



Iron isotope fractionation during fenitization: a case study of carbonatite dykes from Bayan Obo, Inner Mongolia, China

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Abstract

As a powerful tracer in high-temperature geochemistry, Fe isotopes have been studied for their behaviour during fluid exsolution and evolution related to felsic magma system, but that for carbonatite magma system remains unknown. Here we study the Fe isotope fractionation behaviour during fenitization—processes that widely occur associated with carbonatite or alkaline intrusions. Nine fenite/carbonatite samples from carbonatite dykes at Bayan Obo area are analyzed for their Fe isotope compositions as well as elemental compositions. Combined with previous reported carbonatite $\delta^{56}\text{Fe}$ data, the results show that carbonatites range from -0.35‰ to 0.28‰ , with an average of -0.10‰ in $\delta^{56}\text{Fe}$ values, while fenites range from -0.17‰ to 0.30‰ , with an average of 0.11‰ in $\delta^{56}\text{Fe}$ values. This indicates that fenitizing fluids exsolved from carbonatite melts are enriched in heavier Fe isotopes. Such a Fe isotope fractionation trend is different from that for fluid exsolution from felsic magmatism. $\delta^{56}\text{Fe}$ values in fenites are negatively correlated with indicators of fenitization intensity such as (Na+K), Ti, Ba, Th, Nb, U or Pb abundances, likely reflecting that Fe isotopes fractionate during the evolution of the fenitizing fluids. Thus, Fe isotopes are a valuable tool for tracing fluid exsolution and evolution relevant to carbonatite magmatism and related metal mineralization.

Keywords Iron isotopes · Fenitization · Fluid exsolution · Carbonatite · Bayan Obo

1 Introduction

Fe isotopes have been demonstrated as a powerful tracer in high-temperature geochemistry and have been largely applied on silicate igneous rocks and related ore deposits (e.g., Zhu et al. 2002; Wang et al. 2011, 2021; Dauphas et al. 2017). That how Fe isotopes fractionate during fluid exsolution, a magmatic-hydrothermal process in late-stage magmatism that tends to be associated with ore mineralization, has been a subject of intense interest (e.g., Heimann et al. 2008; Wang et al. 2011, 2015, 2021; Wawryk and Foden. 2015). Previous studies show that Fe isotopes fractionate significantly during fluid exsolution and evolution processes relevant to felsic magmatism, where exsolved fluids are enriched in light Fe isotopes relative to the causative intrusion and precipitation of sulfide (Fe oxide) resulting in the residual fluids being isotopically heavier (lighter), making Fe isotopes an effective tool for tracing the fluid evolution processes and related metal mineralization processes (Wang et al. 2011, 2015, 2021). However, how Fe isotopes fractionate during fluid exsolution and evolution relevant to carbonatite magmatism remains unknown, hampering the use of Fe isotopes on carbonatite systems and relevant critical metal mineralization processes (e.g., REE, Nb, etc.).

Fenitization typically occurs associated with alkaline igneous activity, where multiple pulses of alkali-rich fluid exsolved from carbonatitic or alkaline melt, resulting in aureoles of high temperature metasomatically altered country rock, termed fenites (Morogan 1994; Le Bas 2008). Different from those derived from felsic magmas, fenitizing fluids derived from alkali magmas are characterized by enrichment of alkali and critical metal elements such as Fe, Cl, Ba, Na, K, REEs, Th, and U, and they are generally oxidizing, with high $\text{Fe}^{3+}/\text{Fe}^{2+}$ ratios (Bühn et al. 2002; Drüppel et al. 2006; Le Bas 2008). It is unknown whether Fe isotopes fractionate differently or similar between felsic magmatic system and carbonatite magmatic system. Fenites

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record information about fluid exsolution and evolution related to carbonatite or alkali magmatism that is different from those for felsic magmatism, but to date, no $\delta^{56}\text{Fe}$ data has been reported for fenites.

Here in this study, fenites, combined with the corresponding carbonatite rocks, from carbonatite dykes at Bayan Obo, Inner Mongolia, China, are studied for their Fe isotope compositions, with the aim to understand how Fe isotopes fractionate during fenitization.

