1 Mixing-induced chemical disequilibrium creates and modifies

2 basaltic crystal cargoes

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12 Abstract

13 Crystal cargoes of oceanic basalts often preserve records of mantle-derived chemical variability that 14 are erased from their carrier melts by magma mixing. However, the impacts of mixing between 15 similarly primitive but chemically variable magmas remain poorly understood despite ubiquitous 16 evidence of chemical variability in primary melt compositions and chemical disequilibrium in erupted 17 crystal cargoes. Here we report observations from magma-magma reaction experiments performed on 18 analogues of primitive Icelandic lavas from chemically and lithologically distinct mantle sources to 19 determine how crystals respond to mixing-induced chemical disequilibrium. Chemical variability in 20 our experimental products is dominantly controlled by major element diffusion in the melt that alters 21 phase equilibria relations and triggers waves of plagioclase resorption in initially plagioclase-saturated 22 magmas. Isothermal mixing of chemically variable oceanic basaltic magmas may thus play a 23 pervasive role in creating and modifying basaltic crystal cargoes by unlocking plagioclase-rich 24 mushes and driving resorption, (re-)crystallisation and solid-state diffusion.

25 Introduction

26 Chemical variability in primitive oceanic basalts results from variability in mantle melting processes 27 and mantle source compositions^{1,2}. Correlations between the isotopic and incompatible element 28 compositions of erupted basalts reflect how subduction and lithospheric recycling have created 29 isotopic and chemical heterogeneity in the mantle through geological time³. Subduction has also 30 created lithological heterogeneity in the mantle that propagates through melting into correlated variability in the major and trace element compositions of erupted basalts^{4,5}. An important 31 32 consequence of this major element variability is that similarly primitive but chemically variable 33 oceanic basalts have different phase equilibria relations under the same pressure-temperature (P-T)conditions, and therefore evolve along compositionally distinct trajectories^{6,7}. However, mantle-34 derived chemical variability is progressively erased by mixing during magma ascent and evolution^{8,9}, 35 36 meaning that erupted basalts are often less variable than the primary melts from which they have evolved and rarely record their initial chemical variability¹⁰. Fortunately, records of mantle-derived 37 38 chemical variability are often preserved within crystal cargoes and melt inclusions that are more 39 resistant to mixing-induced re-equilibration than the liquids that carry them to the surface and hence 40 provide vital insights into mantle source compositions and magma reservoir properties¹¹⁻¹⁴. 41 Nevertheless, crystal-melt interactions between chemically variable oceanic basalts undergoing mixing are poorly understood, and mixing's role in the creation and modification of basaltic crystal 42 43 cargoes remains largely unexplored.

44 Records of mantle-derived variability in basaltic crystal cargoes are associated with 45 disequilibrium features that range from simple concentric zoning to more complex textures indicative of resorption, rapid crystallisation and diffusive re-equilibration^{15–17}. Crystal-hosted melt inclusions 46 47 provide complementary archives that confirm the presence of considerable mantle-derived chemical variability within individual magma plumbing systems^{14,18}. Indeed, disequilibrium associated with this 48 49 chemical variability may play a fundamental role in the formation of some melt inclusion^{19,20}. Disequilibrium features in crystal cargoes record changes in magma $P-T-H_2O$ activity (a_{H_2O})-oxygen 50 fugacity (f_{Ω_2}) -composition (X) conditions as well as the timescales over which these changes occur, 51

52 and thus enable us to reconstruct magmatic evolution histories. For example, compositional zoning in 53 plagioclase crystals often preserves information about magma reservoir processes and magma transport pathways^{21–23}, while compositional zoning in olivine crystals can preserve information about 54 timescales of magma storage and ascent $^{24-26}$. Although recharge by hot and primitive magmas often 55 creates disequilibrium features in basaltic crystal cargoes^{16,17,27}, near-isothermal mixing between 56 chemically variable oceanic basalts can also occur^{8,9}. Given that the phase assemblages and phase 57 compositions of basaltic systems with different bulk compositions can differ at a under isothermal 58 contisions⁷, isothermal mixing is likely to have important but ill-defined effects on crystal cargoes. 59

60 Experimental simulations of magma mixing typically focus on one of the two processes that occur in natural systems: the physical mingling of melts in dynamic experiments^{28,29}, and the diffusive 61 homogenisation of melts in classic diffusion couples^{30,31}. While physical mingling is an essential 62 63 component of magma mixing, it is diffusive homogenisation that ultimately changes melt 64 compositions and can modify crystal cargoes via resorption, (re-)crystallisation and solid-state 65 diffusion processes. However, melt-melt diffusion couple experiments are typically performed under superliquidus conditions to avoid crystals complicating the characterisation of intra-melt diffusion 66 67 processes^{30,32}; melt-melt diffusion couples provide incomplete insights into magma-magma reactions. 68 Although diffusive controls over crystal growth and resorption have been investigated in some simple systems^{33–35}, current experiments typically define how pressure and temperature rather than melt 69 70 composition affect mineral stabilities. Further experiments are therefore required to constrain how 71 crystals respond to changes in melt compositions triggered by magma mixing.

Mantle-derived chemical variability is particularly well characterised in basalts from
southwest Iceland. Specifically, incompatible element-depleted basalts from lherzolitic mantle sources
have high Al₂O₃ and CaO but low FeO* (total Fe expressed as FeO) and Na₂O contents at any given
MgO content, while incompatible element-enriched basalts from recycled mantle sources have low
Al₂O₃ and CaO but high FeO* and Na₂O contents³⁶. This dichotomy is highlighted by the
Háleyjabunga and Stapafell lavas on the Reykjanes Peninsula that constitute compositional endmembers (Figs. 1a, 1b)¹⁴. Crystallisation experiments performed on synthetic analogues of these lavas

79 illustrate how mantle-derived variability in major element compositions affects phase equilibria 80 relations⁷. Namely, plagioclase crystallises from the Al₂O₃-rich and incompatible element-depleted 81 Háleyjabunga lava analogue at >40 °C higher than it does from the Al_2O_3 -poor and incompatible 82 element-enriched Stapafell lava analogue (Figs. 1c, 1d). Thus, incompatible element-depleted 83 magmas crystallise a greater proportion of their mass for any given decrease in temperature than their 84 incompatible element-enriched counterparts, potentially resulting in the deep sequestration of 85 incompatible element-depleted magmas as plagioclase-rich cumulates and the progressive biasing of 86 more evolved magmas towards incompatible element-enriched compositions derived from recycled mantle sources⁷. Moreover, isothermal differences in phase equilibria relations between Háleviabunga 87 88 and Stapafell lava analogues suggest that mixing between variably enriched magmas will modify 89 crystal cargoes as the chemically distinct magmas attempt to restore chemical equilibrium. The 90 Háleyjabunga and Stapafell lavas thus represent excellent model systems for investigating how 91 mixing-induced chemical disequilibrium affects the basaltic crystal cargoes and the magma plumbing 92 systems from which they erupt.

93 Here we present the results of magma-magma reaction experiments designed to determine 94 how mixing-induced chemical disequilibrium affects the crystal cargoes of chemically variable 95 primitive oceanic basalts. Specifically, we investigate how crystals respond to the diffusive re-96 equilibration of melt major element contents between two chemically distinct magmas. We do this by 97 juxtaposing magma cylinders produced from synthetic analogues of the Háleyjabunga and Stapafell 98 lavas at 300 MPa and 1190 °C for durations of 1 to 96 hours, conditions selected to maximise phase 99 equilibria differences between the two synthetic magmas and ensure that experiments were carried out 100 under realistic magma storage conditions^{7,37}. Having defined how crystals respond to the diffusive re-101 equilibration of their host melts in our experimental system, we explore the implications of our 102 findings for crystal mush stability and the formation of disequilibrium features in natural systems.

103 Results

Magma synthesis experiments. Magma cylinders were synthesised in Fe-preconditioned Au₈₀Pd₂₀
 capsules during crystallization experiments performed on dry glass powders of Háleyjabunga and

106 Stapafell lava analogues. The syntheses were performed in an internally heated pressure vessel 107 (IHPV) at 300 MPa and 1190 °C for 72 hours. The experiments were conducted at low $a_{\text{H}_{2}\text{O}}$ (~0.06,

equivalent to a melt H₂O content of ~ 0.7 wt.%³⁸) and broadly reducing conditions (approximately one

log unit above the fayalite-magnetite-quartz buffer; $FMQ+1^{39}$).

