

LETTERS

Closure in crystal size distributions (CSD), verification of CSD calculations, and the significance of CSD fans

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ABSTRACT

Crystal size distribution (CSD) measurements are susceptible to the closure problem, just like chemical compositions. In its simplest form this means that the total crystal content of a rock cannot exceed 100%. Where chemical or thermal effects limit the total quantity of a single phase, closure can occur at lower volumetric phase proportions. This means that parts of the CSD diagram [ln(population density) vs. size] are not accessible. If the volumetric phase proportion is constant, then straight CSDs will appear to rotate around a point at small sizes giving a fan of CSDs. These fans are significant and do show changes in crystal sizes that can be interpreted in terms of magmatic processes. However, the correlation between the slopes (or characteristic lengths) and intercepts of individual CSDs in a family is not significant, but just a consequence of the constant phase proportion effect. Many other graphs, such as characteristic length vs. volumetric phase proportion, can give more information on magmatic processes.

INTRODUCTION

Crystal size distribution (CSD) studies of igneous and metamorphic rocks have become more popular recently partly because it has been realized that they are a quantitative method of looking at igneous processes that is complementary to geochemical studies (e.g., Cashman 1990; Higgins 1999; Marsh 1998). Another reason may be the recent availability of simple methods of calculating CSDs from measurements in two dimensions, such as thin sections (e.g., Higgins 2000; Peterson 1996).

The crystal content of a rock cannot exceed 100%, hence, as with chemical analyzes, we must always be aware of the closure problem. Closure for an individual phase can also occur at less than 100% crystals. For instance, if a rock is made of 50% plagioclase and 50% olivine then closed-system processes, such as simple textural coarsening, cannot change the phase proportions—they are each fixed at a maximum of 50%. Closure has not, so far, been discussed in published CSD studies. However, I will show that it must be considered in all CSD studies, both of volcanic and plutonic rocks.

The closure effects described in this paper are unrelated to the “Inherited correlation” effect of Pan (2001). That study was flawed by inappropriate use of CSD equations [see comment by Marsh et al. (2002)].

CLOSURE AND CONSTANT PHASE PROPORTION IN CSD MEASUREMENTS

We are concerned here with the effects of constant phase proportion on CSDs, however, I will show later that even quite large variations in volumetric phase proportions can show the same effects. Clearly, closure is just a special case of this more general problem, where the volumetric phase proportion is equal to one.

The volumetric proportion of phase *i*, V_i , is calculated by integration of the volume of all the crystals:

$$V_i = \sigma \int_0^{\infty} n_i(L) L^3 dL \quad (1)$$

where σ = shape factor of phase *i*, equal to the ratio of the crystal volume to that of a cube of side *L* (see below), and $n_i(L)$ = population density of crystals of phase *i* for size *L*. There are many different definitions of crystal size, but in this paper I will follow Higgins (2000) and other authors, and define size as the length of the longest axis of the smallest rectangular parallelepiped that encloses the crystal.

The shape factor, σ , is expanded into a more applicable form:

$$\sigma = [1 - \Omega (1 - \pi/6)] IS/L^2 \quad (2)$$

where Ω is the roundness factor, which varies from 0 for rectangular parallelepipeds to 1 for a triaxial ellipsoid. This definition accords with the roundness factor mentioned in Higgins (2000). *S*, *I*, and *L* are the short, intermediate, and long dimen-

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sions (or axes) of the parallelepiped or ellipsoid. Obviously, most crystals have a more complex shape, but we are concerned here just with a statistical measure. However, Equation 2 should not be applied to crystals with concave surfaces or holes, as they do not necessarily have volumes that lie between that of an ellipsoid and an equivalent parallelepiped.

Equation 1 applies to all CSDs. Marsh (1988) showed that CSDs in many simple magmatic systems are described by the equation:

$$N_i(L) = n_{i0}e^{-L/C_i} \quad (3)$$

where n_{i0} = final nucleation density and C_i = a constant called the characteristic length. It is equal to the mean length of all the crystals in a straight CSD that extends from zero to infinite size. This distribution is linear on a graph of $\ln(\text{population density})$ $\{\ln[n_i(L)]\}$ vs. size (L ; Fig. 1a). The intercept is $\ln(n_{i0})$ and the slope is $-1/C_i$.

