

## GEOCHEMISTRY OF HYDROTHERMAL CHLORITE REPLACING IGNEOUS BIOTITE

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**Abstract**—Hydrothermal chlorite replaces igneous biotite in the Gold Hill, Utah, quartz monzonite. Chemical compositions of coexisting biotite and chlorite determined by electron microprobe and wet chemical methods were used to evaluate chemical mass transfer during the alteration process. The mole ratio Mg/(Mg + Fe) varies from 0.52 to 0.65 in the chlorite and from 0.51 to 0.60 in the parent biotite. The Mg content of the chlorite decreases systematically with increase in the volume percent replacement of biotite. Homogenization temperatures of fluid inclusions in nearby quartz microveinlets indicate that the chloritic alteration took place at approximately 200°C.

Textural relationships suggest that the alteration of biotite to chlorite is isovolumetric, but a comparison of mineral compositions and mineral assemblages with phase diagrams in which Al or volume are conserved among solid phases suggests that the chlorite compositions are best explained as a function of reaction progress in an Al-conservative system. The chlorite composition changes in response to changes in solution composition produced by the dissolution of successive small amounts of biotite. Representative mass balance for the alteration of all of the biotite to chlorite in 1 m<sup>3</sup> of rock containing 336 moles of biotite indicates that 74 moles of Mg, 35 moles of Fe<sup>3+</sup>, 420 moles of H<sup>+</sup>, and 2 moles of Mn are added to the rock and that 311 moles of K, 54 moles of Fe<sup>2+</sup>, 76 moles of Ti, 53 moles of F, and 6 moles of Cl are lost to solution. The mass transfer for partially altered biotite is 11 to 188 moles of K, 2 to 46 moles of Ti, 2 to 44 moles of F, and 0.3 to 6 moles of Cl removed per cubic meter of rock and 1 to 26 moles of Fe<sup>3+</sup> and 20 to 347 moles of H<sup>+</sup> added. The mass transfer of Mg varies from 12 moles added to 32 moles removed per cubic meter of rock depending on mineral composition and extent of replacement.

**Key Words**—Aluminum conservative alteration, Biotite, Chlorite, Electron probe analysis, Fluid inclusions, Hydrothermal alteration, Isovolumetric alteration.

### INTRODUCTION

Hydrothermal chlorite is ubiquitous in propylitic alteration halos associated with many hydrothermal ore deposits. An early summary of the products of hydrothermal alteration of biotite by Schwarz (1958) showed that chlorite was always present. Sales and Meyer (1948) documented biotite alteration to chlorite, rutile, carbonate, epidote, and magnetite and also observed hydrothermal biotite replacing chlorite pseudomorphs of igneous biotite. All twenty of the porphyry copper deposits for which Lowell and Guilbert (1970) reported the mineralogy of outer alteration zones contain chlorite.

Systematic variations in chlorite composition with position relative to known ore bodies have been described in massive sulphide deposits in the Noranda district, Quebec (Price, 1953). Nakamura (1960) found distinctly different chlorite compositions in base metal veins and associated wall rocks; the mole fraction of clinocllore is 0.05 to 0.06 in veins and 0.56 to 0.34 in the wall rocks. Chlorite in metamorphic rocks near the Jerome, Arizona, massive sulphide deposit varies in composition with distance from the known ores, although post-ore regional metamorphism has complicated the interpretation (Nash, 1973). Biotite in granitic rocks from Maine has been converted to chlorite and sphene (Ferry, 1979). Ferry's assumption that Al and

Ti are conserved during the mass transfer accompanying the hydrothermal alteration of the biotite results in an estimated 13% decrease in the volume of solids.

The purpose of the present study was to examine the chemistry of the replacement of biotite by chlorite in the Gold Hill, Utah, pluton using electron microprobe and wet chemical analyses. The temperature of the replacement reaction was determined from homogenization temperatures of fluid inclusions in nearby quartz microveinlets. The systematic variation in mineral composition was correlated with the extent of biotite replacement and with solution chemistry. Chemical reactions in which either Al or volume were conserved among solid phases were formulated to estimate the mass transfer which accompanied the biotite to chlorite reaction.

### GENERAL GEOLOGY

The Gold Hill mining district and pluton are located in Tooele County, northwestern Utah, 88 km south of Wendover. The geology and mineralogy of the Gold Hill district have been described by Butler (1920), Nolan (1935), El-Shatoury (1967), and El-Shatoury and Whelan (1970) and will be dealt with only briefly here. The bedrock of the Gold Hill area consists of Paleozoic sedimentary rocks intruded by the Gold Hill quartz monzonite and related aplite and porphyry dikes, all of

which are probably of Tertiary age (El-Shatoury, 1967). