

## Effect of sonic and ultrasonic frequencies on the crystallization of basalt

KAREN S. BARTELS<sup>1,\*</sup> AND TANYA FURMAN<sup>2</sup>

<sup>1</sup>Department of Earth Science, Northeastern Illinois University, Chicago, Illinois 60625, U.S.A.

<sup>2</sup>Department of Geosciences, Pennsylvania State University, University Park, Pennsylvania 16802, U.S.A.

### ABSTRACT

Experiments were conducted to determine whether vibrations with frequencies in the sonic to ultrasonic range have any effect on the crystallization of basalt. One-atmosphere melting experiments were done at the QFM buffer on a sample of alkali basalt. A piezoelectric quartz crystal and a function generator were used to generate waves of 1.5 MHz and 150 kHz, and waves were propagated along the platinum quench wire to which the experimental charge was attached. Experiments were conducted at 1182 °C; at this temperature the sample was about 25% crystalline and contained olivine and plagioclase. Compared to the static control experiments, vibrated experiments produced plagioclase crystals that were less euhedral and had lower aspect ratios. Crystal size distributions for plagioclase were steeper for the vibrated experiments, with a higher population density of smaller crystals. The effect on olivine crystal size distributions was less. If the effect on plagioclase extends to lower vibrational frequencies, it is possible that this phenomenon may occur in magmas prior to eruption. The experimental technique developed to produce vibrations is potentially useful for improving homogeneity of melt and increasing likelihood of achieving mineral-melt equilibrium in partitioning experiments.

### INTRODUCTION

Careful observation of the textures of igneous rocks is essential for deducing aspects of their solidification history. One way to quantify texture is to measure the distribution of crystal sizes in a rock. Crystal size distributions (CSDs) can provide information on rates of crystal nucleation and growth, residence times of phenocrysts in magma chambers, and processes of crystal fractionation and accumulation (Cashman 1986; Marsh 1988a, 1988b; Cashman and Marsh 1988; Maaloe et al. 1989; Mangan 1990; Marsh 1998). Size data can be obtained from direct measurement of crystals intersected in two-dimensional sections of rocks, but this data must be converted to three-dimensional parameters. Stereological solutions to this problem as applied to geologic samples have been discussed by Sahagian and Proussevitch (1998), Cashman and Marsh (1988), Peterson (1996), and Higgins (2000).

Laboratory experiments can help constrain models of how the thermal history of magmas can be reflected in crystal size distributions. Variables of temperature, pressure, and gas fugacities are controlled in these experiments, simulating conditions in magma chambers, but little attention has been paid to the environmental conditions of infrasonic, sonic, and ultrasonic emissions emanating from volcanoes. In addition to earthquakes caused by motion along faults and injection of magma, earth-

quake-induced fluid resonance and gas bubbling and gas explosions contribute to the wide range in frequency of emissions (Haggerty et al. 2000; Cramer and McNutt 1997; Diodati et al. 2000; Hurst and Vandemeulebrouck 1996; Vandemeulebrouck et al. 2000). This study compares melting experiments conducted under static conditions with those conducted while the sample was subject to vibrational frequencies in the sonic to ultrasonic range to determine whether the vibrational environment has any effect on growth of crystals in silicate melts of basaltic composition.

### EXPERIMENTAL METHODS

#### Starting material

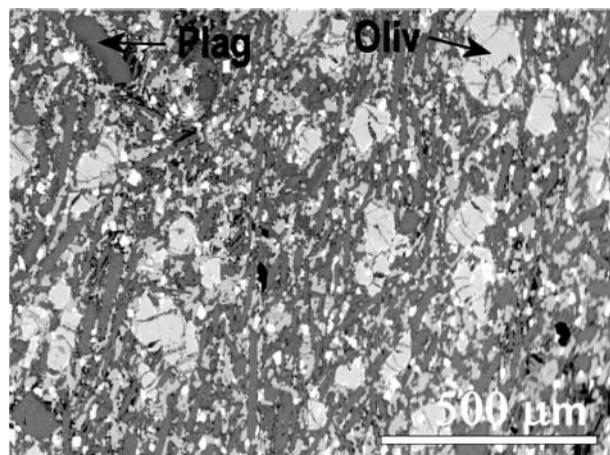
Experiments in this study were conducted on a sample of Virginia Highlands basalt (VAH1) from an Eocene lava flow at Mole Hill, Virginia [Table 1; see Wampler and Dooley (1975); Southworth et al. (1993)]. This alkali basalt is aphanitic and microporphyritic with locally 5 vol% microphe-  
nocrysts of olivine (Fo<sub>90</sub>) in an intergranular, mildly trachytic groundmass of plagioclase and augite microlites (Fig. 1). It also contains scattered xenocrysts of olivine, up to 3 mm in diameter, which are partially resorbed and have cores of Fo<sub>73–76</sub> (unpublished data). The sample was chosen because the phases of interest in this study, olivine and plagioclase, both occur close to the liquidus temperature. Olivine is the liquidus phase, appearing at 1190 °C, followed by plagioclase at 1182 °C, with augite appearing by 1160 °C (Table 2). Small rock chips of

\* E-mail: K-Bartels@neiu.edu

**TABLE 1.** Chemical composition (wt% oxides) of starting material, Virginia Highlands basalt

| SiO <sub>2</sub> | TiO <sub>2</sub> | Al <sub>2</sub> O <sub>3</sub> | Fe <sub>2</sub> O <sub>3</sub> | MnO  | MgO  | CaO   | Na <sub>2</sub> O | K <sub>2</sub> O | P <sub>2</sub> O <sub>5</sub> | Total |
|------------------|------------------|--------------------------------|--------------------------------|------|------|-------|-------------------|------------------|-------------------------------|-------|
| 45.66            | 1.59             | 15.43                          | 11.71                          | 0.21 | 9.92 | 11.98 | 2.36              | 0.70             | 0.38                          | 99.94 |

Notes: Sample analyzed by X-ray fluorescence. Total iron as Fe<sub>2</sub>O<sub>3</sub>.



**FIGURE 1.** BSE image of the rock used as starting material, alkali basalt VAH1 from Mole Hill, Virginia. Darkest-gray crystals are plagioclase, lightest-gray crystals are olivine, small medium-gray crystals in the groundmass are augite, and small white crystals are Fe-Ti oxides (magnetite and ilmenite). Scale bar is 500 micrometers.

**TABLE 2.** Phase appearance sequence

| Experiment | T (°C) | Duration (h) | Phases present                      |
|------------|--------|--------------|-------------------------------------|
| VAH1-1     | 1200   | 18           | glass                               |
| VAH1-6     | 1190   | 28           | glass, olivine                      |
| Ex1s       | 1182   | 38           | glass, olivine, plagioclase         |
| VAH1-5     | 1180   | 25           | glass, olivine, plagioclase         |
| VAH1-2     | 1170   | 63           | glass, olivine, plagioclase         |
| VAH1-4     | 1160   | 21           | glass, olivine, plagioclase, augite |

VAH1 were reduced to a homogenous powder in a SPEX shatterbox using a tungsten carbide grinding vessel, and this powder was used as the starting material for the experiments.

### Experimental procedures

Melting experiments were conducted at 1 atmosphere in a Deltech vertical tube gas-mixing furnace at the University of Virginia using the methods of Tormey et al. (1987). For each experiment, approximately 60 mg of sample powder was pressed into a pellet with polyvinyl alcohol and sintered to a 0.203 mm diameter Fe-Pt alloy loop custom made after the method of Grove (1981) to minimize Fe loss from the experiment. Samples were placed in the furnace at the final temperature of the experiment. Oxygen fugacity was controlled to the QFM buffer by mixing CO<sub>2</sub> and H<sub>2</sub> gases, and the fugacity was monitored using a ZrO<sub>2</sub>-CaO electrolyte cell calibrated at the Fe-FeO buffer. A type S thermocouple (Pt-Pt<sub>90</sub>Rh<sub>10</sub>) calibrated against the melting temperature of Au and Pd was used to monitor temperature (Biggar 1972).