Combining Equations 1 and 3 gives

$$V_i = \sigma \int_0^{\infty} n_{i0} e^{-L/C_i} L^3 dL \quad (4)$$

which can be integrated to give (Marsh 1988)

$$V_i = 6\sigma n_{i0} C_i^4 \quad (5)$$

It should be emphasized that this equation is for straight CSDs and that volumetric phase proportions calculated in this way are sensitive to errors in the shape factor.

A simple straight line CSD is shown in Figure 1a. At low

volumetric phase proportion, for example 1%, the slope of the CSD can change independently of the intercept. For example, if all crystals grow at the same rate and nucleation increases exponentially, then the CSD will move up without changing slope. Once the crystal content reaches 100%, then closure is reached and the CSD is locked. It can only move if the ratio between crystals of different sizes is changed. That is, some crystals must become smaller if others are to enlarge.

A family of straight CSDs for 100% crystallized material define a fan (Fig. 1b). Portions of this fan appear to describe rotation of the CSD around a point commonly close to the left of the diagram. The CSDs together outline a concave-up envelope. Straight CSDs cannot exist in the area above this line. Figure 1c shows another view of the same effect in terms of characteristic length vs. intercept. All straight CSDs can be defined by their intercept and characteristic length. Closure limits describe a curve for each crystal shape. Straight CSDs can only exist below this line.

Closure for a single phase can occur at less than 100% volumetric crystal content, if there are chemical or physical constraints on further crystallization. For instance, if a rock contains 50% plagioclase and 50% olivine, then closure for each phase is at 50%. Thermal conditions may also limit crystal content to a maximum value. If the crystals of a phase in a sample are far from their closure limit, then the intercept and slope can change independently giving two degrees of freedom. However, as the crystal content approaches the closure value, then any process that changes the slope of the CSD must also change the intercept—in this situation, there is essentially only one degree of freedom. Therefore, crystals must dissolve (melt) or divide into sub-grains to en-