- 110 The products of the synthesis experiment performed on the incompatible element-depleted
- 111 Háleyjabunga lava analogue contain crystals of olivine, clinopyroxene and plagioclase (Fig. 2a),

112 whereas experimental products from the incompatible element-enriched Stapafell lava analogue only

113 contain crystals of olivine and clinopyroxene (Fig. 2b). Although the products of the incompatible

element-depleted experiment have a lower average glass content (i.e. melt mass fraction, *F*) than that

the products of the incompatible element-enriched experiment (F = 0.72 and 0.90, respectively,

according to mass balance; all phase analyses are provided in the Supplementary Data), glasses are

117 interconnected throughout the products of both synthesis experiments. Crystals are typically euhedral

to subhedral, consistent with a close approach to equilibrium, and while some clinopyroxene

aggregates occur, degrees of crystal impingement are generally low.

120 Magma-magma reaction experiments. Magma-magma reaction experiments were performed at 121 exactly the same $P-T-a_{H_2O}-f_{O_2}$ conditions as magma synthesis experiments (300 MPa, 1190 °C, a_{H_2O} 122 ~ 0.06, $f_{O_2} \sim FMQ+1$) for durations of 1, 4, 24 and 96 hours; they were also performed in the same 123 IHPV. Two ~3 mm-long cylinders of each synthetic magma analogue were juxtaposed in new Fe-124 preconditioned Au₈₀Pd₂₀ capsules. The dense and FeO*-rich Stapafell analogue was always placed at 125 the bottom of the capsule to minimise advective processes during experimental runs⁴⁰.

The products of magma-magma reaction experiments retain the textural characteristics observed in the products of magma synthesis experiments (Figs. 3), with crystals being both smaller and more abundant in the products of the incompatible element-depleted Háleyjabunga analogue than in those of the incompatible element-enriched Stapafell analogue. Glass composition profiles through the products of magma-magma reaction experiments have variably linear and sigmoidal forms that depend on the element and experimental duration in question (Fig. 4). Profiles near the central axis of

experimental products have similar forms to those near their rims, meaning that parallel profiles could
be stacked following normalisation to common midpoints. Importantly, the similarity of parallel
profiles demonstrates that advection was limited once magma-magma interfaces were established and
that experimental products hence record dominantly diffusive signals.

136 Away from initial magma-magma interfaces, glasses from the Stapafell analogue have lower 137 Al₂O₃ contents and higher FeO*, TiO₂ and K₂O contents than those from the Háleyjabunga analogue 138 after short experimental durations of 1 and 4 hours (~14.5, ~11.0, ~1.7 and ~0.22 wt.% versus ~16.0, 139 ~10.0, ~0.75 and ~0.10 wt.%, respectively; Figs. 4a-c, 4e). Differences in CaO and Na₂O contents are 140 also visible after 1 and 4 hours (~12.0 and ~2.1 wt.% versus ~12.6 and ~1.7 wt.%, respectively; 141 Figs. 4c, 4f), but are comparable in magnitude to 2σ analytical uncertainties (±0.44 and ±1.3 wt.%, 142 respectively). SiO₂ and MgO contents do not vary significantly between the two magma analogues 143 and are not shown in Fig. 5 (full phase compositions are provided in the Supplementary Data). Al₂O₃ 144 TiO₂ and K₂O profiles have sigmoidal forms that show progressively more gradual transitions 145 between the starting glass compositions with increasing experimental duration (Figs. 4a, 4c, 4e). FeO* 146 profiles are sigmoidal in the products of 1- and 4-hour experiments, but linear in the products of 24-147 and 96-hour experiments (Fig. 4b). FeO* contents are also displaced to lower mean FeO* contents in 148 the products of the 24- and 96-hour experiments, possibly as a result of Fe loss to Au₈₀Pd₂₀ capsules in 149 these experiments⁴¹. CaO and Na₂O profiles are linear and CaO and Na₂O contents do not vary 150 beyond analytical uncertainties in the products of 24- and 96-hour experiments, (Figs. 4d, 4f). 151 Diffusive re-equilibration of experimental glasses. Stacking glass composition profiles by 152 normalising distances through experimental products by square roots of experimental durations 153 demonstrates that glass compositions are primarily controlled by diffusive re-equilibration between chemically distinct melts in the starting magma analogues (Fig. 5)^{31,42}. This is because diffusion 154 155 distance scales with the square root of the diffusion coefficient multiplied by the diffusion time (i.e.

156 $x \propto \sqrt{Dt}$, where x is the diffusion distance, D is the diffusion coefficient and t is time)⁴², meaning

157 that composition profiles will overlap once the effects of diffusion over different durations is taken

158 into account (assuming that diffusion coefficients remain constant)³¹. Key implications of the

159	overlapping time-normalised glass composition profiles shown in Fig. 5 are that diffusion appears to
160	have operated coherently during all of our magma-magma reaction experiments and that observations
161	from the products of different experimental runs may thus be meaningfully compared with each other.
162	Effective binary diffusion coefficients estimated by fitting error functions through glass
163	composition profiles to solve Fick's 2 nd Law are consistent with the modest body of published values
164	available (Fig. 6) ^{32–35,43–45} . Most published diffusion coefficients for basaltic melts have been derived
165	from crystal dissolution experiments ^{33–35} or classic melt-melt diffusion couples ^{32,43} . Na diffusivity has
166	also been estimated from radiotracer diffusion experiments ⁴⁴ . Where error functions can be fitted to
167	our glass composition profiles, our effective binary diffusion coefficients are typically within one ln
168	unit of regressions through global datasets that follow Arrhenian relationships ($D = D_0 e^{-E_A/RT}$,
169	where D is the diffusion coefficient, D_0 is the pre-exponential factor, E_A is the activation energy for
170	diffusion and R is the gas constant; Fig. 6). Diffusion coefficients estimated from 1- and 4-hour
171	experiments are particularly close to global regressions for Al ₂ O ₃ , FeO* and CaO ($r^2 = 0.83$, 0.92 and
172	0.79 respectively), suggesting that the behaviour of these elements in basaltic melts can be largely
173	explained without complex multicomponent diffusion models (Figs. 6a, 6b, 6d). While our estimated
174	diffusion coefficients for TiO_2 are similar to published values, the latter span three ln units at any
175	given T and global regression statistics are modest ($r^2 = 0.59$), suggesting that TiO ₂ diffusion may be
176	more strongly affected by matrix effects than the diffusion of other elements (Fig. 6c). Although
177	published diffusion coefficients are scarce for K ₂ O and Na ₂ O ^{43,44} , our estimates fall on plausibly
178	Arrhenian trends with the few data available (Fig. 6e, 6f). Estimated diffusivities of Al_2O_3 and TiO_2
179	are similar to theoretical Eyring diffusivities that relate diffusion coefficients to temperature and melt
180	viscosity an perform well when describing the behaviour of network-forming cations an oxygen
181	$(D_E = k_B T / \lambda \eta$, where D_E is the Eyring diffusivity, k_B is Boltzmann's constant, λ is a jump distance
182	of 0.4 nm related to the atomic spacing of silicate liquids and η is the average viscosity of the two
183	end-member melts, calculated here with the model of Giordano et al. ⁴⁶) ^{31,47} . In contrast, estimated
184	diffusivities of Na ₂ O and, to a lesser extent, K ₂ O are higher, possibly reflecting differences in
185	diffusion mechanisms between network-forming and network-modifying cations (Figs. 6) ^{48,49} .

186	Mineral stabilities and compositions. Olivine crystals are present in the products of magma
187	synthesis experiments on both lava analogues (Fig. 2, 7a). However, they have higher forsterite
188	contents [$X_{Fo} \sim 0.852$ versus 0.843, respectively, where $X_{Fo} = Mg/(Mg+Fe)$ on a molar basis] in the
189	products of the experiment on the FeO*-poor Háleyjabunga analogue than the products of the
190	experiment on the FeO*-rich Stapafell analogue. This difference in olivine compositions is reflected
191	by X_{Fo} steps across magma-magma interfaces in the products of 1- and 4-hour experiments (Fig. 7a).
192	Steps in X_{Fo} also occur in the products of 24- and 96-hour experiments but are less prominent than
193	those in the products of the 1- and 4-hour experiments as a consequence of the olivine undergoing
194	diffusive re-equilibration in response to Fe loss (24-hour experiment in particular) and diffusive
195	homogenisation of melt FeO* contents (96-hour experiment in particular). Indeed, some olivine
196	crystals from these experiments appear to show diffusion profiles (Supplementary Fig. 1).

