at equilibrium is about 7.5 wt.%. In natural systems, which may be seen as chromatographic percolation columns (e.g., Navon and Stolper, 1987), significantly lower instantaneous melt fractions can explain the low contents of Na in the solid phase. Chemical disequilibrium during melt transport also favors stronger partitioning of Na into the melt phase (Oliveira et al., 2020). In any case our results support the supposition that refertilization is an open system process.

4.3. Comparison with partial melting

Partial melting processes can also produce refractory lithologies (**Fig. 3d** and **Fig. 5**). However, our partial melting models do not fully reproduce the spectrum of compositions observed in non-cratonic continental peridotites (**Fig. 3d** and **Fig. 5**). In addition, our mixing model tended to reproduce the rough linear trend represented by the spectrum of natural continental lithospheric data more faithfully than our simplified partial melting model (straight line vs. curved lines for runs PM2 and PM3 in **Fig 3d**). Previous authors (e.g. Niu, 1997) have also pointed out that mixing more successfully reproduces linear trends (FeO vs. MgO and SiO_2 vs MgO) in peridotites than partial melting alone. We note however, that more complex dynamic melting models, such as that of Oliveira et al. (2020) which takes into consideration disequilibrium behavior, heterogeneous source compositions and melt transport rates, can produce a wider variety of compositions than the partial melting runs performed in this study (**Fig. 3d**).

As highlighted in **Fig.1**, partial melting and refertilization likely both contribute to producing lithospheric heterogeneities. Furthermore, refertilization and partial melting sometimes may be genetically linked, as the near absence of lherzolites with unradiogenic osmium isotope compositions in the non-cratonic lithosphere argues that refertilization usually occurs soon after harzburgite

formation (Rudnick & Walker, 2008; Reisberg, 2021). This implies that the original partial melting episodes may potentially be causally linked to subsequent refertilization, which could result from percolation of the remaining melts through the cooling lithosphere. We stress that refertilization as used here refers to the process of transformation of harzburgite by interaction with silicate melt in the non-cratonic mantle lithosphere. This can produce clinopyroxene-enriched harzburgite, lherzolite, or other types of peridotite depending on the percolating melt composition. Other refertilization processes, often referred to broadly as metasomatic (e.g., circulation of various silicate melts, or of hydrous, carbonatitic, or kimberlite fluids/melts) also very frequently affect both the cratonic (e.g., Song and Frey, 1989; Griffin et al., 1999; Jollands et al., 2018) and the non-cratonic lithosphere (e.g., Menzies and Dupuy, 1991; Baker et al., 1998; Raffone et al., 2009; Gu et al., 2018; Azevedo-Vannson et al., 2021). Such processes can lead to enrichment of major or incompatible element abundances, water contents, modification of isotope compositions and/or localized phase precipitation, and can occur at various times throughout the history of a lithospheric domain.

4.4. Other parameters.

One of the goals of our study was to determine the first-order parameters controlling the compositional and modal variability of the SCLM. Unsurprisingly, the melt and the protolith compositions have the strongest effect on the evolution of the system. However, because of the simplicity of our model, the effects of other parameters (e.g., fO_2, presence of volatiles, chemical disequilibrium), may be underestimated. Here we briefly review the potential effects of additional parameters.

Our results show that highly oxidized melts result in slightly higher olivine dissolution and slightly higher pyroxene (mostly orthopyroxene) precipitation compared to the reference run (see run #6b: **Fig. 3a**). Such high degrees of oxidation have been encountered in some oceanic island xenoliths (Cottrell et al., 2021) and might reflect the interaction of the xenolith with a hydrous melt (e.g., Bryant et al., 2007). However, for most conditions relevant to the lithospheric mantle, and in the major melting regime considered in this study, the effect of fO_2 seems to be insignificant. These

518 results are consistent with Shaw et al. (2018). In their calculations, an increase in the fO_2 from FQM-2
519 to FQM+2 resulted in a small decrease of the olivine/orthopyroxene ratio in the initial lithology but
520 the effect of variable fO_2 on the output of the reaction was negligible.

