

PHASE EQUILIBRIA CONSTRAINTS ON LIQUID LINES OF DESCENT GENERATED BY PAIRED ASSIMILATION AND FRACTIONAL CRYSTALLIZATION: TRACE ELEMENTS AND SR AND ND ISOTOPES

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Abstract. Utilizing a phase equilibria based trace element isotope model for low pressure igneous differentiation, it is now possible to update the methods presently being used to evaluate the effect of assimilation and fractional crystallization (AFC) processes. The results of the calculations presented here indicate that bulk partition coefficients (D) for Sr and Nd are strongly dependent on the chemistry of the assimilant and the rate of mixing. Virtually all silicic assimilants will increase bulk D for network-modifying cations because of the positive correlation of most trace element partition coefficients with reciprocal temperature, and the amount of network forming components in the melt. In addition to the effect of temperature and melt composition, the addition of a magma of different composition from the magma chamber will generally cause a change in the fractionating mineral proportions. For example, a peraluminous assimilant will increase the proportion of plagioclase, and a peralkaline assimilant will increase the proportion of augite. This will in turn have an effect on bulk D . For most multiply saturated mafic to intermediate systems differentiating at low pressure, bulk D_{Sr} is >1 , and bulk D_{Nd} is <1 . Thus the effect of a peraluminous assimilant is to drive bulk D_{Sr} away from 1 and a peralkaline assimilant drives bulk D_{Nd} toward 1. Therefore a peralkaline assimilant will generally drive the liquid path away from a simple mix-ing curve (or $D = 1$ AFC curve) and a peralkaline assimilant will drive the liquid path toward a simple mixing curve. Compared to the results derived in this study, existing models using fixed bulk D s calculated from closed system conditions will generate results that underestimate the assimilation rate, $^{87}/^{86}Sr$ and bulk Sr content for a peraluminous assimilant, and over-estimate the assimilation rate, $^{143}/^{144}Nd$ and Nd content of the assimilant for a peralkaline assimilant.

Introduction

Over the past decade, paired assimilation and fractional crystallization (AFC) has been cited with increasing regularity in models of the petrogenesis of igneous systems, particularly in continental and arc environments. Several investigators have derived equations designed to describe trace element and isotopic evolution in these systems [Taylor, 1980; Allegre and Minster, 1978; DePaolo, 1981]. These models make several important simplifying assumptions, the most critical of which is that all partition coefficients (D) are constant over the range of fractionation being modeled. In effect, this assumes that D is independent of temperature and composition. In addition, existing models assume that the fractionating mineral assemblage, from which the bulk D is calculated, is constant as a function of assimilation rate (r), the ratio of assimilant/ fractional crystallization. Finally, the fractionating mineral proportions are assumed to be either equal to the mineral proportions observed in closed system crystallization or to the mineral proportions calculated by least squares models. Some of these assumptions were known to

be at least partly false; however, they were necessary for any predictions to be made.

Recent development of phase equilibria based differentiation models [Ghiorso and Keleman, 1987; Nielsen, 1988] have enabled us to build more complex, realistic models of AFC processes and to refine previously reported predictions. The goal of this paper is to calculate liquid lines of descent for Sr and Nd isotopes and to evaluate the significance of the effects of assimilation on temperature, fractionating mineral proportion, and melt composition.

Model Description

To perform the following calculations, a variant of the program TRACE.FOR [Nielsen, 1988] was used. This model uses a set of single-component mineral melt expressions to calculate the equilibrium temperatures and compositions of minerals for a given natural mafic or intermediate silicate magma. Igneous processes are simulated by the addition or subtraction of components from the system according to a set of input model parameters. For example, in order to model paired assimilation and fractional crystallization, an increment of the liquidus phase, the phase with the highest calculated equilibrium temperature, is removed from the system. An increment of an assimilant is then added to the system. The liquidus phase for this derived magma is calculated, and the procedure is repeated. The ratio of the mass of the assimilant increment size to that of the increment of fractionated liquidus phase is equivalent to (r), the assimilation factor of DePaolo [1981].

Trace elements and Nd and Sr isotopic trends are modeled by approximately the same procedure. As each increment of the liquidus phase is removed, and assimilant added, the trace element and isotopic composition of the liquid are adjusted. Therefore the differentiation paths for major elements, trace elements, and isotopic composition are modeled as a large number of interdependent mass balance calculations. This allows the model conditions, such as temperature and phase composition to change continuously as differentiation proceeds.

The model presented here differs from earlier models in that it is numerical with continuously varying parameters as opposed to analytical with constant parameters. Temperature, trace element partition coefficients, phase proportions, and compositions are calculated within the model, not set externally. The variable model parameters are assimilation rate, assimilant composition, starting composition, oxygen fugacity, equilibrium or fractional crystallization, recharge rate, eruption rate, and crystallization increment size. The assimilant is assumed to be added to the evolving magma as a liquid. There are no restrictions on the value of the assimilation rate or the composition of the assimilant. This assumption implies that there is no coupling of the heat generated by crystallization of the magma to the heat required to produce a liquid assimilant. Thus assimilation is modeled as mixing between a fractionating mafic magma and small increments of liquid assimilant.

