SOME EFFECTS OF VISCOSITY ON THE DYNAMICS OF REPLENISHED MAGMA CHAMBERS

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Some aspects of the dynamical Abstract. behavior of magma chambers, replenished from below with hotter but denser magma, have been modeled in a series of laboratory experiments. In previously reported work the fluids used were aqueous solutions of comparable viscosity, and thus the results should be applicable to basaltic magma chambers, in which the magmas do not vary greatly in viscosity. In that case, the lower layer cools by convective heat transfer to the fluid above, and crystallization causes the density of the residual liquid in the lower layer to decrease. When the density becomes equal to that in the upper layer, sudden overturning and intimate mixing take place. The present paper reports experimental results that allow us to extend the application to systems in which there is a large viscosity ratio between the resident and the injected fluid, for example, to calcalkaline magmas, where magma viscosity can vary by as much as 5 orders of magnitude. The largest viscosity ratio in our experiments (about 3000) was achieved using cold glycerine for the upper layer, above a hot denser KNO₃ solution. The most striking new feature with the very viscous upper layer is that now less dense fluid is released immediately and continuously from the interface and rises as plumes through the upper layer. Further crystallization occurs in the plumes, and the crystals fall out, but there is little mixing, and a layer of depleted KNO3 solution is eventually deposited at the top. The transfer process between the layers is dominated by interfacial effects, with the high-viscosity upper layer acting as a nearly rigid lid that allows buoyant fluid to accumulate just below the interface and then rise in localized plumes across the interface into the viscous layer. This physical picture is supported by a series of experiments in which the viscosity ratio is varied systematically; the mixing behavior changes gradually between that described above for a large viscosity ratio and the sudden overturning characteristic of layers with comparable viscosity. The importance of the viscosity ratio, rather than just an increase in viscosity, is confirmed by experiments in which both viscosities are increased by the same factor; the overturning process is then slower, but symmetrical. Other variations suggested by previous

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Paper number 4B0586. 0148-0227/84/004B-0586\$05.00 experiments are also described; the release of gas by a chemical reaction, to model the release of volatiles following an overturning event in a magma chamber; the effect of a cold, immiscible layer above the cooling crystallizing fluid; the influence of two viscous layers with a density step between them; and the constraining effects of a density (with corresponding viscosity) gradient in the upper region. The experiments indicate that whatever the stratification, whether it be in layers or continuous, the form of the initial motion in the upper fluid is determined by the viscosity ratio between the two fluids immediately adjacent to the interface. Geological applications are not examined in detail in this paper, but the experiments suggest that both sudden overturning (characteristic of magmas of nearly equal viscosity) and continuous release (when the upper layer is much more viscous) are viable mechanisms for magma mixing in the appropriate circumstances.

1. Introduction

During the past few years there has been an increasing awareness of the importance of fluiddynamical processes in the formation of various geological structures. The evolution and differentiation of magmas and the compositional zonation of volcanic sequences or layared igneous intrusions cannot be understood in terms of static processes alone. These geological phenomena depend fundamentally on the density differences and the resulting convective motions that are produced during the crystallization process itself. An outline of the wide variety of dynamical processes that are relevant at various stages of the history of a magma chamber has been given recently in geological terms by Sparks and Huppert [1964] and Sparks et al. [1964].

Part of the physical background to such models has been derived from laboratory experiments using aqueous solutions. Though the properties of such fluids may at first sight seem very different from those of magmas, it has been shown in a series of papers [e.g., Chen and Turner, 1980; McBirney, 1980; Huppert and Turner, 1981a; Turner and Gustafson, 1981; Huppert et al. 1982b] that the overall dynamics of certain processes in magma chambers can be modeled in aqueous systems. We have concentrated so far on two experimental configurations. First, the phenomena of layering and differentiation have been studied in a rectangular container filled initially with homogeneous fluid, with cooling and crystallization taking place at a vertical boundary. Second, we have examined the evolution of a chamber replenished at the base with new, hotter but denser fluid, which crystallizes as cooling takes place by heat transfer to the layer above.

The results described in the present paper represent extensions of the latter type of experiment and were designed to study the effect of viscosity on the behavior of such replenished systems. While we have no doubt that viscous effects will be geologically important, the emphasis in this paper is on the fluid-dynamical concepts, and only a brief outline will be given of pounible detailed applications to geological problems. The presentation will be largely descriptive and visual, using numerous photographs to document the new effects observed.

In the original experiments of Huppert and Turner [1981a] and Huppert et al. [1982b], aqueous solutions that differed little in viscosity from each other were used to represent both the resident magma and the new, denser magma forming the lower layer. This provided a good analog of basaltic magma chambers, in which magmas do not usually vary greatly in viscosity. Thus a single overall convective parameter (the Rayleigh number Ra) could be used in this application to characterize the dynamic behavior of the lower layer, and it was argued that the Rayleigh number in the experiments was sufficiently large that the observed dynamical effects were similar to those expected in basaltic magma chambers. Many other magma systems, on the other hand, can have a very large variation of viscosity. For example, in calcalkaline magmas, magma viscosity can vary over 5 orders of magnitude from fluid basalt to viscous rhyolite. When a low-viscosity magma is emplaced beneath a much higher-viscosity, differentiated magma, one might expect that the dynamical behavior will be changed, and so a complementary series of experiments is needed, covering a range of viscosity ratios.

A few experiments on convection in fluids with variations of viscosity have already been motivated by geological problems, and some of these will be referred to in the later discussion. For example, Turner [1973] used the temperature-dependent viscosity of glycerine to model subducting plates in an experiment analogous to mantle convection. Whitehead and Luther [1975] described experiments and related theories on the slow rise of less dense fluid through another of much larger or smaller viscosity, and suggested applications to the rise of diapirs and to larger-scale phenomena such as mantle plumes. Marsh [1979] reported a series of experiments on the instability of a thin layer of less dense fluid below a more viscous upper layer, and related his results to island arc magmatism.

Two different experiments described in our earlier papers are relevant to the current inventigation. In the first, a layer of hot KNO_3 solution was emplaced below a cold, less dense, and deeper layer of NaNO₃ solution. Rapid transfer of heat across the sharp interface separating them drove vigorous convection in both layers, with insignificant transfer of the solutes. Crystallization of the KNO₃ solution took place as it cooled, leaving behind lighter residual fluid, so that the density of the lower layer gradually decreased. Eventually its density became equal to that in the upper layer, and rapid overturning and mixing occurred. In the other type of experiment, hot KNO_3 solution was added below a stratified solution of cold $K_2\text{CO}_3$. The transfer of heat from below first produced double-diffusive layering in the gradient region, by a mechanism investigated earlier by Turner [1968] and Huppert and Linden [1979]. When crystallization had reduced the density of the lowest layer sufficiently, it overturned, but since its rise was inhibited by the density gradient, it mixed only with the lower part of the stratified and layered upper fluid.

A further variation on these techniques was explored in the experiments reported by Turner et al. [1983]. Nitric acid was mixed with the lower KNO₃ layer, and a carbonate component was added to the upper layer. When overturn occurred, vigorous release of CO₂ bubbles took place due to the reaction between the acid and carbonate, and we have argued that this provides a uneful analog of saturation effects in volatile-rich magma chambers [Huppert et al., 1982a].

The plan of the present paper is as follows. In the next section we describe the properties of the fluids used in the experiments and the methods of setting them up. Section 3 deals in turn with various two-layer systems, starting with the case of a cold viscous layer above a hot, dense, crystallizing aqueous solution. We then describe a sequence of experiments with a systematic variation in the viscosity of the upper layer. The effect of increasing the viscosity in both layers is examined next, followed by the effects of gas release during overturning. We also describe an experiment using immiscible fluids in the two layers. At intervals we also discuss various subsidiary experiments designed to shed light on particular questions raised by the main sequence of experiments. In section 4 we extend the discussion to treat three-layer systems, i.e., mixing of a lower crystallizing layer into two viscous layers above it, with both the viscosity ratio and density difference now being variable. Section 5 describes experiments in which there was a stable density gradient in the cold upper viscous region, with the viscosity either decreasing or increasing with height. In section 6 we briefly outline some of the possible geological applications. In the final section we draw together all the experiments and discuss the differences between them and the general conclusions that can be drawn.

2. Experimental Method

Most of the experiments were carried out in a Plexiglas tank of internal dimensions 200 x 106 mm x 300 mm deep. This was rather smaller than the tank previously employed for "replenishment" experiments, since we wished to reduce the amount of the more expensive glycerine needed for the present study. It was, however, found to be adequate for the purpose, as was checked by comparing the results with one run in the larger tank, $400 \times 200 \times 300 \text{ mm}$ deep. During exploratory runs, we found that when a cold, very viscous upper layer was used, the crystallization and convection were much more strongly affected by the temperature and resulting viscosity gradients



Fig.1. Sketch of the experimental tank, and the system used to fill it with a viscous (and sometimes stratified) upper layer.

produced at the side walls than they had been in earlier experiments using aqueous solutions of comparable viscosities. Consequently, we added a "box" to the bottom of the tank, with internal dimensions $154 \times 60 \times 50$ mm deep, to contain the hot lower fluid and keep it initially 20 mm away from all the side walls. The space between the top of this box and the walls of the tank was sealed to create a partial false floor, as sketched in Figure 1.

The lower layer of KNO was made up to a nominal concentration of 41 wt for most of the experiments, and a small amount of dye was added for visualization purposes. The solution was heated to a temperature of approximately 62°C and poured into the box to a level just below the The more viscous upper layer fluid was rim. prepared in advance and left in a cold room overnight to bring it to the required temperature. For fluids with viscosities not too different from the viscosity of aqueous solutions, the tank was filled using a float with a porous foam bottom, as in previous experiments. With much higher viscosities (especially when the upper layer was nearly pure glycerine) this method was too slow, and the float interfered with the motion in the upper layer. Thus a new filling device was designed and built. This consisted of two tubes of internal diameters 32 and 45 mm sliding inside one another with a water-tight seal between; these were fitled below a reservoir and could be extended over the depth of the experimental tank. A circular plate at the lower end spread the flow out horizontally over the lower layer, and this end was continually raised so that it remained just below the surface as the fluid level increased. A second reservoir was connected to the first to provide the means to stratify the upper layer of viscous fluid when necessary, using the "double bucket" technique, as is also shown in Figure 1 and described in section 5. The rubber stoppers in the tube con-



Fig.2a. The viscosity of the commercial glycerine used in the experiments, measured as a function of temperature. Note the logarithmic viscosity scale.

necting the two reservoirs and at the top of the supply tube to the experimental tank could be removed partially or completely and acted as control valves during the filling process.

The fluids used for the upper layer were glycerine or mixtures of glycerine with aqueous solutions of sugar or NaNO₂. This allowed independent control of both viscosity and density. The viscosity of such solutions varies greatly with both temperature and composition. As a basis for comparison, we have plotted (Figure 2a) the viscosity of the pure glycerine we used over a range of temperatures and (Figure 2b) the viscosity of glycerine-water mixtures at the fixed temperature of 25°C. These should be used for guidance only, and extensive measurements and calibrations were needed to determine the viscosities and densities of the fluids actually used. The viscosities were measured using a series of Cannon-Finske glass viscometers, which



Fig.2b. The effect of water content on the viscosity of glycerine (a different batch to that used for Figure 2a), measured at a constant temperature of 25°C.

