

Warm storage for arc magmas

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Felsic magmatic systems represent the vast majority of volcanic activity that poses a threat to human life. The tempo and magnitude of these eruptions depends on the physical conditions under which magmas are retained within the crust. Recently the case has been made that volcanic reservoirs are rarely molten and only capable of eruption for durations as brief as 1,000 years following magma recharge. If the "cold storage" model is generally applicable, then geophysical detection of melt beneath volcanoes is likely a sign of imminent eruption. However, some arc volcanic centers have been active for tens of thousands of years and show evidence for the continual presence of melt. To address this seeming paradox, zircon geochronology and geochemistry from both the frozen lava and the cogenetic enclaves they host from the Soufrière Volcanic Center (SVC), a long-lived volcanic complex in the Lesser Antilles arc, were integrated to track the preeruptive thermal and chemical history of the magma reservoir. Our results show that the SVC reservoir was likely eruptible for periods of several tens of thousands of years or more with punctuated eruptions during these periods. These conclusions are consistent with results from other arc volcanic reservoirs and suggest that arc magmas are generally stored warm. Thus, the presence of intracrustal melt alone is insufficient as an indicator of imminent eruption, but instead represents the normal state of magma storage underneath dormant volcanoes.

volcano | eruption | arc magma | zircon

etermining the timescale of magma storage and remobili-Determining the timescale of high standing the tempo and magnitude of volcanic eruptions (1–13). Whether a volcano can erupt is controlled by the recharge rate to the magma reservoir (13) (reservoir in this context refers to the portion of the igneous complex that is potentially eruptible), which in turn determines the duration of the "eruption window" [generally defined as the rheological state during which the subvolcanic reservoir is below $\sim 60\%$ crystals and hence capable of eruption (4)]. However, estimates for how long this eruption window remains open vary over four orders of magnitude; this suggests either profound problems in assumptions underlying one or more of these estimates or a continuum of physical mechanisms that resist formulation of a unified model for the state of magma reservoirs before eruption (1-13). The preservation of sharp compositional gradients in plagioclase phenocrysts, assumed to have crystallized >10 ka before eruption, has recently been interpreted to indicate that arc volcanic reservoirs characteristically remain in "cold storage" at temperatures below the eruption window, possibly below the solidus, and thus only capable of erupting during brief recharge events (<10 ka) (1). In contrast, zircon dating and heat budget considerations are difficult to reconcile with this scenario; instead, they are consistent with continuously partially molten reservoirs capable of erupting (i.e., with melt portion $\geq 40\%$) over long durations (>>10 ka) (6-12). Whatever these differences, all agree that understanding the thermal history of the magmatic reservoir is key to constraining the duration of the eruption window (3, 12).

Coupling zircon microgeochronology and geochemistry provides powerful insights into the preeruptive evolution of volcanic centers (14, 15), but thus far this approach has been applied either to volcanic or plutonic end-members. That intrusive magma volumes outweigh their extrusive counterparts is sufficient reason to assume that both may record different aspects of the reservoir's history (15–19); this is because melt-dominated volcanic rocks may only represent a volumetrically minor part of the magma reservoir, whereas plutonic rocks represent conditions in the crystaldominated bulk of the magma reservoir (18). To provide a physical context for our interpretive scheme, we point to simulations of Bergantz et al. (19) that show that the full extent of thermal excursions caused by recharge is only recorded in the immediately adjacent crystal-rich portions of the reservoir.

In this paper, we integrate high spatial-resolution U-series ages of zircon coupled with trace element geochemistry of both the extrusive dacite and cogenetic enclaves (Fig. S1). The enclaves from the Quaternary Soufrière Volcanic Center (SVC), Saint Lucia, Lesser Antilles (Fig. 1), studied here are remarkable in the level of detailed petrologic investigation they have experienced by previous researchers (20, 21). We focus on zircon rims to avoid any inherited cores, but note that due to the continuous growth of zircon and limited depth resolution of secondary ion mass spectrometer (SIMS) U-Th dating, our ages average over the outer ~4 μ m of each zircon.