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Results

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Previous calculations made using earlier versions of this model [Nielsen, 1988] have shown that the mineral propor-

Table 1. Starting and Assimilant Compositions

	Olivine Tholeiite*	High Al Basalt(HAB)†	Peralum. Granite‡	Metalum. Granite‡	Peralkaline Granite§
SiO ₂	49.76	50.58	73.79	75.11	74.00
TiO ₂	1.96	1.22	0.24	0.10	0.21
Al ₂ O ₃	13.43	17.69	13.40	13.17	11.59
FeO	11.27	8.87	1.99	1.36	3.13
MnO	0.23	.21	0.05	0.05	0.08
MgO	10.01	7.31	0.74	0.35	0.36
CaO	10.18	10.05	1.26	1.31	0.36
Na ₂ O	2.35	2.92	2.61	3.48	5.35
K ₂ O	0.62	0.82	4.53	3.97	4.46
P ₂ O ₅	0.13	0.17	0.12	0.05	0.02
Sr	400	400	100,400	400	100,400
Nd	20	20	20	20	20

*Data from Helz [1987].

†Data from Grove et al. [1982].

‡Data from Hine et al. [1978].

§Data from Macdonald [1975].

tions fractionating from an open magma system are dependent upon the composition of the magma composition being added. For example, assimilation of an aluminous granite by a multiply saturated mafic system increases the proportion of plagioclase that will crystallize over a given crystallization interval. Since bulk D is calculated from the fractionating mineral proportions, it is sensitive to any change in those proportions. This is particularly true for elements such as Sr, Sc, and the REE, which are partitioned heavily into a specific mineral in basalts and andesites. In dry mafic and intermediate systems, Sr is preferentially partitioned into plagioclase and Nd into augite. These elements are also of particular interest because of their role as isotopic tracers. Knowledge of the effect of a particular assimilant and the assimilation rate on the fractionating mineral proportions is therefore needed for the development of any realistic models of AFC processes.

To evaluate the potential effects of an assimilant on AFC-generated liquid paths, I have selected two basaltic starting compositions and three assimilant compositions (Table 1). The starting compositions are (1) a high-alumina basalt (HAB) with plagioclase as the liquidus phase followed by olivine, then augite, and (2) an olivine tholeiite with olivine on the liquidus followed by augite, then plagioclase. The assimilants include a range of granite compositions from peraluminous to metaluminous to peralkaline (Table 1).

Addition of a peraluminous granitic magma to an evolving mafic magma system has the effect of moving the liquid composition toward saturation in high-Al phases such as plagioclase. In general, if a magma is fractionating along the olivine-plagioclase cotectic, the addition of a peraluminous granite will move the magma into the plagioclase saturation field. Continued fractionation will drive the magma back toward the cotectic. The higher the assimilation rate, the more time the magma will spend in the plagioclase saturation field and the higher the fractionating proportion of plagioclase.

For HAB the addition of a peraluminous magma increases both the proportion of plagioclase and the period where only plagioclase is fractionating (Figure 1). The proportion of augite steadily drops as the assimilation rate increases, crystallizing pigeonite or orthopyroxene instead. At an assimilation rate of 2.0, the system never encounters augite saturation. These assimilation rates are high compared to the rate permissible if one requires heat balance to be maintained. They are, however, well within the range of cited values for natural systems [Grove and Kinzler, 1986]. In addition, they are justifiable if one assumes that the chemical and thermal

systems of igneous bodies are decoupled. In effect, that melt can be produced from the country rock without it immediately mixing with the residual melts of the system which provided the heat. This is a complex and interesting problem, but beyond the scope of this paper.

Addition of a peralkaline magma suppresses the proportion of plagioclase and increases that of augite (Figure 1). The reason for an increase in the stability of augite caused by the assimilation of peralkaline magma is less obvious than the increase in plagioclase proportion caused by the assimilation of a peraluminous magma. However, addition of alkalis will increase the complexing of Al, decreasing the normative anorthite content of the melt. A natural analog can be found in alkali basalts, where augite is the dominant pyroxene. The effects of a variety of other assimilant compositions are presented by Nielsen [1988].

The effect of the assimilants on the olivine tholeiite phase proportions is similar to that on HAB but modified by the fact that olivine rather than plagioclase is the liquidus phase. Nevertheless, the proportion of plagioclase increases for the peraluminous assimilant and augite increases for the peralkaline assimilant. The other major difference in the trends is due to the greater stability of augite in this olivine tholeiite system.

Variation in fractionating mineral proportions has a first-order effect upon the calculated bulk D. Added to this are the effects of the dependence of D on temperature and melt composition noted above. The addition of a granitic melt to a differentiating mafic magma chamber simultaneously decreases the liquidus temperature of the system and increases the amount of network-forming components in the melt. Because both Sr and Nd are modeled as network modifiers [Nielsen, 1985, 1988], and D_{Sr} and D_{Nd} for plagioclase and augite increase with decreasing temperature, the addition of a granitic assimilant increases D for both elements in most mafic systems. For scenarios such as a peraluminous granitic assimilant, all three, increasing plagioclase proportion, increasing network modifiers, and decreased temperature, reinforce one another, resulting in a rapid rise in bulk D_{Sr} . The same is true for Nd and a peralkaline assimilant. An important point to note is that peraluminous and peralkaline assimilants have opposite