521 The addition of volatiles (H_2O or CO_2) is expected to significantly decrease the solidus temperature of
522 the system (e.g., Asimow and Langmuir, 2003; Dasgupta and Hirschmann, 2006). Hence, based on
523 temperature considerations only, we could expect that a refertilizing hydrous or carbonated melt
524 would result in lower consumption of melt during the reaction. However, this is not supported by
525 experiments. Mallik and Dasgupta (2013) performed reaction experiments between a lherzolite and a
526 CO_2-bearing basaltic andesite melt. In comparison to similar CO_2-free experiments, the addition of
527 CO_2 to the reacting melt has little to no effect on the proportion of residual melt after the reaction.
528 This likely is because the addition of volatiles in the melt increases the reactivity and disequilibrium
529 degree of the melt, resulting in a highly dynamic dissolution-precipitation process (e.g., Keller and
530 Katz, 2016; Lambart et al., 2022; Mallmann et al., 2009; Shaw et al., 2018). Additionally, in
531 comparison to CO_2-free experiments, Mallik and Dasgupta (2013) observed an increase in
532 orthopyroxene (and garnet) in the residual solid after reaction at the expense of olivine and
533 clinopyroxene. Similarly, Wang et al. (2016) demonstrated that the reaction of a hydrous melt with a
534 lherzolite can produce orthopyroxenite. Hence, although orthopyroxene stability also strongly
535 depends on the silica activity of the melt, the addition of volatiles may result in shifting the
536 compositional effect of the melt towards orthopyroxene-rich lithologies (**Fig. 3**). However, the effect
537 of volatiles is likely limited in comparison to that of the major-element composition of the melt. In
538 fact, in the Mallik and Dasgupta (2013) study, similar experiments performed with an alkali basalt
539 melt, but with a CO_2 content more than four times higher than that of the basaltic andesite melt result
540 in much lower consumption of olivine for a similar consumption of melt.

541 Oliveira et al. (2020) demonstrated that the degree of chemical disequilibrium during partial melting
542 can significantly modify the compositional trends of major elements observed in basalt when
543 compared with equilibrium calculations. As these authors noted, disequilibrium polybaric melting
544 increases the fractionation of Na into the melt phase. In their calculations however, both equilibrium

and disequilibrium models produced similar modal evolution of the residual mantle (**Fig. 3d**). The effect of variable potential temperature of the mantle can result in slightly larger variability, but mostly because it results in changing the initial mineralogical composition of the lherzolite between the spinel and the garnet stability field.

5. Conclusions

In order to test the hypothesis of lherzolite formation by refertilization of the non-cratonic continental mantle lithosphere, we employed a simple thermodynamic mixing model of harzburgite with a silicate melt of tholeiitic to picritic composition. We independently investigated the influence of various thermodynamic parameters (pressure, temperature, redox conditions, and the chemical composition of initial peridotite and percolating melt) on the chemical and modal compositions of the resulting lithology. Despite their simplicity, our calculations show that a large part of the compositional variability observed in orogenic peridotites can be explained by the impregnation of a refractory harzburgitic lithospheric mantle with silicate melt of various compositions. We also show that temperature, oxygen fugacity, and small variations of pressure have almost no impact on the evolution of the modal composition of the system; melt composition and initial harzburgite composition are the most influential thermodynamic parameters. The latter directly control the refertilization reaction and so, the phase proportions. To reproduce the natural compositional variability of continental peridotites, initial compositional heterogeneity of the harzburgitic protoliths is needed, as well as impregnation with various silicate melts. The three liquid compositions used here represent a range of mantle melts that could serve as potential refertilizing agents. Our run #2 with an initial average harzburgitic composition (H1) and a picritic basaltic melt (HS) best reproduced the broad trend of the natural data, but other protolith-melt combinations could also explain some of the natural variability. For instance, the composition of a liquid depends on its geodynamic context, which controls the pressure and temperature of melting, as well as the composition of the source. Finally, we highlighted that the main limitation of our model is that the refertilization process is treated as simple melt impregnation rather than melt percolation. The elements that partition preferentially into the melt phase (represented here by Na) are removed from the rock during melt percolation in the natural

system. Our thermodynamic mixing model cannot reproduce this strong fractionation of incompatible elements in the liquid phase during reactive infiltration as in our model, the melt phase is completely consumed in the reaction. This leads to an enrichment of incompatible elements in the resulting lithology in the model, and underscores the open system character of the natural refertilization process. Despite this drawback, our calculations provide thermodynamic support for models suggesting that much of the variability in the major element and modal compositions of the natural data can be reproduced by the impregnation of a harzburgite with silicate melts of various compositions.

