

Modelling the petrogenesis of high Rb/Sr silicic magmas

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ABSTRACT

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Rhyolites can be highly evolved with Sr contents as low as 0.1 ppm and Rb/Sr > 2,000. In contrast, granite batholiths are commonly comprised of rocks with Rb/Sr < 10 and only rarely > 100. Mass-balance modelling of source compositions, differentiation and contamination using the trace-element geochemistry of granites are therefore commonly in error because of the failure to account for evolved differentiates that may have been erupted from the system. Rhyolitic magmas with very low Sr concentrations (≤ 1 ppm) cannot be explained by any partial melting models involving typical crustal source compositions. The only plausible mechanism for the production of such rhyolites is Rayleigh fractional crystallization involving substantial volumes of cumulates. A variety of methods for modelling the differentiation of magmas with extremely high Rb/Sr is discussed. In each case it is concluded that the bulk partition coefficients for Sr have to be large. In the simplest models, the bulk D^{Sr} of the most evolved types is modelled as > 50. Evidence from phenocryst/glass/whole-rock concentrations supports high Sr partition coefficients in feldspars from high silica rhyolites. However, the low modal abundance of plagioclase commonly observed in such rocks is difficult to reconcile with such simple fractionation models of the observed trace-element trends. In certain cases, this may be because the apparent trace-element trend defined by the suite of cogenetic rhyolites is the product of different batches of magma with separate differentiation histories accumulating in the magma chamber roof zone.

1. Introduction

It has been known for a long time (e.g. Nockolds and Mitchell, 1946) that most SiO₂-rich igneous rocks have much higher Rb/Sr than SiO₂-poor igneous rocks and that this ratio can therefore be used as an index of differentiation. This is obviously because Rb is more incompatible than Sr in nearly all magmatic systems. Accurate isotope dilution measurements are now illustrating the remarkable extent of this fractionation. Studies of rhyolitic suites from Yellowstone (Leeman and Phelps, 1981; Hildreth et al., 1991), La Primavera (Mahood and Halliday, 1988) and Long Valley (Halliday et al., 1989) have revealed highly

fractionated magmas with less than 1 ppm Sr (0.5 ppm at Yellowstone, 0.3 ppm at La Primavera and 0.1 ppm at Glass Mountain, Long Valley). In this paper we explain these differentiated magmas in terms of extreme Rayleigh fractionation and discuss the constraints placed thereby on models of rhyolite genesis.

2. Rb and Sr concentration trends

Rb and Sr concentration data for some highly fractionated silicic magmas are shown in Fig. 1 together with data for major granitic suites from around the world. The individual Caledonian granitic suites shown (from Donegal

