LinT, a simplified approach to oxygen-isotope thermometry and speedometry of high-grade rocks: An example from ultrahigh-temperature gneisses of southern Madagascar

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ABSTRACT

Accurate and precise temperatures of metamorphism and magmatism can be obtained from O-isotope compositions of minerals using a system of linear equations that defines fractionation and mass balance of O isotopes among all minerals in a rock. This approach—linear-equation thermometry (LinT)—does not require that cooling rates or absolute O-diffusion parameters be known. If O diffusivities are known, closure temperatures for each phase can be calculated and inverted to determine a temperature-dependent cooling rate that is accurate to a factor of three. The highest and most precise temperatures will be preserved in rocks containing quartz and garnet, because O diffuses slowly in garnet and garnet-quartz O-isotope fractionation is relatively large. The technique will be most useful in rocks with low-variance mineral assemblages in which precise pseudosection analysis is inhibited by wide spacing of reactions in pressure-temperature space. Application of LinT to ultrahigh-temperature gneisses from Madagascar yields temperatures of 927 ± 31 °C, consistent with independent thermometry.

INTRODUCTION

Urey (1947) predicted the temperature (T)-dependent fractionations of O isotopes between coexisting phases and foresaw its application as a geothermometer, specifically to determine paleoseawater temperatures from fossils (Urey et al., 1951). The T dependence of O-isotope fractionation among minerals was first experimentally calibrated in the 1950s through the 1970s (e.g., Clayton and Epstein, 1958; Taylor et al., 1963; O'Neil and Taylor, 1967; Clayton et al., 1972; Bottinga and Javoy, 1973) and allowed calculation of crystallization T for igneous and metamorphic rocks.

Retrograde diffusion can partially reset the isotopic compositions of minerals, leading to T estimates lower than those of peak metamorphism, however the O-isotope disequilibrium can be exploited to calculate cooling rates if diffusivities are known (e.g., Javoy, 1977). Using the Dodson (1973) equations for instantaneous closure T (T_c), Giletti (1986) presented a mathematical approach for quantifying O-isotope disequilibrium as a function of cooling rate, grain size, and mineral modes; this was used to calculate cooling rates for natural granites based on the magnitude of O-isotope disequilibrium among quartz, feldspar, and Ca-amphibole. Eiler et al. (1993) and Kohn and Valley (1998) forward-modeled O-isotope redistribution during cooling of crystalline rocks using the full T dependence of O diffusion rate, rather than the instantaneous T_c used by Giletti (1986). The full-diffusion models provide the advantage of predicting

O-isotope zoning in phases, but bulk-mineral O-isotope compositions are nearly identical to those predicted by the instantaneous T_c method.

The method of Giletti was expanded by Farquhar et al. (1993, 1996), who presented the method of isotope-exchange trajectories (IETs). An IET is an equation that describes O-isotope fractionation between a mineral and the bulk rock as a function of T. The T at which the IET agrees with the measured mineral $\delta^{18}O$ is taken to be T_c of that mineral; each mineral has a different IET. If T_c of one mineral is sufficiently high, IETs can be used to calculate crystallization T of plutons and peak T of ultrahigh-T (UHT) metamorphic rocks.

Similarly, Hoernes et al. (1994, 1995) and Hagen et al. (2008) recognized that the low diffusivities of O in garnet and kyanite-sillimanite allow peak or near-peak T of crystalline rocks to be extracted from garnet–(kyanite-sillimanite)–whole-rock fractionations, without the need to correct for retrograde O diffusion in other phases. The slope of the line connecting $\delta^{18}O_{garnet}$ and $\delta^{18}O_{whole-rock}$ in A-$\delta^{18}O$ space (A is the O-isotope fractionation factor between a phase and quartz) will be proportional to the last T at which the minerals equilibrated.

The method used in this paper is analogous to the methods of Farquhar et al. (1993) and Hoernes et al. (1994), though the mathematical formulation differs. We reiterate the findings of previous studies: O isotopes can be used to extract accurate and precise T of the highest-grade rocks independent from—and complementary to—other thermometric techniques. Mineral diffusivities do not need to be known to calculate T, but if they are known, nonlinear cooling rates can also be calculated.