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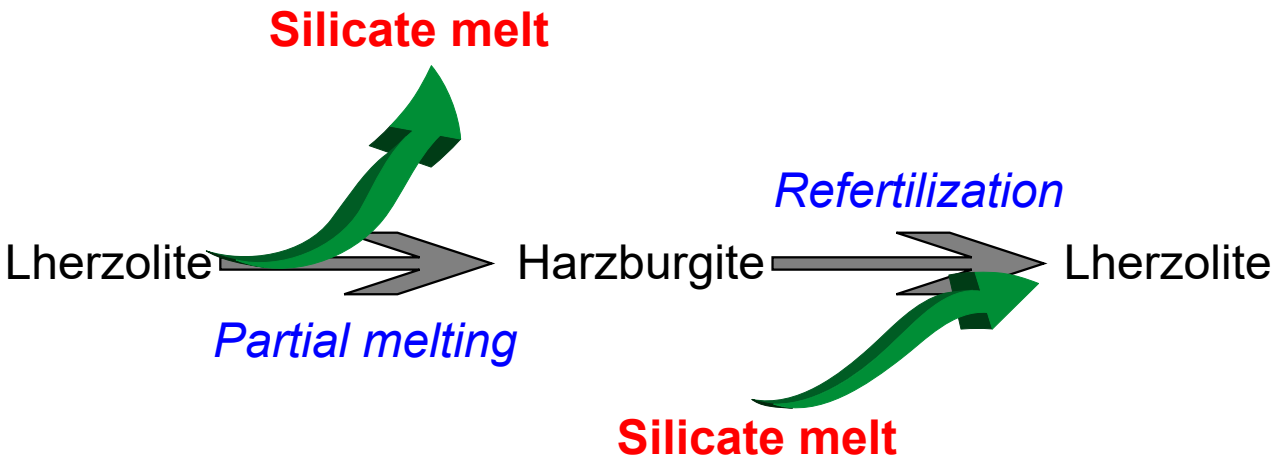
Fig 1. Schematization of the refertilization process in the context of lithospheric dynamics.

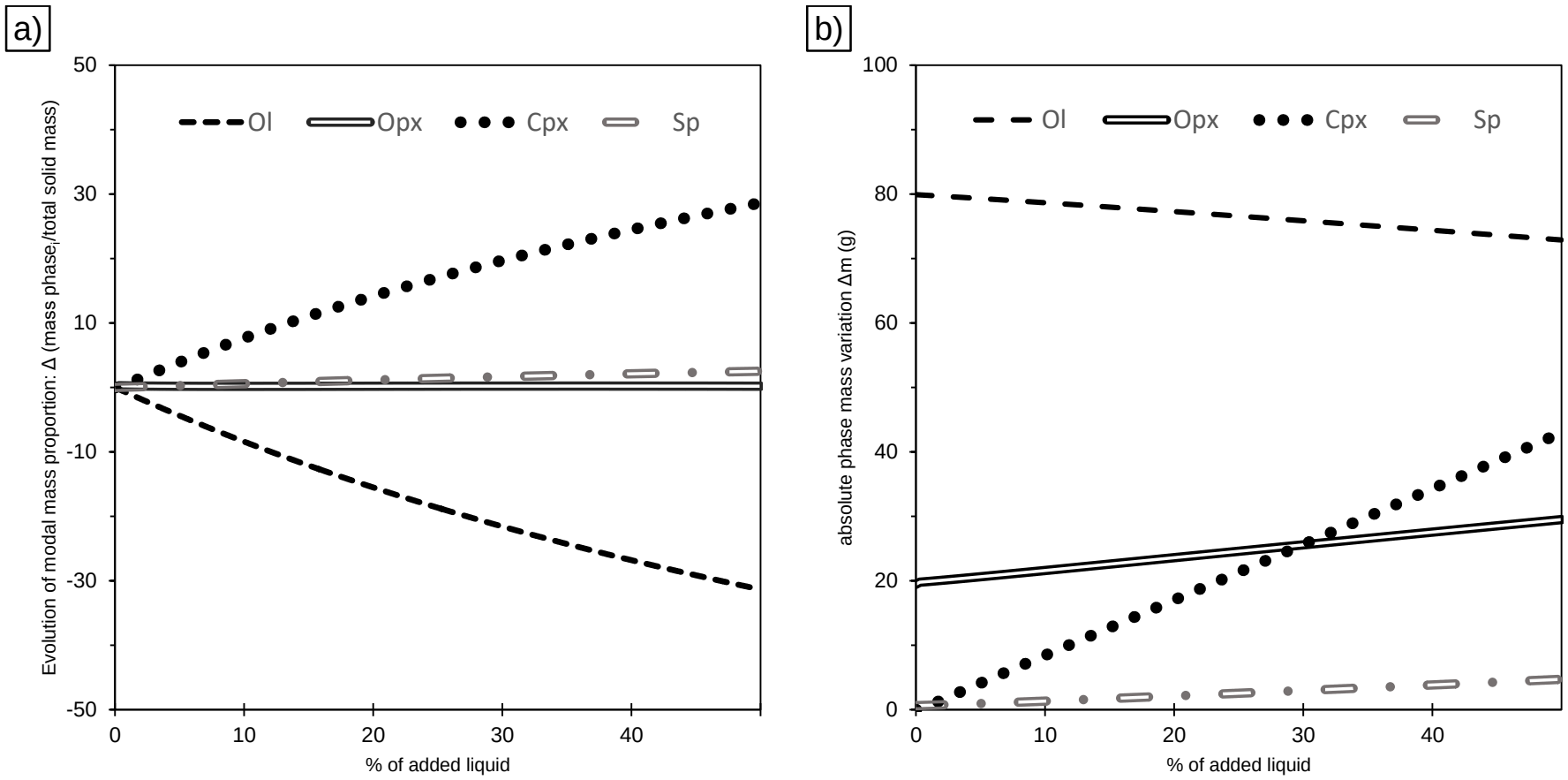
Fig 2. Evolution of modal mass proportions (in wt.%) of the different phases as a function of % of liquid added for run #1. a) Difference between modal proportion of each phase in the impregnated peridotite and its initial modal proportion in the starting material. For example, olivine represents 79.9 wt.% of the initial starting material but only 48.6 wt.% of the solid phase present at the end of the experiment, a change of -31.3. b) Absolute mass variation of each phase, for olivine, 79.9g of material are present at the beginning of the run, but only 72.9g at the end. (Ol: Olivine; Opx: Orthopyroxene; Cpx: Clinopyroxene; Sp: Spinel)