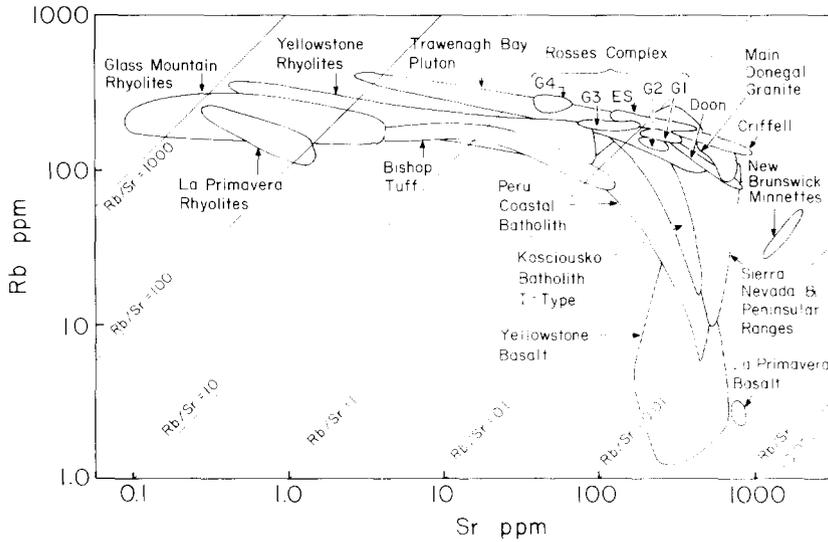


Fig. 1. Rb vs. Sr for rhyolites and associated basalts from western North America (Halliday et al., 1984, 1989; Mahood and Halliday, 1988; Hildreth et al., 1991), and granitic suites from around the world (Halliday, unpublished data on Donegal; Hine et al., 1978; Bachinski and Scott, 1979; Atherton et al., 1979; Bateman and Chappell, 1979; Halliday et al., 1980; DePaolo, 1981b). The data for Rosses (Donegal) are divided into separate phases *ES* (early sheets) and *G1* to *G4*. All Sr concentrations < 50 ppm determined by isotope dilution.

and S. Scotland) were selected specifically for their relatively simple quasi-linear trends when plotted in this manner. Certain key points can be made from examination of this diagram.

(1) In general, the rhyolite suites contain highly fractionated magmas not normally included in our granite sampling. There is therefore a need for detailed studies of the relationship between what are commonly seen as late stage differentiates (aplites and leucogranite minor intrusives) on the one hand and granite pluton geochemistry on the other. These minor intrusives are commonly regarded as esoteric and a minor aspect of the overall story. They are, in fact, possibly of fundamental importance to our understanding of the genesis of granite batholiths. Trying to mass balance crustal contamination, melt compositions and differentiation in granites using bulk geochemistry is of limited value without this information. Most large-volume high-level plutons may be accumulative relative to the once associated, now lost, cogenetic volcanics.

(2) In plagioclase-free differentiation in

which Rb and Sr are both incompatible, a positive slope should result in Fig. 1 (e.g., the New Brunswick minettes). These conditions would be met in the lower crust or upper mantle at pressure-temperature conditions exceeding the stability of plagioclase. In basaltic systems Sr is depleted (in response to plagioclase fractionation and/or crustal assimilation) as Rb increases, corresponding to a more or less vertical trend of data on Fig. 1 (e.g., the Yellowstone basalts). In systems in which Sr depletion is more marked (many granites and rhyolites, for example), a negative trend is anticipated in Fig. 1. A cogenetic suite of compositions evolving from mafic to felsic would therefore be expected to display some kind of an inverted "L" or recumbent "V" shape on this diagram, which is seldom (if ever) observed in perfection, but is possibly the reason for the less perfect inverted "L" shapes of the Peru Coastal Batholith and the Kosciusko Batholith I-types (Lachlan foldbelt). The trend in Fig. 1 will be horizontal if bulk $D^{Rb} \cong 1$ or if $D^{Sr} \gg D^{Rb}$. The former is only likely in assem-

blages fractionating large amounts of biotite (Hanson, 1978). Many evolved granites such as the Trawenagh Bay pluton, Donegal (Fig. 1) have clearly undergone extensive K-feldspar fractionation, as judged by their very high Rb/Sr, Rb/Ba, and Rb concentrations indicative of bulk $D^{Rb} < 1$, bulk $D^{Sr} > 1$ and bulk $D^{Ba} > 1$.

(3) The fact that quasi-linear negative trends are observed in Fig. 1 for "evolved" granites and rhyolites, suggests that there is some simple relationship between the change in Rb and Sr in the magma. It will be shown below that this is indeed explicable with a manipulation of the Rayleigh fractionation equation.

We can take several approaches to the question of how evolved silicic magmas such as those of Long Valley (with more than three orders of magnitude variation in Rb/Sr ratio) are produced. In so doing we are also attempting to place limited constraints on what kind of parent magmas were involved in the genesis of these volcanic rocks, although from the outset we would admit our prejudice that many of the parent magmas were complex mixtures of mantle- and crust-derived components with an overall intermediate to silicic composition (A.N. Halliday in Leake et al., 1980; Halliday et al., 1980; DePaolo, 1981a, b; Hildreth, 1981; Huppert and Sparks, 1988). Isotopic studies prove that some of the arrays shown in Fig. 1 are not simply the result of Rayleigh fractionation. Processes such as crustal contamination can also play a role. Note that contamination cannot be responsible for the very low observed Sr concentrations. In fact, exceedingly low Sr concentrations in a rhyolite are inconsistent with post-fractionation crustal contamination (Halliday et al., 1989).

