Time Scales of Crystal Fractionation in Magma Chambers—Integrating Physical, Isotopic and Geochemical Perspectives

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A simple heat balance model for an evolving magma chamber is used to make predictions of the time scales for magma differentiation, which are compared with geological and isotopic constraints on the rates of crystallization and differentiation. In a 10 km³ magma chamber releasing thermal energy at a rate of 100 MW, basalt and rhyolite magmas should reach 50% crystallization after ~2500 and <1500 years, respectively. The tendency for phenocrysts to remain suspended in a cooling magma increases with melt viscosity and hence the degree of magma differentiation. The time scales of crystallization and differentiation may be estimated on the basis of the U-series isotope compositions of separated crystals and bulk rocks, crystal size distributions, and trace element and Sr isotope profiles in phenocryst phases. The last of these indicate crystal residence ages in the melt of tens of years to 100 years. Short residence and even differentiation times are also obtained from Ra–Th isotope studies of alkali feldspars, and rocks that experienced alkali feldspar fractionation. However, much older ages of \(10^3\)–\(10^5\) years for separated phases have been reported from recent volcanic centres in St Vincent in the Lesser Antilles, Vesuvius in Italy, the Kenya Rift Valley and Long Valley, California. These old crystal ages are all from relatively evolved igneous rocks, as predicted from their higher melt viscosities and simple models of cooling and crystal settling. However, the old ages are also typically obtained for complex minerals that are not in bulk equilibrium with their host rocks, and so, apart from offering a minimum estimate of the age of the particular magmatic system, their significance for models of differentiation of the host magmas is not clear. An alternative approach is therefore to determine the variations in U–Th–Ra isotope compositions of bulk rocks reflecting different degrees of magma differentiation, and such data indicate that differentiation in more mafic magmas takes much longer than in more evolved magmas. For example, 50% fractional crystallization of basanite to produce phonolite on Tenerife took ~10³ years, whereas a further 50% fractional crystallization to generate the more evolved phonolites occurred within a few hundred years of eruption. On Tenerife the more mafic magmas fractionated at greater depths, and the rates of fractional crystallization were higher in the more evolved magmas studied. This is readily explained by a cooling model in which a large volume of primitive magma deep in the crust has a longer cooling time than a smaller body of differentiated magma at shallower depths in the crust.

KEY WORDS: fractional crystallization; time scales; U-series isotopes; crystal settling; magma chambers

INTRODUCTION

Keith Cox was fascinated by the differentiation processes responsible for the chemical variations in igneous rocks, and he contributed much to discussions of the role of fractional crystallization. Magmas crystallize over a range...
in temperature, and, in principle, there is ample opportunity for early formed crystals to separate from the liquid, and for this to be the primary mechanism of differentiation in magmatic systems. The ease with which such separation can happen depends on the density contrast between the crystals and the liquid, the viscosity of the liquid, the size of the crystals and the dynamics of the magma chamber system. Crystal separation can be facilitated by flow differentiation and filter pressing. Crystallization results in a build-up of volatiles in the liquid, which may in turn trigger explosive eruptions; conversely, magma degassing can cause crystallization. Thus, there is considerable interest in the links between crystallization and volcanic eruptions, and in the primary controls on magma differentiation.

The presence of crystals of different sizes in igneous rocks highlights the fact that crystals form under different conditions, particularly of temperature, cooling rate and, in some cases, volatile contents. The crystals have bulk compositions that differ from their host melts, and thus separation of the crystals from the melt causes the magma composition to change. Such models of fractional crystallization have become widely accepted as the principal cause of magma differentiation (Bowen, 1928). In such models, the time scales of magma differentiation depend both on the rate of crystallization and on the mechanism for the physical separation of the crystals and the residual liquid. The rate of crystallization can be a response to falling temperature, whereupon it increases with the rate of cooling, and hence with decreasing depth in the crust. Alternatively, if crystallization occurs in response to magma degassing, it can be extremely rapid (e.g., Brophy et al., 1999).

Once crystals have formed, the mechanism by which the crystals and residual liquid separate from each other depends, inter alia, on their density contrast and whether the crystals are distributed throughout the liquid (allowing operating within magma chambers). Furthermore, the less influenced by being an undergraduate at Oxford short-lived radiogenic isotopes, and the development of systems (Pyle, 1992; Albare `de, 1993; Condomines, 1994). The controls on crystallization rate and crystal settling rate remain important research topics in igneous petrology, as are the links between crystallization events and volcanic eruptions, and the primary controls on magma differentiation. Key aspects are the time scales of crystallization and differentiation. In general, one may distinguish a number of relevant time scales associated with the processes of magma evolution by fractional crystallization: the time taken for a differentiated magma (or liquid) to evolve from its parent, the time that a given magma composition might remain in existence without undergoing any further evolution before its eruption, and the age of the crystals in a magma recording processes occurring within magma chambers. Furthermore, the average residence time of magma in a replenished, erupting chamber, given by chamber volume divided by recharge rate, is a characteristic parameter in open systems (Pyle, 1992; Albarède, 1993; Condomines, 1994).

One way of quantifying these time scales is to use short-lived radiogenic isotopes, and the development of improved mass spectrometric techniques has prompted a significant increase in the number of U-series isotope analyses on volcanic rocks and separated minerals. The more widely used isotopes of $^{230}$Th and $^{226}$Ra have half lives of 75·4 ky and 1·6 ky, respectively, and can be used to investigate processes operative within the last 300 ky. Other potentially useful isotopes in the study of magmatic processes have half lives of a few decades, or even days, and these can be compared with the ages of crystals inferred from crystal size distributions (CSD) (Marsh, 1996), and trace element and Sr isotope profiles in crystals (Knesel et al., 1999; Zellmer et al., 1999). In this paper
we discuss a basic heat balance model for an evolving magma chamber to derive physically based predictions of differentiation time scales and then make comparisons with geological and isotopic constraints on the rates of crystallization and differentiation.

