

Revision 1

Appendix: The transition of the magma plumbing system of Tianchi shield-forming basalts, Changbaishan Volcanic Field, NE China: Constraints from dynamic Fe-Mg diffusion modelling in olivine

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APPENDIX TEXT

Appendix 4: Supplementary remarks

Part 1: Comparison of dynamic diffusion model based on linear cooling and exponential cooling models

The olivine crystals in our study, especially high-Fo populations of TD and BS units, recorded a series of magmatic events such as shallow storage, magma ascent, and lava emplacement. The cooling rates of magmatic environments are varying in different stages. This implies that it is necessary to verify the reliability of using a simple linear cooling model for crystals undergoing complex magmatic processes. Zhang (2008) suggests that the exponential cooling model is more suitable for the cooling history of volcanic systems that underwent pre-eruption and post-eruption processes:

$$T = T_{min} + \frac{T_0 - T_{min}}{1 + \frac{t}{\tau}}$$

The τ is the time when the temperature is 1-1/e of the T_0 . This equation depicts a cooling process in which the cooling rate grows exponentially as the temperature declines. We separately applied the linear and exponential cooling models to the representative compositional profiles of high Fo-olivines from each thin section in the TD and BS units (n =11) while keeping the other variables constant. The results show that both cooling models can fit the composition profile well (Fig. S7) and that adjustments in the cooling

model have no significant effect on the results of the timescale and other variables (Table S1). This demonstrates that although the exponential model aligns with the cooling history of volcanic systems, a simple linear model is as effective. We consider that the following two reasons can explain why the linear model is still well-fitted:

1. The range of temperature variation is small, and a linear model can be regarded as an approximation of exponential models. We rejected this hypothesis as our simulations showed that both the linear and exponential models produced significant temperature ranges (Table S1). Geothermometer constraints based on high-Fo olivine cores and clinopyroxene microlites indicate that magma experienced significant cooling from sub-surface magma reservoirs to lava flows (Fig. 6).
2. In multi-stage magmatic processes of Tianchi basaltic magma systems, the establishment of compositional profile shape is controlled by the magmatic environment of predominant magmatic events, and the modification by the subordinate magmatic environments on profile shape can be moderately ignored. The magma temperature and crystal residence timescale govern the extent to profile shape is modified by diffusion. Petrone et al. (2016) proposed a non-isothermal diffusion incremental step model (NIDIS) to solve the multi-stage diffusion question. In their samples, clinopyroxene crystals in tephra recorded the two-stage processes before the eruption. The high temperature and significant crystal residence timescales in the second stage indicate that the diffusion in the second stage significantly modified the profile shape established in the first stage. However, in our samples, compared to the

timescales of magma ascent and lava emplacement, the high-Fo olivines experienced significantly longer timescales and high temperatures for residence in shallow magma reservoirs. The adequate residence time and relatively high temperature in the reservoirs enable the compositional profile to primarily record the timescales and environmental information of the shallow magma reservoir. The lower cooling rates ($\sim 10^{-7}$ – 10^{-6} °C/s) and slower G_0 ($\sim 10^{-14}$ – 10^{-13} m/s) obtained from the high-Fo populations of the TD and BS unit (Figs. 8e and 8f) imply a hot and relatively adiabatic magmatic environment, thus support the hypothesis that the magma underwent some stagnation in the shallow magmatic storage system. In comparison, timescales of magma ascent and surface emplacement are much shorter (19–66 days obtained from low-Fo populations of the TD and BS units). The shorter timescales and lower magma temperatures make it difficult to further effectively modify the compositional profiles that have already been formed through diffusion re-equilibration in shallow magmatic reservoirs.

Our simulations demonstrated that the dynamic diffusion models based on two cooling models generate well-fitting and comparable timescale results. We still prefer to use simple linear models because they provide cooling rate information to indicate the predominant magmatic environment where the crystals were experienced. It is worth noting that the best-fit linear cooling rate for crystals that record complex magmatic processes is not representative of any specific magmatic event. More accurately, it indicates the principal

magmatic environment that drove the shape of the diffusion profile over multi-stage processes. In contrast, the parameter τ provided by the exponential model represents the time when the initial temperature of the diffusion decreases to its $1-1/e$. This parameter has limited meaning considering that diffusion is only effective at high temperatures.

Part 2: Relevant parameters and detailed calculations of minimum magma supply rate (Q_c)

The minimal magma supply rate (Q_c) was calculated using Equation 4 of Morgado et al. (2017), which was modified based on the model (Eq. 1) of Menand et al. (2015).

$$Q_c = \frac{8}{9} \left(\frac{C_p(T_f - T_{inf})}{L(T_0 - T_f)} \right)^{\frac{9}{4}} \left(\frac{\Delta \rho g k^3 H^3}{\mu} \right)^{\frac{1}{4}} \left(\frac{K_c}{\Delta \rho g} \right)^{\frac{2}{3}}$$

Thermal analysis of magma transport particularly considers physical and thermal properties of the ascending magma (e.g., ascent height, initial and freezing temperature, latent heat) and country rock conditions (e.g., fracture toughness, thermal diffusivity, etc.).

