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ORIGIN OF CHEMICAL VARIATIONS WITHIN IGNIMBRITE COOLING UNITS

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ABSTRACT. Deuteric alteration within large compound ignimbrite cooling units of east-central Nevada redistributed major elements in reactive volcanic glass, creating pronounced vertical chemical variations. K-Na exchanges and Mg and Fe redistributions were the most distinctive deuteric alteration phenomena. Weathering, which is superimposed over the deuteric alteration pattern, affected the more porous zones primarily by leaching sodium and causing growth of Ca-montmorillonite. Local hydrothermal fluids added potassium and removed sodium in the plagioclase phenocrysts and volcanic glass.

INTRODUCTION

Better estimations of the original magmatic compositions of rocks are continually being sought because they can be used in conjunction with other chemical and petrographic data to postulate more closely magmatic chemical and physical conditions. Therefore, it is very critical that a volcanic petrologist should have knowledge of the degree and nature of chemical redistribution mechanisms inherent to post-extrusion alterations. Deuteric fluids alter the original composition of aqueous-rich volcanic units by transfer, exchange, and removal of ions by an aqueous phase during cooling. Hydrothermal activity, commonly unrecognizable in the field, can significantly change the bulk of composition by introduction, redistribution, and removal of ions. Weathering processes, which alter preexisting variation patterns, leach mobile ions and redistribute others by growth of secondary minerals.

The widespread Tertiary ignimbrite sheets of east-central Nevada are particularly suitable for study of post-extrusion alteration. The presence of an aqueous phase during cooling of the ignimbrite created ideal conditions for deuteric alteration. In addition, the degree of welding of ignimbrites is an independent, sensitive record of the temperature and pressure conditions under which deuteric alteration operated. Although the effects of weathering are superimposed upon these ignimbrite sheets, the arid Nevada climate has minimized these effects, leaving the chemical redistribution caused by deuteric alteration relatively undistributed.

A regional correlation by E. F. Cook (1960, 1963, 1966) and the detailed mapping of Tertiary exposures of the Grant Range and vicinity by the author (ms) were used for the sampling program of this study. A location map (fig. 1) shows the positions of measured sections from which samples were obtained for analysis. Figure 2 is a generalization of the Cenozoic stratigraphy of the Grant Range.

Atomic absorption spectrophotometric methods were used to analyze for sodium, potassium, calcium, magnesium, and iron. Silicon and aluminum abundances were measured colorimetrically using methods outlined

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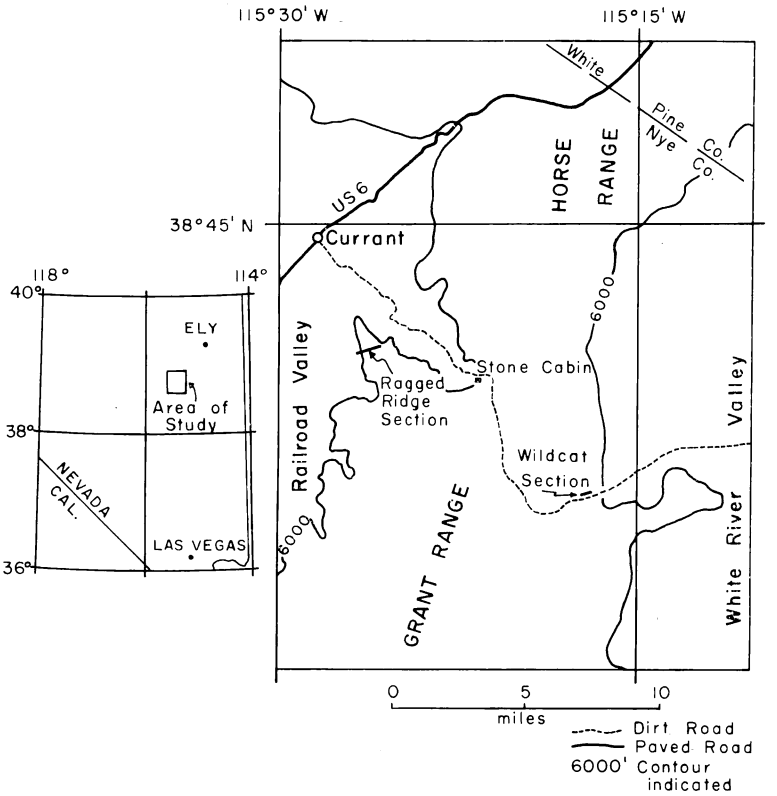


Fig. 1. Map showing the location of measured sections within the Grant Range and an index map of Nevada.

in U. S. Geological Survey Bulletin 1144-A. The analytical precision errors are: Na_2O ± 0.06; K_2O ± 0.05; MgO ± 0.01; CaO ± 0.05; Fe_2O_3 ± 0.12; Al_2O_3 ± 0.8; SiO_2 ± 1.0 weight percent.

CHEMICAL VARIATION WITHIN IGNIMBRITE COOLING UNITS

Distinctive and repetitious trends in chemical variations were found in vertical sequences of samples through cooling units of four major ignimbrite formations in the Grant Range (Scott, 1965), but no lateral trends could be recognized.[1] The closely spaced samples for analysis taken from the compound cooling unit of the Stone Cabin Formation at Wildcat Valley (Wildcat section) demonstrate the degree of vertical variation in greatest detail (pl. 1 and fig. 3).

