Fractional crystallization causes the iron isotope contrast between mid-ocean ridge basalts and abyssal peridotites

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The iron isotope contrast between mid-ocean ridge basalts and abyssal peridotites is far greater than can be explained by mantle melting alone. Here we investigate a suite of mid-ocean ridge magma chamber rocks sampled by the Ocean Drilling Project Hole 735B in the Atlantis Bank of the Indian Ocean. We report major and trace element geochemistry from these rocks and measure their iron isotope compositions to investigate the potential role of fractional crystallization during melt evolution. We observe a large range of δ⁵⁶Fe that defines a significant inverse curvilinear correlation with bulk rock MgO/FeOT. These data confirm that δ⁵⁶Fe in the melt increases as fractional crystallization proceeds but, contrary to expectation, δ⁵⁶Fe continues to increase even when oxides begin to crystallize. We conclude that iron isotope fractionation through fractional crystallization during the evolution of mid-ocean ridge basalts from abyssal peridotites reconciles the disparity in isotopic compositions between these two lithologies.
ur knowledge of the chemical and isotopic compositions of the Earth’s upper mantle comes from the study of mantle melting products at ocean ridges. Mid-ocean ridge basalts (MORB) and abyssal peridotites (AP) are such two products. MORB represent the melt that solidifies to form the ocean crust whereas AP are the residue accreting new growth of the lithospheric mantle. Studies of the two products are expected to reveal consistent information, but if not, some hidden processes must be at work and need understanding. For example, it is considered that MORB have a uniform iron (Fe) isotope composition of $\delta^{56}$Fe = $+0.105 \pm 0.006\%o$ (2 SD/$n$, $n = 43$) irrespective of the extent of mantle melting and magma differentiation. On the other hand, AP have a mean $\delta^{56}$Fe value of $+0.010 \pm 0.014\%o$ (2 SD/$n$, $n = 37$)2, which is indistinguishable from chondrites3,4. A contrast of $\sim 0.1\%$ exists between MORB and AP. Partial melting2,5–7 and fractional crystallization8,9 have been proposed to explain this contrast. Recent studies suggested that the partial melting process alone cannot explain this contrast8,10.

As the erupted MORB are not primary mantle melts, but final products of primary melts that have evolved primarily through varying extent of fractional crystallization in the deep crust. We thus hypothesize that fractional crystallization-dominated MORB melt evolution is the very process that produces elevated $\delta^{56}$Fe values of sampled MORB melts. Investigation of a suite of source homogeneous MORB melts with well-defined liquid lines of descent (LLDs) will be useful9, but MORB are a homogenized mix of variably evolved melts through open-system magma chamber processes11–15 with important details averaged out. Therefore, magma chamber rocks of cumulate origin, once solidified, are largely isolated from subsequent magma chamber processes (i.e., replenishment, crystallization, mixing, and eruption), and thus record in great fidelity of magma chamber processes on mineral scales. In order to better understand the effect of fractional crystallization on MORB Fe isotope variation, we choose to study magma chamber rocks preserved in the lower ocean crust to test this hypothesis. The Ocean Drilling Project (ODP) Hole 735B is so far the only long in situ section of the lower ocean crust ever drilled16. These core samples have been thoroughly studied and well-characterized to record details of MORB melt evolution dominated by fractional crystallization17–20.

The ODP Hole 735B (32°43′S, 57°17′E) is located on the Atlantis Bank, a wave-cut platform on the east side of the Atlantis II Fracture Zone, ~93 km south of the present-day Southwest Indian Ocean Ridge (SWIR) axis (Fig. 1). It has a total penetration of 1508 m below the seafloor (mbst) drilled during Leg 118 and Leg 176. The SWIR separates the African and Antarctic plates with a half spreading rate of ~8 mm/year21, which is classified as a slow- and ultraslow-spreading ridge22. The Hole 735B drill cores are dominated by gabbros and gabbrroic rocks crosscut by minor felsic veins16.

The large and systematic Fe isotope variation of cumulate gabbros with varying modal mineralogy and highly evolved felsic veins from Hole 735B proves the prediction and argument that the magma chamber rocks record details of Fe isotope fractionation in response to MORB melt evolution dominated by fractional crystallization.