In this paper we evaluate and place limits upon the potential importance of Rayleigh crystal fractionation. High Rb/Sr ratios in some alkaline rocks have been explained in terms of sub-solidus fluid-rock interaction (Bonin et al., 1979). However, this is of little relevance to the (mostly glassy) fresh volcanic

rocks with which this paper is mainly concerned.

3. Partial melting vs. fractional crystallization

Trace-element behavior as a result of silicate-melt equilibria is described in a number of familiar equations (e.g., Shaw, 1970; Arth, 1976). Partial melting is generally modelled by an equilibrium batch melting equation such as:

$$C^i/C_o^i = 1 / (F + D^i - FD^i) \tag{1}$$

whereas fractional crystallization is described by the Rayleigh equation:

$$(C^i/C_o^i) = F^{D^i-1} \tag{2}$$

(in which C^i = concentration of element i in the liquid, C_o^i = concentration of element i in the starting mineral-melt assemblage, F = fraction of liquid). If the bulk crystal/liquid distribution coefficient (D) is greater than unity, the trace element (i) is depleted much more rapidly during crystal fractionation than in melting (Fig. 2). Even assuming a high D^{Sr} , it is almost impossible to achieve low Sr concentrations (and correspondingly high Rb/Sr) through equilibrium partial melting. This is true whether modal or non-modal melting equations are considered (as F approaches zero, C^i/C_o^i approaches $1/D^i$). Large deple-

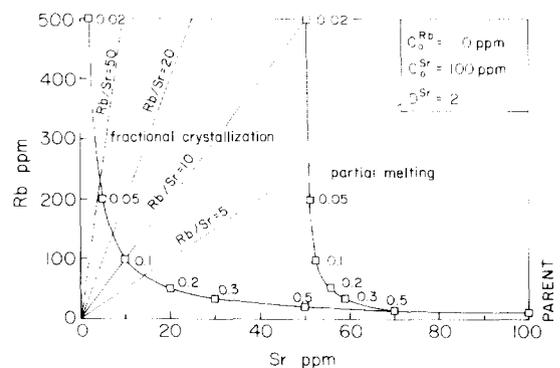


Fig. 2. Model Rb vs. Sr concentrations expected as a result of differing degrees of partial melting as opposed to Rayleigh fractional crystallization for the same parent composition.

tions in Sr could be achieved if an extreme fractional melting model were used, but the physical plausibility of separating infinitesimally small fractions of rhyolitic liquid from a crystalline source is at best questionable (Arzi, 1978; McKenzie, 1985; Wickham, 1987). We are then drawn to the conclusion that the only closed system model capable of generating the Rb–Sr characteristics of highly evolved rhyolites is fractional crystallization (Halliday, 1990). Furthermore, the existence of high Rb/Sr rhyolites can be taken as evidence for a significant crustal magma chamber at the time of eruption. Many precaldera rhyolites with low Sr concentrations are therefore likely to reflect the presence of a large magma system, rather than small pockets of unrelated melt (cf. Bacon et al., 1981; Metz and Mahood, 1985; Huppert and Sparks, 1988; Sparks et al., 1990).

4. Rayleigh fractional crystallization

To achieve three orders of magnitude variation in Sr concentration in a rhyolite system such as the Long Valley caldera or the basalt–rhyolite system of La Primavera by fractional crystallization alone reduces eq. 2 to:

$$D^{\text{Sr}} = (-3/\log F) + 1 \quad (3)$$

This requires a bulk D^{Sr} of 2.5 for an F of 0.01. This translates into thousands of cubic kilometers of cumulates for such systems (i.e., approximately 100 times the volume of erupted rhyolite). The calculated volume of cumulates would be reduced to hundreds of cubic kilometers if the bulk D was 4. The highest mineral/melt K_d that would be suitable from the available literature would be 3.87 for K-feldspar (Philpotts and Schnetzler, 1970) 4.5–7.3 for sanidine (Nash and Crecraft, 1985) and 5.57 for anorthoclase (Sun and Hanson, 1976). These values are not very encouraging in terms of increasing bulk D and reducing the calculated mass of cumulates. D^{Sr} will change during fractionation both due to changing mineral assemblage and as a result of changing equilib-

rium conditions between minerals and melts. For instance, the distribution coefficient for Sr between plagioclase and basaltic melt may be very much less than that between plagioclase and silicic melt (e.g., Korrington and Noble, 1971; Arth, 1976; Hanson, 1978). It has been shown that the mineral/melt distribution coefficients for most elements and minerals increase as the melt becomes more silicic and more polymerized and the concentration of the element becomes very low (Leeman and Phelps, 1981; Mahood and Hildreth, 1983; Watson, 1985). The effective distribution coefficient would therefore be more realistically represented by an integration of D^{Sr} over the range of F . Extremely high K_d -values for Sr in sanidine have been reported (Leeman and Phelps, 1981; Halliday et al., 1989), and below we present further evidence that high bulk D for Sr is required by a simple Rayleigh model.

5. Rayleigh fractionation and the determination of bulk distribution coefficients from Rb and Sr whole-rock concentrations

Another attempt to determine the bulk distribution coefficients and hence constrain a Rayleigh fractionation crystallization model comes from inverting the problem.

We can use the data shown in Fig. 1 to calculate bulk distribution coefficients for Rb and Sr as follows. By taking the logarithm of eq. 2 we obtain:

$$\log C^i - \log C^i_0 = (D^i - 1) \log F \quad (4)$$

We can therefore write:

$$\begin{aligned} \frac{\log \text{Rb} - \log \text{Rb}_0}{\log \text{Sr} - \log \text{Sr}_0} &= \text{slope on Fig. 1} \\ &= n = \frac{D^{\text{Rb}} - 1}{D^{\text{Sr}} - 1} \end{aligned} \quad (5)$$

In Fig. 3 we show an array of lines of different slope ($=n$) which follow from this relationship. The slopes that we observe in evolved granitic (rhyolitic) systems vary from about

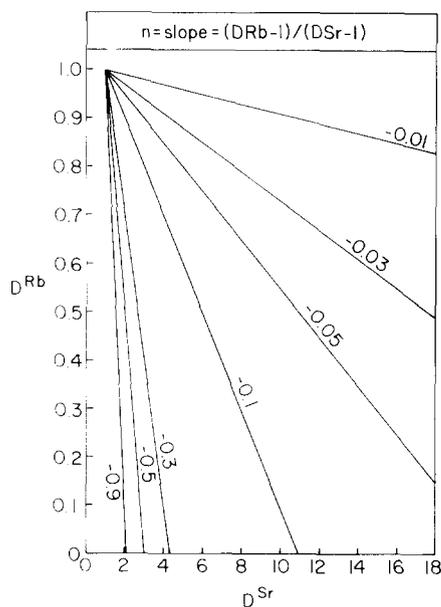


Fig. 3. D^{Rb} vs. D^{Sr} with slopes corresponding to $(D^{\text{Rb}} - 1)/(D^{\text{Sr}} - 1)$ (equivalent to those in Fig. 1).

