1   Revision 2

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3   Chemical fingerprints and residence times of olivine in the 1959 Kilauea Iki eruption, Hawaii:

4   insights into picrite formation

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Abstract

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25   Olivine-rich mafic volcanic rocks (picrites) are a common and important part of ocean

26   island and flood basalt volcanism. Despite their primitive bulk compositions (high MgO, FeO,

27   Mg# and low SiO_2), olivine-rich magmas are typically interpreted as the result of addition of

28   olivine from cumulate zones into more evolved basaltic liquids (MgO $\leq$ ~8 wt%). There are

29   commonly two texturally distinct olivine populations in picrites: type 1 grains with planar

30   dislocation (kink) bands, subgrains or undulose extinction; and type 2 grains that lack these

31   optical textures. Type 1 olivine is similar in texture to olivine from tectonized ultramafic rocks,

32   suggesting that these textures result from plastic deformation, likely within cumulate zones.

33   However, recently it has been proposed that type 1 olivine could also result from growth

34   phenomena or crystal-crystal collisions. In the Kilauea Iki picrite samples used in this study, type

35   1 grains make up only 10–20% of the modal olivine, however they make up 30–65% of the total

36   olivine by volume due to their large size. Therefore, type 1 grains make a large contribution to

37   the overall composition of Kilauea Iki picrites. A combination of textural (optical defects, crystal

38   size distributions and minor element zoning) and geochemical analyses (trace element

39   concentrations, and diffusion of minor elements) suggests that type 1 and type 2 olivine grains

40   have experienced distinctly different petrological histories and that they are antecrysts and

41   autocrysts, respectively. Differences between type 1 and type 2 olivine are evident in the

42   abundances of slow diffusing trace elements (Al, P, Ti, V), which are likely inherited from their

43   distinct parent magmas. Type 1 and type 2 grains also define different slopes in crystal size

44   distributions, and constraints from diffusion of P and Cr suggest that type 1 grains have longer

45   magmatic residence times than type 2 grains. Type 1 grains likely derive from deformed

46   cumulates within the plumbing system of Kilauea volcano, and our work supports the hypothesis

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47 that picrites from Kilauea Iki are formed by accumulation of antecrystic olivine in more evolved

48 basaltic liquid. Our work further supports models that type 1 olivine textures are formed during

49 plastic deformation within cumulate zones and are not the result of growth phenomena. Our

50 methods can be applied to other olivine-rich volcanic rocks to test the cumulate model for the

51 formation of type 1 olivine textures, which are relatively common in picritic and related rocks

52 from other settings.

53 **Keywords:** Kilauea Iki, olivine, picrite, diffusion, crystal size distribution, olivine

54 cumulate

55 **Introduction**

56 The compositions of mafic volcanic rocks from all tectonic settings provide key

57 information for understanding the origin and evolution of the Earth's mantle, and place important

58 constraints on the conditions and processes of mantle melting and the nature of mantle sources

59 (e.g., White 1985; Zindler and Hart 1986; Anderson and Brown 1993; Graham et al. 1998;

60 Révillon et al. 1999; Class and Goldstein 2005; Herzberg et al. 2007; Putirka et al. 2007; White

61 2010; Herzberg 2011; Poland et al. 2014). However, as most mafic magmas also experience

62 modification during transit and storage within the crust, the processes that work to alter the

63 magma composition during transport and crustal residence must also be understood and

64 corrected for where necessary (e.g., Herzberg 2011). In many cases the most mafic (high MgO,

65 Mg#, Ni) rocks in a given suite of lavas are selected as the magma compositions that are the least

66 modified from those in equilibrium with their mantle source. However, many volcanic rocks that

67 meet these primitive chemical characteristics are olivine-rich (e.g., picrites), and although some

68 olivine-rich, picritic rocks are interpreted to represent primary melts from the mantle whose

69 crystallized olivine remains entrained in the host liquid (Francis 1985), most picrites likely result

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70 from the addition of olivine to a more evolved basaltic liquid (e.g., Murata and Richter 1966b;

71 Helz 1987; Albarede and Tamagnan 1988; Clague and Denlinger 1994; Garcia 1996; Révillon et

72 al. 1999; Thomson and Maclennan 2013). In this case much of the olivine present within a picrite

73 did not directly crystallize from the silicate liquid in which they are erupted, but are derived from

74 crustal cumulate zones or other sources (Francis 1985; Albarede and Tamagnan 1988; Clague

75 and Denlinger 1994; Baker et al. 1996; Garcia 1996; Révillon et al. 1999; Sakyi et al. 2012).

76   In detail, olivine accumulated within picritic magmas may be from a number of different

77 sources. These include *autocrysts* (following the nomenclature of Miller et al. 2007) that are

78 derived directly from the liquid fraction of the magma, *xenocrysts* from conduit walls at deep

79 crustal or lithospheric mantle levels, or *antecrysts* derived from disruption of cumulates and

80 other olivine-rich zones within the volcano's plumbing system (e.g., Helz 1987). At Hawaii, and

81 many other ocean island and flood basalt provinces, olivine-rich lavas contain a significant

82 fraction of "deformed" (type 1) olivine grains that exhibit characteristic multiple planar

83 extinction boundaries (kink bands), undulose extinction, or subgrains (Francis 1985; Helz 1987;

84 Albarede and Tamagnan 1988; Garcia 1996; Révillon et al. 1999; Larsen and Pedersen 2000;

85 Natland 2003; Sakyi et al. 2012; Thomson and Maclennan 2013; Welsch et al. 2013; 2014). The

86 traditional interpretation of this texture is that olivine grains are deformed at high temperatures

87 (>1100 °C) while they are stored in dunite cumulate zones (Francis 1985; Albarede and

88 Tamagnan 1988; Clague and Denlinger 1994; Baker et al. 1996; Garcia 1996; Révillon et al.

89 1999; Sakyi et al. 2012), analogous to deformation of olivine during shearing in the upper mantle

90 (e.g., Hansen et al. 2014). At Hawaii, this deformation is thought to be due to both the seaward

91 motion of the south flank of Kilauea and down-rift flow of olivine-rich cumulates (Clague and

92 Denlinger 1994; Clague et al. 1995; Denlinger and Morgan 2014). Type 1 grains derived from

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93   cumulates are thus interpreted as antecrysts, while type 2 grains are considered to be autocrysts

94   that crystallize directly from the host liquid (Francis 1985; Albarede and Tamagnan 1988;

95   Clague and Denlinger 1994; Baker et al. 1996; Garcia 1996; Révillon et al. 1999; Sakyi et al.

96   2012).

97   However, the interpretation that type 1 olivine derives from cumulate zones has also been

98   challenged. Natland (2003) interpreted type 1 olivine textures in picrites from Juan Fernandez as

99   the result of crystal-crystal and crystal-conduit wall collisions that occur during magma mixing.

100   Alternatively, Welsch et al. (2013; 2014) concluded that type 1 olivine grains from Piton de la

101   Fournaise and Hawaii form as a result of initial rapid, dendritic growth of crystals followed by a

102   period of slower growth and infilling (ripening). This could produce the type 1 textural features

103   due to lattice mismatches developed during the dendritic growth phase and accentuated during

104   the ripening phase (Welsch et al. 2013; 2014). Although these alternative models do not exclude

105   the storage of type 1 olivine in cumulate zones, with these alternate models, storage and

106   deformation in a cumulate is not required to form the observed defects. Thus, a correct

107   interpretation of the origin of these olivine textures has real implications for the petrogenesis of

108   picrite magmas.

109   Resolving this question requires a means to fingerprint olivine grains in picrites that are

110   derived from different sources to understand how they are formed and are incorporated into their

111   host magma. This has proven difficult because the olivine grains are likely derived from

112   crystallization of similar magmas and both type 1 and type 2 olivine grains typically overlap

113   substantially in major element composition (e.g., Baker et al. 1996; Garcia 1996; Révillon et al.

114   1999; Welsch et al. 2013). The divalent major elements and most abundant trace elements in

115   olivine (e.g., Fe, Mg, Ni) also diffuse rapidly in olivine, thus they are readily re-equilibrated

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116   during storage at high temperatures (Chakraborty 2010), which could also explain their

117   similarities. Olivine in mafic magmas commonly have geochemically homogeneous cores (not in

118   equilibrium with the host liquid) with narrow zoned rims, suggesting equilibration during

119   extended crystal residence followed by late-stage growth and/or diffusive re-equilibration in the

120   final, carrier magma (e.g., Helz 1987; Garcia 1996; Révillon et al. 1999; Salaün et al. 2010;

121   Thomson and Maclennan 2013). In contrast to rapidly diffusing major elements, slow diffusing

122   trace elements have been shown to record differences between separate olivine populations (e.g.,

123   Neave et al., 2018), and assuming equilibrium partitioning during growth, have the potential to

124   distinguish the petrogenesis of type 1 and type 2 olivine.

125   In this contribution, we utilize both geochemical (olivine trace element concentrations

126   and distributions) and textural information (deformation textures, zoning patterns and crystal size

127   distributions), to explore the sources of olivine in picrites from Kilauea Iki, Hawaii, and test the

128   validity of using textures alone to fingerprint olivine in mafic volcanic rocks. Differences in

129   crystal size distributions (CSD), trace element compositions and constraints from trace element

130   diffusion suggest that the type 1 olivine population is derived from a distinctly different source

131   and has a longer magmatic residence time than type 2 olivine.

132   **Geologic Setting**

133   Kilauea Iki crater is located east of the Kilauea Caldera on the Big Island of Hawaii. The

134   summit eruption of Kilauea Iki lasted from November 14 through December 20, 1959, and

135   consisted of 17 phases of fire fountaining, that partially filled the Kilauea Iki crater (Richter et al.

136   1970). The 1959 eruption is unique for Kilauea summit eruptions in that it produced high

137   fountains, involved abundant picritic lavas, and erupted lavas contained significant amounts of

138   type 1 olivine (Helz 1987; Garcia et al. 2003). The modal abundance of olivine in the erupted

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139   lavas is also correlated with the eruption intensity (high fountaining events), which led to the

140   interpretation that the picritic nature of the lavas is due to the incorporation of olivine crystals

141   from deeper in the system into a volatile rich magma (Murata and Richter 1966a; 1966b; Helz

142   1987). The magma erupted at Kilauea Iki in 1959 is thought to have formed from mixing of a

143   primitive, mantle-derived magma (1959E) with a more evolved, shallowly-stored magma

144   (1959W; Wright 1973; Helz 1987).

145   Olivine is the dominant phenocryst phase (~4 to 30 modal%) present in Kilauea Iki lavas,

146   with minor plagioclase and clinopyroxene present only in the first and the last eruptive phases

147   (Richter and Murata 1966). In general, olivine core compositions are not in Fe/Mg equilibrium

148   with the dominant melts from this eruption (Helz et al. 2014). On the basis of textures, Helz

149   (1987) identified five populations of olivine in Kilauea Iki lavas: (1) blocky grains that contain

150   deformation features (kink-bands or subgrains), (2) euhedral to skeletal grains that are free of

151   deformation features, (3) rounded grains without deformation features, (4) rare, small conchoidal

152   fragments of grains, and (5) rare grains containing abundant sulfide inclusions. Helz (1987)

153   concluded that the class 1 (type 1, herein) olivine are antecrysts derived from the conduit wall in

154   the lower crust, class 2 grains are sourced from the shallow stored magma, and class 5 grains

155   were sourced from deep (but not as deep as class 1) in the plumbing system. Class 3 grains were

156   rounded and class 4 grains were fractured during mixing of the two thermally disparate magmas

157   (Helz 1987). Helz (1987) concluded that both the class 1 grains, and the sulfide-bearing grains

158   were derived from cumulates.