[1] A table listing the results of the chemical analyses of ignimbrites has been deposited as Document no. 8803 with the ADI, Auxiliary Publications Project, Photoduplication Service, Library of Congress, Washington 25, D. C. A copy may be secured by citing the Document number and by remitting $1.25 for photoprints, or $1.25 for 35 mm microfilm. Advance payment is required. Make checks or money orders payable to: Chief, Photoduplication Service, Library of Congress.

TABLE 1

Na_2O	K_2O	CaO	MgO	Total Fe as Fe_2O_3	Al_2O_3	SiO_2
2.92	5.03	1.57	0.30	1.88	14.0	74

An estimation of the original relative abundance of the major elements of the Stone Cabin formation was made from an average of over seventy selected analyses throughout the Grant Range. These samples obviously leached during weathering or affected by hydrothermal fluids were not included in the averaged analyses. The weight percents of major elements are totalled to 100 percent. Obviously, the water content of the original magmatic material cannot be estimated from analyses because of unknown losses and additions of volatiles during extrusion, transport, welding, and alteration.

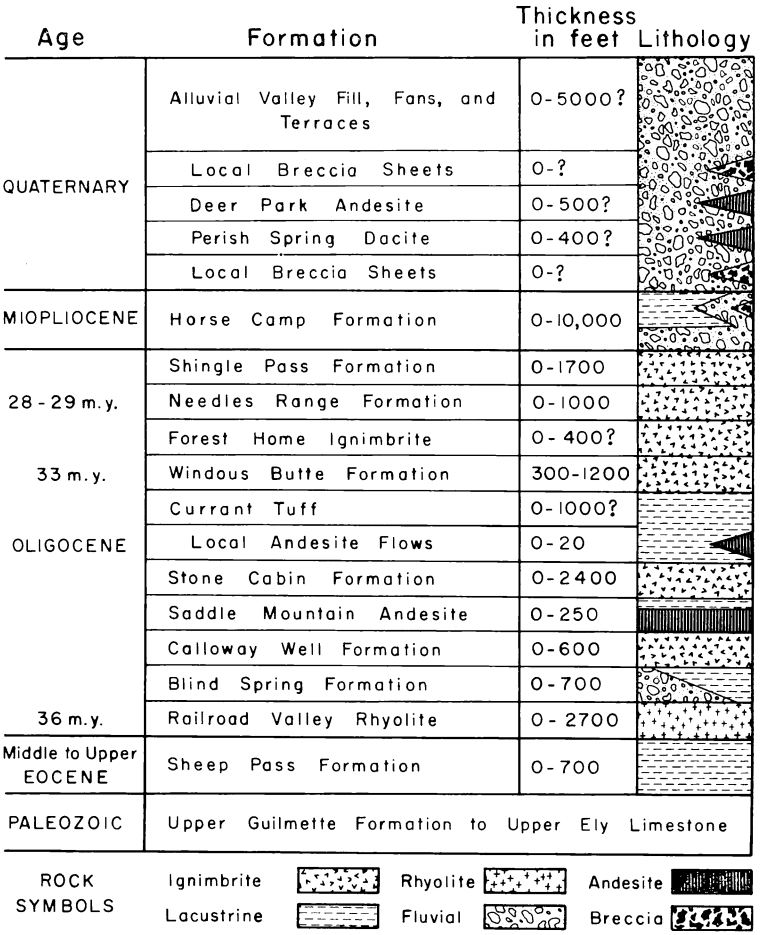


Fig. 2. Generalized Cenozoic section of the Grant Range. Dates in millions of years are published K-Ar dates.

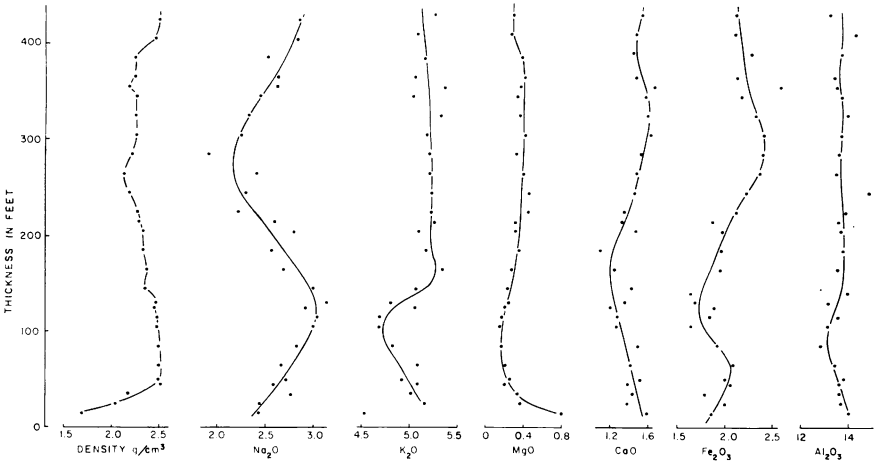


Fig. 3. Chemical and density variations in a vertical section through the Stone Cabin Formation at the Wildcat section. Chemical values are oxide weight percent. Curves on the graphs represent visual estimations of trends suggested by the values shown by points.