–0.4 (high temperature Bishop Tuff, or La Primavera) to –0.2 (Trawenagh Bay pluton) to near zero (Glass Mountain, Long Valley). From Fig. 3 we can see that these shallow slopes require a bulk D for Sr that is about 2.5 or greater provided D^{Rb} is around 0.2. This value will not change greatly in silicic melts in which Rb is relatively concentrated (Hanson, 1978; Mahood and Hildreth, 1983; Watson, 1985). Since sanidine is almost certainly the most important phase to control bulk D in rhyolitic liquids, bulk D^{Rb} must be less than 0.5 (probably 0.1). Rhyolitic suites such as Glass Mountain with a very gentle negative slope on Fig. 1 can clearly only be produced by Rayleigh fractionation if bulk D^{Sr} is extremely high. For example, if $D^{\text{Rb}} < 0.5$, then $D^{\text{Sr}} > 50$! Such high values are virtually impossible on the basis of observed assemblages. In nearly all high silica rhyolites, sanidine and quartz greatly exceed plagioclase in modal abundance.

6. Rayleigh fractionation and the determination of bulk distribution coefficients from whole-rock/glass data

Knowing the concentrations of Rb and Sr in glass separated from non-vesicular whole-rock rhyolite, we can determine bulk distribution coefficients provided we know the proportions of crystalline phases. This approach does have the disadvantage that accurate modal proportions will be difficult to determine in crystal-poor rhyolites. A simpler approach is as follows. The concentration of an element in the bulk crystals can be expressed as:

$$C_{\text{min}} = (C_{\text{wr}} - C_{\text{gl}})M^{-1} + C_{\text{gl}} \quad (6)$$

where C = concentration, min = minerals, wr = whole rock, gl = glass and M is the mass fraction of crystals. From this it follows that:

$$D^{\text{Rb}} - 1 = [\text{Rb}_{\text{wr}} / (M \text{Rb}_{\text{gl}})] - (1/M) \quad (7)$$

and that:

$$\frac{D^{\text{Rb}} - 1}{D^{\text{Sr}} - 1} = \frac{(\text{Rb}_{\text{wr}} / \text{Rb}_{\text{gl}}) - 1}{(\text{Sr}_{\text{wr}} / \text{Sr}_{\text{gl}}) - 1} \quad (8)$$

which is the same as the slope (n) in Figs. 2 and 3 and eq. 5. Whole rock and glass concentrations of Rb and Sr have been measured by isotope dilution for two samples at Glass Mountain (Halliday et al., 1989) and for this we determine values of n of –0.0122 and –0.397 for glasses with 0.13 and 2.3 ppm Sr, respectively. The bulk D^{Sr} required by the first value of n (Fig. 3) is about 50, given $D^{\text{Rb}} < 0.5$, confirming the conclusion based on Rayleigh modelling of Fig. 1.

These indications of very high bulk distribution coefficients for Sr in highly evolved rhyolite systems can be supported in part by some direct measurements data on phenocrysts which is our final approach to the problem.

7. Sanidine/melt partition coefficients for Sr

In Fig. 4 we show the concentration ratios of Sr in sanidine relative to coexisting whole rock

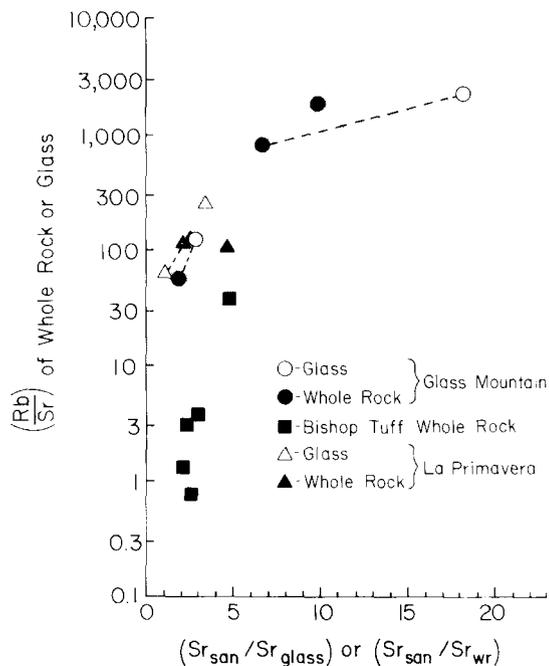


Fig. 4. $\text{Sr}_{\text{san}}/\text{Sr}_{\text{wr}}$ or $\text{Sr}_{\text{san}}/\text{Sr}_{\text{glass}}$ against differentiation index (Rb/Sr) (note log scale) for volcanics of the Long Valley caldera and La Primavera (Halliday et al., 1984, 1989; Mahood and Halliday, 1988).

