Plagioclase archives of depleted melts in the oceanic crust

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ABSTRACT

Mid-ocean ridge and ocean island basalts provide vital but incomplete insights into mantle chemistry. For example, high-anorthite plagioclase is generally too refractory and incompatible-element depleted to have crystallized from the melts that carry it to the surface. Moreover, erupted basalts rarely preserve the extreme isotopic and incompatible-element depletions found in some primitive melt inclusions and residual abyssal peridotites. By integrating experimental observations with analyses of natural crystals and glasses, we infer that high-anorthite plagioclase grows from high-degree melts of refractory but otherwise unexotic mantle sources with depleted incompatible element compositions. The widespread occurrence of high-anorthite plagioclase in oceanic basalts and the oceanic crust hence indicates that refractory melts pervade the uppermost mantle and lower crust. We thus suggest that refractory melts play much a greater role in crustal accretion than typically recognized, and that refractory and feasibly depleted peridotite is more prevalent in the upper mantle than previously thought.
INTRODUCTION

Mid-ocean-ridge and ocean-island basalts (MORB and OIB respectively; oceanic basalts collectively) provide important windows into the chemistry of Earth’s mantle (Hofmann, 1997; Stracke, 2021). Over billions of years, lithospheric recycling at subduction zones has created chemically, isotopically and lithologically enriched mantle domains that are ultimately reflected in the compositions of erupted basalts (Chase, 1981). Concurrent melt extraction and crustal accretion has created refractory and depleted domains that are well documented in abyssal peridotites but rarely expressed in erupted records, reflecting the poor preservation of depleted melts (Salters et al., 2011; Byerly and Lassiter, 2014; Sanfilippo et al., 2021). Melt inclusions in primitive crystals, which are relatively resistant to mixing-induced overprinting, thus provide vital constraints on the chemical and isotopic variability of primitive melts and their mantle sources (Sobolev and Shimizu, 1993; Maclennan, 2008b, 2008a; Stracke et al., 2019). They are however challenging to analyze and remain susceptible to contamination by enriched melts in the deepest reaches of plumbing systems. Fortunately, crystals also record information about the melts from which they grow, and as volumetrically significant components of magmas and cumulates can reflect the abundance of chemically distinct melts at depth.

High-anorthite plagioclase ($X_{An} > 0.8$, where $X_{An} = \text{molar Ca/(Ca+Na+K)}$) is a major constituent of many basalts from ocean islands and slow- to intermediate-spreading mid-ocean ridges (Lange et al., 2013), as well as various cumulates from ophiolites and the lower oceanic crust (Browning, 1982; Lissenberg et al., 2013). However, such crystals are rarely in major-element equilibrium with erupted basalts (cf. Natland et al., 1983). Moreover, they are often out of isotopic and incompatible-element equilibrium with their carrier melts, implying origins from different mantle melt distributions (Halldórsson et al., 2008; Neave et al., 2014; Nielsen et al.,
High-\(X_{An}\) plagioclase cannot be reproduced in experiments on MORB starting compositions with realistic volatile contents (Grove et al., 1992; Kohut and Nielsen, 2003), and calculations with the MELTS algorithm indicate that primitive MORBs (\(\text{MgO} > 8\ \text{wt.}\%\)) typically saturate in \(X_{An} = 0.75–0.80\) plagioclase (Ghiorso and Sack, 1995; Neave et al., 2019). High-\(X_{An}\) plagioclase crystals in oceanic settings are hence thought to grow from refractory, high-\(\text{Ca#}\) melts (where \(\text{Ca#} = \text{molar Ca}/(\text{Ca+Na})\)) with high solidus and liquidus temperatures that seldom erupt (Grove et al., 1992; Neave et al., 2019). Here we integrate observations from natural and experimental systems to explore the origins of high-\(X_{An}\) plagioclase crystals in oceanic settings and discuss the role that high-\(\text{Ca#}\) melts play in oceanic magmatism.