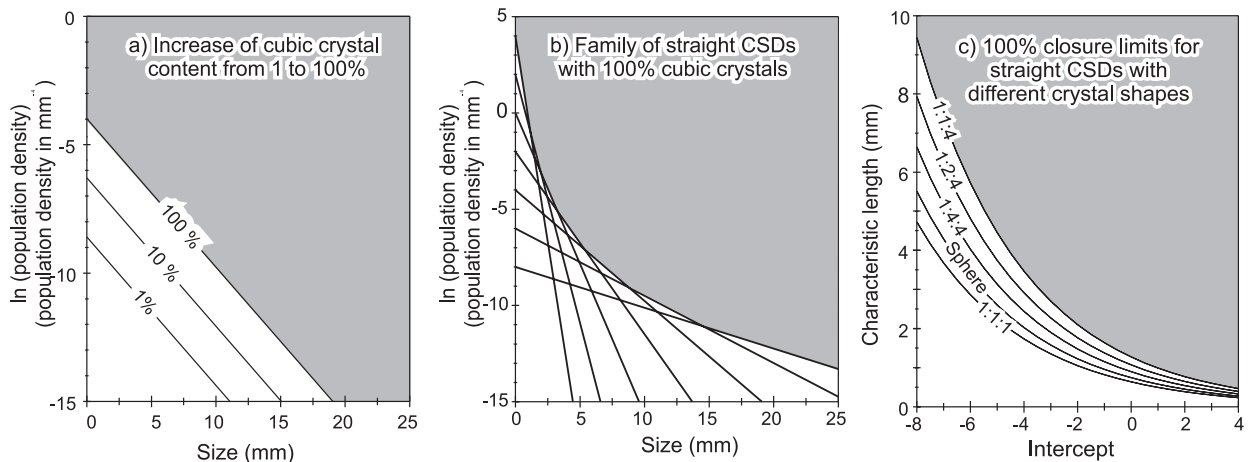


FIGURE 1. (a) The closure problem in CSDs, illustrated for cubic crystals. A straight CSD with a slope of -0.57 (characteristic length 1.73 mm) and an intercept of -8.6 has a volumetric phase proportion of 1%. If the intercept is increased to -4.0 , for example by crystal growth and exponential increase in nucleation rate, then the crystal content will reach 100% and textural changes will stop. At 100% crystals, the CSD is locked and cannot move into the grey area of the diagram. (b) A fan of straight-line CSDs with 100% crystals is tangential to a concave-up curve. Straight CSDs cannot exist in the grey part of the diagram. (c) Straight CSDs can be represented by a point on a graph of characteristic length ($-1/\text{slope}$) vs. intercept. Possible CSDs for 100% crystals proscribe a curve. CSDs can exist below, but not above this line. The position of the line differs for different shapes, here indicated by the aspect ratios of rectangular parallelepipeds and spheres. It is assumed here that the crystals completely fill the volume.

able changes in the slope and intercept of the CSD.

Natural examples of fanning CSDs may not exactly resemble Figure 1b, because the volumetric phase proportion may vary (Cashman and Marsh 1988; Hammer et al. 1999). However, as will be seen later in the example, even in this situation the correlation between characteristic length and intercept is still evident and is little perturbed even by quite large variations in volumetric phase proportions.

VERIFICATION OF CSD DATA

Equations 1 and 5 are a useful way to verify that CSDs have been correctly converted from two-dimensional data, by comparing the measured value of the phase proportion with that defined by the CSD. However, caution must be exercised as the errors in phase proportion estimated from CSDs can be significant, especially for CSDs in which much of the phase volume is contributed by the largest crystals. Such crystals are not numerous and hence their population density is not well known.

Equation 5 can be used directly for straight CSDs. For complex, curved CSDs the corresponding volumetric proportion of the phase can be calculated from the shape factor, the width and mid-points of the size intervals, and the population density of those intervals. For these calculations, a discrete form of Equation 1 is as follows:

$$V_i = \sigma \sum n_j (\bar{L}_j) \bar{L}_j^3 W_j \quad (6)$$

where j is the number of interval, W_j is the width of the interval, and \bar{L}_j is the mean size of the interval j . It should be remembered that $\ln[n_j(\bar{L}_j)]$ is plotted in a CSD diagram. This calculation has been incorporated into version 1.2 of the program CSDCorrections (Higgins 2000; <http://dsacom.uqac.ca/~mhiggins/csdcorrections.html>).

The volumetric proportion of the phase in a rock is equal to the area proportion of the phase in any section (Delesse 1847). The fabric of the rock and the orientation of the section are not important—any plane will give the same area. Crystal intersection areas are commonly measured at the same time as the intersection length and width of the crystals. They can also be determined by point counting or by automatic image analysis. The total crystal area is independent of the length or width measurements used to construct CSDs. Volumetric phase proportions can also be calculated in some cases by mass balance of chemical compositions or even density (for two-phase rocks).

EXAMPLE—LAVAS FROM MOUNT TARANAKI

A series of 14 samples of andesite lava and tuff from Mt. Taranaki (Egmont volcano), New Zealand, provide a good test of the methods developed above (Higgins 1996). Although individual CSDs are slightly curved, they are straight enough that both Equations 5 and 6 can be used to calculate the volumetric phase proportions, and the results of the two methods can be compared with the actual volumetric phase proportion.