2 Fenites at Bayan Obo and sampling

Fenites typically occur around carbonatite or alkali silicate rocks, as have been reported at Fen carbonatite complex (Norway) (Kresten and Morogan 1986; Kresten 1988), Alno (Sweden) (Morogan and Woolley 1988), Bayan Obo (China) (Yang et al. 2000; Le Bas, 2008 etc.), and have been reviewed by Yang et al. (2000), Le Bas (2008), Wang (2015), Elliott et al. (2018) and Yaxley et al. (2022). Fenites can be divided into the potassic type and sodium type according to the ratio of Na and K (Verwoerd 1966; Le Bas, 2008). They are also classified as contact, aureole, or veined fenites based on their spatial relationship to the source intrusion (Kresten 1988), and classified as low, medium, or high-grade based on the fenitization intensity (Morogan 1994).

Fenites associated with Bayan Obo carbonatite have been recognized and described previously, which include K-fenites (pure feldspar rock developed in H9 slates that overly the massive sheet of H8 dolomite body) and Na-fenites (related to carbonatite dykes at Bayan Obo area) (e.g. Le Bas et al. 1992, 2008; Yang et al. 2000; Wang et al. 2018). Although the origin of the large body of “H8” ore-hosting dolomite marble in Bayan Obo REE-Nb-Fe deposit is still under debate (Institute of Geochemistry, Academia Sinica 1988; Sun et al. 2012, 2013; Zhu et al. 2015; Fan et al. 2015; Yang et al. 2019), tens of carbonatite dykes occur at the vicinity of Bayan Obo are undoubtedly of carbonatite origin. Typically, Na-fenites occur adjacent to carbonatite dykes, with the country rocks (normally quartzite) strongly fenitized to blue rocks marked by minerals of riebeckite and aegirine (Fig. 1). Fe oxide phases are observed in those fenites far away from the contact (Le Bas 2008). The fenitization results in an increase of Na, K, Sr, Ba, Sc, V, Zn, Nb, and REE for the country rocks (Table 1 in the Supplementary material and Le Bas 2008).

Nine fenite (Na-fenite) and carbonatite samples are collected from four carbonatite dyke intrusions Northeast of the Bayan Obo deposit, located at areas from North of Boluotou, South of Jianshan, and North of the Main deposit (see detail information in Table 1, Zhang et al. 2004; Sun et al. 2013). There are several types of fenite samples, categorized as

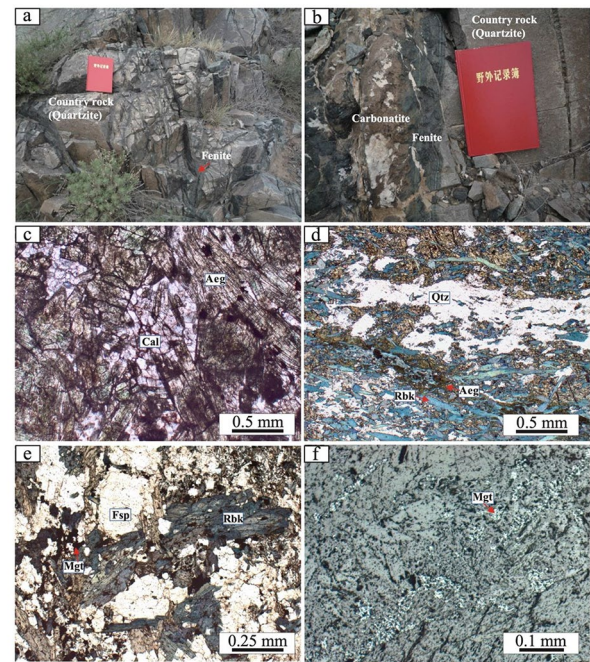


Fig. 1 Field photographs and photomicrographs of fenites in bayan obo area. (a) and (b) Fenites occurred in quartzite, associated with carbonatite dykes; (c) and (f) Photomicrographs showing characters of typical fenites, where quartzite is metasomatized by riebeckite and aegirine. (c) High-grade fenite (BN10-40) contact with carbonatite, containing coarse aegirine and calcite, PPL. (d) High-grade fenite (BN09-18) containing aegirine and riebeckite, with minor residual quartz, PPL. (e-f) Medium-grade fenite containing riebeckite and magnetite, with abundant residual feldspar and quartz, PPL. Abbreviations: Aeg = Aegirine, Fsp = Feldspar, Mgt = Magnetite, Qtz = Quartz, Rbk = Riebeckite

