1	Revision 1
2	Reaction between volatile-bearing eclogite and harzburgite as a function of
3	degree of interaction: experimental constraints at 4 GPa
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9	
10	ABSTRACT
11	The mantle is known to be heterogeneous, mainly composed of peridotite and eclogite.
12	Eclogite-derived hydrous melts may interact with harzburgite at the slab-mantle interface in
13	subduction zones or in the sub-continental lithospheric mantle. In this study, such interactions
14	were simulated by performing hybrid experiments in which a layer of eclogite has been
15	juxtaposed to a layer of harzburgite, in the presence of H <sub>2</sub> O-CO <sub>2</sub> at 4 GPa and 1200°C,
16	conditions where eclogite is super-solidus while harzburgite is sub-solidus. A diamond trap
17	was placed in between the two layers in order to trap the fluid or melt phase, allowing direct
18	determination of their composition. The multi-anvil was rotated in different frequencies in
19	order to examine the effect of increasing degree of interaction on the melt composition as well
20	as the mineral compositions. The interaction of eclogite-derived hydrous melt and harzburgite
21	results in a reaction layer at the interface between the two lithologies, composed of opx and
22	garnet. The harzburgite above the reaction layer is metasomatized, containing various

23 amounts of olivine, opx, cpx and garnet. The eclogitic melt is modified during this interaction. 24 With increasing interaction, a thicker reaction layer is formed. Both the eclogitic and the 25 peridotitic garnet compositions approach each other and become intermediate between the 26 composition of the garnet in the eclogite+H<sub>2</sub>O+CO<sub>2</sub> system and the garnet in the 27 harzburgite $+H_2O+CO_2$  system at these conditions. The Mg# of the peridotitic olivine and opx 28 decreases with increasing interaction. The initial basaltic melt in equilibrium with eclogitic 29 garnet is metaluminous, turning to a peralkaline melt with increasing interaction with the 30 harzburgite. The metasomatizing effect of the eclogite-derived hydrous melt on the 31 harzburgite is observed by increasing the mode of the peridotitic opx, cpx and garnet at the 32 expense of peridotitic olivine and eclogitic garnet. Slight increase in melt fraction occurs as 33 well. This interaction also results in a gradient in the logfO<sub>2</sub>. Relatively more oxidizing 34 conditions occur near the reaction layer, becoming more reduced into the peridotite 35 suggesting that the reaction zones act as partial barriers for the melt to travel through the 36 peridotite. Increased interaction leads to higher logfO<sub>2</sub> values. These experiments demonstrate 37 the influence of the degree of interaction on the range of melt compositions found in volcanic 38 arcs as well as the degree of metasomatism in the mantle found in the sub continental 39 lithospheric mantle.

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#### INTRODUCTION

Heterogeneity in the Earth's mantle has been vastly documented through numerous mineralogical, geochemical, and experimental studies of rocks and magmas (e.g., Allègre and Turcotte 1986; Qian et al. 2022; Qiao et al. 2023; Sobolev et al. 2005; Stachel et al. 2018; Stracke 2012 among others). Geophysical studies have also revealed significant variations in the seismic velocity and density of the mantle, indicating the existence of different domains at

46 depth (e.g., Schaffer and Lebedev 2013). Subducted oceanic crust is the most dominant form 47 of heterogeneity in the mantle. For example, eclogitic lenses and veins are known to be embedded in the peridotitic mantle (e.g., Helffrich and Wood 2001; Hofmann 1988; Hofmann 48 49 1997; Jackson and Dasgupta 2008; Lassiter and Hauri 1998). Melting of hydrothermally 50 altered eclogite occurs at significantly lower temperatures than hydrous or dry peridotite at the 51 same pressure (Dasgupta et al. 2005; Dasgupta et al. 2004; Eggler 1978). Hence, a 52 heterogeneous mantle zone consisting of both peridotite and hydrous eclogite may result in 53 the formation of eclogite-derived hydrous melt interacting with the host peridotite. Such 54 interactions may occur in young subduction zones, where the hot oceanic slab undergoes 55 melting. The eclogite-derived hydrous melt rises into and interacts with the depleted 56 peridotitic mantle just above the subducting slab. In the sub continental lithospheric mantle, 57 eclogite-derived hydrous melt may infiltrate and interact with the surrounding harzburgite 58 (Stachel and Harris 2008).

59 Evidence for the interaction of eclogite-derived hydrous melt and peridotite is abundant in ophiolites, peridotite massifs and xenoliths (e.g., Kelemen et al. 1990; Wang et al. 2016). For 60 61 example, the formation of orthopyroxenite veins in peridotite, found in cratonic (e.g., Bell et 62 al. 2005; Boyd 1989; Kelemen et al. 1992; Kelemen et al. 1998) and orogenic peridotites 63 (e.g., Malaspina et al. 2006), is typically attributed to the infiltration of eclogite-derived 64 hydrous melt into the host peridotite. In places, opx veins are associated with hydrous 65 minerals such as amphibole and phlogopite (Xu et al. 2020), supporting the hydrous nature of the interaction. The thickness of such veins can range from a few centimeters to several 66 meters (i.e., Le Roex et al. 2014; Wang et al. 2016; Xu et al. 2008). Arc lavas have a wide 67 68 range of compositions, from silica saturated to silica undersaturated basalts to dacite.

69 Chemical and isotopic compositions support the idea that many arc magmas have complex 70 multi-source petrogenesis (Atherton and Tarney, 1979; DePaolo, 1981; Hildreth, 1981; Kay, 71 1980). Reaction of eclogite-derived hydrous melt and peridotite has also been advocated to 72 result in the formation of Mg-rich basalts and andesites in volcanic arcs (e.g., Rapp et al. 73 1999; Yogodzinski et al. 1994).

The infiltration of eclogite-derived melt into a peridotite leads to changes in the chemistry of the peridotite minerals (cryptic metasomatism) or the formation of new minerals (modal metasomatism) or both. The interaction also results in changes in the melt chemistry. Thus, the interaction of an eclogite-derived melt with peridotite can lead to the formation of a wide range of rocks and melts. The specific outcome of such an interaction will depend on the conditions under which it occurs (pressure, temperature, composition of the reacting melt and the host peridotite) as well as the nature of the reacting process and the of melt to rock ratio.

81 Many experimental studies simulated mantle metasomatism by adding  $H_2O\pm CO_2$  to a 82 single lithology (peridotite or eclogite) and exposing the system to a wide range of pressure-83 temperature conditions corresponding to the upper mantle [see compilation in Dvir and Kessel 84 (2017) and Elazar et al. (2019)]. These studies are important in establishing the phase 85 relations of the mantle in the presence of volatiles, as well as determining the P-T conditions 86 of melt initiation, i.e., the location of the volatile-bearing solidus of each system. However, 87 these studies cannot simulate complex metasomatic and melting reactions in a heterogeneous 88 mantle, specifically the interaction of eclogite-derived hydrous melts and peridotitic mantle. 89 Thus, hybrid (eclogite+peridotite) experiments are needed.

Two major capsule set-ups are employed in studying hybrid systems. One design consists
of a mixture of an eclogite or an eclogite-derived melt and a peridotite powder in various

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92 ratios in an attempt to study the equilibrium phase relationship as complete reaction was 93 documented in such designs. In such a setup it is assumed that a reactive porous flow occurs 94 in the mantle leading to complete infiltration of the eclogite-derived melt into the peridotite. 95 Gao et al. (2019), Meltzer and Kessel (2020) and Saha et al. (2018) are examples of such an 96 approach when studying the infiltration of hydrous silicate melts into a peridotite. The second 97 design is a layered capsule. An eclogite powder or eclogite-derived melt powder is placed 98 adjacent to a peridotite powder in a single capsule, simulating a scenario where an eclogite-99 derived melt is able to escape the eclogite and rise into the peridotitic mantle via a 100 channelized flow. In such a scenario, representing a more realistic simulation of such 101 processes, the interaction is limited to the melt-rock interface.

102 Most layered hybrid experimental studies were performed on anhydrous systems in order 103 to study the evolution of ocean island basalts or melting during upwelling mantle (e.g., 104 Borghini et al. 2022; Mallik and Dasgupta 2012; Takahashi and Gao 2015; Wang et al. 2010; 105 Yaxley and Green 1998). Only a few studies involved volatiles in the experiments in order to 106 better understand processes occurring at subduction zones or in the subcontinental 107 lithospheric mantle (Carroll and Wyllie 1989; Gao et al. 2019; Gervasoni et al. 2017; 108 Johnston and Wyllie 1989; Rapp et al. 1999; Sekine and Wyllie 1983). Most of these studies 109 focused on the interaction of eclogite-derived hydrous melt and a fertile peridotite at 1-4 GPa, 110 resulting in the formation of high-Mg andesite and dacite melts and enrichment of opx and grt 111 on the expense of olivine in the metasomatized lherzolite. However, in such geological 112 settings, an eclogite-derived hydrous melt mostly interacts with a depleted peridotite. Yet, 113 studies on the interaction of eclogite-derived hydrous melts and harzburgite are rare.

In this contribution, we study the infiltration of a  $H_2O-CO_2$ -bearing eclogite-derived melt into a harzburgite at 4 GPa and 1200°C. The design used in this study is a layered capsule, with a diamond trap separating the two lithologies. Such a design allows us to directly analyze the liquid phase (either fluid or melt), as well as its influence on the mantle mineralogy and chemistry. Experiments were performed in a rocking multi anvil, rotated at various frequencies in order to study the progressive interaction of the eclogite-derived hydrous melt and the host harzburgite.

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### METHODS

122 Starting materials

123 The starting materials used in this study consist of a synthetic carbonated K-bearing 124 basaltic powder (labeled CKB) and a synthetic depleted harzburgite (labeled DHRZ). For the 125 basaltic composition we used an anhydrous eclogite composition similar to ATCM1 (Thomson et al. 2016). This composition was designed to represent subducted oceanic crust 126 127 that has undergone dehydration reactions but no partial melting. It retains a carbonate 128 component ( $\sim 3 \text{ wt}\% \text{ CO}_2$ ), and elevated SiO<sub>2</sub> relative to other basaltic starting compositions 129 (Fig. 1). In contrast to the low  $K_2O$  (0.06 wt%) content used by (Thomson et al. 2016), we 130 used an elevated K<sub>2</sub>O content (0.49 wt%), similar to KMB-7 (Schmidt and Poli 1998). The 131 composition of the depleted harzburgite was taken from a reference natural xenolith GP8, part 132 of a suite of ultra-depleted xenoliths from the southern Zimbabwe Craton (Smith et al. 2009). 133 These peridotitic xenoliths are characterized by high-Mg# olivine (Mg# $\sim 0.94$ ), as well as very low HREE abundances and low Yb/Lu ratios. They are thought to represent some of the most 134 135 pristine examples of residua formed via high degree melting during craton stabilization. This

136 composition was chosen primarily because the mineralogical change due to the metasomatic

137 effect of fluid/melt liberated from the carbonate eclogite is relatively easy to assess.

138 Both starting compositions were prepared using reagent grade oxides. SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, FeO, 139 MgO and Cr<sub>2</sub>O<sub>3</sub> were sintered for one hour, and TiO<sub>2</sub> NiO and MnO were sintered for 4 140 hours, all at 800°C. CO<sub>2</sub> was added to the CKB basaltic powder via  $Na_2(CO)_3$ , CaCO<sub>3</sub> and 141  $K_2(CO)_3$  which were sintered for 4 hours at 400°C. The dried oxides and carbonates where 142 mixed at the appropriate proportions in both powders and ground under ethanol in an agate 143 mortar for several hours, ensuring homogenous powders. Mineral seeds were not added to the 144 starting material. The basaltic starting material was doped with 57 ppm Th. The harzburgitic 145 powder was doped with 36 ppm Cs. Both trace elements were added as internal standards for 146 quantification of LA-ICP-MS analysis of the fluid/melt phase, following the cryogenic 147 technique developed by Kessel et al. (2004). Th was added by diluting 1000 ppm HCl and 148 HNO<sub>3</sub> standard solutions with distilled water. Cs was added as Cs<sub>2</sub>CO<sub>3</sub> dissolved in HNO<sub>3</sub> 149 and was also diluted from a 1000 ppm standard solution using distilled water. The diluted 150 solutions were added to the staring materials and were mixed thoroughly in an agate mortar. 151 Finally, the bulk powders were doped with nano-grains of pure iridium (Ir) metal (<60 152 microns, Chempur® LTD, Poland), such that it constituted ~3% of the powder. The Ir grains 153 acted as a redox sensor for oxygen fugacity determination, following the technique developed 154 by Woodland and O'Neill (1997) and Stagno and Frost (2010). The final synthetic powders 155 were stored at 110°C to avoid absorption of atmospheric water.

The major element compositions of the synthetic powders were determined at the geochemical lab of the Geological Survey of Israel using a Perkin Elmer Optima 3300 ICP-OES against USGS standards, resulting in a confidence level of  $\pm 1\%$  for the measurements.

Aliquots for the determination of Th and Cs concentrations were dissolved in HF+HNO<sub>3</sub> and analyzed by ICP-MS at the Geological Survey of Israel. The compositions are presented in Table 1. The bulk CO<sub>2</sub> content in both starting materials was determined separately, using the QTS (Quartz Tube System) technique developed by Dvir et al. (2013). Two QTS measurements on each powder were performed (Table 1). The eclogite powder contains  $\sim$ 3 wt% CO<sub>2</sub>. Although no CO<sub>2</sub> was added to the harzburgite, it was found to contain  $\sim$ 0.5 wt% CO<sub>2</sub>, most probably the result of contamination during preparation.

166 **Capsule design** 

167 Two control experiments were performed. One experiment contained  $eclogite+H_2O+CO_2$ 168 and the second experiment contained harzburgite+H\_2O+CO\_2. These experiments were run in 169 order to assess the chemistry of the fluid/melt and minerals and their proportions in each 170 system before they interact with each other. In addition, three hybrid experiments were 171 performed, containing both eclogite and harzburgite in order to assess the interaction of 172 eclogite-derived melt and harzburgite in the presence of H\_2O+CO\_2.

173 All experiments were conducted using Au capsules (2.0 outer diameter, 1.9 mm inner 174 diameter). First,  $\sim 1 \mu L H_2O$  was inserted at the bottom of the capsule (measured with a 175 micropipette and confirmed by weight). Then, a layer of powder (5-6 mg) was added, overlain 176 by a layer of 15-25 µm diameter synthetic diamond grains (2-3 mg). Finally, 5-6 mg of 177 powder were placed on top of the diamond layer. In the control experiment, the same powder was added below and above the diamond layer. In the hybrid experiments, eclogite was 178 179 inserted below and harzburgite above the diamond trap. The weight ratio between the eclogite 180 and harzburgite powders in the hybrid experiments was between 0.99-1.11. In all experiments 181 the diamond trap constituted 16-20 wt% of the solid material in the capsule. Capsules were

welded while frozen in liquid nitrogen, in order to minimize the loss of  $H_2O$  and  $CO_2$  during welding and weighed both prior to and after welding and folding. Only capsules that demonstrated no significant loss of material during capsule preparation were used. Finally, the capsules were squeezed to a 2-3 mm long cylinders. The experimental setup for this study is given in Table 2.

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# **Experimental approach**

188 Experiments were performed at constant pressure of 4 GPa and constant temperature of 189 1200°C, conditions where the volatile-bearing eclogite is expected to partially melt while the 190 volatile-bearing harzburgite remains solid. Experiments were conducted in a 675-ton rocking 191 multi anvil press at the Institute of Earth Sciences, the Hebrew University of Jerusalem, Israel. 192 Pressure was calibrated at high temperature (1000-1200°C) for quartz-coesite, fayalite-193  $\gamma$ Fe<sub>2</sub>SiO<sub>4</sub>, and coesite–stishovite phase transitions (Dvir et al. 2011) and is within ±0.2 GPa of 194 the quoted values. Temperature was measured using a  $Pt-Pt_{10}Rh$  (type-S) thermocouple; 195 reported temperatures are not corrected for the effect of pressure on the thermocouples 196 electromotive force and are within  $\pm 2^{\circ}$ C from the thermocouple reading. Experiments were 197 performed using 19 mm gasket edged length MgO octahedra [fabricated from MgO-based 198 castable ceramics (Ceramacast 584)] and tungsten carbide cubes with truncation edge length 199 of 11 mm.

Each experiment contained two identical capsules, vertically placed one on top of the other, although only one capsule in each experiment was used in this study. Each capsule was loaded into a boron-nitride cylinder to minimize hydrogen loss (Truckenbrodt and Johannes 1999; Truckenbrodt et al. 1997). The boron nitride cylinder was placed within a 4.0 mm-outer diameter stepped graphite furnace. MgO spacers and Mo caps were placed above and below

205 the two capsules to ensure electrical contact between the stepped furnace and the tungsten 206 carbide cubes. The thermocouple was placed through the octahedron gaskets, as close as 207 possible to the octahedron center between the two capsules. The temperature gradient along 208 the capsule was not determined in these experiments. However, employing a thermal gradient 209 of 11°/mm assumed to occur in such an assembly (Konzett et al. 2011) along 2-3 mm capsules 210 yields a temperature gradient from 1200°C near the thermocouple down to 1170°C at the far 211 end of the capsules.

212 Previous high-pressure experiments on static multi-anvils containing fluid-bearing systems 213 often resulted in mineralogically segregated charges, sometimes leading to unstable mineral 214 assemblages [see discussion in Schmidt and Ulmer (2004)]. Schmidt and Ulmer (2004) 215 demonstrated that rotation of the multi-anvil gravitationally drives the fluid phase to migrate 216 from one end of the capsule to the other, driving reactions forward and therefore enhancing 217 chemical equilibrium throughout the experimental charge. In all experiments in this study 218 (except the static experiment #40), the multi-anvil was initially rotated 180° constantly (each 219 rotation takes 15 seconds) for the first hour, in order to provide equilibrium nucleation centers 220 throughout the capsule (Melekhova et al. 2007). After 1 hour had elapsed, the rotation was set 221 to occur at a different frequency in each experiment for the remaining duration of the 222 experiment (Table 2). The two control experiments (#30 and #41) were rotated every 15 223 minutes. The hybrid experiment #35 was rotated every 15 minutes while the hybrid 224 experiment #38 was rotated constantly. The hybrid experiment #40 remained static. The 225 higher the frequency of rotation, the frequent migration of the fluid or melt from one end of 226 the capsule to the other, leading to more interaction between the fluid or melt and the rock.

Increasing frequency of rotation is qualitatively taken here as an indication of increasingdegree of interaction.

All experiments were run for 21-24 hours and terminated by turning off the furnace power followed by a slow unloading of pressure. Once pressure was unloaded, the assemblage was taken out and prepared for analyses.

232 Analytical methods

233 High-pressure H<sub>2</sub>O-rich fluids and melts are rich in solute and are thus unquenchable [see 234 discussion in Kessel et al. (2004)]. Being a homogeneous phase at run P-T, the fluid or melt 235 undergoes exsolution during quenching, producing a mixture of H<sub>2</sub>O-rich phase and quench 236 precipitates. Opening the capsule in order to prepare for analyses leads to the escape of the 237 H<sub>2</sub>O-rich phase containing highly volatile cations (such as Na, K, Cs), leaving behind only the 238 quench precipitates. In order to prevent such a fractionation, the fluid or melt phase 239 composition was determined following the cryogenic technique developed by Kessel et al. 240 (2004). In this technique, the capsule is frozen while closed and then cut vertically to expose 241 the diamond trap. The frozen diamond trap is analyzed using a 193 nm RESOlution 193 nm 242 ArF excimer laser system (ASI, Australia) coupled to an AnalytikJena Plasma Quant MS-243 Elite ICP-MS at the Institute of Earth Sciences, the Hebrew University of Jerusalem, Israel. 244 An ablation spot diameter of 50-100  $\mu$ m is used depending on the diamond trap geometry. 245 Data was acquired in blocks of 6-14 individual analyses bracketed by 4-6 analyses of the 246 NIST-SRM-610 standard. Dwell time was typically 40 and 60 seconds. Background was 247 counted for 10-40 seconds and the signal for at least 40 seconds. Analyses were performed for <sup>23</sup>Na, <sup>24</sup>Mg, <sup>27</sup>Al, <sup>29</sup>Si, <sup>39</sup>K, <sup>42</sup>Ca, <sup>44</sup>Ca, <sup>49</sup>Ti, <sup>57</sup>Fe, <sup>53</sup>Cr, <sup>60</sup>Ni, <sup>133</sup>Cs and <sup>232</sup>Th. In addition, <sup>12</sup>C, 248

<sup>13</sup>C, <sup>197</sup>Au and <sup>193</sup>Ir were also analyzed in order to assess the position of the laser beam. Data
was reduced using the SILLS program (Guillong et al. 2008).

251 The Th doped into the eclogite powder is assumed to be a highly incompatible element in 252 eclogitic systems [bulk partition coefficient of 0.002-0.02 between eclogite and H<sub>2</sub>O-CO<sub>2</sub>-253 fluids or melts (Elazar et al. 2019)]. The Cs doped into the harzburgite powder is assumed to 254 be a highly incompatible element in peridotitic systems. Bulk partition coefficients for Cs are 255 in the range of 0.007-0.003 between peridotite and carbonatitic melts (Dasgupta et al. 2009) 256 and 0.002-0.01 between peridotite and hydrous fluids or melts (Kessel et al. 2015). It is, 257 therefore, assumed that all the Th and Cs in the capsule reside in the fluid or melt phase at 258 equilibrium. The known amount of Th and Cs in the fluid or melt is used to convert the 259 elemental ratios derived from the LA-ICP-MS analyses of the diamond trap to the anhydrous 260 composition of the fluid or melt, following the technique developed in Kessel et al. (2004).

261 Following the LA-ICP-MS analysis of the total dissolved solids in the fluid/melt phase, 262 the capsule was defrosted and prepared for analysis of the minerals. Capsules were polished using an ATM<sup>©</sup> Sapphire 520 grinder and polisher using anhydrous silicon carbide (SiC) and 263 264 polycrystalline diamond pastes in order to prevent loss of volatile phases. Phase identification, 265 composition and textural relations were established using a Jeol-JXA-8230 electron probe 266 micro analyzer (EPMA) at the Hebrew University of Jerusalem, Israel. All measurements 267 were preformed using an accelerating voltage of 15 keV and beam current of 15 nA, against 268 silicate and oxide standards. Chemical characterization of the phases was performed with a 269 minimum duration of 30 seconds for the signal and 10 seconds for the background. All phases 270 were analyzed for Si, Ti, Al, Fe, Mg, Ca, Na, K, and Cr. Au capsule walls were also analyzed 271 for elemental contamination.

No hydrous or carbonated crystalline phases were observed in the experiments. Thus, all the  $H_2O$  and  $CO_2$  inserted into the capsule were assumed to reside in the fluid or melt phase. The masses of  $CO_2$  and  $H_2O$  are then combined with the mass of all cations in the fluid or melt to derive the complete major element composition of the fluid or melt phase in each capsule.

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# Phase abundance calculations

The abundance of each phase in each experiment was determined using Monte Carlo calculations based on major element compositions of all minerals, fluid or melt composition and the composition of the bulk material inserted into the capsule. The anhydrous algorithm used a non-weighted least square fit, performing 10,000 calculation runs, by modifying the compositions within the error range given by 1  $\sigma$ . The 10,000 results were collected and the average and 1  $\sigma$  of the phase abundance were calculated.

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# Oxygen fugacity analyses

Oxygen fugacity in these experiments was determined by analyzing the Fe content in Ir grains throughout the capsule. The Ir grains act as  $fO_2$  sensors, following the method of Stagno and Frost (2010). In this approach, Fe in Ir grains is equilibrated with Fe in silicates and the logfO<sub>2</sub> can be calculated by employing activity-composition models for the various phases as well as their  $\Delta G^{\circ}$  (the free energy of the pure end members).

In each experiment, 11-14 Ir grains, located along the capsule from the edge of the diamond trap to the end of the peridotite layer were analyzed via EPMA. Signal time was set to 35 seconds and background time was set to 15 seconds. The concentrations of Si, Mg, Ca, Ti and Cr were monitored in these grains in order to avoid silicate contamination. Only grains

with no apparent contamination (i.e., Si, Mg, Ca and Ti concentrations below 0.2 wt%) werechosen for oxygen fugacity calculations.

296

### RESULTS

The harzburgite+ $H_2O+CO_2$  control experiment (#30) contains mostly olivine (ol) and orthopyroxene (opx) with a small amount of garnet (Fig. 2a). No clinopyroxene (cpx) is observed in the harzburgite. All phases are homogeneously distributed in both sides of the diamond trap. Quench precipitates are trapped in between the diamond grains. The eclogite+ $H_2O+CO_2$  control experiment (#41) is composed of mostly quench glass in and outside the diamond trap (Fig. 2b). Garnet crystals appear around the diamond trap and at the top of the capsule.

304 In all three hybrid experiments, the interaction of eclogite and harzburgite resulted in 305 stratified capsules (Fig. 3a-c) containing a reaction layer between the eclogite and the 306 peridotite. In the static experiment (#40), the sequence from bottom up includes a quench 307 layer composed of small quench precipitates, a layer of eclogitic garnet, the diamond trap, a 308 thin reaction layer containing large grains of garnet and opx, overlain by a peridotitic layer 309 composed of olivine, opx, cpx and small amount of garnet grains, mostly close to the reaction 310 layer. In the two rocking experiments (#35, rocked every 15 minutes and #38, rocked every 311 15 seconds), quench precipitates appear at the bottom of the capsule, overlain by the diamond 312 trap. Above the diamond trap, a thick reaction layer containing garnet+opx±cpx has formed 313 that is overlain by a peridotite layer composed of olivine, opx, cpx and some garnet. Although 314 cpx was not stable in any of the control experiments (harzburgite+H<sub>2</sub>O+CO<sub>2</sub> and 315 eclogite+H<sub>2</sub>O+CO<sub>2</sub> systems), it appears in all rocking experiments as part of the peridotite

layer. In all hybrid experiments, cracks are seen in the reaction layers, perpendicular to thelayer, from the eclogite to the peridotite.