197 Plagioclase crystals are present in the products of the synthesis experiment on the 198 Háleyjabunga analogue; plagioclase is absent from the products of the synthesis experiment on the 199 Stapafell analogue (Fig. 2 7b). In line with published phase equilibria relations⁷, these crystals have 200 high anorthite contents [$X_{An} \sim 0.83$, where $X_{An} = Ca/(Ca+Na)$ on a molar basis]. Similarly, plagioclase 201 is only stable within the portions of magma-magma reaction experiments derived from the 202 Háleyjabunga lava analogue. Although X_{An} variability exceeds analytical uncertainty in the products 203 of all magma-magma reaction experiments, mean X_{An} does not vary systematically as a function of 204 position or experimental duration. In contrast, plagioclase stability depends strongly on experimental 205 duration. While plagioclase is stable throughout the portion of the 1-hour experiment derived from the 206 synthesis experiment on the Háleyjabunga lava analogue (Fig. 3a), the region where plagioclase is 207 stabile progressively shrinks as experimental duration increases (Fig. 3b-d), leaving a >1-mm broad 208 region of plagioclase resorption in the products of 24- and 96-hour experiments. Projecting regions of 209 plagioclase stability onto time-normalised composition profiles indicates that plagioclase resorption 210 occurs when melt Al₂O₃ contents are lowered below ~16 wt.% by diffusive re-equilibration in the 211 melt (Fig. 5a). The region of plagioclase stability in the products of 24-hour experiments is smaller 212 than expected from comparisons with the products of other experiments, which we attribute to the

aforementioned modification of melt compositions by Fe loss leading to corresponding increases in other melt components including Al_2O_3 . Nevertheless, our experiments indicate that plagioclase stability is dominantly controlled by melt Al_2O_3 content (and thus melt Al/Si) rather than melt CaO and Na₂O contents (and thus Ca/Na) as both CaO and Na₂O diffuse considerably faster than Al_2O_3 in both our experimental system and basaltic melts more generally (Fig. 6)⁴⁵.

218 Clinopyroxene crystals are present in the products of synthesis experiments on both lava 219 analogues (Fig. 2). While much variability in clinopyroxene Mg-number $[Mg#_{cnx}, where Mg#_{cnx}]$ 220 Mg/(Mg+Fe) on a molar basis] is related to sector zoning in experimentally produced clinopyroxene 221 crystals⁵⁰, a slight difference in mean $Mg\#_{cpx}$ between the products of the experiments on the 222 Háleyjabunga and Stapafell analogues (Mg $\#_{cpx} \sim 0.835$ versus 0.850, respectively) reflects differences 223 in their melt FeO* contents (Fig. 7c). However, differences between the two magma analogues are 224 more clearly expressed in terms of clinopyroxene TiO_2 contents, with clinopyroxene crystals grown 225 from the incompatible element-enriched Stapafell analogue containing considerably more TiO_2 than 226 those grown from the incompatible element-depleted Háleyjabunga analogue (TiO₂ ~ 0.48 versus 227 0.28 wt.%, respectively; Fig. 7d). Chemical variability associated with clinopyroxene sector zoning 228 masks Mg#_{cpx} steps across magma-magma interfaces in the products of magma-magma reaction 229 experiments (Mg#cpx steps of ~0.01 are difficult to resolve within total Mg#cpx ranges of 0.825-0.855, Fig. 7c). Nevertheless, overall gradients of low Mg#_{cpx} in the Stapafell analogue increasing to high 230 231 $Mg\#_{cox}$ in the Háleyjabunga analogue can be resolved. In contrast, steps in clinopyroxene TiO₂ 232 contents between magma analogues are much more distinct (from $TiO_2 \sim 0.2-0.3$ wt.% in portions 233 from the Háleyjabunga analogue to 0.4–0.6 wt.% in portions from the Stapafell analogue; Fig. 7d), consistent with the slow diffusion of TiO₂ within clinopyroxene preventing intra-crystalline re-234 equilibration over the duration of our experiments⁵¹. Positions of TiO₂ steps do, however, vary with 235 236 experimental duration, with high-TiO₂ clinopyroxene crystals seemingly replacing low-TiO₂ 237 clinopyroxene crystals in the portion of the Háleyjabunga analogue adjacent to magma-magma 238 interfaces in the products of longer duration experiments. Overall, regions of increased clinopyroxene 239 TiO₂ content largely overlap with regions of plagioclase resorption, feasibly reflecting the complete

resorption and re-crystallisation of clinopyroxene crystals, which may also be related to the diffusive re-equilibration of melt Al_2O_3 contents, though further work will be required to clarify this.

242 Discussion

243 Diffusive controls over melt compositions. Although physical mingling is vital for homogenising 244 mantle-derived chemical variability in primitive basalts undergoing mixing, chemical diffusion is 245 ultimately required for mixing to reach completion. Our magma-magma reaction experiments suggest 246 that chemically variable magmas must be mechanically thinned to filaments no more than a few mm 247 wide for diffusive homogenisation to be achieved within the day-to-week timescales typically 248 associated with mixing processes in basaltic plumbing systems^{24,25}. Indeed, chemical variability in 249 olivine-hosted melt inclusions from individual eruptions reflects the entrapment of variably mixed melts^{8,13}, suggesting that crystallisation and diffusive homogenisation of melt compositions occur over 250 251 broadly consistent timescales. However, our experiments also suggest that mixing can create chemical 252 anomalies by fractionating elements with different diffusivities (e.g., K₂O from TiO₂) that are capable 253 of complicating records of mantle-derived chemical variability. There is compelling evidence that 254 mixing induces diffusive fractionation in low-T evolved systems that are predisposed to preserve diffusive features^{31,52,53}, though little comparable evidence has yet been presented for mixing-induced 255 256 diffusive fractionation in high-T primitive systems. Diffusive fractionation has however been 257 implicated in the creation of high field strength elements depletions in some plagioclase-hosted melt inclusions from oceanic basalts^{19,20}, and it is at the µm-scale of melt inclusions that it is likely to have 258 259 its greatest effects. Thus, even transient modifications of melt compositions by diffusion can 260 fundamentally alter our understanding of magmatic processes if they are captured by the melt 261 inclusion archives upon which geological interpretations are often based⁵⁴.

The effective binary diffusion coefficients we estimate are not significantly affected by the presence of moderate crystal proportions (crystal mass fractions of 0.1–0.3). Error function fits through glass composition profiles reproduce compositions from crystal-rich portions of experimental products equally well as those from crystal-poor portions (Supplementary Fig. 2), and diffusivities estimated from these fits are consistent with those estimated from melt-only systems (Fig. 6)^{32–35,43,44}.

Furthermore, melt compositions close to magma-magma interfaces are not detectably affected by
either plagioclase resorption or clinopyroxene re-crystallisation; melt compositions in magma-magma
reaction experiments are dominantly controlled by diffusion within the melt, with Fe loss imposing
second-order controls in longer-term experiments. Our experiments thus suggest that the diffusive
homogenisation of crystal-bearing basaltic liquids can be meaningfully approximated by the diffusive
re-equilibration of pure melts.

Mineral responses to diffusive changes in melt compositions. Primitive oceanic basalts inherit
considerable chemical variability from the mantle³⁻⁵, meaning that different mantle melt batches can
have different bulk compositions at similar temperatures and degrees of magmatic evolution^{8,9}. As a
consequence, isothermal mixing between chemically variable magmas is just as likely to occur as
mixing between cool magmas residing within magma reservoirs and hot recharge magmas^{16,17,24,27}.
Thus, crystal cargoes will not only experience recharge-induced changes in magma temperature but
also mixing-induced changes in magma chemistry that occur under near-isothermal conditions.

280 Plagioclase crystals in magma-magma reaction experiments responded to diffusion-induced 281 changes in local melt compositions by resorbing completely within a few hours. While plagioclase 282 crystals in the products of the magma synthesis experiment on the Háleyjabunga analogue are 283 generally euhedral and tabular (Fig. 2a), those at limits of plagioclase stability in the products of 284 magma-magma reaction experiments are anhedral, embayed and sometimes skeletal (Fig. 8). Our 285 magma-magma reaction experiments have therefore captured plagioclase resorption in action. 286 Specifically, plagioclase mass fractions decrease from far-field values of 0.11 to zero over intervals of 287 no more that $\sim 200 \,\mu m$ that host the most embayed and skeletal plagioclase crystals observed. The 288 small scales of these intervals imply that plagioclase stability in our magma-magma reaction 289 experiments was controlled by the kinetics of Al₂O₃ diffusion in the melt rather than the kinetics 290 plagioclase resorption at crystal-melt interfaces. In other words, the distribution of plagioclase crystals 291 in our experimental products indicates that once local melt Al_2O_3 contents dropped below ~16 wt.%, 292 plagioclase crystals on order of 20 µm long resorbed completely within no more than a few hours. 293 Encouragingly, this observation is consistent with published experiments on anorthite resorption

kinetics, demonstrating that plagioclase crystals can be easily resorbed by mixing-induced changes in melt composition at rates in excess of $10 \,\mu$ m/hour³⁵.