				Lower Li	ayer		Middle	Layer		Upper La	ıyer	
Expt.	Ŧ	Fig.	ц.	ь Г	, L	$_{\rm M}^{\rm T}$	P _M	× ₩ 1	ч П	و_ U	ر ا _	$v_{\mathbf{U}}^{/v_{\mathbf{L}}}$
No.	Type of Experiment	Nos.	(°C)	(kg m ⁻³)	(m ² s ⁻¹ ×10 ⁴)	(°C)	(kg m)	(m^s_x10)	(ວຸ)	(kg m ³)	(m's *10 ⁷)	ĺ
3.1	Two layers, KNO ₃ below glycerine	e	61	1287	0.0054				12	1260	16	3000
3.2a	Two layers, heated NaNO ₃	Бa	9	1271	0.021				8	1266	23	1100
3 Jh	below glycerine Two lawers cooling and	ł	٥۶	1 264					55	1249	2.5	
	crystallizing (Na,K) ₂ CO ₃ below diverine	5	2						}			
3.3a	Two layers, KNO, below a	7	62	1287	0.0054				13	1258	0.11	20
	mixture of NaNO ³ solution											
чс чс	and yrycernie Tho lawers KNO helow a		60	1787	0 0054				13	1262	0.47	06
	mixture of sugar solution		2)) {	,	1
	and glycerine											
3 . 3c	Two layers, KNO, below a	8	60	1287	0.0054				12	1258	7.3	1300
	mixture of sugar solution											
	and glycerine											
3.4a and	Two viscous layers, KNO ₂ plus		71	1258	0.014				15	1230	0.072	ъ
3.4b	glycerine below NaNO ₃ pľus		72	1235	0.05				14	1229	0.40	ω
	glycerine											
3.5	Gas release: KNO ₃ plus HNO ₃ ,	ი	65	1307	0.0059	8	1263	23	ω	1256	3.7	630
	below glycerine plus K ₂ CO ₃ with huffer laver of clycerine											
3.6	Two immiscible fluids: KNO	10	60	1287	0.0054				11	1247		
1	below mixture of kerosene and	ı	ı	I								
	trichlorethylene											
4a-4d	Three layer systems:	11	62	1287	0.0054	12	1260	16	12	1249	9.1	11*
	KNO ₃ solution below a layer	<u>1</u> 2	60	1287	0.0054	12	1260	16	15	1237	3.0	23*
	of glycerine, with glycerine		63	1287	0.0054	12	1260	16	12	1226	1.7	34*
	diluted with water on top		63	1287	0.0054	11	1260	16	11	1216	1.1	44*
5.1	Stratified system: KNO, below	13	62	1287	0.0054	13	1260	14.6	13	1220	1.43	40*
	glycerine, grading upwärd to											
	glycerine plus water at top											
5.2	Stratified system: KNO ₃ below	14	72	1363	0.0057	ъ	1328	0.026	ഹ	1272	4.1	56*
	NaNO ₃ grading upward to											
	glycerine plus some NaNO ₃											
	at top											

The compositions and measured physical properties (temperature, density and kinematic viscosity) of the several layers (or of the gradient region) are recorded. Experiment numbers have been chosen to correspond to the sections in which the experiments are described, and the corresponding figures (where photographs of each experiment are shown) are also indicated. In experiments are described variable, and the corresponding figures the figures the experiment of the variable of each experiment are shown) are also indicated.

layer * p_M - p_U in kg m⁻³.

Table 1. Initial Conditions for Experiments Described in the Text.

were held in a water bath whose temperature could be accurately controlled. The density was determined using a set of hydrometer floats. For pure glycerine it is well represented by $\rho = 1269 (1-5.64 \times 10^{-1} T) \text{ kg m}^3$, where T is the temperature in °C. For glycerine-water mixtures at 25°C it can be represented by $\rho = 1250$ $(1-0.156v) \text{ kg m}^3$, where v is the volume fraction of water.

Some 35 experiments were carried out in the standard tank, and the experimental conditions for those discussed in this paper are summarized in Table 1. In addition, we performed many exploratory experiments before deciding on the optimum techniques, and various other runs to answer specific questions; these do not all appear in the table but are described at appropriate points in the text.

3. Two-Layer Systems

3.1 Very Viscous Upper Layer

We begin by describing experiment 3.1 as given in Table 1. This was a typical experimental run in which the viscosity ratio was at its maximum (\approx 3000 at the beginning of the experiment) and for which pure glycerine was used for the upper layer. The temperature of the upper viscous layer was 12°C, and the other conditions are set out in the table. Essentially the same behavior was observed in other runs under nearly the same conditions; time-lapse movies were made of several of them, and these helped greatly in arriving at the following interpretation.

Immediately after the cold glycerine had spread over the top of the lower hot KNO₂ solution, crystallization began at the interface, particularly along the edges of the lower containing box where there were nucleation sites. Strong thermal convection was also observed in the lower layer, with crystallization occurring at the bottom. Thus compositional convection also became significant in that layer after a few minutes. The most striking difference from the earlier, constant viscosity, experiments was that with a viscous upper layer, less dense fluid was released immediately and continuously from the interface and rose as plumes into the upper layer (see Figure 3). These plumes had a mushroom shaped cap with a thin stalk behind, and the stalks were sometimes interconnected with one another in the form of thin two-dimensional sheets. A more detailed description and interpretation of their behavior is given in the following section. Many of these plumes ori-ginated at the edges where crystals first appeared, but some certainly formed at the interface well away from the sides. The overall behavior was not in fact significantly different in the one "control" experiment we carried out in the larger tank, in which most of the plumes rose from the interface in the interior of the tank and not from the edges of the containing box.

The first plumes showed no sign of crystallization as they ascended, but as the rate of crystallization in the lower layer increased, the rate of release of KNO₃ solution into the upper layer also increased, and crystals formed in the plumes. These were initially carried upward and continued to grow as the solution was cooled, but they eventually became detached from the parent plumes and settled back to the base of the container. Sometimes a long continuous framework of crystals formed in the plume: this sank slowly through the glycerine in one piece, as shown in Figure 3b. The residual (dyed) fluid in the plumes continued to rise, mixing very little with the glycerine and being deposited in a layer right at the top of the tank. At this stage the fronts of the plumes tended to become more compact and pear shaped, rather than thin sheets spread out into mushrooms. Some crystals continued to form in the layer at the top of the tank and gradually fell out.

The release of fluid into the upper layer was somewhat inhibited by the formation of a framework of crystals capping the lower layer, and the exact sequence of events observed in the individual experiments depended on when and where the lower fluid was released through this. The interface between the two layers sank below the level of the capping crystals, and sometimes clear patches of lower-layer fluid remained trapped for some time. The final result was the same, however: the rising plumes deposited a layer of almost unmixed dyed residual KNO, solution, with excess temperature and water concentration with respect to the surroundings, at the top above the viscous glycerine layer. Heat and water were at the same time transferred by horizontal diffusion to the surrounding glycerine, resulting in a decrease in density and, more significantly, in viscosity. The position of a plume was thus stabilized by formation of a local minimum in the viscosity of the fluid surrounding it. A mat of crystals, consisting partly of those grown in situ and partly of those that had settled out, was left at the base. Compositional "fingers" formed below the upper layer, and on a longer time scale these penetrated downward into the clear glycerine. (The mechanism of their formation differs somewhat from that previously documented, and this will be discussed in the next section.) Slow convection in the form of a single cell was frequently visible in the glycerine layer for several days, and this was probably driven by the buoyancy flux through the finger interface above. Some re-solution of crystals formed a denser lower layer at the bottom of the container, with a sharp interface between it and the overlying glycerine.

