

Constraining the time scales of magmatic differentiation with U-Pb zircon geochronology

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ABSTRACT

Quantifying the time scales of magmatic differentiation is critical for understanding the rate at which silicic plutonic and volcanic rocks form. Directly dating this process is difficult because locations with both clear evidence for fractional crystallization and the accessory phases necessary for radiometric dating are rare. Early zircon saturation, however, appears to be characteristic of many high-K, arc-related melts due to their generally elevated initial Zr concentrations. Thus, high-K plutonic series are ideal candidates to study the time scales of magmatic differentiation using zircon U-Pb geochronology. This study focuses on the Dariv Igneous Complex in western Mongolia where early saturation of zircon in a suite of cogenetic, upper crustal (<0.5 GPa) igneous rocks ranging from ultramafic cumulates to evolved granitoids allows us to date magmatic differentiation. Crystallization ages from six samples across the sequence indicate that magmatic fractionation from a basalt to high-silica (>65 wt% SiO₂) melt occurred in $\leq 590 \pm 350$ k.y. This estimate is greater than modeled time scales of conductive cooling of a single intrusion and physical segregation of minerals from a melt, suggesting that continued influx of heat through magmatic activity in the complex may have prolonged cooling and thus time scales associated with the production of silica-enriched melts.

INTRODUCTION

Differentiation of mantle-derived, basaltic magmas to produce more silicic compositions is an essential process to produce the compositional stratification observed in arc crustal sections characterized by a felsic upper crust and mafic lower crust (DeBari and Greene, 2011; Jagoutz, 2014). The process is well studied both experimentally (Sisson et al., 2005; Nandedkar et al., 2014) and through field-based studies of crustal sections (Greene, 2006; Jagoutz, 2010). However, the time scales over which individual bodies of magma differentiate (let alone the entire arc crust) remain poorly understood. Constraints on the time scales of magmatic differentiation come primarily from U-series disequilibrium characterization of modern (<300 ka) volcanic rocks (see the review by Hawkesworth et al., 2000). This approach, however, relies on the assumptions that different volcanic rocks came from the same differentiating magma and that the observed U-series disequilibria is related to the segregation of crystals from melt, which are difficult to verify.

Alternatively, in exhumed plutonic rocks the connection between derivative melts and cumulates may be clearer. For example, the southern portion of the Adamello batholith (Italy) consists of an incrementally assembled, composite body

of ultramafic, gabbroic, tonalitic, and granodioritic lithologies, which formed through fractional crystallization of a mantle-derived melt in the middle to lower crust with variable amounts of crustal assimilation (Ulmer et al., 1983). As such, it has been the focus of intensive geochronological studies to understand the time scales of melt evolution (Schaltegger et al., 2009; Schoene et al., 2012). One limitation of studying the Adamello and similar calc-alkaline plutonic series is that zircons are plentiful only in felsic lithologies and confined to late-stage, pegmatitic segregations of gabbros (Schoene et al., 2012; Samperton et al., 2015). Similarly, zircons have been found in ultramafic/mafic cumulates of the tholeiitic Bushveld Complex, but only in late-crystallizing, highly fractionated intercumulus melts (Zeh et al., 2015). The late saturation of zircon in most crystallizing magmatic systems thus restricts the application of U-Pb zircon geochronology in evaluating time scales of differentiation to a limited compositional range of high-SiO₂ melts and thus a narrow differentiation interval.