After conducting preliminary melting experiments to determine the phase appearance sequence, experiments were undertaken to compare crystallization under static and sonic to

ultrasonic conditions (Table 3). The experimental procedure was modified slightly to permit vibration of the sample in the furnace. Instead of suspending the experimental charge from the ends of two 0.762 mm diameter platinum quench wires to allow for rapid quenching, the free, unplated end of the Fe-Pt alloy loop was wound tightly around the end of one of the quench wires. A piezoelectric quartz speaker element was rigidly attached with epoxy to the other end of this wire, which remained outside of the furnace. A function generator was used to control the frequency of the piezoelectric quartz crystal in the range of 150 kHz to 1.5 MHz. Vibrations were transmitted down the platinum wire to the sample inside the furnace; the precise frequency and amplitude at the sample could not be determined, but were lower than at the source in both cases due to attenuation down the wire. The experiments were quenched by removing the sample assembly from the top of the furnace and directing a stream of air on the sample. Static control experiments were attached to the same quench wire in the same manner as the vibrated experiments, but the function generator was not switched on. Control experiments were quenched in the same fashion as the vibrated ones.

### Analytical methods

The quenched beads of glass were mounted in epoxy and cut in half vertically with a diamond wafering blade. The surface was polished, and the compositions of glass, plagioclase, and olivine were determined using the JEOL 733 Superprobe in the Department of Earth and Planetary Sciences at Washington University in St. Louis. Backscattered electron (BSE) images were obtained at 100×, and the resulting photomosaic of the entire section of the bead was scanned to create jpeg images. Intersection widths and lengths of both plagioclase and olivine crystals were measured over most of the area of these digitized photographs using the SigmaScan image analysis program. The scale bar on the BSE images was used to calibrate distances in units of micrometers. For plagioclase crystals, intersection length was determined by measuring the greatest dimension of the cross section, and intersection width was measured orthogonal to this direction. Olivine crystals were traced to create individual polygons, and intersection length

**TABLE 3.** Experimental conditions

| Experiment | T (°C) | Duration (h) | Vibration condition | Phase proportions*<br>glass: plagioclase: olivine | R <sup>2</sup> † |
|------------|--------|--------------|---------------------|---------------------------------------------------|------------------|
| VAH1-11    | 1160   | 166          | static              | —‡                                                | —                |
| VAH1-12    | 1164   | 47           | 1.5 MHz             | —                                                 | —                |
| Ex1s       | 1182   | 38           | static              | 76 : 13 : 11                                      | 0.36             |
| Ex2v       | 1182   | 35           | 1.5 MHz             | 77 : 12 : 11                                      | 0.20             |
| Ex5v       | 1182   | 21           | 150 Khz             | 75 : 13 : 11                                      | 0.10             |

\* Determined by mass balance using electron microprobe analyses reported in Table 4.

† Sum of squared residuals from mass balance multiple regression.

‡ Not determined.

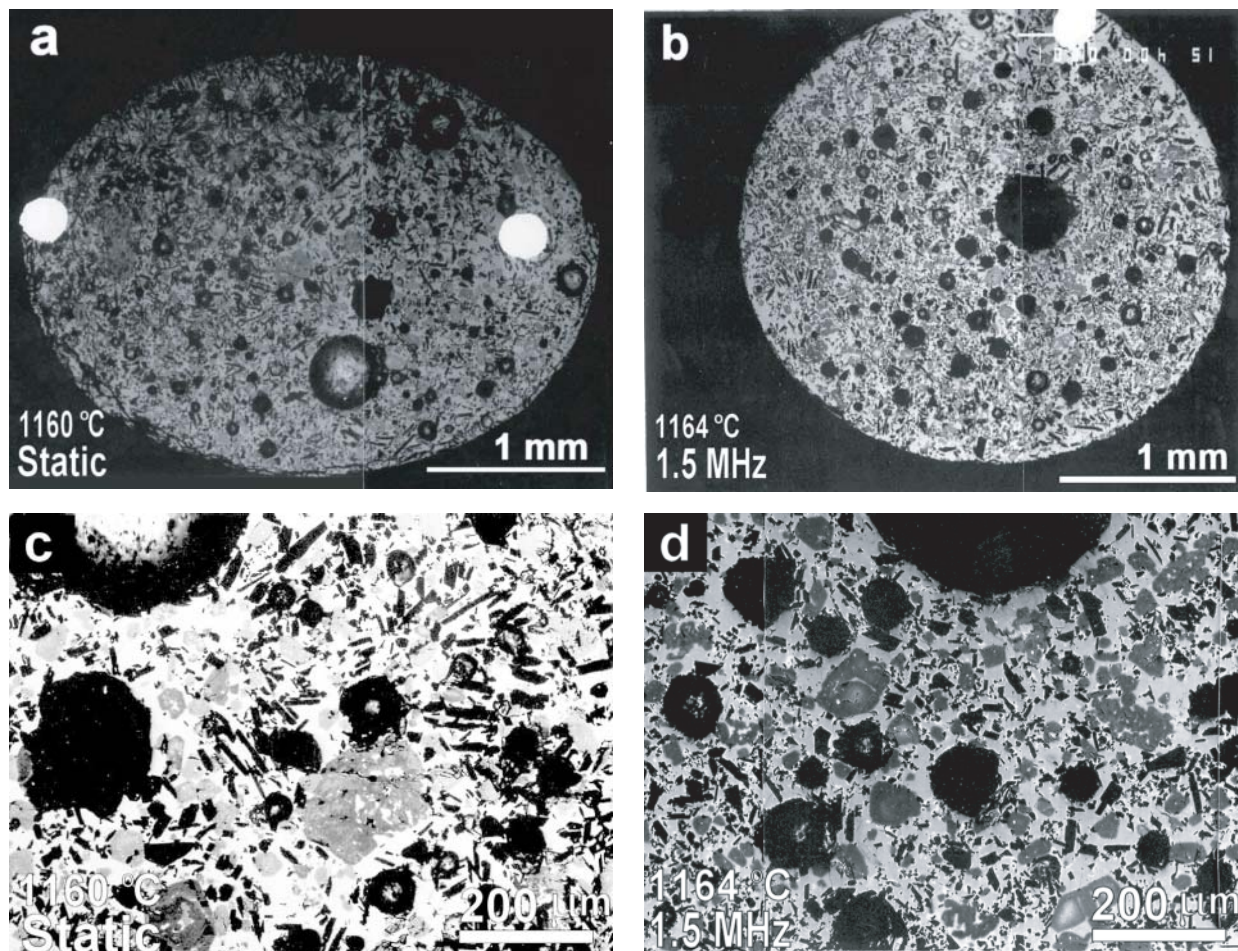
and width were calculated by SigmaScan software as the semi-major and semi-minor axes of these polygonal shapes. Crystal size distributions were calculated from both intersection length and intersection width data using the method of Higgins (2000; software CSDCorrections).

