

# HYDROTHERMAL ALTERATION IN SOME GRANODIORITES

*by*

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## ABSTRACT

The patterns of distribution of the mica and clay minerals resulting from alteration of feldspars are much less regular and well defined in the "porphyry copper" deposits than in the vein deposits such as those at Butte, Montana. There are, however, interpretative problems common to both. Chemically, the destruction of the K-feldspars may be interpreted in the light of laboratory investigations of the system potash-alumina-silica-water at various  $K^+/H^+$  ratios. The experiments indicate that the K-mica (sericite) stability field lies between the K-feldspar and kaolinite fields. The K-mica field is approximately 1.5 pH unit wide, and its boundaries with K-feldspar and kaolinite are parallel below 500°C. The boundaries slope toward higher  $K^+/H^+$  ratios at lower temperatures. Thus, considering the K-feldspar-sericite-kaolinite system of minerals, one might expect field patterns showing a central zone of kaolinite bordered by a zone of K-mica which in turn grades outward into unaltered K-feldspar. Such a pattern is demonstrated by Butte's inner dickite-kaolinite and sericite and possibly by the zoning at San Manuel described by Schwartz. Alteration of plagioclase introduces additional complications into the distribution patterns of clay and mica minerals, the chief of which is the outer argillitic zone so characteristic of both vein and disseminated deposits.

## INTRODUCTION

Detailed re-examination of wall-rock alteration in ore deposits has been much stimulated during the past 20 years by advances in the study of sheet-structure silicates. Ore deposits that are spatially and temporally associated with granodiorites, quartz monzonites, and their porphyries in the Western Cordillera of North and South America have been important focal points of interest in alteration studies. The present review proposes to summarize salient structural and geochemical controls on kaolinite-sericite (muscovite)-K-feldspar associations and distribution patterns, and to appraise possible relationships between them. The kaolinite-sericite-K-feldspar minerals have been isolated for primary attention chiefly for the purpose of making use of some new chemical data on the  $K_2O-Al_2O_3-SiO_2-H_2O$  system. Reference to other important alteration mineral groups, such as the trioctahedral micas and the montmorillonoids, will be made simply to complete the geometrical picture of the mineralogical associations.

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### FIELD RELATIONSHIPS

Striking correlations exist between the structural types of ore deposits in granodiorite, the textures of the rocks, and sizes of the plutons. The large vein deposits (e.g., those at Butte, Montana) are mostly in equigranular rock masses of batholithic size. Many of the "disseminated" deposits are chiefly in or near smaller porphyritic bulb- or pod-shaped intrusives, some of which are demonstrably constricted at depth. Characteristically, the porphyries are "crackled" with numerous closely spaced joints, sometimes producing fragments scarcely a centimeter on a side. By contrast, early joints in the equigranular rocks are commonly several feet or several tens of feet apart.

The wall rocks adjacent to the earliest vein-filled joints in both types of rocks commonly show no destruction of the K-feldspars and may even show feldspar crystallization. At Ajo (Gilluly, 1946), for example, tiny seams of K-feldspar and quartz ramify much of the porphyry in the mine. Under crossed nicols these veinlets show a saccharoidal texture that conforms to the texture of the groundmass. A saccharoidal or aplitic groundmass of K-feldspar and quartz is common in the porphyries that contain disseminated copper deposits. Veinlets of saccharoidal quartz or quartz plus feldspar have also been found at Chuquicamata (Perry, 1952), Potrerillos, Climax (Vanderwilt and King, 1955), Bingham (Stringham, 1953), Bagdad (Anderson, 1950), and in many other "porphyry coppers."

Narrow quartz veinlets which contain little or no feldspar are abundant locally in the equigranular quartz monzonites also, as at Butte. They are discontinuous joint fillings that cut fresh feldspars with no apparent alteration.