The calculated trends in bulk D as a function of composition, assimilation rate, and percent fractionation (Figure 2) have several important characteristics. The bulk D for Sr for both the HAB and olivine tholeiite is near 1 when crystallizing along the olivine, plagioclase, and augite cotectic for closed system crystallization. Remember that at $D=1$,

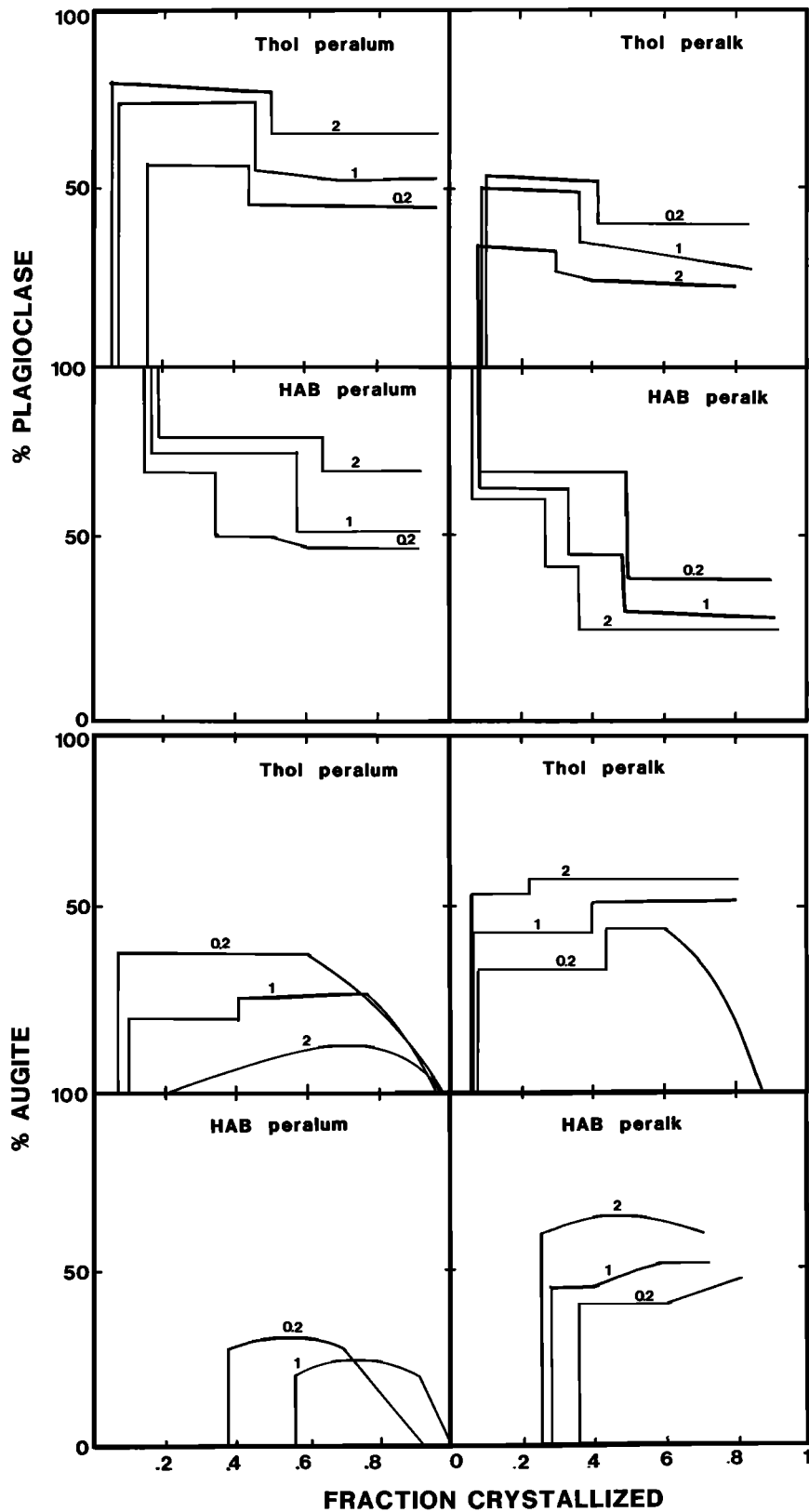


Fig. 1. Calculated fractionating mineral proportions versus percent fractional crystallization for AFC models involving olivine tholeiite and high Al basalt (HAB) starting compositions, and a peraluminous and peralkaline assimilant. Labels on each curve represent r , the assimilation/fractional crystallization ratio.

