4 March 1960, Volume 131, Number 3401 SCIENCE

CURRENT PROBLEMS IN RESEARCH

Granite Problems

The exploration of physical and chemical processes leads to a reorientation of our thinking.

Matt Walton

". . . it has the peculiarity of being elevated the highest . . . and sunk the deepest. . . ."—J. Playfair, *Illustrations of the Huttonian Theory of The Earth* (Edinburgh, 1802).

Near the end of the 18th century James Hutton developed the concept of a major genetic class of rocks formed in the plutonic depths of the crust by crystallization from a molten state. These plutonic rocks he grouped with volcanic rocks, which crystallize at or near the surface of the earth from molten lava, and he dubbed the combined class "igneous" rock to betoken the role of heat in creating their initial molten state. Veins of granite cutting layers of schist and marble at Glen Tilt in the Scottish Highlands served as Hutton's first type locality (*1*) for the igneous origin of rocks formed at depth in the crust; the transgressive relations of granite to stratified rocks (Fig. 1) and its coarse, compact, crystalline fabric were deemed proof of the origin by crystallization from "subterranean lava" or magma, as it has come to be known.

Since then the theory of the igneous origin of plutonic rocks has undergone many developments, some of which amount to revolutions in trends of thinking, above all with respect to granite. These major turns of thought have generally followed major advances in fundamental physical chemistry applied to rocks, or serious breakdowns of igneous theory applied in detail to actual geologic occurrences of supposedly igneous rocks. Right now we appear to be on the threshold of another major reorientation in our thinking, and it has been difficulties in applying igneous theory to granite that have, more than anything else, spurred the advances that are now in the making.

Granite Defined

Not the least aggravating of the problems connected with granite has been the question of what is meant by the name. As a formal term, *granite* applies to a rather restricted rock type, but geologists persistently use *granite* to identify an entire family of rocks which play certain complex roles in the architecture of the crust but which seem to elude the pigeonholes of formal nomenclature. Chayes (*2*, p. 66) who has made careful modal analyses of more rocks-called-granite than anyone else, has attempted to bring the term into line with the way geologists have actually used it by proposing: "The term *granite* should be reserved for massive or weakly oriented rocks in which C.I. [color index] $<$ 20 percent, and 20 percent $<$ quartz $<$ 40 percent; . . ."

Figure 2 shows the volume occupied by granite as defined by Chayes in the composition tetrahedron quartz–alkali feldspar–plagioclase–color index. This definition just about implies that granite is any coarse-grained, reasonably massive rock composed mainly of feldspar and quartz. It departs notably from the conventional systematic classifications by (i) placing no restrictions on the composition and relative proportions of the feldspars; (ii) limiting the modal content of dark minerals to 20 percent but not specifying any particular dark mineral assemblages; (iii) establishing relatively narrow limits to the quartz content instead of allowing quartz to range from near the lower limits of practical detectability to a major percentage of the rock; and (iv) avoiding any genetic implications. The primary justification for Chayes' unconventional definition is that it corresponds to the rocks which geologists have grouped in geologically significant units and have referred to broadly as granite.

Geology of Granite

I have implied that granite is in effect defined by its geologic relations, yet no rock is perhaps so varied in its geology. The thread which unifies the varied occurrences of granite is this: With trivial exceptions, granite is closely associated in time and space to mountain building and regional metamorphism in the so-called geosynclinal belts, where great thicknesses of sedimentary and volcanic rock accumulate. The process that forms mountain chains begins with the downbuckling of such belts, which brings the accumulated mass of sediments and volcanics to depths of tens of kilometers and subjects them to metamorphism under great heat and pressure. Uplift

---

The author is on the staff of the department of geology of Yale University, New Haven, Connecticut.

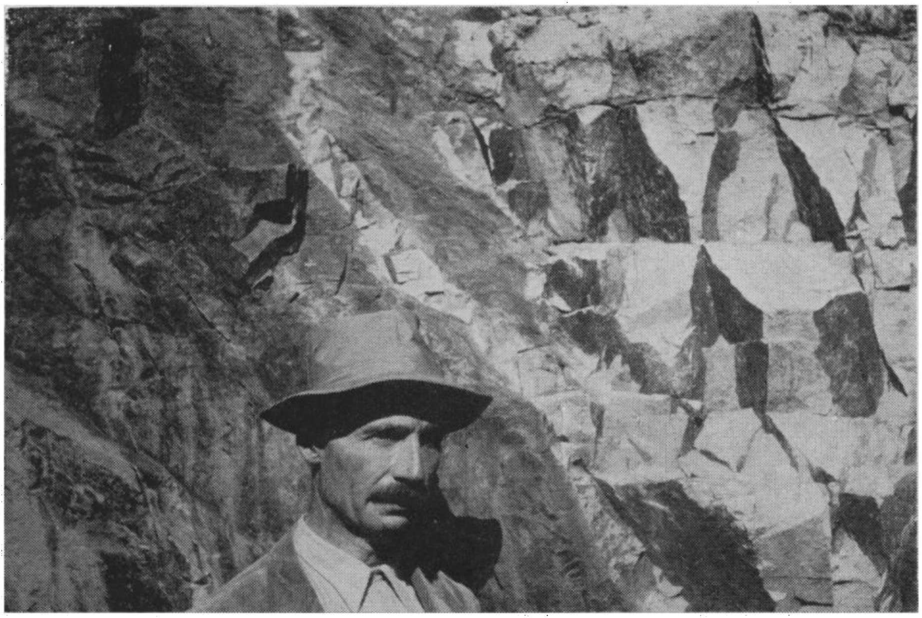

Fig. 1. Granite (light-colored rock with blocky jointing) of Hercynian age in sharp, transgressive contact with Cambrian sedimentary rocks. The granite is chilled to a fine-grained quartz porphyry at the contact, and the sediments are strongly modified to hornfels by heat in a distinct aureole around the granite—classic evidence for igneous origin. E. Raguin, noted French authority on granite (see *50*), stands at the outcrop at Mines d'Azegour, Morocco.

of the deformed and metamorphosed rocks ensues to form mountain chains, and wherever uplift has allowed erosion to bite deeply, great masses of granite are found to have been emplaced among deformed and metamorphosed sedimentary strata to form enormous granite bathyliths in the cores of major mountain ranges (Fig. 3). These are the largest nearly homogeneous bodies of rock accessible to man. Playfair had it backward; granite is sunk the deepest, then elevated the highest.

Extensive terrains of metamorphic rock in the continental shield areas, which are the roots of ancient mountains, are shot through and through with veins of granite or intimately impregnated with granitic materials to form migmatites or "mixed rocks" (Fig. 4), and in many places metamorphic rocks of obvious sedimentary origin pass gradationally through migmatite into granite. Other granite bodies are sharply transgressive and structurally discordant (Fig. 1).

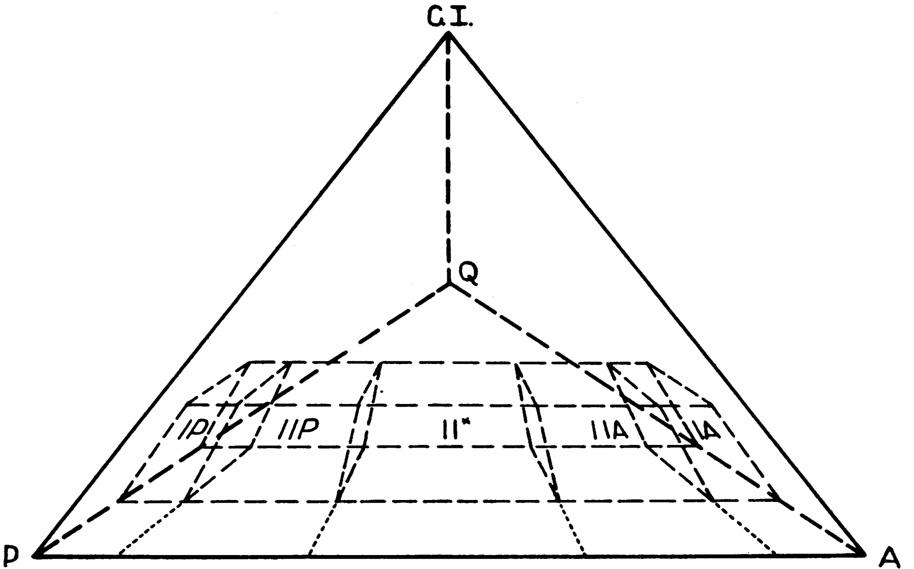

Fig. 2. The volume occupied by granite in the composition tetrahedron of the Chayes classification: *Q*, quartz; *P*, plagioclase feldspar; *A*, alkali (K-Na) feldspar; *C.I.*, color index—the percentage of dark minerals (ferromagnesian minerals) in the rock. Composition is in terms of modal volume percentages. The symbols in the granite volume designate the main classes of granite: *IP*, essentially one feldspar, plagioclase; *IIP*, two feldspars, plagioclase dominant; *II**, two feldspars, neither dominant; *IIA*, two feldspars, alkali feldspar dominant; *IA*, essentially one feldspar, alkali feldspar. [After Crayes (2)]