Results

Samples. The gabbroic rocks (including troctolite, olivine gabbro, gabbro, oxide gabbro, and gabbronorite) are mineralogically dominated by plagioclase and clinopyroxene with a varying abundance of olivine and Fe–Ti oxides (Supplementary Fig. 1a–c). Most of the felsic veins are quartz diorite with minor diorite, trondhjemite, and tonalite17,20, mainly consisting of plagioclase, quartz, and amphibole with varying amounts of Fe–Ti oxides (Supplementary Fig. 1d). The Fe–Ti oxide minerals are ilmenite (Supplementary Fig. 1e) in all the samples but FV62-15 where magnetite occurs as intergrowth with ilmenite (Supplementary Fig. 1f).

We selected a set of 20 representative samples (Supplementary Data 1) covering a compositional spectrum of gabbroic rocks and felsic veins (Supplementary Figs. 1 and 2) from the drill cores and analyzed Fe isotope compositions of bulk rock samples and major Fe-bearing mineral separates (olivine, pyroxene, amphibole, and Fe–Ti oxides) of some larger samples. No plagioclase was analyzed because it has low FeO17 (average of 0.23 wt%17) and contributes little to the bulk rock $\delta^{56}$Fe compositions (see “Methods” section) although it is the most abundant mineral in the drill cores. Samples used in this study have been fully characterized for their mineralogy, and major and trace element compositions17,20.

Fe isotope compositions of ODP Hole 735B samples. Bulk rock Fe isotope, major and trace element compositions for the studied

![Fig. 1 Location of the ODP Hole 735B.](https://example-image-url.com)

**Fig. 1 Location of the ODP Hole 735B.** a Bathymetric map of the Southwest Indian Ridge showing the location of ODP Hole 735B. The map is made using Generic Mapping Tools (GMT45). Topography was taken from ETOP01 1 arc-minute Global Relief Model (https://doi.org/10.7289/V5C8276M). CIR, SWIR, and SEIR refer to Central, Southwest, and Southwest Indian Ridge; NER, Ninety East Ridge; A II, Atlantis II Fracture Zone; GA, Gallieni Fracture Zone; IN, Indomied Fracture Zone; D, Discovery II Fracture Zone. b Three-dimensional topography of the Atlantis Bank is made using the topography data of study46.
samples are given in Supplementary Data 1 and 2. The bulk rock Fe isotope compositions with a large range ($\delta^{56}\text{Fe} = -0.020\pm0.252\%$) show weak/no correlation with most elements (e.g., SiO$_2$, CaO, Na$_2$O, Ni, Y, and rare earth elements, with correlation coefficients $|R| < 0.5$, Supplementary Fig. 3), but strong negative correlation with Mg$^+$ ($R^2 = 0.72$, at >99% confidence level) (Supplementary Fig. 3d) and MgO/FeOT ($R^2 = 0.72$, at >99% confidence level) (Fig. 2b). In this study, we used MgO/FeOT instead of Mg#/ ($[\text{Mg}/(\text{Mg}+\text{Fe}^2+\text{Ti})]$ for discussion to avoid arbitrary assumptions on Fe$^{2+}$/[Fe$^{2+}+\text{Fe}^3+$] for different rock types and samples, so that all these samples/data (MORB, gabbroic rocks, felsic veins, and AP) can be compared. Most of the mineral separates also show a similar negative correlation between $\delta^{56}\text{Fe}$ and MgO/FeOT just like the bulk rock (fall in 95% confidence intervals, Fig. 2b). Compared to Fe-Ti oxides and amphibole, the $\delta^{56}\text{Fe}$ of olivine and pyroxene are closer to those of the bulk rock (Supplementary Fig. 4).

