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Magma transport and metasomatism in the mantle: A critical review of current geochemical models—Reply to Navon et al.

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ABSTRACT

Navon et al. (1996) demonstrated that the Navon and Stolper (1987) model can be formulated to reproduce a pattern of light-ion lithophile trace element (LIL) enrichments produced by a single, small-scale metasomatic process recorded in a composite xenolith from Dish Hill, California (Nielson et al. 1993). The Navon and Stolper model has failed repeatedly to reproduce the shape and lateral positions of LIL enrichment patterns for samples from peridotite massifs, which are of appropriate scale to test the assumption that LIL fractionation takes place in percolating melts over distances >100 m. The model results also produce unreasonably long times for solidification of thin dikes, which imply untenable thermal conditions for lithospheric mantle.

Using parameters drawn from sample compositions, Nielson et al. (1993) demonstrated, and the calculations of Navon et al. (1996) have shown again, that fractionated trace element patterns of a melt are imprinted upon relatively refractory peridotite matrix in zones closest to a melt source. The observed process sequentially extracts LIL into matrix, analogous to the ion-exchange chromatography of water-purification columns. We have never contended that this process is mathematically distinct from the percolation model of Navon and Stolper (1987), which assumes concentration of LIL elements in melt. The choice of parameters defines the result, and one would notice a major difference in the taste of water from an ion-exchange column that traps target ions in matrix compared with one that concentrates those ions in the liquid.

The difference between the models is in the selection of parameters and values: The model of Navon and Stolper (1987) assumes the reaction mechanism, uses theoretical melt compositions, and contains as many as nine unmeasurable parameters. We used the simplified model calculation to avoid reliance on theoretical parameters and to test our assumptions about the process. When the compositions of actual samples are taken as end-members of mantle reactions, the successful results imply that a fractionation-by-percolation process is not applicable to lithospheric mantle. Repetition of the observed small-scale reaction in refractory peridotite must extend the zone of reactions and relative enrichment, centimeter by centimeter, as long as melt aliquots percolate beyond peridotite matrix that had previously reacted to equilibrium with the melt composition. This process satisfactorily explains the wide variations of LIL fractionation patterns over short distances that characterize mantle rocks in xenoliths and massifs, all of which contain complex systems of mafic intrusions with varied LIL fractionation patterns.

INTRODUCTION

We thank Navon et al. (1996) for the opportunity to debate our different approaches to numerical models of magma transport and interactions in the mantle, a debate that incorporates elements of a broader debate about model testing and validation (Bredehoeft and Konikow 1993; Nordstrom 1993; Oreskes et al. 1994). The debates about both mantle and hydrologic models (which are used to assess sites for hazardous waste disposal, for example; Davis et al. 1991) focus on the means of long-distance fluid transport, whether by percolation or in channels (fracture flow). Whatever the social implications, good science requires "the demonstration of agreement between observation and prediction" (Oreskes et al. 1994).

Variable enrichments of light-ion lithophile trace elements (LIL), generally defined by chondrite-normalized abundances of light rare earth elements ($LREE_{cn}$), are common in otherwise refractory peridotite of massifs and xenoliths. Nielson et al. (1993) used simplified versions of ion-exchange chromatography processes to model compositional variations in a composite peridotite-hornblendite xenolith from Dish Hill, California. We used the models to test the hypothesis that the reaction end-members were similar to the compositions of the hornblendite selvage at one margin of the sample and the most refrac-

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tory zone of peridotite at the opposite end. The hornblendite selvage represents the contact between peridotite wall rock and a mafic intrusion (variously termed a dike, dikelet, vein, or veinlet). The success of our various models supported that hypothesis, and the Navon et al. (1996) results provide additional support. We contend that hypothesis testing is the proper use of mantle-process models.