**PHYSICAL PRINCIPLES OF COOLING AND CRYSTALLIZATION**

Magma crystallizes as a result of cooling, degassing (e.g. Brophy *et al.*, 1999), or during concurrent cooling and chemical modification during crustal assimilation or magma mixing. Conversely, crystallization in a magma chamber may be inhibited or reversed by heating caused by fresh inputs of more mafic magma or by magma mixing. In this section we consider cooling of a chemically closed magma, being the simplest reference model with which to address real magma systems. What rates of crystallization (and hence chemical evolution of the remaining liquid) can be expected in such a model system? The rate at which thermal energy is lost from magma chambers maintains geothermal power outputs of 100–1000 MW (e.g. Hochstein, 1995). This is supplied by the cooling and crystallization of a shallow magma body and cooling of already solidified intrusions. The maximum amount of magmatic cooling can be estimated by equating the power output, $P$, to the thermal energy lost from a fixed volume of magma ($V$) divided by the cooling time $t_{cool}$:

$$P = V \rho (|\Delta T| + \Phi L)/t_{cool}$$

where $\rho$ is the magma density, $\epsilon$ the specific heat capacity, $L$ the latent heat of crystallization, $\Delta T$ the temperature drop and $\Phi$ the mass fraction of crystals grown over this temperature range. Rearranging and substituting values representative of basalt magma [$\rho = 2600$ kg/m$^3$, $\epsilon = 1500$ J/kg per K, $L = 4 \times 10^5$ J/kg (Spera, 2000)] leads to

$$t_{cool} = 3.9 \times 10^6 V/|\Delta T| + 267\Phi/P.$$  

In this model, cooling by 75 K accompanied by 50% crystallization takes from 260 to 26 000 years for $V$ and $P$ covering the ranges of 10–100 km$^3$ and 100–1000 MW, respectively. These time scales are not too dissimilar from estimates obtained from models that explicitly couple convective heat flux in the magma to conductive heating of initially cold country rock, an assumed constant heat flux or assumed constant wall-rock temperature (e.g. Spera, 1980; Martin *et al.*, 1987; Wolff *et al.*, 1990; Hort, 1997).

To illustrate the predicted cooling and crystallization behaviour further, we consider a 10 km$^3$ magma chamber releasing 100 MW. This power output may, in general, be achieved by various combinations of $\Delta T$ and $\Phi$ according to equation (1), as illustrated in Fig. 1. Also shown are the paths of increasing crystal mass fraction with falling temperature for a primitive basalt and a rhyolite; intermediate magmas will define curves with intermediate values of the liquidus–solidus temperature interval (e.g. Sutton *et al.*, 2000). The diagram shows that, all else being equal, the basalt reaches 50% crystallization after some 2500 years whereas the rhyolite reaches 50% crystallization in <1500 years because of its narrower crystallization range and lower latent heat of crystallization.

Having derived estimates of crystallization time scales in closed cooling magma chambers, the important question remains whether the crystals growing within the convecting interior of the chamber are retained by the magma or settle to the chamber floor. Adopting the experimentally verified model of Martin & Nokes (1988)
Table 1: Values of the ratio $S = t_{\text{settle}}/t_{\text{cool}}$ for selected magmas, according to equations (2) and (3) and $P = 100$ MW, $V = 10$ km$^3$ and $\Delta \rho = 500$ kg/m$^3$

<table>
<thead>
<tr>
<th>Parent magma</th>
<th>Daughter magma</th>
<th>$\mu$ (Pa s)</th>
<th>$\rho$ (kg/m$^3$)</th>
<th>$c$ (J/kg per K)</th>
<th>$L$ (J/kg)</th>
<th>$\Delta T$ (K)</th>
<th>$\Phi$</th>
<th>$S$</th>
</tr>
</thead>
<tbody>
<tr>
<td>High-magnesia basalt</td>
<td>evolved liquid</td>
<td>$10^4$</td>
<td>2600</td>
<td>$1.5 \times 10^4$</td>
<td>$4 \times 10^4$</td>
<td>30</td>
<td>0.2</td>
<td>0.0061</td>
</tr>
<tr>
<td>Low-magnesia andesite</td>
<td>basalt</td>
<td>$10^4$</td>
<td>2500</td>
<td>$1.5 \times 10^4$</td>
<td>$4 \times 10^4$</td>
<td>75</td>
<td>0.5</td>
<td>0.25</td>
</tr>
<tr>
<td>Dacite</td>
<td>rhyolite</td>
<td>$10^6$</td>
<td>2200</td>
<td>$1.6 \times 10^5$</td>
<td>$2.5 \times 10^5$</td>
<td>40</td>
<td>0.5</td>
<td>47</td>
</tr>
</tbody>
</table>

Setting the chamber height to $H = V^{1/3}$ and considering a canonical example with $\Delta \rho = 500$ kg/m$^3$, $r = 10^{-3}$ m, $V = 10^{10}$ m$^3$ and $P = 100$ MW allows some comparisons between magmas of different composition (Table 1).

The intuitive qualitative result that the tendency to retain phenocrysts in a cooling magma chamber increases with the degree of magma evolution (i.e. melt viscosity) remains. This expectation is borne out by the common increase in modal phenocryst content with increasing silica content (e.g. Marsh, 1981; Ewart, 1982) and the occurrence of plagioclase-accumulative magmas whose melts have densities close to those of plagioclase feldspar (e.g. Cox & Mitchell, 1989).

We end this section with the summarizing statement that closed system magma chambers are expected to cool over time scales measured in decades to tens of thousands of years, depending on their size, power loss and to some extent initial composition. Furthermore, cooling and crystal settling models imply that primitive basalts affected solely by cooling in a chamber will contain a small proportion of near-liquidus phenocrysts whereas more evolved magmas will have an increasing proportion of phenocrysts inherited from early stages of the magma’s history.

Setting the chamber height to $H = V^{1/3}$ and considering a canonical example with $\Delta \rho = 500$ kg/m$^3$, $r = 10^{-3}$ m, $V = 10^{10}$ m$^3$ and $P = 100$ MW allows some comparisons between magmas of different composition (Table 1).
rocks and minerals formed within the last few hundred thousand years. However, because these intermediate isotopes are parts of decay chains, they have the distinctive property that both the parent and the daughter isotopes are radioactive. Thus, in a uranium-bearing system that has been left undisturbed for a few million years, a state of secular equilibrium is established such that the rate of decay, or the ‘activity’, of the daughter nuclide in the chain is equal to that of the parent. If that decay chain is disturbed by the fractionation of a parent from a daughter nuclide it takes time for secular equilibrium to be restored by radioactive decay. In that period before secular equilibrium is restored, the age of the fractionation event can be determined from the measured isotope ratios [see Gill et al. (1992) and Dickin (1995) for fuller discussions of the theory of short-lived isotopes].