The Stone Cabin Formation, a calc-alkalic rhyolite in composition (table 1), is a compound cooling unit consisting of five recognizable individual units (table 2) totaling as much as 2500 feet thick and covering an area of 1500 square miles in the Pancake and Grant Ranges. This ignimbrite contains 43 percent phenocrysts, of which 45 percent are quartz, 30 percent, sanidine (Or_{75}), 20 percent, plagioclase (An_{20-57}), and a trace is biotite and magnetite.[2] The changes in degree of welding of the Stone Cabin Formation at the Wildcat section are shown in plate 1, and variations of density are shown graphically in figure 3. The thick, slightly welded or non-welded upper zones, which are present at more complete but poorly exposed sections, have been eroded at the Wildcat section. The irregular pattern of welding of the cooling unit at the Wildcat section is thought to be the result of at least two extrusive surges separated by a

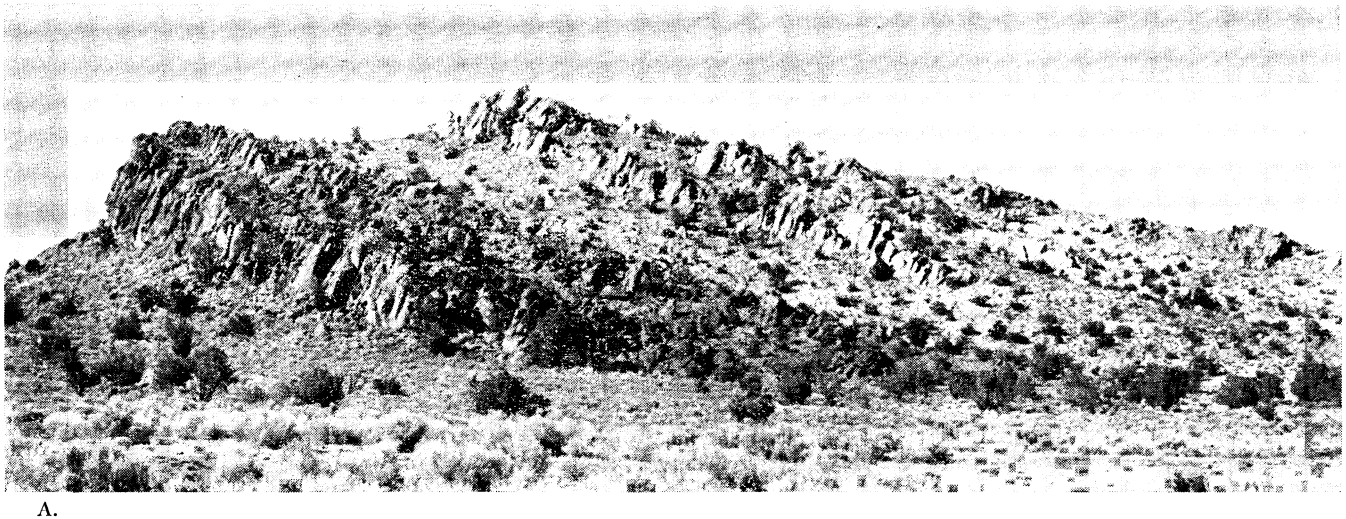
[2] Averaged modal analyses from the 28 samples collected for chemical analysis.

PLATE 1

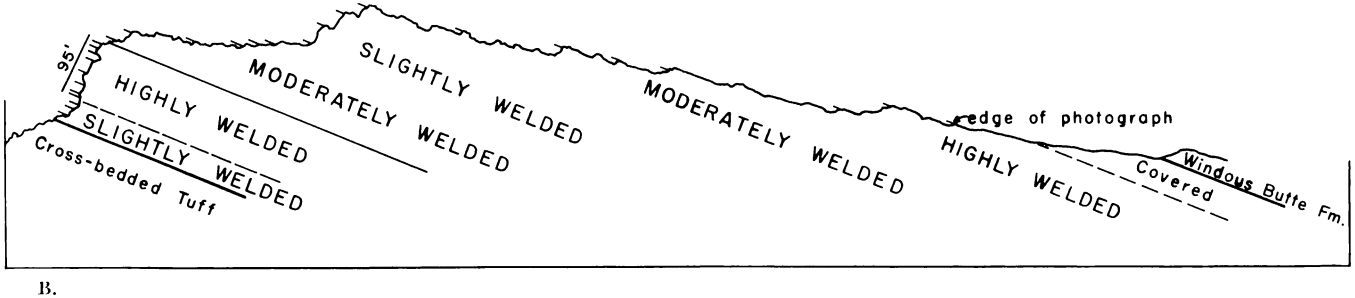
A. Exposures of the Stone Cabin Formation at the Wildcat section as viewed looking north from the dirt road which crosses the Grant Ridge. The ignimbrite is dipping at about 20° to the east.

B. Superimposed upon the outline of the Wildcat section are the zones of welding and the sample locations marked by dashes parallel to the dip of the sheet. The outline is extended eastward beyond the edge of the photograph to the base of the Windous Butte Formation. Below the solid line separating the basal welded zone from the overlying moderately welded zone, the rock is glassy; above the line the rock has been devitrified. The slightly welded zone at the base is commonly called the chilled base. The Stone Cabin Formation rests upon a cross-bedded tuff and underlies the Windous Butte Formation at this locality.

PLATE 1



A.


95'
HIGHLY WELDED
SLIGHTLY WELDED
Cross-bedded Tuff
MODERATELY WELDED
SLIGHTLY WELDED
MODERATELY WELDED
HIGHLY WELDED
edge of photograph
Covered
Windous Butte Fm.

B.

TABLE 2

Unit no.	Sanidine Or%	Plagioclase An%	Degree of zoning of plagioclases	Approx. thickness at Wildcat section
5	68-81	19-47	high	200 +
4	67-69	40-48	moderate	absent at Wildcat section
3	67-73	19-31	slight	100
2	75-78	33-50	high	30
1	72-77	25-30	slight	35

short cooling period. The slightly welded central zone probably represents the top of the first surge which partially cooled; when the second surge was emplaced, this previous top was too cool to become as highly welded as the overlying material.

INDIVIDUAL IGNIMBRITE UNITS WITHIN THE STONE CABIN COMPOUND COOLING UNIT

Gradational vertical changes in bulk chemistry are very pronounced at the Wildcat section (fig. 3).

1. The Na_2O abundance, which varies from 1.9 to 3.1 percent, plots as a smooth curve, increasing from the chilled base to a maximum in the basal welded zone, decreasing to a minimum in the moderately to slightly welded zone, and increasing again where the degree of welding also increases toward the top of the cooling unit.

2. The K_2O abundance ranges from 4.5 to 5.4 percent, decreasing from the chilled base to the most welded basal zone, increasing into the moderately to slightly welded zone, and remaining generally constant above this zone.

3. The graph of MgO content follows a very smooth curve from 0.8 to 0.15 percent. The MgO abundance decreases from the base of the cooling unit to the center of the basal welded zone, increases gradually over the less welded central zone, and decreases again toward the more welded top.

4. The CaO abundance, which ranges from 1.1 to 1.6 percent, decreases from the base to the top of the basal welded zone, increases upward through the slightly welded central zone, and decreases again in the more welded top of the ignimbrite sheet.

5. Total Fe, calculated as Fe_2O_3, has a more irregular trend; it increases between the chilled base and the bottom of the basal welded zone, decreases to the top of the basal welded zone, increases steadily to the upper part of the slightly welded zone, and decreases to the top. The Fe_2O_3 content varies between 1.6 and 2.6 percent.

6. Al_2O_3 is less abundant just below the top of the basal welded zone and the upper welded zone, but elsewhere it is nearly constant at about 13.5 percent. The Al_2O_3 abundance varies between 13.0 and 15.0 percent.

The same general relationships between chemical variations and the degree of welding are shown by other sequences of analyses of samples from the ignimbrites of the Stone Cabin, Windous Butte, Needles Range, and Shingle Pass Formations (see footnote 1).

EXPLANATIONS OF CHEMICAL VARIATIONS

Chemical inhomogeneities within volcanic sheets include those inherent in the original configuration of the unit and those created by post-extrusion alteration. Chemical variations in compound cooling units can be explained in several ways, and the likelihood of different mechanisms is discussed below: (1) The characteristics of the welding and the layering of phenocryst compositions (table 2) of the cooling unit indicate that the tuff was extruded as a series of layers rather than one gush from a vent. Possible original magmatic differences in composition between these layers cannot account for the chemical variations in the Stone Cabin Formation because there are no abrupt changes in the chemical distribution as would be expected if the layers originally had distinct chemistries. (2) Although inhomogeneities in phenocryst composition and distribution do exist (table 2), the changes in phenocryst abundance and composition at the Wildcat section are not of the magnitude or character to explain the observed chemical variations. (3) Hydrothermal alteration by the movement of foreign aqueous fluids through the ignimbrites cannot explain the variations at the Wildcat section because regions subject to hydrothermal alteration do not exhibit the lateral chemical continuity shown by the Stone Cabin Formation. There is no evidence at the Wildcat section that any significant introduction of ions occurred similar to that which affected the hydrothermally altered Ragged Ridge section. (4) Weathering by leaching of some mobile ions and by growth of secondary minerals can be demonstrated; however, the lack of abundant weathering products suggests that the magnitude of weathering alteration is not great enough to explain the large chemical variations found at the Wildcat section. (5) Deuteric alterations probably created most of the chemical variations because the more significant trends of the chemical variations parallel those of laboratory ion exchange experiments and predicted deuteric effects.

Sederholm (1929, p. 870) defines deuteric alterations as "those that have taken place in direct continuation of the consolidation of the magma of the rock itself". Reactions between the hot aqueous phase and glassy phase of an extruded magma are definitely deuteric alterations. Such alterations occurred after emplacement of the ignimbrite sheets during the cooling period and resulted in ionic transfer and exchange between a hot fluid phase and the unstable, reactive volcanic glass. The zones of devitrification, vapor phase crystallization, and fumarolic activity in ignimbrite cooling units described by Smith (1960) are products of deuteric activity.

The types of ionic exchanges and reactions which occurred at different zones within the cooling unit probably were controlled by the

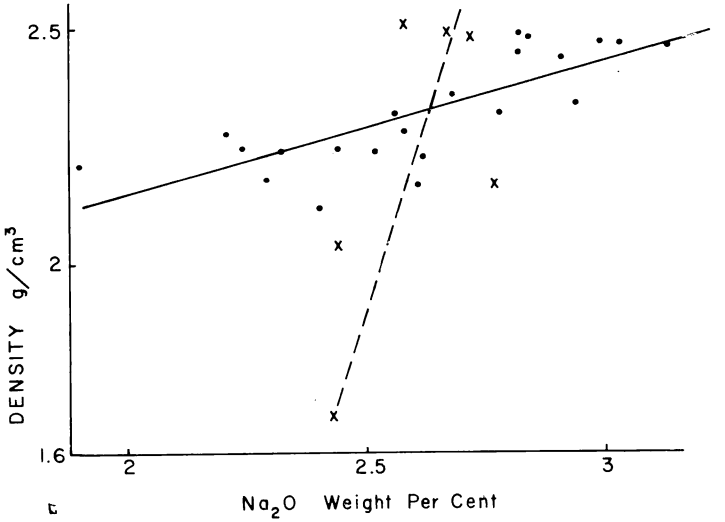


Fig. 4. Relationships between density and Na_2O distribution. The solid line and solid circles represent the relationship of values from above the center of the black glass; the dashed line and crosses represent the relationship of those from below the center of the black glass. All linear curves drawn in figures 4, 5, 6, and 7 are best fit regression lines.

