

MASTER

COMPOSITIONAL GRADIENTS IN NATURAL SILICIC LIQUIDS

By

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ABSTRACT

Electron microprobe analyses of natural rhyolite glass reveal compositional gradients 10-15 μm wide adjacent to microphenocrysts of plagioclase, alkali feldspar, and magnetite. The presence of such gradients has previously been interpreted as indicative of disequilibrium between liquid and solid. If crystals and liquid are not in compositional equilibrium, temperatures determined from the compositions of coexisting mineral phases might be erroneous. Gradients may be attributed, however, to diffusion controlled crystallization. Because convection of magma is indicated based on a comparison of characteristic transport distances and thickness of compositional boundary layers the model of Tiller et al. (1953) is deemed inappropriate. The microprobe data are consistent with the model of Burton, Prim, and Slichter (1953) in which the liquid is stirred by convection and the crystals are surrounded by a diffusive-advective boundary layer. Computed apparent and equilibrium distribution coefficients for natural rhyolites, combined with published diffusion coefficients for K, yield linear growth rates for feldspars on the order of 7×10^{-7} cm sec⁻¹, similar to experimentally determined equilibrium growth rates. The analytical data are consistent with a model of diffusion controlled crystallization in natural silicic liquids, and the utilization of coexisting phases for thermometry remains reasonable in the presence of narrow compositional gradients.

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INTRODUCTION

The assumption of equilibrium between phases is critical to the application of any geothermometer to natural systems. In a previous study of a silicic volcanic system associated with the Roosevelt Hot Springs thermal area (Evans, 1978) use was made of the iron-titanium oxide geothermometer of Buddington and Lindsley (1964) and the two feldspar geothermometer of Stormer (1975), to calculate eruptive temperatures. The present study was undertaken to determine if equilibrium did exist between phases present in two high silica rhyolites. The criterion selected to evaluate the state of equilibrium in the two rhyolites was the presence or absence of compositional gradients adjacent to feldspar and magnetite microphenocrysts.

Previous work on this problem in natural systems is sparse. Bottinga et al. (1966) documented the presence of compositional gradients adjacent to plagioclase crystals in basaltic glass. They suggested that diffusion in a compositional boundary layer next to plagioclase is the rate determining step in plagioclase growth, and that fluctuations in the concentration of constituents in the boundary layer result in oscillatory zoning. Anderson (1973) also reported compositional gradients adjacent to olivine and plagioclase from a Hawaiian basalt. His results were similar to Bottinga's in magnitude and direction, and were also interpreted to be the result of crystal

growth. Smith (1974) in a review of previous work and more recent experimental results concluded that the reported absence of gradients in calcium and sodium by Bottinga et al. (1966) is problematical because these elements would be expected to be depleted adjacent to plagioclase. Experimental determination of the diffusivity of calcium by Magaritz and Hofmann (1977) indicates that gradients in calcium could develop during plagioclase growth depending on the crystal growth rate, as will be discussed in more detail below. Sibley et al. (1976) discussed the development of oscillatory zoning in plagioclase and related its development to transient constitutional supercooling in the interfacial liquid.

In this study, two samples of obsidian containing approximately 5% microphenocrysts were selected for analysis. These samples are from the interior of two volcanic domes in the Mineral Mountains, southwestern Utah (Lipman, et al., 1978). An attempt was made to detect compositional gradients adjacent to microphenocrysts of sanidine, plagioclase and magnetite by electron microprobe analyses. Figure 1 shows photomicrographs of representative grains of sanidine and oligoclase. Average grain sizes for these minerals are 0.3 mm and 0.5 mm, respectively. The feldspar grains are euhedral in habit. The magnetite grains are so small that their morphology is obscure. Most appear to be subhedral cubes or octahedrons.

Figure 1. Photomicrographs of feldspar microphenocrysts. a) Sanidine,
b) Plagioclase (Black dots and black margins on upper and
right side are ink markings).

ANALYTICAL RESULTS

A thorough analysis of the glass matrix far removed from crystals was undertaken to establish the degree of homogeneity of the glasses. Chemical analyses of the two obsidians studied are presented in Table 1. All constants except H₂O were determined with an electron microprobe using a beam diameter of 50 micrometers. The analytical precision is indicated to \pm one sigma. H₂O values were determined on the bulk sample by a modified Penfield method and all water was assumed to be in the glass.

A main objective of this study was to obtain precise chemical analyses of the glass matrix as close to crystalline phases as possible. In order to accomplish this a one micrometer beam was used on our ARL-EMX electron microprobe. The stage of the microprobe was translated manually with the beam initially located on a microphenocryst. A traverse was then made outward across the mineral-glass interface and continued for approximately 30 to 50 micrometers into the glass. A step length of 2 micrometers was used in order to estimate, as nearly as possible, the concentration of elements in the glass immediately adjacent to a mineral grain. This step interval also allowed location of the mineral-glass interface to a precision of 1 micrometer. Step distance was measured on the scale provided on the stage drive of the electron microprobe with backlash removed before each traverse was begun.