These experiments suggest that it is instructive to consider separately the processes of formation of convective plumes at an interface, and their rise through the more viscous layer above. We must first explain the nearly continuous release of buoyant fluid in the present experiments. In common with the experiments using layers of nearly equal viscosity, there is the production of less dense residual fluid, which is released as a result of crystallization. This may be formed by bottom crystallization, so that it mixes through the lower layer, or at the interface, where it may rise immediately into the upper layer. The new feature when there is a more viscous upper layer is the systematic accumulation of buoyant fluid just below the interface, and we suggest that this occurs in the following way.

Continuity of stress across the interface implues that a very viscous upper layer acts like



Fig.3d

Fig.3. Experiment 3.1. Convection in a two-layer crystallizing system with large viscosity ratio (approximately 3000), starting with hot KNO₃ solution below cold glycerine. (a) At 9 min 45 s (photographs with diffuse back lighting). Plumes with mushroom-shaped caps are rising into the upper layer. (b) At 18 min 25 s, crystals are forming in the plumes and dropping back. (c) At 33 min, a layer of residual fluid is forming at the top. (d) At 45 min, plumes continue to rise, with further crystals falling out. (e) At 1 hour, the plume activity has almost ceased, and a deep layer of residual fluid has built up at the top. (f) At 18 hours 18 min (shadowgraph), note the formation of fingers in and below the upper layer.



Fig.3e

Fig. 3. (continued)

Fig.3f

a nearly rigid lid on the lower layer and that, in particular, it causes the velocity to be very small there. Buoyant fluid formed as a result of cooling from above and consequent crystallization near the interface is not swept away by convection in the lower layer but can build up in the nearly stationary boundary layer below the interface, much as it does below a cooled solid lid at which crystallization is taking place [Chen and Turner, 1980]. The accumulation of buoyant fluid will also be facilitated if crystals grow through the interface and tend to stick there. At the same time, slow convergent flows of the form sketched in Figure 4 will be set up in the warm, somewhat less viscous boundary layer in the upper fluid. These flows will separate and break away to form vertically moving channels or plumes, at the base of which the interface will be raised. The lightest, most depleted KNO3 solution in the lower layer will tend to collect under these plumes, and as soon as it is sufficiently buoyant, it will convect upward across the interface to form the cores of these sheet-like ascending plumes. These will continue to rise with a spacing determined by the combined boundary layer itself, but uninfluenced by the depth of the upper layer. Once a low-viscosity channel is established, both by heating the upper fluid and the addition of fluid from the lower layer, buoyant fluid produced subsequently will more readily follow this path, and the plume will persist. Both the interfacial heat flux and the production of light fluid by crystallization will be increased in the convergent flows feeding into the plumes.

Further experiments, designed to shed more light on the processes just described, and the subsequent behavior of the convection in the upper layer, are reported in the following section. 3.2 <u>Subsidiary Experiments with a Viscous Upper</u> Laver

To clarify some of the points raised above, it seemed desirable to separate the process of crystallization from that of the release of less dense fluid at an interface (between a very viscous upper layer and an aqueous lower layer). Accordingly, we built a tank, 200 x 106 x 300 mm deep, containing a coil that could be used to heat or cool the lower layer, independently of any fluxes through the interface.

In the first such experiment, 3.2a, the upper layer was pure glycerine at 8°C and had a specific gravity, S.G., of 1.625 and the lower layer, 50 mm deep, was NaNO₂ solution, initially



IN LOWER LESS VISCOUS LAYER

Fig.4. Sketch of the inferred flow in the boundary layer at the bottom of the upper glycerine layer, and the formation of thin plumes of buoyant, less viscous fluid transported across the interface. The velocities are all much smaller than the convective velocities in the lowviscosity lower layer.



Fig.5a. Experiment 3.2a. Interfacial transport produced by heating a layer of less viscous fluid below glycerine. Note the plumes with thin stalks feeding into the mushroom-shaped caps.

at 5.5°C and S.G. 1.271. Heating through a coil in which the temperature of the circulating water was gradually increased, produced a lighter layer of warmer solution at the top of the lower layer. This in turn heated the boundary layer in the glycerine above, and led to convection in the form of two-dimensional sheets, in which a thin core of lower-layer fluid was incorporated (see Figure 5). The front of the thin rising columns had a mushroom shape, as described for the early part of our experiment in the previous section. The decrease in viscosity due to both heating and dilution of the glycerine seemed to be the dominant process here, with a thin rapid upflow feeding into a much wider cap. The form of motion observed is similar to that described by Whitehead and Luther [1975] for the case of a plume of less viscous fluid released slowly and steadily into a more viscous layer. These authors also comment on the persistence of the following "stalk", which is the path of least resistance through the surrounding very viscous fluid; even if the upward flow stops for a while, it can readily become reestablished in these previous channels.

Returning to the present experiments, we note that the overturning due to heating from below was not complete, since only the upper part of the lower aqueous layer was buoyant enough to rise through the upper layer. Less fluid was deposited at the top than in the experiment with crystallization because the change in density in this experiment depended only on temperature and not on composition, and the density difference between the rising elements and their surroundings decreased with height due to the lateral heat flux.

A second experiment, 3.2b, examined the effect on convection of a change in composition. The upper layer was glycerine of S.G. 1.249 at 33°C, and the lower layer (60 mm deep) a mixture of Na CO_3 and K CO_3 , S.G. 1.264 at 29.5°C, designed to saturate at about room temperature. Circulation of coolant at -3°C produced crystallization round the cooling coils, and a decrease of density of the lower layer. Compositional plumes, containing dyed lower-layer fluid, were rising freely into the glycerine after 21/2 hours, when the temperature of the lower layer was 16°C and the upper 28°C. These upward moving regions were more nearly circular than the thermal plumes and much more compact, suggesting that this shape is characteristic of compositional convection, when there is a smaller variation of temperature and hence viscosity in the surrounding glycerine. They deposited much more of the lower-layer fluid at the top of the tank than did the sheet-like thermal plumes, and again (as in the crystallization experiment), "fingers" grew between this aqueous solution and the glycerine layer below it.

The mechanism of formation of fingers in these experiments is sufficiently different from that previously studied (and reviewed in Huppert and Turner [1981b]) for it to merit a more detailed investigation. Accordingly, we set up a lower



Fig.5b. Experiment 3.2b. Compositional convection produced by cooling and crystallization in an aqueous layer below cold glycerine.



