



Cooling rates of mid-ocean ridge lava deduced from clinopyroxene spherulites



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ABSTRACT

We present a compositional analysis of spherulites and their surrounding groundmass in basaltic andesite lava samples collected from the Pacific–Antarctic Ridge. The spherulites consist of radiating dendritic branches of clinopyroxene, and contain abundant vesicles that are stretched parallel to the branches. Stretched vesicles require that the spherulites grew when the groundmass was liquid, and thus while it was hotter than its glass transition. Although chemical components incompatible in clinopyroxene should have been enriched in the melt by spherulite growth, no compositional variations occur at a scale larger than the 10 μm -wide slit aperture used for laser ablation ICP-MS. The absence of large-scale gradients implies that the spherulites grew faster than the elements could diffuse $\sim 10 \mu\text{m}$. Because diffusion is temperature controlled, cooling must have been rapid enough to prevent movement. If the spherulites nucleated when the melt was at between 1380 and 1223 K (i.e., undercooled by 0 to 157 K), then the constant concentrations of Li, Na, and K, the fastest diffusing elements analyzed, require temperature to have cooled faster than $10^{6-7} \text{ K min}^{-1}$. If, instead, the spherulites nucleated at undercoolings of 357 to 465 K (or as cold as 915 K, equal to the glass transition temperature), then the cooling rate could have been as slow as those inferred for glassy rinds on submarine lavas by relaxation geospeedometry. At all possible cooling rates, however, the spherulites must have grown as fast as $\sim 10^{3-5} \mu\text{m s}^{-1}$ to reach their final sizes of 1.4 to 3.2 mm. We speculate that the rapid growth resulted from low kinetic barriers to clinopyroxene growth in the basaltic andesite melt.

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1. Introduction

The morphology of lava flows erupted along the 55,000 km of the world's mid-ocean ridge varies with spreading rate (Bonatti and Harrison, 1988). Pillow lavas dominate at slow-spreading ridges; sheet flows dominate at fast spreading ridges. The systematic change in flow type implies that eruption dynamics exert a first-order control on morphology (Griffiths and Fink, 1992). Cooling rate may also impact flow morphology, mainly through the rapid formation of a glass layer that insulates the molten interior of the flows from the ambient cold seawater (e.g., Moore et al., 1973; Moore, 1975; Gregg and Chadwick, 1996; Fink and Griffiths, 1998; Gregg and Fornari, 1998). Understanding the flow morphology of submarine lavas thus requires deciphering both their eruption and cooling rates.

Cooling rates of submarine lavas have been inferred using relaxation geospeedometry, which extracts information about the thermal history

locked in the physical state of the glass using differential scanning calorimetry (e.g., Wilding et al., 2000). Because of the nature of the measurement, the inferred cooling rate is that at the glass transition (T_g). Glassy rinds from sheet flows have been found to cool at widely different rates, from 10^1 to $10^{4.8} \text{ K min}^{-1}$ (Bowles et al., 2005; Helo et al., 2013). Cooling rates for submarine pyroclasts and hyaloclastic fragments are equally diverse (Wilding et al., 2000; Potuzak et al., 2008; Nichols et al., 2009). The fastest cooling of any submarine glass measured so far is $\sim 10^{7.1} \text{ K min}^{-1}$, preserved in limo o Pele pyroclastics produced during mild submarine eruptions (Potuzak et al., 2008). The slowest rates measured for hyaloclastic fragments are far slower than those expected during most cooling processes operating during explosive eruptions, and are thus thought to reflect post-eruptive annealing (Wilding et al., 2000; Nichols et al., 2009).

In an effort to further examine the cooling rates of submarine lavas, we analyzed spherulites formed in mid-ocean ridge glass because spherulites grow during cooling (Lofgren, 1971; Fenn, 1977; Swanson et al., 1989). Our goal was to measure compositional gradients in the glassy groundmass surrounding the spherulites. Gradients are thought to form when incompatible elements are expelled from the interior of growing spherulites (Castro et al., 2008; Watkins et al., 2009; Gardner et al., 2012). The nature of the gradient depends strongly on the

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diffusivity of the element in question, and thus because diffusivity varies with temperature the gradient reflects the thermal history (Gardner et al., 2012).