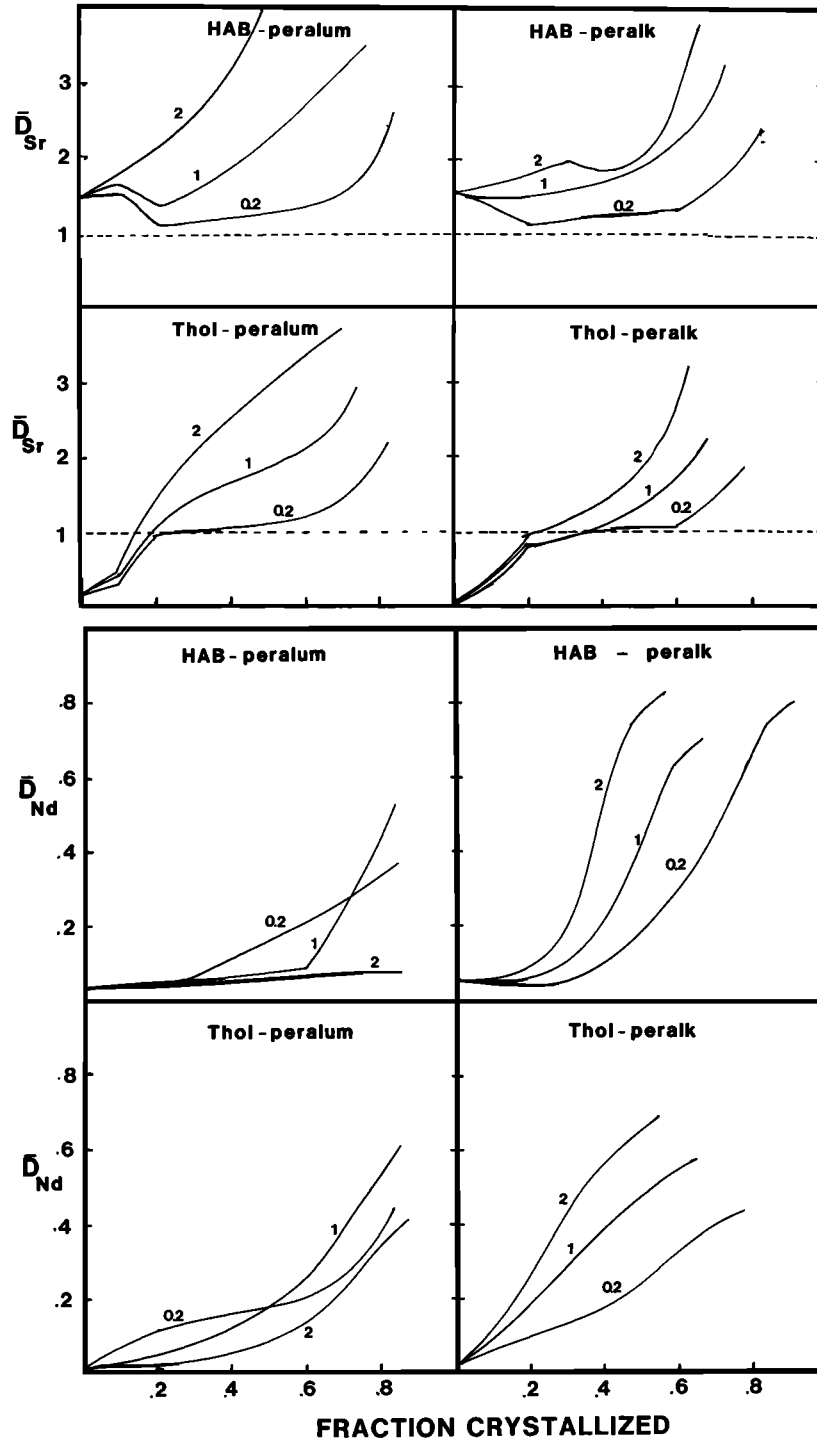


Fig. 2. Bulk D versus percent fractional crystallization. Calculated from mineral proportions given in Figure 1 and mineral D_s calculated from compositionally independent, temperature dependent functions in ISOTOPE.FOR. Note that a peraluminous assimilant drives D_{Sr} away from 1 and peralkaline assimilant drives D_{Nd} toward 1.

AFC-generated curves are identical to the simple binary mixing lines. The peralkaline assimilant has relatively little effect on D_{Sr} in this case except at high-percent fractional crystallization. This is due to the competing effects of reduced plagioclase proportion versus melt composition and temperature dependence of D_{Sr} -plag. The effect of increased r for a peraluminous assimilant has a dramatic effect on D_{Sr} , raising it by a factor of 2-5 compared to the low assimilation

rate scenarios. In all scenarios, bulk D for Sr remains above 1 after plagioclase saturation has been reached, higher than bulk D for closed system crystallization.

The effect of the peralkaline assimilant on D_{Nd} is as great as the effect of the peraluminous assimilant on D_{Sr} . At any individual percent fractionation, D_{Nd} varies by up to a factor of 5-8 (Figure 2). All calculated values, however, are below 1. The suppression of augite by the assimilation of the