Fig.6a

Fig.6b

Fig.6. Two stages in the formation of fingers at an interface between dyed NaNO₃ solution above pure glycerine at room temperature. (a) At 15 min, fingers drop through the lower layer. (b) At 26 hours 45 min, fingers have formed in a thickening interface and have also deposited upper-layer fluid at the bottom of the tank. This lower layer is gradually being eroded away by larger-scale convection driven by the buoyancy flux through the fingers.

layer of glycerine S.G. 1.252 at 24°C to a depth of 112 mm in a tank 50 x 150 x 250 mm deep, with above it a layer of NaNO₃, S.G. 1.206, of compar-able depth and at the same temperature, 24°C. Fingers rapidly formed at the interface, showing that thermal effects are certainly not important (see Figure 6). We suggest that the formation of fingers in this case is due to the destabilizing influence of water and glycerine interdiffusion acting against the stabilizing influence of the diffusion of NaNO₃. Consider a parcel of pure glycerine displaced above the interface. Water diffuses into the glycerine of the parcel, while glycerine from the parcel diffuses into the aqueous environment at a rate approximately 40 times more rapidly [Miner and Dalton, 1953, p. 328]. These effects increase the volume concentration of water in the upward displaced parcel and hence make it less dense. The diffusion of NaNO, into the parcel increases its density. The net effect of these two diffusion processes depends on the density of the upper layer. If it is sufficiently high, i.e., the NaNO₃ concentration is beyond a certain limit, diffusion makes the parcel less dense than its surroundings and it continues to rise. If the upper layer concentration is below this limit, the parcel returns to the lower layer and the two-layer system is stable. A similar argument holds for a parcel of aqueous NaNO₃ displaced below the interface. Experiments indicate that fingering occurs if the upper layer density exceeds 1130 kg m⁻³. In the experiments, the upward and downward fingers extended at comparable rates, thought the convection in the well-mixed layer above the fingers was much more vigorous than that in the lower layer. This indicates the difference between mixing a lowviscosity fluid into a high-viscosity one and vice versa.

3.3 <u>Variation of Viscosity Ratio</u>

After observing the dramatically different influence of a large viscosity ratio on the system described above, we carried out a number of experiments at intermediate values of the upper viscosity. Three of these, with starting conditions listed in Table 1, will be described in turn, and photographs of two of them will also be shown. Note that the initial densities and temperatures of both the layers were kept within narrow ranges for all the experiments, only the upper viscosity being changed by varying the composition as described in section 2. Experiment 3.3a, in which the upper viscosity was a factor of 20 greater than that of the lower layer, evolved in a similar manner to the case of two aqueous solutions with comparable viscosities [Ruppert and Turner, 1981a]. As the density difference decreased, however, pronounced "ridging" occurred at the interface; the thermals that rose into the upper layer were thinner, but nevertheless some lower-layer fluid was taken up by them (see Figure 7). When overturning occurred, the mixing between the two fluids was delayed and appeared more dramatic because of the greater spatial variability in composition that took longer to be made completely uniform. A small vertical stratification remained at the end of the experiment.

The next case, 3.3b, having a viscosity ratio of 90, exhibited features more obviously intermediate between the two extremes. Thermal plumes formed in the upper layer at an earlier stage and began to incorporate some of the liquid released by crystallization. Ridges formed at the interface, and two-dimensional sheets rose out of the interface. Very fine crystals formed in these sheets as they cooled, and then dropped back. Following substantial crystallization in the lower layer, in the form of dendrites that did not appear to be influenced by the presence of glycerine, the interface broke down and the two layers mixed to some extent, though a layer of the released light fluid formed and remained at the top.

In experiment 3.3c, with a viscosity ratio of 1300 at the initial temperature of the experiment, ridges formed at the interface and thermals containing lower-layer fluid began to rise almost immediately. Crystals formed in these plumes, and a layer of residual fluid built up at the top, as shown in Figure 8, with the formation of fingers below it. The crystals fell out of the plumes more rapidly than they did in the experiments with pure glycerine in the upper layer, presumably because of the lower viscosity of the fluid through which they were falling, and there was more mixing of the plumes with their environment on the way up.

3.4 Both Lavers With Increased Viscosity

In order to assess the importance of the viscosity ratio between the two layers, compared with an increase in both viscosities (and a consequent reduction in the Rayleigh number), we carried out two experiments (3.4a and 3.4b), in which glycerine was added to both layers. The viscosity of each was increased by about the same factor compared with the aqueous case, approximately 4 and 20 in the two experiments.

The behavior was similar to that in the aqueous experiments, except that everything proceeded more slowly as the viscosity was increased. The convection was much less vigorous, and hence the heat transfer and rate of crystallization of KNO in the lower layer were reduced. The viscous coupling across the interface was also more prominent, with convective motions on one side clearly affecting the flow of fluid on the other toward a convergence line, and it remained symmetrical in the two layers with plumes rising and falling away from the interface at the same places. When the density difference decreased to zero, overturning occurred, with intimate mixing but at a slower rate. These observations confirm that it is the viscosity gradient across the interface that causes the striking difference in behavior documented in sections 3.1 and 3.3.

3.5 Overturning With Gas Release

The techniques previously used to model the release of volatiles following an overturning event were adapted to investigate the effect of an increased viscosity in the upper layer. Two experiments were conducted, and we describe only the one (3.5) that was (marginally) more success-ful.

The lower layer was a hot 41 wt % KNO₃ solution with a volume just sufficient to fill the lower box, to which 0.7 mol of 70% HNO₃ was added. A buffer layer consisting of 1 L of pure glycerine was placed above this, to keep the acid separated from the deep upper layer during filling. The upper layer consisted of 2.91 kg of pure glycerine, with 1.5 mol of K_2CO_3 dissolved in 0.70 kg of water. The top layer was at 8.4°C and S.G. 1.256, with viscosity 3.65 x 10 m s⁻¹ at 8.4°C.