In contrast, saturation of zircon early in the crystallization sequence may be more common in arc-related high-K (though non-peralkaline) igneous series. Although experimental studies suggest that alkali-rich melts have unfavorable compositions for high-temperature zircon

saturation due to high concentrations of network-modifying cations (i.e., Na + K) and therefore low degrees of polymerization (Watson and Harrison, 1983; Gervasoni et al., 2016), high-K mafic melts are generally enriched in Zr and commonly contain magmatic zircons (e.g., Bergman, 1987) (Fig. 1A). The generally elevated Zr content (up to ~1000 ppm) of high-K basalts is likely an important driver of early zircon saturation. If these melts evolve toward a silica-saturated eutectic or minimum during cooling, closed-system crystallization will increase both melt polymerization and Zr concentrations, enhancing the potential for zircon crystallization. Early saturation of zircon in high-K melts is also suggested by the geochemistry of the plutonic record (Fig. 1B). High-K plutonics and modeled melt trajectories generally show elevated Zr contents at lower SiO₂ as compared to more typical calc-alkaline plutonic series (Fig. 1B). However, zircon saturation (indicated by a decrease in Zr concentrations) occurs at lower SiO₂ contents (~55 wt%) for the high-K plutonics than those of calc-alkaline series (>65 wt% SiO₂) (Fig. 1B). Therefore, high-K intrusive suites may be ideal localities in which to apply U-Pb zircon geochronology in order to constrain the time scales of fractionation over a wide range of melt SiO₂ contents, melt fractions, and temperatures.

Here, we investigate a cogenetic high-K plutonic sequence composed of ultramafic cumulates to granitoids in the Dariv Igneous Complex (DIC) in western Mongolia (Bucholz et al., 2014a) where zircon is an early crystallizing phase, first observed as a primary magmatic mineral in high-Mg lamprophyre dikes and ultramafic cumulates. As such, the DIC affords a unique opportunity to utilize high-precision U-Pb zircon geochronology to constrain the time scales of magmatic differentiation.

GEOLOGIC BACKGROUND

The Dariv Range (southwestern Mongolia) exposes the contact between the Altai Allochthon (Proterozoic high-grade metamorphic rocks) and the Lake Terrane (an Ediacaran to

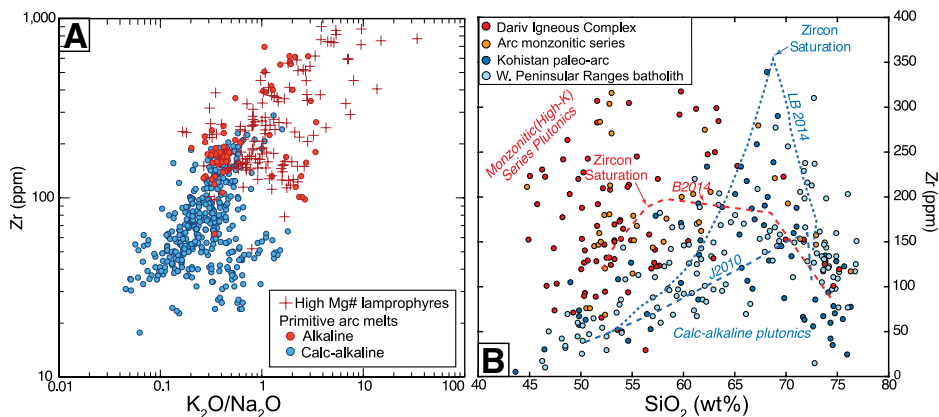


Figure 1. A: Zr (ppm) versus K₂O/Na₂O of primitive or high-Mg# basalts to basaltic andesites. Primitive arc melt compositions are from Schmidt and Jagoutz (2016). Lamprophyres are from the GEOROC database (<http://georoc.mpch-mainz.gwdg.de/georoc/>) and were filtered to only include analyses with major oxide totals between 98 and 102 wt%, molar Mg/(Mg + Fe_{total}) > 0.6, and non-Archean samples. **B:** Zr (ppm) versus SiO₂ (wt%) for plutonic rocks. Data for Dariv Igneous Complex (DIC; western Mongolia), Kohistan paleo-arc (Pakistan), and western Peninsular Ranges batholith (western North America) are from Bucholz et al. (2014b), Jagoutz (2010), and Lee et al. (2007), respectively. Modeled high-K and calc-alkaline melt trajectories are shown as red and blue dashed lines, respectively (J2010—Jagoutz [2010]; B2014—Bucholz et al. [2014b]; LB2014—Lee and Bachmann [2014]). Arc monzodiorites and (quartz) monzonites are from GEOROC database (analyses from western Pacific, Aleutians, western United States, South America, and Mediterranean).