**Discussion**

Previous studies have shown that most of the gabbros are cumulates with bulk compositions determined largely by modal proportions of clinopyroxene and plagioclase with varying small amounts of trapped melt$^{17}$, and that the felsic veins are mixtures of residual melt with incompletely segregated crystals$^{20}$. Unlike incompatible trace elements, iron is a major element in most rocks. It controls and contributes to the phase equilibria, and only significant mass transfer processes are able to shift its isotope composition$^8$. Though some evidence shows that the reactive porous flow may occur in gabbros of the Hole 735B$^{23-25}$, its low volume$^{24,26}$ hardly affects the Fe isotope composition of the gabbros. Therefore, the strong negative correlation of bulk rock $\delta^{56}\text{Fe}$ with MgO/FeOT defined by the gabbroic samples with varying modal mineralogy illustrates significant Fe isotope fractionation during MORB melt evolution dominated by fractional crystallization (Fig. 2b). Using reasonable mineral-melt isotope fractionation factors (see Supplementary Table 1 and “Methods” section)$^{27}$, we can model the Fe isotope composition of melts in equilibrium with these gabbroic samples (Fig. 2a). The result confirms the notion that fractional crystallization of olivine and clinopyroxene with lighter Fe isotope can elevate $\delta^{56}\text{Fe}$ of the residual melt$^{9,28,29}$. The progressive crystallization of olivine and clinopyroxene also increases FeOT and TiO$_2$, but decreases MgO and MgO/FeOT in the residual melt (Figs. 2 and 3) until ilmenite (ilmenite-dominated Fe-Ti solid solutions) begin to crystallize, upon which SiO$_2$ increases rapidly while TiO$_2$ declines in the residual melt (Fig. 3a-c). It is important to note that $\delta^{56}\text{Fe}$ continues to increase in the residual melt throughout the crystallization sequence (Fig. 3d). Mixing between incompletely segregated crystals and residual melt can produce intermediate MgO/FeOT and $\delta^{56}\text{Fe}$ compositions that lie between the compositions of the residual melt and cumulate (Fig. 2b). This is particularly obvious for the three highly evolved FV samples having high SiO$_2$ contents and incompletely segregated ilmenite (Supplementary Fig. 1d) with low $\delta^{56}\text{Fe}$.

The Fe-Ti oxides are compositionally uniform and rapidly cooled liquidus phases$^{20}$, which differ from slowly cooled exsolution aggregates in mafic liquid intrusion$^{30,31}$. Importantly, the Fe-Ti oxides are almost pure ilmenite$^{32}$ (Fe-Ti solid solutions with 91% molar ilmenite, Supplementary Fig. 1e and Data 3), which has lighter Fe than the equilibrium melt with $\Delta^{56}\text{Fe}_{\text{ilmenite-melt}} = -0.07\%$ (see “Methods” section and Supplementary Table 1). Magnetite is rare and is only found in one sample (FV62-15) as intergrowth with ilmenite (molar magnetite: ilmenite ratio of 7:3, Supplementary Fig. 1f), which would have a slightly heavier Fe than the melt with net $\Delta^{56}\text{Fe}_{\text{Fe-Ti oxides-melt}} = 0.08\%$ if $\Delta^{56}\text{Fe}_{\text{magnetite-melt}} = 0.11\%$ is applied (see Eqs. (6–8) in “Methods” section and Supplementary Table 1). The melt in equilibrium with these Fe-Ti oxides, which represents the very late-stage product of mid-ocean ridge basaltic magma evolution$^{20}$, would have $\delta^{56}\text{Fe} = +0.24\pm0.02$‰ (see “Methods” section and Supplementary Data 4). Therefore, the $\delta^{56}\text{Fe}$ of residual melt continues to increase with fractional crystallization of olivine, plagioclase, clinopyroxene, and ilmenite. It is important to note that despite the decreasing FeO and TiO$_2$ and increasing SiO$_2$ in the residual melt as the result of oxide...
crystallization, the $\delta^{56}$Fe of the residual will not decrease, but increase, because the Fe–Ti oxide is ilmenite (TiFeO$_3$) dominated solid solution (Fig. 3; see above), which differs from the conclusion based on the study of variably evolved MORB melts by assuming liquidus oxides of titanomagnetite. Crystallization of magnetite (FeO·Fe$_2$O$_3$) dominated solid solution with heavier Fe will result in $\delta^{56}$Fe decrease in the residual melt. The continued increase of $\delta^{56}$Fe with decreasing MgO/FeO$^+$ (Fig. 2b) is thus expected because liquidus oxides in Hole 735B are essentially all ilmenite or ilmenite-dominated solid solutions (see above). This new observation is also consistent with the modeling that it is ilmenite, not magnetite, that is on the liquidus at the oxygen fugacity quartz-fayalite-magnetite (QFM) buffer most appropriate for MORB. Magnetite is expected to crystallize at lower temperatures when the MORB melt
is about to solidify or the melt may be brought back up to crystallize gabbro or even troctolite by the replenishment of a new batch of melt in open-system magma chambers. The data and observations thus indicate that MORB melt evolution will rarely reach the condition of magnetite crystallization. Consequently, MORB melt evolution will be accompanied by continued $\delta^{56}\text{Fe}$ increase, not decrease, throughout the entire history of MORB melt evolution (Figs. 3d and 4c). This is an important finding. In this context, we predict that basaltic melts with high water contents and higher oxygen fugacity (higher than QFM) may facilitate earlier
crystallization of magnetite and associated δ⁵⁶Fe decrease in the residual melt (e.g., subduction-zone magmatism). The effect of oxygen fugacity on Fe isotope fractionation has been speculated in the literature.