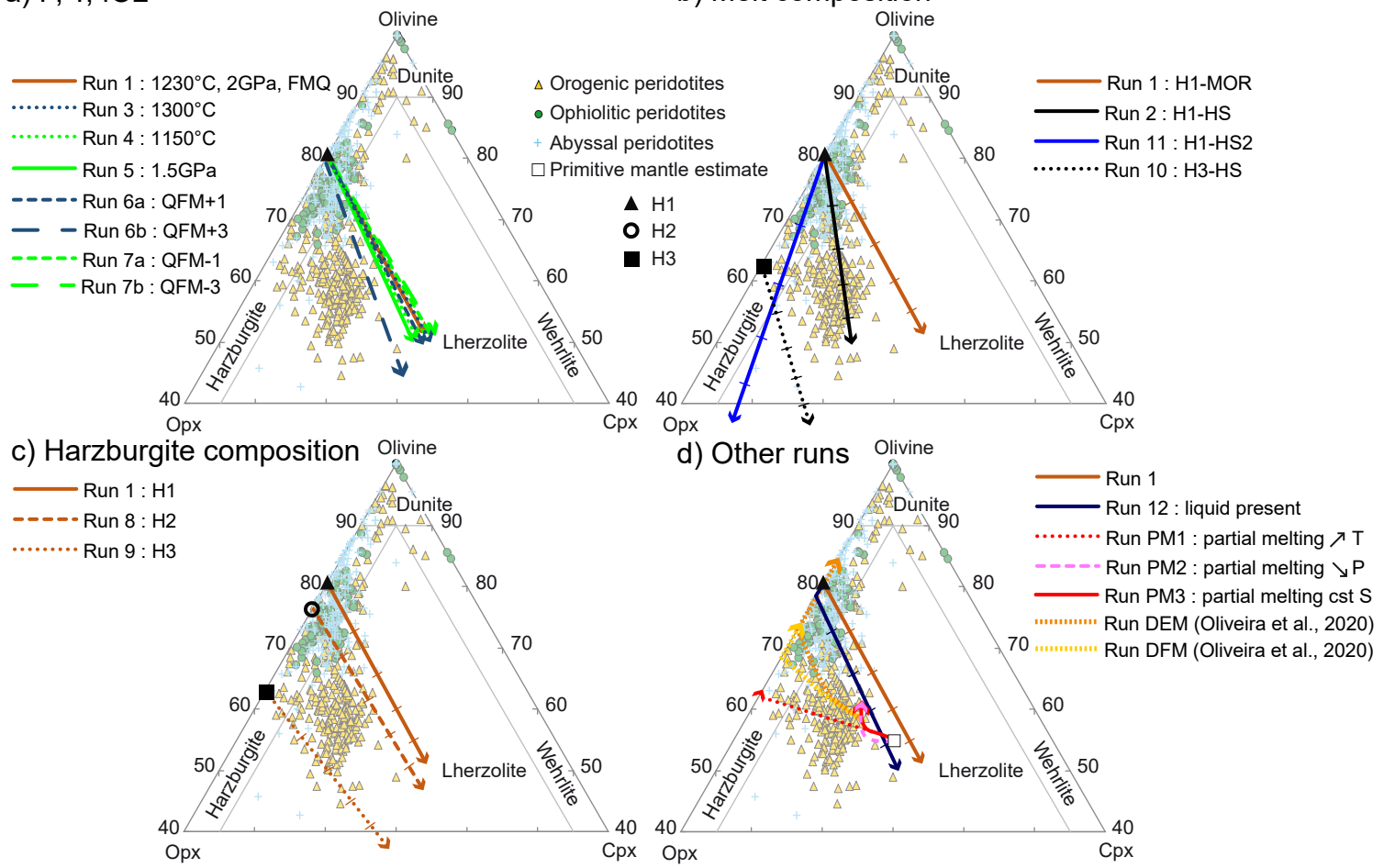
Fig 3. Evolution of modal proportions in models. Arrows indicate the direction of evolution of each run. In the impregnation calculations (runs #1 to #11), each tick mark indicates an addition of 10% of liquid. a) Runs #1 to #7b: influence of variations in pressure, temperature and redox conditions. b) Runs #1, #2, #10, and #11: influence of the chemical composition of the percolating melt c) Runs #1, #8, and #9: influence of the chemical composition of percolated harzburgite. d) Run #12: liquid-phase present run; run PM1: partial melting by isobaric rising temperature; run PM2: partial melting by decompression at constant T; run PM3: partial melting by isentropic decompression (constant S); and run #1: reference run. Partial melting runs by Oliveira et al., 2020 are shown for comparison (DEM: Dynamic Equilibrium Melting; DFM: Dynamic Fractional Melting). Primitive upper mantle estimate is from McDonough, 1995. Results are compared with modal proportions of a global database of tectonically emplaced peridotites (Bodinier & Godard, 2014, and references therein).