As a preliminary step, reconnaissance traverses were made at 5 μ m intervals which established that gradients were present adjacent to small grains of magnetite and feldspar. The results of these

Table 1. Microprobe analyses of glass 2 μ m from crystal-glass interface and bulk composition remote from crystals. Values given in weight percent oxide.

MR75-14

	2 μ m from Sanidine	2 μ m from Plagioclase	Bulk Composition	(1 σ)
SiO ₂	77.3	77.2	76.5	0.5
Al ₂ O ₃	11.9	11.8	12.6	0.2
FeO	0.50	0.55	0.53	0.08
CaO	0.40	0.30	0.38	0.02
K ₂ O	4.2	4.5	4.6	0.1
Na ₂ O	-	-	4.6	0.2
H ₂ O	-	-	(0.14)	

MR75-20

	2 μ m from Sanidine	2 μ m from Plagioclase	Bulk Composition	(1 σ)
SiO ₂	77.0	77.5	76.5	0.5
Al ₂ O ₃	12.3	12.3	12.8	0.2
FeO	0.56	0.58	0.58	0.08
CaO	0.35	0.40	0.40	0.03
K ₂ O	3.6	4.4	4.5	0.1
Na ₂ O	-	-	4.3	0.2
H ₂ O	-	-	(0.10)	

reconnaissance traverses for magnetite are presented in Figure 2. Because of the small grain size of the magnetite detailed traverses were not undertaken.

The direction of the gradients shown in Figure 2 are those to be expected from growth of magnetite. Constituents which are not stoichiometric components of magnetite (SiO_2 , Al_2O_3 , K_2O) are preferentially rejected during growth and consequently build up in the boundary layer. Iron however shows a decline due to its preferential incorporation into magnetite. Calcium and titanium contents are so low that gradients cannot be resolved in the boundary layer. Chemical components are presented as oxides on all figures solely as a matter of convenience for easy comparison with bulk values for oxides in the glass. It should not be inferred that these are the actual species transferred during crystal growth.

Various attempts were made to analyze for sodium, but all failed because of its high volatility in glass under the microprobe beam. If the beam diameter was increased, the gradient could not be resolved, whereas, if sample current was lowered with the beam diameter maintained at one micron, volatilization continued to vanishingly small sample currents. Water as well could not be analyzed. Perhaps an ion microprobe might be able to analyze for water in the glasses studied but it is doubtful if present instrumentation could resolve a gradient in water within a distance 10-20 μm from a crystal boundary.

Analytical results for detailed traverses adjacent to feldspar crystals are presented in Figure 3. In boundary layers adjacent to both sanidine and plagioclase the directions of enrichment or

Figure 2. Reconnaissance traverses of glass adjacent to magnetite microphenocrysts. Error bars show precision of analyses to one sigma. Note: Components plotted as oxides for convenience only.