## RESULTS

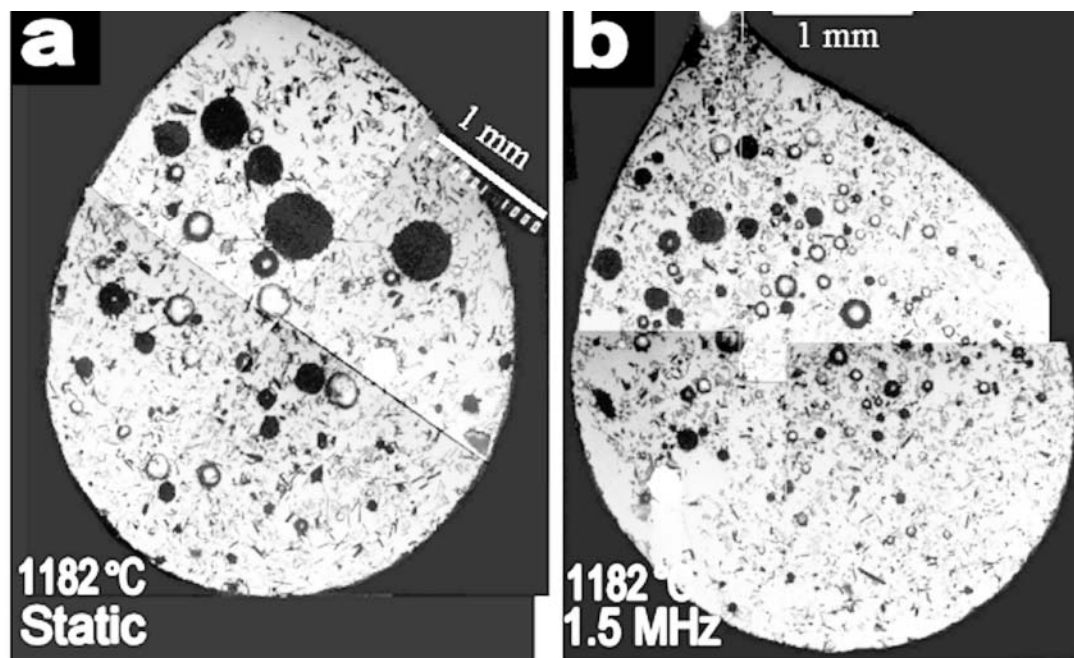
The first experiment designed to test the effect of vibration on crystallization was conducted at 1164 °C (VAH1-12). The sample was attached to the quench wire as described in the methods section above, and the function generator was set to produce a sine wave at 1.5 MHz. Visual inspection of a polished section of this experiment along with that of a similar experiment conducted under static conditions at 1160 °C (VAH1-11) indicated that a noticeable effect on crystallization did occur, especially for plagioclase. BSE images of these two experiments are shown in Figure 2. In the static experiment VAH1-11 (Figs. 2a and 2c), plagioclase crystals are larger, more euhedral, and have a higher aspect ratio compared to experiment VAH1-12 that was connected to the piezoelectric crystal

set to 1.5 MHz (Figs. 2b and 2d). Olivine crystals also appear larger in the static experiment, although the difference is not as pronounced as for plagioclase. Another noticeable difference apparent from Figures 2a and 2b is that plagioclase appears to have floated to the top of the charge in the static experiment, whereas the crystals in the vibrated experiment are distributed more evenly throughout the charge. However, the static experiment was conducted for 166 hours, much longer than the vibrated one at 47 hours, so the differences in crystal sizes and settling could be due to the difference in duration of the two experiments.

Rather than continue with experiments to match the run conditions of VAH1-12, it was decided to conduct the next set of experiments at a higher temperature (1182 °C), thus reducing the degree of crystallinity from about 50% to around 25%, and making it easier to distinguish crystal boundaries and measure apparent crystal sizes. Figure 3 shows BSE images of the entire experimental charge for the static control experiment (Ex1s, Fig. 3a) and the experiment connected to the piezoelectric crystal set to 1.5 MHz (Ex2v, Fig. 3b). Figure 4 shows



**FIGURE 2.** BSE images of first set of static (VAH1-11) and vibrated (VAH1-12) experiments. (a) Entire bead of static experiment VAH1-11. (b) Entire bead of experiment VAH1-12, vibrated at 1.5 MHz. (c) Enlargement of a portion of VAH1-11. (d) Enlargement of portion of VAH1-12. White circular shapes are platinum wires, black circular shapes are vesicles, black lath shapes are plagioclase, medium gray crystals are olivine, and darker gray crystals are augite. The light gray background is glass. Scale bar is 1 mm in (a) and (b) and 200 micrometers in (c) and (d).



**FIGURE 3.** BSE images of (a) static experiment Ex1s and (b) 1.5 MHz experiment Ex2v. White circular shapes are platinum wires, black circular shapes are vesicles, black lath shapes are plagioclase, medium gray crystals are olivine, and light gray background is glass. Scale bar is 1 mm.

enlargements of portions of both of these experiments as well as an additional experiment in which the piezoelectric crystal was set to a frequency one order of magnitude lower (150 kHz; Ex5v). As was the case in the first set of experiments, a qualitative visual inspection reveals that the plagioclase crystals in the static experiment are more euhedral and have a higher aspect ratio than those in the vibrated experiments, with no significant difference observable for olivine.

To test whether chemical equilibrium was reached during the experiments conducted at 1182 °C, the compositions of glass, plagioclase, and olivine were determined by electron microprobe analysis (Table 4). Some of the olivine crystals in each experiment exhibited zoning, with cores of Fo<sub>73–77</sub> and rims of ~Fo<sub>80</sub>. This finding indicates that some of the olivine xenocrysts from the rock powder had not completely equilibrated with the melt during the experiment. Zoning was not

observed in plagioclase crystals (~An<sub>78</sub>). For plagioclase crystals and rims of olivine crystals, the differences in major elements between experiments are within about one standard deviation of the replicate analyzes within an experiment. Glass compositions between experiments are more varied but are still fairly close in composition. Mass-balance calculations yielded similar phase proportions for all experiments, with 75–77% glass, 12–13% plagioclase and 11% olivine (see Table 3). The composition of olivine used in the mass-balance calculation was that of the olivine rims.

To quantify the observed difference in aspect ratio of plagioclase crystals, ratios of intersection width to intersection length (w/l) were calculated for all crystals of plagioclase that were over 10 micrometers in length (at smaller sizes, the width to length ratios become less accurate due to limited resolution). Figure 5 shows histograms of w/l for each of the three experi-

**TABLE 4.** Microprobe analyses of phases in experiments (see Table 3 for experimental details)

| Sample | Phase           | No. | SiO <sub>2</sub> | TiO <sub>2</sub> | Al <sub>2</sub> O <sub>3</sub> | FeO     | MnO     | MgO      | CaO      | Na <sub>2</sub> O | K <sub>2</sub> O | Total |                  |
|--------|-----------------|-----|------------------|------------------|--------------------------------|---------|---------|----------|----------|-------------------|------------------|-------|------------------|
| Ex1s   | gl*             | 3   | 46.2(2)†         | 2.13(2)          | 14.9(0)                        | 11.0(0) | 0.22(1) | 7.41(2)  | 13.5(1)  | 2.11(3)           | 0.82(3)          | 98.7  | <b>Mg</b> 55     |
|        | pl              | 6   | 48.7(5)          | n.d.‡            | 32.1(3)                        | 0.67(5) | n.d.    | n.d.     | 15.6(4)  | 2.32(14)          | 0.16(2)          | 99.6  | An <sub>79</sub> |
|        | ol <sub>r</sub> | 3   | 38.4(1)          | 0.09(5)          | —§                             | 18.4(2) | 0.33(3) | 41.1(1)  | 0.58(4)  | n.d.              | n.d.             | 98.9  | Fo <sub>80</sub> |
|        | ol <sub>c</sub> | 3   | 37.5(0)          | —                | 0.29(8)                        | 22.8(2) | 0.45(4) | 36.7(0)  | 0.81(19) | n.d.              | n.d.             | 98.6  | Fo <sub>74</sub> |
| Ex2v   | gl              | 3   | 46.8(1)          | 2.05(3)          | 14.9(0)                        | 10.9(1) | 0.22(3) | 7.31(2)  | 13.3(1)  | 2.30(2)           | 0.85(2)          | 98.6  | <b>Mg</b> 55     |
|        | pl              | 6   | 48.7(3)          | n.d.             | 31.9(2)                        | 0.75(8) | n.d.    | n.d.     | 15.2(3)  | 2.40(7)           | 0.17(1)          | 99.1  | An <sub>78</sub> |
|        | ol <sub>r</sub> | 4   | 38.5(4)          | 0.08(4)          | —                              | 18.2(2) | 0.32(2) | 41.5(3)  | 0.60(2)  | n.d.              | n.d.             | 99.0  | Fo <sub>80</sub> |
|        | ol <sub>c</sub> | 3   | 37.8(0)          | —                | —                              | 20.5(9) | 0.41(5) | 39.3(1)  | 0.53(5)  | n.d.              | n.d.             | 98.5  | Fo <sub>77</sub> |
| Ex5v   | gl              | 3   | 47.1(3)          | 2.08(4)          | 15.0(1)                        | 11.1(0) | 0.16(2) | 7.23(12) | 13.4(0)  | 2.40(2)           | 0.84(2)          | 99.3  | <b>Mg</b> 54     |
|        | pl              | 6   | 48.8(4)          | n.d.             | 31.7(4)                        | 0.81(4) | n.d.    | n.d.     | 15.3(4)  | 2.42(12)          | 0.17(1)          | 99.2  | An <sub>78</sub> |
|        | ol <sub>r</sub> | 3   | 38.6(2)          | 0.07(2)          | —                              | 19.0(6) | 0.31(5) | 40.7(4)  | 0.61(2)  | n.d.              | n.d.             | 99.2  | Fo <sub>79</sub> |
|        | ol <sub>c</sub> | 3   | 38.1(0)          | 0.06(3)          | —                              | 24.1(8) | 0.49(3) | 36.5(6)  | 0.50(3)  | n.d.              | n.d.             | 99.8  | Fo <sub>73</sub> |

\* Phase abbreviations: gl = glass, pl = plagioclase, ol<sub>r</sub> = olivine rim, ol<sub>c</sub> = olivine core.