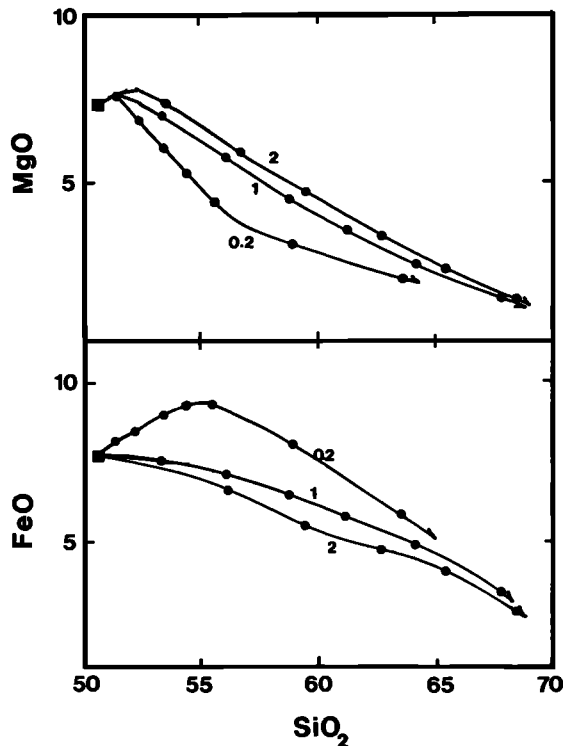


Fig. 3. Calculated liquid paths for the differentiation of a HAB, simulating simultaneous fractional crystallization and assimilation of a peraluminous granitic magma.

peraluminous granite observed in the calculated mineral proportions (Figure 1) is apparent in the calculated bulk D_{Nd} . The result is a relatively narrow range of values for D_{Nd} , rising only at high percent fractionation where the temperature and compositional dependence of D_{Nd} outpace the drop in augite proportion. In the extreme case of a recharge rate of 2 for the HAB, D_{Nd} remains low because augite saturation is never reached.

It is important to note that by the point that these systems have fractionated 60-80 percent, they have evolved into dacite magmas (65-72% SiO_2 , Figure 3). This is important for two reasons, first, because this model is not designed to model the evolution of silicic magmas ($SiO_2 > 68\%$) and second, because minor phases often become increasingly important in this compositional range. In addition, it shows that if the assimilant is granitic, a significant compositional change should accompany an increase in isotopic content.

The effects of the change in bulk D on calculated liquid lines of descent as a function of assimilant and assimilation rate are presented in Figures 4 and 5. The first example of the effects of AFC processes versus simple mixing is the case of a low Sr content (100 ppm), high $^{87}Sr/^{86}Sr$ assimilant and a high Sr content (400 ppm), low $^{87}Sr/^{86}Sr$ mafic starting composition. Figure 4 illustrates the results of the four possible iterations of the two simple assimilants and starting compositions. For the HAB and peraluminous magma scenario, the most notable characteristic is that the trend is toward low Sr contents and relatively low $^{87}Sr/^{86}Sr$ for all cases. This is due to the increase in D_{Sr} with increasing r . As D_{Sr} increases, Sr drops more rapidly than would be predicted using a fixed D_{Sr} , pushing the trends to lower Sr contents at a given $^{87}Sr/^{86}Sr$. The net result is that the trends are virtually superimposed on one another. This implies that for these sets of conditions this system is a poor method for the evaluation of assimilation rate.

For a peralkaline assimilant the calculated liquid lines do not drop to low Sr contents because D_{Sr} is not as high. By the point that the system has dropped to 200 ppm Sr, the bulk composition of the system is nearly granitic. The paths are subparallel and lead to low Sr contents largely because of the relatively high value for D_{Sr} caused by its temperature and compositional dependence.

The paths generated by the differentiation of the olivine tholeiite show the same pattern with a few differences in detail. The most important is the increase in Sr calculated for the low assimilation rate scenarios. This is caused by the initial dominance of olivine in the crystallization sequence. Over 50% crystallization is required before the Sr content of the derived liquids drops below 400 ppm. The increase in Sr is not a major characteristic of the liquid lines at higher assimilation rates because the assimilant drives the liquid to the plagioclase cotectic soon after crystallization begins.

If the concentration of Sr in the assimilant is increased to that in the starting composition, the results are considerably different (Figure 4b). The increased effectiveness of the assimilant at altering the liquid path can be seen by the shift in the paths toward the assimilant as r increases. Nevertheless, the increase in D_{Sr} caused by increased plagioclase, network formers, and decreased temperature causes the liquid paths to curve away from the simple mixing line.

The presence of olivine on the liquidus produces more complex paths for the olivine tholeiite scenarios. The dominance of olivine causes an initial rise in Sr, followed by a drop as plagioclase becomes dominant. However, the systems have fractionated 30-60% before the simple mixing line is crossed. The effect of the peralkaline assimilant is to keep the paths at higher Sr values for more of their evolutionary history compared to the peraluminous assimilant. The most important characteristic illustrated in these plots is that the paths overlap considerably, again making the evaluation of assimilation rate difficult. In none of the scenarios calculated so far does the liquid trend point toward the assimilant composition. Thus neither the assimilation rate nor the characteristics of the assimilant can be readily determined from information from this system.

The effect of variation in bulk D for Sr as a function of assimilant composition at a constant assimilation rate is illustrated in Figure 5. Liquid lines of descent were calculated for four scenarios with r held constant at 1. The starting composition was the olivine tholeiite given in Table 1. Two of the scenarios are the peraluminous and peralkaline assimilant scenarios presented in Figure 4b. The third scenario is for the metaluminous assimilant (Table 1). This assimilant has the effect of increasing the proportion of plagioclase fractionating from the system but not to the same extent as the peraluminous composition. Predictably, the result is intermediate between the peraluminous and peralkaline lines of descent.

