

Apatite CO₂ and H₂O as Indicators of Differentiation and Degassing in Alkaline Magmas

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Abstract

Apatite can incorporate significant amounts of H₂O and CO₂, potentially recording volatile abundance and behavior during magma evolution. Here we conducted *in situ* measurements of CO₂ and H₂O concentrations in apatite, along with elemental compositions, from two contemporaneous alkaline volcanic suites (Tianbao and Tudiling) in the South Qinling belt in central China to better understand the CO₂ and H₂O contents and behavior in evolving alkaline melts. Apatite from alkali basalts in Tianbao contains variable CO₂ contents ranging from <100 ppm to 2.7 wt.% and H₂O contents ranging from 0.1 to 0.6 wt.%. Apatite from REE-enriched trachytes, which evolved from alkali basalt, shows significantly lower CO₂ contents and a continuous decrease in H₂O during magma fractionation. The observations suggest that CO₂ loss commenced at the early stages of magma evolution, whereas significant H₂O loss occurred during subsequent magma fractionation in tandem with REE-enrichment.

In comparison, apatite grains from the Tudiling trachyte, which is genetically linked with carbonatite, contain higher CO₂ contents (0.6 to 1.5 wt.%) but lower REE concentrations than the Tianbao trachytes. Apatite in the Tudiling trachyte is inferred to have crystallized from a carbonated alkaline magma prior to significant CO₂ loss and the separation of Tudiling carbonatitic melts, where subsequent liquid immiscibility led to REE enrichment into the carbonatitic melts. The volatile characteristics of apatite from the two volcanic suites provide valuable insights into two different evolutionary processes of alkaline/carbonatitic magmas, the behavior of CO₂ and H₂O, and the enrichment of REE in alkaline systems.

Key words: *in situ* measurements of CO₂ and H₂O; apatite; alkaline magma evolution; degassing

INTRODUCTION

CO₂ and H₂O play significant roles in various aspects of magma generation, evolution, eruption, and mineralization, impacting both the lithosphere and atmosphere. For instance, partial melting of carbon-rich mantle sources is crucial for generating alkaline and/or carbonatite magmas enriched in REE (Dasgupta *et al.*, 2009), where processes like liquid immiscibility and/or crystal fractionation likely contribute to REE enrichment (Nabyl *et al.*, 2020). The release of substantial amounts of CO₂ and H₂O during volcanic activity can have long-term effects on Earth's climate (Brune *et al.*, 2017). Therefore, accurately tracing and quantifying volatile contents, including CO₂ and H₂O, in magmas is essential for refining our understanding of magma evolution. This has implications for understanding the concentrations of economically relevant elements and for better understanding global-scale processes, such as the volatile cycle.

However, quantitatively measuring CO₂ and H₂O in igneous systems presents challenges. Melt inclusions have been considered the most direct and effective method for investigating volatile abundances in magmas (Metrich & Wallace, 2008), but they are not always present and/or suitable for analysis. An alternative approach is to examine igneous minerals capable of incorporating significant amounts of CO₂ and H₂O during

crystallization. Apatite is an ideal mineral for this purpose, as it can contain weight % levels of H₂O and CO₂ in certain systems, such as alkaline and carbonatitic rocks (e.g. Nadeau *et al.*, 1999; Hammerli *et al.*, 2021). Furthermore, apatite is a common accessory phase that crystallizes throughout the entire process of magmatic evolution (Webster & Piccoli, 2015). Therefore, apatite holds great potential as a tool for estimating CO₂ and H₂O contents in melts, as well as for deciphering magma evolution and degassing processes (Riker *et al.*, 2018; Li *et al.*, 2021).

In this study, we present detailed textural observations, combined with *in situ* trace element analyses and attenuated total reflection-Fourier transform infrared spectroscopy (ATR-FTIR) measurements of CO₂ and H₂O, focusing on apatite grains from two alkaline volcanic suites located in the South Qinling region of central China. Our data set reveals that the CO₂ and H₂O concentrations in apatite offer novel insights into magmatic evolution, degassing/exsolution processes, and potentially the process of REE enrichment.