or glass concentrations, as a function of Rb/Sr ratio of the whole rock or glass (a very suitable indicator of degree of fractionation). In an ideal situation, $\text{Sr}_{\text{san}}/\text{Sr}_{\text{glass}} = \text{mineral}/\text{melt } K_D$, whereas $\text{Sr}_{\text{san}}/\text{Sr}_{\text{wr}} < \text{mineral}/\text{melt } K_D$. Clearly, even minor amounts of crustal contamination of the rhyolitic liquid during eruption are likely to provide false apparent partition coefficients. In short, trying to accurately measure extremely low concentrations of Sr is feasible, but the data may be suspect despite this. However, nature works in our favor in so far as the effects observed (Halliday et al., 1984, 1989; Mahood and Halliday, 1988; Halliday and Bacon, unpublished) suggest preferential addition of Sr to the glass. We have no reason yet to suspect a high apparent K_D in these very evolved compositions as being due to alteration or contamination (i.e., loss of Sr in the glass or liquid).

Highly fractionated liquids do indeed appear to have high mineral/melt distribution

coefficients for Sr in sanidine (Fig. 4). Note the apparent K_D of 18 observed for one Glass Mountain sample. In order to produce bulk D_{Sr} of 50 as suggested by the other lines of evidence, we must also be fractionating plagioclase with even higher K_D . This does not seem unreasonable, although distribution coefficients determined from high precision isotope dilution measurements are needed to substantiate this. We note that Nash and Crecraft (1985) record K_D^{Sr} in plagioclases of 6.8 to 33.

8. Conclusions

(1) Rhyolites exhibit enormous fractionations in Rb/Sr ratios seldom reported from granites. We conclude that our rationale for sampling of granites as representatives of silicic magma compositions is probably flawed and that the evolved portions of the granite magma are lost from the system. It is therefore crucial to examine minor intrusives as clues to the more evolved liquid compositions. Furthermore, the notion that one should be able to mass balance the trace-element or major-element chemistry of granitic batholiths to determine the characteristics of the magma source is possibly erroneous.

(2) First order trace-element modelling indicates that the evolved compositions observed in high-silica rhyolites cannot be produced realistically by crustal melting alone; considerable differentiation of the magma is required.

(3) Several different approaches to the problem of generating high-silica rhyolites by fractional crystallization of a magma with low Rb/Sr indicate that it can only be achieved with reasonably high bulk distribution coefficients for Sr (> 3) and/or large volumes of cumulates. We do not see it as unreasonable to produce evolved rhyolitic magma by fractional crystallization of basalt on these trace-element grounds alone. In certain cases (such as alkaline rhyolite systems) this may be a better explanation than crustal melting, as dis-

cussed by Mahood and Halliday (1988).

(4) Attempts to determine the bulk distribution coefficients for Sr in highly evolved rhyolites yield similar information. The bulk distribution coefficients appear to be high (>2.5) to very high (as high as 50 in extremely evolved liquids). Bulk distribution coefficients this high are difficult to reconcile with observed phenocryst assemblages which are dominated by quartz and sanidine. A possible explanation for this is that the modelling is in error, because D_{Rb} is higher than generally appreciated in very evolved rhyolites. However, this is not supported by observed data. Another possible explanation is that the shallow sloping trends for data from evolved rhyolites shown in Fig. 1 are not the product of single-stage differentiation. Rather they are an artefact of combining a variety of highly evolved, genetically related, but separate differentiates, which accumulate near the roof of the same magma chamber.

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