U–Th and Ra–Th isotope results are usually presented in an isochron diagram [Fig. 2], in which the parent and the daughter isotopes are ratioed to an isotope not involved in the decay scheme. Further, the isotope ratios are expressed as activity ratios, and so in secular equilibrium the activity ratios involving the parent and the daughter isotopes are unity. All samples in secular equilibrium therefore plot on the 1:1 line in an isochron diagram (Fig. 2); this line is known as the equiline (Allegre & Condomines, 1976). The isochron diagram may also be referred to as an equiline diagram. Secular equilibrium is disturbed by any process that fractionates the parent from the daughter nuclides, in this case U from Th, and when that process took place is determined by the difference between the initial and the present-day Th isotope ratios. The initial Th isotope ratio is typically that at the time magma was extracted from its source, and/or subsequently modified en route to the surface. It may therefore be significantly different from the Th isotope ratios of magmas at the time of eruption, and in practice age information is often obtained in one of two ways.

First, samples with different U/Th ratios may plot on a positive linear array, an isochron (Figs 2 and 3), and the slope of that array will correspond to an age. That age will be the time of formation of the different U/Th ratios, and the age will be geologically meaningful if the samples had the same initial Th isotope ratio, and their U/Th and Th isotope ratios have remained in a closed system subsequently. In igneous systems, the samples with different U/Th ratios may be suites of comagmatic whole rocks or mineral separates. However, U and Th are highly incompatible in most common igneous minerals, and so closed system differentiation processes usually do not result in significant variations in U/Th ratios. Exceptions include magnetite, which appears to have very variable U/Th ratios, and other parent/daughter ratios, such as Ra/Th, are fractionated by, for example, the feldspars. Accessory minerals, such as apatite and zircon, may also show strong $^{230}$Th–$^{238}$U disequilibria, and if these minerals crystallized at the time of eruption their U–Th isotope compositions can be used to determine the ages of volcanic rocks (e.g. Condomines, 1997).

Second, it follows that comagmatic igneous rocks may have very similar U/Th ratios, but in some cases they have very different Th isotope ratios. Thus, they plot in a near-vertical array in a U–Th isochron diagram (Fig. 2). In primitive rocks such vertical arrays may reflect dynamic melting processes (e.g. McKenzie, 1985), but in rocks related by fractional crystallization they may reflect the time taken for fractional crystallization to occur. In the latter interpretation it is envisaged that the parental magma had high $^{230}$Th/$^{238}$Th and that the lower values are due to radioactive decay during magma differentiation.

In summary, short-lived isotopes have the potential to allow the time since secular equilibrium was disturbed to the present day to be determined. U–Th–Ra isotopes are the most widely used in the study of igneous rocks,
but there is also considerable potential to exploit isotopes with even shorter half lives, including \(^{210}\text{Pb}\) (22 years), \(^{210}\text{Bi}\) (5 days) and \(^{210}\text{Po}\) (138 days), in addition to \(^{231}\text{Pa}\) (32 000 years). Short-lived isotopes can be used to determine eruption ages, provided that the minerals analysed clearly crystallized at, or very close to, the time of eruption (Fig. 3; Peate et al., 1996; Condomines, 1997; Black et al., 1998a). However, in any attempt to investigate the pre-eruption history of magmatic rocks it is critical to have independent evidence for the eruption age, and precise estimates of the ages of pre-historic eruptions are relatively rare. It is also important to reiterate the kinds of information that can be obtained. These include (1) the ages of crystals, which may or may not be in equilibrium with their host magmas, (2) the time that a magma was in existence before eruption without changing its bulk composition, (3) the time taken for a more differentiated magma to have evolved from its parent magma, and (4) the mean residence time of magma in a continuously replenished and tapped chamber (e.g. Pyle, 1992; Condomines, 1994; Hughes & Hawkesworth, 1999).

Fig. 4. The whole-rock Ra–Th isochron for the main lava pile trachyte samples from Longonot volcano, Kenya, that also have \(^{230}\text{Th}\) excess (Evans, 1999). It is assumed for the sake of the age calculation that the bulk distribution coefficients for Ra and Ra were similar, and the variation in Th/Ba has been modelled by progressive fractional crystallization of an assemblage consisting very largely of sanidine (Evans, 1999).

**MAGMA DIFFERENTIATION**

In principle, differences in the U–Th–Ra isotope ratios of closed system comagmatic rocks offer relatively straightforward information on the time scales of magma differentiation. It may be difficult to establish closed system differentiation, but if that can be demonstrated, it may be easier to interpret the isotope ratios of bulk rocks than those of separated crystals (see below). As noted above, closed system differentiation is unlikely to result in much variation in U/Th ratios, except where accessory minerals are relatively abundant. In contrast, Ra has a higher partition coefficient than Th in plagioclase and it is compatible in alkali feldspar. Thus, shallow level differentiation may result in whole-rock Ra–Th isochrons (see Fig. 4), and variations in Th isotopes with little change in U/Th (Fig. 2). Interpretation of Ra isotope data is complicated when the system has been open to fluids. Ra is strongly partitioned into the fluid phase relative to U and Th, and so the addition or removal of Ra in fluids may result in an Ra–Th isochron age that differs from the U–Th crystallization age (e.g. Black et al., 1998b). In general, however, significant variations in Th isotopes require periods of tens of thousands of years to develop, and variations in Ra isotopes require a few thousand years.