The fourth scenario uses the bulk D calculated for closed system crystallization for the same olivine tholeiite. Inflection points in the curve represent the change in the fractionating mineral assemblage calculated by the phase equilibria model. All the scenarios using variable bulk D exhibit lower Sr contents at a given percent fractionation owing to the higher D_{Sr} caused by the effects of temperature, melt composition, and mineral proportions cited above. Thus the use of a "typical" D_{Sr} calculated on the basis of closed system mineral proportions will generate liquid lines of descent that are higher in Sr compared to those calculated using a phase equilibria based model.

For the scenarios detailed above for Sr the addition of an assimilant had the effect of increasing D_{Sr} away from 1, particularly for a peraluminous assimilant. With increasing r the liquid paths are driven away from the simple mixing line by this effect. The case for Nd is exactly the opposite. As assimilation rate increases, D_{Nd} increases toward a value of 1 if the assimilant is peralkaline. This reinforces the effect of

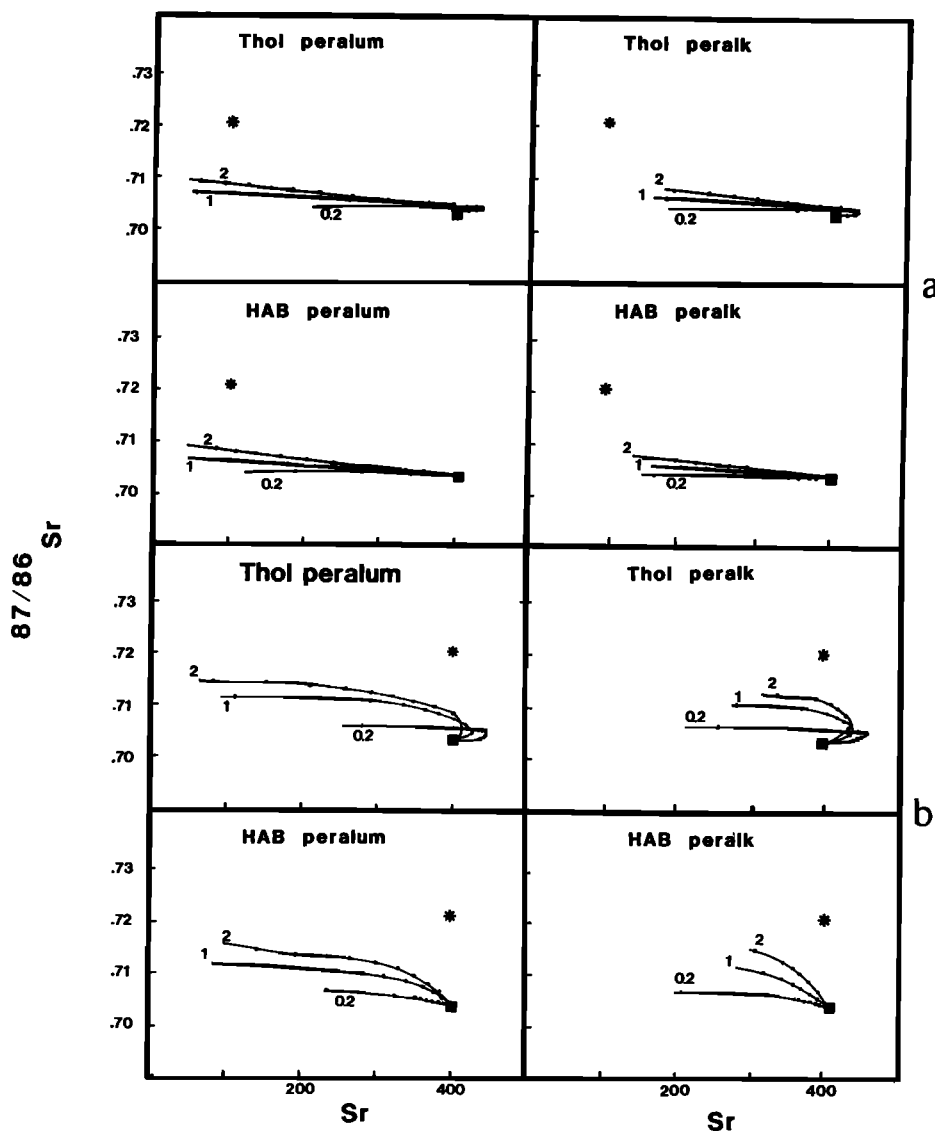


Fig. 4. (a) Calculated $^{87/86}\text{Sr}$ versus Sr (ppm) curves for olivine tholeiite and HAB with peralkaline and peraluminous assimilants. Assimilant (asterisk) contains 100 ppm Sr and $^{87/86}\text{Sr}$ of 0.72. Labels on curves represent r . Dots on curves are spaced at intervals of 10% fractional crystallization. (b) Calculated $^{87/86}\text{Sr}$ versus Sr paths for same scenarios as Figure 4a. Assimilant (asterisk) contains 400 ppm Sr.

mixing, and the liquid paths (Figure 6) converge on the simple mixing line faster than would be predicted using fixed D_s . There is little difference between the trends for a peralkaline versus a peraluminous assimilant at low r . As r increases, the increase in D_{Nd} for the peralkaline assimilant is evident in the proximity of the path to the simple mixing line.