† Units in parentheses represent one standard deviation in terms of least units cited, thus, 46.2(2) should be read as 46.2 ± 0.2.

‡ n.d. indicates element was not determined.

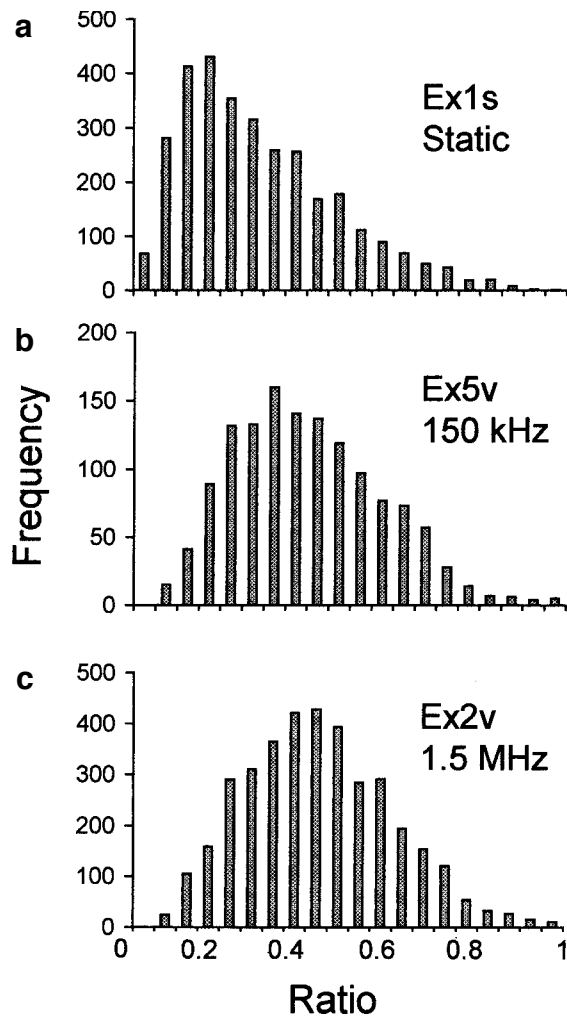
§ Dash indicates element was analyzed, but was below detection limits.



**FIGURE 4.** BSE images of (a) static experiment Ex1s, (b) 150 kHz experiment Ex5v, and (c) 1.5 MHz experiment Ex2v. Black and/or white circles are vesicles, black lath shapes are plagioclase, medium-gray crystals are olivine, and the light-gray background is glass. All experiments were conducted at 1182 °C. Scale bar is 200 micrometers and all photos are at the same scale.

ments conducted at 1182 °C (Ex1s, Ex5v, and Ex2v). The distribution of ratios is heavily skewed toward lower values for Ex1s, the static experiment, whereas the distributions for the vibrated experiments Ex5v and Ex2v are more normal.

For non-equant shapes, statistical data on widths and lengths measured in a two-dimensional cross section can be used to calculate the short:intermediate:long dimensions (S:I:L) of the three-dimensional crystal shape (Higgins 2000). This information on the three-dimensional aspect ratio is needed for calculating crystal size distributions. Stereological corrections are needed to compensate for the sectioning effect; lengths and widths of crystals intersected by the plane of the section must be converted into corrected crystal sizes in three dimensions.



**FIGURE 5.** Histograms of width to length ratios for crystals of plagioclase measured in (a) static experiment Ex1s, (b) 150 kHz experiment Ex5v, and (c) 1.5 MHz experiment Ex2v.

As shown by Higgins (1994), for parallelepiped crystals with no preferred orientation, the value of  $S/I$  is obtained from the mode of the ratio of intersection width to intersection length. The value of  $I/L$  is more difficult to determine, but it can be estimated as:

$$I/L = 0.5 + [(\text{mean } w/l - \text{mode } w/l) / \text{standard deviation } w/l].$$

Statistical data for the  $w/l$  histograms in Figure 5 are shown in Table 5. The S:I:L values calculated from plagioclase width:length data for Ex1s, Ex5v, and Ex2v were 1:5:5, 1:3:3 and 1:2:4 respectively. However, when these aspect ratios were used to calculate CSDs (with the software CSDCorrections,