296 Olivine and clinopyroxene crystals also responded to diffusion-induced changes in local melt 297 compositions in our experimental products, but in different ways from plagioclase crystals. The 298 development of compositional zoning in olivine crystals from longer-duration experiments suggests 299 that they, at least in part, responded to isothermal changes in melt composition by diffusive re-300 equilibration in the solid state (Fig. 7a; Supplementary Fig. 1). Nevertheless, we note that some 301 crystals close to magma-magma interfaces are subhedral, which implies that some textural maturation 302 by resorption and crystallisation also occurred (Fig. 8). The partial re-equilibration of olivine crystals 303 over lengthscales of 10–100 µm within timescales of 96 hours is consistent with the relatively fast Mg–Fe interdiffusion in olivine⁵⁵, and provides independent validation of mixing timescales estimated 304 from diffusion profiles in natural systems²⁵. In contrast, clinopyroxene crystals seemingly responded 305 306 to changes in local melt composition by resorption and re-crystallisation, as evidenced by steps in 307 clinopyroxene TiO₂ content that progressively invade Háleyjabunga analogues with increasing 308 experimental durations (Fig. 7d). Importantly, these steps in clinopyroxene TiO_2 content are offset 309 from magma-magma interfaces, indicating that they cannot have resulted from convection within 310 experimental capsules. Moreover, clinopyroxene is denser than the H₂O-poor basaltic melts 311 considered here (~ 3.3 versus ~ 2.7 g/cm³), confirming that high-TiO₂ clinopyroxene could not have 312 floated from the Stapafell analogue into the overlying Háleyjabunga analogue. However, the 313 implications of our experimentally observed clinopyroxene re-crystallisation for natural systems 314 remain unclear, partly because the small size of experimental crystals (typically $<10 \ \mu m$ in length) 315 makes it difficult to gauge how larger crystals would respond in nature. It is nevertheless plausible 316 that well-documented instances isobaric clinopyroxene resorption in natural basalts may reflect 317 resorption triggered by changes in magma chemistry as well as in magma temperature^{12,56}.

318 Magma mixing creates and modifies basaltic crystal cargoes. Crystal mush disaggregation plays a 319 major role in creating the crystal cargoes carried by many oceanic basalts^{13,57,58}, and the eruption of 320 plagioclase glomerocrysts with textures reminiscent of igneous cumulates is often interpreted as

evidence for pre-eruptive mush entrainment^{21–23}. Although numerical models provide vital insights 321 322 into the behaviour of crystal-rich magmas⁵⁹, the processes by which intergrown and cohesive mushes 323 disaggregate remain largely unclear. For example, the primitive crystal cargoes carried by many 324 oceanic basalts are generally too refractory to have been thermally by the evolved melts that carry them to the surface^{13,22,57}: Assuming magma evolution along a single liquid line of descent, liquids in 325 equilibrium with relatively evolved plagioclase crystals ($X_{An} \le 0.7$) could not resorb grain boundaries 326 327 in mushes dominated by primitive plagioclase crystals ($X_{An} > 0.8$) formed during earlier phases of 328 higher temperature crystallisation. However, primitive crystal cargoes and evolved carrier liquids are often derived from chemically distinct primary melt distributions^{22,60,61}. This means that mush crystals 329 330 may have never been in equilibrium with melts directly related to their eventual carrier liquids and that grain boundary resorption could be driven by isothermal differences in melt chemistry as much as 331 332 by differences in melt chemistry coupled with differences in magma temperature.

333 Crystal mush disaggregation in many basaltic systems may be triggered by chemical 334 disequilibrium associated with the injection of plagioclase-undersaturated magmas into plagioclase-335 saturated magma reservoirs. Very few basalts erupted in oceanic settings are in equilibrium with high-336 $X_{\rm An}$ plagioclase at any stage of their evolution^{13,57,62}. Instead, high- $X_{\rm An}$ plagioclase in oceanic settings 337 likely crystallises from incompatible element-depleted primitive melts that are correspondingly 338 enriched in refractory elements like CaO and Al₂O₃ and rarely survive crustal processing to erupt at the surface^{7,62}. As demonstrated by our magma-magma reaction experiments, the isothermal injection 339 340 of incompatible element-enriched magmas into reservoirs containing incompatible element-depleted 341 magmas can trigger plagioclase resorption in response to local in melt compositions (Figs. 4, 8). We 342 thus suggest that chemical rather than thermal disequilibrium may play an important role in triggering 343 crystal mush disaggregation in some magma reservoirs. Specifically, we propose that mixing-induced 344 changes in melt compositions modify phase equilibria relations to the point where the plagioclase 345 (and possibly clinopyroxene) grain boundaries that bind primitive mushes together can start to resorb 346 (Fig. 9a). As a consequence, resorption-facilitated mush disaggregation could occur in response to 347 transient changes in local melt compositions caused by even volumetrically minor injections of

348 incompatible element-enriched magma into large and incompatible element-depleted reservoirs.

349 Furthermore, the effects of chemical disequilibrium will be greatest close to loci of magma injection,

350 creating positive feedbacks between the chemical and physical processes responsible for mush

disaggregation and crystal entertainment.

352 Chemical and isotopic disequilibria between basaltic melts and high- X_{An} plagioclase crystals 353 suggest that chemical variability plays a widespread role in driving crystal mush disaggregation and 354 creating basaltic crystal cargoes. Chemical disequilibrium is well documented in plagioclase 355 ultraphyric basalts from ocean islands and slow- and intermediate-spreading mid-ocean ridges^{22,60,61}. 356 This is probably because plagioclase ultraphyric basalts typically erupt in locations without long-lived 357 melt lenses capable of efficiently filtering eruption products and erasing mantle-derived chemical variability⁶³. Nevertheless, the occurrence of mush-derived plagioclase crystals in oceanic basalts 358 359 from diverse settings that include fast-spreading mid-ocean ridges suggests that the disaggregation processes we describe may operate on a global scale^{23,64,65}. Finally, we note that variations in the H₂O 360 361 content of arc magmas may have analogous effects to variations in the major element content of 362 oceanic basalts, whereby the injection of H₂O-rich magmas or fluids into H₂O-poor reservoirs may 363 facilitate mush disaggregation by triggering plagioclase resorption⁶⁶.

364 Regardless of their origins, crystal cargoes will be modified as they interact with liquids in 365 equilibrium with different phase assemblages and different phase compositions as a result of magma 366 mixing, with different mineral phases responding in different ways. The transfer of high- X_{An} 367 plagioclase crystals between melts with different CaO and Al₂O₃ contents can result in either 368 resorption or crystallisation depending on the direction of transfer. For example, our magma-magma 369 reaction experiments demonstrate how the diffusive equilibration of incompatible element-depleted 370 and plagioclase-saturated magmas with incompatible element-enriched and plagioclase-371 undersaturated magmas can trigger isothermal plagioclase resorption (Figs. 4, 8). However, it is 372 straightforward to envisage slightly different scenarios in which diffusive influxes of Al₂O₃ into 373 incompatible element-enriched magmas instead trigger waves of crystal nucleation in systems close to 374 plagioclase saturation or waves of rim growth in systems that are already saturated in plagioclase.

375 Although decompression, primitive recharge and boundary layer effects doubtlessly contribute towards the textural complexity of plagioclase crystals in oceanic basalts^{13,17,21,23}, it is that clear that 376 377 isothermal and isobaric changes in melt composition are also capable of significantly modifying 378 crystal textures. Furthermore, mixing-induced changes in melt composition present a compelling 379 mechanism for driving the cycles of resorption and re-crystallisation implicated in the formation of plagioclase-hosted melt inclusions from oceanic settings^{19,20}. Although correlated changes in 380 temperature and melt composition drive the largest diffusive changes in olivine X_{F_0} contents^{24–26}. 381 382 variations in olivine X_{Fo} can also reflect isothermal variations in melt chemistry (Fig. 9c). Indeed, our 383 experiments demonstrate that olivine crystals may largely respond to changes in melt composition by 384 diffusive re-equilibration rather than re-crystallisation. Conversely, our observations suggest that clinopyroxene crystals can respond to changes in melt composition by resorption and re-385 386 crystallisation (Fig. 9c), though more work is required to understand the rates and mechanisms by 387 which this occurs. Some of the textural complexity observed in natural clinopyroxenes may nevertheless result from mixing between chemically distinct primitive melts^{12,56}. 388

389 Magma mixing creates and modifies basaltic crystal cargoes and can therefore alter magmatic 390 plumbing system dynamics and erupted records of mantle-derived chemical variability. Crystal 391 resorption in response to mixing-induced chemical disequilibrium represents an underappreciated yet 392 potentially widespread mechanism for disaggregating crystal mushes that can also alter the viscosity, density and eruptibility of oceanic basalts through the entrainment of buoyant high- X_{An} plagioclase 393 crystals^{22,57}. Magma mixing not only creates crystalline records of chemical variability through the 394 395 crystallisation of concentric zones and the entrapment of melt inclusions but also destroys them 396 through resorption, re-crystallisation and solid-state diffusion. Overall, isothermal mixing between 397 chemically variable primitive magmas likely plays an equally important role in generating the 398 texturally and chemically complex crystal cargoes we observe at the surface as more widely 399 recognised processes invoking mixing between variably evolved and thermally distinct magmas.