In each experiment, 4-37 individual grains of each mineral phase were analyzed throughout the capsule. Care was taken to exclusively measure the rim compositions (the outer 10 microns), which is in equilibrium with their surrounding phases. Mineral compositions are provided in Tables 3-6 and Figs. 4-6. Uncertainties reported throughout this study are one standard deviation of n analyses (1 $\sigma$ ).

323 Quench precipitates are present in the diamond trap in all experiments, indicating the 324 existence of a fluid or melt phase in equilibrium with the surrounding rocks. In each 325 experiment, 5-14 analyses were performed along the diamond trap in order to derive the fluid 326 or melt compositions. The composition of the liquid phase in the eclogite control experiment 327 was calculated based on the Th doped into the starting material, acting as internal standards in 328 the LA-ICP-MS analyses. The composition of the liquid phase in the harzburgite control 329 experiment was calculated based on the Cs doped into the starting material. In all hybrid 330 experiment, the composition of the liquid phase was calculated once based on Th and once 331 based on Cs. The liquid phase composition calculated by both methods (via Th or Cs as 332 internal standard) are similar within uncertainties (Table S1 and Figs. S1-2 in supplement), 333 supporting our assumption of their incompatibility in such systems. The fluid or melt phase 334 compositions presented in Table 7 and Fig. 7 were derived from Th as an internal standard, 335 except in the harzburgite control experiment, where the data was derived from Cs.

336

### Harzburgite+H<sub>2</sub>O+CO<sub>2</sub> control experiment

The mineral assemblage in the harzburgite control experiment (#30) consists of ol, opx and garnet. *Olivine* is the main phase in the harzburgite. It is forsteritic in nature, with Mg#

339	[Mg/(Mg +Fe) in molar unit] of 0.94 (Fig. 4), slightly higher than the #Mg of the initial GP8
340	harzburgite composition (Table 1). It appears subhedral with size ranging from 10-50 $\mu$ m
341	throughout the capsule to 50-100 µm near the diamond trap. Orthopyroxene is enstatite-rich
342	with Mg# of 0.95 (Fig. 4), containing 0.2 wt% CaO and 1.2 wt% Al <sub>2</sub> O <sub>3</sub> . The opx appears
343	subhedral, 20-70 µm in size. Garnet is Cr-rich pyrope (Pyrop <sub>0.73</sub> Alm <sub>0.10</sub> Gros <sub>0.05</sub> ), containing
344	3.2 wt% Cr <sub>2</sub> O <sub>3</sub> and 2.3 wt% CaO (Figs. 5-6), falling within the harzburgitite field (Grütter et
345	al. 2006).

The liquid in equilibrium with the harzburgite at 4 GPa and 1200°C contains 62.6 wt% H<sub>2</sub>O and 2.9 wt% CO<sub>2</sub> (Fig. 7), indicating an aqueous fluid nature. The fluid phase is metaluminous [molar Al/(2Ca+Na+K)< 1] in character, containing 42.2 wt% SiO<sub>2</sub> and 23.2 wt% MgO.

#### 350

#### Eclogite+H<sub>2</sub>O+CO<sub>2</sub> control experiment

The eclogite+H<sub>2</sub>O+CO<sub>2</sub> control experiment (#41) is comprised solely of garnet in equilibrium with a liquid phase (Fig. 2b). Garnet appears euhedral to subhedral, located mostly above the diamond trap. The garnet grains are 10-100  $\mu$ m in size. They are characterized by Mg# of 0.62 and contain 0.01 wt% Cr<sub>2</sub>O<sub>3</sub> and 8.2 wt% CaO, falling within the eclogitic field (Fig. 6). The garnet composition is Pyrop<sub>0.41</sub>Alm<sub>0.25</sub>Gros<sub>0.19</sub> (Fig. 5).

The liquid phase in this experiment contains 6.8 wt%  $H_2O$ , 2.3 wt%  $CO_2$ , 50 wt%  $SiO_2$ and 4.5 wt%  $Na_2O+K_2O$ , thus identified as a metaluminous andesitic melt. This melt is assumed to infiltrate and react with the harzburgite in the hybrid experiments.

359 **Hybrid eclogite+peridotite experiments** 

Three hybrid experiments were performed. All experiments initially contained similar amounts of eclogite and harzburgite powder, with a diamond trap placed in between the two layers (Table 2, Fig. 3). Thus, all three capsules are considered to contain similar bulk compositions. However, each experiment was rotated at a different frequency (static, every 15 minutes, every 15 seconds).

365 In the static hybrid experiment (#40), a layer of eclogitic garnets appears below the 366 diamond trap. Their composition is similar to that in the  $eclogite+H_2O+CO_2$  control experiment, with only slightly higher CaO content and higher Mg# (Figs. 5, 6). Above the 367 368 diamond trap, a thin reaction layer was formed composed of garnet grains. They are 369 intermediate in composition between the eclogitic and harzburgitic garnets in the control 370 experiments, with composition of Pyrop<sub>0.65</sub>Alm<sub>0.16</sub>Gros<sub>0.12</sub>, Cr<sub>2</sub>O<sub>3</sub> content of 0.7 wt% and 371 CaO of 4.8 wt%. Above the reaction layer, a peridotite containing olivine, opx and cpx is 372 stable. The olivine appears as small subheadral grains, forsteritic in composition and the opx 373 is enstatitic, both are characterized by Mg# of 0.93, only slightly lower than in the 374 harzburgite+H<sub>2</sub>O+CO<sub>2</sub> control experiment (Fig. 4). The cpx grains are Mg-rich augite, 375 characterized by Mg# of 0.94; CaO of 20.0 wt%, Al<sub>2</sub>O<sub>3</sub> of 1.4 wt% and Na<sub>2</sub>O of 2.2 wt%.

In both rotating experiments (#35 and #38), no garnet is present in the eclogite side of the experiment. The diamond trap was displaced to the bottom of the capsule and a thick reaction layer was formed above the diamond layer, containing garnet and opx. The garnet grains close to the diamond trap contain 6-7 wt% CaO and  $\sim$ 0.2 wt% Cr<sub>2</sub>O<sub>3</sub>, falling in the eclogitic garnet field. They are intermediate in composition between the eclogitic garnet and the peridotitic garnet in the control experiments (Pyrop<sub>0.65</sub>Alm<sub>0.16-0.17</sub>Gros<sub>0.17-0.18</sub>). The garnet grains in the

far side of the reaction layer are more peridotitic in nature. They are slightly enriched in pyrope and depleted in almandine than the eclogitic garnets in these experiments (Pyrop<sub>0.69</sub>.  $_{0.71}Alm_{0.12-0.18}Gros_{0.13-0.17}$ ), and contain 6-7 wt% CaO and 3-5 wt% Cr<sub>2</sub>O<sub>3</sub>. The olivine in the peridotitic layer is subhedral, 10-50 µm in size. It is slightly less forsteritic than in the harzburgite control experiment and the static experiment (Mg# of 0.92). The opx in these experiments is also slightly less enstatitic with Mg# of 0.93. The jadeite component in the cpx is 0.6-0.8.

389 In all hybrid experiments, a melt phase is trapped in between the diamonds. The melt in 390 the static experiment is metaluminous, containing 50.0 wt% SiO<sub>2</sub>, 15.5 wt% H<sub>2</sub>O, 2.8 wt% 391  $CO_2$  and 4.9 wt% Na<sub>2</sub>O+K<sub>2</sub>O (Fig. 7). The melt in both rocking experiments is peralkaline 392 (molar (Na<sub>2</sub>O/Al<sub>2</sub>O<sub>3</sub> >1), containing 28-34 wt% SiO<sub>2</sub>, 20-25 wt% H<sub>2</sub>O, 5 wt% CO<sub>2</sub> and 6-7 393 wt% Na<sub>2</sub>O+ $K_2O$ . While the melt phase in the static experiment is close in composition to the 394 melt in equilibrium with eclogite only, the composition of the melt in the rocking experiments 395 is an average between the fluid in equilibrium with harzburgite and the melt in equilibrium 396 with eclogite (Fig. 7).

**397 Phase abundance** 

The phase abundance in each experiment is given in Table 8 and shown in Fig. 8. The eclogite+H<sub>2</sub>O+CO<sub>2</sub> system in this study is super-solidus, consisting of ~24% garnet and 76% melt while the harzburgite+H<sub>2</sub>O+CO<sub>2</sub> system is sub-solidus, containing 58% olivine, 25% opx, 1% garnet and 15% fluid. In the static hybrid experiment, the amount of eclogitic garnet in the bulk system is 42%. Its amount decreases to 27-28% in both rocking hybrid experiments. The peridotitic olivine in the static experiment constitutes 16% of the bulk system. Its abundance is reduced to 8-11% with increasing frequency of rotation. The

405 peridotitic opx in the static experiment constitutes 17% of the system, increasing to 26-31% as 406 rotation is set. The peridotitic cpx, absent in the original harzburgite, is formed during the 407 interaction between the eclogite-derived melt and harzburgite. It constitutes 4% of the bulk 408 system in the static experiment, increasing to 8-13% in the rotating experiments. Minor 409 amount ( $\sim$ 3%) of peridotitic garnet is also present in all hybrid experiments. The amount of 410 melt in the system increases from 19 to 25% with increasing rotation frequency.