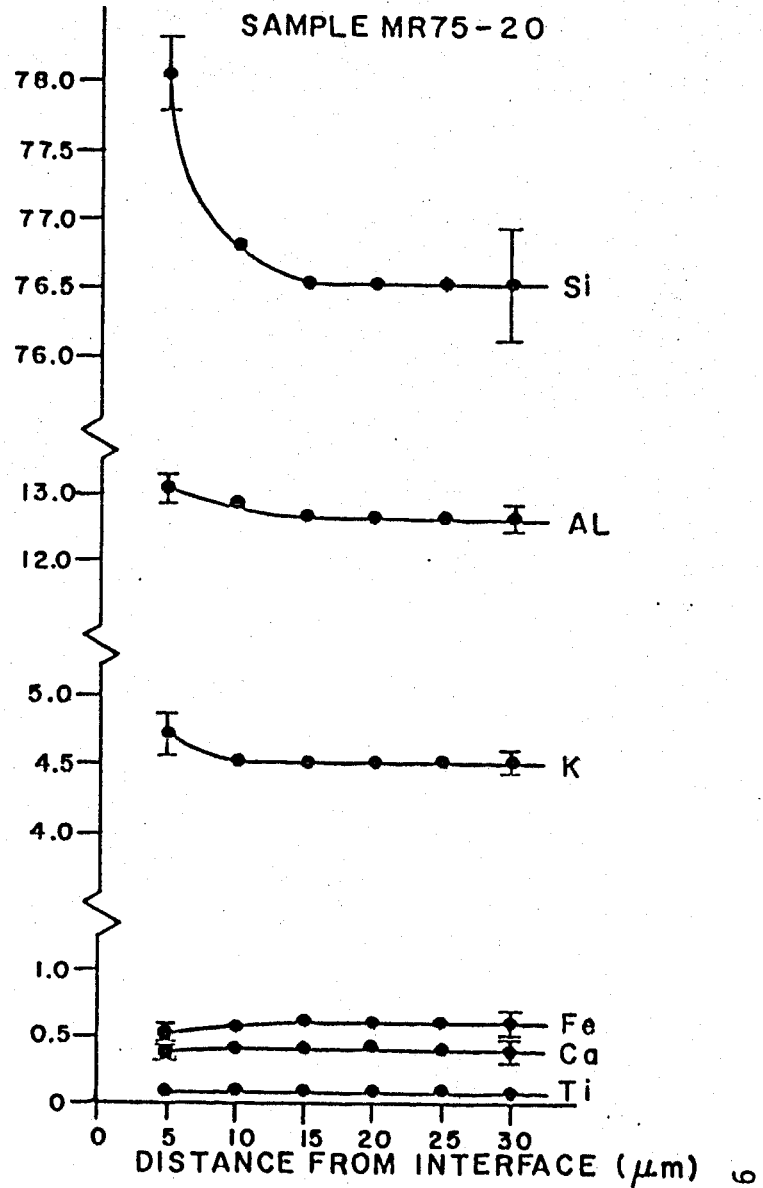
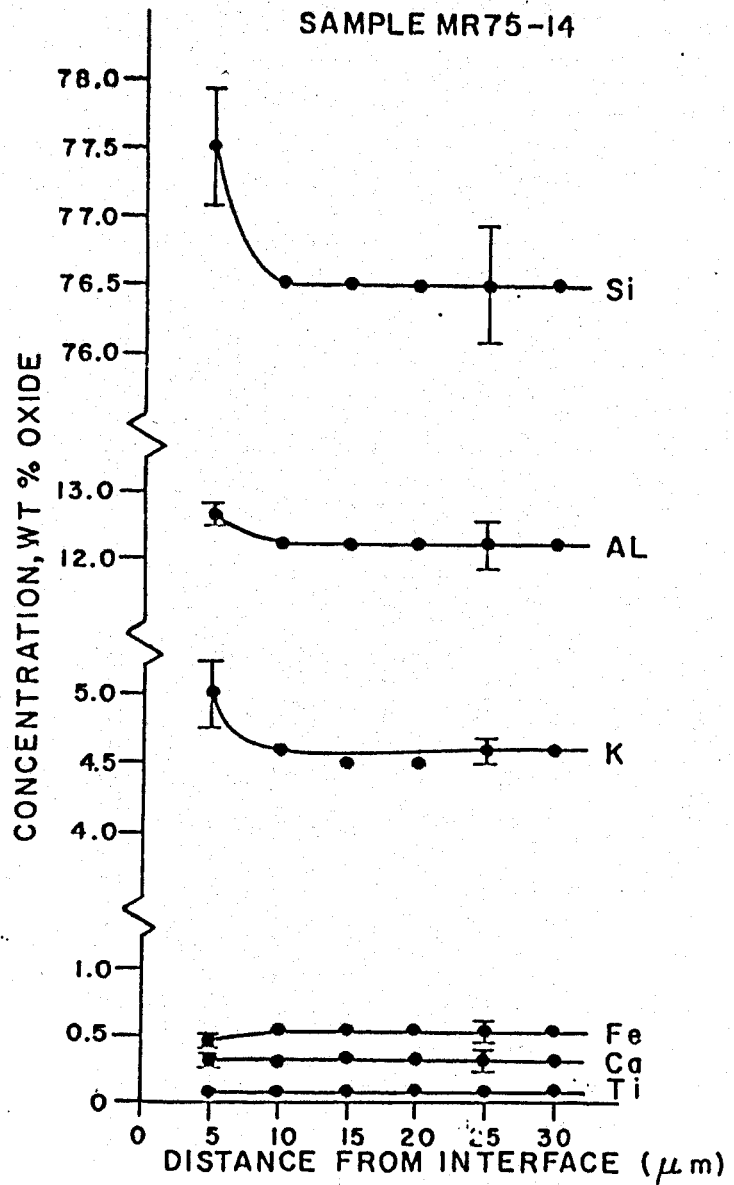


Figure 3a. Detailed traverses at 2 μm intervals of glass adjacent to sanidine and plagioclase microphenocrysts in obsidian sample MR75-14. Shaded areas encompass all analytical results obtained. Note: components plotted as oxides for convenience only. Tick marks on abscissa labeled with chemical symbols are concentrations of components in crystalline phases.