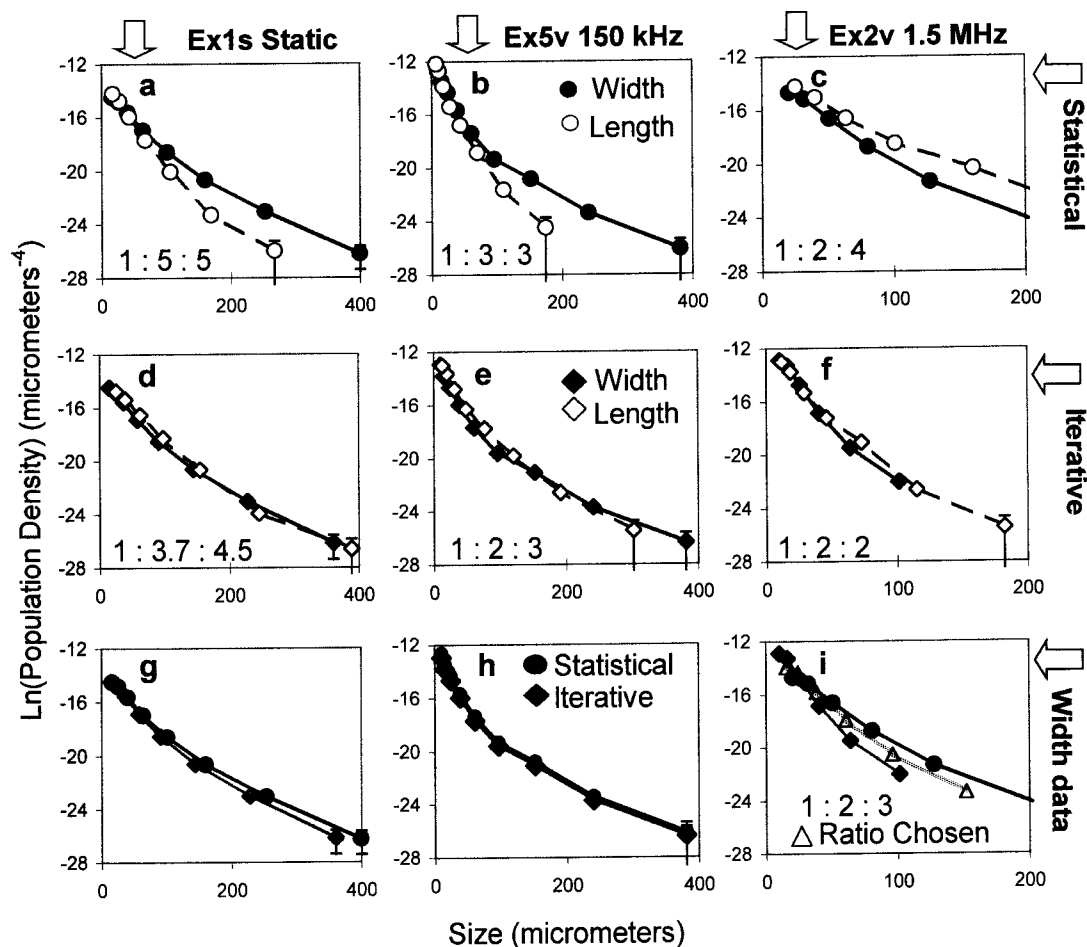


**TABLE 5.** Descriptive statistics for plagioclase width to length ratios

|                                         | Ex1s static | Ex5v 150 KHz | Ex2v 1.5 MHz |
|-----------------------------------------|-------------|--------------|--------------|
| mean                                    | 0.317       | 0.426        | 0.454        |
| mode                                    | 0.20        | 0.35         | 0.45         |
| standard deviation                      | 0.176       | 0.168        | 0.169        |
| count                                   | 3112        | 1330         | 3669         |
| S/I = mode                              | 0.20        | 0.35         | 0.45         |
| I/L = 0.5 +<br>(mean-mode)/stdev        | 1.16        | 0.952        | 0.504        |
| I/S                                     | 5.00        | 2.86         | 2.22         |
| I/S x L/I                               | 4.29        | 3.00         | 4.41         |
| aspect ratio from<br>statistical method | 1:5:5       | 1:3:3        | 1:2:4        |
| aspect ratio from<br>iterative method   | 1:3.7:4.5   | 1:2:3        | 1:2:2*       |

\* Aspect ratio chosen for CSD calculations was 1:2:3. See Figure 6 and text for explanation.

Higgins 2000), the curves calculated from length data did not match up well with those calculated from width data (Figs. 6a–c). As an alternative approach, S:I:L values were chosen by trial and error and iteration until the curves calculated from length data matched up fairly well with those calculated from width data (Figs. 6d–f). This procedure yielded ratios of 1:3.7:4.5 for Ex1s, 1:2:3 for Ex5v and 1:2:2 for Ex2v. Although use of these values resulted in internal consistency, they are not necessarily physically meaningful, as it is likely that there is a range of shapes of varying aspect ratio within individual experiments. CSDs calculated from width curves using the two sets of aspect ratios are compared at the bottom of Figure 6. For Ex1s (Fig. 6g) and Ex5v (Fig. 6h), width curves from the two approaches are in close agreement. However, this is not the case for Ex2v (Fig. 6i), so an aspect ratio of 1:2:3 was chosen for this sample because it is an intermediate value and because it results in using the same aspect ratio as that calculated for Ex5v, the other vibrated experiment. In calculating the pla-



**FIGURE 6.** Crystal-size distributions of plagioclase calculated using aspect ratios obtained by two different methods. The first row of graphs show curves calculated from intersection width and intersection length data for (a) static experiment Ex1s, (b) 150 kHz experiment Ex5v, and (c) 1.5 MHz experiment Ex2v, using an aspect ratio obtained by the statistical method described in the text. The second row of graphs show curves calculated from intersection width and intersection length data for (d) static experiment Ex1s, (e) 150 kHz experiment Ex5v, and (f) 1.5 MHz experiment Ex2v, using an aspect ratio obtained by the iterative method described in the text. The last row of graphs compares the curves obtained from intersection width data using the two methods for (g) static experiment Ex1s, (h) 150 kHz experiment Ex5v, and (i) 1.5 MHz experiment Ex2v. Error bars are not shown for points where the error is smaller than the symbol size.

gioclase CSD curves, the crystals were modeled as block shapes with no rounding for the static experiment and a low degree of rounding (roundness = 0.1 in CSDCorrections) for the vibrated experiments.

Because olivine crystals had a more equant shape, the statistical method of calculating aspect ratio for olivine was less valid, so S:L:L was determined by the trial and error method. For all three experiments, modeling olivine as a block shape with moderate degree of rounding (roundness = 0.5 in CSDCorrections) and a value of 1:1.5:2.3 for the aspect ratio gave good agreement between CSDs calculated from width data and from length data. For consistency, all CSD curves in this paper were calculated using intersection width data and aspect ratios determined by the iterative method (with the exception of Ex2v plagioclase as noted above). Intersection data for plagioclase and the CSD data calculated from them are given in Table 6, and Table 7 presents these data for olivine. The term "size" listed in Tables 6 and 7 (and used on the horizontal axis of the CSD graphs) refers to corrected crystal length calculated by CSDCorrections (Higgins 2000).

Figure 7 shows the CSD curves for both olivine and plagioclase in each experiment. Note that the vibrated experiments have a higher population density of smaller plagioclase crystals compared to the static experiment. The plagioclase distribution curve also appears to become steeper, at least for the

smaller crystal sizes, with increasing frequency of vibration. The CSDs for plagioclase in all three experiments are compared over the entire size range in Figure 8a. Note that although Ex5v, the experiment conducted at 150 kHz, has a steeper CSD at small crystal sizes, at large crystal sizes the CSD curve overlaps that of the static experiment, within errors of the calculation generated by the CSDCorrections program. For all except the largest crystal sizes, error bars are generally much smaller than the symbol size on the diagrams; therefore, to see the variation at small sizes better, Figure 8b is an enlargement of the boxed area in Figure 8a. The natural logarithm of the population density for crystals 15 micrometers in length is -14.5 for the static experiment and approximately -13.8 for both vibrated experiments. Errors in population density at these small sizes are around  $\pm 0.05$ , so the increase in the population density of small crystal sizes in the vibrated experiments is significant.

Consistent with visual observations that the effect of vibration is less pronounced for olivine than it is for plagioclase, there is less difference between CSD curves for olivine than there is between CSD curves for plagioclase. At first glance (Fig. 9a), the olivine distribution curve for the static experiment appears to become steeper at larger crystal sizes, whereas the curves for the vibrated experiments appear to be more linear, with higher population densities for both the smallest (Fig. 9b) and largest sizes of olivine crystals. However, these differ-