Here, we report on compositional gradients associated with clinopyroxene spherulites present in glassy samples from the Pacific–Antarctic spreading ridge (Monecke et al., 2004). Vesicles trapped inside the spherulites are stretched in the general direction of crystal growth, whereas those outside of the spherulites are spherical and undeformed (Monecke et al., 2004). As vesicles can deform only when the melt flows, the textural evidence suggests that the spherulites grew while the groundmass was still molten, implying that growth temperatures were above T_g . Compositional gradients associated with the spherulites could thus potentially reflect cooling at temperatures above those recorded by relaxation geospeedometry.

2. Methods

This study focuses on spherulites contained in glassy mid-ocean ridge lavas collected near 37°47.5'S, at the northern end of an axial high of the central Pacific–Antarctic Ridge, immediately opposite the youngest seamount of the central Foundation seamount chain (Monecke et al., 2004). The samples were collected from glassy sheet flows that laterally transition into pillowed or lobate lava.

Thin sections were cut from two samples to expose spherulites for analysis. Back-scattered electron imaging was performed using a scanning electron microprobe to study the textures of the spherulites and their surrounding matrix. Selected areas were also mapped using X-rays for compositions of Na, Mg, Ca, and K using the JEOL Superprobe electron microprobe at the University of Texas at Austin. Operating conditions were a 40–100 nA beam current, 15 keV accelerating voltage, and a focused beam.

Major-element compositions of the clinopyroxene crystals in the spherulites and in the matrix as well as glass in the surrounding matrix were measured both qualitatively and quantitatively using the electron microprobe. Initially, energy-dispersive X-ray spectroscopy was conducted on crystals within and outside of the spherulites, and major peaks showed that all crystals are compositionally distinct clinopyroxene crystals. Quantitative analyses of major elements were collected using wavelength dispersive X-ray spectroscopy, with a 10 nA beam current, 15 keV accelerating voltage, and a beam focused to $<1 \mu\text{m}$. A focused beam was used because of the small sizes of the crystals and to attempt to avoid adjacent crystals during analysis of groundmass glasses. We report the average of analyses, all of which had analytical totals that exceed 98 wt.% (Table 1). Concentrations of Na_2O are relatively constant, suggesting that sodium migration during analysis was minimal, even though a focused beam was used. Only glass in the groundmass was analyzed.

Trace-element concentrations were measured by LA-ICP-MS at the University of Texas at Austin, using a New Wave Research UP 193-FX fast excimer (193 nm wavelength, 4–6 ns pulse width) laser system coupled to an Agilent 7500ce ICP-MS. The laser system uses a large format laser cell (232 cc with He carrier gas flow of 200 mL min^{-1}) equipped with a roving collector cup for direct sampling of the ablation plume. Reference standards and thin sections were sampled as line scans ($5 \mu\text{m s}^{-1}$), using a rectangular ($10 \times 100 \mu\text{m}$) slit aperture oriented with long-axis normal to the scan direction, an energy density (fluence) of $1060 \pm 29 \text{ J cm}^{-2}$, and 10 Hz repetition rate.

The ICP-MS, operated at an RF power of 1600 W and Ar carrier flow of 1.33 L min^{-1} , monitored masses ^7Li , ^{11}B , ^{23}Na , ^{25}Mg , ^{29}Si , ^{39}K , ^{45}Sc , ^{55}Mn , ^{85}Rb , ^{88}Sr , ^{133}Cs , ^{137}Ba , and ^{208}Pb . Analyte integration times varied between 5 and 100 ms, for a sampling period of 0.7472 s, equivalent to a reading every $3.736 \mu\text{m}$. Time-resolved intensities were converted to concentration (ppm) equivalents using Iolite software (Univ. Melbourne), with ^{29}Si as the internal standard, and a Si index value of 35.8 wt.%. Baselines were determined from 60 s glass

blank intervals measured while the laser was off and all masses were scanned by the quadrupole. USGS BCR-2G was used as the primary reference standard for all analytes. Median elemental concentration for line scans typically exceeded detection limits by factors of 10^2 to 10^4 for all elements except Pb ($34\times$) and Cs ($2\times$). Recoveries (relative 1σ deviations versus GeoREM preferred values) among analytes for secondary standards run as unknowns against primary standards were typically better than 4%. Based on these observations we conservatively assign 5% as relative uncertainties. Large area raster scans using the electron microprobe found no differences in composition between the spherulites and the groundmass. We thus assumed that all have the same silica contents in order to quantify elemental concentrations, equal to the bulk silica content.