A LINEAR-EQUATIONS APPROACH TO O-ISOTOPE THERMOMETRY (LinT)

Assuming that minerals have an essentially instantaneous T_c for O diffusion (Dodson, 1973; Giletti, 1986) and that the rock remains closed to O-isotope exchange during cooling, the last T at which all minerals in the rock were in O-isotope equilibrium—and the O-isotope composition of the minerals at that T—can be uniquely determined from measured mineral $\delta^{18}O$, mineral modes, and the O-isotope fractionation factors among the minerals. This solution comes from solving a set of linear equations that describe the O-isotope fractionations and O mass balance among the minerals. For a rock with three minerals, of which mineral 3 (min3) has the highest T_c, the set of equations can be:

$$\delta^{18}O_{min1}^{T} - (A_{min3} - A_{min1})/T^2 = \delta^{18}O_{min3}^{measured}, \qquad (1)$$

$$\delta^{18}O_{min2}^{T} - (A_{min3} - A_{min2})/T^2 = \delta^{18}O_{min3}^{measured}, \text{ and} \qquad (2)$$
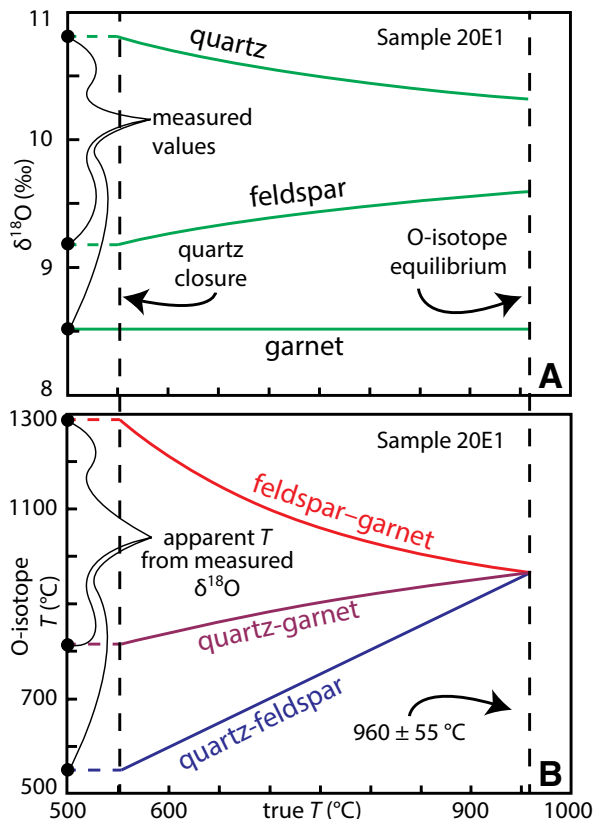
Figure 1. Graphic illustration of system-of-linear-equations approach to high-temperature oxygen-isotope thermometry (LinT) on an ultrahigh-temperature gneiss from southern Madagascar (this study). A: Constrained by mass balance and equilibrium fractionation, $\delta^{18}O_{quartz}$ and $\delta^{18}O_{feldspar}$ are adjusted ($\delta^{18}O_{garnet}$ is held constant, because it has slowest relative oxygen diffusivity) until quartz-garnet and feldspar-garnet temperatures (T) agree. This is the last T at which all three minerals were in equilibrium: either closure T of garnet or peak T of metamorphism, whichever is lower. B: Same data shown in A recast as O-isotope apparent T against "true T:" LinT works by solving where all O-isotope T values agree.
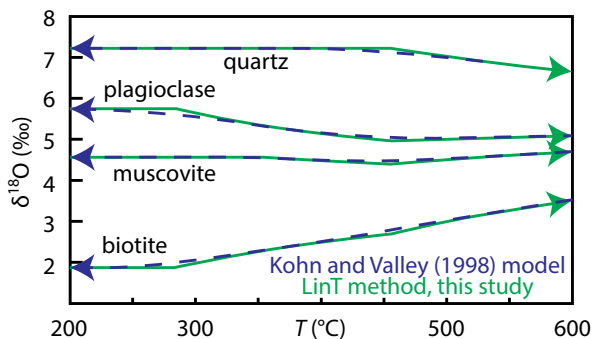

Figure 2. Comparison of forward model of O-isotope reequilibration during cooling (Kohn and Valley, 1998; blue)—using whole-rock composition, temperature (T)-dependent parameterizations of O diffusivity in each mineral, O-isotope fractionation factors, mineral modes, and cooling rate to predict how O-isotope compositions change down-T—with method of this study (analogous to methods of Giletti [1986] and Farquhar et al. [1993]; green), which uses measured mineral $\delta^{18}O$, O-isotope fractionation factors, and modes to calculate $\delta^{18}O$ of each mineral up-temperature. Models differ by ≤0.1‰ for each mineral at all temperatures.