**TABLE 6.** Intersection data and CSD data for plagioclase calculated from CSDCorrections (Higgins 2000)

| Sample | Aspect ratio | Area ( $\mu^2$ )   | From intersection length data |      |                          |         |                 | From intersection width data |      |                          |            |                 |
|--------|--------------|--------------------|-------------------------------|------|--------------------------|---------|-----------------|------------------------------|------|--------------------------|------------|-----------------|
|        |              |                    | Measured length ( $\mu$ )     | No.  | Corrected size ( $\mu$ ) | Ln(PD)* | PD error†       | Measured width ( $\mu$ )     | No.  | Corrected size ( $\mu$ ) | Ln(PD)     | PD error        |
| Ex1s   | 1:3.7:4.5    | $7.89 \times 10^6$ | 398                           | 1    | 388                      | -26.6   | +0.3, $-\infty$ | 100                          | 2    | 361                      | -26.1      | +1.23, -0.53    |
|        |              |                    | 251                           | 6    | 245                      | -23.9   | $\pm 0.45$      | 63.1                         | 19   | 228                      | -23.0      | $\pm 0.23$      |
|        |              |                    | 159                           | 62   | 154                      | -20.6   | $\pm 0.13$      | 39.8                         | 84   | 144                      | -20.6      | $\pm 0.11$      |
|        |              |                    | 100                           | 272  | 97.5                     | -18.2   | $\pm 0.07$      | 25.1                         | 269  | 90.5                     | -18.5      | $\pm 0.06$      |
|        |              |                    | 63.1                          | 653  | 61.5                     | -16.5   | $\pm 0.05$      | 15.9                         | 570  | 57.2                     | -16.8      | $\pm 0.05$      |
|        |              |                    | 39.8                          | 929  | 38.8                     | -15.3   | $\pm 0.04$      | 10.0                         | 878  | 36.1                     | -15.5      | $\pm 0.04$      |
|        |              |                    | 25.1                          | 797  | 25.4                     | -14.7   | $\pm 0.05$      | 6.31                         | 812  | 22.8                     | -14.7      | $\pm 0.04$      |
|        |              |                    | 15.9                          | 415  | 15.4                     | -15.1   | $\pm 0.1$       | 3.98                         | 502  | 14.4                     | -14.4      | $\pm 0.06$      |
|        |              |                    | 10.1                          | 120  |                          |         | 2.51            | 124                          | 9.05 | -16.3                    | $\pm 0.5$  |                 |
|        |              |                    | 6.31                          | 22   |                          |         | 1.59            | 21                           |      |                          |            |                 |
|        |              |                    | 3.98                          | 4    |                          |         |                 |                              |      |                          |            |                 |
|        |              |                    |                               |      |                          |         |                 |                              |      |                          |            |                 |
|        |              |                    |                               |      |                          |         |                 |                              |      |                          |            |                 |
| Ex2v   | 1:2:3        | $7.70 \times 10^6$ | 251                           | 1    | 302                      | -26.2   | +0.7, $-\infty$ | 63.1                         | 9    | 152                      | -23.3      | +0.32/-0.47     |
|        |              |                    | 159                           | 7    | 191                      | -23.4   | $\pm 0.4$       | 39.8                         | 48   | 95.7                     | -20.5      | $\pm 0.15$      |
|        |              |                    | 100                           | 95   | 120                      | -19.8   | $\pm 0.1$       | 25.1                         | 260  | 60.4                     | -17.9      | $\pm 0.07$      |
|        |              |                    | 63.1                          | 256  | 75.8                     | -17.9   | $\pm 0.07$      | 15.9                         | 868  | 38.1                     | -15.8      | $\pm 0.04$      |
|        |              |                    | 39.8                          | 687  | 47.8                     | -16.1   | $\pm 0.04$      | 10.0                         | 1537 | 24.0                     | -14.3      | $\pm 0.03$      |
|        |              |                    | 25.1                          | 1324 | 30.2                     | -14.5   | $\pm 0.03$      | 6.31                         | 1077 | 15.2                     | -13.9      | $\pm 0.04$      |
|        |              |                    | 15.9                          | 1301 | 19.1                     | -13.7   | $\pm 0.04$      | 3.98                         | 363  | 9.57                     | -14.8      | $\pm 0.15$      |
|        |              |                    | 10.0                          | 517  | 12.0                     | -14.4   | $\pm 0.11$      | 2.51                         | 30   |                          |            |                 |
|        |              |                    | 6.31                          | 19   |                          |         | 1.59            | 15                           |      |                          |            |                 |
|        |              |                    | 3.98                          | 1    |                          |         | 1.0             | 0                            |      |                          |            |                 |
|        |              |                    |                               |      |                          | 0.63    | 1               |                              |      |                          |            |                 |
|        |              |                    |                               |      |                          |         |                 |                              |      |                          |            |                 |
|        |              |                    |                               |      |                          |         |                 |                              |      |                          |            |                 |
| Ex5v   | 1:2:3        | $3.65 \times 10^6$ | 251                           | 1    | 302                      | -25.4   | +0.8, $-\infty$ | 159                          | 1    | 381                      | -26.4      | +0.7, $-\infty$ |
|        |              |                    | 159                           | 7    | 191                      | -22.6   | $\pm 0.41$      | 100                          | 6    | 240                      | -23.7      | +0.35, -0.57    |
|        |              |                    | 100                           | 46   | 120                      | -19.8   | $\pm 0.15$      | 63.1                         | 32   | 152                      | -21.1      | $\pm 0.18$      |
|        |              |                    | 63.1                          | 148  | 75.8                     | -17.7   | $\pm 0.09$      | 39.8                         | 58   | 95.7                     | -19.6      | $\pm 0.14$      |
|        |              |                    | 39.8                          | 261  | 47.8                     | -16.3   | $\pm 0.07$      | 25.1                         | 160  | 60.4                     | -17.7      | $\pm 0.08$      |
|        |              |                    | 25.1                          | 472  | 30.2                     | -14.8   | $\pm 0.05$      | 15.9                         | 355  | 38.1                     | -15.9      | $\pm 0.07$      |
|        |              |                    | 15.9                          | 643  | 19.1                     | -13.6   | $\pm 0.05$      | 10                           | 545  | 24.0                     | -14.6      | $\pm 0.05$      |
|        |              |                    | 10.0                          | 535  | 12.0                     | -13.0   | $\pm 0.06$      | 6.31                         | 555  | 15.2                     | -13.7      | $\pm 0.05$      |
|        |              |                    | 6.31                          | 252  | 7.58                     | -13.3   | $\pm 0.13$      | 3.98                         | 527  | 9.57                     | -12.9      | $\pm 0.05$      |
|        |              |                    | 3.98                          | 53   |                          |         | 2.51            | 162                          | 6.03 | -14.1                    | $\pm 0.25$ |                 |
|        |              |                    | 2.51                          | 1    |                          |         | 1.58            | 16                           |      |                          |            |                 |
|        |              |                    |                               |      |                          |         | 1.00            | 2                            |      |                          |            |                 |
|        |              |                    |                               |      |                          |         |                 |                              |      |                          |            |                 |

\* PD = Population density.

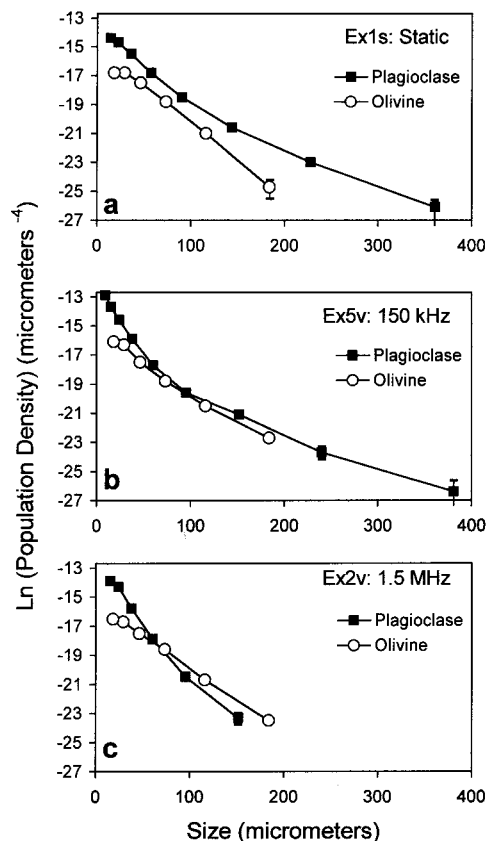
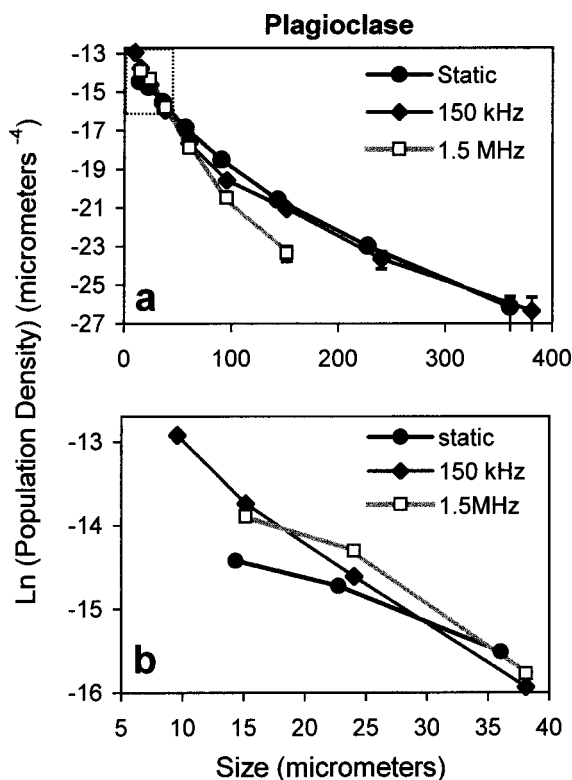
† Error in population density as calculated by CSDCorrections.