400 Methods

Experimental methods. Synthesis experiments were performed in an internally heated pressure 401 vessel (IHPV) in the Institut für Mineralogie of the Leibniz Universität Hannover. Experiment were 402 403 performed at 300 MPa and 1190 °C to maximise differences in resulting phase assemblages and 404 proportions according to published equilibrium phase relations⁷. Synthetic analogues of the 405 incompatible-element depleted Hálevjabunga and incompatible element-enriched Stapafell lavas were prepared by Neave et al.⁷ from reagent grade oxide and carbonate powders that were fused twice in Pt 406 407 crucibles placed in a muffle furnace at 1600 °C. Each fusion was performed for 1 hour, after which 408 fused powders were quenched by pouring them onto clean brass plates. Quenched glass chips were 409 then powdered in an agate disc mill to ensure that starting materials were compositionally 410 homogenous. About 300 mg of each powdered starting material was then loaded into Au₈₀Pd₂₀ 411 capsules with external and internal diameters of 2.6 and 2.2 mm, respectively, and welded shut. Each 412 capsule was ~ 2.5 mm long in order to maximise the volumes of synthetic magmas that could be 413 produced in a single IHPV experiment. Prior to being loaded with starting materials, capsules were 414 preconditioned with ~0.25 wt.% Fe to minimise Fe exchange between capsules and experimental 415 samples⁶⁷ following the procedures described by Husen et al.⁶⁸. 416 Prepared capsules were suspended from a Pt wire in the hot zone of an IHPV⁶⁹. The IHPV was then pressurised to 300 MPa with Ar and heated to 1190 °C. Pressure was monitored with a strain 417 gauge manometer and did not vary by more than 5 MPa; temperature was monitored with four 418 419 unsheathed S-type thermocouples and did not vary by more than 5 °C. Experimental temperatures 420 were approached by heating the furnace from the room temperature to 10 °C below the target 421 temperature at a rate of 50 °C/min; final heating from 1180 to 1190 °C was performed at a rate of 10 °C/min to avoid overshooting. For the magma synthesis experiments, thermal cycling (1190 \pm 422 5 °C) was applied for the first 24 hours to promote the growth of large crystals⁷⁰. Temperature was 423 424 then kept constant for the last 48 hours. Experimental products were quenched after 72 hours by 425 fusing the Pt wire on which the capsules were suspended. The capsules dropped into a cold zone at the

bottom of the vessel cooled at a rate of ~150 °C/s, which was sufficient to avoid the formation of
quench crystals.

All experiments were conducted under nominally dry conditions (no H₂O was added to the dried starting powders), which resulted in melt H₂O contents of ~0.7 wt.% following the reduction of Fe₂O₃ in the starting glasses to FeO and the inward diffusion of trace H₂ from the Ar pressure medium at high temperatures. These H₂O contents are consistent with the experiments having been run under broadly reducing conditions about one log unit above the fayalite-magnetite-quartz redox buffer (FMQ+1): melt H₂O contents of ~0.7 wt.% correspond to an a_{H_2O} of ~0.06³⁸, which is related to the $f_{O_2}^{sample}$ by the relationship $f_{O_2}^{sample} = f_{O_2}^{vessel} \cdot a_{H_2O}^2$, where $f_{O_2}^{vessel}$ is equivalent to FMQ+3.3³⁹.

435 Magma-magma reaction experiments were performed under exactly the same conditions as synthesis experiments, and in the same IHPV. The products of each synthesis experiment were cut 436 437 into four cylinders ~3.5 mm long. The ends of the products of each synthesis experiment were 438 retained to determine the phase assemblages and phase compositions present at the start of magma-439 magma reaction experiments. Magma cylinders from each synthesis experiment were then juxtaposed 440 within new $Au_{20}Pd_{20}$ capsules with the same dimensions and Fe contents as those used for synthesis 441 experiments. Each capsule was then welded shut and magma-magma reaction experiments were 442 performed in the same way as described for synthesis experiments. Capsules were oriented with 443 cylinders synthesised from the dense and FeO*-rich Stapafell lava analogue beneath those synthesised 444 from the less dense and FeO*-poor Háleyjabunga lava analogue to prevent intra-capsule convection⁴⁰. 445 Magma-magma reaction experiments were performed for durations of 1, 4, 24 and 96 hours in order 446 to capture the time-dependent nature of kinetic processes. The products of magma-magma reaction 447 experiments were cut longitudinally and mounted in resin alongside the reserved products of synthesis 448 experiments for subsequent imaging and analysis.

Analytical methods. Experimental products were imaged by field emission gun scanning electron
microscopy (FEG-SEM) on a JEOL JSM-7610F instrument in the Institut für Mineralogie of the
Leibniz Universität Hannover. Backscatter electron images (BSE) were typically collected using an

452 accelerating voltage of 15 kV and a working distance of 15 mm. BSE maps of experimental products453 were acquired using the ESPRIT software from Bruker.

The major and minor element compositions of experimental products were determined by 454 455 electron probe microanalysis (EPMA) on a Cameca SX100 instrument in the Institut für Mineralogie 456 of the Leibniz Universität Hannover. Silicon, Ti, Al, Cr, Fe, Mn, Mg, Ca, Na, K and P were measured 457 in glasses with a beam size of 12 µm, an accelerating voltage of 15 kV and a current of 10 nA. 458 Silicon, Ti, Al, Cr, Fe, Mn, Mg, Ca, Na and K were measured in minerals with a beam size of 1 µm, 459 an accelerating voltage of 15 kV and a current of 15 nA. Elements were counted on peak for 20 s, 460 with the exceptions of Si and Na that were counted on peak for 10 s to minimise detector drift and Na 461 migration, respectively. Background counting times were half on-peak counting times. The following 462 standards were used for calibration: wollastonite (Si and Ca), TiO₂ (Ti), jadeite (Al), Cr₂O₃ (Cr), 463 Fe₂O₃ (Fe), Mn₃O₄ (Mn), MgO (Mg), albite (Na), orthoclase (K) and apatite (P). The following 464 secondary standards were regularly analysed to correct for inter-session drift and to monitor accuracy 465 and precision: VG-2 basaltic glass (NMNH 111240-52; using the preferred MgO content), Kakanui augite (NMNH 122142; using preferred values), San Carlos olivine (NMNH 111312-44) and Lake 466 County plagioclase (NMNH 115900)⁷¹. Accuracy and precision were typically better than 2% and 467 468 2%, and 10% and 10% for major (>1 wt.%) and minor (<1 wt.%) elements, respectively. Typical analyses of standards are provided alongside analyses of experimental products in the Supplementary 469 470 Data.

471 Glass H₂O contents were determined in the products of the synthesis experiments and 1- and 472 96-hour magma-magma reaction experiments by Fourier-transform infrared (FTIR) spectroscopy with 473 a Bruker IFS88 instrument at the Institut für Mineralogie of the Leibniz Universität Hannover 474 following the methods described by Husen et al.⁶⁸. Estimated glass H₂O contents are provided in the 475 Supplementary Data with further information about experimental a_{H_2O} - f_{O_2} conditions.

476 Estimating effective binary diffusion coefficients. Effective binary diffusion coefficients (*D*) were
477 estimated by fitting error functions to glass composition profiles through the products of magma-

magma reaction experiments to solve Fick's 2nd Law: $C(x, t) = C_1 + \frac{C_0 - C_1}{2} \left(1 - \operatorname{erf}\left(\frac{x}{2\sqrt{Dt}}\right)\right)$, where 478 C(x, t) is the concentration in wt.% of the diffusing element C at distance x in m after time t in s and 479 C_0 and C_1 are the initial concentrations of the diffusing element on either side of the couple. Fitting 480 was performed by minimising the χ^2 misfit associated with the following function: $y_{est} =$ 481 482 $a \operatorname{erf}(b(x+c)) + d$, where y_{est} is the predicted concentration of a given oxide in wt.% and a, b, c and d are fitting parameters. The χ^2 misfit was defined as follows: $\chi^2 = \sum_{x=0}^n \left(\frac{(y_{obs} - y_{est})^2}{2\sigma}\right)$, where 483 y_{obs} is the observed concentration of a given oxide in wt.% at a distance of x, y_{est} is the predicted 484 485 concentration of a given oxide in wt.% at a distance of x and σ is the uncertainty associated with analyses of a given oxide in wt.%. Minimisations were performed with the fminsearch() function 486 implemented in the pracma package of \mathbb{R}^{72} . Element diffusivities in \mathbb{m}^2 /s were then calculated using 487 the following relationship⁴²: $D = \frac{((1/b)^2)}{4t}$, where *b* is the fitting parameter described above. 488