The Navon and Stolper (1987) model is founded on the assumption that LIL fractionation in the mantle occurs by disequilibrium percolation of melts over distances >100 m. We do not know of any mantle rocks derived from subcontinental mantle that support this a priori assumption. Instead, both massifs and xenoliths show levels of enrichment and shapes of chondrite-normalized patterns that vary considerably over short distances. Well-documented, melt-peridotite wall-rock reactions in both xenoliths and massif samples are also of small scale, and compositional trends of mafic mantle intrusions show that melts undergo differentiation within mantle conduits (Conquéré 1977; Irving 1980; Bodinier et al. 1987; Nielson et al. 1990). These observations support the origin of LIL enrichment in otherwise refractory peridotite from local, small-scale interactions between variably differentiated melts and depleted peridotite wall rock.

Massif samples are of appropriate scale and contain enough lithologic context to test the hypothesis of long-distance percolation that is built into the Navon and Stolper (1987) model. Depending on the formulation, the calculations can replicate some of the relative LIL enrichments of traverse samples from massifs. Although the equations incorporate time and distance parameters, they fail to reproduce the positions or scales of the patterns. Despite this lack of success, published models of peridotite massif compositions continue to employ the Navon and Stolper (1987) model. That model also has been applied inappropriately to trace element patterns of selected noncomposite xenoliths in suites that largely lack evidence of the preeruptive setting (e.g., Sen et al. 1993). For these reasons we focused our critique on the Navon and Stolper model.

Contrary to Navon et al. (1996), we have never asserted that models of long-distance percolation are inapplicable to all parts of the mantle; in particular we agree with the conjecture of McKenzie (1984) that chromatographic separations of major and trace elements in percolating melt may occur by melt-matrix interactions in the asthenosphere (perhaps represented by massifs derived from the suboceanic mantle), where the thermal conditions likely sustain temperatures that are high enough to prevent rapid solidification of melts. Observations, experimental studies, and theoretical considerations (Hart 1993; references in Nielson and Wilshire 1993, p. 1119) all suggest that fracture flow is the probable means of long-distance melt transport in the colder, more rigid lithosphere. The observational data include lithologic and compositional variations in samples derived from subcontinental and suboceanic lithospheric mantle regions (for xenoliths and massifs, see references in Nielson and Wilshire 1993, p. 1116–1119; also Ocean Drilling Project 1995).

CHROMATOGRAPHIC CALCULATIONS

Nielson and Wilshire (1993) clearly stated (p. 1127) that the finite-plate calculation used by Nielson et al. (1993) models a process that produces a result like that of ion-exchange chromatography and is mathematically similar to the Navon and Stolper (1987) model. Nielson et al. (1993) purposely used a simplified calculation to reproduce the compositional gradients in the Dish Hill xenolith for two reasons: (1) It limits the input to observational data, reducing reliance on a long list of unmeasurable parameters required by the "simple mass-balance calculation" of Navon et al. (1996), and (2) we were testing hypotheses rather than assuming the mechanism of the process. The two hypotheses were as follows: (1) The hornblendite dike selvage and relatively unreacted peridotite in the xenolith represent end-member compositions of metasomatic liquid and matrix, and (2) the observed compositional variations arise from a single-stage process of melt injection and wall-rock reaction. Nielson and Wilshire (1993) focused attention on the positive results of those tests in the hope that other studies might also employ data-based constraints for models of mantle processes and open debate about the actual mechanisms of reaction. Using data of Nielson et al. (1993), Navon et al. (1996) employed the model of Navon and Stolper (1987) to recapitulate one of our successful tests.

DISH HILL XENOLITH

Data and parameters

Xenolith Ba-2-1 from Dish Hill, California, contains reaction fronts of major, minor, and trace elements, isotopic ratios, and volatile species within 4 cm of the contact between centimeter-scale selvage of hornblendite and peridotite wall rock (Nielson et al. 1993). To model the Dish Hill data, Navon et al. (1996) added diffusion terms, which they identify with a change of boundary conditions. In our view, adding diffusion to the calculation constitutes a questionable adjustment of the presumed reaction mechanism of the Navon and Stolper (1987) model, which neglects diffusion effects if the length of the percolation path is large in comparison with the grain size. By this criterion, diffusion should not be important because the percolation distance in the Dish Hill xenolith is greater by a factor of 25 than the average grain size; moreover, the data of Nielson et al. (1993; Fig. 8) show that the slope of the reaction front for Nd ratios is steep enough to discount diffusive effects. The slope of the Sr reaction front is lower and could be ascribed either to limited diffusion or to progressive reequilibration between adjacent volumes of refractory and relatively LIL-enriched peridotite.