temperature gradient, confining pressure, water pressure, composition of the altering fluids, and composition of the volcanic glass. It is particularly significant that the process of welding, that is, the compaction of hot plastic glass shards, was controlled by many of the same physical and chemical factors as was deuteric alteration. Figures 4, 5, and 6 show that the degree of welding, expressed as density, definitely is related to the vertical chemical variations in the cooling unit at Wildcat section. The Na_2O content varies directly with density above the black glass of the basal welded zone but varies inversely and only slightly with density below the black glass zone (fig. 4). The K_2O content, with a minor exception at the chilled base, varies inversely with density. The MgO abundance varies inversely with density throughout the unit (fig. 5). Except for the chilled base and the top of the section, the CaO percent varies inversely with density. The iron content, calculated as Fe_2O_3, increases directly with density from the chilled base to the black glass of the basal welded zone but varies inversely with density above the black glass zone (fig. 6).

A distinction between the superimposed effects of deuteric alteration and weathering is difficult because the degree of weathering is also related to the degree of welding; the more porous, poorly welded zones of the ignimbrite sheet are more weathered than highly welded zones. It is postulated that those chemical trends that can most readily be explained by ionic exchanges should be attributed to deuteric alteration, whereas

those that show evidence of leaching or show association with weathering products should be attributed to weathering. Deuteric alteration probably controlled most of the sodium and potassium variations in the more welded zones of the cooling unit, but leaching of sodium by weathering greatly affected the less welded zones; this is demonstrated by the two distinct relationships shown in the graph of the moles of sodium per kilogram of rock versus the moles of potassium per kilogram of rock (fig. 7). The Na/K ratios of samples from the more highly welded zones in the cooling unit form the first trend; these analyses fall along the line with the slope close to –1, which would be the expected result from an ion-for-ion exchange between sodium and potassium. Orville (1963) has demonstrated experimentally that potassium in an aqueous fluid phase is exchanged with sodium in the solid alkali feldspar phase as the temperature is decreased. Appropriately, the moderately to slightly welded and presumably cooler parts of the ignimbrite sheet are richer in K_2O than are the more highly welded zones, which are richer in Na_2O. The

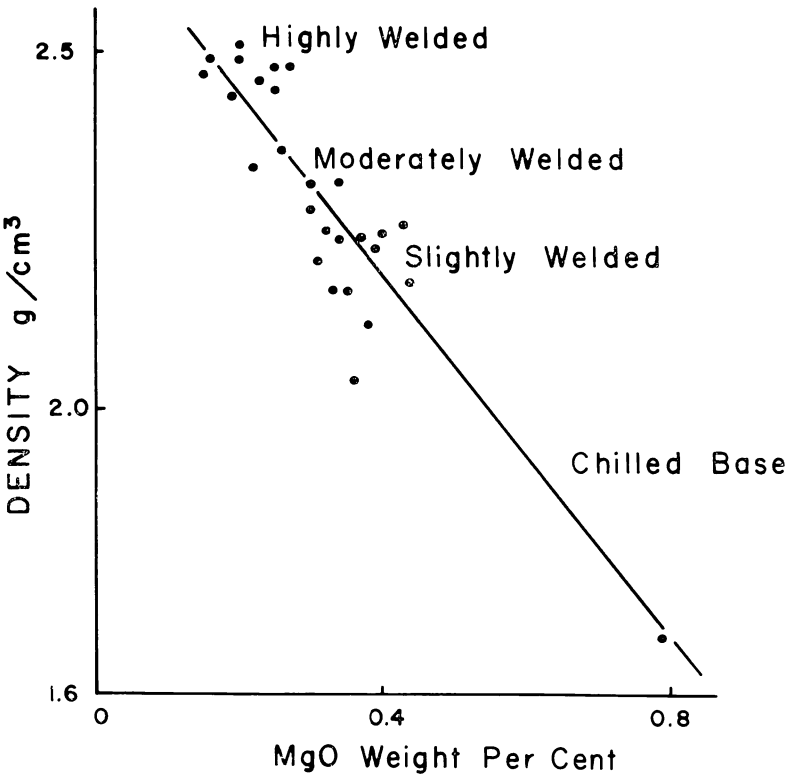


Fig. 5. Density and MgO distribution relationship. The gap in density and MgO content between the one point in the lower right corner and the remaining points indicates that large changes in both values exist over only 10 feet of section in the chilled base.