To better illustrate Fe isotope fractionation during fractional crystallization of MORB melt, we modeled the LLDS using Petrolog33 (see “Methods” section), showing that the δ⁵⁶Fe of the cophesicating minerals and residual melt increases in response to continued cooling and crystallization of the MORB melt (Fig. 3) as our data (Fig. 2b) show in support of a recent study. We can thus conclude that Fe isotope fractionation continues throughout MORB melt evolution, and the lower ocean crust cumulates record this process in great detail.

Bulk rock gabbro and felsic vein samples have varying δ⁵⁶Fe values as low as those of AP and as high as and even higher than those of MORB (Fig. 2). By assuming primary MORB melt representing 5–15% melting of a mantle with δ⁵⁶Fe = +0.02 ± 0.03‰35,36, we obtain the Moho-crossing melt (ultimately solidified to form the bulk ocean crust) with δ⁵⁶Fe of +0.05 ± 0.03‰ (Figs. 3d, 4 and Supplementary Table 2). This confirms the previous study that the Fe isotope composition of primary MORB melt would be only slightly heavier than the mantle source35–38, and the mantle melting residues would be slightly lighter2. It is worth mentioning that AP are not simple melting residues but have excess olivine and incompatible elements35–38 added during melt ascent through the advanced residues atop the mantle, causing AP to have large compositional heterogeneity on hand specimen scales38, which explains the large δ⁵⁶Fe variation (Fig. 2c), but the mean composition is arguably significant. With mantle source heterogeneity considered39,40 the Moho-crossing melt must also vary, but the mean value of δ⁵⁶Fe of +0.05 ± 0.03‰ remains the logical and reasonable approximation for primary MORB melt (Fig. 3d). The 20–80% fractional crystallization of such primary MORB melt can effectively explain the δ⁵⁶Fe range of MORB (Fig. 3d). Sequential fractional crystallization of olivine, pyroxene, and ilmenite with lighter Fe (lower δ⁵⁶Fe) results in a progressive increase of δ⁵⁶Fe in the residual melt, in the cophesicating minerals in equilibrium with the evolving melt and in the bulk cumulate. The bulk cumulate with significantly lower δ⁵⁶Fe characterizes the lower ocean crust (Figs. 3 and 4).

Note that, by mass balance, the elevated δ⁵⁶Fe of the sampled MORB melts is complemented by the low δ⁵⁶Fe of the lower crust cumulate rocks (Fig. 3d). Therefore, fractional crystallization of MORB melt in the deep ocean crust results in the Fe isotope contrast between MORB and AP. We should also note some subtleties. The lower ocean crust made up of cumulate rocks must have δ⁵⁶Fe lower than the δ⁵⁶Fe of Moho-crossing melt, but it must be heterogeneous because of varying amount of trapped melt and locally highly evolved felsic vein lithologies (Fig. 2b), leading to some lower crustal samples having somewhat higher δ⁵⁶Fe than the model bulk cumulate (Fig. 3d). By assuming the ocean crust comprising 40% MORB melts (lavas + dikes) and 60% cumulate lower crust, we have approximately δ⁵⁶Fe [BULK OCEAN CRUST] = δ⁵⁶Fe [PRIMARY MORB MELT] = 40% δ⁵⁶Fe [AVERAGE MORB] + 60% δ⁵⁶Fe [LOWER CRUST CUMULATE] = +0.05 ± 0.03‰.