159   **Samples and Methods**

160   Five samples that were collected during the 1959 eruption of Kilauea Iki by USGS

161   scientists were obtained from the collection of the Department of Mineral Sciences, National

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162 Museum of Natural History for this study. These samples were all collected from the rising lava

163 lake during the ongoing eruption and do not represent pumice samples. One additional scoria

164 sample was collected from the surface along the Devastation Trail in 2009. All samples were

165 prepared as polished thin sections.

166   Electron microprobe analyses of olivine were performed on a Cameca SX-100 at Oregon

167 State University. X-ray element maps were performed on selected olivine grains to map the

168 distribution of P, Al, and Cr in olivine following the techniques outlined by Milman-Barris et al.

169 (2008). This was accomplished using a focused ~1 μm beam diameter, with an accelerating

170 voltage of 15 keV and a beam current of 400 nA. Dwell times for each pixel were between 75

171 and 200 ms and the spacing between pixels was between 2 and 5 μm. Images were exported as

172 16 bit TIFF files to preserve the x-ray intensities. To obtain higher resolution images and

173 increase the signal to noise ratio, while keeping analysis time within reasonable limits, elements

174 were analyzed on multiple spectrometers, and images were combined later using MATLAB and

175 ImageJ (Schneider et al. 2012). Aluminum was analyzed on two spectrometers (one TAP and

176 one LTAP crystal). Phosphorus was analyzed on either three spectrometers (one LPET and two

177 PET crystals), or on one spectrometer with the LPET crystal, with Cr analyzed on the other two

178 spectrometers with PET crystals. Analysis times were between 2 and 17 hours depending on the

179 size of the area of interest and the dwell time and pixel spacing.

180   Trace elements were analyzed by laser ablation using a Photon Machines Analyte G2 193

181 nm ArF excimer laser coupled with a Thermo Scientific Xseries2 Quadrupole inductively

182 coupled plasma mass spectrometer (ICP-MS). Olivine was analyzed using an 85 μm diameter

183 spot, at a pulse rate of 10 hz. Sample traverses were done as rim-to-rim line scans, with a scan

184 speed of 15 μm/s. Care was taken to avoid obvious melt and spinel inclusions; however, some

185   contamination is evident in several analyses and the affected portions of those transects were

186   removed during post analysis processing. Reference glass standard GSE-1G (analyzed every 6-8

187   unknowns) was used for calibration, natural glass standards BHVO-2G and BCR-2G as

188   secondary standards, and ^{29}Si was used as an internal standard. Data were processed using

189   standard calibration techniques (Longerich et al. 1996) to calculate concentrations for individual

190   time slices. Each data point is an average of five time slices, which is similar to the method of

191   Ruprecht and Plank (2013). Each data point represents an analytical area of approximately 85 by

192   120 μm and an analysis depth of ~5 μm. Uncertainties for each analysis were calculated by

193   propagating uncertainties in repeat measurements of the calibration standard in each session with

194   uncertainty in the measured normalized ratio for each set of time slices. Typical uncertainties for

195   these analyses are (1s): <5% for Mg, Al, Fe; <10% for Na, Ca, Sc, V, Mn, Co, Ni, Zn; <20% for

196   Li, Ti, Cr, Cu, Ga, Y, Zr, Mo, Ba, Ce, Dy, Er, Yb; <30% for P, Ge, and <40% for B. The

197   forsterite (Fo) content of analyzed olivine grains was estimated from the laser ablation data using

198   measured Fe/Si ratios. The estimated error in Fo is ± 0.95 mol% (RMSE) based on comparison

199   with electron microprobe analyses on the same locations on select grains. Laser ablation analyses

200   were also compared to the electron microprobe for Hawaiian and Snake River Plain olivine

201   (Bradshaw 2012) to assess the accuracy of this technique. The percent difference in composition

202   of select elements between the two techniques were calculated, and they agree well, but with

203   some systematic bias. The laser ablation results are higher for Mg (5%), Mn (30%), Fe (20%)

204   and Ni (10%) and lower for Ca (35%).

205   Electron backscatter diffraction (EBSD) analysis was done in the Linus Pauling Science

206   Center at Oregon State University using an FEI Quanta dual beam scanning electron microscope

207   equipped with an EDAX EBSD detector. Analyses were done using an accelerating voltage of 15

208   kV, current of 0.85 or 1.7 nA (adjusted to reduce charging of the sample). Samples were tilted to

209   an angle of 70° with a typical working distance between 12 and 14 mm. Samples were lightly

210   carbon coated to reduce sample charging.

211   Crystal size distributions were measured by outlining individual crystals in Adobe

212   Illustrator using high resolution scans of thin sections and verified by visual inspection using a

213   petrographic microscope. Type 1 and type 2 grains were outlined in different layers to allow a

214   comparison of their size characteristics. Individual crystals in glomerocrysts were measured

215   separately by visual inspection using a petrographic microscope. We measured 120 to 230

216   olivine grains in each of the five thin sections (Table 1). Outlined crystal intersections were

217   imported into ImageJ (Schneider et al. 2012) to calculate the area, orientation, length and width

218   of a best-fit ellipse. CSDslice (Morgan and Jerram 2006) was used to estimate the overall crystal

219   shape (short:intermediate:long dimensions) based on the 2D intersection data. There were too

220   few crystals in the individual samples to produce robust CSDs, so the combined CSD for all

221   samples was used. The best fit shape parameter from all crystals of 1.0:1.0:1.4 was calculated

222   using CSDslice (Morgan and Jerram 2006). These data were input into CSD Corrections 1.4

223   (Higgins 2000) to correct the 2D intersection data to 3D crystal size distributions on a vesicle-

224   free basis. For input into CSD Corrections, we specified the rock fabric as massive and estimated

225   the roundness to be 0.5, the same value estimated by Vinet and Higgins (2010; 2011) for Kilauea

226   olivine, including grains from the Kilauea Iki lava lake. As we focus on the pre-eruptive storage

227   conditions of these lavas, only grains larger than about 0.1 mm were outlined, and our

228   interpretations are based on grains larger than ~0.2 mm.

229   **Results and Discussion**

230   **Olivine Textures**

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231   We have identified the same five textural classes identified by Helz (1987) in our sample

232   set, but we focus on the two major types, type 1 (class 1 of Helz) and type 2 grains (classes 2 and

233   3 of Helz). Due to analytical reasons (small size and abundant inclusions), and their rarity, class

234   4 and 5 grains are not included in this study. Type 1 grains are typically blocky and commonly

235   contain embayments, which we interpret as evidence of resorption (Figure 1), which is most

236   evident along the boundaries between kink bands. Type 1 grains are present in all samples, with

237   a modal proportion of 12 to 22% of the total olivine crystals (Table 1). Although there are fewer

238   type 1 crystals in our samples, they are larger than type 2 crystals and represent ~30 to 65% of

239   the crystal content by volume (Table 1), and therefore have a large effect on the bulk

240   composition of picrites. Type 2 grains (Helz 1987) are typically euhedral and are the most

241   common crystals in these samples (Table 1).

242   Type 1 and type 2 crystals were identified optically, which due to crystal size and also

243   sectioning effects may result in misidentification of type 1 grains. Vinet et al. (2011) used in situ

244   x-ray diffraction to study the deformation of single olivine crystals from Mauna Ulu and Kilauea

245   Iki, Hawaii, and were able to identify type 1 grains as small as 0.15 mm. In the optical methods

246   applied here, we did not identify type 1 grains smaller than 0.28 mm, therefore some small, type

247   1 grains may have been misidentified. Type 1 grains can also be misidentified due to their

248   orientation as optical defects are not visible in olivine grains whose a-axes are parallel to the

249   plane of the thin section (Boudier 1991). Type 1 olivine textures were successfully identified

250   optically in grains with their a-axes oriented as close as 5 degrees from the plane of the section,

251   as determined by EBSD. Therefore, assuming all crystals are randomly oriented (verified by

252   EBSD analysis; Table 2), only about 6% of type 1 grains would be misidentified due to their

253   orientation within the plane of the thin section. In the work of Vinet et al. (2011) only ~6% of

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254   type 2 grains identified optically (3 out of 54 total grains) were determined to be type 1 grains

255   using in situ x-ray diffraction, which may suggest that those grains were also misidentified

256   optically due to their orientation.

257   **Trace Element Variations in Olivine**

258   Intra-grain trace element variations are evident in LA-ICP-MS transects across both type

259   1 and type 2 olivine and also in X-ray element maps of minor elements (P, Al and Cr; Figures 2,

260   3; Supplemental Material). These within-grain variations in both olivine types are most evident

261   in P (c.f., Milman-Barris et al. 2008; Welsch et al. 2013; 2014; Xing et al., 2017), but minor

262   zoning in Al, Ti, V, Cr, Cu, Sr, Y, and Zr is also present in some grains. Zoning of P is common

263   in both type 1 and type 2 olivine and is also observed in most P X-ray element maps (Table 3,

264   Figures 2, 3). The variation in P and equivalent zoning in Al and Cr, as observed in element

265   maps, is likely the result of kinetic effects during rapid crystal growth (Milman-Barris et al.

266   2008; Grant and Kohn 2013; Welsch et al. 2013; 2014).

267   To compare the compositions of type 1 and type 2 grains, regions of transects with

268   elevated P contents (compared to the remainder of the transect) were removed to eliminate any

269   kinetic effects from analysis of rapid growth zones (Milman-Barris et al. 2008; Grant and Kohn

270   2013; Welsch et al. 2013; 2014). To accomplish this, the laser ablation transects were compared

271   to their associated P maps to identify and remove the locations along the transects affected by

272   high P zones (Figures 2, 3). For grains without P maps only the portions of the profiles with the

273   lowest P in grains with clear P zonation were included in this analysis. By means of comparison

274   we also performed the comparison between the two olivine types without removing zones of

275   elevated P.

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276   When comparing type 1 and type 2 grains, data are plotted as cumulative distribution

277   functions (CDF), which can show differences, or similarities between populations even where

278   there is considerable overlap in the data (Figure 4). Two-sample Kolmogorov-Smirnov tests

279   (which quantifies the difference between two CDFs, Press et al. 1992) were performed to assess

280   whether type 1 and type 2 grains are sampled from the same population (Table 4), using a

281   significance level of 95% ($\alpha= 0.05$). To avoid overweighting of larger crystals (which contain

282   more individual analyses) the Kolmogorov-Smirnov tests were performed using the median

283   concentrations of the laser transects for each grain (both including and excluding high P zones).

284   Type 1 and type 2 grains, from Hawaii and elsewhere, have previously been recognized

285   as indistinguishable in major element compositions (e.g., Helz 1987; Baker et al. 1996; Garcia

286   1996; Welsch et al. 2013). This is also evident here as the p-value for the Kolmogorov-Smirnov

287   test is high for Fo ($p = 0.56$), and other major and minor elements ($p > 0.26$ for Mg, Ca, and Ni)

288   in Kilauea Iki olivine (Table 4), meaning that the null hypothesis that the major elements from

289   type 1 and type 2 grains sample the same population cannot be rejected at our selected level of

290   confidence (95%). However, we can reject this null hypothesis for Al, P, Ti and V ($p < 0.05$;

291   Table 4) in type 1 and type 2 grains, suggesting these two types of olivine contain distinctly

292   different abundances of these elements and thus sample different populations. When high P

293   zones are not removed, the same slow diffusing elements (Al, P, Ti and V) still have

294   significantly low p-values ($p < 0.01$; Table 4).

295   This apparent discrepancy between Fe, Mg, Ca, and Ni (and other divalent cations)

296   compared to Al, P, Ti, and V, where the former sample a single population and the latter two

297   different populations can be best explained by the different diffusion rates among these elements.

298   The major elements and many divalent trace elements (Mg, Mn, Fe, Co, Ni) in olivine have

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299 relatively rapid diffusion rates (> 10^{-16} m^2/s at 1300 °C; Chakraborty 2010; Cherniak and Liang

300 2014; Watson et al. 2015); therefore their original compositions will be modified by diffusive re-

301 equilibration more readily than slower diffusing elements such as Al, Cr, P and V, which are

302 more likely to preserve their original compositional information.