Evans (1999) analysed U–Th–Ra isotope ratios in a stratigraphic package of trachyte lavas from the volcano Longonot in the Gregory Rift, Kenya. These rocks were erupted in the period 5650–3280 years BP (Clarke et al., 1990), and the degree of fractionation increases up-sequence. The rocks are aphyric and they have been
modelled as the products of closed system fractionation of an assemblage in which Ba and Ra were highly compatible. Thus, they have large ranges in Ba/Th and Ra/Th ratios, and the samples with excess 230Th represent a range of up to ~15% fractional crystallization (Evans, 1999). These isotope data are illustrated in an equiline diagram of (226Ra)/Ba–(230Th)/Ba (Fig. 4), normalized to Ba abundances because Ra and Ba were thought to have similar partition coefficients in common igneous minerals (Williams et al., 1986). The whole-rock samples from Longonot, representing ~5 km³ of magma, define a linear array with a slope corresponding to an age of 4274 ±1389 years, consistent with their eruption ages of 5650–3280 years BP inferred from 14C dating (Clarke et al., 1990). It is likely that Ra is more compatible in sanidine than is Ba (Blundy & Wood, 1991; Zellmer, 2000). However, that does not alter the main conclusion that the Ra–Th isotope system yields an age for magma differentiation for the Longonot trachytes, and that age is indistinguishable from the age of eruption. The implication is that differentiation took place over short periods of time, and only just before eruption.

In a similar study, Condomines et al. (1995) reported 238U–230Th–226Ra values on lavas erupted from Mt Etna in Sicily over the last 2000 years. Initial (226Ra/230Th) values decrease with increasing differentiation, suggesting a link between residence time in the crust and the degree of differentiation. Some of the magmas were affected by crustal contamination and the higher initial (226Ra/230Th) values tend to be in the more contaminated rocks. None the less, Condomines et al. (1995) concluded that differentiation from hawaiite to mugearite occurred on a time scale of ≤200 years. The Th contents range from 7 to 12 ppm and, assuming the simplest closed system model of fractional crystallization and $D_{Th} = 3 \times 10^{-4}$ (e.g. La Tourette et al., 1993), the degree of fractionation is therefore ~40%. The post-1977 eruptions appear to have bypassed the main magma chamber, and this may have been a factor in these comparatively short residence times (perhaps even tens rather than hundreds of years). In contrast to the results from Longonot and Etna, a recent study by Thomas (1999) suggests that differentiation from basanite to phonolite on the island of Tenerife in the Canaries may have taken ~200 ky.

Figure 5 summarizes selected data from the Tenerife rocks. Tenerife is the largest island (2058 km²) in the Canaries, and it is dominated by the imposing 3718 m Mt Teide within the Las Cañadas caldera in the centre of the island. Since 1.6 Ma there have been four distinct volcanic cycles, each separated by periods of dormancy of between 120 and 250 ky (Ancochea et al., 1990; Martí et al., 1994). Each cycle involved the extrusion of mafic to intermediate magmas followed by voluminous explosive phonolitic eruptions linked to caldera collapse. The samples for the U–Th–Ra isotope study were taken from the basanites to phonolites of the last cycle, which started ~175 ky ago.

A key feature of these rocks is the lack of evidence for significant amounts of crustal contamination or magma mixing. Most of the samples analysed have restricted $^{87}Sr/^{86}Sr = 0.70298–0.70326$, and more scattered values are observed only in the highly evolved rocks with <30 ppm Sr, for which it is difficult to distinguish the effects of alteration from small amounts of crustal contamination (Thomas, 1999). More compelling perhaps is the tight, slightly curved array of, for example, Rb vs Zr (Fig. 5a), as Rb/Zr ratios decrease in the most evolved rocks (attributed to alkali feldspar fractionation, which also resulted in a sharp decrease in Ba/Zr), and yet most crustal contaminants have higher Rb/Zr ratios than mantle-derived magmas. Thus, closed system fractional crystallization (of up to ~77%) appears to have been the dominant process of differentiation for these lavas, consistent with the relatively large volumes of mafic and small volumes of more differentiated rock types (70% and 30%, respectively).
The initial \((230\text{Th}/238\text{U})\) values in most of the basanites and phonolites range from 1·5 to 1·05, and there is just one sample (a cumulate) with \((230\text{Th}/238\text{U}) < 1\) (Fig. 5b). The basanites tend to have higher \((230\text{Th}/238\text{U})\) values, and these decrease with increasing differentiation, \((230\text{Th}/238\text{U})\), changes with time, and Zr with the amount of fractional crystallization, and in a diagram of \((230\text{Th}/238\text{U})\) vs Zr most of the data plot in a concave-up array. This suggests that early fractionation in the more mafic rocks from, for example, 250–500 ppm Zr took longer than the subsequent fractionation to more differentiated magmas. The basanites have a very similar average U/Th ratio to the phonolites (0·26 and 0·27, respectively). are consistent with long time scales of magma di

fractional crystallization, and in a diagram of \((230\text{Th}/238\text{U})\) to decay from 1·5 to 1·06 is \(~ 230\text{ky}.\) Provided that the younger phonolites were derived from parental magmas with similar \((230\text{Th}/238\text{U})\) to the older basanites, as indicated by the available chemical and stratigraphic data, it appears that the erupted phonolites evolved from magmas that had been emplaced into the deep crust \(~ 230 \pm 70\text{ ky}\) previously. The youngest basanite erupted in 1909 has a lower \((230\text{Th}/238\text{U})\) value of 1·23, but even if this was more representative of the primary magmas the calculated time for differentiation from basanite to phonolite is still almost 100 ky.

In detail, the mafic magmas and intermediate lavas are thought to have evolved in deep crustal storage zones at 6–9 kbar \(P_{\text{max}}\), and then subsequently differentiated in a shallow chamber at 1·5–2·5 kbar \(P_{\text{max}}\) (Ablay, 1997; Ablay et al., 1998). Thus, the concave-up form of the \((230\text{Th}/238\text{U})\) Zr array indicates that differentiation in the deep crust took much longer than that at shallow levels. Such a model is clearly consistent with slower crystallization rates at the higher temperatures in the deep crust and more rapid crystallization at shallow levels. It is noteworthy that the oldest phonolite in the youngest magmatic cycle was erupted just 2000 years ago, and that whole-rock \(226\text{Ra}/230\text{Th}\) data on young basanites to phonolites yield an age of 2300 \pm 80\text{ years} (Thomas, 1999). Thus differentiation to the most evolved phonolites happened within a few hundred years before eruption. Finally, it is intriguing that the overall time of \(~ 230\text{ ky}\) for differentiation from basanite to phonolite is broadly similar to the periodicity of the eruption cycles on Tenerife over the last 1·6 My. The simplest interpretation is that this may represent the time scale of the release of primary basalts from their mantle source regions.

Elsewhere, a \(U\)–Th isotope study of the Laacher See tephra by Bourdon et al. (1994) indicated that, although some of the younger pumices (13 \pm 3 ky) had short magma residence times of a maximum of a few thousand years (from analyses of separated minerals), \(~ 100\text{ ky}\) were needed to explain the difference in \(Th\) isotopes between the inferred parental basanitic magmas and the erupted phonolite. Widom et al. (1992) suggested that the time of evolution from alkali basalt to trachyte on Sáo Miguel in the Azores was 90 ky, and Reagan et al. (1992) inferred a time of \(< 150\text{ ky}\) for differentiation from basanite to phonolite beneath Mt Erebus in Antarctica. Reagan et al. (1992) reported average \(226\text{Ra}_{\text{th}}/230\text{Th}\) ages of 2380 years for anorthoclase–glass pairs, and concluded that the residence time of phonolitic magmas at shallow levels was \(~ 3000\text{ years}\). Overall, there are still too few data to draw sweeping conclusions, but the present data are consistent with long time scales of magma differentiation of \(~ 10^7\text{ ky}\) for basanitic magmas deep in the crust, and with very much shorter periods of up to a few thousand years at shallower depths when feldspars, and particularly alkali feldspars, are on the liquidus.

ZONED MAGMA CHAMBERS

Evidence from compositionally zoned ash flow tufts and from systematic compositional variations in lava sequences indicates that the upper parts of magma chambers may become vertically zoned. Phenocryst growth often post-dates the development of zonation (Smith, 1979; Hildreth, 1981). Provided that eruption does not result in mixing of layers, internal isochrons can, in principle, be used to determine residence times of magmas within layers and differences in crystallization ages of magmas from different layers.

We are aware of only one detailed study of zoned units using \(U\)-series isotopes, although Widom et al. (1992), Bourdon et al. (1994) and Evans (1999) concluded that the rate of formation of zoned magma bodies was rapid. Black et al. (in preparation) applied the \(230\text{Th}/238\text{U}\) method to the first caldera-forming ash flow tuff erupted from the Menengai volcano, Gregory Rift, Kenya. The earliest erupted products were strongly peralkaline (pantelleritic) trachytes and these were followed by less peralkaline (comenditic) varieties later in the eruption. Compositional variation in the tuff is essentially continuous and it involved, in the inferred zoned chamber, upward enrichment in \(FeO^+, Na_2O, CaO\) and incompatible trace elements, and decreases in \(Al_2O_3, MgO, Ba\) and Sr. \(U\) /Th varied from 0·42 to 0·17 and \((230\text{Th}/238\text{U})\) from 1·3 to 1·0 in nine samples. Evidence was interpreted to show that the \(U\) enrichment was related to the exsolution and upward migration, shortly before eruption, of a \(U\)-rich carbonate melt from the trachyte magma (Black et al., in preparation). It appears that the presence of a carbonate-rich melt further

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complicates attempts to relate U-series isotope disequilibria in silicic magmas to the nature of their source rocks or the melting process.

AGE INFORMATION FROM CRYSTALS
In principle, the ages of crystals can be determined from measurements of radiogenic isotopes, CSD, and trace element and isotope profiles. However, it is important to have independent evidence that the minerals analysed were in equilibrium with the host rock. Minor phases, such as magnetite, and accessory minerals either in the groundmass or as inclusions in phenocrysts, have been successfully used to determine eruption ages (Fig. 3; Peate et al., 1996; Condomines, 1997). Elsewhere phenocrysts have yielded ages that are indistinguishable from the ages of eruption (e.g. Volpe & Hammond, 1991; Volpe, 1992; Black et al., 1996a), but if the age of eruption is not known it can be difficult to determine whether such minerals formed at the time of eruption, rather than earlier in the evolution of the magma. Similarly, it is critical to establish the extent to which the compositions of phenocrysts are consistent with crystallization from the host magma, or whether they are xenocrystic in origin. For example, relatively old crystals may be preserved in the erupted magma, even though any crystal settling, and hence magma differentiation, may have occurred very recently. Similarly, crystals that are much older than the host lava may be those that crystallized from earlier magma batches and were simply incorporated in younger, and possibly unrelated, magma batches that were erupted through the same conduits.

Radiogenic isotopes
A number of studies have used the Rb-Sr and Ar-Ar isotope systems to document isotope heterogeneities, and to infer ages from crystals and groundmass in high-silica systems, as at Long Valley in California (e.g. Halliday et al., 1989; van den Bogaard & Schirnick, 1995; Davies & Halliday, 1998, and references therein), the Bandelier Tuff, New Mexico (Wolff et al., 1999), and the Kenya Rift Valley (Heumann et al., 1995). These studies utilize the unusually high Rb/Sr ratios (10¹⁰) of the highly differentiated rhyolites (Halliday et al., 1991), which result in rapid changes of \(^{87}Sr/^{86}Sr\) with time. With high-precision thermal IR multispectral scanning (TIMS), time scales of \(\sim 1000\) years can be resolved, and if the diffusion coefficients between minerals and melt are low, it may even be possible to determine the time difference between the formation of different minerals within cogenetic suites (Davies et al., 1994).

One debate over the origin of high-silica rhyolites is whether they evolve in long-lived, large-volume shallow-level magma chambers (Halliday et al., 1989), or are the products of repeated episodes of crustal melting and short magma residence times (Sparks et al., 1990). In one set of models crystals reside in the magma for \(>10^6\) years (Halliday et al., 1989; Christensen & Halliday, 1996); in another they are remobilized from a rind of crystalline mush around the margins of the magma body (Mahood, 1991). Glass-whole-rock Rb/Sr isochrons, and isochrons including plagioclase and alkali feldspars and both glass and biotite inclusions, from Glass Mountain, yielded ages that are up to 700 ky older than the eruption ages (Halliday et al., 1989; Davies et al., 1994). In situ laser \(^{40}Ar-{ }^{39}Ar\) analyses of sanidine phenocrysts and matrix glass confirmed an eruption age of 760 ± 1 ka for the Bishop Tuff, but melt inclusions in quartz yielded ages of 1·9–2·3 Ma (van den Bogaard & Schirnick, 1995). Thus, both melt compositions and crystals preserve ages that are significantly older than the age of eruption, and the presence of ‘old’ melt inclusions would appear to require that their host minerals had been present for hundreds of thousands of years at the time of eruption. Such ages are significantly older than those inferred above for differentiation processes involving felsic magmas in intraplate settings, and the data cannot be used to resolve whether the crystals resided in the magma or a marginal crystal rind. However, Wolff et al. (1999) reported Sr isotope ratios from sanidines, glomerocryst glasses, and quartz with glass inclusions from the Bandelier Tuff, and argued that the quartz crystallized from contaminated magma closer to the magma chamber walls than the glass and sanidine samples. In this case the inferred Sr isotope gradient towards the walls of the chamber offers the possibility of resolving the relative positions of the different phases analysed (Fig. 6). U-Th isotope studies in zircons in rhyolites from Long Valley also indicate pre-eruption residence times of \(~100\) ky (Reid et al., 1997) whereas zircon separates from Taupo rhyolites have U/Th disequilibria implying pre-eruptive ages of \(~10\) ky (Charlier et al., 2000). In one case where the Rb/Sr and U-series methods have been applied to the same rocks, the Olkaria peralkaline rhyolites of the Kenya Rift Valley, analyses of separated phases by both methods point to residence times of \(10^3–10^5\) years (Heumann et al., 1995; Black et al., 1997).

Heath et al. (1998) reported four U-Th mineral-whole-rock isochrons from basaltic andesites erupted from the Soufrière volcano on St Vincent in the Lesser Antilles are in the last 4000 years (Fig. 7). All the minerals had lower U/Th ratios than the whole rocks, and so not all the phases present had been analysed. The U and Th abundances in the mineral separates were higher than expected, indicating that the minerals contained significant inclusions. The data were scattered with mean

999
square weighted deviations (MSWDs) of 2·6–19·3, but they yielded consistent ages of 46–77 ka. The scatter of the data about the ‘errorchrons’ implies that they may represent a mixture of crystals of various ages, but even in that interpretation some of the crystals analysed would appear to be tens of thousands of years old. Heath et al. (1998a) noted how only the rims of the plagioclase phenocrysts were in equilibrium with the host magma, and the cores of these crystals may have been remobilized from significantly older crystal mush, perhaps from the margins of the magma chamber. However, the presence of the groundmass compositions on, or close to, the mineral isochrons from Soufriere suggests that the bulk magma was also tens of thousands of years old, and that is the best estimate of the residence times of these magmas within the crust.

Hughes & Hawkesworth (1999) explored the effects of open system magma replenishment processes on $^{230}$Th–$^{234}$U isotopes. Magmas can be maintained out of secular equilibrium for long periods of time by periodic mixing with new magma batches. For subduction-related magmas, which typically have high ($^{238}$U/$^{230}$Th) ratios because of the preferential remobilization of U from the subducted slab (Gill & Williams, 1990; Hawkesworth et al., 1997), new magma batches will tend to plot below any isochron defined on the basis of the pre-existing crystals (Fig. 7). Thus, mixing associated with the injection of fresh magma will displace the resultant mixed magma to lower ($^{230}$Th/$^{232}$Th), and relatively low groundmass ($^{238}$U/$^{232}$Th) values may be evidence for magma replenishment processes during differentiation. Such low ($^{238}$U/$^{232}$Th) values are observed in the groundmass of one sample from Soufriere, but in general the groundmass data do not plot as far below as might be expected from a steady-state magma replenishment system (Hughes & Hawkesworth, 1999). This suggests that either magma infilling was relatively infrequent or the ability of new magma to mix with older magma was limited. The high eruption frequency of Soufriere, possibly >140 eruptions in 5000 years (Heath et al., 1998b), might imply frequent injections of new magmas if such injections act as eruption triggers (Sparks et al., 1977).

New $^{226}$Ra analyses on the Soufriere rocks yield ($^{226}$Ra/$^{230}$Th) values of up to ~2 (Chabaux et al., 1999; P. Evans, unpublished data, 1999). As $^{226}$Ra/$^{230}$Th returns to isotope equilibrium in ~8000 years, the presence of $^{226}$Ra excess suggests...
in rocks that preserve much older U–Th ages might indicate that the U–Th and Ra–Th isotopic systems are in some way decoupled. However, preliminary modelling, following Hughes & Hawkesworth (1999), shows that $^{230}$Th–$^{238}$U and $^{238}$Ra–$^{230}$Th disequilibria can be sustained for long periods of time provided that the system is frequently replenished with reasonable volumes of new magma on time scales of tens to hundreds of years. Such a system might well preserve old crystal ages but groundmass samples would not be expected to plot on old mineral isochrons.

Old U–Th mineral isochrons that yield ages of $39^{\pm 9}_{\pm 8}$ and $12^{\pm 5}_{\pm 5}$ ky have also been reported for cumulate and lava samples from the 1944 eruption of Vesuvius (Black et al., 1998b). However, such evidence for long magma residence times contrasts with results recently obtained by Zellmer et al. (2000) for Santorini in the Aegean Volcanic Arc, Greece. A whole-rock plagioclase–magnetite U–Th mineral isochron from a dacite that erupted $67^{\pm 9}$ ky ago ($^{40}$Ar–$^{36}$Ar) yielded an age of $85^{\pm 30}_{\pm 30}$ ka (1σ). Similarly, a U–Th mineral isochron from a dacite of a historic eruption yielded an age indistinguishable from zero years, and such young ages are in good agreement with those estimated from Ra-Th isotope data, and trace element profiles (see below). These rocks are also calc-alkaline, but they are more evolved than those from Soufriere (Heath et al., 1998a), and yet they provide no evidence for long magma residence times in the crust. The reasons for such differences are unclear, but they may relate to differences in the periodicity of magma injection into the crust, and perhaps to the average melt generation rates in the two areas. There have also been a number of studies of Ra–Th ages on mineral separates and they tend to yield ages close to, or just slightly older than the eruption age (Reagan et al., 1992; Schaefer et al., 1993; Evans, 1999).