489 Data availability

490 EMPA data and BSE maps are provided in the Supplementary Data.

491 References

- Hofmann, A. W. & White, W. M. Mantle plumes from ancient oceanic crust. *Earth Planet*.
 Sci. Lett. 57, 421–436 (1982).
- 494 2. Gast, P. W. Trace element fractionation and the origin of tholeiitic and alkaline magma types.
 495 *Geochim. Cosmochim. Acta* 32, 1057–1086 (1968).
- 496 3. Hofmann, A. W. Mantle geochemistry: the message from oceanic volcanism. *Nature* 385,
 497 219–229 (1997).
- 498 4. Hirschmann, M. M. & Stolper, E. M. A possible role for garnet pyroxenite in the origin of the
 499 'garnet signature' in MORB. *Contrib. to Mineral. Petrol.* 124, 185–208 (1996).
- 5. Hauri, E. H. Major-element variability in the Hawaiian mantle plume. *Nature* 382, 415–419
 (1996).
- Michael, P. J. & Chase, R. L. The influence of primary magma composition, H₂O and pressure
 on mid-ocean ridge basalt differentiation. *Contrib. to Mineral. Petrol.* 96, 245–263 (1987).
- Neave, D. A., Namur, O., Shorttle, O. & Holtz, F. Magmatic evolution biases basaltic records
 of mantle chemistry towards melts from recycled sources. *Earth Planet. Sci. Lett.* 520, 199–
 211 (2019).
- 507 8. Maclennan, J. Concurrent mixing and cooling of melts under Iceland. J. Petrol. 49, 1931–1953
 508 (2008).
- 509 9. Shorttle, O. Geochemical variability in MORB controlled by concurrent mixing and
 510 crystallisation. *Earth Planet. Sci. Lett.* 424, 1–14 (2015).
- 511 10. Rubin, K. H. & Sinton, J. M. Inferences on mid-ocean ridge thermal and magmatic structure
 512 from MORB compositions. *Earth Planet. Sci. Lett.* 260, 257–276 (2007).
- 513 11. Saal, A. E., Hart, S. R., Shimizu, N., Hauri, E. H. & Layne, G. D. Pb isotopic variability in
- melt inclusions from oceanic island basalts, Polynesia. *Science* (80-.). **282**, 1481–1484 (1998).

- 515 12. Winpenny, B. & Maclennan, J. A partial record of mixing of mantle melts preserved in
 516 Icelandic phenocrysts. *J. Petrol.* 52, 1791–1812 (2011).
- 517 13. Neave, D. A., Passmore, E., Maclennan, J., Fitton, J. G. & Thordarson, T. Crystal-melt
- 518 relationships and the record of deep mixing and crystallization in the AD 1783 Laki eruption,
- 519 Iceland. J. Petrol. 54, 1661–1690 (2013).
- 520 14. Maclennan, J. Lead isotope variability in olivine-hosted melt inclusions from Iceland.
 521 *Geochim. Cosmochim. Acta* 72, 4159–4176 (2008).
- 522 15. Kuo, L. C. & Kirkpatrick, R. J. Pre-eruption history of phyric basalts from DSDP legs 45 and
 523 46: Evidence from morphology and zoning patterns in plagioclase. *Contrib. to Mineral. Petrol.*524 79, 13–27 (1982).
- 525 16. Dungan, M. A. & Rhodes, J. M. Residual Glasses and Melt Inclusions in Basalts from DSDP
 526 Legs 45 and 46: Evidence for Magma Mixing. *Contrib. to Mineral. Petrol.* 67, 417–431
 527 (1978).
- 528 17. Rhodes, J. M., Dungan, M. A., Blanchard, D. P. & Long, P. E. Magma mixing at mid-ocean
 529 ridges: evidence from basalts drilled near 22°N on the Mid-Atlantic Ridge. *Tectonophysics* 55,
 530 35–61 (1979).
- 531 18. Sobolev, A. V. & Shimizu, N. Ultra-depleted primary melt included in an olivine from the
 532 Mid-Atlantic Ridge. *Nature* 363, 151–154 (1993).
- 533 19. Michael, P. J., McDonough, W. F., Nielsen, R. L. & Cornell, W. C. Depleted melt inclusions
 534 in MORB plagioclase: Messages from the mantle or mirages from the magma chamber? *Chem.*535 *Geol.* 183, 43–61 (2002).
- 536 20. Neave, D. A., Hartley, M. E., Maclennan, J., Edmonds, M. & Thordarson, T. Volatile and light
 537 lithophile elements in high-anorthite plagioclase-hosted melt inclusions from Iceland.
 538 *Geochim. Cosmochim. Acta* 205, 100–118 (2017).
- 539 21. van Gerve, T. D., Neave, D. A., Almeev, R. R., Holtz, F. & Namur, O. Zoned crystal records

- of transcrustal magma transport, storage and differentiation: insights from the Shatsky Rise
 oceanic plateau. *J. Petrol.* 61, egaa080 (2020).
- 542 22. Neave, D. A., Maclennan, J., Hartley, M. E., Edmonds, M. & Thordarson, T. Crystal storage
 543 and transfer in basaltic systems: the Skuggafjöll eruption, Iceland. *J. Petrol.* 55, 2311–2346
 544 (2014).
- 545 23. Bennett, E. N., Lissenberg, C. J. & Cashman, K. V. The significance of plagioclase textures in
 546 mid-ocean ridge basalt (Gakkel Ridge, Arctic Ocean). *Contrib. to Mineral. Petrol.* 174, 49
 547 (2019).
- 548 24. Kahl, M., Chakraborty, S., Costa, F. & Pompilio, M. Dynamic plumbing system beneath
 549 volcanoes revealed by kinetic modeling, and the connection to monitoring data: An example
 550 from Mt. Etna. *Earth Planet. Sci. Lett.* **308**, 11–22 (2011).
- 551 25. Mutch, E. J. F., Maclennan, J., Shorttle, O., Edmonds, M. & Rudge, J. F. Rapid transcrustal
 552 magma movement under Iceland. *Nat. Geosci.* 12, 569–574 (2019).
- 553 26. Costa, F., Shea, T. & Ubide, T. Diffusion chronometry and the timescales of magmatic
 554 processes. *Nat. Rev. Earth Environ.* 1, 201–214 (2020).
- 555 27. Ubide, T. & Kamber, B. S. Volcanic crystals as time capsules of eruption history. *Nat.*556 *Commun.* 9, 326 (2018).
- 557 28. Kouchi, A. & Sunagawa, I. Mixing basaltic and dacitic magmas by forced convection. *Nature*558 304, 527–528 (1983).
- 559 29. De Campos, C. P., Dingwell, D. B. & Fehr, K. T. Decoupled convection cells from mixing
 560 experiments with alkaline melts from Phlegrean Fields. *Chem. Geol.* 213, 227–251 (2004).
- 30. Kress, V. C. & Ghiorso, M. S. Multicomponent diffusion in MgO-Al₂O₃-SiO₂ and CaO-MgOAl₂O₃-SiO₂ melts. *Geochim. Cosmochim. Acta* 57, 4453–4466 (1993).
- 563 31. González-Garcia, D. *et al.* Diffusive exchange of trace elements between alkaline melts:

- 564 Implications for element fractionation and timescale estimations during magma mixing.
 565 *Geochim. Acta* 233, 95–114 (2018).
- LaTourrette, T., Wasserburg, G. J. & Fahey, A. J. Self diffusion of Mg, Ca, Ba, Nd, Yb, Ti, Zr,
 and U in haplobasaltic melt. *Geochim. Cosmochim. Acta* 60, 1329–1340 (1996).
- 568 33. Chen, Y. & Zhang, Y. Olivine dissolution in basaltic melt. *Geochim. Cosmochim. Acta* 72, 4756–4777 (2008).
- 570 34. Chen, Y. & Zhang, Y. Clinopyroxene dissolution in basaltic melt. *Geochim. Cosmochim. Acta*571 73, 5730–5747 (2009).
- 572 35. Yu, Y., Zhang, Y., Chen, Y. & Xu, Z. Kinetics of anorthite dissolution in basaltic melt.

573 *Geochim. Cosmochim. Acta* 179, 257–274 (2016).