### 411 **Oxygen fugacity**

Following Stagno and Frost (2010), the equilibrium between Ir grains, olivine and opx fixes the log $fO_2$  of the assembly. Only garnet and melt are stable in the eclogite+H<sub>2</sub>O+CO<sub>2</sub> control experiment, thus no  $fO_2$  calculations are available for this experiment.

415 Olivine, opx and Ir grains are present in the harzburgite+H<sub>2</sub>O+CO<sub>2</sub> control experiment as 416 well as in all three hybrid experiments. The compositions of olivine and opx in each of these 417 experiments are homogeneous throughout the capsule (Fig. 4). The Fe content in Ir grains was 418 analyzed in each capsule from the edge of the diamond trap to the end of the peridotite layer 419 and the local log $fO_2$  was calculated (Fig. 9). The mole fraction of Fe in the Ir grains ( $X_{Fe}^{Ir}$ ) in 420 the harzburgite+ $H_2O+CO_2$  control experiment is 0.02-0.10, with no apparent dependence on 421 the location in the capsule. The  $\log O_2$  registered by the Ir grains ranges between -7.7 to -6.1 422 (1.89±0.40 log units below the QFM buffer), considered constant within the uncertainties of the method. The  $X_{Fe}^{Ir}$  in the static experiment (#40) and the hybrid experiment rotated every 423 424 15 minutes (#35) varies systematically from 0.21 in the edge of the reaction layer to 0.31 in 425 the far end of the peridotite layer, corresponding to a decrease in  $\log fO_2$  from -8.7 to -9.5 (3.4 426 to 4.2 log units below QFM, respectively). A similar trend is observed in the hybrid experiment rotated every 15 seconds (#38), albeit at slightly higher values. The log/O<sub>2</sub> 427

428	decreases from 2.0 log units below QFM to 2.9 log units below QFM. All logfO2 values
429	recorded in these experiments are below EMOD (enstatite-magnesite-olivine-diamond)
430	reference curve, which is the limiting reaction for the diamond stability field [taken from
431	Eggler and Baker (1982)].

432

#### DISCUSSION

433 Approach to equilibrium

434 The initial synthetic powders in all capsules were converted to high-pressure and high-435 temperature mineral assemblies. Phases are homogeneously distributed in each layer and no 436 major internal zoning is observed in the minerals. The amount of Fe diffused into the Au 437 capsule is less than 0.2 wt% close to the experimental charge, indicating no significant Fe loss 438 to the capsule. Close to the edge of the Au capsule, Fe content is below detection limit (0.02 439 wt%). All these observations suggest that all capsules, including the hybrid experiments, demonstrate an equilibrium set up. In addition, the chemical compositions of opx and cpx in 440 441 the hybrid experiments indicate an equilibrium temperature of 1180-1260°C (Table 2), based 442 on the Mg-Fe exchange between the two minerals (Putirka 2008). These temperatures are within the range of temperatures experienced by the capsules. 443

444

#### Mineral and melt compositions

In both eclogite+H<sub>2</sub>O (Kessel et al. 2005) and eclogite+H<sub>2</sub>O+CO<sub>2</sub> (Elazar et al. 2019) systems, the second critical endpoint was found to be between 5 and 6 GPa, indicating the existence of a solidus at 4 GPa. Thus, the eclogite+H<sub>2</sub>O+CO<sub>2</sub> control experiment in this study (4 GPa and 1200°C) is assumed to be at super-solidus conditions. The capsule is composed of only garnet in equilibrium with melt. Mass balance calculations indicate that 76% melt is present in the capsule (Table 8). Elazar et al. (2019) studied a similar eclogite+H<sub>2</sub>O+CO<sub>2</sub>

451 system. Their eclogite, similar to SCLM1 (Dasgupta et al. 2004), is slightly depleted in SiO<sub>2</sub> 452 and Al<sub>2</sub>O<sub>3</sub> relative to the eclogite in this study (43 and 11 comparted to 51 and 15 wt%, 453 respectively, Fig. 1). While a similar  $H_2O$  content was added in both Elazar et al. (2019) and 454 this study (9 and 10 wt%, respectively), higher CO<sub>2</sub> of 10 wt% was added in Elazar et al. 455 (2019) compared with only 3 wt% in this study. As a consequence, at 4 GPa, the solidus in 456 Elazar et al. (2019) was found to be between 1000 and 1100°C. At 1200°C, the system in Elazar et al. (2019) is composed of garnet, cpx, rutile, magnesite and kyanite, in equilibrium 457 458 with 70% melt containing 13 wt% H<sub>2</sub>O and 15 wt% CO<sub>2</sub>. In the current study at 1200°C, 459 higher amount of melt was found, containing lower amounts of  $H_2O$  (7 wt%) and  $CO_2$  (2 460 wt%) indicating a lower solidus temperature than in Elazar et al. (2019). The melt in equilibrium with eclogite in this study is metaluminous, similar to the near-solidus melt in 461 462 Elazar et al. (2019).

463 Most previous studies on peridotite $+H_2O+CO_2$  systems focused on lherzolitic composition 464 [Dvir and Kessel (2017) and references therein]. All the studies mentioned in Dvir and Kessel 465 (2017) determined the location of the solidus at 4 GPa, albeit its location varies due to 466 differences in bulk composition. Nevertheless, these studies demonstrate that the system is 467 below its second critical endpoint. Studies on harzburgite+H<sub>2</sub>O+CO<sub>2</sub> are limited so no direct 468 comparison to the current study is available. The harzburgite+H<sub>2</sub>O+CO<sub>2</sub> control experiment in 469 this study is sub-solidus, containing a fluid phase in equilibrium with 58% olivine, 25% opx 470 and 1% garnet. Wyllie (1987) has determined the solidus of such a system to be above 1400°C 471 at 4 GPa, in agreement with our findings of the system to be sub-solidus at 1200°C. A 472 metaluminous fluid is in equilibrium with the harzburgite at these conditions.

473 All three hybrid experiments were performed containing eclogite and harzburgite, 474 juxtaposed one above the other, in the presence of H<sub>2</sub>O and CO<sub>2</sub>. The H<sub>2</sub>O-CO<sub>2</sub>-bearing melt 475 formed in the eclogite control experiment is assumed to interact with the harzburgite in the 476 hybrid experiments. In all scenarios examined in this study, a H<sub>2</sub>O-CO<sub>2</sub>-bearing melt was formed. Cracks, seen in all reaction layers in the hybrid experiments crossing from the 477 478 eclogite to the peridotite (Fig. 3), allow the eclogite-derived hydrous melt to move through the 479 reaction layer into the peridotite layer (i.e., allowing channelized flow) promoting melt-rock 480 interaction. The appearance of cpx in the peridotite layer indicates that some interaction 481 occurs also at the far end of the capsule as well. However, the degree of interaction, shown 482 both by changes in mineral and melt compositions as well as the amount of newly formed 483 minerals, is a function of frequency of rotation, thus degree of interaction.

484 The static interaction between eclogite and harzburgite resulted in the formation of a 485 metaluminous melt, characterized by only slightly higher Mg# and lower Ca# 486 [Ca/(Ca+Mg+Fe)] than the melt in equilibrium with only eclogite (Fig. 7e). Rotation of the multi-anvil led to the formation of a melt intermediate in composition between the fluid in 487 488 equilibrium with harzburgite and the melt in equilibrium with eclogite in the control 489 experiments. Increasing frequency of rotation lowered the SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> contents of the 490 melt while increased the MgO, H<sub>2</sub>O and CO<sub>2</sub> and alkali contents towards the composition of 491 the fluid in the harzburgite control experiment. These changes in chemistry lead to a change 492 in character from metaluminous to peralkaline melt. The more frequent the rotation is, the 493 more peralkaline the melt is (Fig. 8).