The great bulk of granite is emplaced at a depth and under conditions where metamorphism affects all rocks, but masses of granite of significant size and number reach levels of the crust where regional metamorphism is slight or even unnoticeable and are associated with volcanic rock of comparable composition. Such bodies are generally in less deeply eroded, younger, mountain chains and are believed to pass into metamorphic zones at depth. The relationship of granite to geosynclinal sedimentation and mountain building means that granite is almost unknown in the great ocean basins unaffected by epicontinental sedimentation. Basalt is the rock of the ocean basins.

"Classic" Igneous Theory

Early in the 19th century geologists began to develop the idea that the wide variety of igneous rocks evolves from a limited variety of primary magmas by chemical fractionation processes broadly referred to as magmatic differentiation. Field relationships suggested this even before the potentialities of chemical fractionation in the course of crystallization of complex solutions were generally recognized (*3*). Real progress began with insight into the theory of heterogeneous equilibrium and systematic experimental investigations of silicate melt crystallization.

The classic synthesis of experimental investigations in the crystallization of silicate melts and its application to natural associations of igneous rock was, and still remains, Norman L. Bowen's *The Evolution of the Igneous Rocks*, published in 1928 (*4*). Much experimental work has since been done, by Bowen himself in association with O. F. Tuttle (*5*), as well as by a number of other investigators, notably in systems involving high vapor pressures of water, a vital component of natural magmas at depth. Later work has only served to strengthen the main argument and to resolve some of the dilemmas on which Bowen could only speculate at the time.

Bowen developed in detail the application of the "Bowen reaction series" (Fig. 5) to the fractional crystallization and differentiation of rock magmas. He showed that, by the separation and reaction of appropriate liquid and crystal fractions at various stages in the crystallization of basaltic magma (basalt is the most common of all volcanic rocks), sequences of mineral assemblages can be obtained which correspond to most naturally occurring sequences of igneous rocks. He showed, moreover, that courses of crystallization in rock melts tend to produce residual liquids which approach compositions in the low-temperature region of "petrogeny's residua system"—$KAlSiO_4$-$NaAlSiO_4$-SiO_2 (Fig. 6)—at the heart of which is the "granite system," $KAlSi_3O_8(Or)$-$NaAlSi_3O_8(Ab)$-$SiO_2(Q)$ (*6*).

At this point igneous theory reached a certain classic purity and unity. The rocks preserved from earliest geologic time down to present active volcanism demonstrate that great quantities of basaltic magma are repeatedly, if not continuously, generated in the earth. There is no need to speculate on the origin of special magmas to account for special rock associations; the potentiality to produce them all exists in ordinary, ubiquitous basalt, or perhaps in two or three common types of basalt, and granite magma was presumably the residual liquid normally remaining from the fractionation of the common continental type of basalt. Moreover, it seemed attractive at that time, for various other reasons as well, to suppose that at the base of the crust a substratum of basalt existed in a vitreous, pressure-rigidified condition ready to erupt if tectonically disturbed (*7*). Nevertheless, igneous theory in this classic form presented such difficulties, especially for granite, that it rekindled a long-smouldering "granite problem."

Not that there was anything wrong with Bowen's chemistry or his application of it to the fractionation of basaltic magma; it was and will remain a keystone of petrology. But there was a crucial tacit assumption involved in tying igneous theory so rigidly to a single model in which the evolution of most igneous rocks is dependent on the upward intrusion of basaltic magma into a level of the crust where it cools, crystallizes, and fractionates. The same chemistry can be applied to other models. The question, to which American geologists especially were, in general, somewhat insensitive, was whether this magmatic differentiation model outlined an actual sequence of events which really operated in nature to produce granite. I recall the characteristically modest and graceful words with which Bowen declined to comment on a particularly baffling petrologic problem before a meeting of the Petrologists' Club held at U.S. Geological Survey headquarters in Washington. "After all," he said, "I only pretend to know

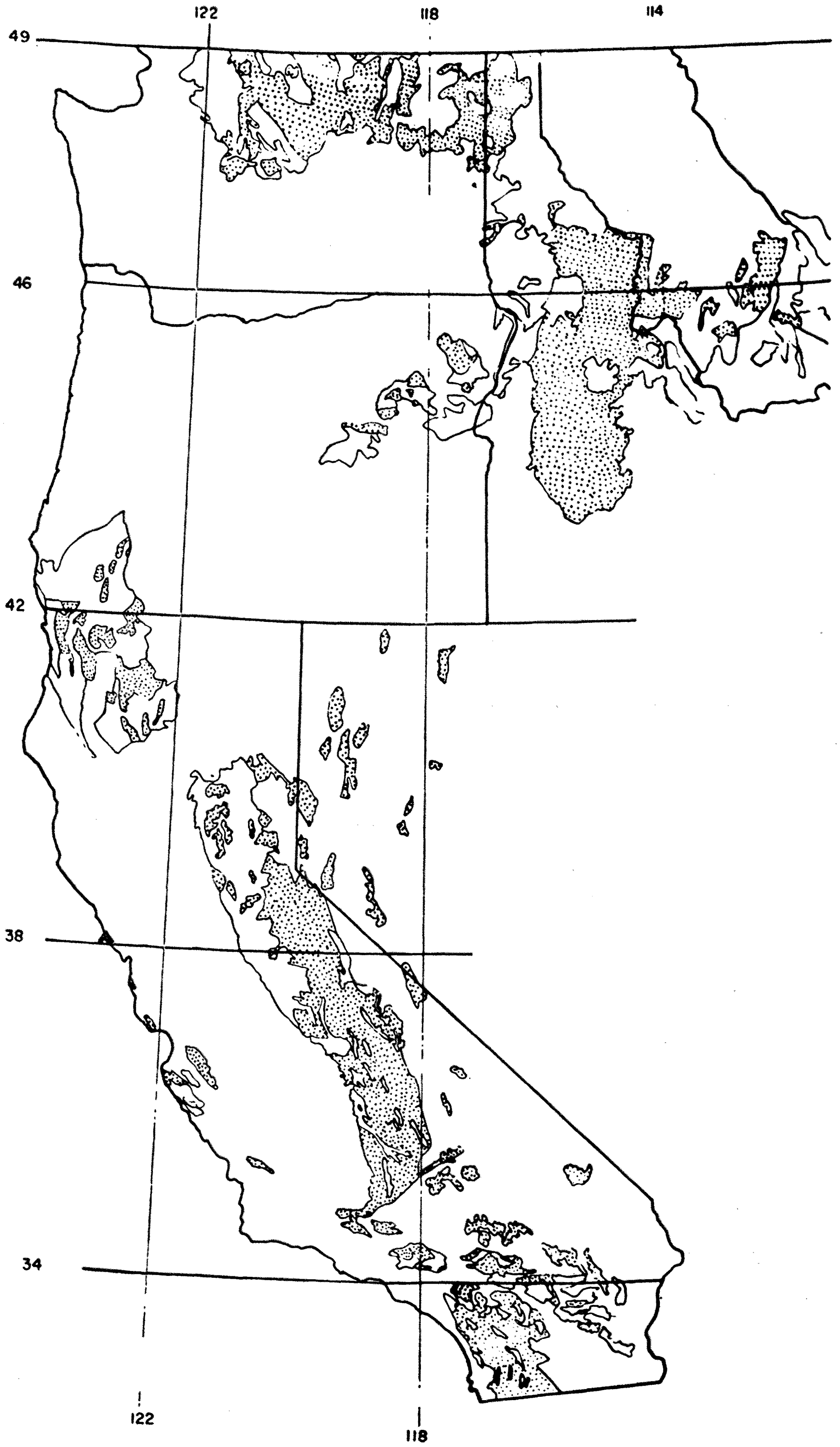

Fig. 3. Major bathyliths associated with Mesozoic and Tertiary mountain building in the far western United States. Granitic rocks are stippled, and associated areas of deformed and metamorphosed sedimentary rocks are outlined. [Data from Am. Assoc. Petrol. Geologists, *Tectonic Map of the United States* (*51*)]