early Paleozoic island arc system) (Dijkstra et al., 2006). The kilometer-scale DIC belongs to the latter and comprises of a suite of high-K plutonic rocks, including phlogopite-bearing wehrlites and clinopyroxenites, monzogabbros, monzodiorites, (quartz) monzonites, and late-stage felsic and lamprophyre dikes that were emplaced at 0.2–0.5 GPa (see Bucholz et al. [2014a] for detailed field and petrographic descriptions). The observed lithologic variability, petrographic observations, and mineral and whole-rock geochemistry suggest that the plutonic rocks can be ascribed to a common fractionation sequence defined by olivine + clinopyroxene ± Fe-Ti oxides → biotite + apatite ± titanite ± zircon → K-feldspar + plagioclase → amphibole + quartz (Bucholz et al., 2014a, 2014b). Several primitive lamprophyre dikes have compositions appropriate to be parental to the observed cumulate lithologies, and their liquid line of descent was modeled by Bucholz et al. (2014b). Major and trace element trends of the sequence appear to be dominantly controlled by fractional crystallization. Although feldspar-bearing plutonic lithologies range from cumulative to melt-like in character, some of the monzodiorites, most of the quartz monzonites, and all of the aplite dikes have major and trace element compositions that suggest that they are differentiated melts.

PETROGRAPHY AND SAMPLE SELECTION

Notably, zircon is an early-crystallizing mineral in the fractionation sequence that defines the DIC, observed as a primary magmatic

phase in both ultramafic cumulates (Fig. 2B) and high-Mg lamprophyre dikes (Fig. 2A). In more evolved monzogabbros, monzodiorites, and (quartz) monzonites, zircons are abundant (Fig. 2C; Fig. DR6 in the GSA Data Repository¹). In all lithologies, textural observations, such as enclosure within clinopyroxene and biotite, confirm that zircon is magmatic in origin and crystallized either before or contemporaneously with the volumetrically dominant mineral phases (Fig. 2).

Six samples were selected for U-Pb zircon geochronology from an ~250-m-thick section of the DIC with continuous exposure of ultramafic cumulates to quartz monzonites cross-cut by both basaltic and felsic dikes (Fig. 3A). We selected samples that encompassed the range of compositions observed in the field, from primitive basaltic dikes to ultramafic and mafic cumulates to evolved “melt-like” lithologies. Of the coarse-grained lithologies, two cumulate samples (MO-11-10, phlogopite clinopyroxenite; MO-11-12, monzogabbro) and two “melt-like” lithologies (MO-11-14, monzodiorite; MO-11-26, quartz monzonite) were selected. Two dikes that cross-cut the sequence were also dated: a primitive lamprophyre (MO-11-19) similar in composition to the estimated parental melt for the sequence and a felsic dike (MO-11-16).

¹GSA Data Repository item 2017003, field relationships, sample descriptions, geochronological and modeling methods, and CL images of zircons, is available online at www.geosociety.org/pubs/2017.htm or on request from editing@geosociety.org.