$$x_{min1} \times \delta^{18}O_{min1}^{at\ T} + x_{min2} \times \delta^{18}O_{min2}^{at\ T}$$

$$= x_{min1} \times \delta^{18}O_{min1}^{measured} + x_{min2} \times \delta^{18}O_{min2}^{measured}, \qquad (3)$$

where T is in K; x = mineral mode × O atoms per formula unit / molar volume; and A_{minX} = O-isotope fractionation factor between mineral X and quartz. Equations 1 and 2 derive from the fundamental relationship of O-isotope fraction as a function of T: $\Delta^{18}O = A/T^2$. For n minerals in a rock, there are $n!/[2(n-2)!]$ possible equations of this form; only $n-1$ are necessary to define the O-isotope fractionations among all phases. Equation 3 is a mass balance of O isotopes among the minerals in which O diffused during cooling. For each additional mineral in the rock beyond three, additional terms of the form $x_{min} \times \delta^{18}O_{min}$ must be added to both sides of Equation 3 (example sets of equations for four- and five-mineral rocks are given in the GSA Data Repository[1]).

Temperatures calculated from these equations will be the peak T of metamorphism (or crystallization T of a pluton) or T_c^{min3}, whichever is lower. Uncertainties in the calculated T are a function of analytical uncertainty in $\delta^{18}O$, uncertainty in fractionation factors, magnitude of fractionation among minerals, and uncertainties in mineral modes; all uncertainties are easily propagated by Monte Carlo simulation. A graphical illustration of the LinT method applied to an UHT garnet-quartz-feldspar gneiss from Madagascar (data described below) is shown in Figure 1.

If the effective grain size of each mineral and diffusion parameters are known, the instantaneous cooling rates (dT/dt) at each mineral T_c can be calculated using the equations of Dodson (1973). If dT/dt is calculated at T_c for multiple minerals within a rock, dT/dt can be defined as a function of T and integrated with respect to time to yield a temperature-time path for the rock—a fundamental goal in petrology (Sharp, 1991).

COMPARISON OF LinT WITH MODELS AND REAL ROCKS

To evaluate the accuracy of the LinT approach, we (1) apply LinT to synthetic data output from cooling models using fully temperature-dependent expressions for O diffusion (Kohn and Valley, 1998), and (2) apply LinT to three UHT gneisses from Madagascar and compare the resulting temperatures to independent temperature estimates from the same terrane.

Comparison with the Model Schist of Kohn and Valley

Kohn and Valley (1998) forward-modeled O-isotope reequilibration during cooling of a hypothetical quartz-plagioclase-muscovite-biotite schist using a fully temperature-dependent description of O diffusion rate in each mineral and allowing zoning to develop within each mineral. LinT was applied to the final O-isotope compositions and modes of each mineral in the Kohn and Valley model (i.e., the values that would have been measured, had it been a real rock). From LinT, the last temperature at which all minerals were in equilibrium was 456 °C (interpreted to be T_c^{quartz}), and the last temperature at which muscovite, biotite, and plagioclase were in equilibrium was 353 °C (interpreted to be $T_c^{muscovite}$). These T_c values are nearly identical to the Dodson T_c values calculated from the dT/dt and grain sizes prescribed in the forward model: 470 °C (quartz) and 335 °C (muscovite). Plotting each of the linear equations in $\delta^{18}O$-T space (effectively the same as generating IETs by the method of Farquhar et al. [1993]) shows that the calculated $\delta^{18}O$ of all minerals at all temperatures differs by <0.1‰ from the full diffusion model of Kohn and Valley (Fig. 2). Assuming a hypothetical analytical uncertainty of 0.1‰ (2σ) for $\delta^{18}O$ in each mineral, the precision of the T_c values calculated by LinT is ±12 °C (T_c^{quartz}) and ±48 °C ($T_c^{muscovite}$).

Instantaneous dT/dt values calculated at the LinT T_c values of quartz and muscovite are 2.2 and 13 °C/m.y., respectively. The prescribed dT/dt

in the Kohn and Valley forward model was a constant 5 °C/m.y., suggesting that dT/dt calculated by inversion of LinT temperatures should only be considered accurate within a factor of three (similar to the 0.5-log_{10}-unit accuracy suggested for cation diffusion by Spear and Parrish [1996]). This is due to the exponential relationship between closure T_c and dT/dt in the Dodson (1973) equation; small uncertainties in closure temperature correspond to large uncertainties in dT/dt.