MR 75-14

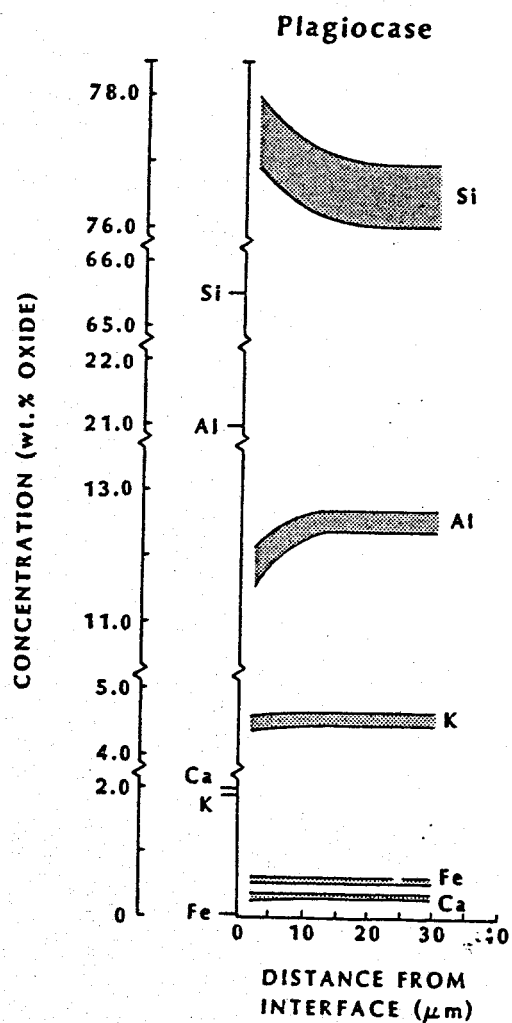
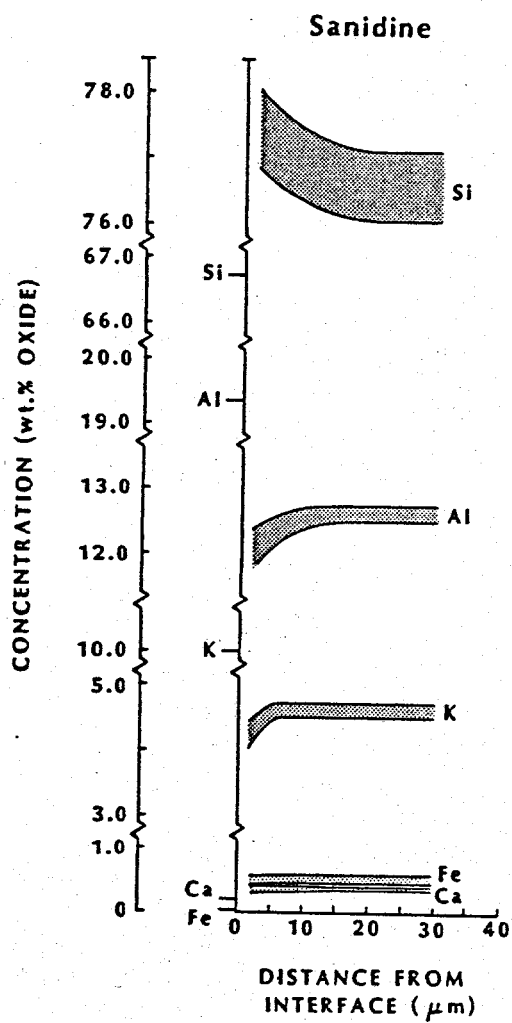
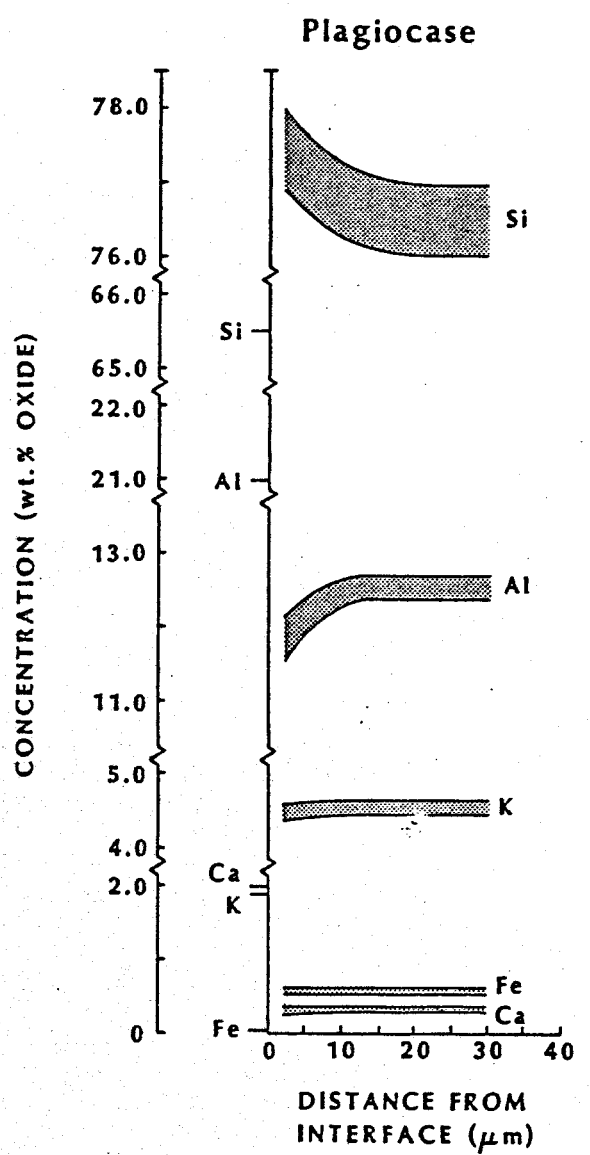
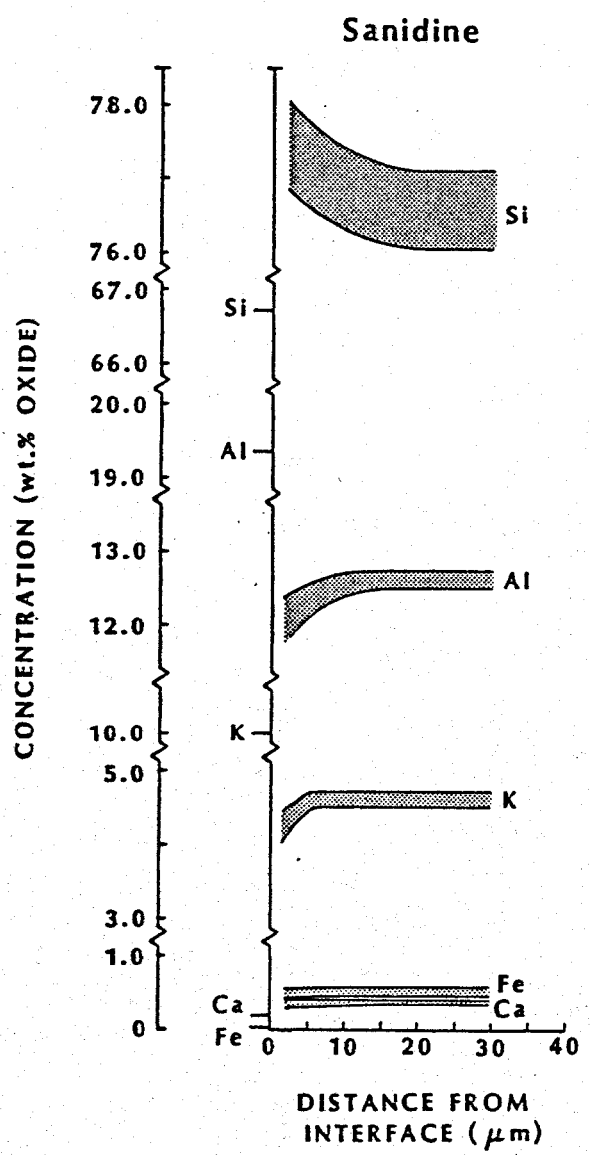