In sum, the data (Fig. 2) and quantitative understanding (Fig. 3) demonstrate that MORB melts periodically erupted from magma chambers of the varying extent of fractional crystallization have variably higher δ⁵⁶Fe than that of the Moho-crossing primary mantle melt, which explains in simple clarity that fractional crystallization causes the Fe isotope contrast between MORB and AP (Fig. 4).

Methods

Fe isotope analysis. The studied samples were selected from the drill cores during Leg 17637. Sample information is given in Supplementary Data 1. Analytical methods, standards, and data for major and trace elements have been detailed17,20.

All suspicious surface contaminants such as pen marks, saw marks, and sticker residues were thoroughly removed. The samples were then reduced to 1–2 cm size fragments and ultrasonically cleaned in Milli-Q water before being dried and ground into powder using an agate mill in a clean environment. Some larger samples were crushed into 40–100 mesh for handpicking mineral separates under a binocular. Major Fe-bearing minerals (e.g., olivine, pyroxene, amphibole, and Fe–Ti oxides) were separated for Fe isotope analysis (Supplementary Fig. 4). Plagioclase and quartz were not analyzed. Mineral separates were cleaned in Milli-Q water for 10 min, at least three times, in an ultrasonic bath. The iron isotope analysis was done in the Laboratory of Ocean Lithosphere and Mantle Dynamics, Institute of Oceanology, Chinese Academy of Sciences. About 5–20 mg of the iron separated residues were dissolved in distilled HNO₃ + HCl + HF mixture, and a 10 μl PFA Teflon beaker at 190 °C for 15 h, and then re-dissolved, after evaporation, with distilled 3 N HNO₃ for 2 h until complete dissolution. Iron was purified with a polypropylene column filled with 1 ml AG-MP-1 resin (200–400 mesh) in a 9 N HCl medium following the procedure41, which was improved upon previous methods42,43. Iron was collected using 1.5 ml 1 N HCl. The total procedural blank for these samples is 32 ng, which is negligible compared to the amount of material processed. The purified solutions were doped with GSB Ni standard (an ultrapure single elemental standard solution from the China Iron and Steel Research Institute) as an internal mass bias monitor with Ni: Fe ratio of ~1:4.145.

The iron isotope compositions were determined using Nu plasma II multiple collector inductively coupled plasma mass spectrometer (MC-ICP-MS) in wet plasma mode with medium resolution (a mass resolution >7500). Reference material GSB Fe standard (a substitution of IRMM-014: δ⁵⁶FeIRRM-014 = δ⁵⁶FeGSB + 0.729, δ⁵⁶FeIRRM-014 – δ⁵⁶FeGSB + 1.073‰) was used for bracketing each sample. Iron isotope compositions are reported as δ-notation relative to the international standard of IRMM-014: δ⁵⁶Fe(‰) = [(Fe⁵⁶/Fe⁵⁴)sample / (Fe⁵⁶/Fe⁵⁴)IRMM-014] × 1000, where i refers to mass 56 or 57. Our analyzed δ⁵⁶Fe values for USGS standards agree well with recommended values in the literature44,45; GSP-2 (0.14 ± 0.05‰; 2 SD, n = 12), BCR-2 (0.05 ± 0.07‰; 2 SD, n = 8), AGV-2 (0.09 ± 0.07‰; 2 SD, n = 12) and BHVO-2 (0.13 ± 0.07‰; 2 SD, n = 12).

Mineral composition analysis. Major and minor element compositions of olivine, clinopyroxene, amphibole, and Fe–Ti oxides were analyzed on polished thin sections using a JEOL Electron Probe micro analyzer (EPMA 8230) at the Laboratory of regional geology and resources research institute of Hebei Province. An accelerating voltage of 15 kV, a beam current of 20 nA, and a beam diameter of 1 μm were employed. For major elements, the analytical precision is better than 2%. The data are given in Supplementary Data 3.