SIMS U-Th zircon geochronology and trace element analysis of zircon (*SI Text*) were undertaken for a suite of SVC enclaves, yielding crystallization ages up to 130 ka (Fig. 2 and Figs. S1–S3; Datasets S1 and S2; sample description in *SI Text*), and the host lava of the Belfond Dome, which erupted at 13.6 \pm 0.4 ka (21) (Fig. 1). Ti contents of zircons in cogenetic enclaves show spikes in crystallization temperatures (Ti-in-zircon) (22, 23) (see *SI Text*, Fig. S4, and Dataset S3 for methodology and discussion

Significance

The increasingly popular notion that steady-state magma chambers are highly crystallized, and thus only capable of erupting during brief (<1 ka) reheatings, implies that melt detection beneath volcanoes warns of imminent eruption. By integrating the microgeochronology and geochemistry of zircons from lavas with those from components crystallized within the magma chamber and incorporated during eruption, we show that the Soufrière (Saint Lucia) volcanic reservoir was instead eruptible over long (>100 ka) timescales. Together with data from other volcanic complexes, we show that arc magmas may generally be stored warm (are able to erupt for >100 ka). Thus geophysical detection of melt beneath volcanoes represents the normal state of magma storage and holds little potential as an indicator of volcanic hazard.

The authors declare no conflict of interest.

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Fig. 1. (Left) Location of Saint Lucia in the Lesser Antilles with positions of active volcanoes within the arc; shades of blue relate to bathymetry (darker indicates deeper). (Right) Geological sketch map of the SVC with main eruptive phases and (U-Th/He) eruption ages (21). Stars represent sampling locations of pyroclastic flow deposits that are related to the SVC eruption phases (see color coding) and whose eruption was dated by (U-Th)/He zircon methods (21).

regarding a_{TiO2}) that coincide with the main eruption phases recorded in the SVC (phase 2 and 3; Figs. 1 and 2A). These spikes are interpreted as resulting from recharge of juvenile magma into the magma reservoir. We estimate a lower bound for the maximum temperature reached during the recharge event from the highest calculated value of 880 ± 20 °C (Fig. 2A), which would dissolve most of the preexisting plagioclase and quartz (24). Indeed, the subdued Eu/Eu* in the enclave zircon likely records the release of Eu^{2+} back into the melt (Fig. 2*B*) due to near-complete resorption of plagioclase. Because quartz and plagioclase are >40% of the mineral assemblage of the cogenetic enclaves (which at the typically lower temperatures recorded outside the spike intervals would be a crystal mush) (SI Text), the prograde thermal excursion locally reduced the proportion of crystals below 60% and thus placed at least part of the reservoir into the eruption window.

The overall longevity of the SVC magma reservoir is supported by the abundance of zircons extracted from the lava that show an extended and continuous period of crystallization spanning \sim 250 ka (Fig. 2 and Dataset S2). In contrast to the plutonic zircons, the volcanic zircons do not exhibit any resolved spikes in temperature or Eu/Eu*, instead showing a constant decrease in Eu/Eu*, suggesting continuous plagioclase fractionation throughout their history (Fig. 2B). These results show that the ultimately erupted part of the magma reservoir beneath the SVC remained above its solidus (~700 °C at 1-2 kbar; ref. 25; reservoir depth estimation following geophysical model from other volcanoes in the Caribbean arc from ref. 26) over at least the last 140 ka and likely back to 250 ka (Figs. 1 and 2). This contrasting behavior recorded by the lava and enclave zircons is best reconciled by the latter forming during cooling from a heating event associated with recharge by juvenile magma. The lack of temperature spikes in the volcanic zircons requires that that portion of the reservoir having remained above the solidus between rejuvenation events was of sufficient size that such recharge events did not affect the entire reservoir, but only its proximal surroundings. The evidence is consistent with the view that cogenetic enclaves reflect individual recharge events that heated only parts of the reservoir, whereas the volcanic zircons are derived from a thermally buffered reservoir that continuously retained melt. That is, the lava only experienced the recharge event that led to its removal from the reservoir. Our inference is consistent with simulations that show that the effects from recharge are likely to only be recorded in the crystal-rich part of the reservoir (19). The fact that older zircon rims are preserved throughout the magmatic history suggests that they were armored by (i) modal mineral phases or (ii) younger, subsequently resorbed rims, or that the melt-present part of the magma reservoir waxed and waned, permitting incorporation of some crystals from intermittently solidified margins before eruption (Fig. S1). Both components of the magma reservoir were sampled during the Belfond Dome eruption with most volcanic zircons crystallizing at steady state in supersolidus storage conditions, whereas enclave zircon crystallized during or immediately after transient rejuvenation from magma recharge. The coinciding trace elemental spikes in plutonic zircon and SVC eruption ages suggest to us a link between recharge and eruption. We note that zircon crystals from some cogenetic enclaves display correlated spikes in Ti-in-zircon temperature and Eu/Eu* (e.g., Gran 1.1, 1.4c, or 1.4d), whereas others do not (e.g., SLJL52; Fig. 3A), likely because zircon stability is a function of zirconium abundance [Zr], melt chemistry, and temperature (27, 28), and some cogenetic enclaves may not have saturated zircon at a sufficiently high temperature to record the spike. Indeed, we find a correlation between the magnitude of the temperature spike and the enclave [Zr] (Fig. 3B), illustrating that only the Zr-rich enclaves record the temperature excursion. The coupled geochemical and geochronological data from the SVC cogenetic enclaves provide the first direct constraints for remelting of a partially crystallized reservoir (crystal mush) (4, 29).