In summary, unexpectedly old ages have been reported on crystals from a number of magmatic systems. It is too early to establish why old ages are preserved in some systems and not others, but those rocks with old mineral ages are relatively evolved (see Fig. 8 and the associated discussion). Often the minerals are not in bulk equilibrium with the whole-rock composition, and it remains difficult to establish the links between the analysed crystals and the age of the liquid. If the groundmass plots on a mineral isochron corresponding to an old age, it would appear that the liquid itself was also old (e.g. tens of thousands of years, Fig. 7). In general, old mineral ages provide direct evidence for the minimum life span of a particular magmatic system.

**Additional sources of information**

Additional information on rates of crystallization and crystal fractionation in magma chambers can potentially come from sources other than isotopic measurements. These include the extent of continuous normal zonation in phenocrysts, the characteristics of crystal size distributions, and progressive evolution through dated stratigraphic sequences. Furthermore, diffusive loss of initially sharp trace element profiles in phenocrysts offers a way of estimating the time spent at a particular temperature to produce the observed diffusive equilibration (e.g. Humler & Whitchurch, 1988; Zellmer et al., 1999). For example, if isotopic studies suggest that phenocrysts in a magma grew a long time before the magma was erupted (e.g. Halliday et al., 1989) then a lengthy period at high temperature should have degraded any trace element zonation in the phenocrysts. Moreover, such degradation of the initial trace element zonation is much less likely to occur if the crystals had been held in a mush or rock at lower temperatures until being mobilized by heating a short time before eruption. In rhyolites that give large discrepancies between crystallization and eruption ages, the temperatures involved are so low that diffusion is
particularly sluggish. This does have the advantage, how-

ever, of preserving trace element changes caused by the
magmatic processes (e.g. Blundy & Shimizu, 1991; Hervig &
Dunbar, 1992; Knesel et al., 1999) that affected these
evolved magmas.

Although kinetic effects during crystal growth may

complicate the interpretation of trace element profiles,
trace elements are generally very useful as at low con-
centrations their partitioning between crystal and melt is
governed by the partition coefficient $D$. Often the par-

tition coefficient is dependent on the major element
composition of the crystal and on magmatic temperature
only and thus can be determined accurately. For example,
the partition coefficient $D_{\text{Sr}}$ for Sr between plagioclase
and liquid is given by the empirical relation

$$RT \ln D_{\text{Sr}} = 26,800 - 26,700 X_{\text{An}}$$

(5)

where $R$ is the universal gas constant and $X_{\text{An}}$ is the mole
fraction of anorthite (Blundy & Wood, 1991).

Using this relation, the equilibrium partitioning of Sr
between two parts of a plagioclase crystal with variable
anorthite content can be determined at any given tem-
perature. However, if, for example, the Sr content of the
liquid changed during crystallization of the plagioclase
crystal, there will be an initial variation in Sr abundances
across the crystal reflecting the Sr contents of the liquid
and its distribution coefficient. Then, if magmatic tem-
peratures are sufficiently high, diffusive equilibration of
the trace element profile will occur such that the initial
Sr abundance profile equilibrates with time to form an
‘equilibrium concentration profile’ governed by variations
in the anorthite content of the crystal. As the diffusivity
of Sr in plagioclase can be estimated for different anorthite
contents and magmatic temperatures (Giletti & Casserly,
1994), it is possible to calculate the ‘age’ of an observed Sr
abundance profile that is not (yet) in diffusive equilibrium.

Assuming initial Sr concentration profiles with large
variations in Sr content across zoned plagioclase crystals,
Zellmer et al. (1999) used Sr diffusion systematics to
determine maximum residence times of selected plagioclase
crystals from recent dacites from the Kameni islands,
Santorini, in the Aegean Volcanic Arc. Although a num-
ber of crystals showed equilibrium concentration profiles,
preventing determination of maximum crystal residence
times, two crystals had not equilibrated (see Zellmer et al.,
1999, fig. 6). For these, maximum crystal residence
times of 100 years and 450 years, respectively, were
obtained. Similarly, a plagioclase crystal from a recent
andesite from Soufrière, St Vincent, in the Lesser Antilles
island arc, was shown to have grown <150 years ago.

In principle, further information can be obtained by
determining the variations in both Sr abundances and
Sr isotope ratios across feldspars. A number of studies
have constructed Sr isotope profiles across plagioclase
and sanidine crystals from selected dacites and rhyolites
using microdrilling techniques (Davidson & Tepley, 1997;

Knesel et al., 1999). These show that such crystals preserve
variations in $^{87}$Sr/$^{86}$Sr, and they have been used to
demonstrate that the magmatic system from which these
crystals grew was subject to both assimilation and sub-
sequent recharge with more mafic material. However,
microdrilling techniques involve sampling on a larger
scale than that for diffusion studies, and so the combined
application of Sr isotopes and trace element profiles
probably awaits improved laser ablation techniques for
Sr isotope measurements. Estimates of crystal residence
times could then come from studies of the different
degrees of diffusive equilibration of Sr isotope ratios
because of self-diffusion and of Sr concentration because
of (slower) chemical diffusion.

The time scales of crystallization, and hence the res-
idence times of crystal populations, can also be con-
strained from estimates of the numbers of crystals, their
size and morphology (e.g. Cashman, 1990; Marsh, 1996).
Many igneous rocks show a log-linear correlation of
crystal population density ($n$, in units of number of crystals
per volume of rock per crystal size) with crystal size
(Marsh, 1996). The total number of crystals ($N_\text{V}$) of a
specific phase is given [Marsh, 1996, equation (4)] by

$$N_\text{V} = n^2 G t_b = J t$$

(6)

where $G$ is crystal growth rate, $J$ is nucleation rate, $n^2$ is
nucleation population density at zero crystal size and $t_b$
is the characteristic time to complete crystallization.

Magmas reach the point of critical crystallinity, i.e. the
point beyond which eruption becomes impossible, at
$N \approx 50$–55% (Marsh, 1996). A characteristic time for a
single-phase system to reach 55% crystallization is given
[Marsh, 1996, equation (6)] by

$$t_b = 0.93 (JG)^{-1/4}$$

(7)

Using the estimates of $J$ and $G$ given by Marsh (1996)
suggests that magmas reach the point of critical crys-
tallinity in tens of years, and so CSD studies measure
time scales much shorter than is possible with most short-
lived isotope determinations. CSD analyses may also be
used to assess whether more than one crystal population
is present, as when megacrysts have been incorporated
from previous magma batches (Higgins, 1996a). CSD
analyses of plagioclase crystals from Kameni volcano,
Thera, Greece, and Egmont volcano in New Zealand
suggested crystal residence times of up to 100 years
(Higgins, 1996a, 1996b).