SAMPLE DESCRIPTION

Samples of contemporaneous volcanic rocks from the Tianbao and Tudiling regions of the South Qinling belt in central China

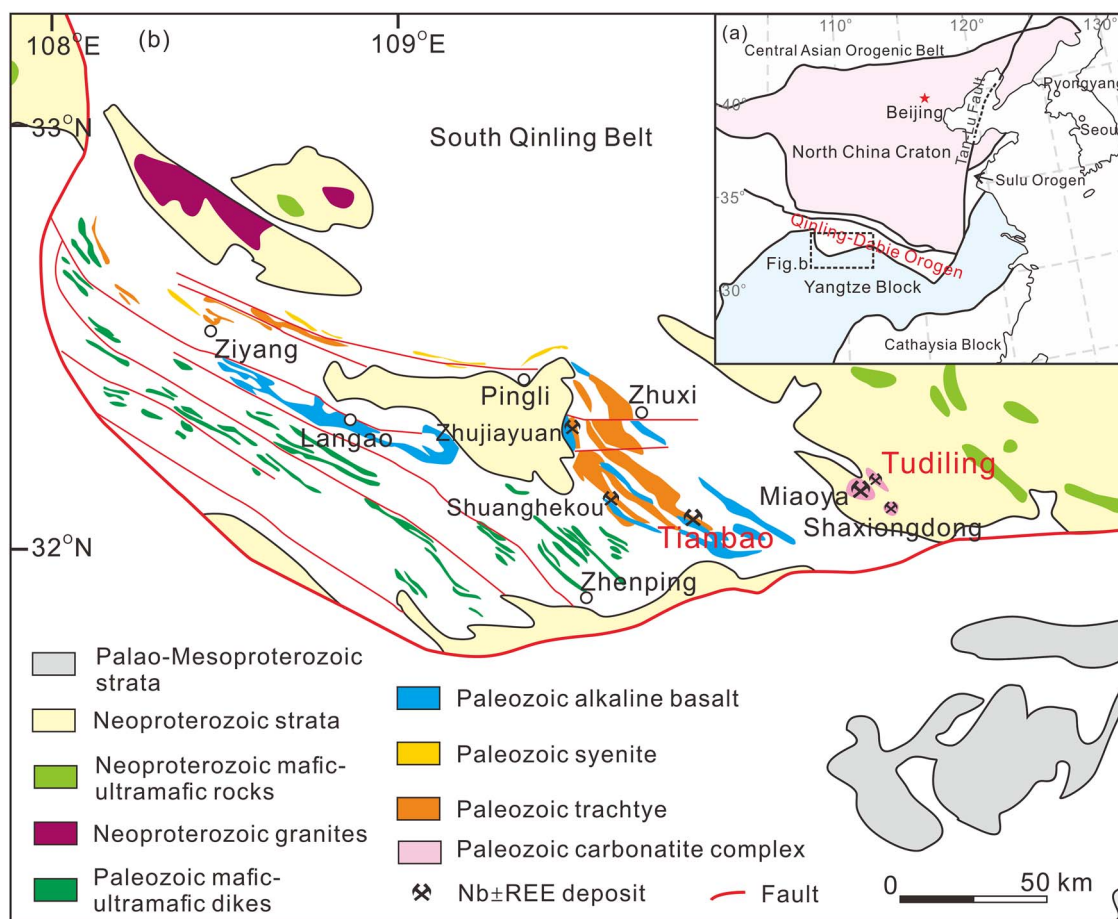


Fig. 1. Geotectonic maps showing the location of the Qinling orogen (a) and the distribution of early Silurian alkali magmatic rocks and related Nb-REE deposits in the South Qinling Belt (b).

(Fig. 1) were collected for trace element and CO_2 and H_2O analyses. These volcanic rocks together with the contemporaneous intrusive rocks that emplaced around 440 to 430 Ma (Yan *et al.*, 2021; Su *et al.*, 2023) are believed to have formed in a continental rift setting (Wu & Zheng, 2013).

Samples from the Tianbao igneous suite include alkali basalt and two types of phenocryst-bearing trachyte, referred to as trachyte I and trachyte II, based on their phenocryst mineral and whole rock trace element compositions (Su *et al.*, 2023). In microscope cathodoluminescence (CL) images, apatite grains from the Tianbao alkali basalt exhibit purple or blue colors (Fig. 2a). In trachyte I, most apatite grains are blue or light blue, with a few grains displaying purple CL signals (Fig. 2a; Supplementary Fig. S1). Apatite grains in trachyte II appear dark purple (Fig. 2a). The Tudiling trachyte, located approximately 50 km away from Tianbao (Fig. 1), is associated with carbonatite. Apatite grains from the Tudiling trachyte exhibit exclusively reddish colors in CL images (Fig. 2a; Supplementary Fig. S1).

RESULTS

Apatite grains within the examined volcanic suites contain high but variable REE concentrations (Fig. 2b; see Supplementary Table S1 for complete dataset). Apatite grains in the Tianbao alkali basalt contain REE concentrations ranging from 4351 to 7701 ppm, with slightly negative Eu anomalies ($\delta\text{Eu} = [2 \cdot \text{Eu}_N] / [\text{Sm}_N + \text{Gd}_N]$: ~ 0.88 on average). Purple apatite grains from

Tianbao trachyte I display similar total REE contents (~ 3300 ppm) and δEu ratios (~ 0.90) as those from the alkali basalt. Interestingly, the CL-blue and light blue domains within the same sample (trachyte I) exhibit significantly higher REE contents of 16 657 to 44 747 ppm but lower δEu ratios (~ 0.78). Tianbao trachyte II apatite grains contain the highest REE contents (38 511 to 71 378 ppm) with pronounced negative Eu anomalies (δEu : ~ 0.44). In contrast, apatite grains from the Tudiling trachyte contain significantly lower REE contents (6636 to 7362 ppm) without notable Eu anomalies (δEu : ~ 0.98).

CO_2 and H_2O contents of apatite were measured via ATR-FTIR following the same approach as described in Hammerli *et al.* (2021), which is summarized in Supplementary Material 1 and results can be found in supplementary Table S3. The CO_2 and H_2O contents of apatite show systematic variations among different rock types in the Tianbao volcanic suite (Fig. 3a, b). Apatite grains in alkali basalts contain variable CO_2 and H_2O contents ranging from < 100 ppm to 2.73 wt.% and from 0.1 to 0.6 wt.%, respectively. No correlation between CO_2 and H_2O concentrations is observed. CL-purple apatite grains from trachyte I contain 1.0 to 2.1 wt.% CO_2 and 0.18 to 0.38 wt.% H_2O , which is similar to CO_2 and H_2O contents in apatite from the alkali basalt. However, most CL-blue to light blue grains within the trachyte I sample contain CO_2 contents below the detection limit (< 100 ppm) but contain 0.09 to 0.42 wt.% H_2O . Furthermore, the CO_2 and H_2O contents of apatite grains in Tianbao trachyte II are mostly below the detection limits (< 400 ppm H_2O and < 100 ppm CO_2). In contrast to Tianbao, apatites in the Tudiling trachyte display high CO_2 contents of 0.61

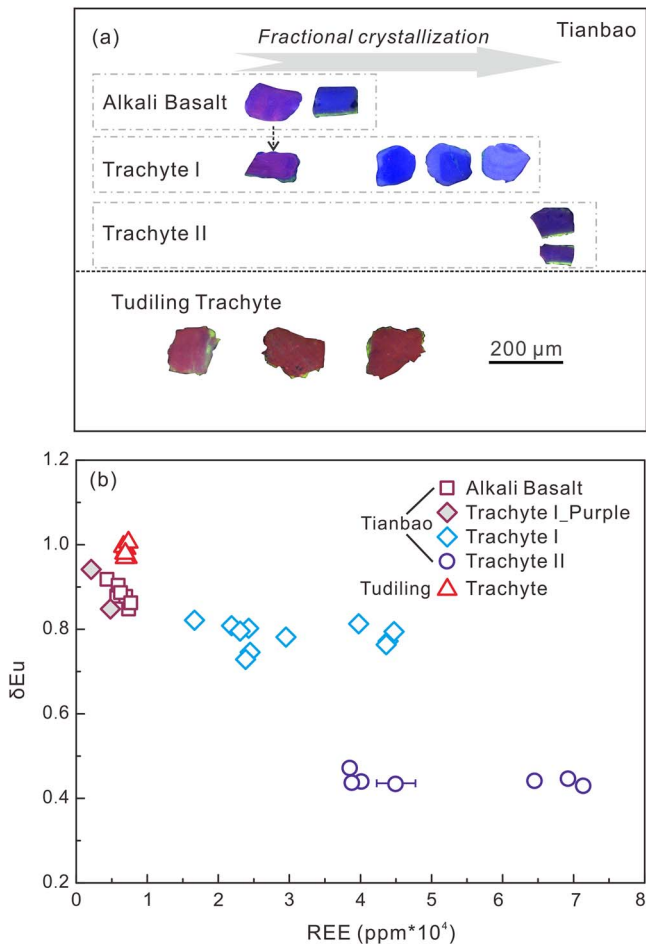


Fig. 2. (a) Cathodoluminescence (CL) images displaying representative apatite grains in Tianbao and Tudiling volcanic rocks; (b) Plot showing the δEu vs. REE relationship of apatite. Note that the analytical errors for trace elements of apatite are less than 5% and most error bars of REE concentrations and Eu anomalies (δEu) are smaller than the symbols (see Supplementary Table S1 for details).

to 1.45 wt.% but low H_2O concentrations ranging from <400 ppm to 0.13 wt.%.

DISCUSSION

Interpretation of CL properties for apatite

Cathodoluminescence (CL) imaging was used to distinguish between different apatite types and generations (see Fig. 2 and Supplementary Fig. S1) in the investigated samples prior to *in situ* micro-analysis. Apatite grains from the alkaline volcanic rocks in Tianbao and Tudiling exhibit various CL colors ranging from blue to purple to red (Fig. 2; Supplementary Fig. S1). Generally, different CL colors are a result of the presence of lattice defects (such as vacancies and broken bonds) or the incorporation of specific trace elements, including their valency (e.g. Mn^{2+} , $\text{REE}^{2+/3+}$; Götze, 2012). The incorporation of these so-called activator elements into the mineral lattice is influenced by the specific physicochemical conditions during crystallization of the minerals (Götze, 2012; Decrée et al., 2020). The different CL-colors in our study are a result of the response to the electron transitions of the REE in the minerals (Supplementary Fig. S2). Therefore, the colors of the CL images correlate to different REE concentrations and patterns (e.g. Sm^{3+} , Dy^{3+} , and Eu^{2+}), which have been confirmed by our

laser ablation-inductively coupled plasma-mass spectrometer (LA-ICP-MS) analysis of apatite minerals from each of the different populations.

Apatite recording different evolutionary histories of magmas

Based on consistent ages, Sr-Nd isotope signatures, and major and trace element compositions, previous studies have suggested that the alkaline rocks in the South Qinling belt are the products of fractional crystallization of similar alkali basaltic parental magmas (Wang et al., 2021; Yan et al., 2021; Su et al., 2023). Our study of the trace elements in apatite supports this interpretation as trace elements in apatite suggest an evolutionary trend from basaltic to trachytic rocks. Negative Eu anomalies in apatite increase from alkali basalt to trachyte I and trachyte II in Tianbao, suggesting an incremental fractionation of plagioclase during magma evolution (Larsen, 1979). Weakly negative Eu anomalies are also present in apatite from the alkaline basalts (Fig. 2b), implying that the alkali basalts also underwent a low degree of differentiation prior to eruption. It is worth noting that CL-purple apatite grains from Tianbao trachyte I contain trace elements concentrations that are similar to those from the alkali basalts but are distinct from CL-blue to light blue apatite grains within the same sample (Fig. 2b). These CL-purple trachyte I apatite grains also contain CO_2 and H_2O contents that are indistinguishable from those in alkali basalt (Fig. 3a), suggesting that the CL-purple apatite grains in trachyte I likely crystallized from early-stage alkali basaltic melts and were subsequently entrained by trachytic magmas. The significant differences in trace elements and volatile contents of apatite grains from the different rock types align with the interpretation of magma differentiation from alkaline basalt to trachyte II.