The real difference between the models is comprehended by the values for compositional parameters, the real

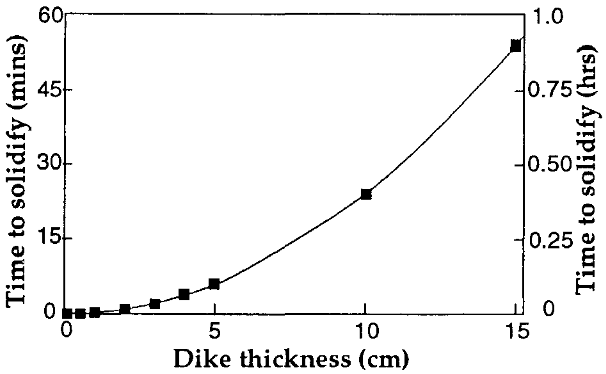

Figure 1. Time required for freezing of thin dikes, for a thermal contrast of 200–250 °C (calculation by A.V. McGuire).

boundary conditions, that are substituted into mathematically similar equations. Navon et al. (1996) used the "higher than expected" clinopyroxene D values calculated by Nielson et al. (1993) from sample data but suggested that those values are high because of processes related to the deposition of interstitial amphibole, or of melt, within the rock. Both amphibole and melt blebs are present within the peridotite, decreasing systematically in abundance with distance from the contact between hornblendite and peridotite in the xenolith (Noller 1986; Nielson and Noller 1987), but calculated modes for the xenolith show that leached clinopyroxene compositions are sufficient to account for whole-rock $\mathrm{LREE_{cn}}$ compositions (Nielson et al. 1993). The Nd isotopic ratios are higher in clinopyroxene separates than in the whole-rock volumes, and Sr ratios are much lower, suggesting that clinopyroxene controlled the distribution of LREE and that other LIL were deposited as melt blebs, in grains of secondary minerals, or both. These relations indicate that the hornblendite was the likely melt source, and we successfully tested that assumption by using the hornblendite composition for the melt end-member (Nielson et al. 1993). In contrast, the discussion of Navon et al. (1996) assumes that similarities between the $\mathrm{REE_{cn}}$ fractionation patterns of hornblendite, minerals, and whole rocks, and the success of models based upon this observation are coincidental.

Temporal and thermal implications

Temporal information that may be derived from the Navon and Stolper (1987) model has important, although unacknowledged implications. In the case of the calculations of Navon et al. (1996) for the Dish Hill xenolith, the minor REE fractionation ($\mathrm{Ce/Yb_{cn}} = 2.5$) required eight months to develop over a lateral distance of ~4 cm. This result requires that the melt source of the Dish Hill fractionation patterns must have remained molten and the conduit must have remained open for at least eight months, and therefore the ambient temperature in the reaction zone must have been near the peridotite solidus for that extended length of time.

Standard thermal models do not support the Navon et al. (1996) result (Fig. 1). Intrusions of a few centimeters to several tens of centimeters in thickness probably represent small volumes of partial melt that stagnate and solidify in minutes to hours (Turcotte and Schubert 1982, p. 172–174). The time (t_s) required for an intrusion of width $2b$ to solidify is calculated from the equation

$$t_s = \frac{b^2}{4\alpha\lambda_2^2} \tag{1}$$

where α is the thermal diffusivity of the wall rock (~0.5 × 10^{-6} m²/s; as great as 1 × 10^{-6} m²/s) and λ_2 is a family of solutions for the part of a transcendental equation that describes the migration of the solidification boundary. Boundary conditions are defined by the movement of the surface of thermal contrast, expressed in the dimensionless variable $\eta = y/(2\sqrt{\kappa t})$, where y is the linear distance in wall rock ($y = 0$ at the intrusive contact) and $\sqrt{\kappa t}$ is the characteristic thermal diffusion distance (Turcotte and Schubert 1982, p. 158–159). In this case η for the melt (η_m) is also expressed as $\eta_m = -\lambda_2$.