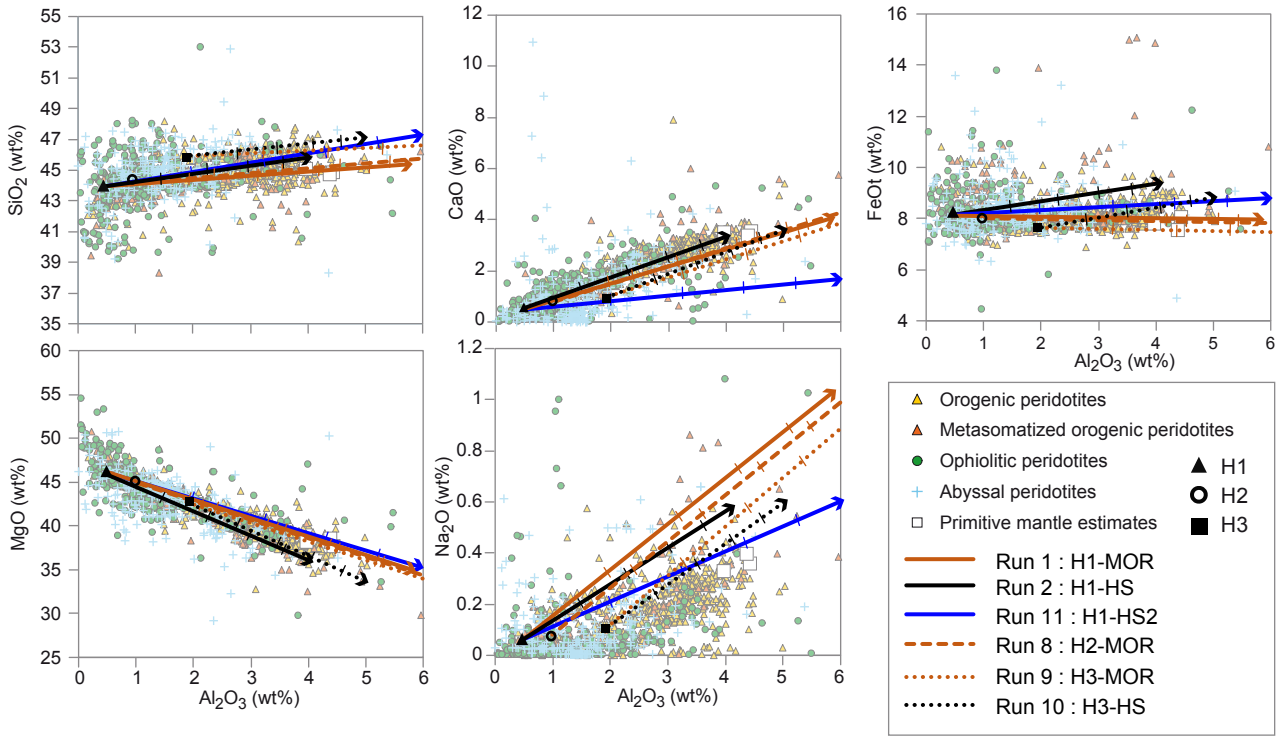
Fig 4. Chemical diagrams displaying the variation of major element oxides (wt%) of the 6 runs comparing the influence of compositional variations (runs #1, #8, and #9: variation of the harzburgite composition; runs #1, #2, #10, and #11: variation of the percolating melt composition). Arrows indicate the direction of evolution of each test. Crosses on slopes are every 10% of liquid added. FeO_t: total iron content expressed as FeO. Results are compared with a global database of peridotites (Bodinier & Godard, 2014, and references therein).