medium-grade to high-grade or contact fenites, according to the extent of fenitization (Fig. 1). Fenites contact with carbonatite is characterized by coarse aegirine and contain carbonate minerals (Fig. 1c). High-grade fenite samples are generally composed mainly of the riebeckite and aegirine, with minor residual quartz (Fig. 1d). Medium-grade fenite samples are characterized by riebeckite and may contain magnetite, with abundant residual feldspar/quartz (Fig. 1e-f).

3 Analytical methods

Major and trace element analysis was undertaken at the ALS Chemex (Guangzhou) Co., Ltd. Major elements were measured using fusion beads and X-ray fluorescence (XRF) spectrometer and trace elements were measured using ICP-MS (X-series).

Fe isotope analysis was undertaken at the Laboratory of Isotope Geology, Institute of Geology, Chinese Academy of Geological Sciences, with the protocol for sample dissolution, chemical separation and isotope analysis of Fe

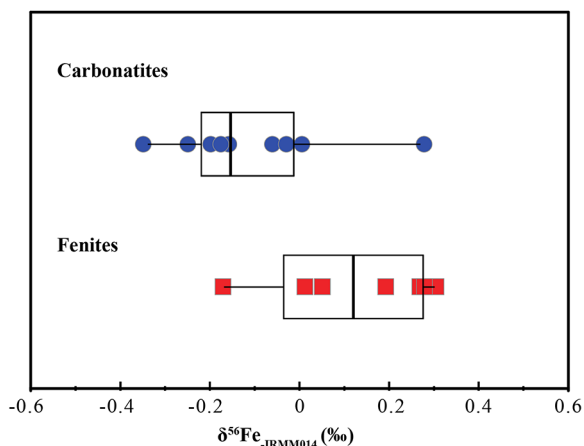


Fig. 2 Comparison of Fe isotope compositions between fenites and carbonatites from Bayan Obo area (see data source in Table 1). The ends of the boxes are the upper and lower quartiles, while the median is marked by the line inside the box. The ends of the whiskers are the highest and lowest values in each group

have been described previously (e.g. Zhu et al. 2002, 2008; Tang et al. 2006; Sun et al. 2010, 2013). In brief, fenite samples were digested with HNO₃ and HF. Carbonatite samples were digested with HCl and minor HF. Chromatographic separation of Fe was performed using AG MP-1 anion exchange resin (100–200 mesh). Matrix elements were eluted with 6 M HCl+0.001% H₂O₂ and Fe was collected with 2 M HCl+0.001% H₂O₂. Fe isotope ratios were determined by Nu Plasma HR MC-ICP-MS, with the mass discrimination corrected using standard-sample bracketing (SSB) approach. The average Fe isotope values obtained for CAGSR-1 (GBW-07105) during this study are $\delta^{56}\text{Fe} = 0.14 \pm 0.07\text{‰}$ (2SD, n = 11), identical to those obtained by Craddock and Dauphas (2011).

4 Results

Fe isotope compositions, as well as major and trace element compositions, of fenites and carbonatites at the Bayan Obo area are listed in Table 1, combined with those reported previously by Zhang et al. (2003) and Sun et al. (2013). For carbonatites, varieties of both calcio-carbonatite and magnesio-carbonatite are included, and they have similar $\delta^{56}\text{Fe}$ values, ranging from -0.35‰ to 0.28‰ for $\delta^{56}\text{Fe}$, with an average of -0.10‰ (Table 1). Fenites are typically enriched in Na, Fe and are variable in K, Ba, Nb, Sr, Sc, REEs and Th. They vary from -0.17‰ to 0.30‰ for $\delta^{56}\text{Fe}$, with an average of 0.11‰ (Table 1). Overall, both carbonatites and fenites are variable in $\delta^{56}\text{Fe}$ values, but the former is mostly negative in $\delta^{56}\text{Fe}$ values while the latter is mostly positive in $\delta^{56}\text{Fe}$ values (Fig. 2).