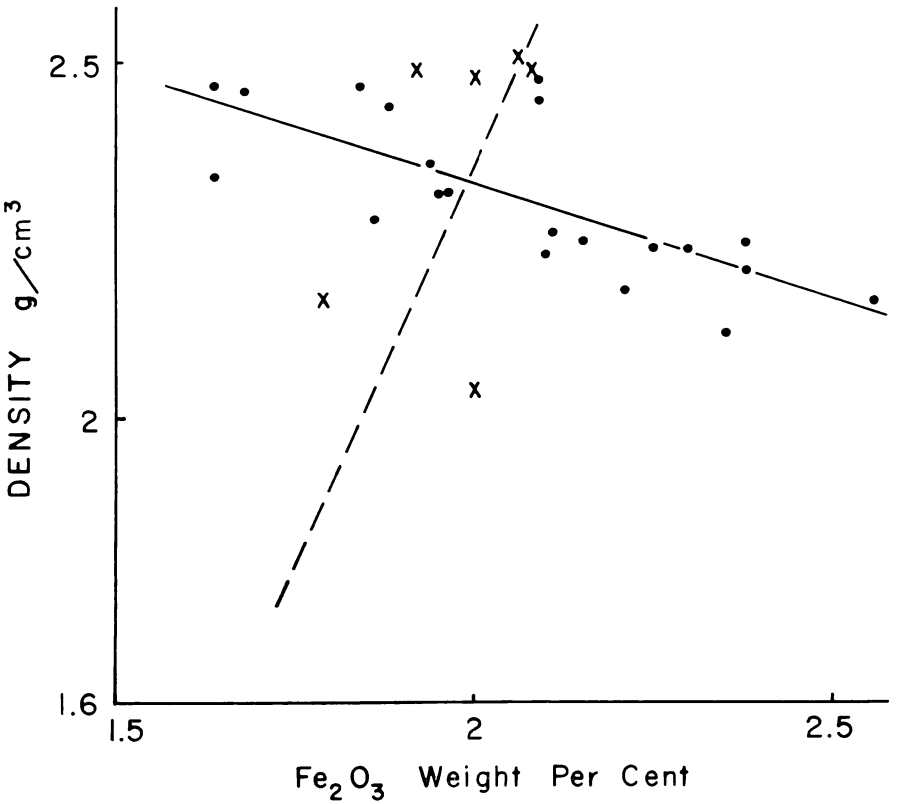


Fig. 6. Relationship between density and Fe_2O_3 distribution. The values above the center of the black glass zone are represented by solid circles and a solid line. The dashed line and crosses show the relationship for values below the center of the black glass zone.

author postulates that a deuteric alteration exchange phenomenon similar to that experimentally produced by Orville operated in the more welded portions of the ignimbrite cooling unit, causing most of the potassium and sodium variation. The experimental exchange was between an aqueous phase and alkali feldspars, but the natural one was between an aqueous phase and a presumably unstable, reactive volcanic glass. Lipman (1965) found similar inverse relationships between the K and Na distribution in ignimbrites of the Nevada Test Site.

The second trend in figure 7 is formed by the remaining data from analyses of the more poorly welded parts of the cooling unit. This trend shows almost no potassium variations in rocks relatively low in sodium, thus suggesting that sodium, which is particularly mobile during incipient weathering, has been selectively leached from the porous zones in the ignimbrite sheet. Plagioclase feldspars have been altered during weathering (and possibly deuteric alteration) by the growth of calcite