Comparison with UHT Granulites of Madagascar

Ultrahigh-T granulites in the Anosyen domain of southern Madagascar are metapelitic, calc-silicate, and felsic paragneisses metamorphosed between 590 and 510 Ma (e.g., Holder et al., 2018). Because the peak temperatures have been consistently estimated at 900−1000 °C (Jöns and Schenk, 2011; Boger et al., 2012; Horton et al., 2016; Holder et al., 2018), these rocks provided an ideal case study to compare results of *LinT* with other modern thermometric techniques in UHT metamorphic rocks. We chose three samples from near Tôlanaro (city; see the Data Repository) because of their simple, un-retrogressed mineral assemblages; all three are garnet–quartz–ternary-feldspar gneisses with <5 total vol% other phases (mainly sillimanite, ilmenite, and Al-spinel).

Twenty-five (25) cm^3 of each sample was crushed and separated into 350–850-μm-diameter fragments. Unweathered, inclusion-free grains of quartz, perthite, and garnet were then picked by hand. Aliquots of 1−2 mg of each mineral were measured at the Center for Stable Isotopes at the University of New Mexico, Albuquerque (USA), using the laser-fluorination method of Sharp (1990). The analytical precision of the measurements was ~0.1‰ based on repeated analyses of a reference quartz measured periodically throughout the analytical session. Quartz and feldspar mineral modes were determined from pixel counting of backscattered electron images of whole thin sections. For comparison, two O-isotope fractionation factors were used for quartz-garnet: that of Sharp (1995) and that of Valley et al. (2003), the former yielding higher temperatures. The quartz-feldspar fractionation factor used is from Clayton et al. (1989) for Ca/(Ca+K+Na) = 0.15 ± 0.05. Results are shown in Table DR1 in the Data Repository and in Figure 3.

Using the Valley et al. (2003) value for quartz-garnet fractionation, the calculated temperatures at which the quartz, feldspar, and garnet were

in O isotope equilibrium ranges from 899 to 960 °C with a weighted mean of 927 ± 31 °C (2σ). Using the quartz-garnet O isotope fractionation factor of Sharp (1995), temperatures range from 1009 to 1069 °C with a weighted mean of 1037 ± 34 °C (2σ). The Valley et al. calibration yielded temperatures in agreement with peak metamorphic temperatures calculated from feldspar-solvus thermometry (>930 °C; Holder et al., 2018), Zr-in-rutile thermometry (930 ± 45 °C; Horton et al., 2016), and pseudosections (880–920 °C: Boger et al., 2012; or >930 °C: Holder et al., 2018), whereas the Sharp calibration yielded higher temperatures. The agreement of LinT temperatures with independent estimates of peak metamorphic temperature suggests that T_c^{garnet} is equal to or greater than the peak temperature of metamorphism, consistent with interpretations of previous studies (Hoffbauer et al., 1994; Sharp, 1995). The temperatures calculated from $\delta^{18}O_{\text{feldspar}}$–$\delta^{18}O_{\text{quartz}}$ ($\Delta^{18}O_{\text{feldspar-quartz}}$) are 498–638 °C and are interpreted to be the T_c values of quartz. The temperatures calculated from $\Delta^{18}O_{\text{quartz-garnet}}$ are 740–819 °C (calibration of Valley et al., 2003; Fig. 3) or 810–896 °C (calibration of Sharp, 1995); these are minimum temperatures of metamorphism, because $\Delta^{18}O_{\text{quartz-garnet}}$ can only increase during cooling (Fig. 1).

Instantaneous dT/dt can be calculated from T_c^{quartz} using the equations of Dodson (1973). The grain sizes for the minerals in each sample are similar, but each mineral has a distribution of grain sizes such that the "effective grain radius" to be used in the Dodson equation is unclear. To account for this variability, we propagate a large uncertainty in grain radius (0.6 ± 0.3 mm) through each cooling-rate calculation; this is the largest contribution to the uncertainty in dT/dt presented below. Quartz was modeled as a plane sheet, because O diffusion in quartz along the c-axis is much faster than perpendicular to the c-axis (e.g., Sharp et al. 1991). The lack of hydrous, retrograde minerals in these rocks suggests that these rocks did not have a free-fluid phase during cooling. Using the "dry" diffusion parameters of alpha quartz (parallel to the c-axis) from Sharp et al. (1991) yields dT/dt of 1–55 °C/m.y. at the times of quartz closure. This range of dT/dt calculated for the three samples reflects the range in permissible values for the combined uncertainties in grain size, T_c values, and $\delta^{18}O$ of minerals.