494 The initial harzburgite constitutes ol, opx and garnet. In all hybrid experiments the 495 peridotite contains a similar mineral assemblage of ol, opx, cpx, grt. However, the

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496 compositions of the minerals change with increasing interaction between the harzburgite and 497 the eclogite-derived hydrous melt (Figs. 4-6). In the static experiment, the eclogitic garnet is 498 similar in composition to the garnet in the eclogite control experiment. The peridotitic garnet 499 is intermediate in composition between the eclogitic and peridotitic garnets in the control 500 experiments. The Mg# of the peridotitic of and opx is slightly lower than that of the 501 harzburgite control experiment. More pronounced changes in mineral composition are 502 observed in the rotation experiment. Both the eclogitic garnets and the peridotitic garnets 503 approach each other in composition, both in Mg# as well as in CaO and  $Cr_2O_3$  contents. The 504 Mg# values of ol and opx are significantly lower than the values in the harzburgite control 505 experiment. Clinopyroxene, absent in both the initial harzburgite and eclogite, appear in all 506 hybrid experiments. The Mg# of the cpx in the hybrid experiments is also lower and the 507 jadeite component is higher compared to the static experiment.

508 Increasing interaction between the eclogite and the harzburgite at a constant 509 eclogite:harzburgite ratio at a given P-T conditions led to increasing change in mineral 510 chemistry towards an intermediate composition between the eclogite and harzburgite. 511 However, the enhanced interaction did not cause the disappearance of the reaction layer 512 between the two lithologies. Rather, increasing interaction led to increasing thickness of the 513 reaction layer. This observation is in accord with Bulatov et al. (2014) who demonstrated that 514 a reaction layer remains even after long duration of metasomatism, preventing complete 515 equilibration.

516

## Oxygen fugacity during metasomatic reaction

517 The oxygen fugacity registered by the Ir-Fe grains in the harzburgite control experiment is
518 2.0±0.2 log units below QFM, ranging from 1.6 to 2.2 log units below QFM buffer (Fig. 9),

 $\sim 0.6 \log$  units below the enstatite-magnesite-olivine-diamond (EMOD) buffer. The  $\log fO_2$ values are identical within uncertainties throughout the capsule, regardless of the location of the Ir grain, indicating a high degree of equilibrium in this experiment.

522 In all hybrid experiments, the  $log fO_2$  is more reducing than in the harzburgite control 523 experiment and exhibit a gradient from relatively more oxidizing conditions near the reaction 524 layer, towards more reducing conditions further away into the peridotite layer. In both the 525 static and the slow rotating experiment, log/O<sub>2</sub> changes systematically from QFM-3.4 to 526 QFM-4.2. In the fast-rotated experiment (#38), the  $\log O_2$  exhibits similar gradient, albeit the 527  $fO_2$  is ~1 log unit more oxidizing, from QFM-2.3 close to the reaction layer down to QFM-2.9 528 in the far end of the peridotite. The  $\log fO_2$  close to the reaction layer is similar to the  $\log fO_2$  in 529 the harzburgite control experiment.

530 The bulk composition in the harzburgite control experiment contains  $\sim 10 \text{ wt}\% \text{ H}_2\text{O}$  and 531 0.4 wt% CO<sub>2</sub>. The fluid in equilibrium with harzburgite contains  $\sim 63$  wt% H<sub>2</sub>O. Water acts as 532 an efficient oxidizing agent. Kelley and Cottrell (2009) showed that the H<sub>2</sub>O concentrations in 533 the fluid or melt and the oxidation state of Fe linearly correlate. High H<sub>2</sub>O content in the fluid in this experiment suggests high  $Fe^{3+}/\Sigma Fe$  ratio, imposing relatively oxidizing conditions. 534 535 Since the experiment was constantly rotated and the fluid freely traversed throughout the 536 capsule, a homogeneous logfO<sub>2</sub> values are registered in the harzburgite on both sides of the 537 diamond trap.

538 In the hybrid experiments, eclogite and harzburgite were mixed in equal proportions. The 539 bulk H<sub>2</sub>O in the capsule is 7-8 wt% H<sub>2</sub>O and 1.6 wt% CO<sub>2</sub>. The melt in all three experiments 540 contains 15-25 wt% H<sub>2</sub>O and 2-5 wt% CO<sub>2</sub>, lowering the activity of H<sub>2</sub>O in the melt. Lower 541 H<sub>2</sub>O content correlates with lower Fe<sup>3+</sup>/ $\Sigma$ Fe ratio, suggesting a more reducing conditions in

542 these capsules. The melts in these experiments originated in the eclogite layer, travelled and 543 filled the diamond trap and interacted with the harzburgite, forming a reaction zone. The 544 logfO<sub>2</sub> values registered at the edge of the reaction zones reflect the more reducing conditions 545 in these melts compared to the harzburgite control experiment. The decrease in  $\log fO_2$  from 546 the edge of the reaction zone into the peridotite suggests that the reaction zones act as partial 547 barberries for the melt to travel through the peridotite. The logfO<sub>2</sub> profile of experiment #38 548 (rotation every 15 seconds) is at higher values than of experiments #40 and #35 (static and 549 rotation every 15 minutes). This is probably due to the fast rotation in this experiment, 550 allowing better interaction of the melt with the peridotitic layer.

### 551

# Metasomatic interaction of eclogite-derived hydrous melt and harzburgite

The phase abundance in each experiment (Table 8) allows us to quantify the changes occurring as a function of increasing interaction between the eclogite and harzburgite due to the rotation of the multi-anvil (compared to the static experiment) at 4 GPa and 1200°C. The effect of increasing interaction is shown in Fig. 8 and can be summarized in the following reaction:

557 
$$0.7 \text{ ol} + 2.4 \text{ grt}_{ecl} = 1.3 \text{ opx} + 0.7 \text{ cpx} + 0.2 \text{ grt}_{perd} + 1.0 \text{ melt}$$
 (1)

Reaction (1) demonstrates the metasomatizing effect of the eclogite-derived hydrous melt on the harzburgite, by increasing the mode of the peridotitic opx, cpx and garnet (mostly as a reaction layer between the eclogite and harzburgite) on the expense of peridotitic olivine and the eclogitic garnet. Slight increase in melt fraction occurs with increasing interaction.

562 The formation of a reaction layer with sharp boundaries during the interaction of hydrous 563 eclogite-derived melt with harzburgite in this study is in accord with previous studies.

564 However, the minerals composing the reaction layer vary in previous studies as a function of 565 the eclogite and peridotite compositions and the melt/peridotite ratio. (Sekine and Wyllie, 566 1983), (Carroll and Wyllie, 1989) and (Johnston and Wyllie, 1989) performed experiments 567 containing water-saturated silicate melt and peridotite at 1.5-3 GPa and 850-1150°C and 568 observed opx+cpx±grt±phlogopite reaction layers. (Kelemen et al., 1990) conducted 569 experiments containing olivine-tholeiite melt and hydrous harzburgite at 0.5 GPa and 1050-570 1150°C and identified an opx-rich reaction layer. (Rapp et al., 1999) performed experiments 571 containing amphibolitized basalt and harzburgite at 3.8 GPa and 1100-1150°C. The reaction 572 layer in this study consisted of grt±opx. They demonstrated that opx is formed when an 573 eclogite-derived melt infiltrates a depleted peridotite, while cpx is formed when the melt 574 infiltrates a fertile peridotite. In this study, it is demonstrated that increasing reaction between 575 the eclogite-derived hydrous melt with harzburgite at a constant pressure and temperature 576 increases the reaction layer, i.e., increases the amount of opx and cpx formed on the expense 577 of peridotitic olivine and eclogitic garnet. The fraction of melt increases as well.

### 578 Implications to interaction of eclogite-derived melts and harzburgite

579 The eclogite-harzburgite interaction experiments documented here may provide further 580 understanding of the chemical and modal heterogeneity in the mantle and the wide range of 581 melts. This interaction can occur in subduction zones where eclogite-derived hydrous melts 582 infiltrate into harzburgitic mantle adjacent to the subducting slab or in sub continental 583 lithospheric mantle. Static interaction between eclogite and harzburgite simulates a scenario 584 where the eclogite-derived melt rises through the harzburgitic peridotite with little time to 585 interact with the harzburgitic walls. This experiment demonstrates that little interaction occurs. 586 The composition of the melt is similar to the melt derived from melting of volatile-bearing

587 eclogite, a metaluminous andesitic melts. In such a setting, the eclogite-derived hydrous melt is 588 only slightly reactive towards the surrounding peridotite. Close to the boundary between the 589 eclogite and harzburgite, the eclogite-derived hydrous melt consumes the harzburgitic olivine 590 while the mode of opx increases and new cpx grains are formed in a narrow boundary layer. The 591 addition of cpx, homogeneously distributed throughout the peridotitic layer, transforms the 592 harzburgitic peridotite to lherzolitic peridotite. This re-fertilization of the mantle by metasomatic 593 melt is called stealth metasomatism (O'Reilly and Griffin 2013). New phases are formed while 594 slight changes in the composition of the pre-existing minerals occur. Grütter and Menzies (2017) 595 suggested that at low metasome/rock ratio, interaction between Al<sub>2</sub>O<sub>3</sub>-bearing fluid with an Al-596 depleted harzbguite can lead to the formation of new garnet growth. This metasomatic garnet 597 growth can also be manifested as core-to-rim zonation vectors in CaO-Cr<sub>2</sub>O<sub>3</sub> space, which reflect 598 changing garnet Cr/(Cr+Al) in response to changing bulk Cr/(Cr+Al). The compositional vectors 599 for the large dataset of garnets form the Newlands kimberlite (Fig. 1 of Grütter et al. 2017) are 600 similar to that found in the equilibrium assemblage in our reaction experiments (Fig. 6), 601 suggesting that an eclogite-derived fluids or melts can promote such reactions.