303   The differences evident in chemical composition in the slow diffusing trace elements also

304 suggest that these two textural populations of olivine derive from different composition magmas,

305 and/or formed under pressures and temperature conditions sufficient to affect element

306 partitioning into olivine. However, it is also difficult to relate these differences directly to

307 magmatic compositions. Higher overall Al and Ti in type 1 olivine (Figure 4) might suggest that

308 type 1 olivine formed from melts more enriched in incompatible elements, but P contents in type

309 1 crystals are lower. In addition, there has also likely been some intra-grain diffusive

310 redistribution of these slow diffusing elements, as evidenced by the relaxation or elimination of

311 Al zoning (see below). Within-grain diffusion acted to remove elemental zoning produced during

312 crystal formation (e.g. Millman-Barris et al., 2008), but if this occurred while the grains were in

313 equilibrium with their original liquids, or within a cumulate pile (Thomson and Maclennan,

314 2011) this can still maintain overall composition differences. We also note that some other slow

315 diffusing elements (e.g. Y, REEs) do not appear to sample separate populations. Although this

316 might reflect the possibility that no differences exist between these two olivine populations with

317 respect to these elements, we note that these are also elements present at lower abundance in

318 olivine, and thus also have higher analytical uncertainties, which makes resolving different

319 populations more difficult (Figure 5).

320   Although both olivine textural types overlap completely in major element composition,

321 Helz (1987) reports various patterns of zoning in Fo content in Kilauea Iki olivine, with olivine

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322   from classes (types) 1 and 2 exhibiting both normal (core compositions > Fo_{86}) and reverse

323   zoning (cores < Fo_{86}). This was attributed to olivine being derived from different depths beneath

324   the summit and varying degrees of equilibration (Helz, 1987). We observe a slightly larger range

325   in Fo content than Helz (1987; Fo_{85-88}), with 6 grains having a median Fo content < 84 mole %

326   (three from each textural type), and a total range from Fo_{82-89}, and note the presence of some

327   reversely zoned crystals. However, in our samples there are no obvious Fo-delineated

328   populations, and the trace element abundances are similar between high (> 86%) and low Fo (<

329   86%) olivine. What does emerge from our data is the trace element differences between type 1

330   and type 2 olivine, suggesting that these populations had distinct histories regardless of apparent

331   storage depth.

332   **Crystal Size Distributions**

333   Crystal size distributions of Kilauea Iki olivine grains were determined to further

334   understand the processes that control the formation of these two olivine populations. The CSD

335   data are shown in Figure 6, where type 1 and type 2 grains are plotted separately. There are too

336   few grains from individual samples to plot robust CSDs, so we focus on the overall results from

337   all samples combined. Vinet and Higgins (2011) also performed CSD analyses for Kilauea Iki,

338   mainly focusing on lava lake samples, but they did analyze three scoria samples collected from

339   the surface. They identified two main olivine populations (based on two linear segments of

340   curved CSDs), a smaller crystal population (G2a), consisting of mostly type 2 olivine, and a

341   larger crystal population (G2b), with a significant proportion of type 1 grains (Vinet and Higgins

342   2011). Mangan (1990) measured CSDs for type 2 grains in 8 of the eruptive phases of the 1959

343   eruption and combined them to improve precision. Vinet and Higgins (2011) recalculated the

344   original crystal size measurements of Mangan (1990) using the shape parameters from olivine

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345 from the lava lake and area to volume estimates from CSDCorrections (Higgins 2000). In this

346 study, in order to examine the differences between type 1 and type 2 crystals in more detail we

347 explicitly separate them to investigate the contribution of each olivine texture to the overall CSD.

348   The type 1-only and type 2-only curves in Figure 6 are both approximately linear, with

349 the type 1-only curve having a shallower slope and extending to larger crystals than the type 2-

350 only curve. The type 1-only curve does not extend to the smallest crystal sizes, which could be

351 the result of physical processes during crystal growth and storage that cause differences in CSD

352 shape (Higgins 2006a) but could also be the result of misidentification, as small, type 1 grains

353 are more difficult to identify optically. Although this type of misidentification would also have

354 an effect on the slope of the CSD, we note that along with the evidence presented above, there is

355 not a substantial downturn of the type 1 CSD at the smallest crystal sizes, suggesting that the

356 misidentification of small, type 1 grains is minimal. Any type 1 grains that were misidentified

357 from being oriented with the a-axis parallel to the section should affect both the type 1 and type 2

358 CSDs equally as these grains should be misidentified equally across all grain sizes because the

359 crystals are randomly oriented (Table 2). Therefore, the effect of misidentification due to crystal

360 orientation would only shift the position of the curves, with no change in slope. Our CSD results

361 are similar to the previous studies of Mangan (1990) and Vinet and Higgins (2011). The type 1

362 CSD slope and intercept are similar to the G2b population of Vinet and Higgins (2011), which

363 contains significant type 1 crystals (Table 5). The type 2 crystals are similar to the G2a

364 population of Vinet and Higgins (2011) and the type 1-only CSD of Mangan (1990) (Table 5).

365 As with previous studies (Helz 1987; Garcia 1996; Vinet and Higgins 2010; Vinet et al. 2011;

366 Vinet and Higgins 2011; Sakyi et al. 2012), we also find that type 1 grains extend to larger

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367   crystal sizes than type 2 grains. The largest type 1 grain identified in our samples is 5.56 mm,

368   and the largest type 2 grain is 1.95 mm (long axis).

369   The differences in the shapes of these CSD curves also suggest that there are significant

370   differences in the crystallization and storage histories of these two different textural crystal

371   populations. The shallow slope of the type 1-grain CSD curve (Figure 6; Table 5), could be the

372   result of extended growth (residence) time of type 1 olivine, higher growth rates, textural

373   coarsening, accumulation of olivine or a combination of these (Higgins 2006a). Faster growth

374   rates for the type 1 olivine population could explain the shallower slope and larger crystals

375   compared with the type 2 olivine population. In the dendritic growth model of Welsch et al.

376   (2013) it would be expected that crystals with P zoning would grow large due to a rapid initial

377   growth phase. However, this is at odds with the observation that both type 1 and type 2 olivine

378   are zoned in P, but only the type 1 grains are large, which argues against a single process

379   producing both olivine textures, unless the formation of defects require even more rapid growth.

380   If P-enrichment occurs in a similar way regardless of growth rate – above a certain threshold

381   growth rate – then defects could form in *very* rapidly grown olivine and could produce larger

382   type 1 grains with similar zoning patterns to type 2 grains; however, this is unlikely because as

383   we show below, zoning of P, Al, and Cr should be affected by growth rates, therefore we would

384   expect to see a difference in the concentrations of these elements and their zoning patterns. An

385   alternative explanation for the difference in crystal size between these two olivine populations is

386   textural coarsening. Vinet and Higgins (2010; 2011) also observed a shallower slope for several

387   CSDs that included type 1 grains in Mauna Ulu and Kilauea Iki lava lake samples, which they

388   interpreted as the result of textural coarsening. Textural coarsening (Ostwald Ripening) occurs

389   when larger grains grow at the expense of smaller grains (Marsh 1988; Higgins 2006a; 2011),

390 and is a process that could readily occur during residence within a cumulate zone (Vinet and

391 Higgins 2010; Higgins 2011). The steeper slope and lack of a downturn at the smallest sizes for

392 the type 2 crystals suggest that such coarsening did not affect this population. Although we agree

393 with Vinet and Higgins (2010; 2011) that coarsening has affected the crystals in the Kilauea Iki

394 samples, we suggest it only affected the type 1 grains in the surface samples. The recognition of

395 textural coarsening of type 1 grains potentially implies longer residence than type 2 grains, as

396 additional time would be needed for the type 1 grains to coarsen, assuming the two populations

397 had similar initial crystallization histories (as evidenced by P zoning). Several type 1 grains also

398 have resorption textures, which suggests that they experienced disequilibrium conditions that

399 resulted in some reduction in size prior to eruption. However, this minor amount of resorption

400 could occur late in the development of the crystals, after significant coarsening, resulting in the

401 overall large sizes of type 1 crystals.

402   The Kilauea Iki eruption is well known to be a product of magma mixing (Wright 1973;

403 Helz 1987; Garcia et al. 2003; Helz et al. 2014; Poland et al. 2014; Wright and Klein 2014),

404 which may also have affected the shape of the CSD. Mixing of two magmas with different CSD

405 slopes and intercepts will produce a kinked CSD with two linear segments, representing the

406 slopes of the CSDs for the original magmas (e.g., Higgins 2006a; Salisbury et al. 2008; Kent et

407 al. 2010). The total crystal CSD (both type 1 and type 2 grains) for all samples (Figure 6) has

408 two nearly linear segments (also observed by Vinet and Higgins, 2011) that resembles a mixing

409 situation; however, the larger crystal size population is made up mostly of type 1 grains.

410 Therefore, the curvature of the total CSD is primarily the result of incorporation of type 1

411 olivine. Although two distinct segments are not evident in the single population CSDs, the type 2

412 olivine population exhibits a slight curve, as indicated by the lower value of the goodness of fit

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413 (Q) parameter (Table 5; Higgins 2006b). This may indicate that more subtle effects of magma

414 mixing are present, possibly representing the mixing of the 1959E and 1959W endmembers of

415 Helz (1987), which mixed two similar (but not identical) populations of type 2 grains.

416 **Minor element zoning**

417   The Kilauea Iki samples used in this study have many grains with zoning in the minor

418 elements P, Al and Cr (Figures 2, 3, Supplemenal Material), as has been observed in olivine from

419 Hawaii and a number of other systems (e.g., Milman-Barris et al. 2008; Sakyi et al. 2012;

420 Welsch et al. 2013; 2014). Zoning of these elements in Kilauea Iki olivine is evident in electron

421 microprobe x-ray maps (Figures 2, 3). Most of the grains imaged by this technique show zoning

422 in P, and several of those grains also show corresponding zoning in Al and Cr (Table 3), which

423 has also been observed in natural and experimentally grown olivine (e.g., Milman-Barris et al.

424 2008). The zoning in these elements is mostly independent of any zoning in major elements (Fo)

425 and in many cases is not obviously related to the final crystal shape or to the presence of planar

426 defects (Figure 3). Crystals commonly have hopper-shaped, high-P cores that can be either sharp

427 or diffuse. Most crystals, whether or not they are zoned in the cores, have one or more thin, sharp

428 zones at or near the rim that match the final shape of the crystals, suggesting they are late-stage

429 growth zones. One hopper-shaped groundmass crystal exhibits zoning that matches the crystal

430 shape, and one highly resorbed grain has oscillatory zoning that is disrupted by dissolution

431 surfaces (Figure 2). Zoning in P is generally not disrupted by crystal edges, however, there are a

432 few examples of zoning that is truncated by dissolution surfaces (~11/66 grains; Figure 7).

433 Identifying truncated P zones is somewhat ambiguous because P zonation is typically

434 discontinuous within crystals (see images herein and in Milman-Barris, 2008; Welsch et al.,

435 2013; 2014). Also, in the cases where P zoning is truncated, there is typically a narrow, low-P

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436 area between the high P zone and the crystal edge, indicating that some crystallization has

437 occurred subsequent to dissolution. Of the crystals with identified truncations in P zoning, 8 are

438 type 1 and 3 are type 2 crystals. This truncation of P zoning shows that these crystals

439 experienced disequilibrium conditions sometime before eruption and could be the result of their

440 incorporation into magmas of different composition, either by disruption of a cumulate or during

441 magma mixing.