If the effect of assimilant composition on bulk D and the fractionating mineral proportions is an important problem, the evidence should occur in the cumulate sequences of large mafic to intermediate magma systems. The mineral proportions in the cumulates should reflect the proportions of the phases that were removed from the system, modified by the effect of the process of separation (settling, reequilibration, in situ fractionation). This is not a simple question; however, we may be able to clarify the importance of these phase equilibria constraints if we begin to look at the petrographic and field information from cumulate sequences with this in mind.

Periodic Mixing

In natural systems it may be more likely that mixing of mafic and silicic components takes place periodically, separated by periods of fractionation. To address this point, I have made several calculations simulating this process. For these simulations I used the olivine tholeiite, the peraluminous assimilant, and an r of 1. The results (Figure 7) show that the general patterns seen in the continuous assimilation model are preserved. This is because the major factors responsible for the trends are still in effect, the increase in D_{Sr} caused by drop in temperature and the increase in plagioclase proportion and network-forming components versus closed system fractionation. If mixing and sampling occur at more irregular intervals, the resultant pattern of compositions will exhibit the scatter characteristic of natural suites [Nielsen, 1988].

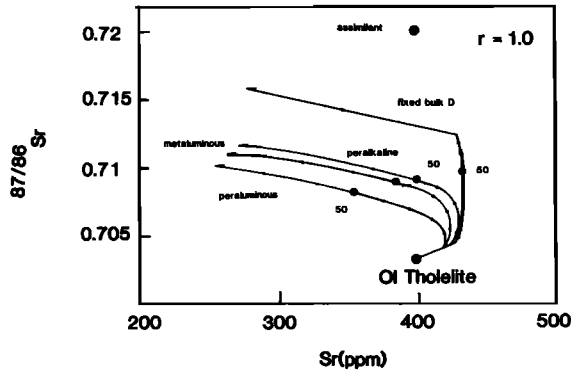


Fig. 5. Calculated $^{87/86}\text{Sr}$ versus Sr paths for peralkaline, metaluminous, and peraluminous granitic assimilants. Assimilation rate (r) equals 1. Fixed D calculations use the bulk D_{Sr} calculated from ISOTOPE.FOR assuming closed system conditions: 0-7% - $D_{\text{Sr}} = 0$; 7-40% - $D_{\text{Sr}} = 0.9$; 40-80% - $D_{\text{Sr}} = 1.0$; 80-90% - $D_{\text{Sr}} = 1.3$; 90+ - $D = 2.5$.

Speculations on the Effects of Increased P and Volatiles

The model used here is designed to model only low-pressure, anhydrous phase equilibria. Obviously, this is not appropriate for many systems. The production of a reliable model for high-pressure phase equilibria awaits the data base of mineral melt experiments necessary to derive the required expressions. Nonetheless, there are several points that can be made about the extrapolation of these results to middle and lower crustal pressures. The most important is the suppression of plagioclase caused by the increase in water and pressure. This would have the effect of decreasing D_{Sr} and of postponing the appearance of plagioclase. However, the basic conclusions still stand. As a peraluminous

assimilant is added to an evolving mafic magma, the proportion of plagioclase fractionating from that system will increase.

The effect of increased pressure on Nd would be opposite to the effects on Sr. Increased pressure would increase the role of augite. Addition of a peralkaline assimilant would increase the fractionating mineral proportions even more. The effect of a peraluminous assimilant, however, may stabilize orthopyroxene rather than augite, as it does at low pressure (Figure 1). In this case, D_{Nd} would drop with increased r . The most important consequence of increased pressure and water would be the increased stability of amphibole in the place of augite. The effect of this is at present impossible to predict quantitatively.

Conclusions

Development of models that combine phase equilibria with the simulation of trace element and isotope systematics provides the means of updating the conclusions of existing models on the results of AFC processes. The major conclusions of this work are based upon a set of simulations of AFC-generated liquid paths. The results indicate that isotope ratio trace element paths (e.g., $^{87/86}\text{Sr}$ versus Sr) are dependent upon the nature of the assimilant. A peraluminous assimilant will increase bulk D_{Sr} owing to an increase in the proportion of plagioclase fractionating from the system. A peralkaline assimilant will increase D_{Nd} owing to the increase in the proportion of augite. An increase in the assimilation rate, r , for a peraluminous assimilant will increase D_{Sr} from ~ 1 and keep D_{Nd} relatively low, driving the liquid paths away from the simple mixing curve. This in effect buffers the effect of increased assimilation rate. Thus the $^{87/86}\text{Sr}$ versus Sr ratio is not very sensitive to the assimilation rate if the assimilant is peraluminous, especially if the Sr content of the assimilant is low.