Fig 5. Variation of bulk major element concentrations (wt%) for the residual solid phase in runs #1, #12, PM1, PM2 and PM3. Run #12 is an impregnation run performed with a liquid phase present. Run PM1 is an isobaric partial melting run. Run PM2 is a partial melting run by decompression at constant T. Run PM3 is a partial melting run by isentropic decompression. Run #1 is shown for comparison. Arrows indicate the direction of evolution of each test. For runs #12 and #1, tick-marks on arrows indicate every 10% of liquid added. FeO_t: total iron content expressed as FeO. Results are compared with a global database of peridotites (Bodinier & Godard, 2014, and references therein).


Silicate melt
Lherzolite
Harzburgite
Lherzolite
Refertilization
Partial melting
Silicate melt


a)
b)
Ol
Opx
Cpx
Sp
Evolution of modal mass proportion: Δ (mass phase/total solid mass)
absolute phase mass variation Δm (g)
% of added liquid
% of added liquid
50
30
10
-10
-30
-50
100
80
60
40
20
0
0
10
20
30
40


a) P, T, fO2
Run 1 : 1230°C, 2GPa, FMQ
Run 3 : 1300°C
Run 4 : 1150°C
Run 5 : 1.5GPa
Run 6a : QFM+1
Run 6b : QFM+3
Run 7a : QFM-1
Run 7b : QFM-3
Orogenic peridotites
Ophiolitic peridotites
Abyssal peridotites
Primitive mantle estimate
H1
H2
H3
b) Melt composition
Run 1 : H1-MOR
Run 2 : H1-HS
Run 11 : H1-HS2
Run 10 : H3-HS
c) Harzburgite composition
Run 1 : H1
Run 8 : H2
Run 9 : H3
d) Other runs
Run 1
Run 12 : liquid present
Run PM1 : partial melting ↗ T
Run PM2 : partial melting ↘ P
Run PM3 : partial melting cst S
Run DEM (Oliveira et al., 2020)
Run DFM (Oliveira et al., 2020)
Olivine
Dunite
Harzburgite
Lherzolite
Wehrlite
Opx
Cpx
90
80
70
60
50
40


SiO2 (wt%)
CaO (wt%)
FeOt (wt%)
MgO (wt%)
Na2O (wt%)
Al2O3 (wt%)
Orogenic peridotites
Metasomatized orogenic peridotites
Ophiolitic peridotites
Abyssal peridotites
Primitive mantle estimates
H1
H2
H3
Run 1 : H1-MOR
Run 2 : H1-HS
Run 11 : H1-HS2
Run 8 : H2-MOR
Run 9 : H3-MOR
Run 10 : H3-HS