Cooling through the interface produced crystallization in the lower layer, releasing light fluid into the buffer zone. Gradually this penetrated into the upper layer, and after some 20 min the reaction between the acid and the carbonate released gas bubbles, which contributed greatly to the buoyancy of the thermals and the vigor of the convection and mixing (see Figure 9). A mat of crystals formed over the lower layer at the level of the containing box and slowed up the release of lower fluid, and hence the reaction and release of gas. Small bubbles were trapped below the crystals and accumulated to form larger ones, which caused considerable stirring as they escaped and rose through the upper layer. Vigorous gas release continued for about 30 min after it began, but stopped rather rapidly 70 min from the beginning of the experiment. Crystals continued to grow both in the lower layer and in the rising plumes during this time. Some released fluid was deposited at the top, but not as much as in the viscous experiments without bubble formation, because of the mixing generated by the gas release.

3.6 Immiscible Fluids

For comparison with the cases already discussed, we carried out an experiment (3.6) using immiscible fluids: hot KNO₃ below a cold mixture of trichlorethylene and kerosene, a mixture that could be adjusted to have a density somewhat less than that of the lower layer.

Not surprisingly, surface tension effects dominated in this case. As crystallization reduced the density of the lower-layer fluid, large rounded blobs formed, escaped through the interface, and rose to the top of the tank (see Figure 10). They gradually built up a layer of depleted fluid at the top, in which further crystallization continued. Some of the crystals migrated to the interface at the base of this upper layer, and intermittently fell out and dropped to the bottom of the tank. After several hours, most of the dyed lower fluid escaped from the crystal



Pig.7c

Fig.7d

Fig.7. Experiment 3.3a. Convective overturning produced by crystallization, with a viscosity ratio of 20. (a) At 13 min, crystallization occurs at the bottom of the lower layer only, and thermal plumes rise in the upper layer. (b) At 16 min, sudden overturning takes place as the density of the lower-layer fluid becomes equal to that of the upper layer. (c) At 16 min 45 s, mixing takes place more slowly than with equal viscosities in the two layers. (d) At 20 min, mixing is now nearly complete.



Fig.8c

Fig.8d

Fig.8. Experiment 3.3c. Convection in a two-layer crystallizing system with a viscosity ratio of 1300. (a) At 9 min 09 s, plumes are formed by crystallization at the interface. Further crystals form in the plumes and fall out. (b) At 23 min, the rate of crystallization in the plumes has increased markedly. (c) At 36 min, a layer of residual lower-layer fluid (dyed KNO₃) has been deposited at the top. (d) At 2 hours 56 min 20 s (shadowgraph), note the dyed layer of KNO₃ deposited at the top, with fingers below it, and the large-scale convection in the viscous layer.



Fig.9a

Fig.9b

Fig.9. Experiment 3.5. Convection in a crystallizing system having a large viscosity ratio, with gas release following mixing. A buffer layer of pure glycerine initially separates the two reacting layers. (a) At 16 min, crystallization near the interface produces plumes that pull lower-layer fluid upward. A reaction leading to gas release, and further crystallization, then occurs in the upper layer. (b) At 21 min 30 s, the release of gas causes much more mixing in the upper viscous layer, and less residual fluid is deposited at the top.

layer on the bottom, to be replaced by fluid from the upper layer, but some of it was retained in the interstices between the crystals.

In some respects these results are similar to those with a very viscous upper layer, in that the depleted residual fluid is deposited at the top, with little mixing with the intervening layer, although the form of the convecting elements is different. Perhaps the more surprising result is that a large viscosity ratio can be almost as effective a barrier to mixing as complete fluid "immiscibility".

4. Three-Layer Systems: Two Viscous Upper Layers

In order to shed further light on the relative importance of interfacial effects, and the later rise of convective elements through the viscous layer, we conducted a series of experiments with three layers under the following conditions. The lower layer was 41 wt % KNO₂, as for the "viscous upper layer" experiment, placed in the lower box at ~ 61°C. Above this was a cold layer of pure glycerine l25 mm deep, and above this again a layer, also 125 mm deep, of glycerine diluted with water to produce density differences, $\Delta\rho$, compared with the middle layer, ranging (in different experiments) from 1.1% to 4.4%. The viscosity of the upper layer therefore also decreased systematically, in a manner that was closely related to the dilution and the density.

We can regard the single upper layer glycerine experiments as being at one end of the range (with zero density difference between the upper layers) and experiment 4d with $\Delta \rho = 44$ kg m at the other. In both, the motions were virtually the same, as described in section 3.1, except that in 4d the flow was confined to half the depth, with an exceedingly small amount of the lower fluid penetrating into the top layer. Some weak thermal plumes did rise above the interface, but the lower dyed fluid built up into a layer that remained below the interface, i.e., this acted like a lid or free surface. The similarity of the behavior in these two cases is consistent with the notion that depth, and indirectly the overall Rayleigh number, has no quantitative influence on the motion over the range of conditions considered.

In fact for all of these experiments, the evolution of the lower KNO₃ layer as it crystallized, and the motions in the lower part of the pure glycerine layer above it, were virtually identical. The differences appeared at the interface between the two upper layers. Plumes that initially propagated freely through the middle glycerine layer ponded at the interface with the upper layer. With the smallest interfacial density difference ($\Delta \rho = 11$ kg m⁻³ in experiment 4a), the interface bulged upward, and as further



Fig.10a

Fig.10b

Fig.10. Experiment 3.6. Convection with hot crystallizing KNO_3 in the lower layer and a cold immiscible upper layer. (a) At 29 min 08 s, large rounded elements of depleted KNO_3 break through the interface and rise through the upper layer. (b) At 1 hour 50 min³ 30 s, eventually most of the depleted fluid is deposited at the top, and a crystal matrix remains at the bottom.

crystallization took place, even lighter residual fluid was released, which rose right to the top of the tank (Figure 11). Eventually, plumes extended right through the depth with not much, if any, deceleration, and most of the depleted lower-layer fluid accumulated at the top, with very little at the interface. The level of the interface between the two glycerine layers dropped as released fluid poured past it and carried fluid from the middle layer up with it. We noted also that there were fewer crystals in the upper layer in this case, presumably since the major cooling and crystallization had already taken place in the middle glycerine layer.

For the experiment with the next larger density step ($\Delta \rho = 23$ kg m⁻³ in experiment 4b, for which a movie was also made) the behavior was already very different. In this case most of the fluid released from the bottom KNO₃ layer accumulated at the interface, with only a small fraction penetrating to the top of the upper layer (see Figure 12). In experiment 4c with $\Delta \rho = 34$ kg m⁻³, virtually no released fluid came through to the upper layer. In all cases the bottom box was eventually drained, and the layer of released fluid formed fingers, which in time propagated throughout the layer below it (whether this was at the top of the tank or below the interface).