Rotation of hybrid eclogite-peridotite system simulates enhanced interaction between the eclogite-derived hydrous melt and the surrounding harzburgite. Increasing interaction between the eclogite-derived hydrous melt and the harzburgite leads to increasing proportion of the peridotitic olivine that is consumed while the proportions of opx and cpx increase, resulting in a thicker reaction zone. The mineral compositions trend towards homogeneity throughout the mixing zone. The reactive melt becomes peralkaline, intermediate in composition between the  $H_2O-CO_2$  eclogitic melt and  $H_2O-CO_2$ -peridotitic fluid.

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609 The interaction between eclogite-derived hydrous melt and harzburgite play an important 610 role in the mineralogical and chemical modification of the subcontinental mantle lithosphere 611 and the mantle wedge, as well as the formation of a wide range of magmas. The above 612 experiments suggest that the degree of interaction between the slab-derived melts with the 613 hosting peridotitic rocks is an important parameter, together with the initial eclogite and 614 peridotite compositions, in forming the wide range of melt compositions, from metaluminus 615 to peralkaline. The degree of interaction also influences the mineral chemistry in the residual 616 metasomatized mantle and the width of the reaction layer between the two lithologies. 617 ACKNOWLEDGEMENT 618 This research was supported by Israel Science Foundation grants (167/14; 760/18). We 619 thank Omri Dvir for his help with the LA-ICP-MS analyses and Yael Kempe for her help with 620 the EPMA analyses. Constructive reviews by Veronique Le Roux and an anonymous reviewer 621 helped to improve this manuscript and are greatly appreciated. 622 **REFERENCES CITED** 623 Allègre, C.J., and Turcotte, D.L. (1986) Implications of a two-component marble-cake mantle. 624 Nature, 323(6084), 123-127. Atherton, M.P., and Tarney, J. (1979) Origin of Granite Batholiths: Geochemical Evidence: 625 Based on a Meeting of the Geochemistry Group of the Mineralogical Society. Springer. 626 627 Bell, D.R., Grégoire, M., Grove, T.L., Chatterjee, N., Carlson, R.W., and Buseck, P.R. (2005) 628 Silica and volatile-element metasomatism of Archean mantle: a xenolith-scale example 629 from the Kaapvaal Craton. Contributions to Mineralogy and Petrology, 150, 251-267. 630 Borghini, G., P, F., and E, P. (2022) Melt-rock interactions in a veined mantle:pyroxenite-631 peridotite reaction experiments at 2 GPa. 34, 109-129. Boyd, F. (1989) Compositional distinction between oceanic and cratonic lithosphere. Earth and 632 633 Planetary Science Letters, 96(1-2), 15-26. 634 Bulatov, V.K., Brey, G.P., Girnis, A.V., Gerdes, A., and Höfer, H.E. (2014) Carbonated sediment-peridotite interaction and melting at 7.5-12GPa. Lithos, 200, 368-385. 635 636 Carroll, M.R., and Wyllie, P.J. (1989) Experimental phase relations in the system tonalite-637 peridotite-H2O at 15 kb; implications for assimilation and differentiation processes near

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	Eclogite (CKB)		Harzburgi	te (DHRZ)
wt%	OE-1	OE-2	OE-3	OE-4
SiO <sub>2</sub>	49.47	49.33	45.15	45.24
TiO <sub>2</sub>	1.63	1.64	-	-
Cr <sub>2</sub> O <sub>3</sub>	-	-	0.75	0.76
Al <sub>2</sub> O <sub>3</sub>	15.10	15.08	0.62	0.63
FeO <sup>b</sup>	9.63	9.68	5.64	6.19
MnO	-		0.09	0.09
CaO	10.86	10.86	0.51	0.52
MgO	7.03	7.10	46.20	45.52
Na <sub>2</sub> O	2.51	2.55	-	-
K <sub>2</sub> O	0.49	0.49	-	-
NiO	-		0.31	0.31
$H_2O$				
$CO_2^{b}$	3.08-	3.11	0.42	-0.46
Total <sup>c</sup>	99.80	99.82	99.74	99.28
Th (ppm)	58	56	-	-
Cs (ppm)	-		35	36
Mg# <sup>c</sup>	0.57	0.57	0.94	0.93
$Ca\#^d$	0.38	0.38	0.01	0.01

TABLE 1. Chemical compositions of starting material

<sup>a</sup> All Fe is expressed as FeO.

<sup>b</sup> The wt% CO<sub>2</sub> was measured via the QTS technique (Dvir and Kessel 2013).

<sup>c</sup> Mg# (molar) = Mg/(Mg+Fe)

<sup>d</sup> Ca# (molar) = Ca/(Ca+Mg+Fe)

Total wt% wt% Rotation  $T_{opx\text{-}cpx}$ duration Run# Capsule# lithology Harz/Ecl powder H<sub>2</sub>O<sup>c</sup> diamond<sup>a</sup>  $(hr)^{d}$  $(^{\circ}C)^{e}$ time  $(mg)^{b}$ EHR17 30 15 min Harzburgite 16 15.00 10 21 -EHR18 41 20 10.29 9 20 Eclogite -15 min EHR19 40 hybrid 0.99 19 13.81 10 24 1179 static EHR14 35 hybrid 1.05 8 1209 16 11.74 15 min 24 EHR16 38 hybrid 1.03 17 11.42 9 15 sec 21 1262

 TABLE 2. Capsule set up and experimental conditions

<sup>a</sup> wt% diamonds – wt% of diamond powder in the system powder +  $H_2O + CO_2$  + diamonds.

<sup>b</sup> wt%  $H_2O - wt$ % of  $H_2O$  powder in the system powder +  $H_2O + CO_2$ .

<sup>c</sup> wt%  $CO_2 - wt$ % of  $CO_2$  powder in the system powder +  $H_2O + CO_2$ .

<sup>d</sup> Rotation time – the time elapsed between rotations.

<sup>e</sup> Equilibrium temperature calculated based on the Mg-Fe exchange between orthopyroxene and clinopyroxene (Putrika 2008).

	Control		Hybrid	
	Harzburgite	static	15 min	15 sec
Capsule#	30	40	35	38
n <sup>a</sup>	28	10	23	10
SiO <sub>2</sub>	41.60(40) <sup>c</sup>	41.18(52)	41.44(40)	41.19(50)
TiO <sub>2</sub>	$\operatorname{Bdl}^d$	0.03(2)	0.07(3)	0.05(3)
$Cr_2O_3$	0.12(9)	0.14(4)	0.10(6)	0.08(2)
$Al_2O_3$	0.03(4)	0.06(5)	0.04(4)	0.02(2)
FeO <sup>b</sup>	5.63(67)	6.61(65)	7.54(28)	7.94(54)
CaO	0.02(1)	0.05(5)	0.04(3)	0.04(2)
MgO	52.31(57)	51.59(1.03)	50.11(44)	50.11(63)
MnO	0.09(2)	0.0(3)	0.07(2)	0.04(2)
NiO	0.30(10)	0.32(13)	0.37(5)	0.40(7)
Na <sub>2</sub> O	bdl	0.01(1)	0.01(1)	0.02(2)
K <sub>2</sub> O	0.01(1)	0.01(1)	bdl	0.01(1)
Total	100.13	100.07	99.79	99.90
Mg# <sup>e</sup>	0.943(7)	0.933(6)	0.922(3)	0.918(6)

TABLE 3. Olivine compositions in this study

<sup>b</sup>All Fe is expressed as FeO

.

<sup>c</sup>Numbers enclosed in parentheses indicate 1 standard deviation of the last digit quoted, i.e., 41.60(40) should be read as 41.60±0.40.

<sup>d</sup>bdl – below detection limit.