The original plagioclase intersection data were recalculated using the methods of Higgins (2000; Fig. 2a). The crystal shape was determined following Higgins (1994): the crystals are

euhedral hence the roundness equals zero (parallelepipeds). The mode of the intersection width/length ratios is 0.33 hence the ratio $I/S = 3.0$. The skewness of the intersection width/length ratios suggested that the ratio L/I is close to one, giving an overall aspect ratio of 1:3:3. The actual volumetric phase proportions were determined from the total area of the plagioclase crystals in thin section. There is a good correlation between the actual volumetric phase proportion and that determined from the CSD using Equation 6 and the CSD calculated with the program CSDCorrections (Fig. 2b). Most of the CSDs are almost straight, hence the volumetric phase proportion also can be calculated with Equation 5, using the slope and characteristic length from the regression of the actual CSD. Again there is a good correlation between the actual volumetric phase proportions and the calculated values, except for two slightly more curved CSDs (Fig. 2c). The overall agreement between the two methods of calculating the volumetric phase proportions from the CSD and the values determined independently from the crystal areas, indicates that the CSDs have been correctly measured and can now be interpreted.

It is difficult to compare a series of samples on a “classic” CSD diagram such as Figure 2a as they all tend to overlap. Another way of looking at the variation between straight or almost straight CSDs is to regress the CSD and plot the intercept against the characteristic length (Fig. 2d)—these parameters were used to calculate the volumetric phase per cent used in Figure 2c. It is clear that despite the relatively low volumetric phase proportion of plagioclase, and its variability (10–30%), the correlation between intercept and characteristic length is the most important component of the variation in this diagram. The variation normal to the closure limit is much less evident and is controlled by the volumetric phase proportions.

There are several better ways of looking at these data. One such graph plots characteristic length against volumetric phase proportion (Fig. 2e). This diagram has the advantage that it is completely accessible. That is, there are no forbidden zones, as is the case for the slope vs. characteristic length diagram (e.g., Figs. 1b and 2d). This accessibility is because the two parameters are completely independent.

The Taranaki CSDs fall into three groups. Fanthams Peak lavas have the smallest characteristic lengths and volumetric phase proportions. This area is the youngest part of the volcano, hence it would be expected that the lavas have lower residence times and are crystal poor. The Burrell ash (sample 12) also falls in this group suggesting that the current magma chamber may be similar or identical to that which produced other lavas of Fanthams Peak. The Staircase lavas have the greatest characteristic lengths. This result indicates that the magma chamber was evolving slowly at this time, either because it was large or because the eruption rate was slow. The Summit and Castle lavas have intermediate characteristic lengths, but the greatest range in volumetric phase abundance. The most crystal-rich samples are from the base of thick flows, suggesting either that there was crystal settling in the lava flows or that the first part of the magma chamber to be sampled was richer in plagioclase.