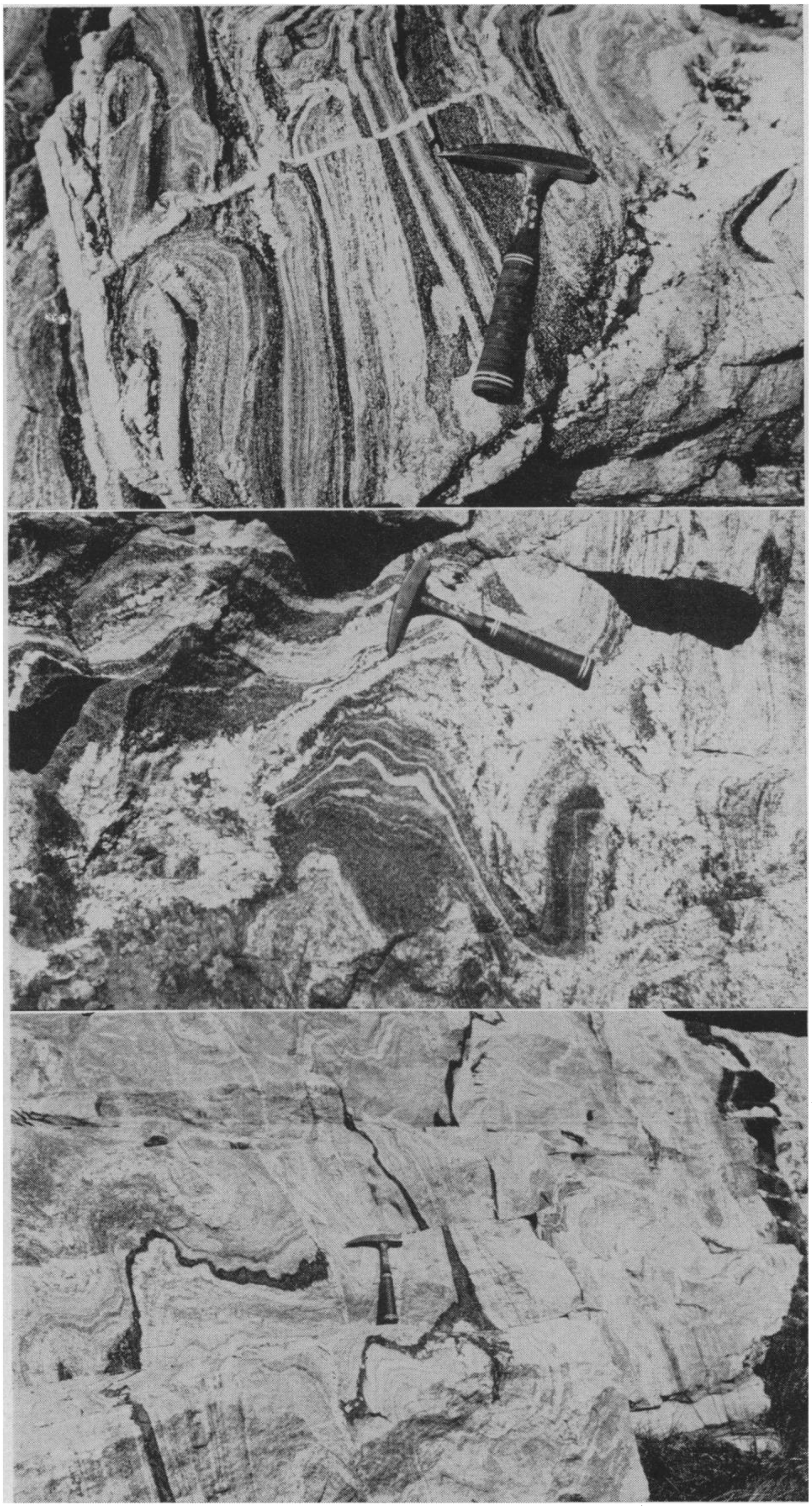

Fig. 4. Migmatite: (Top) Feldspathic schist (darker rock) interlayered with thin sheets of granitic material (lighter gray) and transected by granitic pegmatite dikes (coarse-grained, white). (Middle) Schist intimately impregnated with granitic material and passing into a granite gneiss, cut by irregular granite pegmatite dikes. (Bottom) Gneissic granite filled with dark, micaceous relics of schist in irregular streaks and wisps. "Mamacoke gneiss,"[9] southeastern Connecticut. [Lawrence Ashmead]

a little about rocks as they should be, and you are all experts on rocks as they are."

The Trouble with Igneous Theory

There were several major difficulties with the theory when applied to "rocks as they are." First of all, there is a disappointing lack of clear-cut, uncomplicated field examples to show that simple fractional crystallization normally produces granitic sequences of rocks (Fig. 7). Second, the theory offers no particular explanation of why granite should be so closely tied to orogeny and metamorphism and insignificant in other igneous environments, yet this is the crucial fact in the geology of granite. Third, to produce a quantity of granite requires the complete fractionation of a huge quantity of basaltic magma—a process for which there is little, if any, direct evidence—not to mention a staggering amount of dislocation to move this material into place. Finally, there is the rock against which magmatic theory has repeatedly foundered—migmatite (the "mixed rocks" of metamorphic terrains).

Granitization

The trouble with migmatite is that it quite commonly looks like various arrested stages in processes by which preexisting rocks, generally metamorphosed sandy and shaley sediments, have been converted into granite without much change in volume or mechanical disturbance. The evidence for transformation of other types of rock into granite cannot, in the nature of the case, be founded in direct observations of the processes at work many kilometers down in the depths of the crust. It rests on interpretations of the geologic configuration left behind by long-dead processes, but it ranges from the broadest regional relationships to details of rock textures and crystal chemistry. An attempt to review the extensive literature on this subject in a short summary article would degenerate into mere "name-dropping." I have only outlined the nature of the evidence in Table 1 and cited representative examples.

Geologic evidence is seldom so complete or conclusive that it is not open to dispute and to alternative interpretations. None of the attempts to prove that major bodies of granite have formed by granitization have gone unchallenged, nor have magmatists lacked ingenuity in presenting alternative "igneous" interpretations of the features in rocks ascribed to granitization. Nevertheless, most geologists who have worked in terrains where granite and migmatite are intimately interrelated have had some kind of hypothesis of metasomatic granitization forced on them by the geology of migmatite. In fact, there is now no real argument that there is no such thing as granitization, but there is vigorous controversy over the nature of the process and the extent to which it is responsible for forming granite. Staggering chemical transport problems are of course involved in converting a large mass of rock into granite.

"Per Migma ad Magma"

Classic igneous theory, following the main line of German and Anglo-American thought at the turn of the century, holds that granitization is a localized phenomenon in places where magma comes in contact with country rocks under conditions that permit gaseous or liquid "emanations" given off during crystallization of the magma to penetrate the country rock intimately and modify it in varying degrees toward granite. But other concepts of granitization have equally venerable antecedents, especially in the French and Scandinavian literature, where difficulties were long recognized for straightforward magmatic theory in vast migmatite terrains and extensive bodies of granite that preserve throughout the relics of metamorphic rocks and structures. Michel Levy (*8*), Lacroix (*9*), Termier (*10*), and Sederholm (*11*), among others, conceived in varying degrees of a more organic inter-relationship between metamorphism and granitization, in which fluids mobilized in the depths of the earth beneath the orogenic belt rise into the deeply buried supracrustal rocks of the geosyncline, spreading like a "*tache d'huile*," and, along with the rise in temperature, effect the metamorphism, the metasomatic replacement, and ultimately the dissolution and fusion of rocks, giving rise to granitic magmas which rise as intrusive masses to higher levels. Thus, "normal" magmatic granite characterizes mainly the higher levels of granite emplacement, and in these views there is clear recognition of a sequence in depth of granite relationships correlated with increasing metamorphism and migmatization downward. Sederholm, in addition to the term *migmatite*, introduced the terms *palingenesis* (literally, "rebirth"), to denote the remelting of older rocks to form new magmas, and *anatexis*, to describe the extraction of granitic liquids from heterogeneous rocks by selective fusion and solution.