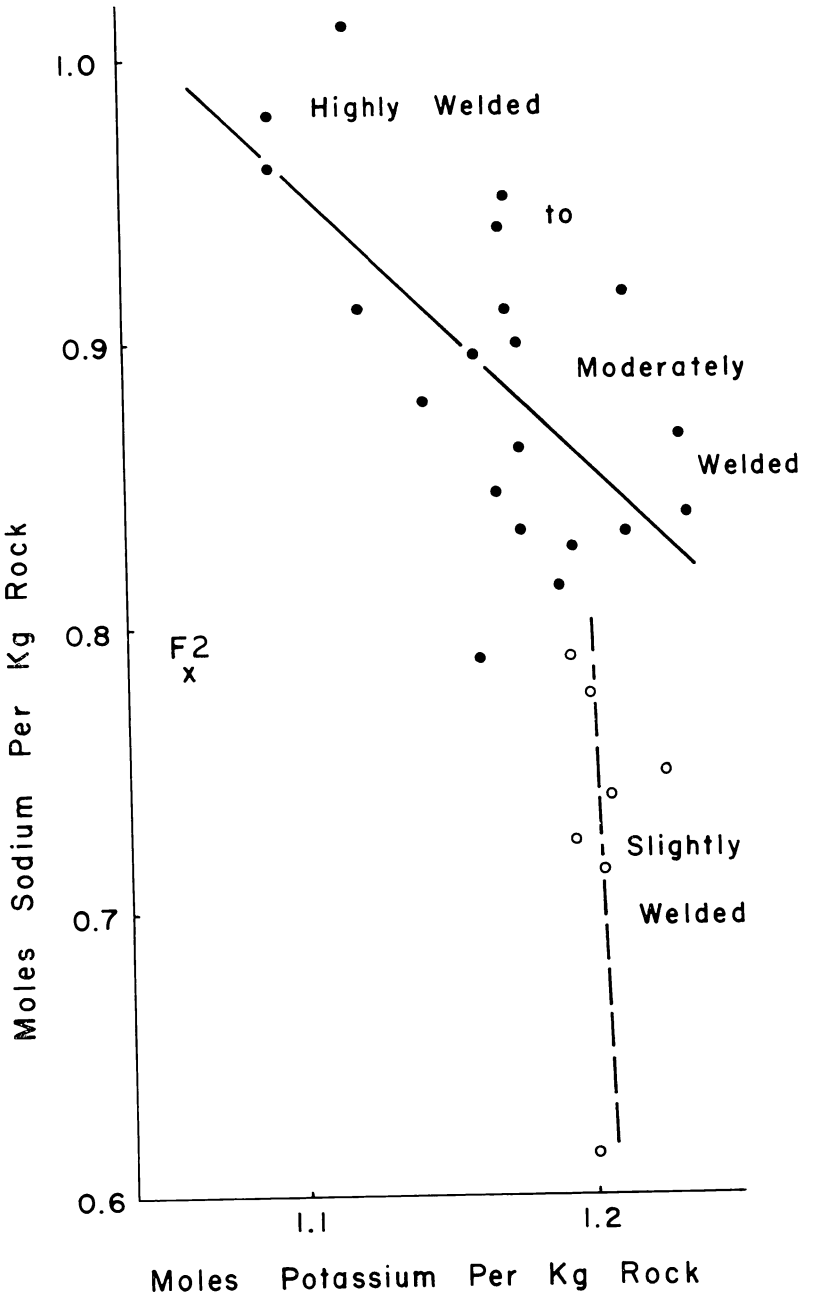


Fig. 7. Moles of potassium per kilogram of rock versus moles of sodium per kilogram of rock. The trend of highly and moderately welded samples is shown by the solid line and solid circles; slightly welded samples marked by open circles form the dashed trend. Sample F2 was collected from the base of the entire section.

and Ca-montmorillonite and by the leaching of sodium. In a study of weathering of a gneissic granite, Goldich (1938) found a similar leaching of sodium during early stages of weathering and noticed that plagioclase feldspars were affected by weathering before the alkali feldspars. Porous, poorly welded tuffs on the Nevada Test Site are low in both sodium and silicon; correspondingly, analyses of ground water taken from these tuffs are highest in silicon and sodium, indicating that leaching of these elements is probable (Lipman, 1965). The extremely porous and highly weathered sample at the chilled base (F2) is low in both sodium and potassium; this can be interpreted as an indication of fairly intense weathering.

Magnesium variations at the Wildcat section are probably the result of deuteric alteration, although magnesium is among the more mobile ions in most studied examples of weathering. If the magnesium was mobile during weathering, it would be leached from the more porous zones; however, the magnesium content is higher in the more porous, weathered zones of the tuff (fig. 5). An undetermined deuteric alteration mechanism must have controlled the migration of magnesium. This variation of magnesium content from some presumably homogeneous, original content occurred in the volcanic glass rather than in the magnesium-bearing phenocrysts. Unaltered biotite phenocrysts, which are the only magnesium-bearing phenocrysts in the Stone Cabin Formation, are homogeneously distributed and contain only one third of the average MgO content in the rock. The sites of magnesium gains and losses are not identifiable in the volcanic glass. Lipman (1964) has suggested that some of the magnesium may be in montmorillonite, which is more abundant in porous zones. To explain all the magnesium variations, however, 20 to 40 percent of the glass would have to be altered to montmorillonite in the most porous zone. The poor quality basal X-ray diffraction peaks observed certainly do not suggest such an abundance of montmorillonite.

The ferric oxide abundance, which generally varies inversely with density throughout the cooling unit, with the exception of the chilled base (fig. 6), may be partly controlled by movement of iron in a fluid phase during deuteric alteration. Iron oxides, mostly hematite, fill joints and small voids and coat glass shards and phenocrysts in the more porous zones of the ignimbrite sheet. In the basal chilled zone (figs. 3 and 6) the iron has a direct relationship to density; this may represent some leaching as ferrous iron by cooler acidic deuteric solutions.

Calcium variations do not have strong direct or inverse relationships to the degree of welding. Although gradational trends are distinct, more scatter is found in the CaO values than in other oxides (fig. 3). Plagioclase phenocrysts and devitrified glass shards are partially replaced with calcite and Ca-montmorillonite in the more porous samples. This evidence suggests that most of the calcium variations are the result of weathering which mobilized and redeposited the calcium in patches of

calcite and montmorillonite. Pettijohn (1949, p. 379) reports that in several weathering studies calcium is one of the most mobile ions.

PRODUCTS OF ALTERATION

Several alteration minerals have been identified, although X-ray graphs of impure separates of devitrified glasses and thin sections were difficult to interpret. In the devitrified glass and in small vugs or vesicles, a few wedge-shaped tridymite crystals were found in thin sections but were not detected by X-ray diffraction. Abundant radial growths of chalcedony are easily recognizable in highly devitrified glasses. Alkali feldspars (Or_{36}) have been identified by X-ray diffraction in glass separates but were not recognized in thin section. Hematite dust or stain outlines glass shards in the more porous zones of the cooling unit. Ca-montmorillonite was found in the porous weathered glass of the tuff, in weathered pumice fragments, and flattened vesicles. No pyroxene microlites were identified in any of the ignimbrite glass shards.

THICKNESS CONTROL OF THE DEGREE OF DEUTERIC ALTERATION

Lipman and Christiansen (1964) found relatively little chemical variation in a thin, simple cooling unit, the Yucca Mountain member of the Piapi Canyon formation in southern Nevada. Leaching of sodium and the growth of Ca-montmorillonite observed by Lipman and Christiansen are probably the result of weathering. As the authors pointed out, "no measurable effect on the bulk composition of the rocks" by deuteric alteration was found, with the possible exception of magnesium. Their magnesium value for the chilled basal zone was significantly higher than those from the upper zones. Magnesium values from the Wildcat section are in agreement with this trend and magnesium was more extensively redistributed relative to its abundance than other major elements in the thick Grant Range ignimbrites. Lipman and Christiansen concluded that the deuteric activity did not significantly affect the major element distribution. The Yucca Mountain cooling unit is thinner (maximum of 250 feet thick) than the Stone Cabin cooling unit and does not exhibit the vitrophyric black glass zone, which is about 95 feet thick in the Stone Cabin Formation (pl. 1). Evidently, the Stone Cabin Formation cooled over a much greater period of time, allowing more deuteric fluid to percolate through the reactive volcanic glass. Possibly the thin Yucca Mountain member lost most of its deuterically active fluid by evaporation into the atmosphere or much of its original heat during transportation before welding.

