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Caveats and challenges in geospeedometry: A reply to Faak et al.'s critique of the Mg–REE coupled geospeedometry

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ABSTRACT

The commentary note from Faak et al. questioned the new Mg–REE coupled geospeedometry and its application to Hess Deep gabbros in the recent paper of Sun and Lissenberg (2018; denoted as S&L). Characterizing S&L's approach as "flawed", Faak et al. mistakenly asserted that S&L's results regarding rapid hydrothermal cooling of the fast-spreading lower oceanic crust were "incorrect". Instead, they claimed that the Mg-in-plagioclase geospeedometry used in Faak et al. (2015) showed "good precision as well as accuracy", which produced slower cooling rates consistent with conductive cooling. We strongly disagree with Faak et al.'s claims but appreciate the opportunity here allowing us to further elaborate S&L's approach in greater detail. In this reply, we show that the erroneous claims of Faak et al. are derived from their misunderstandings on geospeedometry regarding the calibration of exchange thermometers and bulk-diffusion modeling for multi-mineral systems.

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1. Introduction

The motivation of Sun and Lissenberg (2018; denoted as S&L hereafter) and Faak et al. (2015) is to resolve the debate on formation of fast-spreading lower oceanic crust regarding its cooling style and igneous accretion. A key to this problem is the quantitative determination of cooling rates and crystallization temperatures for *in situ* gabbros across the lower oceanic crust at Hess Deep. The aforementioned two studies used different approaches to this problem but obtained contrary results of cooling rates for Hess Deep gabbros (see Fig. 6d of S&L). Apparently, their distinct approaches are responsible for the opposite results of cooling rates.

To argue against S&L's approach, Faak et al. listed five points in their comments. In Points I&II, they described S&L's Mg-exchange thermometer as "problematic", because it was not calibrated based on "direct measurements" and "failed" to include silica activity. In Points III&IV, they considered S&L's bulk-diffusion approach as "incorrect" due to the "neglect" of anorthite zoning and "inappropriate" spherical geometry. In Point V, they questioned the accuracy of S&L's chemical analysis as they found large standard deviations of MgO in plagioclase. Meanwhile, they ignored the critical limitations of their own geospeedometry and claimed that their

approach had "good precision as well as accuracy". Overall, their comments are misleading due to a number of misinterpretations on the fundamental aspects of geospeedometry. Thus, it is crucial to remark major caveats and cautions in geospeedometry to set the stage for clarification of Faak et al.'s concerns.

2. Geospeedometry: caveats and cautions

Quantitative extraction of thermal histories from rock records requires accurate geospeedometers to measure the kinetic responses of minerals to changes in temperature. Without significant crystal growth, such kinetic responses manifest in mineral compositional variations and are primarily controlled by the diffusive exchange between co-existing minerals. Since the chemical diffusivity is dramatically reduced at lower temperatures, the rate of diffusive exchange decreases in a cooling petrological system and effectively diminishes after a certain time, yielding stabilized compositional profiles in minerals (i.e., closure profiles; Dodson, 1973). The average or local concentrations of the closure profile correspond to apparent equilibrium temperatures (i.e., bulk or local closure temperatures, respectively; Dodson, 1973), which can be determined by relevant exchange thermometers calibrated based on the temperature-dependent partitioning of the element of interest. Using the familiar diffusion equation (see S&L's Appendix A), one could model the closure profiles (or local closure temperatures) of single crystals to estimate cooling rates, i.e., the grain-scale diffusion approach; alternatively, one could also model the average

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concentrations or bulk closure temperatures of coexisting minerals to determine cooling rates, i.e., the bulk-diffusion approach. Both approaches have been discussed in Dodson (1973). However, Faak et al. mistakenly claimed that “there is no reason to expect the bulk plagioclase and clinopyroxene compositions provide any information about the thermal history” (see their Point II).

The accuracy of determined cooling rates strongly depends on the knowledge of several basic parameters, including diffusion coefficients of individual minerals, partition coefficients between coexisting minerals, geometry, size and proportion of each mineral, and initial temperature and composition. Among these, the initial condition, mineral abundance, and crystal geometry are often simplified through certain approximations. Notable exceptions are single crystals with well-preserved stepwise concentration profiles, presumably recording initial conditions and insensitive to diffusion geometry (e.g., Watson and Cherniak, 2015). Perhaps the most commonly used assumptions for the initial condition and mineral proportion are those from Dodson (1973), who assumed (1) negligible influence of the initial condition for systems with extensive diffusive re-setting and (2) surrounding phases acting as a homogeneous infinite reservoir. Without knowing the initial conditions, it is actually ambiguous to estimate the extent of diffusion and to further justify the validity of Dodson’s first assumption. The effect of surrounding phases in a multi-mineral system is generally believed to be insignificant for crystals with much lower chemical concentrations and/or modal abundances than the surrounding (e.g., Liang, 2015). However, this assumption becomes invalid when the crystal and surrounding mineral show considerable differences in diffusivities and/or grain sizes (see Eq. (8a–c) in Sun and Liang, 2017). Thus, these Dodson-type assumptions should be applied to multi-mineral systems with extreme caution. Here “Dodson-type” assumptions are not related to Dodson’s equation, which Faak et al. misunderstood in their commentary.