Figure 3b. Detailed traverses at 2 μ m intervals of glass adjacent to sanidine and plagioclase microphenocrysts in obsidian sample MR75-20. Shaded areas encompass all analytical results obtained. Note: Components plotted as oxides for convenience only. Tick marks on abscissa, labeled with chemical symbols, are concentrations of components in crystalline phases.

MR 75-14



depletion are in accord with a crystal growth model. Constitutents incorporated preferentially into the solid (Al_2O_3 , K_2O) are depleted in the boundary layer, whereas SiO_2 , which is lower in feldspar than in the liquid, is concentrated in the boundary layer. The direction of the gradient for CaO is also compatible with crystal growth, as it is depleted adjacent to plagioclase. Average values for components $2\mu\text{m}$ from the crystal-glass interface, obtained during the gradient studies, are also given in Table 1.

Feldspar microphenocrysts were carefully analyzed on the electron microprobe as part of the present study in order to assess intra-grain zoning and variation of composition between grains. Figure 4 presents the results of microprobe analyses of approximately twenty grains from the two obsidians studied. As can be seen, zoning within individual feldspars is minimal, averaging approximately 2-3 mole percent. Inter-grain variability is also minimal, again on the order of 2-3 mole percent. The average compositions of feldspars are given in Table 2. Previous analyses of oxide phases separated from glasses by heavy liquid extraction indicated they were homogeneous (Evans, 1978).