HYDROTHERMAL ALTERATION

An introduction of potassium and a removal of sodium by hydrothermal fluids occurred at Ragged Ridge, affecting the ignimbrites of the Stone Cabin Formation through the Shingle Pass Formation. About 1.9 weight percent Na_2O has been lost on an average at Ragged Ridge, and 3.4 percent K_2O has been added on an average. The resulting abnormal

alkali content (about 9 per cent K_2O and 1 percent Na_2O) must be a result of alteration because no known magmatic process can produce this composition. The plot of moles of sodium against moles of potassium (fig. 8) has a negative slope similar to that found at Wildcat section. Replaced plagioclase rims, which are found at Ragged Ridge but are absent elsewhere in the Grant Range, optically appear to be alkali feldspar with a $2V_x = 26°$. The Or_{90} alkali feldspar measured in some of the Ragged Ridge samples by X-ray, in addition to the normal Or_{70-80} sanidine, has been interpreted as the rim around the plagioclase phenocrysts.

It is proposed that an exchange process similar to that experimentally achieved by Orville (1963) controlled most of the movement of sodium and potassium during hydrothermal alteration of the type that affected the part of Ragged Ridge sampled for this study. Potassium-rich hydrothermal waters probably percolated through these ignimbrites, exchanged the introduced potassium for sodium in the rims of plagioclase phenocrysts and in the glass, and carried the dissolved sodium elsewhere. Lipman (1965) found comparable potassium introduction in hydrothermally altered tuffs. The exchange occurred to a greater degree in more porous, poorly welded zones than in highly welded ones (fig. 8).

The distribution of the other oxides, CaO, MgO, and Fe_2O_3, is more erratic at the Ragged Ridge section than at regions unaffected by hydrothermal fluids, but the same average abundance is present. Hornblende and biotite phenocrysts in the more mafic-rich formations are highly oxidized. The ground-mass of all the ignimbrites at Ragged Ridge show relic shard texture only faintly; the growth of tiny secondary feathery crystals is much more pronounced.

SUMMARY AND CONCLUSIONS

The pronounced vertical chemical variations found in the large ignimbrite cooling units of the Grant Range are probably caused by a combination of deuteric alteration and weathering. The characteristic relationship between sodium and potassium can readily be explained by an ionic exchange of sodium and potassium between deuteric fluids and reactive volcanic glass. Sodium was leached by weathering from highly porous samples. The magnesium distribution is the result of an undetermined deuteric mechanism. Much of the iron distribution pattern may have resulted from deuteric action that dusted glass shards and phenocrysts with hematite in porous portions of the tuff. Typical weathering products such as Ca-montmorillonite and calcite in both glass and plagioclase phenocrysts indicate that weathering agents affected the calcium distribution, but the relative degree of weathering or deuteric action on calcium cannot be determined. Also the aluminum pattern cannot be attributed definitely to a particular alteration mechanism.

The patterns produced by deuteric and weathering alteration form distinct direct and inverse relationships with the density or degree of welding of the ignimbrite. Deuteric alteration was probably controlled

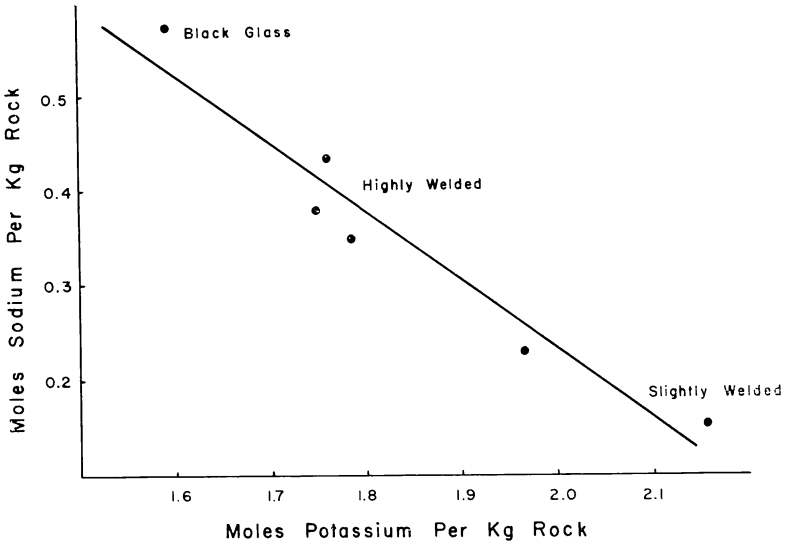


Fig. 8. Relationship between sodium and potassium at the hydrothermally altered Windous Butte Formation at Ragged Ridge. An average of 0.6 moles of sodium per kilogram of rock has been lost, and an average of 0.7 moles of potassium per kilogram of rock has been added to the Windous Butte Formation at Ragged Ridge.

by temperature, pressure, and other factors which also controlled the welding of the tuff. The degree of weathering also is related to the degree of welding because more porous tuffs are more easily weathered.

Local hydrothermal alteration of Ragged Ridge was characterized by the introduction of potassium into the rock by hydrothermal fluids, an ionic exchange of potassium for sodium in the plagioclase phenocrysts and in the glass, and a removal of that sodium from the rock.

ACKNOWLEDGMENTS

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