**PLAGIOCLASE-LIQUID EQUILIBRIA**

The exchange of anorthite (An; CaAl\(_2\)Si\(_2\)O\(_8\)) and albite (Ab; NaAlSi\(_3\)O\(_8\)) components between liquid and plagioclase depends on many variables (Namur et al., 2012, and references therein). In summary, equilibrium plagioclase \(X_{An}\) correlates positively with melt \(\text{Ca#}\), melt \(\text{Al#}\) (where \(\text{Al#} = \text{molar Al}/(\text{Al+Si})\)), melt \(\text{H}_2\text{O}\) content and temperature, and negatively with pressure. The melt \(\text{H}_2\text{O}\) content of oceanic basalts is uniformly low when compared with arc basalts (≤1 wt.%; Michael, 1995), and thus exerts little influence over plagioclase \(X_{An}\). Conversely, melt \(\text{Ca#}\), which varies greatly in oceanic basalts, is inextricably linked with equilibrium \(X_{An}\) and the timing of plagioclase saturation (Bowen, 1913). For example, observations from experiments on synthetic analogues of Icelandic lavas at 300 MPa demonstrate that plagioclase saturates at higher melt \(\text{MgO}\) contents and temperatures, and with higher \(X_{An}\) contents, during the crystallization of high-\(\text{Ca#}\) primitive melts from depleted peridotite sources with refractory major element compositions than during the crystallization of low-\(\text{Ca#}\) primitive melts from modally enriched sources with more fusible compositions (Neave
et al., 2019). To place these observations within the polybaric context of crustal magmatism, we performed new crystallization experiments on the same lava analogues from Iceland at 100 and 600 MPa (high-Ca# Háleyjabunga lava, refractory source; low-Ca# Stapafell lava, fusible source; details in the Supplementary Material).

Plagioclase-liquid equilibria at 100 and 600 MPa are summarized in Fig. 1 alongside published equilibria at 300 MPa from Neave et al. (2019). The high-Ca# Háleyjabunga analogue saturates in plagioclase at higher melt MgO contents (and temperatures) than the low-Ca# Stapafell analogue. While isobaric differences in plagioclase saturation between the two starting compositions reflect mantle-derived variability in melt Ca# and Al#, polybaric differences reflect differences in plagioclase and clinopyroxene stability, with plagioclase generally saturating at lower temperatures (and melt MgO contents) when clinopyroxene stability is greater at higher pressures (Fig. 1). Equilibrium plagioclase $X_{An}$ also correlates negatively with clinopyroxene stability and pressure. Overall, melt composition, which correlates with pressure and temperature as well as source composition, exerts the main control over $X_{An}$, with high-$X_{An}$ plagioclase only crystallizing from the high-Ca# Háleyjabunga analogue (up to $X_{An} = 0.88$ and 0.85 in the products of 100 and 300 MPa experiments, respectively; the highest $X_{An}$ crystallized from low-Ca# Stapafell analogue was 0.71). Importantly, our new experiments demonstrate that high-$X_{An}$ plagioclase can be produced from refractory oceanic basalts under realistic pressure and temperature conditions (cf. Grove et al., 1992; Kohut and Nielsen, 2003). Even if the composition of Háleyjabunga lava is at the limit of erupted variability (Fig. 3), our findings thus suggest high-$X_{An}$ plagioclase crystals reflect the crystallization of refractory but essentially unexotic melts derived from refractory mantle sources that experienced high degrees of prior melt extraction, whether recent or ancient.
PREDICTING PLAGIOCLASE-LIQUID EQUILIBRIA

By predicting equilibrium plagioclase $X_{An}$ as a function of melt composition it is possible to evaluate plagioclase-liquid equilibria in diverse natural systems. While thermodynamic models allow phase relations to be extrapolated across wide parameter spaces (Ghiorso and Sack, 1995; Holland et al., 2018), empirical models can be more precise when applied within their calibration ranges (e.g., Namur et al., 2012). Moreover, it is possible to avoid introducing erroneous assumptions about crystallization conditions by predicting equilibrium $X_{An}$ from melt compositions alone; pressure and temperature are implicit in melt compositions.

Performing multiple linear regression through calibration data ($n = 98$) from experimental studies on basalts from mid-ocean ridges, an oceanic plateau and an ocean island yields the following relationship between plagioclase $X_{An}$ and melt composition (Fig. 2a; $r^2 = 0.88$; standard error = 0.03; details and sources in the Supplementary Material):

$$X_{An} = 0.92(0.07) \cdot Ca\#_{melt} + 1.63(0.24) \cdot Al\#_{melt} + 0.24(0.05) \cdot \text{(molar Ca/Al)}_{melt} - 0.54(0.06)$$ (1)

Test data ($n = 36$) from experimental studies on basalts from mid-ocean ridges and a continental hotspot with $X_{An} \sim 0.6–0.9$ are reproduced well by equation 1 ($r^2 = 0.92$; standard error = 0.02), albeit with a slight offset to lower $X_{An}$, possibly because of Na loss during some experiments (Fig. 2b; sources in the Supplementary Material).

ORIGINS OF HIGH-ANORTHITE PLAGIOCLASE

High-$X_{An}$ plagioclase has been described in lavas from many mid-ocean ridge segments and ocean islands (e.g., Lange et al., 2013). Here we apply our model to published Icelandic and MORB glass compositions, though our findings are likely to apply in other settings erupting H$_2$O-poor basalts. Equilibrium plagioclase $X_{An}$ predicted from Icelandic ($n = 190$) and MORB ($n = 1687$) glass compositions collated by Shorttle and Maclellan (2011) and Gale et al. (2013),
respectively, are shown in Fig. 3; results were filtered for plagioclase saturation using a stability
criterion from Gale et al. (2014) (details in the Supplementary Material).