5 Discussions

5.1 Fe isotope fractionation during fluid exsolution from carbonatite melts

Fe isotope behaviour during fluid exsolution process related to felsic magmatism has been studied to show that the exsolved fluids are enriched in lighter Fe isotopes (Wang et al. 2011, 2015), likely due to that Fe in fluids derived from the felsic magmas is present as Fe²⁺-Cl (FeCl₂⁰) species, which tends to be isotopically lighter than magnetite or silicate melt in equilibrium (Heimann et al. 2008; Wang et al. 2015). But it is still unknown how Fe isotopes fractionate during fluid exsolution related to carbonatite magmatism.

Fe isotope behaviour during fluid exsolution from carbonatite melts is investigated here by comparison of Fe isotope compositions between carbonatites and fenites. As the protolith of fenites is quartzite with little Fe contents, while fenites are enriched in Fe (Table 1), the effect of protolith on $\delta^{56}\text{Fe}$ signature of fenites is expected minimal. The observation that heavier $\delta^{56}\text{Fe}$ values in fenites than those in carbonatites indicates that the exsolved, fenitizing fluids derived from carbonatite magmas are enriched in heavier Fe isotopes. Such a Fe isotope behaviour is different from that for felsic magmatism.

Different from that of exsolution of Fe²⁺-Cl species in fluids derived from felsic magmas, the mechanism causing Fe isotope fractionation during fenitization likely due to the enrichment of Fe³⁺ in the exsolved fluids from carbonatite magmas. Fenitizing fluids are generally suggested to be highly oxidizing, as high Fe³⁺/Fe²⁺ ratios have been measured in fenites and Fe oxide minerals commonly occur in fenites (Bühn et al. 2002; Drüppel et al. 2006; Le Bas 2008). For Bayan Obo fenites, it is also observed that disseminated Fe oxide mineral, magnetite, occurs in some medium-grade fenites (Fig. 1e-f). The oxidizing conditions of fenitizing fluids can be due to that carbonatite melts are generated at a mantle source of relatively oxidizing conditions (Braunger et al. 2020) or the “alkali ferric iron effect” (Carmichael and Nicolls, 1967). Besides, carbonatite melts or crystallization of carbonate minerals likely tend to host Fe²⁺.

5.2 Fe isotope fractionation during fenitizing fluid evolution

Temporal and spatial zonations of Fe isotopes in skarn type Cu-Fe deposits have been observed in Tongling District, China, indicating regular Fe isotope fractionation during fluid evolution related to felsic magmatism, which is mainly controlled by precipitation of Fe-bearing mineral types (e.g., sulfide and magnetite) during fluid evolution (Wang et al. 2011, 2015). As sulfide minerals precipitate, evolved