442   Although the electron probe x-ray mapping technique used to identify minor element

443 zoning is time consuming, we have imaged a sufficiently large number of crystals to investigate

444 whether there is a relationship between the presence of P and Cr zoning in the two olivine

445 populations. Both type 1 and type 2 grains commonly exhibit zoning in P, with 24/25 type 1

446 grains and 39/44 type 2 grains showing P zoning (Table 3). Using the binomial distribution, it is

447 possible to evaluate whether these proportions are significantly different from one another. The

448 binary distribution is used to calculate the probability of obtaining one of two outcomes when the

449 same population is sampled randomly and independently (e.g., Taylor 1997). If we take the

450 probability of a crystal having P zoning as equal to the combined proportions of crystals

451 observed with P zoning (e.g., 63/69 or $p = 0.91$) then the probability of obtaining the observed

452 proportion of type 1 and type 2 crystals with P zoning is 23 and 16% respectively, and at 95%

453 confidence ($\alpha = 0.05$) we cannot reject the null hypothesis that the proportions of olivine with P-

454 zoning are the same in type 1 and type 2 crystals. In contrast, the proportions of crystals with

455 observed Cr zoning evident are much lower for type 1 crystals (4/21) than for type 2 (12/21).

456 Applying the same methodology and assuming the proportion of Cr zoning in all crystals is the

457 combined proportions (16/42, $p = 0.38$) the probability that either type 1 or type 2 crystals show

458 Cr zoning in the observed proportions is $<< 5\%$, allowing us to reject the null hypothesis that

459　there is no difference in the proportion of crystals with Cr zoning between type 1 and type 2

460　grains. If we instead use the observed proportions of Cr zoned crystals in type 1 and type 2

461　crystals separately to set the p value then probabilities that Cr zoning in type 1 olivine occur in

462　the same proportions as in type 2 grains, and vice versa, are even lower ($\ll$ 1%), again allowing

463　us to reject the null hypothesis. Applying this methodology to Al, the probability that crystals

464　show Al zoning in the observed proportions is 12% for type 1 and 10% for type 2 olivine, thus

465　the null hypothesis cannot be rejected at the 95% confidence level. However, if we use the

466　observed proportions of Al zoning in type 1 and type 2 crystals separately to set the p value, then

467　the probabilities that Al zoning occurs in the same proportions are 2% for type 1 grains and type

468　2 grains and the null hypothesis can be rejected.

469　　　　Although there are differences in the number of crystals that exhibit zoning in P, Al, and

470　Cr between type 1 and type 2 grains, it is likely that the incorporation of these three elements

471　into olivine is a linked process. Phosphorus may be incorporated in olivine at elevated

472　concentrations during periods of rapid crystal growth by both equilibrium partitioning of

473　elevated P contents in enriched boundary layers and also by non-equilibrium solute trapping—

474　where rapid olivine growth "runs over" these P enriched boundary layers (Milman-Barris et al.

475　2008; Grant and Kohn 2013; Welsch et al. 2014). To explain the similar zoning of P, Al, and Cr

476　in natural and experimentally grown olivine, Milman-Barris et al (2008) concluded that P is

477　incorporated at high concentrations with a coupled substitution with Al and Cr. Grant and Kohn

478　(2013) did not observe a correlation between P with Al in their equilibrium experiments, but

479　speculated that under non-equilibrium conditions (solute trapping), P may partition with a

480　growth rate-dependent coupled substitution with Al and Cr. Alternatively, Watson et al. (2015)

481　propose that the growth enrichment model (Watson and Liang 1995), which invokes differential

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482   partitioning behavior of the surface layer of a growing crystal with the interior and intra-

483   crystalline diffusion better explains the enrichment of P in olivine. Regardless of the exact

484   partitioning behavior of these elements, evidence suggests that Al and Cr are likely incorporated

485   in olivine along with P. In olivine, Al and Cr also diffuse more rapidly than P (Ito and Ganguly

486   2006; Milman-Barris et al. 2008; Watson et al. 2015), and Milman-Barris et al (2008) argued that

487   Al and Cr zoning profiles in the natural phenocrysts are erased during extended storage at high

488   temperatures while P zoning was preserved, which agrees with our observations.

489   To further establish that Al and Cr are incorporated in olivine in higher concentrations

490   where elevated P is observed we have applied a boundary layer model (Smith et al. 1955; Baker

491   2008). This approach models the enrichment of incompatible elements at the boundary of a

492   growing crystal, taking into account the crystal growth rate, the olivine-liquid partition

493   coefficient, and the diffusivity of the element in the melt. Our goal is to understand the

494   conditions that lead to P-rich boundary layers during olivine crystallization in Kilauea Iki and

495   other basaltic magmas, and to assess whether we can reasonably expect Al and Cr to be

496   consistently enriched in these zones by this same process. Watson et al (2015) performed similar

497   models for P, and here we also model Al and Cr to understand their behavior. Welsch et al.

498   (2013) predicted that the incorporation of P in olivine at high concentrations occurs during

499   periods of rapid, dendritic growth, which occurs at growth rates of $\sim 10^{-6}$ m/s and that subsequent

500   ripening, of the crystal occurs at a slower growth rate of $\sim 10^{-9}$ m/s. We therefore modeled the

501   boundary layer of a crystal after 10 μm of growth using this range of expected growth rates,

502   similar to the method of Watson et al. (2015).

503   Boundary layer models for P, Al and Cr (Figure 8), show that these elements are enriched

504   at the boundary of the growing olivine at high growth rates (10^{-6} m/s). Our results for P match

505   the results of the same modeling by Watson et al. (2015), and show enrichments factors of >4

506   and >1.5 at growth rates of 10^{-6} and 10^{-7} m/s, respectively (Figure 8). At the highest growth rate,

507   Al and Cr are also enriched by factors of 1.4 and ~1.2, respectively (Figure 8). The observed

508   enrichment factors from our laser ablation data for P ranges from 1.5 to 11 (median of 2.5),

509   which agree with the highest growth rates in the boundary layer models. The growth enrichment

510   model (GEM), proposed by Watson et al. (2015) to explain P zoning in olivine, may also

511   produce enrichments of Al and Cr in olivine. However, the partitioning behavior for these

512   elements in the surface layer of olivine is not well constrained, so we did not attempt to model

513   this here. Importantly for our purpose, our modeling suggests that P, Al, and Cr are co-enriched

514   at the olivine-liquid boundary during rapid crystal growth, and that instances where enrichment

515   of P occurs within olivine during rapid growth without enrichment of Al and Cr are unlikely. It is

516   also important to note that although diffusion may erase the Al and Cr zonation, there are

517   observed, significant differences in the absolute concentrations of Al (and P, Ti, and V) between

518   type 1 and type 2 olivine (Table 4) that are unrelated to mineral zoning. This suggests that

519   although Al is incorporated at higher concentrations along with P and diffusive re-equilibration

520   of Al may erase the initial zoning, these elements also record the last equilibration conditions of

521   these crystals. Also, the concentration of P is higher in type 2 crystals than type 1crystals (with

522   and without excluding the P-rich zones from the analysis), which is the opposite of what would

523   be expected for the dendritic growth model if elevated P is indicative of type 1 textures.

524   Zoning of trace elements in olivine provides a means to constrain the timescales of

525   crystal residence at magmatic temperatures in the Kilauea system. Although diffusion of divalent

526   cations in olivine is frequently used to infer the timescales of eruptive processes (e.g., Nakamura

527   1995; Coombs et al. 2000; Pan and Batiza 2002; Costa and Chakraborty 2004; Kahl et al. 2011;

528   2013; Marti et al. 2013; Ruprecht and Plank 2013; Longpre et al. 2014), we are more interested

529   in the longer timescales of crystal residence and thus focus on the slower diffusing elements, Cr

530   and P.

531   To effectively estimate diffusion timescales, appropriate diffusion coefficients are

532   needed. Ito and Ganguly (2006) measured diffusion coefficients for Cr in olivine and found that

533   it is highly anisotropic with little dependence on oxygen fugacity, at least from the wustite-iron

534   buffer to two log units above this buffer (Ito and Ganguly 2006). The oxygen fugacity of these

535   experiments was lower than what is reasonable for Hawaiian magmas; however, the calculated

536   diffusivites were the same over two orders of magnitude change in fO_2 (Ito and Ganguly 2006).

537   Therefore, it is likely that this diffusion coefficient is also appropriate for our samples. Diffusion

538   of Cr is anisotropic (Ito and Ganguly 2006), therefore the diffusion coefficient for Cr along the

539   measured transect was calculated using the equation

540          $$D_{traverse} = D_a(\cos\alpha)^2 + D_b(\cos\beta)^2 + D_c(\cos\gamma)^2 \qquad\qquad (1)$$

541   where D_a, D_b, D_c are the Cr diffusion coefficients along the a, b, and c-axes respectively, and α,

542   β, γ are the angles between the traverse and the a, b, and c-axes respectively as determined by

543   EBSD (Costa et al. 2008). Watson et al. (2015) report a diffusion coefficient for P in olivine, and

544   determined that diffusion is isotropic and not dependent on fO_2 (experiments performed at NNO

545   and WM buffers). Watson et al. (2015) also confirm that P diffuses at a slower rate than Cr, as

546   had been previously inferred (Ito and Ganguly 2006; Milman-Barris et al. 2008). To date, the

547   diffusivity of Al in olivine has not been determined, therefore Al is not modeled here. We

548   evaluate the uncertainties in the diffusion modeling using a Monte Carlo approach to take into

549   account the uncertainties in the diffusion coefficients and temperature (Ito and Ganguly 2006;

550   Putirka 2008; Watson et al. 2015) and conclude that the uncertainties for these two diffusion

551   coefficients are less than a factor of 2.5. The spatial resolution of the geochemical analysis is

552   another source of uncertainty for short diffusion timescales and was evaluated using the method

553   of Bradshaw and Kent (2017). Using the spatial resolutions (step size in individual x-ray element

554   maps) and diffusivities for individual crystals, we calculate the shortest timescales that can be

555   accurately calculated between 0.4 and 4.8 days for Cr diffusion and between 6 and 30 days for P

556   diffusion.

557   Diffusion is strongly temperature dependent, and as the temperature at which diffusion

558   occurs is not well constrained, this is often a large source of uncertainty in estimated timescales

559   (Costa et al. 2008). Many of the olivine grains in this study are inferred to be antecrystic,

560   therefore the temperature at which they formed could not be estimated using the observed whole

561   rock or glass compositions. Therefore, we calculated the equilibrium liquid from the composition

562   of individual olivine grains and equilibration temperatures using the MgO-liquid thermometer of

563   Helz and Thornber (1987) for Kilauea Iki and has an uncertainty of $\pm$ 10 °C. The equilibrium

564   liquid Mg# (Mg/(Fe+Mg)) was estimated using a K_D of 0.28 and a $Fe^{3+}/(Fe^{2+} + Fe^{3+})$ ratio of

565   0.17 (Helz et al. 2017). The MgO content of the equilibrium liquid was then estimated using the

566   observed trend of MgO and Mg# for Kilauean lavas (compilation taken from the GeoRoc

567   database: georoc.mpch-mainz.gwdg.de/). Results are given in Table 6 with an average from all

568   grains of 1182 $\pm$ 21 °C (one standard deviation, 1s) for both type 2 and type 1 grains. We used

569   the temperatures estimated from individual crystals using a FeO/MgO $K_D^{Olivine/melt}$ of 0.28 for

570   diffusion modeling. For comparison, the average temperature estimates using other published K_D

571   values range from 1251 $\pm$ 30 °C using a K_D of 0.30 (Roeder and Emslie 1970), to 1277 $\pm$ 32 °C

572   using a K_D of 0.33 (Putirka 2014), to 1286 $\pm$ 32 °C using a K_D of 0.34 (Matzen et al. 2011).

573   Using the temperatures derived from these higher K_D values would result in shorter absolute

574   diffusion timescales. For example, using a K_D of 0.34 results in estimated residence times that

575   are shorter by approximately a factor of 2.