A culmination in this trend of thought is perhaps Wegmann's paper "Zur Deutung der Migmatite" (*12*), in which migmatite and granite are depicted as products of mobilization of material attending the deformation and metamorphism of the geosynclinal belt. With increasing heat and the advance of fluids and magmas mobilized deeper in the crust, the mineral assemblages of sedimentary rocks become unstable, and widespread migrations begin of those components which are not stable in solid phases under the new conditions. This migration, Wegmann suggested, need not wait upon the injection of magma but may take place by intergranular diffusion of ionic and molecular particles in a dispersed state. He conceived of waves of diffusing ionic and molecular particles expelled from lower levels and followed by fluid solutions to produce advancing fronts, first of metamorphism, then of migmatiza-

Table 1. Arguments for granitization. The literature cited is typical, giving evidence in support of, and uses of, these arguments, but the examples have not been selected especially for historical priority, nor are they unequivocal. Practically all of them have been vigorously contested on factual grounds or differently interpreted on theoretical grounds. Most of these authors also use other arguments besides the one given.

Structural

Room. The space occupied by a body of granite is not matched by commensurate dislocations in the structural pattern of the enclosing rocks; there is no "room" for intrusive magma (*33*).

Continuity. A granite contains structures, such as gneissic layering, or relics of a preexisting rock with structures, such as bedding or foliation, which are in structural alignment with the structure of the surrounding terrane (*34*).

Gradation. A structural pattern such as bedding or foliation in enclosing rocks passes gradationally into a pattern such as gneissic structure in a granite body (*35*).

Mechanics. The structural pattern of a granite body and its enclosing rock is such that the pattern could only develop if there was at no time a body of liquid corresponding to the present body of granite (*36*).

Pseudomorphism. A granite contains variations in texture and composition which simulate in pattern primary sedimentary or volcanic structures such as ripple marks, bedding, and pillows (*37*).

Involvement. Granitic materials are too intimately and complexly involved with sedimentary or metamorphic rocks to have been emplaced by injection of a magmatic liquid (*38*).

Stratigraphic

Substitution. A rock unit in a known sequence is missing, and in its place is a body of granite (*39*).

Relic stratigraphy. A granite contains isolated included layers which are part of a preexisting sequence, but these included layers maintain relative stratigraphic position within the granite (*40*).

Ghost stratigraphy. A body of granite contains a sequence of variations in its texture, composition, or structure which is analogous in some way to the stratigraphic sequence in the surrounding terrane (*41*).

Chemical

Basic front. Rocks in the vicinity of a body of granite are relatively enriched in components which must be driven out of preexisting rock to convert it to granite (*42*).

Basic behind. There has been impoverishment of granitic components in a mass of rock from which components have been extracted to make a related granite (*43*).

Gradation. A preexisting rock passes gradationally into granite by progressive enrichment in granitic components and impoverishment in nongranitic components (*44*).

Physical-chemical and crystal-chemical

Relic phases. Mineral phases in a granite may be identified as mineral phases belonging to preexisting sedimentary or metamorphic rock (*45*).

Mineral replacement and pseudomorphism. Minerals in a granitic rock are pseudomorphs or replacements of sedimentary or metamorphic minerals (*46*).

Metamorphic phase assemblage. Mineral assemblages in granite are related to the metamorphic facies of surrounding rocks (*47*).

Subsolidus assemblage. A phase assemblage is present in a granite which cannot form by crystallization from a magma (*48*).

Subsolidus or crystalloblastic paragenesis. The growth relationships among the mineral phases of a granite cannot be formed by crystallization from a magma but indicate growth by solid-state reactions (*19*).

Subsolidus temperature. The ambient temperature as evidenced by mineral phase relationships in the surrounding rocks was at all times below the stability range of a magma (*49*).

tion, and finally of selective fusion (anatexis), culminating in the mobilization (palingenesis and anatexis) of the migmatite zone and its injection in tongues and lobes into higher zones as essentially magmatic intrusions. Thus there evolved the concept "through migma to magma," which virtually reverses the roles of migma and magma as conceived in classic igneous theory. This point of view has received extended expression over the past 20 years in H. H. Read's development of the "granite series" concept (*13*).

Chemical Transport Problem

The emphasis placed by Wegmann on the role of intergranular diffusion gave recognition to a problem that up to about this time had received little systematic attention. Geologists attempting to form concepts adequate to explain the extensive migmatite terrains spoke freely of the introduction of mysterious granitizing substances by the spreading through the rock of "*agents mineralisateurs*," "solutions," "granitic juices," "pore fluids," or "ichor" (*14*). Vague terms such as *injection, soaking*, or *imbibition* were freely used. Yet it is plain that these are essentially metaphors rather than descriptions of observed physical processes, for very little is known experimentally of the transport characteristics of rock at pressures of a few thousand atmospheres and at temperatures of a few hundred to nearly a thousand degrees centigrade, yet this range is of most concern in the origin of granite. Furthermore, the capacity of dilute solutions to transport much material by fluid flow through a body of solid rock seems questionable.

Many geologic phenomena involve solid diffusion within dimensions of the order of crystal grains, but because of the extreme sluggishness of experimentally measured rates of solid diffusion in silicate minerals (*15*), solid diffusion was long dismissed as a mechanism for transporting large amounts of material within reasonable spans of time (*16*). However, in 1934 R. Perrin (*17*) cited reactions between slags and furnace linings in metallurgical experiments where substantial chemical exchanges took place between the charge and the refractory lining within periods of a few hours or less. He attributed these phenomena to solid diffusion and suggested by analogy that many geologic phenomena attributed to fluid transport actually involve solid diffusion. Perrin vigorously pursued this thesis and in 1939, with Roubault, made a detailed application of it to the formation of granite.

In retrospect the experimental evidence adduced by Perrin seems slender indeed compared to the broad application which he and others made of it to geology, for his melts and refractory linings were complex systems in which the phase relations were incompletely known, and no real evidence was given to rule out the participation of fluid phases. Not only might capillary action draw liquid or vapor from the melt into the brick but, to quote Greig (*18*): "Silica bricks are made from crushed quartzite mixed with a small amount of some other substance which on firing fluxes a part of the silica and forms a bond. When the brick is in use at a high temperature it is partly crystalline, partly liquid."

Indeed, the experiments from which so much geologic speculation proceeded are of present interest mainly because they stimulated a sweeping reappraisal of a great deal of geologic data from a new point of view. The real argument for diffusion as a major transport mechanism was drawn from the rocks themselves, even in the writings of Perrin and Roubalt (*19*).

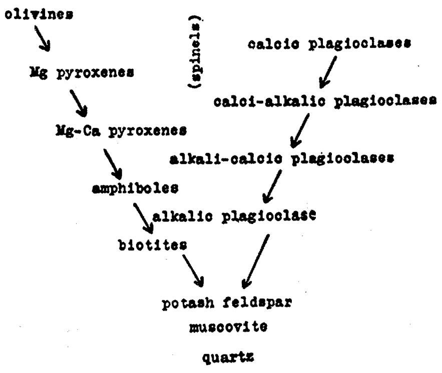

Fig. 5. The Bowen reaction series, showing the sequence in which the principal rock-forming minerals crystallize from magmas. The ferromagnesian ("dark") minerals on the left-hand branch form a discontinuous series with intervening reactions. The plagioclase feldspars form a continuous solid-solution series in the right-hand branch. In Bowen's view, common basaltic magmas with strong fractionation should yield a granitic residuum. The principal modifications suggested by modern research would be to show the Mg and the Mg-Ca pyroxenes in parallel, not in sequence, and to suggest a range of residual assemblages from the silica-enriched (quartz-bearing) to the silica-deficient (feldspathoid-bearing). [Bowen (*4*)]

Transformism

The awakening of interest in diffusion as a major transport mechanism in rock-forming processes heralded a new stage in the development of granitization theory. A theoretical rationale for long-range solid diffusion as the mechanism of emplacement of granite was soon forthcoming from Ramberg (*20*) and Bugge (*21*); it may be summarized as follows: The diffusivity of a particle in a phase may be given by a generalized thermodynamic or statistical mechanical function; for example, the thermodynamic expression

$$D = D_0 e^{-Q/RT} \qquad (1)$$

where D is diffusivity, D_0 is ideally a constant for a given mechanism of diffusion in a homogeneous phase, and Q is an activation energy. Since D_0 and Q are finite quantities, any particle always has finite diffusivity in a phase; therefore any chemical potential gradient will cause a finite migration of matter. To show that this should operate in the crust of the earth to produce granite, various thermodynamic and crystal-chemical arguments are mustered to show that the chemical potentials of the elements with large relative abundance in granite increase more with increasing temperature and stress than do the potentials of the less abundant ones. Consequently, some elements, such as potassium and sodium will tend to migrate out of deeper regions of the earth or regions of intense metamorphism and deformation to become fixed again as migmatite and granite in regions of lower stress and temperature by displacing other elements, notably iron and magnesium. Since not all the elements that occur in relatively great abundance in granite have closely comparable crystal-chemical parameters—notably silicon relative to the alkalis—the theory is not without difficulties in detailed application.