Predicted plagioclase $X_{An}$ correlates with melt MgO content and Ca# in Icelandic and
MORB datasets (Figs. 3A and 3B), though high-Ca# melts occur across a wide range of melt
MgO contents (8–11 wt.%). Crucially, some glasses from both datasets return stable high-$X_{An}$
compositions ($n = 33$ and $22$, respectively). Although these glasses are at the limit of natural
variability in the case of MORB, their occurrence nonetheless substantiates rare observations of
equilibrium high-$X_{An}$ in nature ($X_{An}$ up to 0.89; Natland et al., 1983). Icelandic glasses return
higher maximum $X_{An}$ contents than MORB glasses (up to $X_{An} = 0.89$ and 0.85, respectively),
likely reflecting differences in sampling density and mantle melting conditions.

As well as being associated with high melt Ca# at any given melt MgO content (Figs. 3A
and 3B), high-$X_{An}$ plagioclase is typically associated with low melt K$_2$O contents (often
$<0.1$ wt.%; Figs. 3C and 3D), recapitulating the incompatible-element-depleted character of
erupted high-$X_{An}$ plagioclase (Neave et al., 2014; Nielsen et al., 2020). Such high-Ca#, low-K$_2$O
melts are commonly generated by shallow melting of refractory peridotites that have experienced
high degrees of prior melting (e.g., Grove et al., 1992; Shorttle and Maclellan, 2011). High-$X_{An}$
plagioclase is also associated with low melt FeO* contents (total Fe as FeO) at any given melt
MgO content (Figs 3E and 3F). This is particularly clear for Iceland, where low-FeO* primitive
melts (FeO* $< 10$ wt.%) from relatively refractory peridotites are predicted to be equilibrium in
with high-$X_{An}$ plagioclase but high-FeO* primitive melts (FeO* $> 10$ wt.%) from modally
enriched sources (pyroxenites sensu lato) are not predicted to be in equilibrium with plagioclase
of any composition (Fig. 3E). Refractory melts capable of crystallizing high-$X_{An}$ plagioclase thus
do not mix substantially with melts from modally enriched sources during ascent and are derived
from dominantly peridotitic sources (Shorttle and Maclennan, 2011); rare K$_2$O-rich melts in
equilibrium with high-$X_{An}$ plagioclase may have interacted with depleted harzburgites (Fig. 3D;
Nielsen et al., 2020). We hence argue that high-$X_{An}$ plagioclase crystals are the solid products of
melts from sources that have experienced high degrees of prior melting. In some cases, isotopic
depletions suggest that this melt extraction was ancient (Halldórsson et al., 2008; Nielsen et al.,
2020). Indeed, we speculate that high-$X_{An}$ plagioclase may be associated with ultra-depleted
sources recorded by some melt inclusions and abyssal peridotites (Sobolev and Shimizu, 1993;
Salters et al., 2011; Stracke et al., 2019), though closer integration of isotopic and major-element
observations is required to test this further.

**WIDESPREAD REFRACTORY MELTS AT DEPTH**

High-$X_{An}$ plagioclase occurs throughout the oceanic realm (Fig. 4). In Iceland, it is
especially well documented in the Eastern Volcanic Zone (Fig. 4A), where isotopically and
incompatible-element-depleted high-$X_{An}$ plagioclase may constitute $>30$ vol.% of lavas with
comparatively enriched isotopic and incompatible-element compositions (Halldórsson et al.,
2008). High-$X_{An}$ plagioclase has also been described in depleted picrites from Iceland’s Northern
Volcanic Zone of Iceland, as well as in diverse basalts from Galápagos, Réunion and Kerguelen
(Fig. 4A). Overall, these observations suggest that refractory melts with depleted incompatible-
element compositions are more prevalent beneath ocean islands than the incompatible-element
enriched character of OIBs would otherwise imply (e.g., Stracke et al., 2019).

High-$X_{An}$ plagioclase crystals have been widely reported in plagioclase-rich MORBs
from slow- to intermediate-spreading mid-ocean ridges (Fig. 4B; Lange et al., 2013; Nielsen et
al., 2020). With a few notable exceptions (Fig. 4B; Natland et al., 1983), these high-$X_{An}$ contents
are found in crystal cores surrounded by more evolved rims and are out of equilibrium with their
carrier melts, mirroring observations from ocean islands that high-Ca# melts from refractory
sources are probably more widespread at depth than suggested by erupted archives. Indeed, melt
inclusion entrapment pressures typically place high-$X_{An}$ plagioclase crystallization at or below
the Moho (Drignon et al., 2018), preventing the eruption of such plagioclase in many settings.
Recent experiments have also suggested that phase relations at >500 MPa may play a key role in
generating $X_{An} \sim 0.9$ plagioclase (Ustunisik et al., 2022).