A curious feature is the presence of albite rims on some sanidine microphenocrysts in each of the two glasses. These rims are 1-2 μm wide, barely analyzable on the microprobe, and remarkably uniform in composition. Analyses of these rims are shown in Figure 4. Rims are present on approximately one quarter of the sanidines analyzed and do not affect the magnitude or direction of gradients as compared to those adjacent to sanidines without rims. The rims are seldom continuous around a given grain but do tend to follow any

Table 2. Average feldspar compositions determined by electron microprobe.

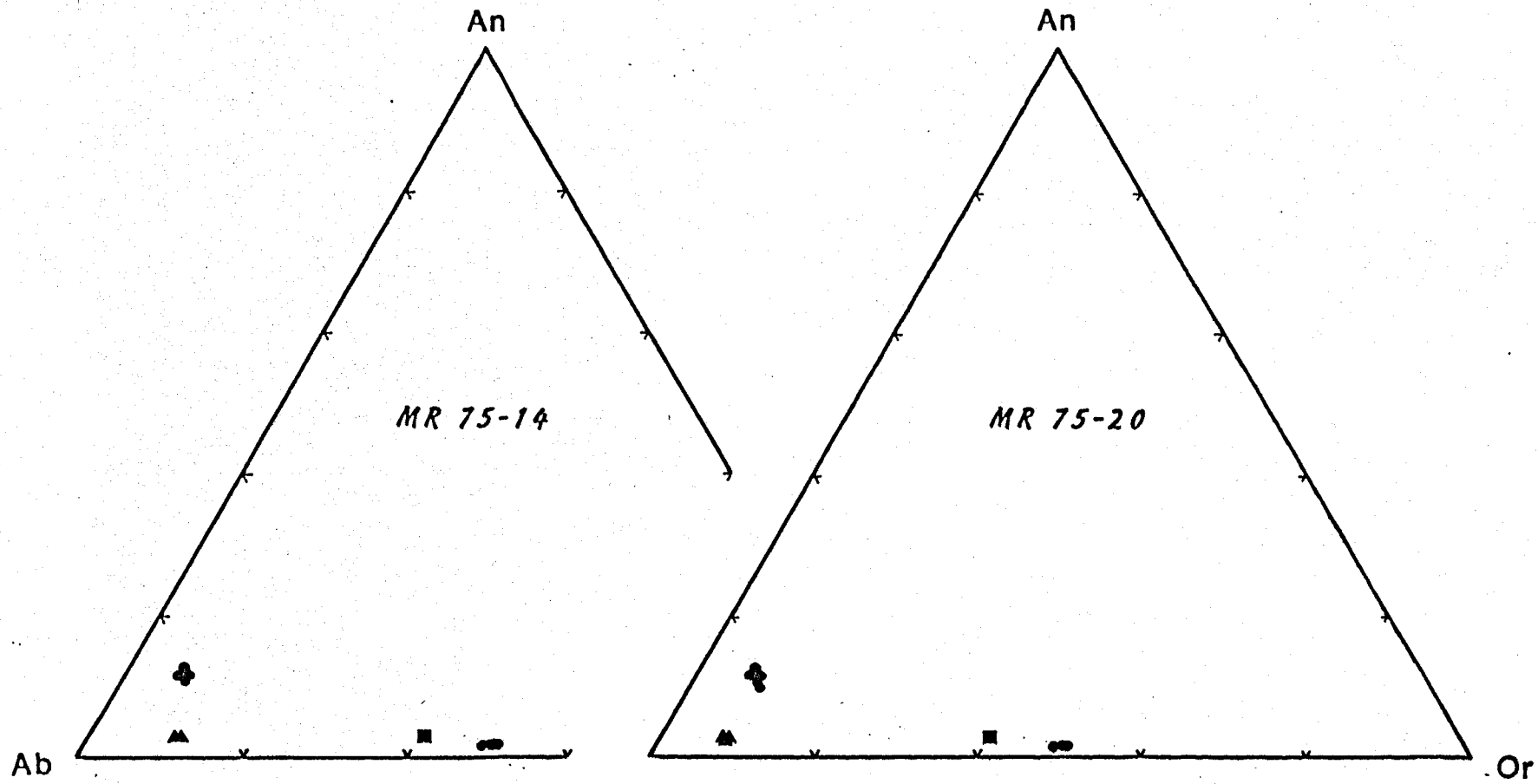
	MR75-14		MR75-20	
	Sanidine	Plagioclase	Sanidine	Plagioclase
SiO ₂	66.7	65.5	66.5	65.3
Al ₂ O ₃	19.4	20.9	19.7	21.4
Fe ₂ O ₃	0.21	0.24	0.20	0.22
CaO	0.20	1.91	0.20	1.88
K ₂ O	10.0	1.83	10.2	1.79
Na ₂ O	4.45	9.31	4.76	9.52
TOTAL	101.0	99.7	101.6	100.1

Two feldspar
geothermometer
temperature
(Stormer, 1975)

720°C

715°C

Figure 4. Feldspar analyses plotted in terms of mole percent. Filled circles represent sanidine and plagioclase microphenocrysts, triangles are albitic rims discussed in text, and squares are whole rock analyses plotted in terms of normative feldspar content.



irregularities present. Similar rims have been described by Gilbert (1938) and Hildreth (1979) for feldspar of the Bishop Tuff, and were interpreted by them to be late stage vapor deposited phenomena. This mechanism is a possible explanation for the rims in the Mineral Mountains obsidians but their presence on sanidines in compact glasses rather than porous pumices is difficult to interpret.