**TABLE 7.** Intersection data and CSD data for olivine calculated from CSDCorrections (Higgins 2000)

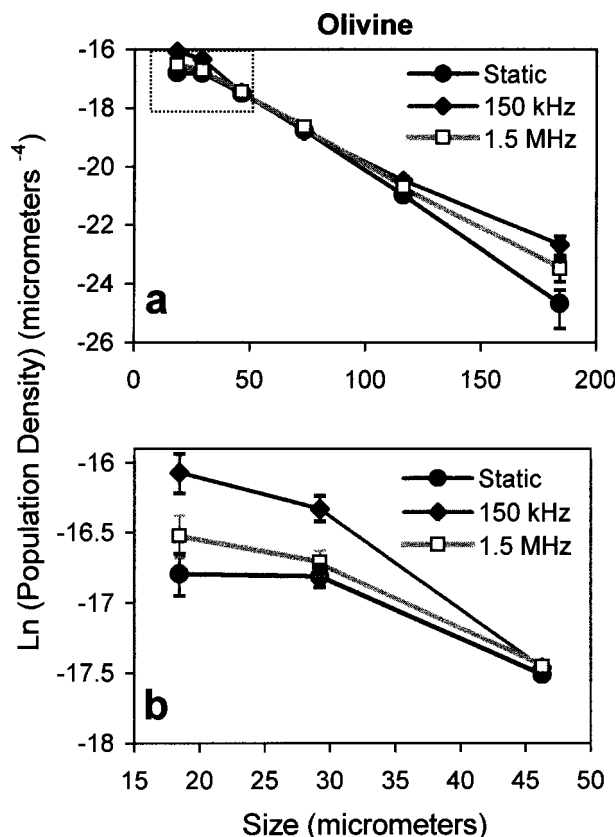
| Sample | Aspect ratio | Area ( $\mu^2$ )   | From intersection length data |     |                          |                        |                          | From intersection width data |     |                          |                       |                         |
|--------|--------------|--------------------|-------------------------------|-----|--------------------------|------------------------|--------------------------|------------------------------|-----|--------------------------|-----------------------|-------------------------|
|        |              |                    | Measured length ( $\mu$ )     | No. | Corrected size ( $\mu$ ) | Ln(PD)* ( $\mu^{-4}$ ) | PD error† ( $\mu^{-4}$ ) | Measured width ( $\mu$ )     | No. | Corrected size ( $\mu$ ) | Ln(PD) ( $\mu^{-4}$ ) | PD error ( $\mu^{-4}$ ) |
| Ex1s   | 1:1.5:2.3    | $8.17 \times 10^6$ | 159                           | 1   | 195                      | -25.5                  | +0.7, - $\infty$         | 100                          | 3   | 184                      | -24.7                 | +0.45, -0.9             |
|        |              |                    | 100                           | 36  | 123                      | -21.0                  | $\pm 0.17$               | 63.1                         | 49  | 116                      | -21.0                 | $\pm 0.15$              |
|        |              |                    | 63.1                          | 153 | 77.5                     | -18.7                  | $\pm 0.08$               | 39.8                         | 180 | 73.3                     | -18.8                 | $\pm 0.08$              |
|        |              |                    | 39.8                          | 262 | 48.9                     | -17.3                  | $\pm 0.07$               | 25.1                         | 272 | 46.3                     | -17.5                 | $\pm 0.07$              |
|        |              |                    | 25.1                          | 266 | 30.9                     | -16.4                  | $\pm 0.07$               | 15.9                         | 246 | 29.2                     | -16.8                 | $\pm 0.08$              |
|        |              |                    | 15.9                          | 164 | 19.5                     | -16.2                  | $\pm 0.12$               | 10.0                         | 137 | 18.4                     | -16.8                 | $\pm 0.15$              |
|        |              |                    | 10.0                          | 33  |                          |                        |                          | 6.31                         | 24  |                          |                       |                         |
|        |              |                    | 6.31                          | 2   |                          |                        |                          | 3.98                         | 6   |                          |                       |                         |
| Ex2v   | 1:1.5:2.3    | $5.76 \times 10^6$ | 159                           | 4   | 195                      | -23.8                  | +0.4, -0.7               | 100                          | 7   | 184                      | -23.5                 | +0.32, -0.48            |
|        |              |                    | 100                           | 38  | 123                      | -20.6                  | $\pm 0.17$               | 3.1                          | 46  | 116                      | -20.7                 | $\pm 0.15$              |
|        |              |                    | 63.1                          | 113 | 77.5                     | -18.6                  | $\pm 0.1$                | 39.8                         | 146 | 73.3                     | -18.6                 | $\pm 0.09$              |
|        |              |                    | 39.8                          | 185 | 48.9                     | -17.3                  | $\pm 0.08$               | 25.1                         | 205 | 46.3                     | -17.5                 | $\pm 0.08$              |
|        |              |                    | 25.1                          | 238 | 30.9                     | -16.1                  | $\pm 0.08$               | 15.9                         | 190 | 29.2                     | -16.7                 | $\pm 0.09$              |
|        |              |                    | 15.9                          | 138 | 19.5                     | -16.0                  | $\pm 0.12$               | 10.0                         | 118 | 18.4                     | -16.5                 | $\pm 0.15$              |
|        |              |                    | 10                            | 43  | 12.3                     | -17.2                  | +0.45, -0.83             | 6.31                         | 44  | 11.6                     | -17.4                 | +0.44, -0.82            |
|        |              |                    | 6.31                          | 10  | 7.75                     | -16.7                  | +0.35, -0.66             | 3.98                         | 13  | 7.34                     | -16.7                 | +0.31, -0.47            |
| Ex5v   | 1:1.5:2.3    | $3.65 \times 10^6$ | 159                           | 7   | 195                      | -22.8                  | +0.32, -0.47             | 100                          | 10  | 184                      | -22.7                 | +0.28, -0.38            |
|        |              |                    | 100                           | 29  | 123                      | -20.4                  | $\pm 0.20$               | 63.1                         | 37  | 116                      | -20.5                 | $\pm 0.16$              |
|        |              |                    | 63.1                          | 80  | 77.5                     | -18.5                  | $\pm 0.12$               | 39.8                         | 81  | 73.3                     | -18.8                 | $\pm 0.12$              |
|        |              |                    | 39.8                          | 103 | 48.9                     | -17.4                  | $\pm 0.11$               | 25.1                         | 128 | 46.3                     | -17.5                 | $\pm 0.10$              |
|        |              |                    | 25.1                          | 147 | 30.9                     | -16.1                  | $\pm 0.09$               | 15.9                         | 165 | 29.2                     | -16.3                 | $\pm 0.09$              |
|        |              |                    | 15.9                          | 122 | 19.5                     | -15.5                  | $\pm 0.11$               | 10.0                         | 106 | 18.4                     | -16.1                 | $\pm 0.14$              |
|        |              |                    | 10.0                          | 76  | 12.3                     | -15.2                  | $\pm 0.18$               | 6.31                         | 44  | 11.6                     | -16.8                 | +0.37, -0.62            |
|        |              |                    | 6.31                          | 11  |                          |                        |                          | 3.98                         | 4   |                          |                       |                         |

\* PD = Population density.

† Error in population density as calculated by CSDCorrections.

**FIGURE 7.** Crystal size distributions of plagioclase and olivine calculated from intersection width data for (a) static experiment Ex1s, (b) 150 kHz experiment Ex5v, and (c) 1.5 MHz experiment Ex2v. Error bars are not shown for points where the error is smaller than the symbol size.**FIGURE 8.** Crystal size distributions for plagioclase calculated from intersection width data (a) over the entire size range calculated and (b) at the smallest crystal sizes. CSDs are shown for all three experiments (Static, Ex1s; 150 kHz, Ex5v; and 1.5 MHz, Ex2v). Error bars are not shown for points where the error is smaller than the symbol size.





**FIGURE 9.** Crystal size distributions for olivine calculated from intersection width data (a) over the entire size range calculated and (b) at the smallest crystal sizes. CSDs are shown for all three experiments (Static, Ex1s; 150 kHz, Ex5v; and 1.5 MHz, Ex2v). Error bars are not shown for points where the error is smaller than the symbol size.

ences are basically within the error in the population density calculation, so this study concludes that there is no discernable difference in olivine CSDs as a result of vibration.