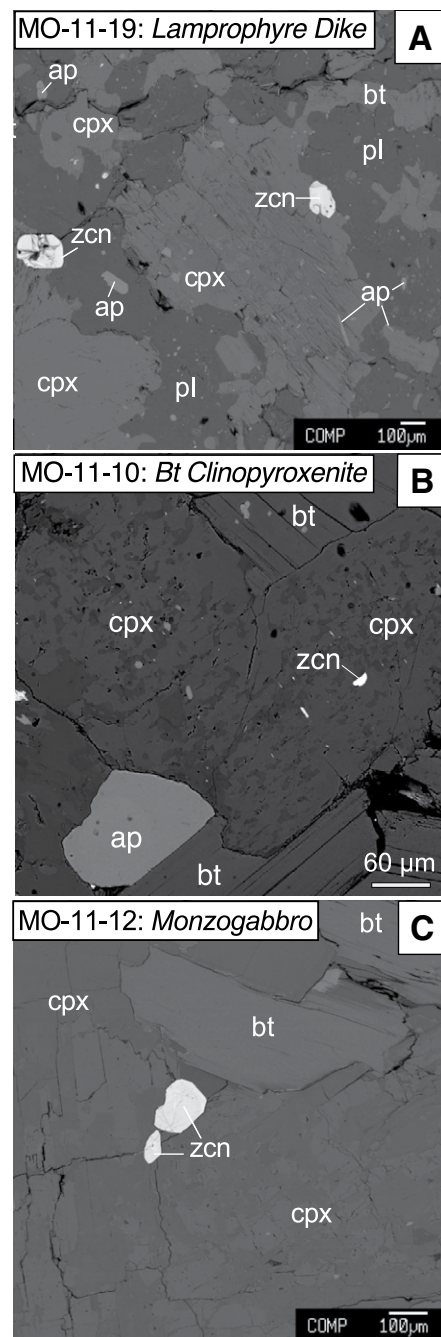


Figure 2. Backscatter electron photomicrographs of selected samples showing zircon textural relationships with other phases. A: Sample MO-11-19. B: Sample MO-11-10. C: Sample MO-11-12. Abbreviations: zcn—zircon; bt—biotite; cpx—clinopyroxene; pl—plagioclase; ap—apatite.

U-Pb ZIRCON GEOCHRONOLOGY

Zircons were analyzed using chemical abrasion–isotope dilution–thermal ionization mass spectrometry (CA-ID-TIMS). Detailed methods, sample locations, isotopic data, and cathodoluminescence (CL) images can be found in the Data Repository. Because the ²³⁸U–²⁰⁶Pb isotopic system gives the most precise date for rocks of this age, we use Th-corrected ²⁰⁶Pb/²³⁸U dates for