In Butte these quartz veinlets are in turn cut by other quartz veinlets which contain selvages of molybdenite (Sales and Meyer, 1956). Rocks adjoining the earliest of the quartz-molybdenite veinlets still show fresh feldspars, but narrow envelopes of sericitization enclose later veinlets of quartz-molybdenite composition, especially if pyrite is present in addition to the molybdenite. In a few areas in the north-central part of the district, pyrite is the most abundant mineral in these early joint fillings. Immediately adjacent to the veinlets both K-feldspar and plagioclase are sericitized, but an inch or two from the veinlets the sericite has attacked the plagioclase preferentially relative to K-feldspar. Here sericite and K-feldspar are in contact, and there is no argillic (clay-rich) zone between sericitized plagioclase and fresh plagioclase. Because of the absence of an argillic zone these early alteration envelopes afford a sharp contrast to the ubiquitous argillization in the outer alteration envelopes around structurally later main-stage veins.

Quartz-molybdenite veinlets are also very common in the porphyry coppers. As at Butte, the quartz-molybdenite veinlets may or may not be

enveloped in sericitized walls. Well-developed bands of sericitization are visible adjacent to the quartz molybdenite veins at Chuquicamata without parallel outer bands of argillization, but sericitization appears to have been more diffuse and interspersed with K-feldspar at Climax (Vanderwilt and King, 1955, p. 45). At Bagdad, Arizona, molybdenite-quartz-orthoclase-pyrite veins cut quartz-pyrite-chalcocopyrite. "Locally sericite is abundant and appears to be essentially contemporaneous with molybdenite" (Anderson, 1950, p. 616). Anderson concludes that lack of well-defined penetrations of one by the other suggests essential contemporaneity of orthoclase and sericite, and he believes that the sulfides also crystallized in the same episode.

#### *Zoned Sericitization and Argillization Adjacent to Veins*

Lovering's paper of 1941 on the tungsten veins of Boulder County, Colorado, was a major stimulus to modern study of alteration mineralogy. He described an outer zone of alteration characterized by clay minerals that replace the plagioclase feldspars of a quartz monzonite host rock. The clay minerals are allophane and montmorillonoid (plus a little "hydrous mica") in the outer portion of the argillized zone, but they change to dickite-rich clay at its inner or veinward edge. As illustrated in Fig. 3 of Lovering's paper, the clay minerals are converted to sericite in the zone between the argillic zone and the vein. The sericite zone replaces the clay at a sharp front.

K-feldspar is not much affected in either zone. This important observation means that sericitization of clay proceeded ahead of, or in preference to, sericitization of K-feldspar. As in nearly every well-studied case in the porphyry coppers where K-feldspar and sericite exist together in the same zone, the sericitic zones at Boulder County show addition of  $K_2O$  over the amount either in fresh rock or in the argillized zones (Lovering, 1941, p. 244). Tooker (1956) also records substantial increases of  $K_2O$  in the sericitized zones of the Central City District, Colorado.

Parallel zones of sericitization and argillization are present also at Butte, Montana, adjoining large copper, zinc, and manganese veins that cut a batholithic mass of medium-grained quartz monzonite. Detailed structural studies for more than 50 years in Butte's 2,800-cdd miles of underground workings have established the fact that fracturing of the host rock was a recurrent process before, during, and after the introduction of the ore components. Much of the relief of late stresses was by strike faulting of earlier fissures, but it is also demonstrable that many of the later faults, which are themselves ore-bearing, cut and offset sulfide vein-filling in the earlier veins and strike out through previously unaltered wall rocks between the earlier veins. It is significant therefore that concentric envelopes of sericitization and argillization border all ore-bearing fractures, with the sericite envelope adjacent to the vein and the argillic envelope ubiquitously between the sericite envelope and fresh rock. Argillization could not have been an exclusively early process in the Butte district.