This type of theoretical argument was the final link in the evolution of a whole new scheme of petrogenesis for granitic rocks, which has been styled "transformism," and which holds that chemical potential gradients drive diffusion mechanisms which are capable of producing granite in practically all its geologic configurations without the participation of a magmatic phase. Enthusiastic transformists needed only one fell swipe of Occam's razor to remove magmatic granite from serious geologic consideration as ruthlessly as though it were a morbid vermiform appendix, and it has been held by some that magmatic granite, if it forms at all, is only produced by localized melting at an advanced stage of granitization or as an insignificant minor product of basaltic differentiation. However, it seemed to many geologists that there was a missing link in the transformist argument.

The Trouble with Transformism

Magmatists have a wealth of experimentally established physical chemistry which they have had trouble fitting to the geology of granite. Transformists, on the other hand, have a hypothesis to explain the geology of granite which is difficult to found in orthodox chemistry. To some extent the transformists have reacted by developing a "geochemistry" of their own to fit the geology, but much of this special geochemistry essentially rests on a negative argument: Conventional chemistry is beset by many difficulties in the geology of granite, therefore diffusion is presumed to do what it has to do to explain that which cannot otherwise be explained. Undismayed by this *ad hoc* quality, some transformists have styled it the "actualistic" approach to petrology.

The missing link in the transformist scheme of petrogenesis is the independent experimental demonstration that the potentials resulting from nonequilibrium configurations and dislocations in the lithosphere really do drive diffusion to the exclusion of other processes such as solution, melting, mechanical motion, and heat flow. Without an adequate experimental foundation, the rationale in terms of chemical potential gradients does not demonstrate the operation of diffusion to the exclusion of other processes but is nothing more than an abstract and operationally clumsy restatement of the basic problem: What processes do the potentials activate, at what rates, and under what conditions?

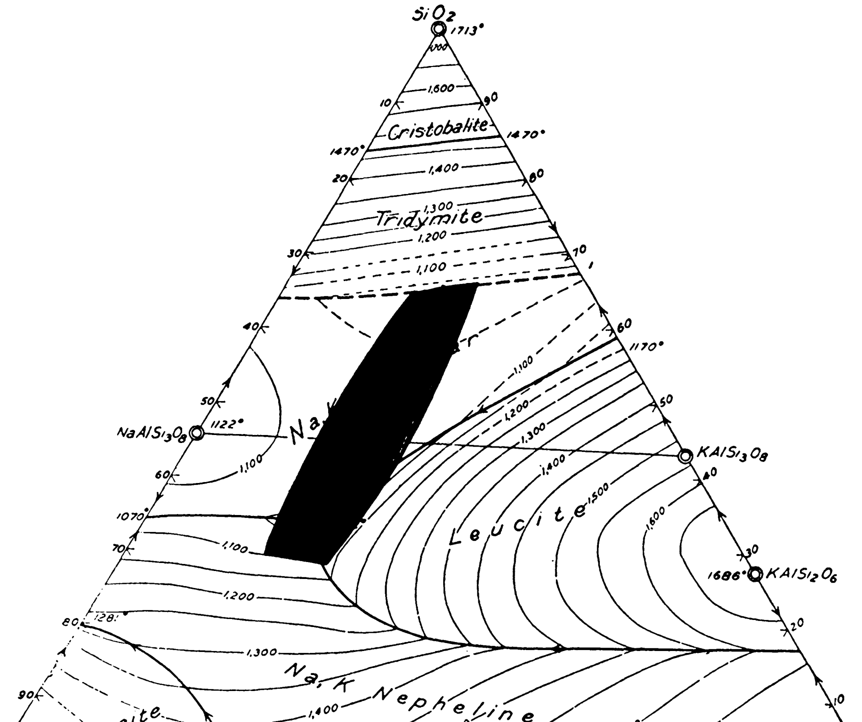

Fig. 6. "Petrogeny's residua system"; equilibrium relations of the principal components of granite in the dry melt, showing liquidus temperatures. Granites fall within the triangle SiO_2-$NaAlSi_3O_8$-$KAlSi_3O_8$. The trough containing the compositions toward which residual liquids proceed with falling temperatures of crystallization is shown in solid black. [Bowen (*52*)]

Diffusion in the Geologic Realm

Measured lattice diffusion rates in silicate crystals are too sluggish to produce large-scale geologic effects in reasonable spans of time. Below magmatic temperatures, measured rates are typically less than 10^{-8} cm^2 sec^{-1} by one or more orders of magnitude. Diffusion rates as high as 10^{-8} cm^2 sec^{-1} allow mean displacements of only a few meters in a million years. There is considerable geologic evidence that masses of granite are emplaced in orogenic belts within time spans of less than a million to a few million years (*22*), so it is probably idle to attempt to magnify the possible role of lattice diffusion by assuming vastly longer spans of geologic time.

There are other kinds of diffusion in solid aggregates, such as grain boundary and surface diffusion, for which there are few or no experimental data directly applicable to rocks. However, a strong geologic argument may be made *against* the large-scale geologic effectiveness of *all forms* of diffusion in *entirely solid* aggregates. Lattice, grain boundary, and surface diffusivity are properties possessed in common by all rocks. Yet many metamorphic and granitic rock assemblages contain numerous small-scale nonequilibrium features such as zoned crystals, reaction rims or coronas between minerals, and adjacent layers of rock with sharply contrasting compositions and mineral assemblages. Something other than a general property of rocks must be involved in the effective transport of matter through rock, otherwise nonequilibrium configurations would be wiped out over all relatively short distances in regions where solid diffusion is supposedly active enough to granitize huge masses of rock.

However, diffusion in an intergranular fluid phase may permit the necessary flexibility by allowing large-scale diffusion where the intergranular medium is present, in sharp contrast to sluggish and incomplete reactions where the solid phases alone must serve as the vehicle. The coefficient of diffusion of a particle at infinite dilution in a fluid medium is approximated by the well-known Stokes-Einstein equation:

$$D = RT/4\pi N\eta r_s \qquad (2)$$

where R is the gas constant, T is the absolute temperature, N is Avogadro's number, r_s is the radius of the particle, and η is the viscosity of the fluid medium (*23*). $R/4\pi N$ is a constant which equals 1.1×10^{-17} erg deg^{-1}; thus,

$$Dr_s = \frac{T}{\eta}\, 1.1 \times 10^{-17} \text{ erg deg}^{-1} \qquad (3)$$

I have attempted to evaluate the function Dr_s for several temperatures within the range of strong metamorphism and granitization and for pressures up to 2000 atmospheres (see Fig. 8), using some recent work by Fano *et al.* (*24*) on the extrapolation of viscosity data for water to the high pressures and temperatures for which Kennedy (*25*) has supplied experimental *PVT* data.

The characteristics of the function Dr_s have interesting geologic implications. The ions and molecules mainly involved in forming granite have radii (r_s) ranging from about 0.5×10^{-8} to about 3×10^{-8} cm. Since Dr_s attains slowly varying values of near or less than 1×10^{-11} at very moderate geologic pressures, it appears that in low concentrations the diffusion rates (D) of most rock-forming components in supercritical water vapor should be on the order of 10^{-3} to 10^{-4} cm^2 sec^{-1} without much change over a wide geologic range of P and T.

Now, diffusion rates on the order of 10^{-3} to 10^{-4} cm^2 sec^{-1} would be adequate to produce very large transfers of material over considerable distances within very moderate periods of geologic time and with reasonably small concentration gradients if diffusion actually takes place according to the Stokes-Einstein law through an *unobstructed* gaseous medium, which of course is certainly not exactly the case in rocks.