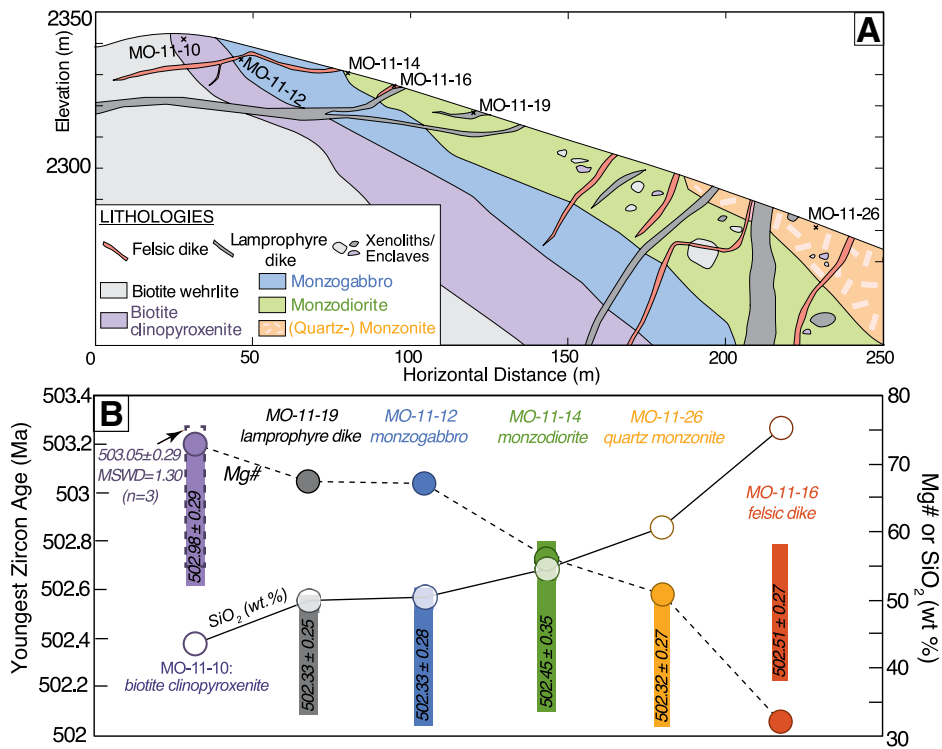


Figure 3. A: Detailed cross-section across high-K fractionation sequence with locations of analyzed samples. Lithologies become more evolved left to right, from phlogopite wehrlites to quartz monzonites. Contacts are drawn as sharp lines for clarity, but boundaries between lithologies are gradational. B: U-Pb geochronology and whole-rock data for analyzed samples showing Th-corrected $^{206}\text{Pb}/^{238}\text{U}$ dates (in Ma) for individual zircons. Colored bars represent youngest zircon ages from each sample with 2σ uncertainty. Weighted mean for sample MO-11-10 is shown with internal uncertainties (MSWD—mean square of weighted deviates). Whole-rock Mg# and SiO_2 (wt.%) are from Bucholz et al. (2014b).

all of our interpretations and present 37 single-grain analyses in Figure DR1 and in Table DR1 in the Data Repository. Our interpreted crystallization ages for the samples are in Figure 3B.

Both the quartz monzonite and the lamprophyre dike contain zircon age dispersion beyond what can be explained from analytical uncertainty alone (Fig. DR1). This apparent age dispersion may result from incorporation of xenocrysts (zircons inherited from country rock), antecrysts (zircons inherited by the magma but that originated in broadly the same magmatic system), protracted antecrystic zircon growth, or a combination of all of these processes. All 37 zircons analyzed in this study appear to be either auto- or antecrysts. It is unlikely that the lamprophyre dike experienced protracted *in situ* crystallization due to its probable rapid cooling because of its small width (~60 cm) and elevated magmatic temperatures compared to its host rock. Further, field evidence for high-temperature mechanical mixing of multiple melt generations is abundant in the quartz monzonite, including mafic lamprophyre enclaves, diffuse intrusive contacts, and K-feldspar xenocrysts (Fig. DR2). Therefore, we prefer an antecrystic origin for the oldest grains in these samples and suggest that they were transported to the

emplacement depth from a long-lived magmatic system at depth. In general, we consider the youngest zircon grains from each sample to be representative of the age of final crystallization.

The youngest zircon dates in the monzogabbro, monzodiorite, and quartz monzonite overlap within uncertainty and likely represent the end of melt crystallization within the system at ca. 502.35 Ma. Because field evidence suggests that the felsic and lamprophyre dikes were intruded into a highly crystalline mush (Fig. DR2) and the ages of the youngest grains from the samples from these two dikes also overlap with those of the more evolved, coarsely crystalline plutonics (Fig. 3), we conclude that the final crystallization event occurred at 502.39 ± 0.20 Ma (average value of youngest zircons from five samples). We were able to extract and date three grains from the clinopyroxenite after considerable effort (see the Data Repository). The ages from these grains all overlap within uncertainty and give a weighted mean date of 503.05 ± 0.29 Ma (mean square of weighted deviates = 1.30). The youngest zircon age from the phlogopite clinopyroxenite is 502.98 ± 0.29 Ma (Fig. 3B). Both of these ages for the phlogopite clinopyroxenite are distinctly older than the other dated samples.