Calculation of Δ⁵⁶Fe mineral−melt. The theory states that the heavy Fe isotope is preferentially partitioned into the phase with the highest bond strength or bond stiffness27,46. Ferric iron (Fe³⁺) is predicted to have heavier Fe (i.e., higher δ⁵⁶Fe) because the smaller ionic radii and higher valence state of Fe results in Fe featuring stronger bond10. Fe isotope can fractionate when iron incorporates into different phases, leading to different δ⁵⁶Fe between different phases. The fractionation between crystallizing mineral and melt is expressed as Δ⁵⁶Fe mineral−melt (δ⁵⁶Fe mineral − δ⁵⁶Fe melt), which is inversely proportional to temperature and is calculated using the following Eq. (1):47

\[ Δ^{56}\text{Fe}_{\text{mineral−melt}} = \frac{290}{T} \left( \frac{K_{\text{mineral}} - K_{\text{bulk}}}{T^2} \right), \]

where K is the force constant and T is the temperature in Kelvin. The force constants of chromite, olivine, plagioclase, pyroxene, ilmenite, magnetite, and melt are given in Supplementary Table 1.

The coordination and valence state of Fe are the two very factors that control the bond strength and affect the Fe isotope fractionation10,27,46. According to previous studies10,27,47,49, Fe²⁺ and Fe³⁺ are in V- or VI-fold average coordination in the basaltic melt, and in IV-fold average coordination in the rhyolitic melt, so the force constant of the rhyolitic melt is higher than in basaltic melt at given Fe³⁺ / ΣFe. In this study, we focus on the force constant of MORB evolution, Fe³⁺ / ΣFe increases because Fe³⁺ (vs. Fe²⁺) behaves as a slightly "incompatible" element33 until magnetite (Fe³⁺ O four Fe²⁺ O₄) may begin to crystallize. However, the liquidus oxides in Hole 735B are ilmenite (TiFe²⁺O₄) and ilmenite-dominated solid solutions (nearly 91%), which means that the Fe²⁺/ΣFe of MORB melt is low and hardly reaches magnetite saturation. Hence, we apply a constant force constant (233 ± 17 N/m) by assuming melt Fe²⁺/ΣFe = 0.16 following the previous study10 for melt during MORB evolution.

Force constants for minerals (Supplementary Table 1) are taken from the literature26,27; except for plagioclase. With no experimental data, we estimated the force constant for plagioclase using an empirical equation27 by assuming Fe²⁺/ΣFe = 0.7 and a coordination number of 43. The calculated Δ⁵⁶Fe mineral−melt differs from the values obtained from natural samples (e.g., Δ⁵⁶FeFeO₄ = −0.1 to −0.3‰26; Δ⁵⁶FeFe₄O₉ = 0.022 ‰ − 1.155 ‰, where Ab is albite mode of plagioclase). Significant Fe isotope difference between olivine and melt was observed from phenocrysts and matrix glass of the Hawaiian Kilauea Iki lava28. However, the surface oxides indicate that these olivine–melt pairs are not in equilibrium and the diffusive processes cause Fe isotope fractionation10,26,49. A previous study33 suggests that feldspar has heavier Fe isotope composition than its coexisting
magnetite. However, these data are limited to plagioclase with high Ab and alkali feldspars, which may be applicable for plagioclase in felsic vein samples but unsuitable for plagioclase in the gabbroic samples (with an average of 42% Ab37).