While the volatile properties (CO_2 and H_2O contents) of the evolving magmas are inferred to be recorded and archived in apatite, limited experimental data are available regarding their partitioning behavior, hindering efforts to solely quantify CO_2 and H_2O contents in alkaline magmas using apatite data. Riker et al. (2018) have experimentally demonstrated that apatite H_2O - CO_2 contents are correlated with those in silicate melt, and that CO_2 contents in apatite also correlate with $\text{CO}_2/(\text{H}_2\text{O} + \text{CO}_2)$ ratios in the melt. Our study reveals that apatite grains in the alkali basalts contain the highest CO_2 contents (up to 2.73 wt.%), which are even higher than those previously reported in carbonatites (Nadeau et al., 1999; Hammerli et al., 2021). This observation suggests that the parental alkali magmas in the South Qinling belt were CO_2 -rich with high $\text{CO}_2/\text{H}_2\text{O}$ ratios. However, some apatite grains in the Tianbao alkali basalt also show CO_2 -depleted signatures (i.e. <100 ppm), indicating the emission of CO_2 -dominated fluids or gases during the early-stage evolution of relatively primitive alkali basalts. Interestingly, most apatite grains in alkali basalt and trachyte I retain high water contents despite their low CO_2 contents, which is especially the case of trachyte I apatite (Fig. 3a). This phenomenon may reflect that early magmatic evolution did not result in a significant release of H_2O . However, subsequent magma evolution (i.e. fractionation) from the trachyte I to trachyte II stage leads to a significant decrease in apatite H_2O contents to <400 ppm, suggesting that significant H_2O exsolution or degassing takes place after earlier CO_2 loss.

To determine whether apatite from the analyzed volcanic rocks can qualitatively record relative CO_2 and H_2O contents in alkaline magmas, we modeled CO_2 and H_2O concentrations in alkali magmas via MELTS (Gualda et al., 2012; Ghiorsio & Gualda, 2015), using the Tianbao alkali basalt as the starting composition (Su et al., 2023). The modeling results indicate that CO_2 contents in magmas