- 574 36. Shorttle, O. & Maclennan, J. Compositional trends of Icelandic basalts: Implications for short575 length scale lithological heterogeneity in mantle plumes. *Geochemistry, Geophys. Geosystems*576 12, 1–32 (2011).
- 577 37. Neave, D. A. & Putirka, K. D. A new clinopyroxene-liquid barometer, and implications for
 578 magma storage pressures under Icelandic rift zones. *Am. Mineral.* 102, 777–794 (2017).
- 579 38. Aranovich, L. Y. & Newton, R. C. Experimental determination of CO₂-H₂O activity-
- composition relations at 600-1000 °C and 6-14 kbar by reversed decarbonation and
 dehydration reactions. *Am. Mineral.* 84, 1319–1332 (1999).
- 58239.Botcharnikov, R. E., Koepke, J., Holtz, F., McCammon, C. & Wilke, M. The effect of water
- 583 activity on the oxidation and structural state of Fe in a ferro-basaltic melt. *Geochim*.
- 584 *Cosmochim. Acta* **69**, 5071–5085 (2005).
- 585 40. Zhang, Y., Walker, D. & Lesher, C. E. Diffusive crystal dissolution. *Contrib. to Mineral.*586 *Petrol.* 102, 492–513 (1989).
- 587 41. Barr, J. A. & Grove, T. L. AuPdFe ternary solution model and applications to understanding

- the fO₂ of hydrous, high-pressure experiments. *Contrib. to Mineral. Petrol.* 160, 631–643
 (2010).
- 590 42. Zhang, Y. Diffusion in minerals and melts: theoretical background. *Rev. Mineral.*591 *Geochemistry* 72, 5–59 (2010).
- 43. Lundstrom, C. C. An experimental investigation of the diffusive infiltration of alkalis into
 partially molten peridotite: Implications for mantle melting processes. *Geochemistry, Geophys. Geosystems* 4, (2003).
- Lowry, R. K., Henderson, P. & Nolan, J. Tracer diffusion of some alkali, alkaline-earth and
 transition element ions in a basaltic and an andesitic melt, and the implications concerning
 melt structure. *Contrib. to Mineral. Petrol.* 80, 254–261 (1982).
- 598 45. Zhang, Y., Ni, H. & Chen, Y. Diffusion data in silicate melts. *Rev. Mineral. Geochemistry* 72, 311–408 (2010).
- 600 46. Giordano, D., Russell, J. K. & Dingwell, D. B. Viscosity of magmatic liquids: A model. *Earth*601 *Planet. Sci. Lett.* 271, 123–134 (2008).
- 47. Glasstone, S., Laider, K. J. & Eyring, H. The Theory of Rate Processes. (McGraw-Hill, 1941).
- 48. Dingwell, D. B. Effects of structural relaxation on cationic tracer diffusion in silicate melts.
 604 *Chem. Geol.* 82, 209–216 (1990).
- Mungall, J. E. Empirical models relating viscosity and tracer diffusion in magmatic silicate
 melts. *Geochim. Cosmochim. Acta* 66, 125–143 (2002).
- 50. Neave, D. A. *et al.* Clinopyroxene–liquid equilibria and geothermobarometry in natural and
 experimental tholeiites: the 2014–2015 Holuhraun eruption, Iceland. *J. Petrol.* 60, 1653–1680
 (2019).
- 610 51. Cherniak, D. J. & Liang, Y. Ti diffusion in natural pyroxene. *Geochim. Cosmochim. Acta* 98,
 611 31–47 (2012).

- 612 52. Perugini, D., De Campos, C. P., Dingwell, D. B., Petrelli, M. & Poli, G. Trace element
 613 mobility during magma mixing: Preliminary experimental results. *Chem. Geol.* 256, 146–157
 614 (2008).
- 615 53. Laeger, K. *et al.* High-resolution geochemistry of volcanic ash highlights complex magma
 616 dynamics during the Eyjafjallajökull 2010 eruption. *Am. Mineral.* 102, 1173–1186 (2017).
- 617 54. Kent, A. J. R. Melt inclusions in basaltic and related volcanic rocks. *Rev. Mineral.*618 *Geochemistry* 69, 273–331 (2008).
- 55. Dohmen, R. & Chakraborty, S. Fe-Mg diffusion in olivine II: Point defect chemistry, change
 of diffusion mechanisms and a model for calculation of diffusion coefficients in natural
 olivine. *Phys. Chem. Miner.* 34, 409–430 (2007).
- 622 56. Caricchi, L. *et al.* A Data Driven Approach to Investigate the Chemical Variability of
 623 Clinopyroxenes From the 2014–2015 Holuhraun–Bárdarbunga Eruption (Iceland). *Front.*624 *Earth Sci.* 8, 1–15 (2020).
- 57. Lange, A. E., Nielsen, R. L., Tepley, F. J. & Kent, A. J. R. The petrogenesis of plagioclasephyric basalts at mid-ocean ridges. *Geochemistry, Geophys. Geosystems* 14, 3282–3296
 (2013).
- 628 58. Holness, M. B. *et al.* Textures in partially solidified crystalline nodules: A window into the
 629 pore structure of slowly cooled mafic intrusions. *J. Petrol.* 48, 1243–1264 (2007).
- 630 59. Bergantz, G. W., Schleicher, J. M. & Burgisser, A. Open-system dynamics and mixing in
 631 magma mushes. *Nat. Geosci.* 8, 793–796 (2015).
- 632 60. Halldórsson, S. A. *et al.* Isotopic-heterogeneity of the Thjorsa lava-Implications for mantle
 633 sources and crustal processes within the Eastern Rift Zone, Iceland. *Chem. Geol.* 255, 305–316
 634 (2008).
- 635 61. Lange, A. E., Nielsen, R. L., Tepley, F. J. & Kent, A. J. R. Diverse Sr isotope signatures
 636 preserved in mid-oceanic-ridge basalt plagioclase. *Geology* 41, 279–282 (2013).

- 637 62. Grove, T. L., Kinzler, R. J. & Bryan, W. B. Fractionation of Mid-Ocean Ridge Basalt
- 638 (MORB). in *Mantle Flow and Melt Generation at Mid-Ocean Ridges, Geophysical*639 *Monograph 71* 281–310 (American Geophysical Union, 1992).
- 640 63. O'Neill, H. S. C. & Jenner, F. E. The global pattern of trace-element distributions in ocean
 641 floor basalts. *Nature* 491, 698–704 (2012).
- 642 64. Costa, F., Coogan, L. A. & Chakraborty, S. The time scales of magma mixing and mingling
 643 involving primitive melts and melt-mush interaction at mid-ocean ridges. *Contrib. to Mineral.*644 *Petrol.* 159, 371–387 (2010).
- 645 65. Ridley, I. W., Perfit, M. R., Smith, M. C. & Fornari, D. J. Magmatic processes in developing
 646 oceanic crust revealed in a cumulate xenolith collected at the East Pacific Rise, 9°50'N.

647 *Geochemistry, Geophys. Geosystems* 7, Q12O04 (2006).

- 648 66. Sisson, T. W. & Grove, T. L. Experimental investigations of the role of H₂O in calc-alkaline
 649 differentiation and subduction zone magmatism. *Contrib. to Mineral. Petrol.* 113, 143–166
 650 (1993).
- 651 67. Gaetani, G. A. & Grove, T. L. The influence of water on melting of mantle peridotite. *Contrib.*652 *to Mineral. Petrol.* 131, 323–346 (1998).
- 653 68. Husen, A., Almeev, R. R. & Holtz, F. The Effect of H₂O and Pressure on Multiple Saturation
 654 and Liquid Lines of Descent in Basalt from the Shatsky Rise. *J. Petrol.* 57, 309–344 (2016).
- 655 69. Berndt, J. *et al.* A combined rapid-quench and H₂-membrane setup for internally heated
- 656 pressure vessels: Description and application for water solubility in basaltic melts. *Am.*657 *Mineral.* 87, 1717–1726 (2002).
- 658 70. Erdmann, M. & Koepke, J. Experimental temperature cycling as a powerful tool to enlarge
 659 melt pools and crystals at magma storage conditions. *Am. Mineral.* 101, 960–969 (2016).
- 660 71. Jarosewich, E., Nelen, J. A. & Norberg, J. A. Reference samples for electron microprobe
- 661 analysis. *Geostand. Newsl.* **4**, 43–47 (1980).

662 72. R Development Core Team. R: A Language and Environment for Statistical Computing. *R*663 *Found. Stat. Comput.* (2016).

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669 Author contributions

- 670 DAN designed the project. PB performed the experiments and EPMA analyses under the supervision
- of DAN and FH. HB performed the FTIR analyses. All authors contributed to data interpretation.
- 672 DAN wrote the manuscript with contributions from PB, HB and FH.



673



- 685 element enrichment: plagioclase crystallises at higher temperatures and in greater amounts from
- 686 incompatible element-depleted magmas that are correspondingly enriched in refractory elements like
- 687 CaO and Al₂O₃. Phases are labelled as follows: gl, glass; pig, pigeonite; cpx, clinopyroxene; pl,
- 688 plagioclase; and ol, olivine.