An increase in r for a peralkaline assimilant increases D_{Nd} from near 0 and keeps D_{Sr} relatively low by increasing the

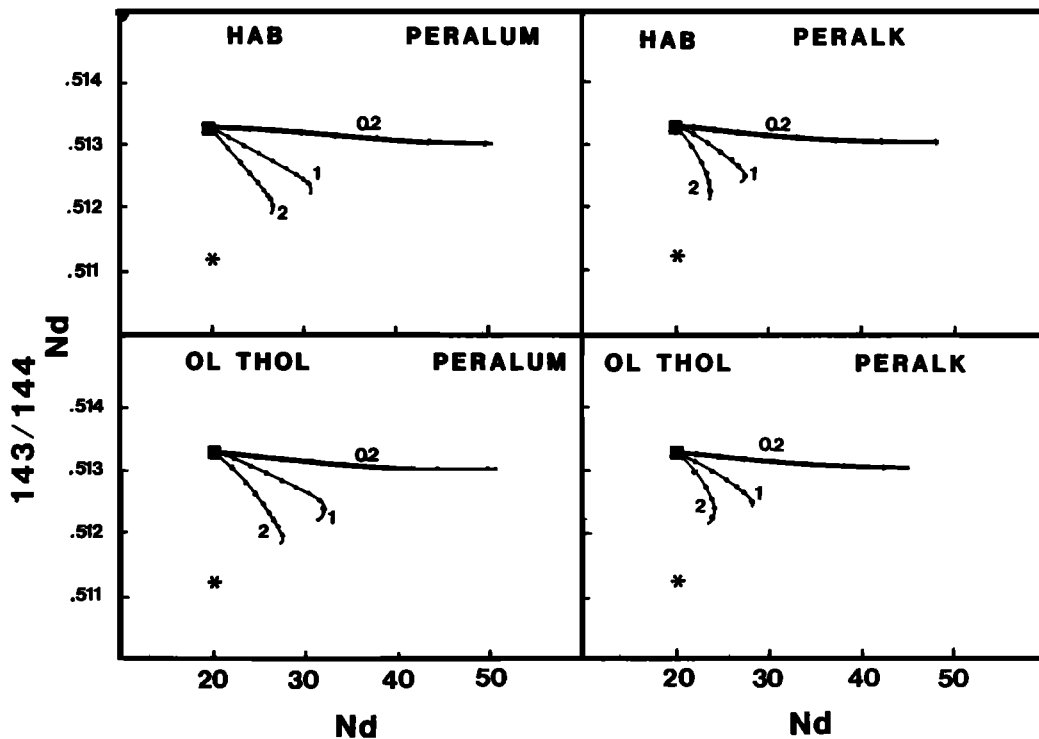


Fig. 6. Calculated $^{143/144}\text{Nd}$ versus Nd (ppm). Labels and compositions are the same as Figure 4.

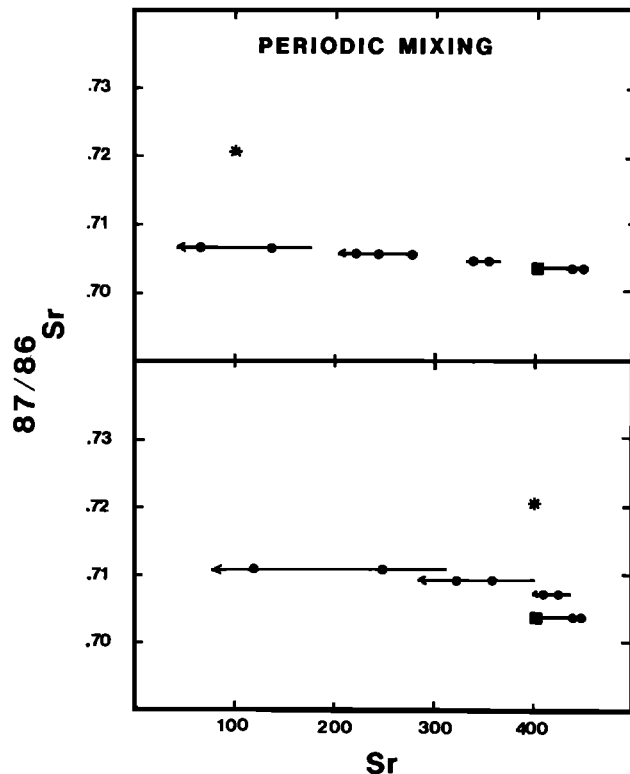


Fig. 7. Calculated $^{87}/^{86}\text{Sr}$ versus Sr (ppm) for case of olivine tholeiite and peraluminous assimilant, $r = 1$. Assimilation is periodic, with the system fractionating 25% between episodes of mixing.

role of augite versus plagioclase fractionation. This causes the liquid paths to approach the simple mixing curve as assimilation increases.

The addition of a granitic magma to an evolving mafic to intermediate magma system will cause the temperature of the system to drop and add significant amounts of network-forming components to the system. Both of these will have the effect of increasing D for Sr and Nd for plagioclase and augite. Therefore, for most scenarios, bulk D will rise regardless of the changes in fractionating mineral

These results indicate that trends calculated using fixed D s calculated from closed system fractionating mineral proportions could lead to the underestimation of the assimilation rate or the $^{87}/^{86}\text{Sr}$ and Sr content of the assimilant if the assimilant were peraluminous. If the assimilant were peralkaline, results calculated for Nd systematics would lead to the overestimation of $^{143}/^{144}\text{Nd}$ or Nd content of the assimilant or the assimilation rate.

One of the most important conclusions of this study is that even though a new generation of phase equilibria based models are now becoming available, it remains our responsibility to consult the field, petrographic, and chemical data to decide what model parameters are and are not geologically reasonable.

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