Nevertheless, if my evaluation of Dr_s has even order-of-magnitude validity, it would appear that the diffusion rate in the fluid medium is not the rate-limiting factor for geologic processes, provided the rock has significant effective directional porosity (*26*). The real rate control may be exercised by the effective concentration gradients possible in the intergranular fluid, and these depend on the solubility of the diffusing particles in the fluid medium, the rate of solution where the rock is being attacked, and the rate of removal at the interface where the particles are going back into solid phases; the last factor in turn depends on solid diffusion through the reaction-product layer. As the concentration and variety of particles in solution increase and the dimensions of intergranular openings decrease, complex effects are possible, some of which may increase and some of which may decrease diffusion rates (*27*). At present we are quite unable to assign even order-of-magnitude values to these effects.

From the geologic evidence it appears to me that rocks which are inherently "dry" (composed of anhydrous mineral assemblages, and of low porosity) are poor media for diffusion and generally show only small-scale effects involving any kind of chemical transport. Certainly rocks that become "dry" in this sense are notably resistant to further recrystallization and chemical change during metamorphism unless water is reintroduced into them and "retrograde" metamorphism takes place. On the other hand, where rocks suffer dilatant strain and have water introduced into them, or contain hydrous minerals such as clays, chlorite, and hydro-micas and reach temperatures where these minerals break down into less hydrous assemblages plus a separate fluid phase, the rock of necessity contains a pore fluid and becomes open to diffusion in the fluid phase until the water itself diffuses or flows away. Loss of water is the reason for the "irreversibility" of metamorphism and granitization. It is realistic to suppose that intergranular water becomes available in the environment of granitization because water-bearing rocks such as clay shales evolve into mica schists and ultimately into feldspathic gneisses during regional metamorphism with the evolution and loss of several percent of original combined water, quite apart from the question of the introduction of juvenile water.

The New Synthesis

If it is indeed water (with other volatiles, to be sure) which gives rock the major part of whatever capacity it has to transport material by diffusion on a geologically large scale, then exciting possibilities open up for reconciling many anomalies in the geology of granite and migmatite with growing experimental knowledge of the chemistry of silicates. O. F. Tuttle's summation of the work he and Bowen did in the system $NaAlSi_3O_8$(Ab)-$KAlSi_3O_8$(Or)-SiO_2(Q)-H_2O has now been published (*28*, *29*). These are the components at the apex of the reaction series

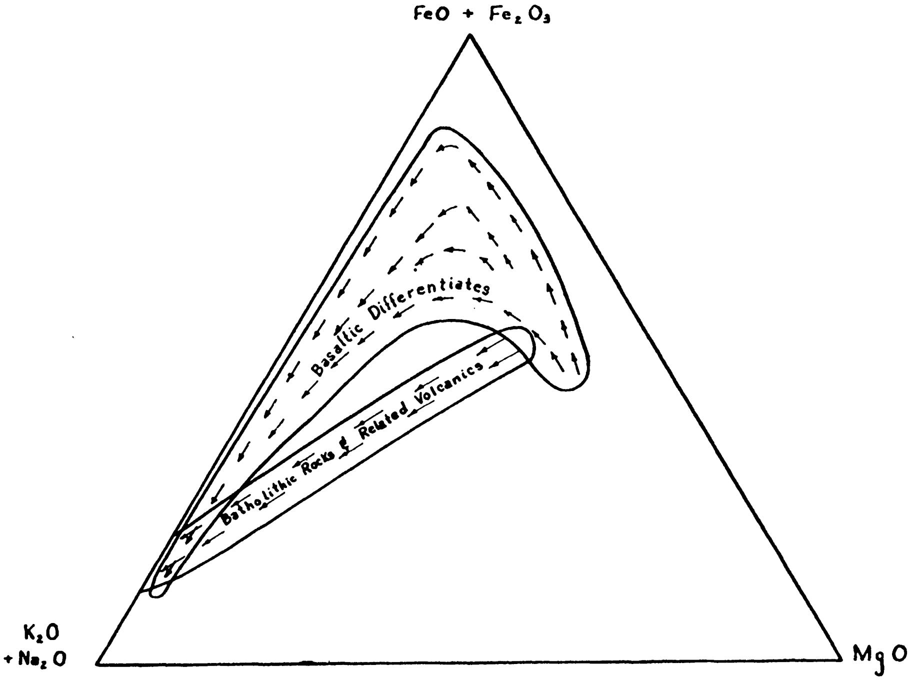

Fig. 7. Trends of variation, from field examples of clearly related suites of rocks, with respect to total Fe, Mg, and the alkalis. Rocks clearly derived from the fractionation of basaltic magma follow trends toward iron enrichment in the middle stages which are not shown by rocks related to granitic sequences. [After Tilley (*53*)]

that impart to granite its principal chemical characteristics, and this is a fitting final legacy of Bowen to the science he served, for, having contributed the reaction series to geology and having done so much to work out its consequences for magmatic differentiation by crystal fractionation with loss of heat from basaltic magmas, he joins posthumously with Tuttle now in giving us some initial insights into another petrogenic model.

Tuttle and Bowen present sufficient experimental data to define important characteristics of the system Ab-Or-Q-H_2O at liquidus temperatures and up to more than 4000 atmospheres of water-vapor pressure (Fig. 9). They show how crystallization of melts in this system tends to produce mineral assemblages of alkali feldspars and quartz approaching granites as water-vapor pressure increases. Crystallization proceeds to a cotectic minimum at which quartz and a single homogeneous K-Na feldspar crystallize simultaneously up to a water vapor pressure of about 3600 kg cm^{-2}. At higher water concentrations separate K and Na feldspars begin to crystallize, and the isobaric minimum in the system is a ternary eutectic (Fig. 9, bottom left). Work at the Geophysical Laboratory by Yoder, Stewart, and Smith (*30*) shows that with very modest amounts of Ca feldspar in the ternary feldspar-water system $NaAlSi_3O_8$(Ab)-$KAlSi_3O_8$(Or)-$CaAl_2Si_2O_8$(An)-H_2O two feldspars crystallize at even lower water concentrations, one K-rich and the other a sodic plagioclase. Thus, the magmatic crystallization of the quartz-feldspar assemblage of granite has been experimentally confirmed, with water playing a crucial role.

Tuttle argues that granitic liquids must be important in rocks that contain water and the components of granite because the stability field of granitic liquids extends well into a range of temperature and pressure which is attained at depth in zones of high-grade regional metamorphism. Moreover, the close relationship of the composition of granite to the locus of the cotectic minimum in the experimental system is strong evidence that granite-melt equilibria exercise a high degree of control over the composition of granite. This is powerful experimental support for the role of magma in the forming of granite, but it also leads to a highly significant departure from the classic model of igneous theory.

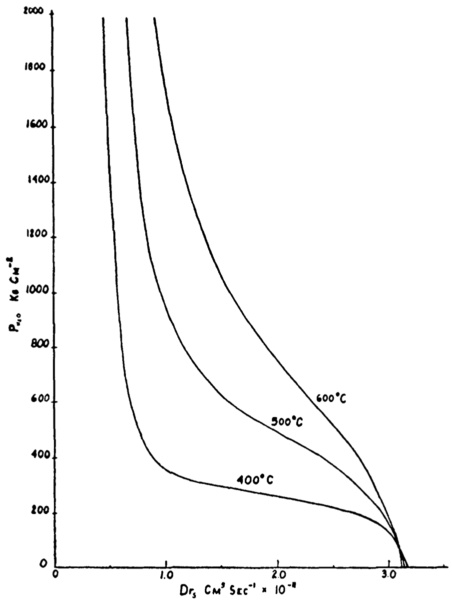

Fig. 8. Characteristics of the Stokes-Einstein diffusion function for various pressures and temperatures of water vapor.

I have already pointed out that the old magmatic model considered the consequences of introducing a relatively hot silicate melt derived from some deep-seated source into an envelope of rock at a lower temperature. The ensuing events and final products then depend primarily on flow of heat from the melt to the envelope and secondarily, on chemical and mechanical interaction between the envelope and the body of magma. Contrast this with an alternative model toward which Tuttle is pointing, a model presaged in Sederholm's concept of anatexis.