DISCUSSION

There is little doubt that the compositional gradients are caused by crystal growth, this is substantiated by both the magnitudes and directions of the gradients determined. In order to assess possible departure from equilibrium which may be represented by the compositional gradients preserved in the glasses studied, some judgement must be made regarding the mode of crystal growth. Growth may have been either steady state or transient in nature.

There are various theoretical models for crystal growth which may be applied to the analytical data. For steady state growth the models of Tiller et al. (1953) or Burton et al. (1953) may be applicable. Other theoretical models are available for transient growth (see Rosenberger, 1979, Chapter 6).

Transient crystal growth is growth for which the rate of crystallization is time-dependent. Two criteria may be used to decide whether crystal growth was transient or not. The first, and most obvious, is the presence or absence of zoning in crystals. Zoning is absent or minor in feldspars in the obsidians studied. If a crystal is zoned this indicates that the crystal growth rate varied during crystallization. The functional dependence between growth rate and zoning has recently been modeled by Haase et al. (1980) for plagioclase. They show that oscillatory zoning results from the interplay of: (1) the dependence of growth on melt and solid surface composition, (2) the stoichiometry of the solidification reactions, (3) the diffusion of melt species, and (4) the motion of the growing crystal rim. If zoning is absent steady state crystallization is

assumed to be the mode of growth and no time dependence of the four factors above existed during growth of the crystal.

A second criteria of steady state crystallization is the presence of well developed facets on the feldspar crystals. If growth rate varied with time preferential growth of certain crystal faces would have taken place. If transients in growth rate were extreme the development of atoll and swallow-tail morphologies would be expected. Petrographic study indicates feldspar crystals in the glasses studied are tabular to equidimensional and are therefore interpreted to have grown under steady state conditions.

The approach to equilibrium between sanidine and plagioclase may be assessed by comparing growth rate data from experiment (Swanson, 1977) and calculated growth rates for the feldspars in the analyzed samples. In order to proceed with a calculation of growth rate two items merit discussion. First, what is meant by "equilibrium"? In crystal growth from the melt, equilibrium is confined locally to the crystal-liquid interface. This local equilibrium is a function of the interfacial mass transfer kinetics controlling transport of material from the liquid to the solid. How far this interfacial or local equilibrium deviates from bulk equilibrium between solid and liquid is controlled by transport kinetics in the liquid and the crystal growth rate. Secondly, the convective state of the liquid needs to be known in order to decide on an appropriate crystal growth model. If the liquid is stagnant then the model of Tiller et al. (1953) is appropriate. However, if the liquid is convective, either free or forced, and has reached a steady state condition, then the model of

Burton et al. (1953) is appropriate.

In order to decide upon an appropriate liquid flow regime a characteristic distance for mass transport near the crystal-liquid interface can be conveniently calculated. This distance is $Y' = D_K/V_C$ or diffusivity divided by crystal growth rate (Rosenberger, 1979). For feldspars this distance is on the order of 100 to 1000 μm ; assuming a diffusivity for potassium of $1 \times 10^{-8} \text{ cm}^2/\text{sec}$ and growth rates of $1 \times 10^{-6} \text{ cm/sec}$ to $1 \times 10^{-7} \text{ cm/sec}$ (data from Margaritz and Hofmann, 1978 and Swanson, 1977). Because this characteristic distance is substantially greater than the observed thickness of the boundary layer in the glasses studied, it serves as strong evidence for convective motion during the growth of microphenocrysts. The impetus for convection is a matter of speculation: free convection in a magma chamber caused by a thermal gradient, or forced convection caused by movement of the magma through a volcanic conduit are possibilities.

Additional evidence for thorough mixing of the magma is seen in chemical analyses of a rhyolite flow erupted prior to the domal material described here. Four analyses (Evans, 1978), one at the distal end of the flow, two at an intermediate distance from the source and one analysis close to the vent are chemically indistinguishable from one another. This flow is approximately 0.1 to 0.2 km^3 in volume, a volume considerably greater than that of any of the domes. It is apparent then that substantial volumes of compositionally homogeneous magma existed from which crystalline phases could grow. Beyond the compositional boundary layers there is

evidence that this homogeneity was maintained by convective mixing of the magma, but on what scale this convection operated is unknown.