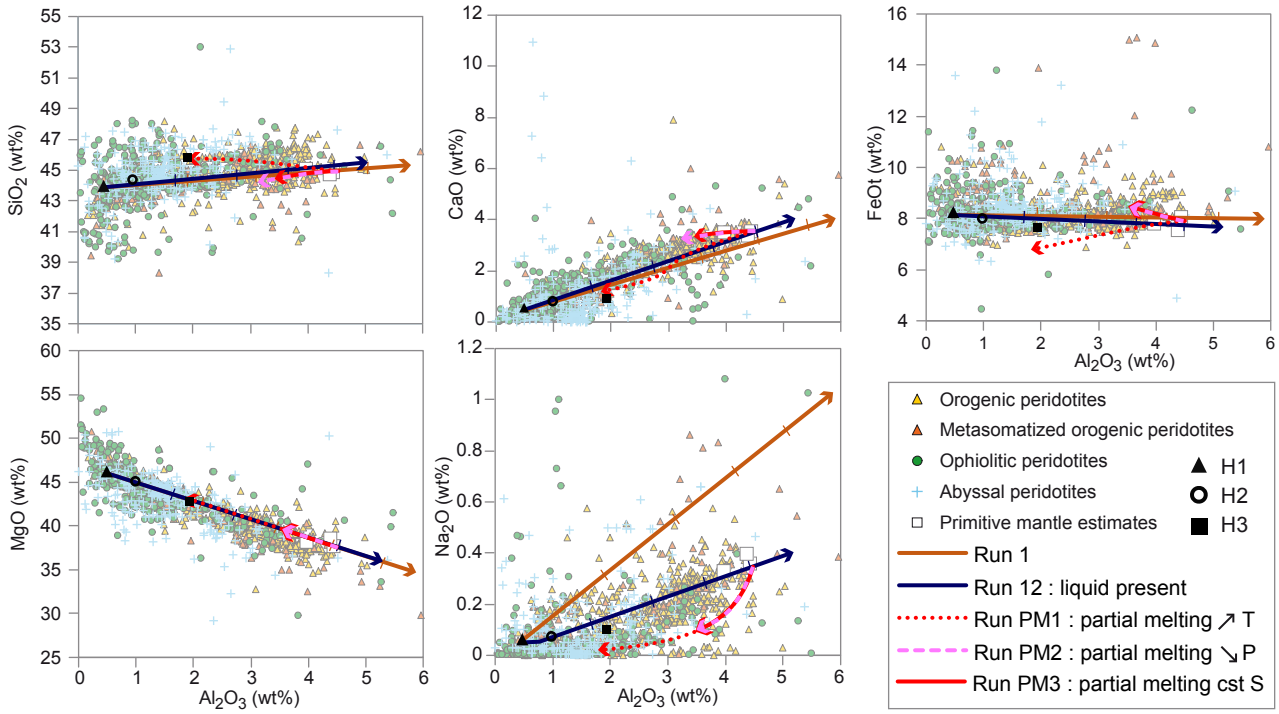

SiO2 (wt%)
CaO (wt%)
FeOt (wt%)
MgO (wt%)
Na2O (wt%)
Al2O3 (wt%)
Orogenic peridotites
Metasomatized orogenic peridotites
Ophiolitic peridotites
Abyssal peridotites
Primitive mantle estimates
H1
H2
H3
Run 1
Run 12 : liquid present
Run PM1 : partial melting ↗ T
Run PM2 : partial melting ↘ P
Run PM3 : partial melting cst S

Table 1. Summary of thermodynamic parameters used in the calculations: temperature (T), pressure (P), oxygen fugacity (fO_2), starting composition and basaltic melt composition. (PM: Partial Melting; FMQ: Fayalite-Magnetite-Quartz oxygen buffer; H: Harzburgite; PUM: Primitive Upper Mantle; MOR: Mid-Ocean Ridge; HS: Hot Spot)

Run n°	T (°C)	P (GPa)	fO_2	Starting composition	Melt composition
1	1230	2	FMQ	H1	MOR
2	1230	2	FMQ	H1	HS
3[a]	1300	2	FMQ	H1	MOR
4	1150	2	FMQ	H1	MOR
5[a]	1230	1.5	FMQ	H1	MOR
6a	1230	2	FMQ+1	H1	MOR
6b	1230	2	FMQ+3	H1	MOR
7a	1230	2	FMQ-1	H1	MOR
7b	1230	2	FMQ-3	H1	MOR
8	1230	2	FMQ	H2	MOR
9	1230	2	FMQ	H3	MOR
10	1230	2	FMQ	H3	HS
11	1230	2	FMQ	H1	HS2
12[a]	1400	2	FMQ	H1	MOR
PM1	1200 - 1520	2	FMQ	PUM	-
PM2[b]	1230	2 – 0.6	FMQ	PUM	-
PM3[c]	1396 - 1213	2 – 0.6	FMQ	PUM	-

[a] Liquid-present runs

[b] Isothermal run

[c] Isentropic run

Table 2. Chemical compositions of peridotites used in calculations.[a] (H: Harzburgite; PUM: Primitive Upper Mantle)

	SiO_2	Al_2O_3	FeOt	MnO	MgO	CaO	Na_2O	Cr_2O_3	TiO_2
H1[b]	43.91	0.47	8.19	0.13	46.57	0.38	0.05	0.31	0.01
H2[b]	44.34	0.97	7.99	0.13	45.39	0.64	0.06	0.46	0.02
H3[b]	45.82	1.92	7.49	0.13	42.95	0.75	0.09	0.82	0.04
PUM[c]	45.03	4.45	8.06	0.14	37.83	3.55	0.36	0.38	0.20

[a] All compositions are normalized to 100 wt %. FeO_t: total iron content expressed as FeO.

[b] Le Roux et al.,2007; [c] McDonough, 1995.

Table 3. Chemical compositions of percolating basaltic melts used in calculations.[a] (MOR: Mid-Ocean Ridge; HS: Hot Spot)

	SiO_2	Al_2O_3	FeOt	MnO	MgO	CaO	Na_2O	Cr_2O_3	TiO_2	a_{SiO2} [b]
MOR[c]	48.76	16.65	7.20	0.14	11.48	11.75	3.03	0.12	0.87	0.372
HS[d]	50.01	11.37	11.48	0.18	13.98	9.26	1.66	0.00	2.06	0.485
HS2[e]	54.32	17.18	9.93	0.18	11.98	3.87	1.73	0.12	0.69	0.551

[a] *All compositions are normalized to 100 wt %. FeO_t: total iron content expressed as FeO.*