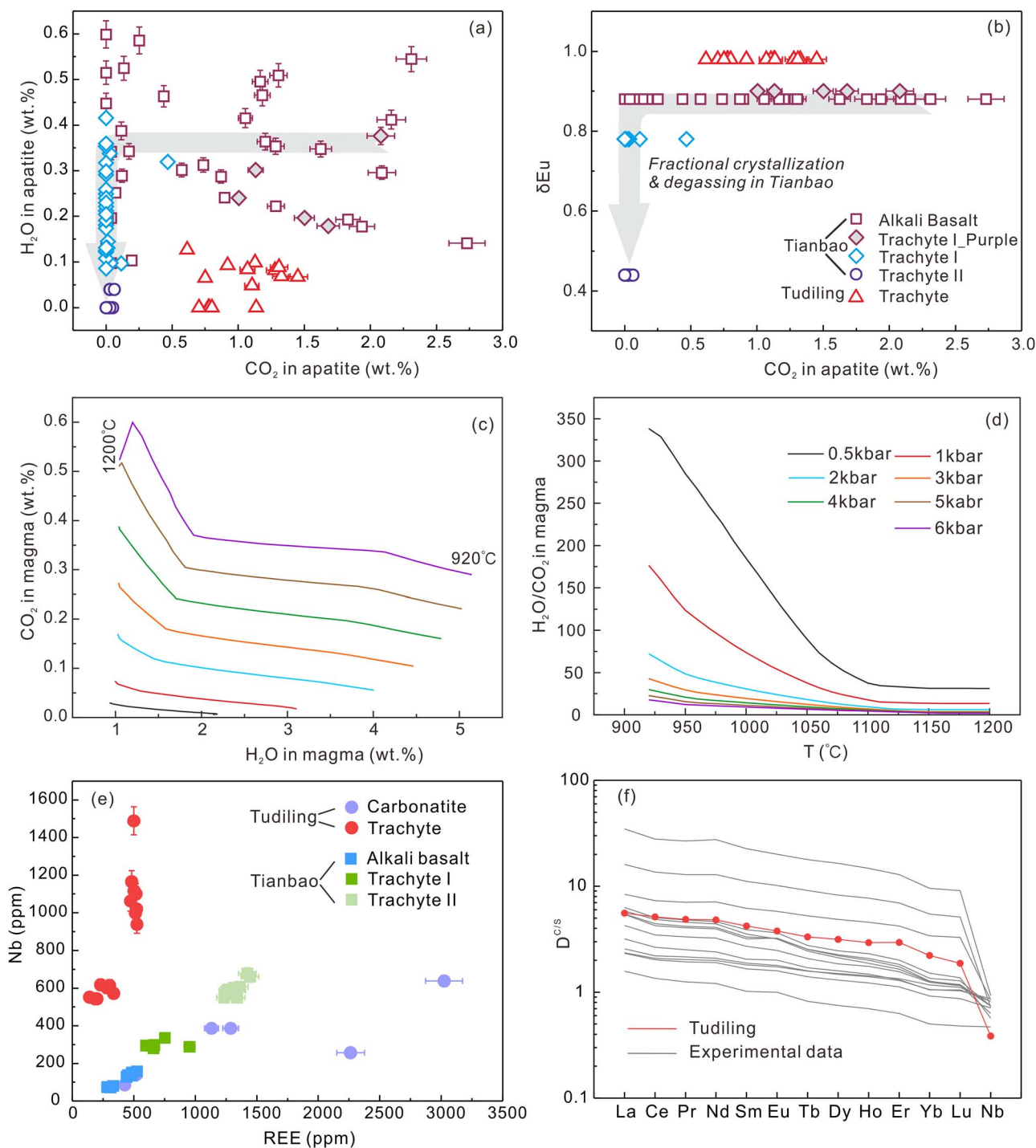


Fig. 3. (a)–(b) Apatite CO₂ and H₂O contents in Tianbao and Tudiling volcanic rocks. Note that individual analysis uncertainty is less than 5% at 95% confidence interval (Hammerli et al., 2021; see Supplementary Table S3); (c)–(d) MELTS modeling outcomes revealing CO₂ and H₂O concentrations in Tianbao alkali magmas at pressures of 0.5–6 kbar; (e) REE and Nb concentrations in different types of rocks from Tianbao and Tudiling (This study; Yan et al., 2021; Su et al., 2023). Note that individual analysis uncertainty is less than 5% (see Supplementary Table S2); (f) Comparison of Tudiling carbonatite-trachyte ratios for REE and Nb with experimental carbonatite-silicate partition coefficients ($D^{C/S}$) from Nabyi et al. (2020).

decrease with decreasing temperatures and pressures. Particularly, there is a notable decline in CO₂ concentration in magmas during the early stages of magma cooling when the temperature decreases from 1200 to 1100°C under various pressures (0.5 to 6 kbar; Fig. 3c, d). These findings align with the observed CO₂ contents in apatite from the alkaline magmas in South Qinling, indicating that apatite grains can record the early release of CO₂ from alkali basalt to trachyte I. Alkaline magmas can contain

slightly higher water contents as magma temperatures decrease, resulting in increased H₂O/CO₂ ratios. However, water contents in magmas decrease with decreasing pressure (Fig. 3d), consistent with previous experimental studies on H₂O solubility of alkaline magmas (e.g. Shishkina et al., 2014; Buono et al., 2020). Therefore, the decreasing trend of H₂O contents in apatite from trachyte I to trachyte II is likely governed by magma crystallization under different pressures when fluid exsolve as the magma ascends.

Although no mafic (e.g. basaltic) rocks have been identified in the Tudiling area, elemental and Sr-Nd isotope studies have suggested that the Tudiling and Tianbao volcanic rocks were derived from a similar mantle source (e.g. Yan *et al.*, 2021; Su *et al.*, 2023). Therefore, it is inferred that the Tudiling trachyte likely formed through fractional crystallization from a parental magma similar to the Tianbao alkali basalt (Yan *et al.*, 2021). However, apatite grains from the Tudiling trachyte exhibit significantly different CO₂ and H₂O contents compared to those of Tianbao trachyte. They display much higher CO₂ contents and high CO₂/(H₂O + CO₂) ratios, which are similar to those of the Tianbao alkali basalts. Such high CO₂ contents in apatite suggest the crystallization of apatite from a CO₂-rich magma source. Therefore, we speculate that apatite grains from the Tudiling trachyte crystallized from an earlier carbonated silicate melt before the separation of carbonatitic melt, and that no significant loss of CO₂ occurred during the early fractionation of the silicate melt.