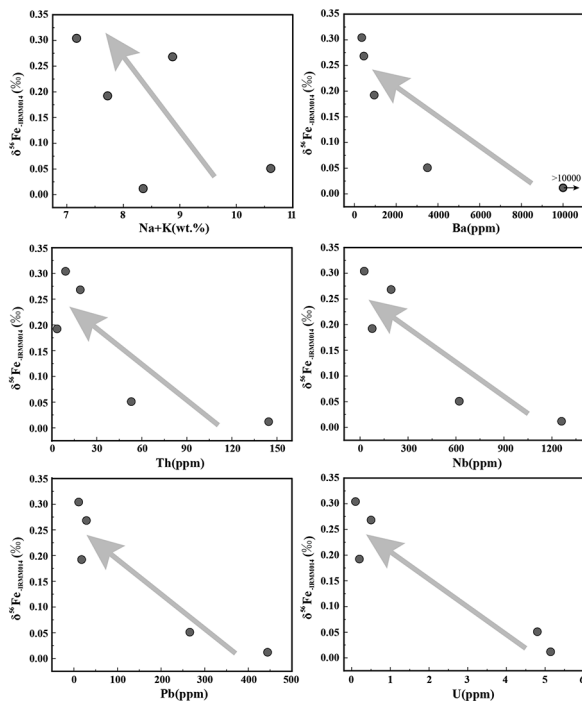


Fig. 3 Plots showing a relationship between $\delta^{56}\text{Fe}$ values and (Na + K), Ba, Th, Nb, Pb, and U in fenites (see data source in Table 1). The arrows represent the trend of fluid evolution

fluids turn isotopically heavier (Wang et al. 2011, 2015). But it is still unknown how Fe isotopes fractionate during fluid evolution relevant to alkali fenitization.

$\delta^{56}\text{Fe}$ variation in Bayan Obo fenites is as heavy as 0.47‰, and the values are negatively correlated with indicators of fenitization intensity such as (Na + K), Ti, Ba, Th, Nb, Th, U, or Pb abundances (Fig. 3). This means that a more weakly fenitized rock, which is likely farther away from the carbonatite dykes, is enriched in heavier Fe isotopes. In other words, the early-stage fenites tend to incorporate lighter Fe isotopes, and as fenitizing fluids evolve, their Fe isotopes turn to be heavier. This is consistent with the mineral and $\text{Fe}^{3+}/\text{Fe}_{\text{tot}}$ ratio evolution during fenitization. For the studied Bayan Obo fenites, it is observed that the dominated Fe-bearing minerals are silicates but Fe oxide mineral tends to occur for more weakly fenitized rocks (Fig. 1 and Le Bas 2008), indicating the increase of $\text{Fe}^{3+}/\text{Fe}_{\text{tot}}$ ratio or oxygen fugacity in fenites far away from the contact. Such a trend has also been reported in other fenite occurrences. For example, mineral compositions and S isotopes from Swartbooisdrif carbonatite, Namibia, indicate an increase of oxygen fugacity during fenitization (Drüppel, 2006). In Alno carbonatite complex from Sweden, $\text{Fe}^{3+}/\text{Fe}_{\text{tot}}$ ratio in fenites was measured to increase as far away from the contact, although a decreasing trend for some fenites may also occur (Morgan et al. 1988, 1989). Thus, the early-stage

fenites, dominated by silicate minerals with relatively lower $\text{Fe}^{3+}/\text{Fe}_{\text{tot}}$ ratio, preferentially incorporate lighter $\delta^{56}\text{Fe}$, and as fenitizing fluids migrate and evolve, the late-stage fenites, containing Fe oxide minerals with relatively higher $\text{Fe}^{3+}/\text{Fe}_{\text{tot}}$ ratio, have heavier $\delta^{56}\text{Fe}$.

6 Conclusions and implications

With carbonatites dykes from Bayan Obo, Inner Mongolia, China as a case, Fe isotope geochemistry during fluid exsolution and evolution processes relevant to carbonatite magmatism is studied. The main findings are:

1. Fenites are heavier than carbonatites in $\delta^{56}\text{Fe}$ values, indicating that fenitizing fluids exsolved from the carbonatite melts are enriched in heavier Fe isotopes. This is different from the way of Fe isotope fractionation during fluid exsolution from the felsic magmatism reported previously, where the exsolved fluids are enriched in lighter Fe isotopes.
2. $\delta^{56}\text{Fe}$ values in fenites are negatively correlated with indicators of fenitization intensity such as (Na + K), Ti, Ba, Th, Nb, U or Pb abundances, suggesting that as fenitizing fluids evolve, their Fe isotopes turn to be heavier.

This study confirms that Fe isotopes are a valuable tool for tracing fluid exsolution and evolution and related metal mineralization, but attention should be paid that Fe isotope behaviours during these processes may be different for different magmatic-hydrothermal systems.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s11631-022-00531-4>.

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Declarations

Conflict of interest We declare no conflicts of interest in this study.

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