Recent studies of experimentally determined growth rates for feldspars have been conducted by Fenn (1977) and Swanson (1977). Swanson's data are particularly pertinent because his liquid compositions are comparable to those of the glasses of this study. Fenn searched for compositional gradients adjacent to his synthetic feldspars and found none. However, the technique he used would not have revealed gradients of the magnitude detected in the present study. Swanson made no attempt to measure gradients in his experiments.

Growth rates may be calculated by adopting the model of Burton, Prim and Slichter (1953) to describe the segregation of potassium in the liquid during crystal growth. It is important to stress that the Burton-Prim-Slichter model is a hybrid; in the original paper mixing of the fluid was forced by rotating the growing crystal, a technique common in metallurgy. This technique generates a forced convective regime around the growing crystal which gives rise to a momentum transfer boundary layer accounted for in their model as a stagnant boundary layer. Within this stagnant boundary layer diffusion was purported to be the dominant mode of mass transfer. This model is now known to be physically unrealistic. Rosenberger (1979) has pointed out that the B.P.S. relationship is not restricted to rotated systems and applies to any diffusive-convective growth situation so long as the boundary layer thickness is properly defined. Recent papers by Wilson (1978a, 1978b) further clarify the definition of the boundary

layer in the B.P.S. relationship.

For the crude calculation of growth rate to follow the B.P.S. relationship will be used in the form:

$$k = \frac{k_0}{k_0 + (1-k_0) \exp(-\delta' V_c/D)}$$

where k_0 is the "equilibrium" interfacial segregation coefficient, k is the bulk segregation coefficient, V_c is the crystal growth rate, D is the diffusivity of the component under consideration and δ' is the boundary layer thickness within which diffusive-advective transport dominates over convective transport. Another assumption necessary for application of the B.P.S. relationship is that the densities of the liquid and solids are equal (or nearly so), which is generally satisfied for geological materials.

It is possible to estimate k_0 for potassium using analyses at $2\mu\text{m}$ from the interface to approximate the interfacial equilibrium values. k may be calculated from the bulk values for glasses and feldspar. The estimates for k and k_0 for both glasses are given in Table 3. The diffusivity for potassium is calculated from the relationship given by Magaritz and Hofmann (1978):

$$D_K = 0.0050 \exp(-26.0 \times 10^3/RT)$$

The temperature used was that calculated from the two-feldspar geothermometer of Stormer (1975) (Table 2). Finally δ' , the diffusion boundary layer width, is determined directly from the analytical data presented in Figure 3. The boundary layer thickness for potassium is $10\mu\text{m}$ (10^{-3} cm). For sample MR75-14 a crystal growth rate for sanidine is calculated to be 7×10^{-7} cm/sec and for sample MR75-20 the rate is

Table 3. Equilibrium interfacial segregation coefficients (k_0) and effective segregation coefficients (k) for potassium between sanidine and liquid (glass).

Sample	k_0	k
75-14	2.38	2.17
75-20	2.83	2.26

1×10^{-6} cm/sec. These values compare favorably with growth rates determined by Swanson for an undercooling of 100-200°C. (Swanson's diagrams are difficult to interpret because the abscissa is apparently mislabeled in his Figure 3a.) It is possible that the undercooling below the liquidus for sanidine might be less than indicated above, as would be expected in order to develop the euhedral morphology of sanidine observed in the glasses of the present study. In the work of Fenn (1977), undercoolings of 20°C to 40°C lead to the development of crystal morphologies similar to those shown in Figure 1.

CONCLUSIONS

The presence of compositional gradients adjacent to feldspars apparently do not imply severe disruptions of crystal-liquid equilibrium during crystallization. Rates obtained computationally, based on equilibrium assumptions, are comparable to those determined experimentally for sanidine. The generation of compositional gradients is to be expected for any reasonable growth rate.

Equilibrium between mineral pairs constituting geothermometers, such as feldspars, is taken to mean that local equilibrium exists at the crystal-liquid interface, and that through successive local equilibria (i.e. by virtue of microscopic reversibility) equilibrium between crystals and liquid is obtained and hence equilibrium between mineral pairs also exists. In the context of igneous petrology the mere presence of compositional gradients is not a sufficient condition to discount equilibrium between minerals useful as geothermometers. Rather it is important to emphasize that compositional gradients are a natural consequence of crystal growth from a melt even under steady state conditions in which values for the diffusivities of migrating components are small. Future studies of igneous systems in which mineral pair geothermometers are to be utilized should include an investigation of homogeneity of the minerals and subsequent assessment of growth mode. In glassy samples, an attempt should be made to detect compositional gradients, bearing in mind that careful

microprobe work may be required to resolve them when they are 10-20 μm wide.

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