## DISCUSSION

The increase in population density of small crystal sizes for plagioclase in the vibrated experiments could be due to an increase in the number of nucleation sites. Ultrasonic treatment of melts of light alloys has been shown to increase the number of nucleation sites when casting ingots, producing a more uniform and finer-grained texture (Eskin 1994). In contrast to this, ultrasonic treatment of aqueous solutions of salts has been shown to result in a larger average particle size upon crystallization, perhaps by increasing the critical nucleus size (Enomoto et al. 1992).

Another possible explanation for the observed effect on plagioclase is that vibrations in the melt disrupt the process of crystal growth. The fact that the CSDs for olivine are not affected as much by the vibrations may be related to the fact that olivine crystals tend to grow with approximately equant shapes whereas plagioclase crystals tend to grow as needles, thus mak-

ing them more susceptible to longitudinal vibrations. In addition, because olivine is a orthosilicate and plagioclase a tectosilicate, it may be easier to disrupt plagioclase growth because of the higher degree of organization of silica tetrahedra that is required. Experiments should be conducted on pyroxene growth to see whether the effect is intermediate to that of olivine and plagioclase.

The frequencies used in this study were in the ultrasonic range. The piezoelectric crystal was attached to a function generator that was set to produce either a 1.5 MHz or 150 kHz sine wave. However, it is not known what the actual vibrational frequencies were inside the experimental charge. Even when the apparatus was set to 1.5 MHz, a high-pitched sound could be heard coming from the furnace. The upper end of the sonic range is 20 kHz, so significant attenuation of frequency must have occurred somewhere within the experimental apparatus. Seismic activity lies within the infrasonic range, at much lower frequencies than those used in this study. However, other types of volcanic activity can produce vibrations of higher frequencies. Exsolution of gases and gas explosions produce frequencies in the sonic range, and even ultrasonic volcanic emissions from volcanoes have been observed (Diodati et al. 2000). Further experiments should be done to test the lower frequency limit of this effect.

This technique also has implications for methods in experimental petrology. The increase in population density of smaller crystal sizes could be an advantage in partitioning experiments, increasing the likelihood of achieving equilibrium by increasing surface area to volume ratios of the crystals in the melt. If vibrations disrupt the flotation or settling of crystals, as results of the first set of experiments suggest (Fig. 2), perhaps this technique could be used to increase the homogeneity of the melt throughout the charge.

## ACKNOWLEDGMENTS

Facilities support was provided by the College of Arts and Sciences at the University of Virginia. Additional support came from NSF EAR-9508112 to T. Furman. Thanks to Alison Cohen for helping with the initial stages of the project, and to Daniel Kremser, Department of Earth and Planetary Sciences at Washington University, for valuable assistance with electron microprobe analysis. We thank Michael Higgins, Bruce Marsh, and Michael Toplis for their critical and constructive comments in review. The Committee on Organized Research at Northeastern Illinois University provided partial support for completion of this work.

## REFERENCES CITED

- Biggar, G.M. (1972) Diopside, lithium metasilicate, and the 1968 temperature scale. *Mineralogical Magazine*, 38, 768–770.
- Cashman, K.V. (1986) Crystal size distributions in igneous and metamorphic rocks. Ph.D. Thesis, Johns Hopkins University.
- Cashman, K.V. and Marsh, B.D. (1988) Crystal size distribution (CSD) in rocks and the kinetics and dynamics of crystallization. II. Makaopuhi lava lake. *Contributions to Mineralogy and Petrology*, 99, 292–305.
- Cramer, C.H. and McNutt, S.R. (1997) Spectral analysis of earthquakes in the 1989 Mammoth Mountain swarm near Long Valley, California. *Bulletin of the Seismological Society of America*, 87, 6, 1454–1462.
- Diodati, P., Bak, P., and Marchesoni, F. (2000) Acoustic emission at the Stromboli Volcano: Scaling laws and seismic activity. *Earth and Planetary Science Letters*, 182, 3–4, 253–258.
- Enomoto, N., Sung, T., and Nakagawa, Z. (1992) Effect of ultrasonic waves on crystallization from a supersaturated solution of alum. *Journal of Materials Science*, 27, 5239–5243.
- Eskin, G.I. (1994) Influence of cavitation treatment of melts on the process of nucleation and growth of crystals during solidification of ingots and castings from light alloys. *Ultrasonics Sonochemistry*, 1, S59–S63.
- Grove, T.L. (1981) Use of Fe/Pt alloys to eliminate the iron loss problem in 1-atm

- gas-mixing experiments: theoretical and practical considerations. *Contributions to Mineralogy and Petrology*, 78, 298–304.
- Haggerty, M.T., Schwartz, S.Y., Garces, M.A., and Protti, M. (2000) Analysis of seismic and acoustic observations at Arenal Volcano, Costa Rica, 1995–1997. *Journal of Volcanology and Geothermal Research*, 101, 1–2, 27–65.
- Higgins, M.D. (1994) Determination of crystal morphology and size from bulk measurements on thin sections: numerical modelling. *American Mineralogist*, 79, 113–119.
- Higgins, M.D. (2000) Measurement of crystal size distributions. *American Mineralogist*, 85, 1105–1116.
- Hurst, A.W. and Vandemeulebrouck, J. (1996) Acoustic noise and temperature monitoring of the Crater Lake of Mount Ruapehu Volcano. *Journal of Volcanology and Geothermal Research*, 71, 1, 45–51.
- Maaloe, S., Tumyr, O., and James, D. (1989) Population density and zoning of olivine phenocrysts in tholeiites from Kauai, Hawaii. *Contributions to Mineralogy and Petrology*, 101, 176–186.
- Mangan, M.T. (1990) Crystal size distribution systematics and the determination of magma storage times: the 1959 eruption of Kilauea Volcano, Hawaii. *Journal of Volcanology and Geothermal Research*, 44, 295–302.
- Marsh, B.D. (1988a) Crystal size distribution (CSD) in rocks and the kinetics and dynamics of crystallization. I. Theory. *Contributions to Mineralogy and Petrology*, 99, 277–291.
- Marsh, B.D. (1988b) Crystal size distribution (CSD) in rocks and the kinetics and dynamics of crystallization. II. Makaopulu lava lake. *Contributions to Mineralogy and Petrology*, 99, 292–305.
- Marsh, B.D. (1998) On the interpretation of crystal size distributions in magmatic systems. *Journal of Petrology*, 39, 553–599.
- Peterson, T.D. (1996) A refined technique for measuring crystal size distributions in thin section. *Contributions to Mineralogy and Petrology*, 124, 395–405.
- Sahagian, D.L. and Proussevitch, A.A. (1998) 3D particle size distributions from 2D observations; stereology for natural applications. *Journal of Volcanology and Geothermal Research*, 84, 3–4, 173–196.
- Southworth, C.S., Gray, K.J., and Sutter, J.F. (1993) Middle Eocene intrusive igneous rocks of the central Appalachian Valley and Ridge Province—setting, chemistry, and implications for crustal structure. *U.S.G.S. Bulletin* 1839J, 24.
- Torney, D.R., Grove, T.L., and Bryan, W.B. (1987) Experimental petrology of normal MORB near the Kane Fracture Zone: 22°–25° N, mid-Atlantic ridge. *Contributions to Mineralogy and Petrology*, 96, 121–139.
- Vandemeulebrouck, J., Sabroux, J.C., Halbwachs, M., Surono, I., Poussielgue, N., Grangeon, J., and Tabbagh, J. (2000) Hydroacoustic noise precursors of the 1990 eruption of Kelut Volcano, Indonesia. *Journal of Volcanology and Geothermal Research*, 97, 1–4, 443–456.
- Wampler, J.M. and Dooley, R.E. (1975) Potassium-argon determination of Triassic and Eocene igneous activity in Rockingham County, Virginia. *Geological Society of America Abstracts with Programs*, 7, 547.

MANUSCRIPT RECEIVED JANUARY 23, 2001

MANUSCRIPT ACCEPTED OCTOBER 13, 2001

MANUSCRIPT HANDLED BY MICHAEL TOPLIS