576   Compositional gradients for diffusion modeling of P and Cr were extracted from the

577   electron microprobe x-ray element maps, which give better spatial resolution than LA-ICP-MS

578   analysis, and allows us to see the zoning in two dimensions to select transects that are normal to

579   zoning (Figure 9). Profiles of distance versus counts (as greyscale intensity) were taken directly

580   from the 16 bit x-ray element maps using ImageJ (Schneider et al. 2012). Diffusion modeling

581   was performed using an analytical solution of the diffusion equation for diffusion of an initial

582   compositional step function, assuming isothermal diffusion, after Crank (1975)

583             $$C = C_0 + \frac{(C_1 - C_0)}{2}\left[erfc\left(\frac{x}{2\sqrt{Dt}}\right)\right] \qquad (2)$$

584   where C is the concentration of the modeled profile, C_0 and C_1 are the initial concentrations on

585   either side of the step function interface at time zero, D is diffusivity (m^2/s), t is the diffusion

586   time (seconds), and x is distance (m) along the compositional gradient relative to the center of

587   the step function. The assumption of initial step function geometry results in maximum

588   timescales for the case where zoning has not been completely erased. To estimate the initial

589   zoning profiles for grains with P zoning but without observed Cr zoning, profiles with sharp P

590   zoning were selected, and the range of initial Cr intensity was derived from analyses of olivine

591   grains with similar P variation that also had Cr zoning. The calculated timescales for the case

592   where Cr zoning has been erased are minimum estimates as more time could have elapsed after

593   the initial zoning was erased (Figure 9).

594   For type 1 grains, the modeled diffusion timescales are between 10 days and 43 years for

595   Cr diffusion and between < 33 and 238 days for P diffusion (Table 7, Figure 10, Supplemental

596   Material). For type 2 grains, the timescales are between < 2 and 82 days for Cr diffusion, and <

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597   22 and 127 days for P diffusion (Table 7, Figure 10). Several grains produced timescales that are

598   below the lowest accurate timescale estimate and cannot be more accurately constrained with our

599   spatial resolution (Table 7; Bradshaw and Kent, 2017); these are depicted as arrows in Figure 10.

600   There is some discrepancy in the timescales estimated for Cr and P diffusion in individual grains,

601   which is most evident in type 1 grains that do not have observed zoning in Cr (Table 7). We also

602   note that the choice of initial conditions for estimating the timescales needed to erase Cr zoning

603   have a relatively large effect on the calculated timescales. We chose the initial conditions based

604   on the Cr intensities in type 2 olivine grains with similar magnitudes of P zoning, however, if the

605   initial zoning of Cr was less pronounced, then the calculated timescales would be shorter.

606   Preliminary modeling of one such profile (Iki-Scoria-14E), suggests that for any case where Cr

607   zoning was initially present, it would take years to erase the Cr zoning.

608   An important observation is that most type 1 grains do not have equivalent Cr zoning

609   evident with P, whereas more of the type 2 grains do (Tables 2, 6), and as shown above, this

610   difference is statistically significant. Assuming there was initially Cr zoning present, long

611   residence times (years to decades) are thus needed to remove all indication of enriched Cr.

612   However, the timescale estimates from P for these same profiles are much lower and overlap

613   with P and Cr timescales from type 2 grains (Figure 10). This discrepancy may be because our

614   assumption that Cr zoning always accompanies P zoning in type 1 grains is not correct.

615   However, constraints from kinetic modeling and the presence of Cr zoning in similar type 2 (and

616   some type 1) grains suggest that type 1 olivine crystals did have initial Cr zoning. Type 1 and

617   type 2 grains have significantly different proportions of olivine with Cr zoning, suggesting that

618   there is some fundamental difference between these two olivine populations. Our data is more

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619   consistent with Cr zoning being erased by diffusion at high temperatures, with P not being

620   affected as much.

621   The sharp P zoning and short apparent diffusion times along with erased Cr zoning giving

622   longer apparent diffusion timescales is consistent with observations from mafic and ultramafic

623   intrusions (e.g., Welsch et al., 2014; Xing et al., 2017). Olivine in intrusions would be expected

624   to have diffuse P-zoning due to extended storage at high temperatures and slower cooling

625   compared with olivine in rapidly quenched lavas. However, olivine from Mid-Atlantic Ridge

626   gabbros, the Rum intrusion (Welsh et al., 2014), and the Abulangdang and Baima lherzolite and

627   dunite intrusions in SW China (Xing et al., 2017) retain P zones that are sharp, suggesting that

628   diffusion in these P-rich zones may occur at even slower rates than currently measured (Watson

629   et al., 2015). If this is the case then the timescales we have calculated from P diffusion may be

630   shorter than the true residence times.

631   There are uncertainties for diffusion modeling, which effect the absolute timescales. The

632   most prominent of these is the temperature estimates, which in this case were derived from

633   estimates of the equilibrium liquid composition based on the Fe-Mg concentrations in individual

634   olivine grains. This approach yields uncertainties in the equilibrium liquid composition, which

635   along with the uncertainty in the Fe-Mg liquid geothermometer (Helz and Thornber, 1987)

636   produce the uncertainty in the final temperature estimate. Also, the calculated equilibrium liquid

637   composition represents the last equilibrium conditions experienced by the olivine, which might

638   not reflect the long-term storage conditions of the magma. However, despite the uncertainties in

639   the absolute residence time estimates, we interpret our observations to suggest that type 1 grains

640   resided at high temperatures significantly longer than type 2 grains.

641   **Kilauea Iki Picrite Formation**

642 **Summary of Observations and Interpretations**

643   There are several observations presented above, which need to be satisfied by any model

644 for the formation of Kilauea Iki (and other) picrites and their contained olivine.

645   • Presence of two distinct olivine populations based on optical characteristics: type

646   1 and type 2 grains (e.g., Helz 1987; Natland 2003; Welsch et al. 2013).

647   • The two olivine populations are geochemically distinct, most evident in slow

648   diffusing trace elements (P, Al, Ti, V), which likely reflects differences in source

649   magma compositions or formation and storage conditions.

650   • The two olivine populations have different CSD slopes (with the type 1-only

651   slope being shallower), and that the type 1 olivine grains may have experienced

652   textural coarsening.

653   • Residence time estimates, from Cr and P diffusion models suggest that the type 1

654   olivine grains have experienced a longer magmatic residence than the type 2

655   grains.

656 **Existing Models for Picrite Formation**

657   Central to any model for the assembly of picrites must address the origins of the two

658 textural and geochemical populations of olivine. The traditional model (Francis 1985; Albarede

659 and Tamagnan 1988; Clague and Denlinger 1994; Baker et al. 1996; Garcia 1996; Révillon et al.

660 1999; Sakyi et al. 2012) states that type 1 olivine is sourced from deforming cumulates beneath

661 the volcano, and type 2 olivine crystallizes from the host liquid. Recent models have suggested

662 that type 1 olivine textures may be the result of collisions (Natland 2003), or growth effects

663 (Welsch et al. 2013; 2014). In this section, we explore the existing models for the formation of

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664   olivine, and test whether their predictions satisfy the observations for the origins of distinct

665   olivine populations.

666   Below, we focus on mechanisms that might produce type 1 and type 2 texture in

667   autocrystic or antecrystic olivine, rather than lithospheric olivine incorporated as xenocrysts. As

668   argued elsewhere (e.g., Larsen and Pedersen 2000), the low to moderate Ca contents and

669   presence of melt inclusions in both textural types argue against olivine derived directly from

670   mantle peridotite (Simkin and Smith 1970; Helz 1987; Garcia et al. 2007; Vinet and Higgins

671   2010; 2011). Low Ca content is consistent with these olivine grains crystallizing from magmas at

672   low pressure (e.g., Simkin and Smith 1970), and are not likely due to re-equilibration as it would

673   take hundreds of years for Ca to diffuse into the largest type 1 crystals (using diffusion

674   coefficients from Chakraborty, 2010).

675   **Non-cumulate models.** Non-cumulate models must account for the formation of type 1

676   olivine textures during growth and transport within a liquid. Natland (2003) suggests that olivine

677   grains experience non-hydrostatic stresses resulting from crystal-crystal or crystal-conduit wall

678   collisions during flow, which could cause type 1 olivine textures. However, Welsch et al. (2013)

679   noted that there is little evidence for collisions seen on the surfaces of type 1 olivine. The model

680   of Natland (2003) also does not account for the differences in the geochemistry we observe

681   between type 1 and type 2 olivine (assuming equilibrium partitioning) because, although it would

682   require two magmas with different olivine compositions and textures to mix, the collisions that

683   would produce deformation are thought to be enhanced during magma mixing, so olivine from

684   both magmas would have type 1 textures. The collisions suggested to produce deformation also

685   are predicted to fracture the grains to produce the conchoidal fragments recognized by Helz

686   (1987), but this fragmentation would cause a net decrease in the size of type 1 grains, which is

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687   not consistent with crystal size observations. This model also does not explain the different

688   residence times of the two olivine populations. The two magmas that mixed during the Kilauea

689   Iki eruption were an older, shallow, stored magma (1959W) and a younger magma that bypassed

690   the summit reservoir (1959E) and intersected the stored magma (Wright 1973; Helz 1987;

691   Poland et al. 2014; Wright and Klein 2014). In this scenario, the olivine from the younger

692   ascending magma would be more likely to be deformed during its rapid ascent (Helz 1987) and

693   would produce two olivine populations with type 1 grains with *shorter* residence times and type

694   2 grains with *longer* residence times, the opposite of our modeling results.

695   The other alternative model is presented by Welsch et al. (2013; 2014) who suggest that

696   type 1 olivine grains are the result of a two-stage growth history, with initial rapid, dendritic

697   growth followed by slower growth and ripening of the crystals. In essence, as a crystal grows

698   rapidly, crystal "buds" grow dendritically from a single crystal, then during the slow-growth

699   phase those "buds" eventually grow into one another (Welsch et al. 2013). The resulting mosaic

700   of crystals would have an overall shape similar to euhedral olivine, but any lattice mismatches

701   among the different crystal "buds" would cause their boundaries to be misaligned and create type

702   1 olivine textures (Welsch et al. 2013), which would explain the first observation.

703   For the dendritic growth model of Welsch et al. (2013; 2014) to explain our other

704   observations, the chemical differences we observed require that the type 1 and type 2 grains were

705   sourced from separate magmas or that crystallization occurred at far-from-equilibrium conditions

706   that were different for type 1 and type 2 grains. In our samples, about the same proportion of

707   type 1 and type 2 olivine exhibit P zoning (Table 3), which is a major prediction of the dendritic

708   growth model for *type 1* grains (Welsch et al. 2013; 2014). Therefore, either the two-stage

709   growth predicted to produce type 1 olivine is not the cause of type 1 textures or the lattice

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710 mismatches that produce these textures are rare. The rapid growth that enriches olivine in P,

711 would also enrich it in Al (and Ti, V, and Cr), therefore, we would expect type 1 olivine to have

712 higher overall P, Al, Ti, and V contents that type 2 olivine. However, we observe that P

713 concentrations are *lower*, and Al, Ti, and V are *higher* in type 1 grains than type 2 grains (Figure

714 4), which further suggests that zoning in P and Al is not correlated with defects in olivine.

715   The dendritic growth model of Welsch et al. (2013; 2014) also does not explain the

716 different textural features of type 1 and type 2 grains, such as the different CSD slopes, crystal

717 shapes, and crystal sizes. These textural differences could be explained if the different olivine

718 populations were sourced from separate magmas that mixed during the eruption. However, as

719 discussed above, both populations have P zoning and thus likely underwent similar growth

720 histories, and thus should have similar textures and sizes.