Where H is the depth from which magma can ascend to the surface without completely solidifying. In this case, we chose 20 km (maximum crystallization depth of the primitive olivine modelled by rhyolite-MELTS) which represents the maximum vertical distance that an ascending magma needs to travel before eruption for the Tianchi magmatic system. T_0 and T_f are the initial and freezing temperatures of the ascending magma, respectively. Crystallization temperatures of olivine cores (1138–1227 °C) calculated by Al-in-olivine thermometry (Coogan et al. 2014) were used to represent initial magma temperatures. We used rhyolite-MELTS to model a series of isobaric cooling models and evaluated the

evolution of crystallinity with cooling temperature. The freezing temperature is defined as the temperature corresponding to the melt volume fraction of 0.6–0.7, representing the maximum particle packing fraction for magmas to retain the rheological properties of a melt (Cashman et al. 2017). The freezing temperatures range from 998 °C to 1121 °C for the considered pressure range resulting in temperature differences of 17 °C to 229 °C between the initial and freezing temperatures. $\Delta\rho$ is the density difference between the magma and the country rocks. We used the Tianchi average upper crust density of 2.65 g/cm³ to represent the average density of country rocks (Guan et al. 2020). The density range of the ascending magma (2.39–2.65 g/cm³) was assumed to correspond to the temperature evolution between initial and freezing magma temperature using rhyolite-MELTS.

μ is the dynamic viscosity of the ascending magma. To calculate μ , we first selected the corrected bulk-rock composition of TD-2 equilibrated with the Fo₈₂ olivine (Part 5 of Appendix¹) and the bulk-rock composition of LFZ-4 (Part 1 of Appendix¹) representing the most primitive magma composition and the most evolved composition, respectively. Such compositions are employed in the equation of Giordano et al. (2008) to calculate the melt viscosity with no crystals. We calculate the melt viscosity as about 10 Pa•s based on a temperature of 1201 °C (olivine core average temperature of the TD unit) for the most primitive melt under the water content of 1 wt%, and about 114 Pa•s using a temperature of 1059 °C (clinopyroxene microlite average temperature of the LFZ unit) for the most

evolved melt under the water content of 2.5 wt% (We assumed the water is perfectly incompatible, the initial water content is 1 wt% and the minimum melt fraction is 0.4). The dynamic viscosity of magma can be defined as the product of the melt viscosity combined with the effect of suspended crystals and vesicles (e.g., Harris et al. 2008):

$$\mu_{(magma)} = \mu_{(melt)} \times \mu_{(crystal)} \times \mu_{(vesicle)}$$

Here, we only consider the effect of suspended crystals, because vesicles are only present sparsely in our samples, and vesicles most probably formed after the eruption. The dynamic viscosity of the crystal-bearing magma was obtained using the equation of Krieger and Dougherty (1959) which considers the amounts of suspended crystals as well as crystal shapes. The crystal volume fraction was set to 5% of olivine and 10% of plagioclase. The olivine and plagioclase were assumed as spherical and prolate shapes respectively. Geometries of all crystals give the same magnitude of relative viscosity for a given crystal volume fraction ≤ 0.25 (Mueller et al. 2010). The maximum particle packing fraction is set as a melt volume fraction of 0.6-0.7 (Cashman et al. 2017). Therefore, the maximum magma dynamic viscosity is about 18.3–20.5 Pa·s for the most primitive melt and 182.7–205.3 Pa·s for the most evolved melt.

C_p is the specific isobaric heat capacity which ranges from 1300 J/kg K to 1700 J/kg K for natural silicate melts as suggested by Lesher and Spera (2015). L is the magma latent heat (2.5×10^5 - 5.5×10^5 J/kg), K_c is the fracture toughness of the country-rock ($10^6 \times 10^9$ Pa m^{1/2}), κ is the country-rock thermal diffusivity (0.3×10^{-6} - 2×10^{-6} m²/s) and T_{inf} is the crustal far-

field temperature (50-600 °C) that represent the crustal temperature horizontally distal from the heat source. These values were taken from Menand et al. (2015) and references therein. g is the gravitational acceleration ($9.8 \text{ m}^2/\text{s}$). Detailed ranges of all parameters are compiled in Table S2. We adopted the Monte Carlo simulation method to obtain the probability distribution of Q_c and the sampling number is 50,000 to ensure the reliability of the model results. Detailed modelled results of the expected range of critical supply rates for basaltic magmas are shown in Fig. S10. The Q_c presented in the main text define as the minimum boundary values of the complete range, i.e., the minimum magma supply rate for avoiding freezing of ascended magma in a ~20 km long dyke.

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