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690 Fig. 2 Backscattered electron (BSE) images of the products of synthesis experiments. Synthesis 691 experiments were performed on Háleyjabunga and Stapafell lava analogues at 300 MPa and 1190 °C. 692 These conditions were chosen to maximise differences in equilibrium phase assemblages and 693 proportions⁷. **a** Products of the synthesis experiment on the incompatible element-depleted 694 Háleyjabunga (Hál) lava analogue contain crystals of olivine (ol), clinopyroxene (cpx) and plagioclase 695 (pl) in relatively modest amounts of glass [gl; melt mass fraction (F) ~ 0.72]. **b** Products of the 696 synthesis experiment on the incompatible element-enriched Stapafell (Sta) lava analogue contain 697 crystals olivine (ol), clinopyroxene (cpx) in relatively large amounts of glass (gl; $F \sim 0.90$).





experiments. Magma-magma reaction experiments were performed by juxtaposing quenched magma

701 cylinders from synthesis experiments on both Háleyjabunga (Hál) and Stapafell (Sta) lava analogues

within new capsules. These new capsules were then subjected to exactly the same conditions used

- during synthesis experiments (300 MPa and 1190 °C). a-d BSE maps show the products of
- experiments run for 1 hour (**a**), 4 hours (**b**), 24 hours (**c**) and 96 hours (**d**). Magma cylinders
- synthesised from the incompatible element-depleted Háleyjabunga lava analogue are on the right, and
- magma cylinders synthesised from the incompatible element-enriched Stapafell lava analogue are on
- the left. Grey lines show the positions of original interfaces between juxtaposed magma cylinders.
- 708 Plagioclase (pl) is the lowest BSE intensity phase (purple, only towards the right), followed, in order
- of increasing intensity, by olivine (ol, blue), clinopyroxene (cpx, blue-green) and glass (gl, green to
- 710 yellow).



712 Fig. 4 Glass composition profiles through the products of magma-magma reaction experiments. 713 Glass composition profiles are centred on the interfaces between juxtaposed magma cylinders, which 714 are indicated by vertical grey lines. Analyses from the products of experiments with different 715 durations are shown with different colours and symbols. Solid lines show moving averages calculated 716 by applying a Gaussian filter with a 0.25 mm bandwidth to the raw analyses. Regions of the 717 experimental products where plagioclase (pl) is stable are shown with dashed vertical lines and 718 horizontal arrows. Compositions of glasses in the products of synthesis experiments are shown with 719 grey symbols. Characteristic 2σ analytical uncertainties are shown. **a-f** Glass composition profiles are shown in order of increasing element diffusivity⁴⁵: Al₂O₃ (**a**), FeO* (**b**), TiO₂ (**c**), CaO (**d**), K₂O (**e**) 720 721 and Na₂O (f). No analyses are shown for SiO₂ or MgO because along-profile compositional variability 722 does not exceed analytical uncertainty for these elements (Supplementary Data). a-c Relatively slow-723 diffusing elements typically show composition profiles with sigmoidal shapes for all experimental durations. Sigmoidal profiles are especially clear in the cases of Al_2O_3 (a) and TiO_2 (c) but obfuscated 724 by Fe-loss for experimental durations \geq 24 hours in the case of FeO* (b). **d-f** Relatively fast-diffusing 725 726 elements only show composition profiles with sigmoidal shapes for short experimental durations. This 727 is emphasised most by $Na_2O(\mathbf{f})$ for which all initial variability is erased within 24 hours.



Fig. 5 Time-normalised glass composition profiles through the products of magma-magma

730 reaction experiments. Normalising glass composition profiles by the square roots of experimental

- durations eliminates the effects of time on the evolution of diffusively controlled composition
- 732 profiles, which thus collapse onto single curves for each element^{31,45}. Symbols are the same as in Fig.
- 4. **a**, **b** Closely overlapping time-normalised glass compositions profiles for Al_2O_3 (**a**) and TiO_2 (**b**)
- confirm that diffusion in experimental melts was the dominant process controlling chemical
- variability in the products of our magma-magma reaction experiments.



737 Fig. 6 Element diffusivities in basaltic melts. Effective binary diffusion coefficients (D) were estimated by fitting error functions to glass composition profiles to solve Fick's 2nd Law, and are 738 739 summarised in 1000/T versus $\ln(D)$ space alongside published diffusivities from experiments on 740 basaltic systems^{32–35,43,44}. Diffusion coefficients estimated from our experiments are only presented for 741 element-duration combinations for which composition profiles have clearly sigmoidal forms. 742 Temperature- and viscosity-dependent Eyring diffusivities calculated with a characteristic diffusive jump length of 0.4 nm are shown as orange lines^{31,47}. Grey lines show regressions with 95% 743 744 confidence intervals through global datasets. Dashed lines show tentative regressions for elements 745 with sparse diffusion data. a Estimated Al₂O₃ diffusivities are broadly consistent with Eyring diffusivities and slightly faster than predicted from published diffusivities^{33–35,43}. **b** Estimated FeO* 746 diffusivities are similar to Eyring diffusivities and consistent with published diffusivities 35,43 c 747 748 Estimated TiO₂ diffusivities are similar to Eyring diffusivities and consistent with many published diffusivities, though published values span up to three ln units at any given temeprature^{32-35,43}. **d** 749

- 750 Estimated CaO diffusivities are slightly faster than Eyring diffusivities but appear slightly slower than
- published diffusivities $^{32-35}$. e Estimated K₂O diffusivities are a ln unit faster than Eyring diffusivities
- but define a plausible array in 1000/T versus $\ln(D)$ space with the limited number of published
- 753 diffusivities available⁴³. **f** Estimated Na₂O diffusivities are three ln units faster than Eyring
- diffusivities. They are also coherent with some published diffusivities⁴⁴, but not others³⁴.





766 contents triggered the diffusive re-equilibration of $X_{\rm Fo}$ in some crystals (one example is outlined in 767 black box). **b** Distribution of plagioclase compositions across the products of magma-magma reaction 768 experiments summarised in terms of plagioclase anorthite content $[X_{An}, where X_{An} = Ca/(Ca+Na)$ on a 769 molar basis]. The region where plagioclase is stabile contracts significantly as a function of increasing 770 experimental duration as a result of plagioclase resorption in response to diffusively driven changes in 771 melt compositions. Although X_{An} is somewhat variable, potentially as a consequence of 772 disequilibrium crystallisation during the initial phases of synthesis experiments, it does not vary 773 systematically with position or experimental duration. c Distribution of clinopyroxene compositions 774 across the products of magma-magma reaction experiments summarised in terms of clinopyroxene 775 Mg-number [Mg $\#_{cpx}$, where Mg $\#_{cpx} = Mg/(Mg+Fe)$ on a molar basis]. While much of variability in $Mg\#_{cpx}$ reflects the development of clinopyroxene sector zoning⁵⁰, mean $Mg\#_{cpx}$ contents are slightly 776 777 higher in the FeO*-poor Háleyjabunga analogue than the FeO*-rich Stapafell analogue. **d** Distribution 778 of clinopyroxene compositions across the products of magma-magma reaction experiments 779 summarised in terms of clinopyroxene TiO₂ contents. Clinopyroxene crystals from the incompatible 780 element-enriched Stapafell analogue are richer in TiO₂ than those from the incompatible element-781 depleted Háleyjabunga analogue. Moreover, clinopyroxene crystals within regions of plagioclase 782 resorption have high TiO₂ contents, suggesting that plagioclase resorption is spatially correlated with 783 the resorption and re-crystallisation of clinopyroxene, as indicated by the black arrow.



784

785 Fig. 8 Backscattered electron (BSE) image showing plagioclase resorption in the products of the

786 96-hours magma-magma reaction experiment. Many plagioclase (pl) crystals close to plagioclase

resorption fronts show resorbed, and the last remnants of resorbing plagioclase crystals are especially

- 788 irregular. Some olivine (ol) crystals close to plagioclase resorption fronts are anhedral and embayed,
- 789 whereas clinopyroxene (cpx) crystals retain euhedral to subhedral textures.



Fig. 9 Cartoon summarising how mixing-induced chemical disequilibrium creates and modifies
basaltic crystal cargoes. Primitive and incompatible element-depleted magmas crystallise high-

anorthite [high- X_{An} , where $X_{An} = Ca/(Ca+Na)$ on a molar basis] plagioclase (pl) alongside olivine (ol) and clinopyroxene (cpx) during storage in the crust^{7,62}. Even under isothermal conditions, recharge by

incompatible element-enriched and plagioclase-undersaturated primitive magmas can trigger changes

in the crystalline portions of incompatible element-depleted and plagioclase-saturated magma

reservoirs. **a** The infiltration of plagioclase-dominated mushes by plagioclase-undersaturated magmas may trigger mush disaggregation by resorbing grain boundaries, as indicated by chemical and isotopic disequilibria between some basaltic melts and their cargoes of high- X_{An} plagioclase crystals^{22,60}. **b** The

- transfer of high- X_{An} plagioclase crystals between variably incompatible element-enriched magmas
- that are also variably saturated in plagioclase produces resorption surfaces, concentric zoning,
- 802 overgrowth rims and melt inclusions^{19,20,23}. **c** Olivine and clinopyroxene crystals may re-equilibrate,

- 803 and resorb and re-crystallise, respectively, in response to isothermal mixing of variably incompatible
- 804 element-enriched magmas with different major element compositions.