Popular geometries for diffusion modeling include the plane-sheet, cylinder, and sphere, approximating crystal shapes in one- (1-D), two- (2-D), and three-dimensional spaces (3-D), respectively. The spherical (3-D) and cylindrical (2-D) geometries give rise to apparent “advection” terms in the diffusion equations and hence show relatively faster diffusion than the plane sheet (1-D). This geometric effect on diffusive exchange has been well recognized in the community since the pioneering work of Dodson (1973). Hence, it is important to choose appropriate geometries for accurate estimation of cooling rates. In general, the choice of geometry should closely resemble the crystal dimension, for instance, plane-sheets for bladed crystals (e.g., mica), cylinders for columnar crystals (e.g., apatite), and spheres for rounded crystals (e.g., olivine). Caution, however, must be exercised when modeling diffusion in minerals with anisotropy. A notable example is mica. Although mica is a sheet silicate, its effective shape is considered as an infinite cylinder for oxygen diffusion, because oxygen diffusivity parallel to the cleavage is about 3 to 4 orders of magnitude faster than that perpendicular (e.g., Fortier and Giletti, 1991).

3. Clarification of Faak et al.’s concerns

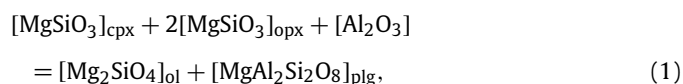
The major differences between S&L’s and Faak et al.’s approaches include four aspects: (1) Mg partitioning model (i.e., Mg-exchange thermometer), (2) compositional effect on Mg diffusion in plagioclase, (3) diffusion geometry, and (4) Dodson-type assumptions (see Section 2). With the Dodson-type assumptions, Faak et al. (2015) numerically modeled the Mg closure profiles of single plagioclase crystals using the 1-D diffusion equation to estimate the cooling rates of Hess Deep gabbros. Their Mg partitioning model was calibrated only at 1100–1200 °C and strongly relies on the knowledge of silica activity (see S&L’s Fig. 1a). The

strong compositional effect on Mg diffusivity in plagioclase (see S&L’s Eq. (A.14)) was neglected in their diffusion modeling.

Taking advantage of the differential diffusive responses of Mg and REE, S&L determined the crystallization temperatures and cooling rates simultaneously for individual Hess Deep samples through a combination of bulk-diffusion modeling and numerical inversions. Their bulk-diffusion modeling was achieved by considerable numerical simulations of bulk closure temperatures in various aggregates of spherical (3-D) plagioclase and clinopyroxene. For accurate cooling rate estimation, S&L also calibrated a new Mg-exchange thermometer for a broad range of temperatures (800–1430 °C) and new composition-dependent Arrhenius equations for Mg and REE diffusivities in plagioclase. Yet Faak et al. erroneously disapproved all effort made by S&L as well as their chemical analysis. Following are our detailed explanations for clarifying Faak et al.’s misunderstandings on S&L’s (1) thermometer calibration, (2) diffusion equation, and (3) chemical analysis.

3.1. Thermometer calibration

The Mg-exchange thermometer of S&L is distinct from that of Faak et al. by the exchange reaction and calibration strategy. Because natural petrological systems often involve multiple components and phases, equilibrium exchange of an element of interest between two coexisting phases does not necessarily follow a unique path. For instance, we could name another reaction to describe Mg exchange between clinopyroxene (cpx) and plagioclase (plg).



which involves olivine (ol) and orthopyroxene (opx) instead of silica activity. However, this reaction is not practically useful because coexisting ol-opx are often absent in oceanic gabbros (e.g., Lissenberg et al., 2013). Given that S&L’s reaction equation (cf. their Eq. (3a)) only involves plagioclase and clinopyroxene, the two major cumulus minerals in oceanic gabbros, it provides a much simpler mechanism for the thermometer calibration. In addition, silica activity is ambiguous to constrain since it could be defined in different ways depending on whether melts are present, as exactly discussed in Faak et al.’s Point II.

Ideally, exchange thermometers should be calibrated through well-controlled phase equilibria experiments for a broad range of temperatures, pressures and compositions. However, this is not the case for either Faak et al.’s or S&L’s Mg-exchange thermometers. Faak et al.’s thermometer was calibrated through diffusion experiments at 1100–1200 °C and 1 bar, assuming chemical equilibrium at plagioclase rims. Instead, S&L took an indirect approach by combining the phase equilibria experiments (800–1430 °C, 1 bar–27 kbar) with coexisting plagioclase, clinopyroxene and silicate melt from the literature and the experimentally calibrated plagioclase–melt Mg partitioning model from Sun et al. (2017). A similar strategy has also been successfully implemented in previous studies for calibrating REE-exchange thermometers (Liang et al., 2013; Sun and Liang, 2015, 2017) through mineral–melt partitioning models (Sun and Liang, 2012, 2013a, 2013b, 2014; Yao et al., 2012; Sun et al., 2017). Regardless of calibration methods, key criteria of accurate thermometers are (1) whether they could reproduce the calibration dataset and more importantly (2) if they are testable by independent data. As shown in their Fig. 3, S&L’s Mg-exchange thermometer obviously satisfies these criteria.

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