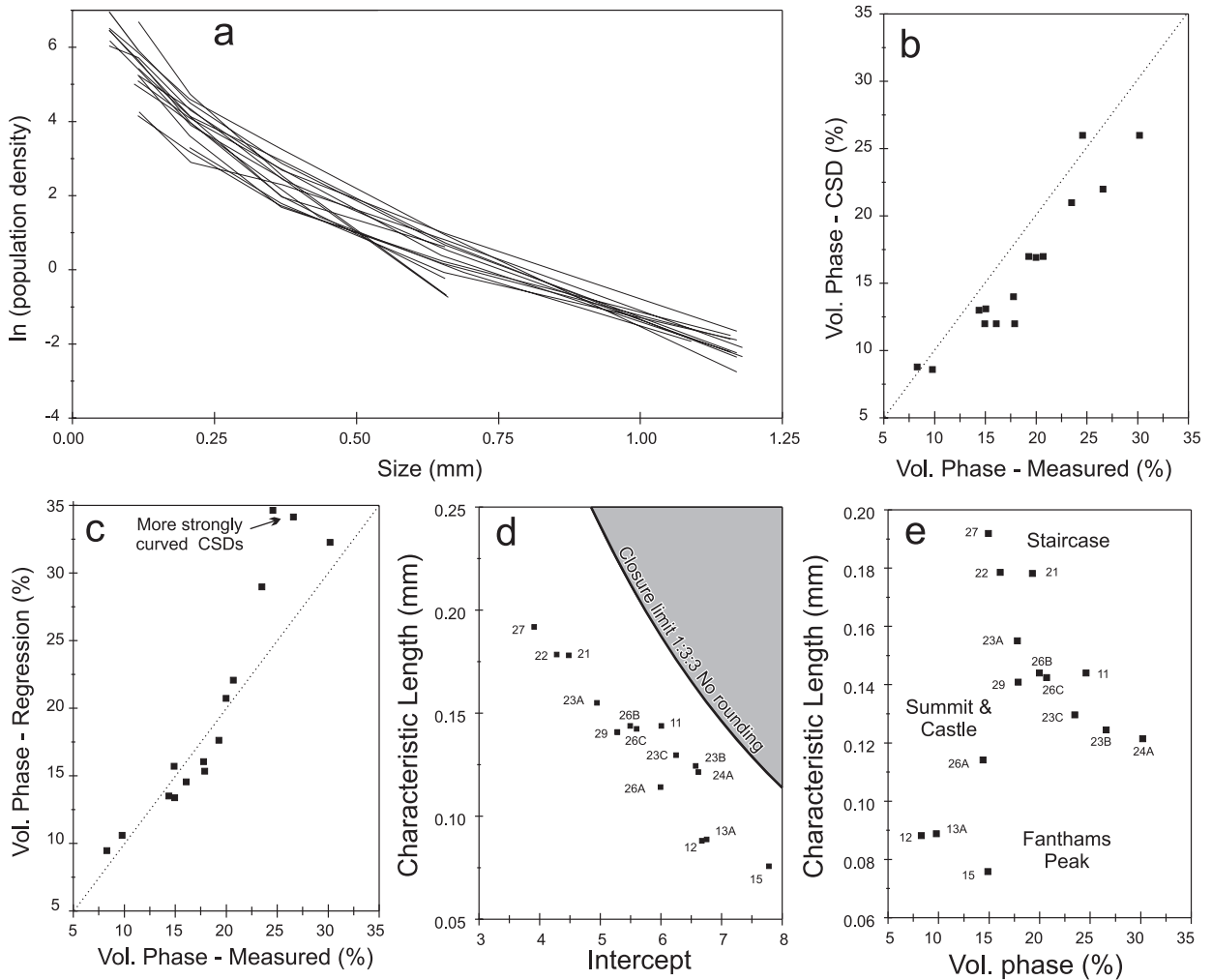


FIGURE 2. CSD data from 14 samples of andesite lava, Egmont volcano, Mt. Taranaki, New Zealand (Higgins 1996). Original 2D data have been converted to 3D values using the program CSDCorrections 1.2 (Higgins 2000). (a) Crystal size distribution diagrams. Most individual CSDs are almost straight. However, the envelope of CSDs describes a curve. This is the effect described as CSD fanning. (b) Comparison of volumetric phase proportion in percent determined from the total area of crystals in thin section (measured) vs. that calculated from the CSD and crystal shape using Equation 6. (c) Comparison of measured volumetric phase proportion vs. that calculated for a straight CSD with intercept and slope derived from a regression of the actual CSD using Equation 5. The two samples that lie above the correlation line are more strongly curved than the other samples. (d) Characteristic length vs. intercept of the regressed CSDs. Samples cannot lie above the closure limit. Although the Taranaki samples are far from the closure limit, there is a strong correlation between characteristic length and intercept. The prefix NZ has been omitted from the labels. (e) Volumetric phase proportion vs. characteristic length. The different groups of lavas are clearly separated in this diagram.

CONCLUSIONS AND RECOMMENDATIONS

(1) Closure limits form an easily calculated, convenient, and useful reference for CSD studies. They indicate how much liberty is possible for CSD movement during petrologically important processes. They can also show if correlated characteristic lengths (or slopes) and intercepts of straight CSDs are significant or not. Closure limits should be indicated on published CSDs where possible.

(2) Calculation of volumetric phase proportions from CSDs

can be used to verify that CSDs have been correctly determined from two-dimensional measurements. Common errors, such as in the determination of crystal shapes or by the use of inappropriate equations, can be easily recognized and corrected.

(3) It is commonly observed that there is a strong correlation between the characteristic length and intercept of CSDs. Restricted variations in volumetric phase proportions can account for this effect. A more informative diagram may be that of characteristic length vs. volumetric phase proportion.

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