The Anatectic Model

In the "anatectic model" we consider a body of rock containing water into which there is a large flow of heat as a consequence of crustal downbuckling and other orogenic processes. The temperature and water-vapor pressure rise toward the point where a granite melt becomes stable. Water reacts with the rock silicates, first to form alkali silicate solutions and then to form melt, the quantity and composition of the solution or melt being such, of course, that the chemical potentials of water and the other components are equal in the coexisting gaseous, liquid, and solid phases. For example, at a temperature somewhat less than 700°C and at a water-vapor pressure of 2000 kg cm^{-2} a granite melt can form, and if the components of granite are available in a rock, melting will proceed until a melt is formed containing a little more than 6 percent water. One percent of water in a rock, under these conditions, could melt up to about 16 percent of the rock, provided the rock contained this much granitic material. According, therefore, to the temperature, confining pressure, composition of the rock, and water available, melting may be either partial or complete.

As Tuttle points out, realistic conditions may be set up where crystallization proceeds nearly adiabatically, isothermally, or isobarically. The classic model of secular cooling of the melt may not be at all realistic in the plutonic environment, where the melt may be in equilibrium at all times with the ambient temperature of metamorphism and where crystallization proceeds by loss of more mobile fractions to the surroundings. Realistically, then, the system may proceed toward equilibrium under the influence of a pressure gradient and a continuous heat input from the deep-seated sources of energy which drive the orogenic process. The heat flow will then induce chemical and mechanical mass transport tending to produce a new geologic configuration in which the whole rock mass is more stable with respect to a higher temperature gradient.

It will take a great deal of additional experimental work and careful analysis to work out the major consequences of this complex conception of igneous processes in the forming of granite. Fractional crystallization is only one means of reorganizing a body of rock by the separation of phases in different states. In effect, the term *igneous* is endowed here with great generality to refer to all rock-forming processes in which changes of state in rock silicates from fluid to solid or solid to fluid are important (*fluid* refers to both liquid and gasesous states). The experimental data show that such changes must be important in the realm in which granite forms.

The anatectic model is somewhat less concerned with fractionation by the separation of crystals from the melt, which is the main theme of the classic model, because the melts most readily formed are bound to be near the cotectic minimum composition and there is no place much for them to go by crystal fractionation. Because of the key role played by water vapor under high pressure in forming granite melts, the possible role of chemical transport and fractionation by diffusion in an aqueous fluid phase extending into ranges of temperature and water-vapor pressure well below the range of silicate melts becomes a major theme of the anatectic model. The transport properties of these fluids and the relative solubilities and mobilities of rock-forming components in them are already a factor in the experimental work, although these phenomena have not yet received much systematic attention. One of the most annoying problems encountered in experiments with silicates at high water-vapor pressures is that some components, notably silica and alkalis, won't stay put in the crucible. Rapid transport takes place in the vapor phase throughout the pressure vessel (*5, 28, 31*). The ratios in which the components of granite are transported in the vapor phase are not the same as their ratios in the melt, but favor silica over the feldspars and Or over Ab. Phenomena of this kind can explain many anomalous geologic relationships which are difficult to explain in terms of melt equilibria.

Much of the geologic data that has been adduced in favor of transformist or magmatist interpretations must be reexamined in the light of the new experimental data and the anatectic model. Much of the argument for granitization rests on the persistence within granites of what seem to be relics of preexisting rocks, minerals, and structures. To what extent are these relics simply residues from which the granitic fraction has been extracted or redistributed by anatectic mobilization? Once the granite fraction becomes mobile, how far might it move? How much chemical mobility is there in intergranular fluids below the stability field of granite liquids, and to what extent is a melt phase necessary to segregate a granite fraction from rocks? When a melt fraction forms in rocks, to what extent does the rock as a whole lose cohesion and become subject to semiviscous flow, and under what conditions does the melt phase segregate from the unmelted residue? Up to what stage will the geologic configuration continue during these processes to simulate the ghost stratigraphy and structural relationships on which the granitization hypothesis rests? These and a host of other intriguing questions suggest themselves.

H. H. Read has said that the granite problem is essentially a field problem (*32*). I must offer a resounding dissent. Whenever a geologist seeks to discover the origin of a configuration of rocks he must bring to it some conception of the physical and chemical processes which are possible in rocks. The geologist interested in surficially formed rocks can often study his processes at work in the field, but this opportunity is not vouchsafed the student of granite except by extrapolation and analogy from the crucible and the high-pressure furnace.

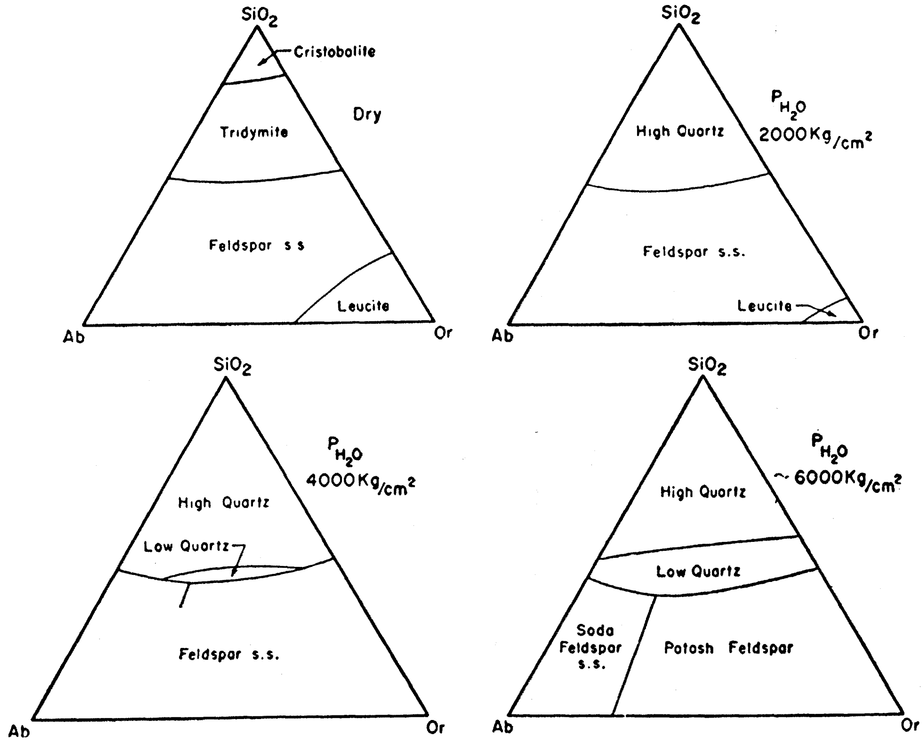

Fig. 9. Stability fields at the liquidus in the system $NaAlSi_3O_8$-$KAlSi_3O_8$-SiO_2-H_2O for various pressures of water vapor. Liquidus temperatures fall toward a trough on the join between feldspar and the silica minerals. Two feldspars begin to crystallize, and a ternary eutectic appears at water-vapor pressure of about 3,500 kg cm^{-2}. The minimum temperature on the liquidus falls from about 950°C for the dry melt (compare Fig. 6) to about 650°C with water-vapor pressure of 4000 kg cm^{-2}. [Tuttle and Bowen (*28*)]

Looked at from this point of view, the granite controversy has been a noble effort by geologists to pull themselves up by their bootstraps, to discover the origin of granite before the physics and chemistry of the system could be investigated experimentally or theoretically in the appropriate regions of pressure, temperature, and composition. The magmatist effort has been at least superficially conservative. It has been an effort to interpret the geology in terms of models based only on well-known fundamental physical and chemical processes—basically, heat flow, mechanical motion of solid and fluid bodies, crystal-melt equilibria, and fractional crystallization. Where the geologic configuration has seemed to be at odds with the ostensible consequences of these processes, the magmatist has sneaked behind his conservative façade to make *ad hoc* assumptions about the working of his processes under conditions at depth.

The transformists have in a sense been more openly imaginative and more radical in turning to the poorly known processes of diffusion where the well-known ones seem to fail. Speculative though a good deal of their special geochemical doctrine may be, it has performed a great service in shaking the complacency which always overcomes those who believe they are being conservative. It has forced geologists to look harder at their rocks, to define the limitations of the processes, to question oversimplified or inflexible models, and to extend their experimental investigations into increasingly critical regions. Transformists who have read these words may well be smiling to themselves with the thought that at last the magmatists have "discovered" granitization. The difference is that modern experimental research is beginning to explore rather than hypothesize physical-chemical processes which can ultimately reconcile the many puzzles presented by the geology of granite.