To constrain the minimum duration of the temperature spikes, we use the three cogenetic enclaves (Gran 1.1, 1.4d, and 1.10; Fig. 2) that record a well-defined excursion in Ti abundance and Eu/Eu* before the Belfond Dome eruption $(13.6 \pm 0.4 \text{ ka})$ (21). Using our zircon age and geochemistry data and the independently



Fig. 2. Zircon U-Th model ages (in ka) vs. Ti-in zircon temperatures (A) and Eu/Eu* (determined using the geometric mean) (B) for the cogenetic enclaves and the Belfond lava, with main SVC eruptive phases and (U-Th)/He eruption ages (21). Errors are 1σ.

known eruption age, we calculate a duration of 14 ± 2 ka for the cooling following the magma recharge before the Belfond Dome eruption (Fig. 4 and SI Text). This time interval also encompasses an eruption at ~20 ka, which formed the extensive pyroclastic flow deposits of the Belfond unit covering a large portion of southwestern Saint Lucia (21). We note that the duration calculated here represents the amount of time that the reservoir spends in the eruption window during the cooling that follows from the recharge event, and is the minimum estimate because cooling following the thermal spike was interrupted by the Belfond Dome eruption. Furthermore, zircon saturation (and hence zircon crystallization) was only reached during cooling following the rejuvenation, and therefore our data can only constrain a lower bound on the duration of magma remobilization, because any increase in temperature immediately raises the [Zr] required for saturation and thus zircon tends to become unstable during heating. Results from other volcanic centers suggest that the rejuvenation itself could occur

on a very short timescale (<1 y) (30). Our data are also best explained by the occurrence of a single, robust thermal spike (≥880 °C) rather than multiple heating episodes occurring on timescales that cannot be resolved by our zircon dates. In a multiple-spike model, the reservoir would have tended to dissolve zircon after each recharge, and thus the last rim would have crystallized just before, and not 15 ka before, eruption. The lower abundance of older enclave zircons supports the contention that recharge events destroyed earlier generations. That is to say, the cogenetic enclave population is likely biased toward younger ages because preexisting crystal-rich domains may become mixed into and assimilated by the magma or, alternatively, growth of new rims may obscure any previously recorded spikes. Zircon data from two of the studied enclaves also provide evidence for reservoir rejuvenations before the last spike. Enclave 1.4c contains zircons dating back to the reservoir reactivation during SVC eruption phase 2, with a spike defined by temperature and Eu/Eu* at ~60 ka (Fig. 2 and Dataset S2). By



Fig. 3. (*A*) Zircon U-Th model ages (ka) vs. Ti-in zircon temperatures for cogenetic enclaves Gran 1.1 (spike recorded) and SLJL52 (no spike recorded). (*B*) Correlation between the extent of spike recorded and Zr whole rock abundance (δ T °C is maximum – minimum temperature recorded in each enclave).

examining the range in zircon ages from 1.4c, we estimate a spike duration ranging from 20 to 40 ka for phase 2 (Fig. 2). The fact that many zircon crystals in enclave 1.4c postdate the SVC eruption phase 2 may reflect an incomplete chronostratigraphy for the SVC with not all eruptions being identified or dated. Enclave 1.3 contains even older zircons with a spike between 90 and 130 ka that may be related to the onset of the penultimate eruptive phase of the SVC (phase 2 in ref. 21) (Fig. 2 and Dataset S2). Additionally, we note that our calculated eruption window durations agree well with independently constrained cooling and crystallization timescales for the shallow-level calc-alkaline Elba Island pluton, where U-Pb zircon dating and thermal modeling indicated that rhyolitic magma remained at conditions conducive for eruptive melt extraction over $\sim 10-40$ ka (6, 7).