At the outer edge of the argillic envelope andesine is attacked along its cleavage by predominantly aluminous montmorillonoids interlayered with a small amount of kaolinite. Toward the vein the kaolinite proportion in the mixed clay increases. Locally, patches of amorphous clay in the plagioclase grains are ensheathed by montmorillonoid plates. No amorphous clay is now in contact with feldspar. Biotite is marginally chloritized at the outer fringes of alteration, but in many cases the chlorite apparently is reconverted to fine-grained biotite closer to the vein in the kaolinite subzone. Quartz is everywhere unaffected in contact with kaolinite. No detectable 0:1:4 ( $K_2O:Al_2O_3:SiO_2$ ) clay is found between them. K-feldspar shows only slight marginal attack by kaolinite right up to the sericitized front.

In the sericitized zone the clay minerals, biotite, and K-feldspar are all ultimately converted to white mica. The iron of the biotite goes to pyrite. Total sericitization of all aluminous minerals at a single sharp front takes place in the outer or peripheral parts of the Butte District. However, at deeper zonal levels, close to the Central Zone of pervasive sericitization and presumably closer to the source of solution, unaltered or incompletely altered K-feldspar and sericite are common associates. This is the same association which was described by Lovering for the Boulder County tungsten veins, and which is common in the "porphyry coppers." Sericitization of clay is found farther from the vein than sericitization of K-feldspar.

Small amounts of dickite and alunite are present at Butte in the deeper parts of the Central Zone. These minerals are found both in the vein associated with covellite, pyrite, and digenite, and also as narrow alteration zones partly replacing sericite in the adjacent wall rocks.

The mineralogical changes in the alteration envelopes at Butte take place without evident volume change. Therefore analyses plotted on a volumetric basis (Sales and Meyer, 1948) show that  $SiO_2$  is abundantly leached from the argillized zone but actively precipitated at the sericite front. Alumina stays close to the amount in fresh rock except where it is flushed out by silicification adjacent to a vein. MgO is in a white, essentially dioctahedral mica, apparently chiefly as a result of Mg-Si substitution for Al-Al. White micas showing as much as 2 percent MgO, such as those which replace biotite, are commonly predominantly dioctahedral 1M or 3T polytypes using Yoder and Eugster's (1955) criteria for interpreting powder x-ray patterns. Associated micas replacing clay minerals are generally dominantly 2M polytypes. Those replacing K-feldspar are 1M or 2M polytypes. Both the latter groups usually show less than one-half of one percent MgO on analysis.

The amount of  $K_2O$  in the rock is little changed regardless of the alteration mineralogy. Butte shows less change in  $K_2O$ , zone to zone, however, than most districts of similar alteration mineralogy.

Lime and soda are almost completely removed from the clay and mica zones of alteration at Butte. Except for calcite joint fillings in peripheral areas of the Butte deposit, these constituents have been expelled from the observable part of the hydrothermal system.

*Argillization in the Porphyry Copper Deposits*

The mineralogical types of alteration in the porphyry coppers have been summarized by Schwartz (1947). Briefly, the amount of argillization ranges from virtually zero (Bagdad), to intense, as in Morenci (Schwartz, 1947) or Chino (Kerr, and others, 1950). Virtually all clay mineral groups are represented—allophane, kaolin, montmorillonoid, and the so-called “mica-like” clays. Generally, the deposits having the weaker clay alteration have a higher percentage of montmorillonoid as compared to kaolin, as, for example, in Ajo or Ely. The reverse proportion holds in the porphyries where argillic alteration is more intense.