The behavior of the aqueous plumes in the uppermost layer varied systematically in a manner that can be understood, at least qualitatively, in terms of the variation of both density and viscosity in that layer. The nearly spherical fronts ahead of the much thinner plumes tended to be largest in the single upper glycerine layer experiments, and decreased in size progressively as the density and viscosity of the upper layer decreased. It is worth noting that this is consistent with the simplified theory given by Whitehead and Luther [1975] for the slow rise of a "dome" of less viscous fluid in viscous surroundings, though it would take us too far from our present theme to pursue this comparison in detail here.

> 5. Upper-Layer with a Density and Viscosity Gradient

5.1 Viscosity Decreasing with Height

The next experiment to be described is a natural extension of those with two viscous layers above the crystallizing solution, except that now we set up a linear gradient of density ($\Delta \rho$ about 3% top to bottom) instead of an abrupt step. The tank was filled using the special two-bucket device designed for use with viscous



Fig.lla



Fig.ll. Experiment 4a. Convection in a crystallizing system, with a hot KNO₃ layer below two cold viscous layers having a small density difference ($\Delta \rho = 11$ kg m³) between them. (a) At 20 min, plumes produced by crystallization at the lower interface rise through the lower layer and, after further crystallization, continue to the top of the tank. (b) At 47 min 30 sec, when crystallization is nearly complete most of the residual fluid from the lower layer has been deposited at the top of the tank. Note that the middle interface has fallen as a result of the upward transport.

fluids, which is sketched in Figure 1. Pure glycerine was placed in the stirred container, which led directly to the filling tube, and glycerine plus water (at the same low temperature) in the second identical container connected to it. We should note that while the water content and density variations produced in this way were linear with height, the viscosity variation was very nonlinear, as can be seen from Figure 2b.

The lower layer of hot crystallizing KNO₃ had the same properties as before, and just above it was pure glycerine at 13°C. Thus it is not surprising that the initial behavior, as crystallization began in the lower layer and near the interface, was similar to that with a large viscosity ratio and an unstratified viscous fluid above. Plumes of released fluid began to rise almost from the start, with mushroom-shaped caps much larger than the following channels. The ascent of these plumes was, however, halted by the stable density gradient. A layer of mixed fluid built up at the base of the gradient region, and further layers developed above it, driven partly by compositional differences, and partly thermally (see Figure 13). These layers were very inhomogeneous in the horizontal, reflecting the history of the plumes responsible for their formation. As more fluid escaped from the lower layer and further crystallization occurred, the plumes extended upward again, to produce more convecting layers that gradually became nearly horizontally homogeneous. At much later times a series of slowly evolving "compositional" layers was observed, with diffusive interfaces between them. The mechanism whereby these were maintained is possibly the inverse of that proposed for the fingers described in section 3.2, which depends on the different rates of diffusion of water into glycerine and glycerine into water.

5.2 <u>Viscosity Increasing With Height</u>

By using a hotter and denser KNO_3 solution in the lower layer, we were above to set up a stratified system in which the density gradient in



Fig.12a

Fig.12b

Fig.12. Experiment 4b. A similar three-layer experiment to 4a, except that the density difference between the two upper viscous layers is larger ($\Delta \rho = 23$ kg m). (a) At 17 min (diffuse back lighting), plumes produced by crystallization at the lower interface rise through the lower layer. Most residual fluid is accumulating at the middle interface, but some KNO rises through it to the top of the tank. (b) At 52 min (shadowgraph), note again, at this later stage, the partial penetration of the middle interface by the residual fluid.

the upper fluid, and the density step across the interface, were the same as in the preceding case, but in which the viscosity increased toward the top of the tank. The fluid feeding into the top of the gradient was pure glycerine, and that at the bottom aqueous NaNO solution, both at 5°C before filling using the double-bucket technique.

In this case, the fluid immediately in contact with the crystallizing KNO₃ layer was an aqueous solution of comparable viscosity. The early evolution of the system was similar to that described by Huppert et al. [1982b] for an experiment with a gradient of entirely aqueous solution above the lower layer. This shows again that the interfacial processes are the most important in determining the motion in the upper fluid.

Thermal convection, driven by cooling across the interface, was visible in the lower layer from the beginning, and this was soon enhanced by crystallization and compositional convection from the bottom. Heat transferred to the upper, compositionally stratified region produced rising thermals, but their rise was limited by the gra-

dient, and a convecting layer developed that slowly increased in depth. A second layer formed above this, as the thermal boundary layer at the top of the first NaNO layer became unstale. Up to this time, practically no KNO (or dye) was transferred upward, but when the density of the lowest (KNO,) layer reached that of the first convecting Jayer above it, a sudden overturning occurred, with rapid and thorough mixing through the lowest (NaNO₂) layer only (see Figure 14).

The further evolution of the system was much slower and was monitored over several days. Long after residual temperature differences had decayed by conduction through the side walls, convection in layers persisted. This must be a "compositional convection" effect, and a probable mechanism has already been mentioned in section 5.1. In the course of time, the series of layers extended upward (more and more slowly as the viscosity in the new layers increased). We note for future reference the potential application of these results to stratified magma chambers, which will often have a strong viscosity gradient in this sense, as well as density and composition gradients.



Fig.13a



Fig.13b



Fig.13c

Fig.13d

Fig.13. Experiment 5.1. Convection in a crystallizing system, with a cold fluid above having a gradient of density and viscosity decreasing with height. (a) At 10 min (diffuse back lighting), convecting plumes rise above the interface. (b) At 40 min (shadowgraph), the plumes are brought to rest and spread out in the density gradient. (c) At 1 hour 42 min, layers are forming in the released fluid. (d) At 1 hour 51 min 30 s, a further breakthrough occurs, and plumes of lower-layer fluid rise higher into the gradient region. Note the layers at the top of the tank, which are due to side-wall heating of the cold gradient.



Fig.14a

Fig.14b

Fig.14. Experiment 5.2. Convection in a crystallizing system, with a cold fluid above having a gradient of density and viscosity increasing with height. (a) At 12 min 30 s, heating of the aqueous solution above the interface produces a convecting layer, while there is cooling and crystallization in the lower layer. (b) At 16 min 50 s, the interface breaks down, and lower-layer fluid suddenly mixes upward into the first convecting layer at the bottom of the gradient region. Another convecting layer is beginning to form above. (c) At 1 hour, overturning is complete, but the residual fluid 1s still confined to the lowest convecting layer. (d) Nearly 5 days after the start of the experiment, compositional convection has gradually extended the series of convecting layers up into the more viscous fluid at the top of the gradient.