References and Notes

1. J. Hutton, *Trans. Roy. Soc. Edinburgh* **3**, 77 (1794).
2. F. Chayes, *Geol. Mag.* **94**, 58 (1957).
3. C. Darwin, *Geological Observations on the Volcanic Islands* (Smith, Elder, London, 1844).
4. N. L. Bowen, *The Evolution of the Igneous Rocks* (Princeton, Univ. Press, Princeton, N.J., 1928).
5. ——— and O. F. Tuttle, *Bull. Geol. Soc. Am.* **60**, 439 (1949).
6. Or, Ab, and Q are abbreviations for the mineral phases of orthoclase feldspar, albite feldspar, and quartz but are also used in a chemical context to mean any phase of corresponding composition.
7. R. A. Daly, *Igneous Rocks and the Depths of the Earth* (McGraw-Hill, New York, 1933).
8. M. Michel Levy, *Carte géol. France* **5**, 317 (1893).
9. M. A. Lacroix, *ibid.* **10**, 241 (1898).
10. P. Termier, *Compt. rend. congr. intern. géol. 9^{e} Congr. Vienna, Austria* (1904), vol. 2, pp. 572-586.
11. J. J. Sederholm, *Bull. comm. géol. Finlande No. 23* (1907), English summary.
12. C. E. Wegmann, *Geol. Rundschau* **26**, 305 (1935).
13. H. H. Read, *The Granite Controversy* (Interscience, New York, 1956).
14. *Ichor* is a Greek word of unknown origin. In classical mythology it signified the blood of the gods.
15. W. S. Fyfe, F. J. Turner, J. Verhoogen, *Geol. Soc. Am. Mem. No. 73* (1958).
16. A. Harker, *Metamorphism* (Methuen, London, 1932); W. J. McCallien, *Bull. comm. géol. Finlande No. 104* (1934), pp. 11-27.
17. R. Perrin, *Ann. mines, Mém. No. 13* (1934), pp. 135-170.
18. J. W. Greig, *ibid.* **13**, 133 (1927).
19. R. Perrin and M. Roubault, *Carte géol. Algérie Bull. No. 4* (1939).
20. H. Ramberg, *Norsk. Geol. Tidsskr.* **24**, 98 (1944); *Medd. Dansk Geol. Foren.* **11**, 13 (1946).
21. J. A. W. Bugge, *Avhandl. Norske Videnskaps-Akad. Oslo Mat-Naturv. Kl.* **13** (1945).
22. A. Knopf, *Bull. Geol. Soc. Am.* **57**, 649 (1948).
23. The numerical coefficient in the denominator of the Stokes-Einstein equation as derived from Stokes' law is 6. However, it has been found that the value 4 gives better agreement with experimental results in dense gases (P. A. Lyons, Yale University, personal communication).
24. L. Fano, J. H. Hubbell, C. W. Beckett, "Compressibility factor, density, specific heat, enthalpy, entropy, free-energy function, viscosity, and thermal conductivity of steam," *National Advisory Committee on Aeronautics, Tech. Note No. 3273* (1956).
25. G. C. Kennedy, *Am. J. Sci.* **248**, 540 (1950); ——— and W. T. Holser, *ibid.* **256**, 744 (1958); ———, *ibid.* **257**, 71 (1959).
26. R. M. Garrels, R. M. Dreyer, A. L. Howland, *Bull. Geol. Soc. Am.* **60**, 1809 (1949).
27. J. H. Wang, *J. Phys. Chem.* **58**, 686 (1954); W. T. Holser, *Econ. Geol.* **42**, 384 (1947).
28. O. F. Tuttle and N. L. Bowen, *Geol. Soc. Am. Mem. No. 74* (1958).
29. I wish especially to acknowledge the courtesy of O. F. Tuttle and his staff for putting the galley proofs of the Tuttle and Bowen memoir at my disposal in advance of its publication date.
30. H. S. Yoder, D. B. Stewart, J. R. Smith, *Carnegie Inst. Wash. Year Book No. 56* (1957), pp. 206-214.
31. G. W. Morey, *Econ. Geol.* **52**, 225 (1957).
32. H. H. Read, in "Origin of Granite," *Geol. Soc. Am. Mem. No. 28* (1948).
33. B. C. King, *Geol. Mag.* **84**, 145 (1947).
34. F. D. Eckelmann and A. Poldervaart, *Bull. Geol. Soc. Am.* **68**, 1225 (1957).
35. G. H. Anderson, *ibid.* **48**, 1 (1937).
36. H. Ramberg, *ibid.* **67**, 185 (1956).
37. D. R. Bowes, *Quart. J. Geol. Soc. London* **109**, 455 (1954).
38. P. Ljunggren, *The Region of Hâlia in Dalecarlia, Sweden* (Bergendahls, Göteborg, 1954).
39. H. G. Backlund, *Bull. Geol. Inst. Univ. Upsala* **27**, 219 (1937).
40. P. Misch, *Am. J. Sci.* **247**, 209, 372, 673, (1949).
41. W. S. Pitcher, *Quart. J. Geol. Soc. London* **108**, 413 (1952).
42. D. L. Reynolds, *Geol. Mag.* **84**, 33 (1947).
43. A. E. J. Engel and C. G. Engel, *Bull. Geol. Soc. Am.* **69**, 1369 (1958).
44. G. E. Goodspeed, *Am. J. Sci.* **246**, 515 (1948).
45. A. Poldervaart and F. D. Eckelmann, *Bull. Geol. Soc. Am.* **66**, 947 (1955).
46. B. Stringham, *ibid.* **64**, 945 (1953).
47. H. Ramberg, *J. Geol.* **57**, 18 (1949).
48. E. Nickel, *Opuscula mineral. et geol. Veröffentl. Mineral u Geol. No. 2* (1954).
49. G. Kullerud and H. Neumann, *Norsk Geol. Tidsskr.* **32**, 148 (1953).
50. E. Raguin, *Géologie du granite* (Masson, Paris, ed. 2, 1957).
51. Am. Assoc. Petrol. Geologists, *Tectonic Map of the United States* (1944).
52. N. L. Bowen, *Am. J. Sci.* **33**, 1 (1937).
53. C. E. Tilley, *Quart. J. Geol. Soc. London* **106**, 37 (1950).

Strontium-90 Content of the Stratosphere

A low concentration of strontium-90 in the stratosphere indicates a short stratospheric residence time.

Herbert W. Feely

Prediction of the future hazard to the human population from radioactive fallout from past and future tests of nuclear weapons requires an estimate of the amount of debris held in the stratosphere and its rate of release to the troposphere and thence to the ground. Estimates of the stratospheric burden and residence time of strontium-90, generally considered to be the most hazardous fission product, have been made in the past (*1, 2*) by combining assumed stratospheric bomb yields with measurements of fallout in rain and soil. However, each of these numbers is known only approximately. Some data are available from Project Ashcan (*3*), a project in which filter sampling of air is carried out by means of balloon-borne samplers. Unfortunately, uncertainties in the sampling efficiency and in the radiochemical analyses have limited the usefulness of these data (*2, 4, 5*).

Research Program

Since August 1957, long-range, high-altitude Lockheed U-2 aircraft of the U.S. Air Force have been systematically sampling the radioactive debris in the stratosphere as part of the High Altitude Sampling Program sponsored by the Defense Atomic Support Agency. The scientific direction of the sampling program, the radiochemical analysis of samples, and the meteorological interpretation of the data are the responsibility of Isotopes Inc., Westwood, N.J. (*6*).

After an initial training and testing phase, from August to October 1957, at Laughlin Air Force Base, Texas, the sampling aircraft were based at Plattsburg Air Force Base, New York, and Ramey Air Force Base, Puerto Rico. From these two bases a north-south sampling corridor along the 70th meridian (W) extending from latitude 66° N to 6° S, was monitored systematically from November 1957 through May 1958. During June and July 1958, sampling was conducted only in the vicinity of Puerto Rico. From September 1958 through August 1959 the aircraft operated from Ramey Air Force Base and from Ezeiza Airfield, near Buenos Aires, Argentina, in order to sample the atmosphere in the South-

---

The author is senior research scientist at Isotopes Inc., Westwood, N.J.