Chuquicamata and the western part of the Ajo orebody afford examples of well-developed zones of sericite adjoining pyrite-bearing fractures which cut virtually unargillized rock. Chino, Morenci, Bingham, Ely, San Manuel, and others show moderate to intense argillization between the crisscrossing sericite bands. None of the porphyries, however, shows clear-cut zones of argillization parallel to the sericite zones and to the veinlets in a well-defined pattern such as that present in vein deposits in equigranular rocks like Butte, Boulder County tungsten, and Central City. Argillization, whether weak or strong, is much more diffuse and pervasive in the porphyries. Moreover, there is distinct asymmetry on a district-wide scale of the distribution of the various argillic alteration types even with respect to one another, as well as with respect to the mica-dominant types. This is demonstrated by the maps of Schwartz (1953) at San Manuel, Stringham (1953) at Bingham, and Kerr and others (1950) at Santa Rita. Peterson, Gilbert, and Quick (1951), however, found a broadly symmetrical pattern of argillization adjacent to a core of sericitization at Castle Dome, with the ore body to one side of the most intense sericitization.

Many of the writers on alteration in both the porphyry coppers and vein deposits apparently conclude (though with varying degrees of conviction) that intense sericitization—the main muscovite-forming stage—was separated from earlier predominantly argillic stages by a time break during which solutions were changing composition at source, or at least changing below the level of the exposed part of the district in question. Certainly this is one possible interpretation of the patterns cited as evidence. But if the patterns of mineral distribution of alteration products and ores in the natural habitat of structurally and chemically complex mining districts are to be useful in the search for system among the various processes of ore concentration, those patterns must be evaluated in such a way as to avoid mistaking geometrical plausibility for geometrical proof. For example, crisscrossing sericitic bands in a uniform field of clay *may* result from sericitization along fractures which formed after the process of argillization had ceased in the exposed part of the district. But the same pattern *may* result during simultaneous argillization and sericitization if argillic zones merely coalesced during growth at their outer edges to make a uniform field of clay, while the seri-



in Figs 1 and 2. The importance of temperature and the ratio of  $KCl_{aq}/HCl_{aq}$  in defining the stability fields of the mineral phases involved is evident. The values of  $mKCl/mHCl$  for the mica decomposition equilibrium are more than a factor of 10 lower than for the corresponding feldspar equilibrium. These observations indicate that K-feldspar and kaolinite could not exist together stably. Both equilibria are displaced to lower  $mKCl/mHCl$  ratios (greater acidity) with increasing temperature. Thus, at a given  $mKCl/mHCl$  ratio the stability fields of kaolinite, K-mica, and K-feldspar

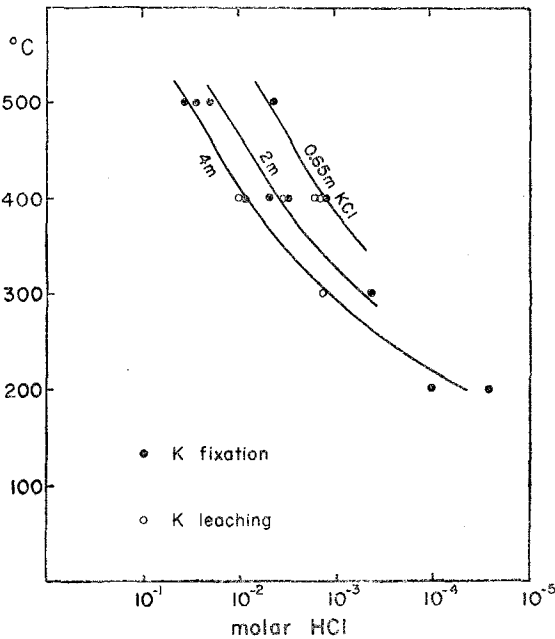


FIGURE 1.—Experimental equilibrium curves for K-feldspar-K-mica-silica. K-fixation denotes approach to equilibrium by conversion of mica plus silica to feldspar. K-leaching denotes the reverse process.

are successively traversed with increasing temperature. Similarly, at constant temperature the same fields are traversed with increasing  $mKCl/mHCl$  ratio. At about  $350^{\circ}C$  (at somewhat higher temperatures in more dilute solutions) mica decomposes to pyrophyllite-boehmite rather than to kaolinite. The transition in assemblage is apparently quite smooth and is indicated by the dashed line in Fig. 2.