Values of λ_2 are obtained from the relation

$$\frac{L\sqrt{\pi}}{c(T_m - T_0)} = \frac{e^{-\lambda_2^2}}{\lambda_2(1 + \operatorname{erf}\lambda_2)} \tag{2}$$

where L is the latent heat of fusion (≅300–400 × 10^3 J/kg), $T_m - T_0$ is the thermal contrast between melt in the dike (T_m) and wall rock (T_0), and c is the wall-rock heat capacity [≅10^3 J/(kg·K)]. The parameter $\operatorname{erf}\lambda_2$ is the error function calculated for η_m (values of $\operatorname{erf}\lambda_2$ are tabulated by Turcotte and Schubert 1982, p. 161).

Average pyroxene compositions from the Dish Hill xenolith produce temperatures between 1008 and 1018 °C by the Wells (1977) method. These values are relatively high for refractory peridotite and may well reflect the ambient temperature of the xenolith source region during melt injection, thus implying a peridotite-dike thermal contrast of 200–250 °C. Substitution of the ranges of values given above (mineral temperatures in kelvins) indicate that mantle intrusions, which commonly are <20 cm thick, would crystallize in less than one hour (Fig. 1).

Discussion

Assessment of the Navon and Stolper (1987) model

The ability of the Navon and Stolper (1987) model to produce the compositional gradient from a single-stage, short-scale, melt-matrix reaction contrasts with the significant lack of success when the model is applied to samples of appropriate scale to test the hypothesis of fractionation by long-distance percolation. As modified, the Navon et al. (1996) percolation model produced the same result as model calculations that show a process by which ion-exchange chromatography traps target ions in matrix.

The success of these calculations should not be taken as a proof for either of the reaction mechanisms.

The value of a model cannot be known until it is tested under the conditions for which it was formulated. The Navon and Stolper (1987) hypothesis has failed to recreate the positions and scales of compositional variations of massif samples on traverses of appropriate scale to test the basic assumptions. In the particular case of a 65 cm traverse across a talus boulder in the Lherz massif (Bodinier et al. 1990), both diffusion-controlled and percolation-controlled models failed to reproduce the scales and positions of significant compositional variations (Bodinier et al. 1990; Nielson and Wilshire 1993). For example, the diffusion-controlled model failed to predict correctly the most highly $\mathrm{LREE_{cn}}$-enriched sample in a 25 cm thick zone of amphibole harzburgite 12 cm from the contact with an amphibole pyroxenite dike. The percolation-controlled model was able to reproduce the observed systematic increase of $\mathrm{Ce/Yb_{cn}}$ in a zone of amphibole-free harzburgite 25–65 cm from the dike contact but at a scale that is 10–1000× greater than the maximum dimension of the boulder. The model also required 26000 yr to develop the relatively moderate LREE/HREE fractionation ($\cong$18× chondrite), which implies that the melt source (probably an intrusion similar to the 10 cm wide amphibole pyroxenite dike in the boulder) remained above its solidus temperature for tens of thousands of years.

Sources of discrepant model results

We believe that the success of the Navon et al. (1996) calculation applied to the Dish Hill xenolith is the result of the relatively simple relations produced by the single-stage reaction and of the use of sample compositions for the *D* values in the calculation. These are the same bases of success for our simplified model calculations (Nielson and Wilshire 1993). The poor performance of the percolation model applied to the Lherz boulder could be ascribed to inappropriate scale (<100 m) for that sample, except that a similar percolation model for a 100 m sampling traverse across peridotite zones and mafic dikes in the Horoman massif, Japan, also produced significant misfits between the actual and expected sequence of fractionation patterns (Takazawa et al. 1992).

We contend that the discrepancies result from the unexamined assumptions about the process mechanism and scale that are inherent in the model and which require substitution of hypothetical values for as many as nine unmeasurable parameters (porosity, melt and matrix velocities and densities, diffusivity of species in the melt, percolation distance, melt volume, and matrix column cross section). The product of mantle melt-matrix interactions is largely influenced by end-member compositions and relative volumes of reactants; of these, the only parameters amenable to observation are end-member compositions. To achieve separations of mechanically similar elements in percolating liquids, calculations that are based on the Navon and Stolper (1987) model use conjectural melt compositions. These compositions are orders of magnitude richer in LIL in comparison with chondrites than the compositions of most mafic dikes or veinlets found in contact with mantle peridotite.