For garnet, the agreement of LinT with independent estimates for peak metamorphic temperatures indicate that T_c^{garnet} is equal to or greater than the peak temperature or metamorphism. No "dry" diffusion data are available for almandine-rich garnet, but if they were, they could be used to calculate minimum dT/dt.

The 75 m.y. difference in age between the onset of cooling at 555 ± 11 Ma (oldest monazite leucosome date; T of ~930 °C; Holder et al., 2018) and 530–510 Ma monazite He dates (Montel et al., 2017; 300 ± 50 °C) implies an average dT/dt of 17 ± 7 °C/m.y., consistent with the range of permissible dT/dt calculated from T_c^{quartz}. In more mineralogically complex samples, LinT could be used to calculate different instantaneous dT/dt for each mineral T_c, defining dT/dt as a function of T. Alternatively, in rocks with effectively binary mineral assemblages in which one mineral is much more abundant than the other (such as minor magnetite in a marble), dT/dt can be evaluated as a function of T by measuring $\delta^{18}O$ of the minor phases as a function of grain size (Sharp, 1991). Although dT/dt values inverted from LinT temperatures are imprecise, the uncertainties are similar in magnitude to those obtained by cation diffusion chronometry (e.g., Spear and Parrish, 1996); dT/dt values inverted from LinT temperatures will be most useful for identifying periods of anomalously fast cooling relative to the time-integrated average (e.g., Spear and Parrish, 1996) or "short-duration" metamorphic events (Viete and Lister, 2016).

CONCLUSIONS

We presented a simplified mathematical approach (LinT) to the application of O-isotope thermometry that requires only mineral $\delta^{18}O$, modes, and fractionation factors. Grain size, cooling rate, and diffusion parameters are not needed for the extraction of meaningful temperature
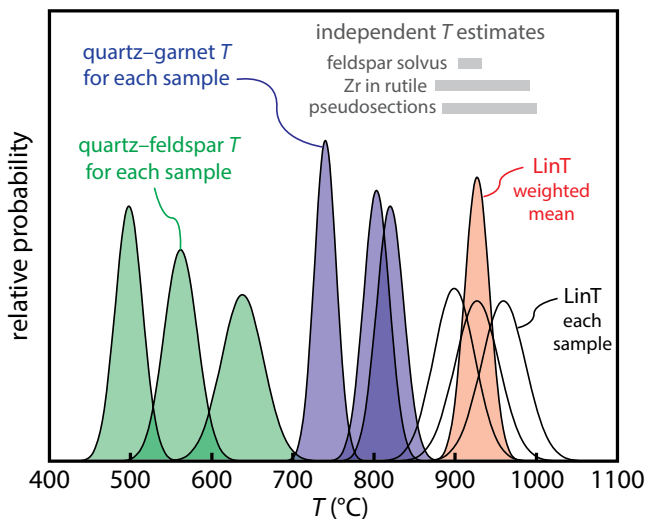

Figure 3. Despite different amounts of retrograde O-isotope exchange between quartz and feldspar in each sample (differences among quartz-feldspar and quartz-garnet apparent temperatures [T] among samples), O-isotope T values calculated by linear-equation thermometry (LinT) agree with each other and with independent T estimates (Boger et al., 2012; Horton et al., 2016; Holder et al., 2018).

information, but if they are known, mineral T_c values can be used to calculate temperature-dependent cooling rates to within a factor of two to three. Temperatures were calculated for UHT gneisses from south Madagascar (921 ± 30 °C or 1037 ± 34 °C, depending on the calibration used) consistent with two-feldspar thermometry, Zr-in-rutile thermometry, and pseudosections. Cooling rates calculated from T_c^{quartz} are consistent with the cooling rate calculated from thermochronology.

The most-precise O-isotope T will be obtained from rocks that contain minerals with a large range in fractionation factors, such as quartz with garnet, pyroxene, or kyanite-sillimanite (Fig. DR1 in the Data Repository; Sharp, 1995; Valley et al., 2003; Valley, 2003). The highest T values will be obtained if the diffusion rate of O in at least one mineral is slow enough that its T_c is equal to or greater than the peak T of metamorphism or the emplacement T of a pluton. The best candidates for this are garnet and kyanite-sillimanite (Hoffbauer et al., 1994; Sharp, 1995; this study). O-isotope thermometry is particularly well suited to high-T quartz-garnet–bearing rocks with low-variance mineral assemblages (rocks that are problematic for precise phase-equilibria modeling).

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