The experimental  $mKCl/mHCl$  quotients are approximate expressions of the thermodynamic equilibrium constants of these several reactions and thus express the trend of the equilibrium constants to lower values at higher temperatures. Values of the experimental equilibrium quotients for the mica

and K-feldspar decomposition reactions at 400°C are given in Table 1. The agreement is surprising in view of the difficulties of experimentation.

TABLE 1.—EXPERIMENTAL EQUILIBRIUM QUOTIENTS AT 400°C

	K-feldspar-Mica-Silica	Mica-Pyrophyllite-Boehmite
4 mKCl	$10^{2.66}$	$10^{1.33}$
2 mKCl	$10^{2.70}$	$10^{1.28}$
0.65 mKCl	$10^{2.70}$	$10^{1.30}$

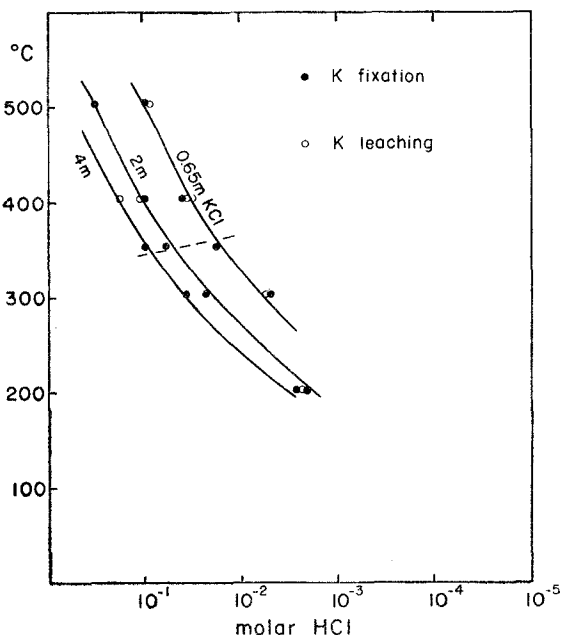


FIGURE 2.—Experimental equilibrium curves for K-mica-kaolinite and K-mica-pyrophyllite-boehmite.

Determination of the thermodynamic equilibrium constants or activity expressions from these data involves extrapolation of activity coefficient ratios of KCl and HCl to the high pressure and temperature conditions of the experiments. Also it will be observed that the activity expression for the mica decomposition reaction below the temperature of formation for pyrophyllite and boehmite would involve  $a_{\text{H}_2\text{O}}$  as well as  $a_{\text{KCl}}$  and  $a_{\text{HCl}}$ . On the basis of the limited thermodynamic data available these extrapolations can not be done accurately, though it appears likely that the experimental



equilibrium quotients would not differ from the true constants by more than a factor of about  $10^{0.5}$  at  $200^\circ$  and  $300^\circ\text{C}$ . The experimental uncertainty of the results appears to be small. The reproducibility and consistency of the data suggest that errors arising from sources such as back reaction on quenching are insignificant. It is unlikely that the errors are negligible, but the uncertainty in the equilibrium quotient may be no more than  $10^{-3}$  or  $10^{-4}$ .

In the application of these experimental results to geological problems, ratios of the activity of aqueous potassium to the activity of acid in the decomposition reactions for K-feldspar and K-mica will be referred to simply as the  $\text{K}^+/\text{H}^+$  ratio. Such overall activities are the important consideration thermodynamically regardless of what species of acids or potassium compounds may actually be present geologically and regardless of their degrees of dissociation.