In the case of the Lherz boulder, the application of a percolation-controlled model to the amphibole-free harzburgite zone was predicated on the assumption that the zone had been pervasively altered by metasomatism. Woodland et al. (1992) discovered that the entire boulder contains discrete zones of submicroscopic secondary minerals, demonstrating nonpervasive infiltration of metasomatic agents and disproving the assumption. Bodinier et al. (1990) assumed a single melt source even though as many as five other candidate sources of fractionated melts are present in the zone of amphibole peridotite and the most highly $\mathrm{LREE_{cn}}$-enriched sample, at 12 cm from the amphibole pyroxenite-dike margin, is in the contact zone of a veinlet (Nielson and Wilshire 1993). Like Dish Hill, the shapes of LIL fractionation patterns in peridotite coincidentally mimic those of the intrusions, at lower abundance levels.

Model implications

The extremely long crystallization times for melt sources produced by the Navon and Stolper (1987) model are based on hypothetical rates of percolation through a matrix of hypothetical porosity and density. The long crystallization times have two direct consequences: (1) The sources (probably intrusions) that supplied the percolating melts must have remained liquid for extensive periods of time, requiring that (2) ambient temperatures must have been close to the solidus of peridotite in the percolation-reaction zones for as long as 26000 yr (Nielson and Wilshire 1993, p. 1118). These results are unreasonable and provide the clearest demonstration that the Navon and Stolper (1987) model is inapplicable to lithospheric mantle processes.

Temperatures that are close to the solidus of peridotite are more likely characteristic of the asthenosphere, where long-distance percolation is likely to be the most significant means of melt transport. The parts of upper mantle that we see at the surface are mostly too cold and too refractory to allow small volumes of melt to percolate more than a few centimeters without crystallizing. The change of thermal conditions at the asthenosphere-lithosphere boundary likely favors the coalescence of percolating melts into cracks for transport through the lithosphere (Nicolas 1990).

Models of the Dish Hill compositional gradients show that a small volume of relatively LIL-enriched melt that percolates from its source would add incompatible elements to the matrix; according to Navon et al. (1996), this effect is predicted by the Navon and Stolper (1987) model. Contrary to the doctrine of the percolation hypothesis, the observed end-member compositions of relatively refractory mantle peridotite and fractionated, LIL-enriched mafic intrusions probably represent the reaction end-members. Refractory peridotite in the wall rock reacts with the contrasting melt and is imprinted with the fractionated melt composition. For small melt volumes, each additional aliquot of melt with the original composition percolates unchanged short distances through previously enriched zones (which have reached equilibrium with the melt) but reacts as soon as it encounters matrix of relatively refractory composition. Long-distance disequilibrium percolation of melts and enrichment of peridotite volumes >100 m from a melt source do not occur unless melt volumes are large enough to saturate such huge volumes of refractory lithospheric peridotite, but we are not aware of any mantle samples that exhibit large-scale LIL enrichments in otherwise refractory peridotite.

The observed mantle process resembles the migration of reaction fronts, which characterize water-purification ion-exchange columns (Nielson and Wilshire 1993, Fig. 1c). In contrast, the Navon and Stolper (1987) hypothesis assumes that lithospheric mantle processes imitate laboratory chromatographic columns that concentrate target ions in solution. Navon et al. (1996) derided this distinction between water-purification and analytical extraction processes, asking, "How can one model do what the other cannot when they are almost mathematically identical?" Our answer echoes the observation of astrophysicist David Weinberg, "... you may be able to choose parameters in slightly different ways and get different answers" (quoted in *Science,* v. 268, p. 1849, June 30, 1995). The results of model calculations are determined by the parameters built into the calculations and the values assigned to them (cf. also Oreskes et al. 1994). Navon et al. (1996) stated that the assumed mechanisms of the Navon and Stolper (1987) model constitute boundary conditions. We argue the contrary, that end-member compositions and D values are the controlling boundary conditions.