6. Some Geological Applications

The experiments provide further examples of the diversity of convective phenomena that can occur in crystallizing systems. They, together with earlier experimental studies, strongly suggest that crystallization and phase changes, rather than purely thermal effects, will be dominant in controlling convection in magma chambers. The experiments also show that viscosity variations will be important in determining how magmas can interact and mix together or separate from one another [McBirney, 1980].

A topic of recent petrological interest concerns the circumstances that allow basaltic magmas to mix with more differentiated magmas of higher viscosity. A substantial body of data and observations exists to support the view that mixing is a common phenomenon among calcalkaline magmas. In some cases, mixing is incomplete, with inclusions of mafic pumice being found within more silicic pumice and lava [Eichelberger, 1978, 1980; Sparks et al., 1977]. Compositionally banded pumice is also common [Smith 1979]. In other examples, mixing is more complete, and hybrids are often recognized during detailed petrological studies [for example, Anderson, 1976; Eichelberger, 1978; Luhr and Carmichael, 1980; Sakuyama, 1981; Gerlach and Grove, 1982].

Two models of mixing have been proposed, which involve the emplacement of wet mafic magma into the base of a high-level magma chamber containing more differentiated magma [Eichelberger, 1980; Huppert et al., 1982a]. Both models envisage the dense mafic magma initially forming a separate layer beneath the more differentiated magma. In the model of Huppert et al. [1982a] the mafic magma cools and crystallizes and becomes volatile saturated, allowing vapor bubbles to grow. Eventually the bulk density of the mafic magma becomes less than that of the overlying magma, and an abrupt overturn can occur, leading to intimate mixing of the magmas. Eichelberger [1980] envisaged that crystallization and bubble growth were restricted to the interfacial region between the magmas and that continuous release of vesiculated blobs and plumes of mafic magma



Fig. 14. (continued) Fig.14d

ascended and mixed into overlying magma.

The present experiments indicate that both these mechanisms are viable, and which occurs will depend, among other factors, on the viscosity differences between the layers. With a large viscosity contrast, the experiments have documented a continuous release of buoyant fluid in blobs and plumes induced by crystallization and viscous coupling at the interface. The mechanism is quite reminiscent of Eichelberger's model for mixing between rhyolite and basalt, and the experiments give support to his model as a mechanism for the formation of vesiculated mafic inclusions found in silicic volcanic rocks.

When the viscosity contrast between the layers is not as marked, the behavior could be closer to that envisaged by Huppert et al. [1982a], with an abrupt overturn leading to mixing. This model should be more applicable to mixing between magmas that do not differ greatly in viscosity, such as basaltic andesite and andesite.

There are many other situations in nature where magmas of contrasted viscosities are generated in the same system. Consequently, we suspect that there will be many applications of these experiments apart from those sketched here.

7. Discussion and Conclusions

The series of experiments described in this paper has produced a wide range of individual phenomena. Clearly, the basic concepts behind the observed motions should be explored more fully before attempting to apply the results in

specific geological contexts. We can, however, already make some important qualitative statements about the role of viscosity in layered convection with crystallization, based on the striking contrasts between our observations under different extreme conditions. For a layered system driven toward instability by cooling and crystallization in the lower layer, we have reached the following general conclusions:

1. It is the viscosity ratio between the two layers, not viscosity per se, that determines the form of the convection and breakdown.

2. A comparable increase in viscosity in both layers slows down the processes of heat transfer and crystallization as predicted by theory [Huppert and Sparks, 1980], but the motion remains symmetrical across the interface.

3. In the case of nearly equal viscosities, overturning and mixing occur when the density of the lower layer reaches that of the upper, and there is little compositional transfer until that stage.

4. With a large viscosity ratio (the larger viscosity being in the upper layer), residual fluid is released upward soon after crystallization begins, and the process is nearly continuous, with no sudden overturn.

5. In the latter case the transfer process is dominated by interfacial effects, with the upper layer acting as a nearly rigid lid that allows buoyant fluid formed by crystallization in the lower layer to accumulate below the interface and then rise across it into the viscous layer, rather than being swept away by unconstrained convective motions in the less viscous fluid.

6. Whatever the stratification, whether it be in layers or continuous, the form of the initial motion in the upper layer is determined by the viscosity ratio between the two fluids immediately adjacent to the interface.

7. The relative importance of thermal and compositional effects determines the later form of the rising plumes. Heating produces thin, less viscous upflows in the form of sheets, feeding into much wider mushroom-shaped caps. Compositionally lighter upflows of comparable viscosity are more compact and nearly cylindrical, with a rounded top.

8. A large viscosity ratio inhibits mixing between the rising elements and their surroundings and allows a layer of depleted fluid to be deposited at the top with practically no mixing.

As indicated briefly in the preceding section, the experiments are considered to be applicable to the understanding of mixing processes between magmas of contrasting viscosity. We have illustrated one situation (mixing of calcalkaline magmas) where these experiments give new insights into the important processes.

The experiments also indirectly raise some general questions that cannot yet be answered but seem worth more attention from geologists and fluid dynamicists: for example, what exactly is the mechanism whereby a large viscosity ratio inhibits mixing? What determines the time for which heterogeneities can survive in a stirred fluid of variable viscosity? What is the relative importance of diflusion and disruption by shearing motions? Is there a characteristic size of elements of a known viscosity, subject to specified motions in a fluid of different viscosity? Progress in this field will depend on the formulation of some explicit physical models, which can be tested quantitalively in the laboratory. The qualitative observations already made, and reported here, leave us in no doubt that further, more detailed research in this area could be very productive.

Acknowledgments. These experiments wore carried out during the latter half of 1982, while H.E.H. and R.S.J.S. were Visiting Fellows at the Research School of Earth Sciences, Australian National University. We are grateful for the financial support from the ANU and the Royal Society, which made the visit possible. We acknowledge the able assistance of Derek Corrigan, Joe Micallef, and Ross Wylde-Browne at all stages of the experimental work and in the preparation of the photographs and diagrams.

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(Received August 22, 1983; revised April 10, 1984; accepted April 11, 1984,)