#### APPLICATION OF EXPERIMENTAL DATA

It has been pointed out in the simplest case of zonal alteration at Butte, that geological evidence indicates contemporaneous zonal growth of sericitic and argillic alteration envelopes outward from each vein during much of the alteration process. The outward migration of each front of alteration presumably was controlled by diffusion rates. The clay minerals in the argillic zone originate chiefly from decomposition of plagioclase; orthoclase remains fresh. But from the equilibrium study it is apparent that K-feldspar and kaolinite can only be metastably associated. In any given cross section of a vein envelope both sericite and clay zones must have been growing in essentially an isothermal (Lovering, 1950) and isobaric environment. Hence K-feldspar persists and the clays are formed from plagioclase in the argillic zone in a P-T environment favorable to white mica. The persistence of the K-feldspar must depend on kinetic factors. A full understanding of the chemistry involved also requires consideration of equilibrium relations between feldspar, mica, and clays when Na-Ca are major components of these phases. Information on these kinetic and equilibrium problems is not yet complete. It would be expected that the alteration products of andesine would be montmorillonite at high alkali/ $\text{H}^+$  ratios, that is, under conditions approaching the equilibrium pH of solutions in the fresh rock. The observed alteration patterns bear this out. At somewhat lower alkali/ $\text{H}^+$  ratios the montmorillonite would be expected to go to kaolinite. Soda mica does not appear at any point, presumably because in this alteration system of fairly high lime content, the activity of soda is never sufficiently high to produce and maintain paragonite. The  $\text{Na}^+/\text{H}^+$  ratio is probably never within the Na-mica stability field.

In the kaolinite subzone the absolute concentration of potassium is probably very low, so that in spite of a  $\text{K}^+/\text{H}^+$  ratio corresponding to the K-mica stability field, the recrystallization of kaolinite to muscovite does not usually take place at a geologically significant rate. The only major K fixation that

occurs is the recrystallization of chlorite to biotite. The existence of  $\text{AlSi}_3\text{O}_{10}$  groups in chlorite may facilitate fixation of the K to make the biotite.

Outward growth of the sericite zone itself, then, must have been controlled by a kinetic process which initiated the breakdown of K-feldspar and the reaction with the kaolinite to form the thermodynamically stable mica. This reaction front may be controlled by a critical value or range of values of the  $\text{K}^+/\text{H}^+$  ratio presumably in the interval between the equilibrium values for the K-feldspar decomposition and the mica decomposition respectively. Values of  $\text{K}^+/\text{H}^+$  lower than the equilibrium values for the decomposition of mica may account in part for the sporadic development of dickite at Butte at the veinward side of the sericitic envelope deep in the Central Zone.

The usually sharp mineralogical transition from the argillic to the sericitic envelopes at high zonal levels at Butte demonstrates the inter-dependence of the mineral-forming reactions there. At this interface three things happened simultaneously: sericitization of K-feldspar and the clay minerals; pyritization and sericitization of biotite; and precipitation of quartz. If there existed a decreasing gradient in acid activity which, for example, would be the result of a diffusion gradient for hydrogen ions outward from the vein, there might exist at some point a critical level of hydrogen ion activity required for activation of a pH-dependent reaction such as decomposition of K-feldspar. The resulting inflection or discontinuity in the  $\text{H}^+$  diffusion gradient may then cause other reactions of a pH-dependent type. Two such reactions may be the breakdown of K-feldspar at a critical  $\text{K}^+/\text{H}^+$  ratio, releasing potash for sericitization, and the pyritization of biotite. A rise in pH with the release of  $\text{K}^+$  ions from K-feldspar would shift the  $\text{H}_2\text{S}$ -water equilibrium in such a way as to give a higher activity of  $\text{S}^{2-}$  ion, which might in turn trigger pyritization of the Fe in the biotite. Similarly, silica migrating toward the vein may be precipitated at this pH front.