721   **Cumulate model.** A cumulate model best explains our observations for the assembly of

722 Kīlauea Iki and other lavas containing type 1 olivine. In this model type 1 textures in olivine are

723 formed by plastic deformation in a deforming olivine cumulate, which are later disrupted and

724 incorporated in unrelated ascending magmas (Helz 1987; Albarede and Tamagnan 1988; Clague

725 and Denlinger 1994; Garcia 1996; Garcia et al. 2007; Vinet and Higgins 2010). This explains the

726 presence of geochemically distinct populations of type 1 and type 2 olivine in the same magma,

727 with the type 1 grains being sourced from the disaggregated cumulate, and the type 2 grains from

728 the carrier magma(s). This process would also produce CSDs with different slopes and average

729 grain sizes for the type 1 and type 2 grains, as the type 1 grains would likely experience textural

730 coarsening during storage in the cumulate, whereas the CSD for the type 2 grains would only

731 record their crystallization history. Type 1 grains also show more evidence for dissolution and

732 fewer have euhedral shapes than type 2 grains, which is likely the consequence of partial

32

733   dissolution during the disruption of the cumulates. This dissolution appears to be a minor part of

734   the history of type 1 olivine, and it likely occurred after a significant amount of coarsening

735   occurred within the cumulate, thus preserving the large sizes of these crystals. This model also

736   explains how the type 1 population has a longer residence time than the type 2 population due to

737   extended storage in a cumulate.

738   Type 1 olivine from cumulate zones would be readily available as these cumulate zones

739   have been estimated to comprise 14 vol% of Hawaiian volcanoes (Clague and Denlinger 1994),

740   and have been imaged as zones of high seismic wave velocity beneath the summits and rift zones

741   of Hawaiian volcanoes (e.g., Okubo et al. 1997; Kauahikaua et al. 2000; Denlinger and Morgan

742   2014). The seaward flow of these dense cumulates, the same process interpreted to form

743   deformation textures in olivine, may also be the source of gravitational instability that leads to

744   large landslides that dissect these volcanoes (Clague and Denlinger 1994; Clague and Sherrod

745   2014; Denlinger and Morgan 2014).

746   A criticism of the deforming cumulate model is that some of the type 1 crystals have

747   euhedral outlines (Welsch et al. 2013; 2014). Welsch et al. (2013; 2014) argue that because some

748   type 1 grains have euhedral outlines, they could not have been plastically deformed, and

749   therefore type 1 olivine textures must be a primary growth feature. Overall, in Kilauea Iki

750   magmas, type 1 crystals with euhedral habits are less common than euhedral type 2 crystals. Of

751   the crystals analyzed in this study, <20% of type 1 crystals were classified as euhedral, whereas

752   60% of type 2 grains are euhedral. Type 1 grains may develop euhedral outlines during cooling

753   after disaggregation from a cumulate as shown in the melting experiments of Boudier (1991).

754   The experiments of Boudier (1991) produced euhedral crystal faces of deformed grains during

755   the relatively slow cooling (several minutes) after experimental melting of peridotite, which

756   allowed the olivine grains to obtain equilibrium where they were in contact with the liquid

757   produced during melting. Although euhedral forms are not produced during melting (e.g., Soulie

758   et al., 2017), the experiments of Boudier (1991) show that they may form relatively quickly

759   during the slower cooling obtained in that particular experimental setup, as opposed to the rapid

760   quenching in other experiments. This process observed during slowly-cooled melting

761   experiments, may be analogous to the processes involved in the disaggregation of a crystal-rich

762   cumulate as the crystals may achieve equilibrium with the surrounding intercumulus liquid

763   during cooling and could explain the ubiquity (yet low abundance of) euhedral crystals. The

764   euhedral, type 1 crystals in this study commonly contain thin, sharp P zones near to and parallel

765   with the crystal rims, suggesting late-stage rapid crystallization related to rim growth, which may

766   record the crystallization after disaggregation from a cumulate. Several crystals (8 type 1 and 3

767   type 2) also have evidence for P zoning truncation near resorption surfaces, suggesting they

768   experienced some dissolution, which may be the result of partial dissolution during disruption in

769   a cumulate. Boudier (1991) also showed that dissolution of olivine is more pronounced along

770   zones of weakness such as deformation boundaries, which may explain the common

771   embayments at the edges of type 1 grains (Figure 1), as opposed to them being dendritic crystal

772   buds as proposed by Welsch et al. (2013; 2014).

773   **Type 2 Olivine Source.** Type 2 olivine crystals likely have a less complicated history

774   than type 1 olivine. The most likely source for type 2 grains is from one or both of the magmas

775   that mixed to form the Kilauea Iki eruption. The type 2 grains are commonly euhedral, which has

776   been interpreted to mean that they crystallized, uninhibited in a melt (Helz 1987; Welsch et al.

777   2013), although euhedral crystals may also form during cooling after partial melting (e.g.,

778   Boudier 1991). The slight curvature of the type 2-only CSD (Table 5) suggests that there may be

779   two slightly different populations of type 2 grains. If this is the case it is likely to be the result of

780   mixing between the 1959E and 1959W endmember magmas as suggested by Wright (1973) and

781   Helz (1987). These magmas are both similar, and crystallize similar olivine grains, thus the

782   curvature on the CSD is small and we see no obvious compositional bimodality in the type 2

783   population, suggesting that any difference in minor element (P, Al, Cr, V) composition between

784   olivine from these two sources is smaller than differences between type 1 and type 2 olivine.

785   **Implications**

786   Our work suggests that type 1 and type 2 olivine in Kilauea Iki picrites require different

787   conditions for their formation. Type 1 and type 2 olivine likely experience similar initial growth

788   histories as evidenced by similar P zoning; however, type 1 grains require non-hydrostatic stress

789   to develop deformation textures. Type 1 olivine textures do not appear to result from growth

790   phenomena or subsequent crystal collisions. These results suggest that the type 1 grains in

791   Kilauea Iki magmas were sourced from an olivine-rich cumulate zone deeper in the plumbing

792   system, where prolonged crystal residence and plastic deformation occurs. These type 1 grains

793   were likely sampled by the ascending deep magma (1959E) during times of rapid ascent, as

794   evidenced from the high fountaining events. The type 2 grains are autocrysts sourced from both

795   the deep magma (1959E), and the older magma (1959W) stored beneath Kilauea Iki. Similar to

796   Kilauea Iki, many other volcanoes erupt picrites that contain both type 1 and type 2 olivine (e.g.,

797   Francis 1985; Albarede and Tamagnan 1988; Révillon et al. 1999) and may have experienced

798   similar processes in their formation.

799   Our methods can be applied to olivine-rich volcanic rocks from other locations to further

800   test the cumulate model for the formation of deformation textures in olivine. Our work shows the

801   value of combining an array of textural and geochemical observations in studying the origin of

802   olivine crystals in picritic magmas and emphasizes the important role of olivine-rich cumulate

803   zones in controlling the production of olivine-rich picritic magmas.

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Figure Captions

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1043   Figure 1. Cross polarized photomicrographs of type 1 (**a-c**) and type 2 grains (**d**).

1044   Resorption is evident at the ends of the kink bands in grains a and b and is indicated by red

1045   arrows. White arrows indicate the locations of LA-ICP-MS transects.

1046

1047   Figure 2. Phosphorus X-ray maps of type 2 olivine, where brightness corresponds with

1048   higher P concentrations. (**a**) Resorbed olivine with P zoning that is disrupted by dissolution

1049   surfaces. (**b-d**) Olivine grains with P zoning that have equivalent Al and Cr zoning (not shown).

1050   White arrows indicate the locations of LA-ICP-MS transects, the dashed portions of the arrows

1051   show the high-P regions of the transects that were removed to compare the compositions of type

1052   1 and type 2 olivine.

1053

1054   Figure 3. Phosphorus X-ray maps of type 1 olivine. Dashed red lines indicate locations of

1055   dislocation bands. (**a**) Olivine grains with P zoning that has equivalent zoning in Al and Cr (not

1056   shown). (**b-d**) Olivine grains with P zoning only. White arrows indicate the locations of LA-ICP-

1057   MS transects, the dashed portions of the arrows show the high-P regions of the transects that

1058   were removed to compare the compositions of type 1 and type 2 olivine.

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1060   Figure 4. Cumulative distribution functions for select elements in olivine. Significant

1061   differences between type 1 (n = 42) and type 2 (n = 42) olivine grains are evident in slow

1062   diffusing elements Al, P, and Ti (**a-c**) No significant differences evident in the more rapidly

1063   diffusing elements, Cr (**d**), and Fe/Mg (Fo content) (**e-f**). The histogram of Fo content is shown

1064   in (**f**) for comparison with the CDF (e). The p-values for the two sample Kolmogorov-Smirnov

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1065   test are shown, which indicates the probability that the two samples are derived from the same

1066   population. The median values of individual crystals were used to create the CDFs and

1067   histogram, when all the data are used, the CDFs are smoother and have the same general

1068   characteristics.

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1070   Figure 5. Kolmogorov-Smirnov test p-values plotted against the median concentration for

1071   each element in individual olivine grains. Slow diffusing elements (< 10^{-17} m^2/s at 1,300 °C; red

1072   symbols) that have high p-values all have low abundances in olivine (<1 μg/g), whereas the slow

1073   diffusing elements Al, Ti, P, and V have higher abundances and lower p-values. Faster diffusing

1074   elements (blue symbols) have high p-values and high concentrations. Dashed line is $\alpha = 0.05$.

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1076   Figure 6. Olivine crystal size distributions for all Kilauea Iki samples. Type 1 grains have

1077   fewer small crystals and extend to larger sizes than type 2 grains. The type 1 and type 2 olivine

1078   populations also have different slopes.

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1080   Figure 7. Phosphorus X-ray maps of type 1 olivine showing resorption along the rims that

1081   disrupt P zoning. Red arrows indicate areas where zoning is disrupted. In areas where zoning is

1082   disrupted, there are P-poor zones between the zoning and the resorbed rim indicating some

1083   regrowth after dissolution.

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1085   Figure 8. Models showing the relative enrichment of the boundary layer around a crystal

1086   growing at different rates. Total crystal growth is 10 μm in each model. Each element is enriched

1087   at growth rates needed for incorporation of elevated P (Welsch et al., 2013). Element diffusion

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1088   coefficients in the melt for Al and P are from Baker (2008) and Cr from Zhang et al. (2010).

1089   Partition coefficients for P, Al and Cr are from Grant and Kohn (2013), Beattie (1993) and

1090   Spandler and O'Neill (2010), respectively.

1091

1092   Figure 9. Example forward diffusion models with best-fit residence times. (**a-d**) Type 2

1093   grain (Iki-36-27-A) that has P and associated Cr zoning. (**e-h**) Type 1 grain (Iki-Scoria-14-A)

1094   that has P zoning but no associated Cr zoning. The Cr diffusion model for this grain is the

1095   minimum time necessary to erase the assumed initial Cr zoning.

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1097   Figure 10. Residence time estimates based on diffusion of Cr (**a**) and P (**b**) in individual

1098   olivine grains (17 profiles from 10 olivine grains). Error bars show the uncertainty in the

1099   diffusion times from the uncertainty in the diffusion coefficient and temperature estimates. The

1100   diffusion profiles that produced timescales below the shortest accurate timescales are depicted

1101   with an arrow.

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Tables

Table 1. Olivine textures

Sample Name	NMNH SI Catalog #	Olivine Proportion (vol %)[a]	Type 1 Olivine (vol %)[a]	Type 1/ Total Olivine (modal %)[b]	Type 1/ Total Olvine (vol %)[a]	Number of Crystals[b]	
						Type2	Type 1
All Samples		15.2	7.7	16.0	50.7	757	144
Iki-36	116111-36	14.6	6.8	13.9	46.6	196	34
Iki-43	116111-43	10.1	3.3	13.9	32.7	199	32
Iki-66	116111-66	11.7	5.6	11.5	47.9	131	17
Iki-75	116111-75	30.5	17.4	22.4	57.0	135	39
Iki-Scoria		17.6	11.7	18.6	66.5	96	22

Notes: [a] volume from 2D crystal intersections, steriologically corrected to 3D shapes (Higgins, 2006a).

[b] Mode from number of crystals in the thin sections.