 $^{e}Mg\#$  (molar) = Mg/(Mg+Fe)

	Control		Hybrid	
	Harzburgite	static	15 min	15 sec
Capsule#	30	40	35	38
n <sup>a</sup>	18	9	37	24
$SiO_2$	58.48(41) <sup>c</sup>	56.31(1.31)	57.70(57)	57.86(59)
$TiO_2$	$\operatorname{Bdl}^d$	0.06(4)	0.14(3)	0.13(2)
Cr <sub>2</sub> O <sub>3</sub>	0.74(8)	0.49(10)	0.55(16)	0.54(14)
$Al_2O_3$	1.24(34)	1.39(51)	1.53(42)	1.08(15)
FeO <sup>b</sup>	3.69(24)	4.54(29)	4.79(35)	4.60(42)
CaO	0.20(3)	0.32(6)	0.71(37)	0.51(18)
MgO	36.59(33)	36.16(80)	34.73(19)	35.61(69)
MnO	0.08(2)	0.05(2)	0.03(1)	0.03(2)
NiO	0.11(3)	0.19(6)	0.19(5)	0.14(4)
Na <sub>2</sub> O	0.01(1)	0.11(5)	0.15 (4)	0.13(3)
K <sub>2</sub> O	bdl	0.01(1)	0.01(1)	0.01(1)
Total	101.14	99.63	100.53	100.64
Mg# <sup>e</sup>	0.946(4)	0.934(4)	0.928(5)	0.932(6)

TABLE 4. Orthopyroxene compositions in this study

<sup>b</sup>All Fe is expressed as FeO

<sup>c</sup>Numbers enclosed in parentheses indicate 1 standard deviation of the last digit quoted, i.e., 58.48(41) should be read as 58.48±0.41.

<sup>d</sup>bdl – below detection limit.

 $^{e}Mg\#$  (molar) = Mg/(Mg+Fe)

		Hybrid	
Capsule#	static	15 min	15 sec
	40	35	38
n <sup>a</sup>	4	22	18
$SiO_2$	54.85(33) <sup>c</sup>	54.58(56)	54.90(35)
TiO <sub>2</sub>	0.03(2)	0.35(6)	0.26(5)
$Cr_2O_3$	1.42(53)	0.99(60)	0.62(41)
$Al_2O_3$	1.40(27)	3.00(35)	2.10(38)
FeO <sup>b</sup>	2.00(24)	2.55(22)	3.01(31)
CaO	20.02(88)	19.07(58)	18.96(79)
MgO	18.45(1.19)	17.66(52)	18.48(81)
MnO	0.04(5)	0.01(1)	0.02(2)
NiO	0.04(2)	0.06(3)	0.05(3)
Na <sub>2</sub> O	2.15(33)	2.62(33)	1.97(33)
K <sub>2</sub> O	0.04(3)	0.01(1)	0.02(1)
Total	100.44	100.89	100.38
$Mg \#^d$	0.943(4)	0.924(5)	0.916(7)

TABLE 5. Clinopyroxene compositions in this study

<sup>b</sup>All Fe is expressed as FeO

<sup>°</sup>Numbers enclosed in parentheses indicate 1 standard deviation of the last digit quoted, i.e., 54.85(33) should be read as 54.85±0.33.

 $^{d}Mg\#$  (molar) = Mg/(Mg+Fe)

 TABLE 6. Garnet compositions in this study

Control			Hybrid					
Capsule	Capsule Harzburgite Eclogite		40		35		38	
			Harzburgite	Eclogite	Harzburgite	Eclogite	Harzburgite	Eclogite
N <sup>a</sup>	28	10	6	8	5	5	6	8
$SiO_2$	42.19(24) <sup>c</sup>	42.05(48)	43.44(36)	42.05(36)	42.24(80)	40.19(50)	41.83(97)	41.22(14)
$\mathrm{TiO}_2$	bdl	0.56(16)	0.17(19)	0.98(19)	0.87(14)	0.59(30)	0.96(23)	0.69(19)
$Cr_2O_3$	3.19(78)	0.01(1)	0.75(4)	0.05(4)	4.78(54)	0.17(5)	3.04(1.07)	0.23(27)
$Al_2O_3$	22.27(67)	23.80(43)	23.40(44)	23.52(44)	19.63(90)	24.42(61)	19.93(1.22)	22.79(44)
FeO <sup>b</sup>	6.27(18)	13.61(27)	9.28(25)	11.36(25)	6.14(1.02)	8.85(49)	7.23(61)	8.33(25)
CaO	2.28(20)	8.20(39)	4.85(60)	9.77(60)	6.64(98)	6.92(1.03)	6.35(69)	6.34(49)
MgO	23.15(30)	12.67(42)	19.15(42)	13.07(42)	19.67(1.23)	17.64(94)	19.29(1.12)	18.36(30)
MnO	0.31(3)	0.02(2)	N.A. <sup>e</sup>	0.03(2)	0.07(4)	0.07(1)	0.01(2)	0.08(2)
NiO	0.02(2)	0.01(1)	N.A. <sup>e</sup>	0.01(1)	0.03(3)	0.02(2)	0.01(2)	0.01(2)
Na <sub>2</sub> O	0.01(1)	0.18(4)	0.05(14)	0.23(14)	0.18(5)	0.10(1)	0.11(3)	0.07(1)
K <sub>2</sub> O	$\operatorname{Bdl}^d$	0.01(1)	bdl	bdl	0.01(1)	0.01(1)	0.01(2)	0.01(1)
Total	99.69	101.13	101.08	101.09	100.26	99.89	98.78	98.12
$Mg \#^{\rm f}$	0.868(3)	0.624(8)	0.784(5)	0.672(8)	0.850(28)	0.780(18)	0.826(19)	0.797(6)

<sup>b</sup>All Fe is expressed as FeO

<sup>c</sup>Numbers enclosed in parentheses indicate 1 standard deviation of the last digit quoted, i.e., 42.19(24) should be read as 42.19±0.24.

<sup>d</sup>bdl – below detection limit.

<sup>e</sup>N.A. – Not analyzed.

 $^{f}Mg\# (molar) = Mg/(Mg+Fe)$ 

	Con	Hybrid			
	Harzburgite	Eclogite	static	15 min	15 sec
Capsule#	30	41	40	35	38
N <sup>a</sup>	13	14	7	5	6
$SiO_2$	13.41(71) <sup>c</sup>	50.03(2.45)	53.13(91)	27.69 (82)	33.97(1.22)
$TiO_2$	0.011(1)	1.64(13)	1.07(24)	1.64(11)	1.95(20)
$Cr_2O_3$	0.08(2)	bdl	0.02(2)	0.02(1)	0.008(5)
$Al_2O_3$	1.11(16)	12.78(1.95)	8.78(1.13)	8.06(62)	6.20(62)
FeO <sup>b</sup>	3.17(1.21)	6.11(1.33)	3.77(77)	4.83(42)	6.93(96)
CaO	3.34(30)	10.50(38)	8.88(2.66)	5.44(34)	8.45(75)
MgO	13.28(1.28)	5.39(1.00)	7.62(42)	14.58(69)	10.79(1.18)
MnO	0.08(2)	bdl	0.009(0)	0.04(1)	0.03(1)
NiO	0.01(1)	bdl	bdl	0.007(1)	0.012(7)
Na <sub>2</sub> O	bdl <sup>d</sup>	3.98(1.12)	4.72(45)	5.52(14)	5.25(3.18)
K <sub>2</sub> O	bdl	0.46(6)	0.73(21)	1.54(19)	1.24(20)
$H_2O$	62.63(2.67)	6.82(87)	15.52(2.10)	25.20(1.48)	20.42(1.75)
$CO_2$	2.86(12)	2.28(29)	2.76(37)	5.43(32)	4.74(41)
Total	100.00	100.00	100.00	100.00	100.00
Mg# <sup>e</sup>	0.88(3)	0.61(1)	0.78(3)	0.84(18)	0.75(2)
$Ca\#^{f}$	0.11(2)	0.46(5)	0.39(8)	0.18(1)	0.30(5)

<sup>b</sup>All Fe is expressed as FeO

<sup>°</sup>Numbers enclosed in parentheses indicate 1 standard deviation of the last digit quoted, i.e., 42.19(24) should be read as 42.19±0.24.

<sup>d</sup>bdl – below detection limit.

 $^{e}Mg\#$  (molar) = Mg/(Mg+Fe)

fCa# (molar) = Ca/(Ca+Mg+Fe)

	Control			Hybrid		
	Harzburgite	Eclogite	static	15 min	15 sec	
Capsule#	30	41	40	35	38	
Olivine	$0.58(2)^{a}$	_	0.16(2)	0.08(3)	0.11(4)	
opx	0.25(2)	_	0.17(5)	0.31(5)	0.26(6)	
cpx	b	_	0.04(3)	0.13(4)	0.08(6)	
Garnet(ecl)	_	0.24(7)	0.42(7)	0.28(10)	0.27(34)	
Garnet(Hrz)	0.01(1)	_	0.02(8)	0.03(13)	0.03(38)	
Fluid	0.15(2)	_	_	_	—	
Melt	_	0.76(7)	0.19(3)	0.17(2)	0.25(9)	

 TABLE 8. Modal phase abundance

<sup>a</sup>Numbers enclosed in parentheses indicate 1 standard deviation of the last digit quoted, i.e., 0.58(2) should be read as  $0.58\pm0.2$ .

<sup>b</sup> – indicates absent from the run product assemblage.















Fig. 5