The outward growth of the 2 zones of alteration would be sustained so long as there was a continuing supply of hydrothermal fluid at a temperature and  $\text{K}^+/\text{H}^+$  ratio appropriate to muscovite's stability field. Through progressive thermal and chemical reaction each increment of fluid would lose heat and acidity as it migrated outward through the strictly epigenetic part of the hydrothermal system. But over the span of relatively constant temperature and pH at "source," each successive increment of upward migrating fluid to arrive at a specific point of reference on the channel walls might have at that point a higher temperature and acidity than its predecessor, owing to progressive thermal and chemical insulation along its preceding path. The same is true with respect to the chemical diffusion gradients within the wall rocks, so the sericite and clay fronts would gradually grow away from the veins in a migrating steady-state system controlled by diffusion.

Earlier reference was made to the overlap of more rapid outward growth of sericitization of clay than of sericitization of K-feldspar, leading to the association of K-feldspar and muscovite found in the deeper portions of the

Intermediate Zone at Butte. In terms of the equilibrium reactions this circumstance may be controlled by a higher activity of potassium or a higher  $K^+/H^+$  ratio than that which obtained farther away from the "source" of the hydrothermal fluid. The higher activity of potassium would tend to impede destruction of K-feldspar as well as accelerate the sericitization of clay, promoting the sericitization of clay farther from the vein than sericitization of K-feldspar at any given time. It is possible, of course, by extrapolation to visualize a situation approaching the "source" where at moderately high K/H and temperature values, no clay minerals would be developed at all by breakdown of plagioclase, and sericite would be the only alteration. At still higher  $K^+/H^+$  values and temperature, K-feldspars would be stable instead of muscovite. Here channels of circulation would presumably show no alteration envelopes of mica or clay.

In the deepest exposed levels of the Butte district, neither the clay nor the mica is actually eliminated adjacent to the main ore-bearing fractures. However, it is interesting to speculate that the overlap of sericite and orthoclase suggests a first approach to that situation as the "source" is approached. In this connection reference may be made again to the joint fillings of quartz-molybdenite and pyrite which, at exposed levels in Butte, are cut by the ore-bearing structures. It will be remembered that the earliest of these veinlets show only sericitized envelopes of alteration or no alteration of the feldspars at all.

It is clear on structural grounds that these veinlets may represent the transition in time (at a specific point of reference) between the feldspar-stable magmatic crystallization of the quartz monzonite and the feldspar-destructive epigenetic hydrothermal veins. But it is interesting to speculate that these "early" veinlets may also represent the chemical transition at a given *time* between orthomagmatic activity and shallower ore deposition. According to this suggestion the quartz-molybdenite veinlets are cut by the main-stage hydrothermal activity at the level of current exposure only because they were formed earlier *at that level* in the batholith, not necessarily because they preceded hydrothermal activity in the batholith as a whole. At any rate they are suggestive of a changing chemical system under conditions of falling temperature and  $K^+/H^+$  ratio progressing from magmatic to hydrothermal stages at a given level of exposure. A specific increment of hydrothermal fluid may start its migration out of the system at high temperatures stable against the fresh feldspars. But the slope of the stability curves indicates that as it cooled, even at constant  $K^+/H^+$  ratio along its upward path, it could by losing heat readily develop into the leaching hydrothermal fluid which produced the familiar sericitization and argillization in the Butte veins. Evidently convectional supply in the vein channels maintained the level of acidity and temperature necessary to produce sericite as an insulating layer all along the exposed channel, with diffusion in the walls controlling the breakdown of plagioclase to make the argillic zone.

The diffuse distribution of argillization of the plagioclases in the porphyry coppers presents difficult interpretative problems on any hypothesis regarding

timing relative to sulfides and sericite bands. Certainly control of argillization by fractures is much less evident in the porphyries than in the peripheral zone at Butte. Still, broadly speaking, argillization of plagioclase seems strongest in those parts of Ajo and Yerington, for example, where the criss-crossing sericite-bordered veinlets are also best developed. Is the lack of well-developed argillic banding parallel to the fractures merely the result of closer approach to the feldspar stability field, as in the deeper levels at Butte? Suggested answers to this problem may be forthcoming from studies now in progress on the equilibria in systems of plagioclase composition.

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