Calculation of melt composition in equilibrium with the gabbroic samples. Studies on Hole 735B gabbroic rocks show significant correlations of anorthite (An) content in plagioclase with MgO in olivine, clinopyroxene, and orthopyroxene, indicating that the bulk of the coexisting minerals in each sample were coprecipitated from a common liquid undergoing cooling31. Using the Fe-Mg exchange relationship between liquid and olivine57,

$$K_{Fe-Mg}^{liquid} = (\frac{X_{Fe}}{X_{Mg}})_{liquid} / (\frac{X_{Fe}}{X_{Mg}})_{olivine} = 0.30,$$

(2)

we can calculate MgO/FeO of basaltic melts in equilibrium with olivine. The liquidus temperatures of olivine and basaltic melts are derived from well-established experimental petrology data summarized in study37, from which we use the modified equation relevant to MORB melt evolution

$$T_{liquidus} = (1055.1 + 193.8) \times (MgO/FeO)_{melt}^{-1} - 46.966 \times (MgO/FeO)_{melt}^{-1},$$

(3)

to calculate the liquidus temperature of olivine. The liquidus temperature of clinopyroxene can also be calculated using $K_{Fe-Mg}^{liquid} = 0.2438$. Most of the gabbroic samples plot onto the band defined by the liquidus olivine and clinopyroxene, suggesting that the bulk of the coexisting minerals in these samples are in equilibrium with their parental melts in terms of MgO/FeO (Supplementary Fig. 5). To calculate the compositions of basaltic melts in equilibrium with the gabbroic samples, we assume the $K_{Mg}^{gabbro} = 0.27 \pm 0.03$, and the result are given in Supplementary Data 4.

For the gabbroic samples, using their mineralogy, $\alpha_{mineral-melt}$ and average FeO content of each minerals therein (Supplementary Table 1 and Data 4), we can calculate the $56^{Fe}$ of the melt (weighted mean) in equilibrium with the bulk rock sample as follow:

$$56^{Fe}_{melt} = \frac{56^{Fe}_{melt}}{56^{Fe}_{sample}} \times \frac{56^{Fe}_{sample}}{56^{Fe}_{Rdd}} - 1,$$

(4)

where $56^{Fe}_{Rdd}$ and $56^{Fe}_{sample}$ are, respectively, the $56^{Fe}$/54Fe isopic compositions of the melt and bulk rock sample. The isotopic fractionation factor $\alpha$ between gabbro rock melt and can be calculated using Eqs. (6–8):

$$\alpha_{RR-melt} = \alpha_{mineral-melt} \times f_{mineral} + \alpha_{mineral-2-melt} \times f_{mineral2} + \cdots + \alpha_{mineral-1-melt} \times f_{mineral1},$$

(5)

$$f_{mineral} = \left[ C_{FeO, mineral X} / C_{FeO, RR} \right] \times \left[ X_{mineral X} / X_{RR} \right],$$

(6)

d gives rise to intermediate MgO/FeO and $56^{Fe}$ compositions that lie between the compositions of the residual melt and cumulate (Fig. 2b). The plagioclase with high Ab may be the host of heavy Fe isotope in the solidified residual melt samples35.

The Fe isotope composition of the primary MORB melt. In order to calculate Fe isotope composition of the primary MORB melt, we use a batch non-modal melting model (calculating parameters are given in Supplementary Table 2) by using a mantle source composition of $56^{Fe}_0 = +0.02 \pm 0.03‰$35,36 as a starting point. The $56^{Fe}$ of source, melt, and residue satisfy Eqs. (9–11): 

$$\Delta^{56}Fe_{source} = (\frac{C_{FeO,melt} / C_{FeO,source}}{C_{FeO,melt} / C_{FeO,source}}) \times 56^{Fe}_{melt} - (1 - \frac{C_{FeO,melt} / C_{FeO,source}}{C_{FeO,melt} / C_{FeO,source}}) \times 56^{Fe}_{residue}.$$

(9)

Using the $\Delta^{56}Fe_{mineral-melt}$ given in Supplementary Table 1, the $56^{Fe}$ of the melt can be calculated using:

$$56^{Fe}_{melt} = \frac{56^{Fe}_{source} - (1 - C_{FeO,melt} / C_{FeO,source}) \times 56^{Fe}_{residue}}{56^{Fe}_{melt} - 56^{Fe}_{residue}},$$

(10)

where

$$\Delta^{56}Fe_{residue} = \left( C_{FeO, mineral X} / C_{FeO, residual} \right) \times \Delta^{56}Fe_{mineral-melt} + \cdots + \left( C_{FeO, mineral X} / C_{FeO, residual} \right) \times \Delta^{56}Fe_{mineral X - melt}.$$

(11)