Table 2.   Mean EBSD results for repeat spot analyses of individual grains

Crystal	Texture	n	phi1	PHI	phi2
Iki-36-1	Type 1	7	264	116	355
Iki-36-1A	Type 1	2	90	64	182
Iki-36-6	Type 2	3	115	35	348
Iki-36-6A	Type 2	2	289	147	248
Iki-36-7	Type 1	3	277	43	10
Iki-36-12	Type 2	3	237	59	70
Iki-36-25	Type 1	2	262	137	353
Iki-36-27a	Type 2	5	100	107	337
Iki-36-27b	Type 2	2	280	73	22
Iki-36-30	Type 2	6	167	92	70
Iki-36-33	Type 2	2	288	145	183
Iki-36-34	Type 2	2	118	39	350
Iki-36-35	Type 2	4	99	143	351
Iki-43-9	Type 1	4	37	40	39
Iki-43-13	Type 2	2	111	51	169
Iki-43-23	Type 1	3	7	151	338
Iki-43-25	Type 1	2	149	137	42
Iki-43-GM-2	Type 2	2	78	93	293
Iki-43-GM-3	Type 2	2	220	173	21
Iki-66-13	Type 1	3	234	166	107
Iki-66-GM-2	Type 2	5	38	47	7
Iki-66-GM-5	Type 2	2	197	55	186
Iki-Scoria-1	Type 2	2	222	172	136
Iki-Scoria-2	Type 1	3	125	102	312
Iki-Scoria-3	Type 1	3	323	142	109
Iki-Scoria-8	Type 2	3	60	119	34
Iki-Scoria-14	Type 1	3	86	100	335
Iki-Scoria-29	Type 2	3	85	76	185
Iki-Scoria-32	Type 1	2	78	95	333
Iki-Scoria-34	Type 2	2	97	82	335
Iki-Scoria-38	Type 2	3	66	126	219
Iki-Scoria-39	Type 2	3	172	141	304

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Table 3.   Minor element zoning

Olivine Texture	Imaged Grains		Grains with Zoning			Percent with Zoning		
	P and Al	Cr	P	Cr	Al	P	Cr	Al
Type 1	25	21	24	4	6	96	19	24
Type 2	44	21	39	12	16	89	57	36

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Table 4.	Two sample Kolmogorov-Smirnov test	
	Excluding high P zones	Including high P zones
Element	p-value	p-value
P	0.01	<0.01
Ti	0.03	<0.01
V	0.03	<0.01
Al	<0.01	<0.01
Dy	0.35	0.57
Er	0.26	0.76
Yb	0.33	0.23
Y	0.18	0.52
Cr	0.56	0.10
Ca	0.75	0.13
Na	0.10	0.16
Co	0.56	0.62
Sc	0.91	0.41
Ni	0.26	0.48
Fe	0.56	0.55
Mg	0.39	0.71
Mn	0.91	0.63
Li	0.40	0.98

Note: Elements listed in approximate order of increasing diffusivity.

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Table 5.   CSD Results

	CSD Slope	Intercept	Goodness of Fit (Q)
Type 1	-1.23 (0.08)	-1.81 (0.32)	0.46
Type 2	-2.87 (0.10)	1.52 (0.14)	7.5×10^{-5}
V&H G2a	-2.7	1.3	
V&H G2b	-1.11	-2.28	
Mangan	-2.88	1.03	

Note: Values in parenthesis are 1 s uncertainties

V&H: Vinet and Higgins (2011), Mangan: Mangan (1991)

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Table 6.	Equilibrium liquid temperature estimates											
	$K_D = 0.28$		$K_D = 0.30$		$K_D = 0.33$		$K_D = 0.34$	
	Average T (°C)	Standard deviation	Average T (°C)	Standard deviation	Average T (°C)	Standard deviation	Average T (°C)	Standard deviation
All	1234	21	1195	22	1214	23	1220	23
Type 1	1233	21	1194	22	1213	23	1219	24
Type 2	1236	21	1197	21	1216	23	1222	23

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Table 7. Results of diffusion modeling

Sample	Cr diffusion time (days)	P diffusion time (days)	Comments	Orientation[a]		
				α	β	γ
Type 2						
Iki-36-27-A	22	< 46		56	119	58
Iki-36-27-C	82	127		96	69	158
Iki-36-30-C	< 5	84		119	21	83
Iki-43-GM-2b-A	4	< 22		109	47	49
Iki-66-GM-2b-D	< 2	< 22		19	116	81
Iki-66-GM-5b-B	5	< 22		75	57	143
Iki-66-GM-5b-C	35	< 22		98	55	143
Iki-Scoria-1-C	5	< 43		86	173	85
Type 1						
Iki-43-23-A	2665	238	No Cr zoning	179	83	82
Iki-43-25-A	2279	< 33	No Cr zoning	71	49	133
Iki-43-25-B	1111	34	No Cr zoning	112	31	111
Iki-66-13-A	10	< 53		81	171	88
Iki-Scoria-14-A	3932	< 72	No Cr zoning	113	73	151
Iki-Scoria-14-D	3747	< 72	No Cr zoning	33	109	127
Iki-Scoria-14-E	1591	71	No Cr zoning	41	117	61
Iki-Scoria-2-B	15746	200	No Cr zoning	53	118	130
Iki-Scoria-2-C	1967	113	No Cr zoning	75	89	165