Dissolved H_2O contents in the groundmass were analyzed by Fourier Transform Infrared (FTIR) spectroscopy with a Thermo Electron Nicolet 6700 spectrometer and Continuum IR microscope. To analyze water, samples must be thinned greatly, to about $20 \mu\text{m}$, to allow transmitted light to be focused. We therefore focused on analyzing a spherulite from a different sample than those analyzed by LA-ICP-MS. For that spherulite, the H_2O contents were measured along one analytical traverse. Outside of the spherulite, a total length of $300 \mu\text{m}$ was analyzed, consisting of spectra collected every $20 \mu\text{m}$. Inside the spherulite, a traverse of $200 \mu\text{m}$ was analyzed every $20 \mu\text{m}$. For the entire traverse, the aperture was set at $20 \mu\text{m} \times 40 \mu\text{m}$, with the long axis orientated parallel to the spherulite-groundmass contact.

Each spectrum consists of 60 scans at a resolution of 4 cm^{-1} , measured in transmittance mode in the mid-IR region (4000 to 650 cm^{-1}) using an infrared light source and a KBr beamsplitter. Contents of H_2O ($\text{H}_2\text{O}_{\text{tot}}$) were determined from absorbance at $\sim 3500 \text{ cm}^{-1}$, assuming a density of 2470 g L^{-1} . Molar absorptivity (ϵ) is poorly constrained for glasses of basaltic andesite to andesite composition, but is probably similar to values determined for Fe-bearing andesite ($62.32 \text{ L mol}^{-1} \text{ cm}^{-1}$; Mandeville et al., 2002) and Mt. Hood andesite ($70.32 \text{ L mol}^{-1} \text{ cm}^{-1}$; cited in Mandeville et al., 2002). Based on those measurements, a value for ϵ of $66 \text{ L mol}^{-1} \text{ cm}^{-1}$ was used. The sample thickness where each spectrum was collected was measured optically using a petrographic microscope by focusing on the top and bottom of the sample and measuring the depth with the focus drive linear encoder attached to the microscope. No carbon, either as molecular CO_2 or as dissolved carbonate, was detected.

Table 1

Compositions of the mid-ocean ridge lava, different types of clinopyroxene, and glass in the surrounding groundmass.

	Lava (average)	Clinopyroxene (in spherulites)	Clinopyroxene (in groundmass)	Glass (in groundmass)
SiO_2	52.14(10)	50.67(173)	51.49(166)	58.03(47)
TiO_2	2.24(1)	2.24(11)	2.13(18)	2.69(12)
Al_2O_3	13.12(1)	12.49(74)	12.72(62)	15.26(24)
FeO^*	13.20(5)	15.21(57)	14.65(107)	12.04(77)
MnO	0.24(1)	0.32(4)	0.29(4)	0.13(5)
MgO	3.81(5)	5.65(94)	5.43(106)	1.89(37)
CaO	8.25(2)	10.57(126)	11.03(142)	5.27(41)
Na_2O	3.68(7)	1.40(40)	1.40(19)	3.28(26)
K_2O	0.41(1)	0.24(5)	0.24(3)	0.47(4)
P_2O_5	0.50(1)	n.d.	n.d.	n.d.
H_2O	n.d.	n.d.	n.d.	0.90(2)
Total	97.57(10)	98.80(98)	99.38(111)	100.19(57)
<i>n</i>	3	6	5	4

All compositions are averages of *n* number of analyses. Lava composition by XRF from Monecke et al. (2004). Clinopyroxene and groundmass analyses are from this study. Major elements analyzed by electron microprobe, with all Fe reported as FeO; H_2O analyzed by FTIR spectroscopy; all oxides are in wt.% (n.d. = not determined). Values in parentheses are estimated standard deviations in terms of least units cited, such that 52.14(10) indicates a standard deviation of 0.10 wt.%.

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