Calculation of Fe isotope fractionation during fractional crystallization. To better understand Fe isotope fractionation during MORB melt evolution, we explore the fractional crystallization model using Petrolog34 with a starting composition of MORB sample (3Sc/3) from an off-axis site nearby the Atlantic Platform59. This MORB composition is corrected to $M^s = 0.72$ in equilibrium with mantle olivine of $F_{O2}^{1200}$. The original and corrected major element compositions are given in Supplementary Table 3. The corrected MORB melt is set to crystallize spinel, olivine, plagioclase, clinopyroxene, orthopyroxene, ilmenite, and magnetite at a pressure of 0.2 GPa and oxygen fugacity at QFM buffer (Petrolog34), using the mineral-melt equilibria models57,61–64. A controlled amount of the crystal mass (0.01 wt%) calculation step) increases at each step of the calculation. The model stops at 15% liquid remaining. With decreasing temperature, chromium spinel crystallizes first, followed by olivine, plagioclase, clinopyroxene, ilmenite, and orthopyroxene (Fig. 3c). The SiO$_2$ of residual melt does not increase until oxides (ilmenite) begin to crystallize (Fig. 3a–c). Magnetite does not appear on the liquidus to crystallize in modeling, which agrees with the observation that magnetite is rare throughout Hole 735B (Fig. 3c).

Using the result of fractional crystallization model with Petrolog33, and assuming the starting MORB melt represents the primary MORB melt with $56^{Fe}$ of $-0.05 \pm 0.03‰$ (5–15% melting of the mantle with $56^{Fe}_0 = +0.02 \pm 0.03‰$, Supplementary Table 2), the fractionation of Fe isotope for each step of MORB melt evolution can be evaluated using Eq. (12):

$$\Delta^{56}Fe_{melt} = \left( C_{FeO,melt} / C_{FeO,CMB} \right) \times X_{Fe,CMB} + F_{melt},$$

(12)

where $F_{melt}$ is mass fraction of residual melt; $X_{CMB}$ is mass fraction of coprecipitating minerals (CMB); $\alpha$ is the current step within the evolution of the magma and $n$ represents the previous step. The fractionation of Fe isotope for coprecipitating minerals can be calculated using Eqs. (13–16):

$$\Delta^{56}Fe_{CMB} = \alpha_{CMB} \times 56^{Fe}_{melt} - 1,$$

(13)

where

$$\alpha_{CMB} = f_{sp} \times f_{il} + f_{Cpx} \times f_{il} + f_{Opx} \times f_{il} + f_{il} \times f_{Cpx} + f_{Opx} \times f_{il} + f_{il} \times f_{il} + f_{il} \times f_{il},$$

(14)

$$X_{CMB} = \left[ X_{mineral X} / X_{CMB} \right] \times C_{FeO, mineral X} / C_{FeO, CMB},$$

(15)

According to Eqs. (8) and (1) as well as force constants (Supplementary Table 1), we can calculate $\alpha$ of each step (with different temperature). The fractionation of Fe isotope for bulk cumulate (BC) can be calculated using the mass balance equation (17):

$$\Delta^{56}R_{BC} = \left( C_{FeO,melt} / C_{FeO,BC} \right) \times \Delta^{56}Fe_{melt} / \left( F_{melt} - F_{frac} \right).$$

(17)

The modeling result is shown in Fig. 3d.

Data availability. Topography data for Fig. 1a (ETOPO1 1 arc-minute Global Relief Model) and Fig. 1b-66 are available at the NOAA National Centers for Environmental Information (https://www.nccs.noaa.gov/). Other relevant data necessary to reproduce the figures presented in this paper are available in supplementary files and references given in the captions. Data are also available online the FigsHare Data repository at https://doi.org/10.6084/m9.figshare.14135012.v1.
Code availability
Petroleo3 is available for free download from http://petroleo.web.ru/. Figure 1a was plotted using GMT5/6, which can be accessed at https://www.generic-mapping-tools.org/.

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Author contributions

Y.N. designed the project, Y.C. and Y.N. wrote the manuscript, Y.C. did an analysis with the assistance of M.D., H.G., and P.G.

Competing interests

The authors declare no competing interests.

Additional information

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