[b] *Silica activities were calculated using the MELTS supplemental calculator.*

[c] *Lambart et al.,, 2009;* [d] *Norman & Garcia, 1999;* [e] *Jakobsson, 1978.*

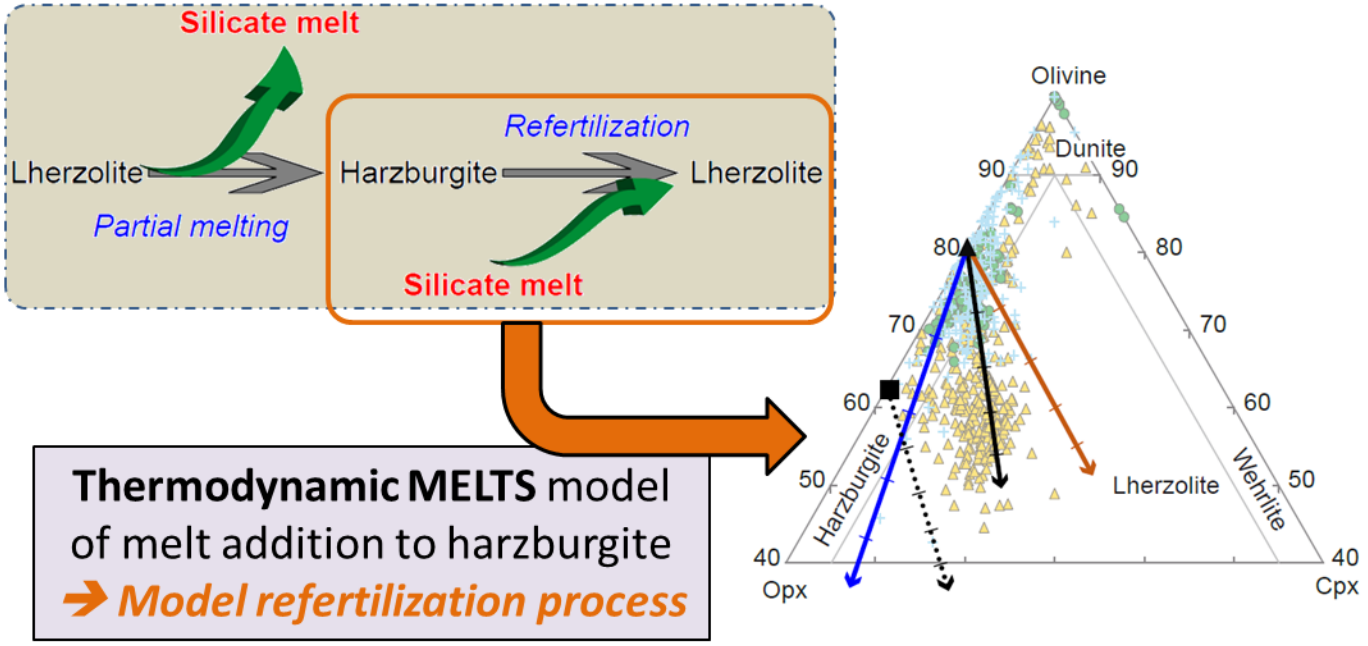
Table 4. Final phases proportions for all runs (in wt.%). (PM: Partial Melting)

Run n°	Liquid	Olivine	Orthopyroxene	Clinopyroxene	Spinel
1	0.0	48.6	19.7	28.6	3.1
2	0.0	48.7	30.2	19.5	1.5
3	0.9	47.6	19.4	28.9	3.1
4	0.0	48.6	21.2	27.0	3.3
5	1.6	47.3	21.7	26.2	3.3
6a	0.0	47.5	20.8	28.4	3.3
6b	0.0	42.5	25.8	27.4	4.4
7a	0.0	49.2	19.0	28.8	3.1
7b	0.0	49.7	18.4	28.9	3.0
8	0.0	44.7	22.3	29.3	3.7
9	0.0	35.0	31.3	29.0	4.6
10	0.0	34.6	42.4	19.8	3.1
11	0.0	35.1	52.5	8.5	3.9
12 [a]	7.5	44.0	21.8	23.8	3.0
PM1	29.8	43.8	25.2	0.0	1.2
PM2 [b]	7.5	56.2	20.9	12.7	2.8
PM3 [c]	4.9	57.7	21.6	13.0	2.7

[a] Liquid-present run

[b] Isothermal run

[c] Isentropic run


Silicate melt
Lherzolite
Partial melting
Refertilization
Harzburgite
Lherzolite
Silicate melt
Olivine
Dunite
90
80
70
60
50
40
Harzburgite
Lherzolite
Wehrlite
Opx
Cpx
Thermodynamic MELTS model
of melt addition to harzburgite
Model refertilization process