Note: [a] α, β, and γ are the angles between the measured transect and the a, b, and c axes, respectively
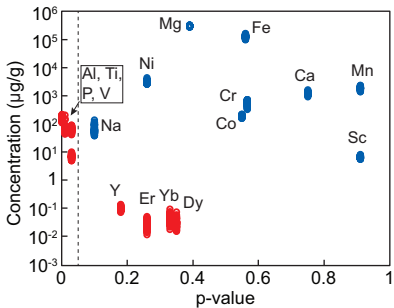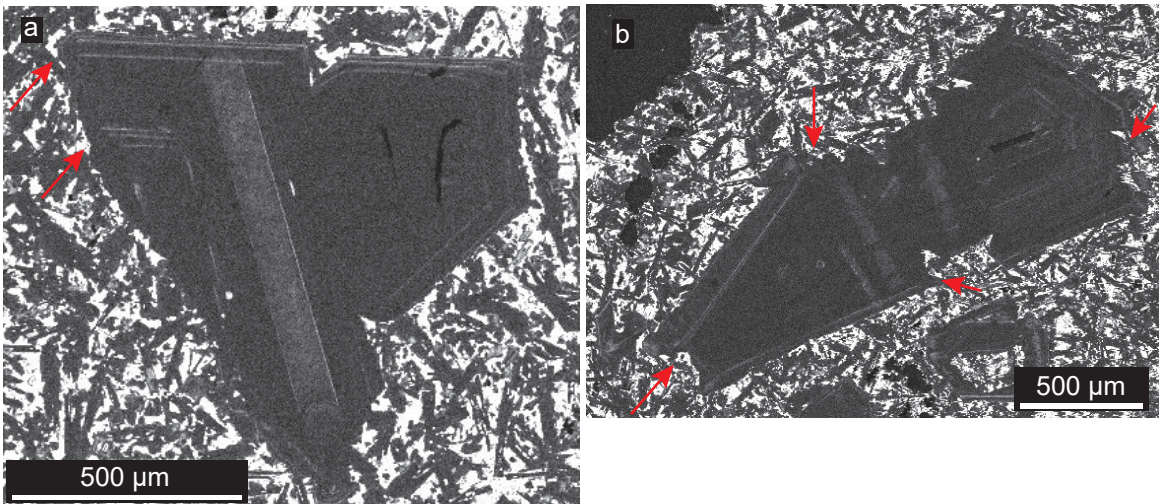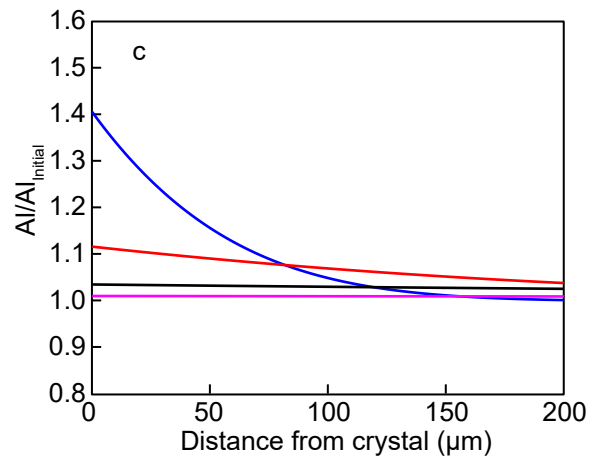
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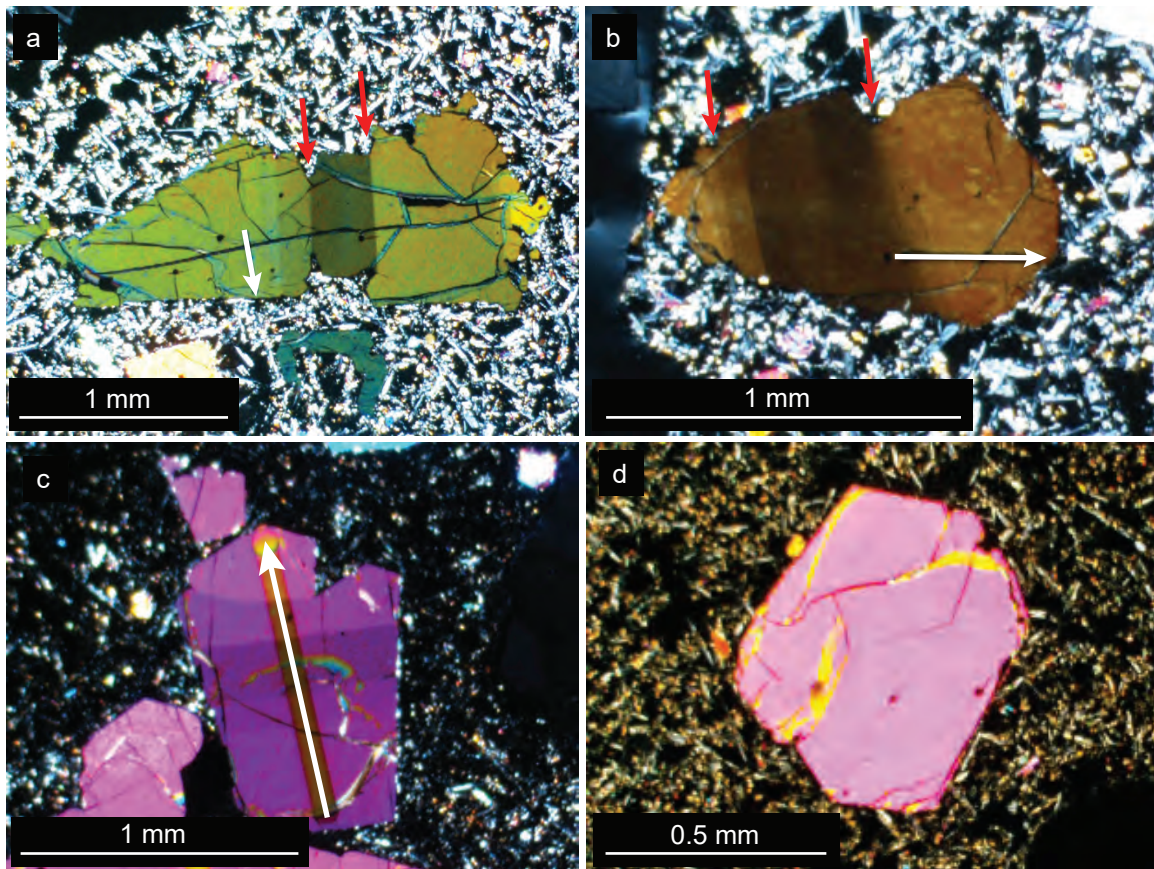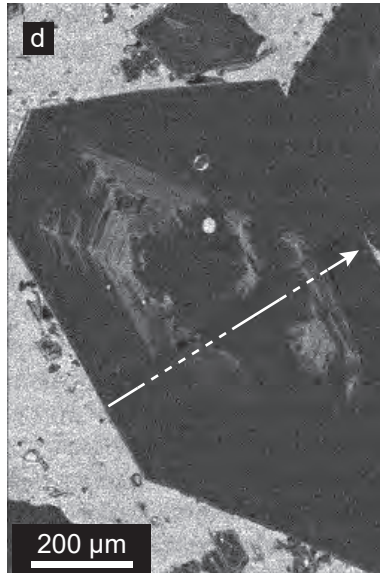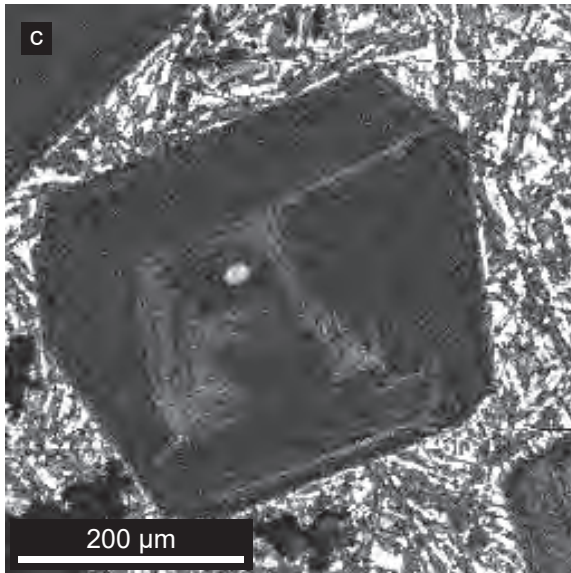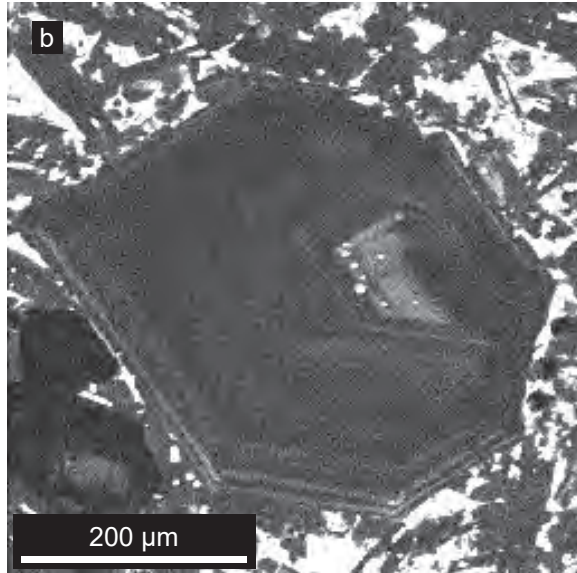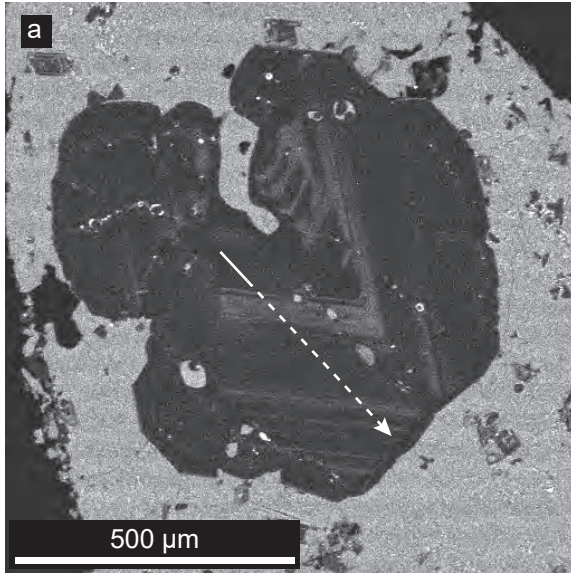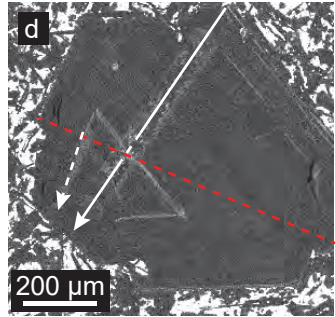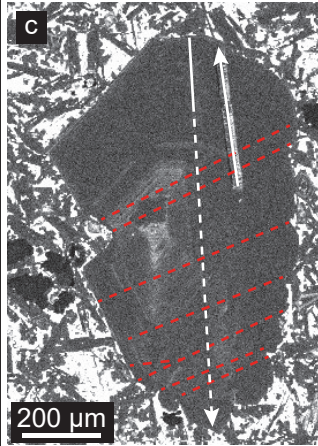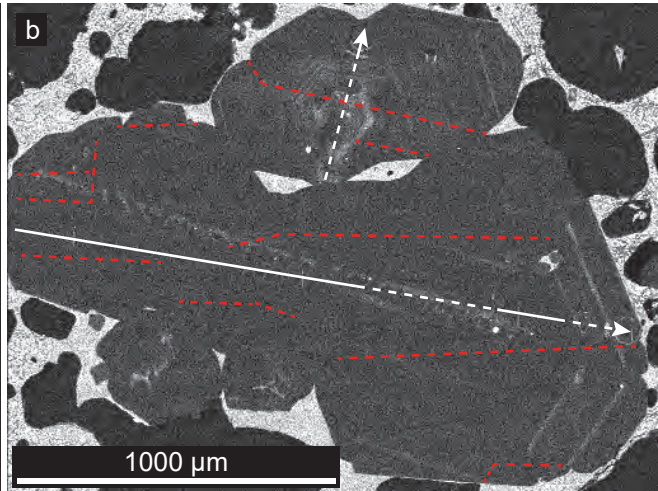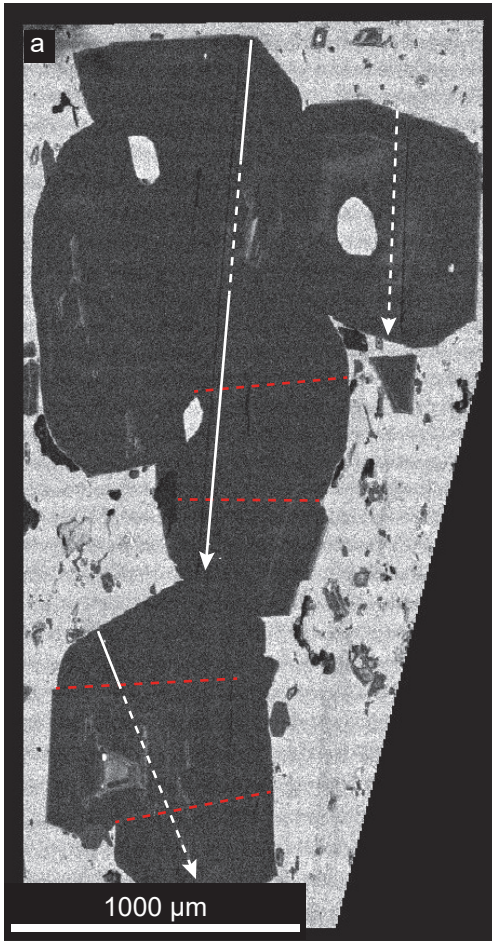
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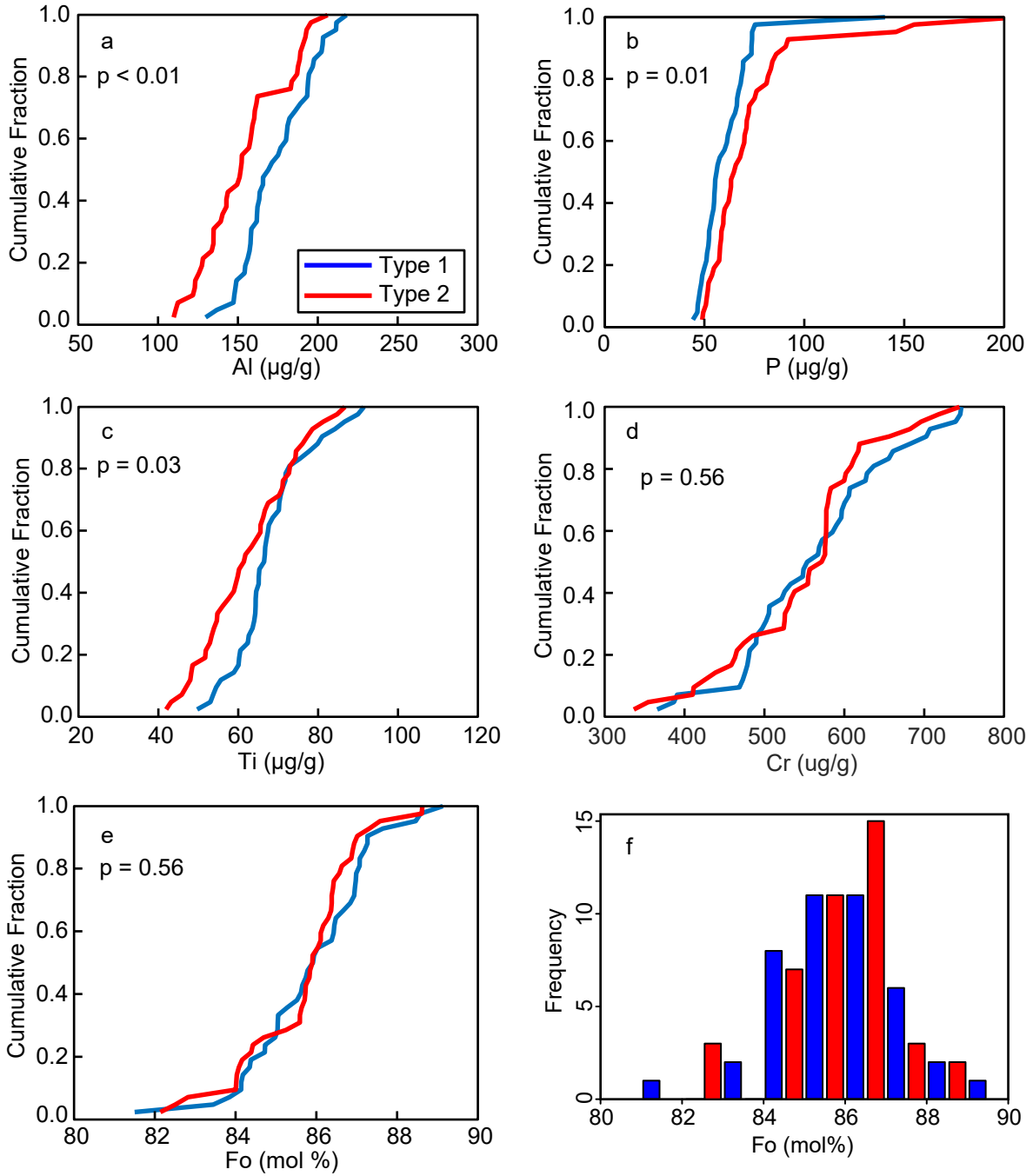
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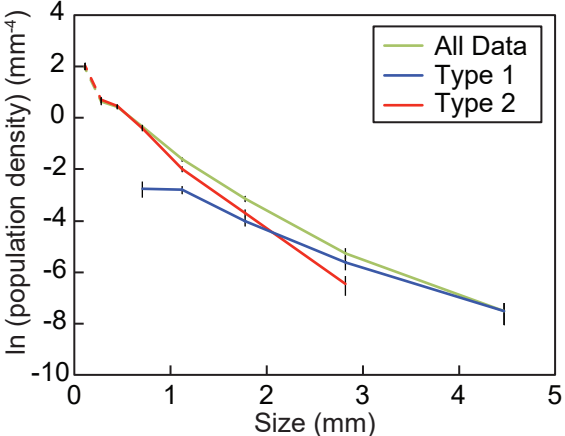
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Figure 8

Figure 9

Figure 10