DISCUSSION

Time Scales of Magmatic Fractionation

The time difference (Δt) between the ages of the youngest grain of the most primitive cumulate (phlogopite clinopyroxenite) and that of the inferred final crystallization for the more evolved plutonic rocks is 590 ± 350 k.y. ($502.98\text{--}502.39$ Ma, 2σ errors summed in quadrature), providing a direct approximation of the time scale of melt differentiation and crystallization within this system. Assuming that crystal settling is an efficient process in low-viscosity basaltic melts and occurs on time scales more rapid than that of crystallization (Martin and Nokes, 1988), the rate of cooling is the primary control on the time scales associated with fractional crystallization. To place time constraints on the cooling history of the DIC, we modeled the closed-system thermal evolution of a 500–1000-m-thick sheet-like magma body emplaced at an initial temperature of 1200°C into a country rock at 400°C (see the Data Repository for details). The results of this modeling suggest that ~15–45 k.y. is required to cool the magma body to near its solidus temperature, which is a minimum time required for cooling to occur in a non-convecting, closed magma system. The modeled cooling time scale of tens of thousands of years is less than our minimum estimate of ~240 k.y. for melt differentiation to occur. This discrepancy suggests that either the dated grains from the phlogopite clinopyroxenite are antecrystic in origin and do not provide an *in situ* crystallization age for the cumulate, that zircon ages from this sample are too limited in number ($n = 3$) to resolve the full duration of crystallization, or that continued magmatic activity or recharge prolonged the cooling time scales on the order of several hundred thousand years. An antecrystic origin is unlikely as this sample is cumulative in nature and does not represent a melt with a complex mixing history. We cannot rule out the uncertainty associated with the small sample size of zircons from the biotite clinopyroxenite, and it is plausible that more zircon ages from this sample would lessen the discrepancy between our modeled cooling-induced crystallization time scales and those inferred from the geochronology. Due to these uncertainties, we consider the duration of 590 ± 350 k.y. as a maximum duration for fractional crystallization.

If crystallization rates in crustal intrusions are primarily a function of cooling and if the effects of decompression are negligible, rates of fractionation will be strongly dependent on the size and depth of the magmatic system as well as the dynamics of magma chamber replenishment (Annen et al., 2006). The detailed section we dated (Fig. 3A) is a small part of the DIC, which comprises many smaller intrusive bodies covering an area of $\sim 5 \times 10$ km and representing an ~6-km-thick crustal section (Bucholz et al.,

2014a). Although emplaced in the upper crust, the DIC likely experienced a prolonged thermal history due to the multiple intrusive events involved in its formation. After intrusion of the melt batch that gave rise to the studied section, initial cooling below the liquidus promoted crystallization of ultramafic cumulates at high melt fractions and temperatures. This crystallization period may be recorded in the distinctly older population of zircons in the phlogopite clinopyroxenite. Subsequent prolonged cooling may have delayed the crystallization of more evolved lithologies and thus zircon crystallization. Indeed, cooling time scales of 10^5 – 10^6 yr, as recorded in the zircon geochronology of this study, are reasonable for multiply injected upper crustal intrusions (Blundy and Annen, 2016). Fractional crystallization of basalts in the lower crust may occur on much more protracted time scales due to greater ambient temperatures at depth and the potentially more frequent influx of hot basalts (Dufek and Bergantz, 2005; Annen et al., 2006).

CONCLUDING REMARKS

This study represents the first direct measurement of fractional crystallization time scales over a large range of melt compositions from basaltic to high-silica melts using high-precision CA-ID-TIMS U-Pb geochronology in zircon. The presented zircon ages from the DIC indicate that fractional crystallization of basaltic melts to produce more evolved granites in the upper crust occurred in $\leq 590 \pm 350$ k.y. This time scale is longer than that predicted through conductive cooling of a single intrusion, possibly due to continued magma influx into the upper crust, prolonging the time scales of crystallization and thus fractionation. Further detailed field studies of high-K intrusive suites demonstrating early saturation of zircon combined with high-precision U-Pb zircon geochronology will provide much needed temporal constraints on the efficiency of fractional crystallization at different crustal levels. In particular, younger complexes (<100 Ma) could yield even more precise ages, permitting the study of this process at the 10 k.y. time scale.

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