The eruption windows for the SVC, as determined using combined zircon geochronology and geochemistry (from ~ 15 up to ~ 40 ka), are more protracted than those proposed using diffusion profile modeling of compositional gradients in plagioclase from arc volcanic reservoirs (i.e., ≤ 10 ka) (1). However, studies using diffusion profiles cannot assign crystallization ages to individual plagioclase crystal because current U-Th disequilibrium dating techniques requires multigrain aliquots (up to several grams) (1). Therefore, it cannot be ruled out that those crystals exhibiting disequilibrium profiles formed immediately before eruption. That is to say, apparent diffusion profiles more likely reflect the individual plagioclase crystal formation age rather than storage conditions. Relating bulk plagioclase U-Th ages to diffusion profiles in individual crystals requires that they formed simultaneously, a requirement that is unlikely to be met in magmatic reservoirs. Support for this hypothesis comes from Sr profiles in plagioclase from Kameni Island (Greece) and Soufrière (Saint Vincent) (31), where some are in equilibrium and others show apparent diffusion profiles. If interpreted to reflect storage conditions, the magma would simultaneously be stored cold and warm, whereas the alternative proposal that plagioclase crystallizes continuously does not result in such an internally inconsistent interpretation. This recognition effectively precludes using diffusion modeling of modal minerals to infer storage durations.

Based on (U-Th)/He zircon dating, the SVC experienced at least five eruptions during the last 120 ka (21) (Figs. 1 and 2). Combining the requirement from the mush model (4) that each eruption is linked to at least one recharge with zircon age and trace element constraints for eruption windows lasting 15-40 ka, we conclude that parts of the SVC reservoir remained within the eruption window for at least 120 ka. Further support for this inference comes from the continuous zircon crystallization and plagioclase fractionation in the volcanic host of the cogenetic enclaves (Fig. 2), which contradicts cold-storage models (1) for which calculated temperatures are <700 °C and hence below the solidus (SI Text). Additional support for our preferred scenario of continuous magma presence in the thermally buffered zone from which volcanic zircons are derived comes from Ti-in-zircon data from the Tarawera arc-volcano (New Zealand) where volcanic zircons crystallized continuously over 150 ka and show little resolvable variation in their crystallization temperatures (14).



Fig. 4. (*A*) Minimum modeled spike duration constrained by the Belfond Dome eruption. Errors on modeled spike duration are 2σ . (*B*) Summary of mush rejuvenation events recorded by zircon U-Th model ages and Ti-in zircon temperatures within selected cogenetic enclaves and Belfond Dome lava. Errors are 1σ . Mush rejuvenation duration from our model. $T_{solidus}$ estimated by the lowest Ti-in zircon temperature from the zircon in the lava. $T_{liquidus andesite}$ from ref. 35. Minimum spike temperature estimated by the highest Ti-in zircon temperature from the cogenetic enclaves.

We conclude that eruptible magma reservoirs are generally long-lived (>100 ka), can continuously retain significant amounts of melt (<60% crystal fraction), and are stored broadly isothermally for timescales of tens to hundreds of thousands of years such that they can be rapidly mobilized and erupt. Volatiles liberated by hydrous magma recharge during decompression aid in rejuvenating the crystal mush by triggering the remelting process and facilitating eruption by decreasing melt viscosity (32).

The consistency of observations from two volcanic centers (Saint Lucia and Tarawera) and a shallow level plutonic reservoir (Elba) suggests our observation may be broadly applicable to other arc volcanoes of similar size and composition (e.g., Mount Saint Helens, Mount Pinatubo) (33, 34). Because the vast majority of modern continental volcanism is related to arcs, these findings have general implications for volcanic hazard assessments. Previous studies proposed that because magma chambers are ephemeral, detection of intracrustal melt might be a sign of imminent eruption (1). However, protracted crystallization ages and contrasting chemical variations in zircon from cogenetic volcanic and plutonic rocks call for the magma reservoir beneath the SVC being kept in warm storage (i.e., above the solidus, with portions residing continuously in the eruption window). Thermal energy considerations (3, 10, 12, 13) require a high rate of rejuvenation that leads to thermal excursions with durations >10 ka. Therefore, geophysical detection of melt beneath a volcano may hold little value as an indicator of volcanic hazards (1). By contrast, the extended use of the method applied in this study (U-Th ages combined with trace element on zircon from cogenetic volcanic and plutonic rocks) can

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provide superior insights into magma chamber evolution in potentially hazardous arc volcanoes based on absolute timescales.

Methods Summary

U-Th zircon analyses were performed on the University of California, Los Angeles (UCLA) CAMECA *ims*1270 and the Heidelberg University CAMECA *ims*1280-HR SIMS using the protocol described in ref. 36. Zircon trace elements abundances were acquired using the UCLA CAMECA *ims*1270, the Heidelberg University CAMECA *ims*1280-HR, and the Lausanne University CAMECA *ims*1280-HR (Swiss SIMS) ion probes following the analytical procedure described in ref. 37. U-Th and trace element analyses were made on the same spot on the zircon uppermost rim (crater average depth is ~4 µm). Description of the samples, SIMS protocol, Ti-in-zircon thermometry constraints, details on the spike duration modeling, and additional information on the Sr diffusion model of ref. 1 are given in *SI Text*.

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