The differences in CO₂ and H₂O between Tianbao and Tudiling volcanic rocks indicate that they likely underwent different evolutionary processes. Interestingly, the behavior of volatiles (especially CO₂) in magmas can be correlated with REE enrichment in the studied volcanic rocks. In Tianbao, whole-rock geochemical data demonstrate a continuous increase in REE contents from alkali basalt to trachyte II (Su *et al.*, 2023), consistent with the trend observed in apatite REE contents. The Tudiling trachyte has REE contents much lower than those of Tianbao trachyte and just similar to Tianbao alkali basalt, suggesting that REE are likely fractionated into the carbonatite phases. This observation is consistent with previous experimental data indicating that REEs tend to concentrate in carbonatite melts when immiscible from trachytic melts (Fig. 4; Nabyl *et al.*, 2020). Thus, we propose that the Tudiling trachyte represents a residual silicic endmember after the unmixing of carbonatite. The Tudiling trachyte contains the highest Nb content among all the volcanic rocks in the South Qinling, also supporting that they represent an evolved magmatic endmember (Yan *et al.*, 2021). In this consideration, these apatite grains were crystallized from earlier parental carbonated trachytic magma and were subsequently inherited by residual trachyte melt after unmixing of carbonate melts. Consequently, the unmixing of carbonate melts leads to a significant drop in REE contents in the coexisting trachytic melts in Tudiling. In contrast, the continuous evolution and ascent of Tianbao alkali basalt do not result in the formation of carbonatitic melt but rather the continuous release of CO₂ and H₂O through gases or aqueous fluids. This, combined with magma fractionation, leads to the concentration of REE in trachyte II melts.

CONCLUSIONS

In this study, we present detailed textural and *in situ* elemental, CO₂, and H₂O compositions of apatite from two alkaline volcanic suites in the South Qinling belt, central China. Our findings provide valuable insights into two distinct processes of REE enrichment during the evolution of alkaline/carbonatite magmas. In the Tianbao alkaline suite, the fractionation of alkali basalts involved early CO₂ loss and subsequent H₂O release, ultimately leading to the formation of REE-rich trachytes. Conversely, in Tudiling, the early evolution of the melt did not involve significant CO₂ loss, and instead resulted in the separation of REE-rich carbonatite. The distinct magmatic evolution and degassing processes in alkaline magmas are effectively captured by the CO₂ and H₂O contents in apatite. When combined with other volatile elements (such as S, Cl, F) and isotopes (including Sr, Nd, O), apatite exhibits

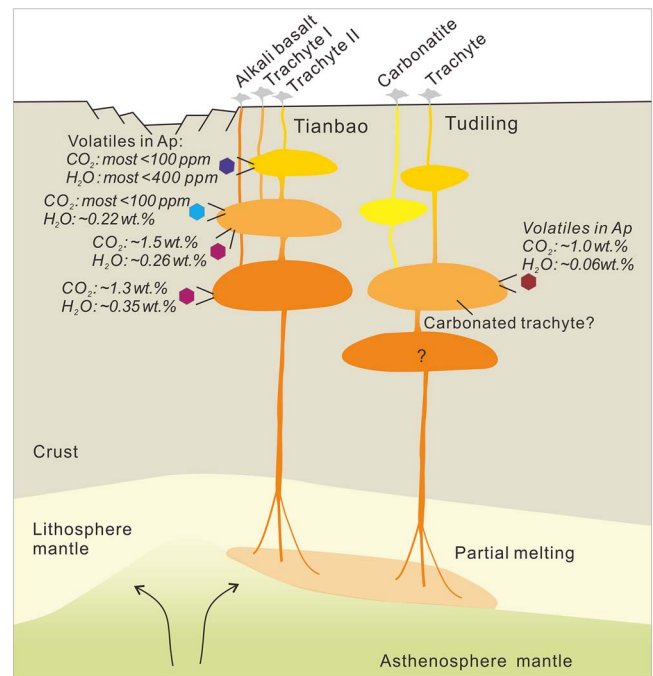


Fig. 4. Petrogenetic models illustrating the evolution of Tianbao and Tudiling volcanic rocks. Apatite in Tianbao preserves evidence of early CO₂ loss during magma evolution from alkali basalt to trachyte I, followed by a transition to H₂O-dominated release in trachyte II melts. Conversely, there is no significant CO₂ loss during the early melt evolution in Tudiling, ultimately leading to the unmixing of REE-rich carbonatite. Note that the apatite grains with high CO₂ content in Tianbao trachyte I were crystallized from early-stage alkali basaltic melts and were subsequently entrained by trachytic magmas, see text for details.

significant potential for deciphering fluid properties, exsolution phenomena, and potentially even metal transfer within magmatic-hydrothermal systems.

Supplementary Data

Supplementary data are available at *Journal of Petrology* online.

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Data Availability

All data are available in the main text or the online supplementary materials.

Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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