Thus the information from CSD studies (Higgins,
1996a, 1996b) and from trace element and Sr isotope
variations across crystals (Davidson & Tepley, 1997;
Tepley et al., 1999; Zellmer et al., 1999) indicates relatively
short crystal residence time scales of tens of years. These
are broadly consistent with results of a number of studies
of Ra–Th isotopes (Evans, 1999; Thomas, 1999; Zellmer
DISCUSSION

In principle, short-lived isotopes offer a powerful way to investigate the ‘ages’ of crystals and liquids at the time of eruption, and these may be tested against independent methods that rely on element diffusion profiles, or other geological information. In a chamber where crystals are forming throughout and settling towards the floor, U-series internal isochrons can potentially date the time of diagenesis. Alternatively, magma chambers may be surrounded by mushy solidification fronts (Marsh, 1996). At the low-viscosity end of such fronts (i.e. the end further from the wall), crystals and melt are free to move relative to each other and may be erupted as magma. Provided that the advancing front has not captured ‘tramp’ crystals and there has been no magma mixing within the front, such magma may be treated as an essentially closed system for U-series work. In all such discussions the key point is to establish independently the relationship between the analysed crystals and the host rock, either by testing for chemical equilibrium or normal crystal size distributions.

Figure 8 summarizes some of the available data on the apparent ages reported from recent volcanic rocks at the time of eruption, plotted against an index of differentiation, molar Si + Al. These are framework-forming elements in the liquid, and there is a marked increase in the viscosity of liquids at Si + Al ≈ 66 (e.g. McBirney, 1984). The ages illustrated in Fig. 8 are from separated phases, mostly with U–Th isotopes, and almost all the old pre-eruption ages are from rocks with relatively high Si + Al. Thus the simplest observation is the intuitive one that the likelihood of erupted magmas containing old crystals is much greater in the higher-viscosity liquids. Simple cooling and crystal settling models also imply that primitive basalts affected solely by cooling in a chamber will contain a small proportion of near-liquidus phenocrysts whereas more evolved magmas will have an increasing proportion of phenocrysts inherited from earlier stages of the magma’s history. In detail, it is difficult to evaluate the role of these ‘old’ crystals in the differentiation of the erupted magmas, not least because particularly in calc-alkaline rocks only the rims of phenocrysts may be in equilibrium with the host magma. Non-equilibrium minerals with old ages may be xenocrysts incorporated by the magma en route to the surface, and as such provide little insight into magma residence times or the time scales of magma differentiation. One possible exception is provided by the errorchrons reported by Heath et al. (1998b), as they include samples of groundmass and bulk rock, suggesting that in this case both the liquids and the separated crystals were tens of thousands of years old at the time of eruption.

The pattern of more old ages from the more evolved rock compositions is in sharp contrast to that emerging from studies of changing whole-rock U–Th–Ra isotope compositions with degree of differentiation (Fig. 8). Here differentiation from, for example, basanite to phonolite appears to take ~107 years (Reagan et al., 1992; Widom et al., 1992; Bourdon et al., 1994; Thomas, 1999), whereas differentiation to the more evolved phonolites, and particularly those involving alkali feldspar, typically occurred within 100–200 years of eruption (Reagan et al., 1992; Condonines et al., 1995; Thomas, 1999; Zellmer et al., 1999). The extent to which such inferred time scales are a feature of more alkaline magmatic systems compared with, for example, subduction-related calc-alkaline suites, is yet to be established. In the rocks from Tenerife (208Th/238U), decreases from 1.5 to 1.06 as the Zr abundances have increased by a factor of two (230–500 ppm), representing 50% fractional crystallization in 230 ± 70 ky, and from ~1.06 to 1.04 as the Zr contents increase to 1100 ppm, representing a further 25% fractional crystallization in less than a few thousand years (Fig. 5; Thomas, 1999). The available evidence suggests that these two idealized stages in differentiation took place at very different depths in the crust, and each represents ~50% fractional crystallization of the liquid that was present at the onset of each stage. Although the relative amounts of fractional crystallization were similar, their rates were higher in the more evolved magmas studied. This is readily explained by the cooling model described above and the geological inferences regarding the depths and sizes of the magma bodies in the two stages of evolution. Thus, a large volume of primitive magma cooling slowly at a deep crustal level will have a longer cooling time than a smaller body of differentiated magma cooling more rapidly in shallow crust. For example, 390 km3 of basalt cooling at 100 MW requires 100 ky to cool by 175 K and produce 50% crystals. In contrast, 88 km3 of intermediate magma cooling at 1000 MW requires just 2000 years to cool by 50 K and crystallize by 50%.

Finally, we note that over the last 20 years there has also been considerable interest in the roles of crustal
REFERENCES


contamination and magma mixing in magma differentiation. Pyle et al. (1988) presented \(^{143}Nd\)–\(^{144}Nd\) data for a basaltic andesite from Santorini, showing that the crystal population came from two sources that had mixed just before eruption, a mafic crystal mush and a dacite magma. The populations had similar crystallization ages of 79.4±1.0 ka and 93.2±2.2 ka, respectively. A tephra sample from the 1944 eruption of Vesuvius apparently records mixing of the two phenocrystal populations at 18.2±2 ka and 0.4±2 ka (Black et al., 1998b). Whereas the time scales of mixing are presumably short, those of contamination by assimilation of country rocks depend on the volume and temperature of the magma and the composition and temperature of the country rocks. None the less, Edwards & Russell (1998) suggested that assimilation-fractional crystallization may take place in weeks or years.

Keith Cox was a strong advocate of the importance of clear methodology, and he had a firm belief in the self-organizing ability of magmatic systems: after all, the mechanism of compensated crystal settling demands sophisticated controls of crystal behaviour. The new techniques for investigating time scales, and the emerging evidence from eruptive rocks of crystal mushes, in many cases mixtures of old and new crystals, present new challenges that would have intrigued him. One aspect that he might have highlighted is the need for a clearer understanding of the information available from the various chronometers that can now be applied to crystals and bulk rocks in magmatic systems.

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