Navon and Stolper (1987) assigned D values on the basis of the preconception that so-called incompatible LREE (the target ions for this process) travel faster through peridotite matrix than do the HREE. The Dish Hill xenolith shows that this assumption is not correct, but even if it were correct, sorption of the LREE would occur in the mantle column only where D values were different from those along the percolation path. Thus, we would expect mineralogic differences between peridotite matrix of percolation paths and those of sorption areas. According to Navon et al. (1996), such considerations are "not a boundary condition," and the location of the sorption area ("top of the column," cf. Navon and Stolper 1987) "is not important." By this assertion Navon et al. (1996) seem to argue that the presence of a fractionation pattern in peridotite demonstrates the validity of a hypothetical process.

The location of the sorption area may not be important for a laboratory column that contains a matrix that concentrates target ions in percolating solutions because the ions are collected in liquids emerging from the column. The mantle column does not collect metasomatic liquids; instead, the fractionated compositions are recorded in matrix (Nielson and Wilshire 1993, Fig. 1b). Ion-exchange columns that quantitatively remove target ions in matrix either extract ions all along the column (water purification), as we believe happens in mantle lithosphere, or else the D value of column matrix is different from that in the zone where ions are extracted. There is no evidence to indicate that peridotite with variable LIL enrichments had different D values toward mantle melts than nearby zones of unaltered peridotite. Navon et al. (1996) added a third possibility: The melt-fractionation pattern is preserved in matrix at the site where percolation stops. If so, the whole volume of percolating melt must be added to the matrix in that region and therefore should contain overwhelming evidence of modal metasomatism, such as melt blebs and secondary minerals.

Limited trace element compositions (Kelemen et al. 1992) suggest that processes of melt extraction in oceanic asthenosphere do not show the same kinds of fractionation patterns with distance from melt conduits that are seen in xenoliths and massifs of the continental lithosphere. Geochemical data from large tabular dunite zones in such massifs as the Trinity peridotite thus may provide a positive result for the equilibrium-percolation hypothesis of McKenzie (1984). The dunite zones (scales from centimeter to ~400 m wide and up to several kilometers long) in subocean-derived massif have been interpreted as focused flow channels in which percolating olivine-saturated melts dissolved and removed pyroxene from the peridotite (for example, Dick and Natland 1993; Kelemen et al. 1994; Quick and Gregory 1995). Such zones are structurally controlled by the deformation of upwelling asthenosphere at spreading ridges and could result from flow of melts through zones of numerous closely spaced shears or conjugate fractures, accompanied by local percolation effects (cf. Savel'yeva et al. 1980; Hart 1993; Quick and Gregory 1995).

CONCLUSIONS

Fundamentally, the debate about mantle processes arises from attempts to understand processes that create the compositions of erupted lavas, many of which carry xenoliths. We believe that asthenospheric processes, such as equilibrium melt percolation (McKenzie 1984) or disequilibrium flow in vein networks (Spiegelman and Kenyon 1992; Hart 1993), may explain lava compositions but do not explain the observed short-scale variations of trace element and isotopic fractionation patterns that characterize peridotite samples from the subcontinental lithosphere. Trace element patterns in peridotite derived from suboceanic mantle thus may represent the thermal conditions of the asthenosphere and may be more amenable to modeling by processes of long-distance melt percolation. The poor results of Navon and Stolper's calculations for massifs likely result from imposing preconceived mechanisms and inappropriate parameters onto samples from the colder, more rigid subcontinental lithosphere in areas that underwent multiple melt injections. Problematic models should be carefully tested under a range of conditions to reveal their confounding variables and assumptions. Above all, all model makers should be aware that a theoretical calculation can be adjusted to imitate observations even if it does not incorporate the operative mechanisms. The modeler's "sin comes in believing a causal hypothesis is true because [the modeler's] study came up with a positive result, or believing the opposite because [the modeler's] study was negative" (Sander Greenland, epidemiologist, quoted in *Science,* v. 269, p. 169, July 14, 1995).

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