High-$X_{An}$ plagioclase crystals are found in lower crustal sections of the fast-spreading
Samail ophiolite in Oman (Fig. 4C; Browning, 1982), though elevated melt H$_2$O contents likely
modify plagioclase-liquid equilibria in ophiolitic settings (Koepke et al., 2021). Despite its
relative scarcity, perhaps as a result of melt rock-reaction (Sanfilippo et al., 2020), $X_{An} \sim 0.8$
plagioclase does occur in crustal samples from the fast-spreading East Pacific Rise, slow-
spreading Mid-Atlantic Ridge and very slow-spreading Southwest Indian Ridge (Fig. 4D; Dick
et al., 2002; Lissenberg et al., 2013; Sanfilippo et al., 2019), suggesting that high-Ca# melts
occur and contribute to crustal accretion throughout the global ridge system. Observations from
the East Pacific Rise also illustrate how magmatic processes bias records of chemical variability,
with high-$X_{An}$ present at depth but absent from seafloor lavas as a consequence of filtering by
axial melt lenses.

In line with observations from melt inclusions and abyssal peridotites, we argue that the
global occurrence of high-$X_{An}$ plagioclase demonstrates that refractory and incompatible
element-depleted melts occur widely in the uppermost mantle and lower crust. Two key
implications of this are that melts from refractory mantle sources are likely to play a greater role
in crustal accretion than currently recognized and that refractory (and feasibly depleted) domains
may be more prevalent in the upper mantle than previously thought.
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Figure 1. Plagioclase-liquid equilibria in synthetic analogues of the high-Ca# Háleyjabunga (Hál) and low-Ca# Stapafell (Sta) lavas from Iceland (where Ca# = molar Ca/(Ca+Na)) summarized in terms of glass MgO content versus (A) glass Ca# and (B) glass molar Ca/Al. New results at 100 and 600 MPa are presented alongside results at 300 MPa from Neave et al. (2019). Plagioclase crystallizes earlier and with a higher anorthite content ($X_{An}$, where $X_{An} = \text{molar Ca/(Ca+Na+K)}$) from high-Ca# melts. Vectors show the crystallization of 5 wt.% olivine (ol), clinopyroxene (cpx) and plagioclase (plag) according to Danyushevsky (2001).
Figure 2. (A) Performance of the multiple linear regression (Eq 1) used to predict plagioclase (plag) anorthite content ($X_{An}$) as a function melt composition. The black line shows a regression through calibration data from experimental studies on basalts from an ocean island, mid-ocean ridges and an oceanic plateau. Dark and pale grey envelopes show 95% confidence and prediction intervals, respectively. (B) Performance of Eq 1 on test data from mid-ocean ridges and a continental hotspot. The dashed black line and dark grey envelope show a regression through the test data and its 95% confidence interval, respectively; the pale grey envelope shows the 95% prediction interval of Eq 1. Details and sources in the Supplementary Material.
Figure 3. Plagioclase (plag) anorthite contents ($X_{An}$) predicted to be in equilibrium with Icelandic (A, C and E) and MORB (B, D and F) glasses from Shorttle and Maclellan (2011) and Gale et al. (2013), respectively. The evolution of glass compositions is shown for elements emphasizing fractionation (MgO), degree of source enrichment ($K_2O$ and $FeO^*$) and control on plagioclase-liquid equilibria (Ca#). Glass compositions that fail a plagioclase stability criterion from Gale et al. (2014) are transparent and those in equilibrium with high-$X_{An}$ plagioclase ($X_{An} > 0.8$) are outlined in black; glasses from Háleyjabunga (Hál) and Stapafell (Sta) are highlighted.
Figure 4. Kernel density estimates summarizing how plagioclase (plag) anorthite contents ($X_{An}$) predicted to be in equilibrium with (A) Icelandic and (B) MORB glasses from Shorttle and Maclennan (2011) and Gale et al. (2013), respectively, compare with natural plagioclase compositions from (A) ocean island basalts, (B) mid-ocean ridge basalts (MORB), (C) the Samail ophiolite, Oman, and (D) the oceanic crust (OCCs, oceanic core complexes). Rare high-$X_{An}$ rims from MORB are highlighted (Natland et al., 1983; N83). Sources in the Supplementary Material.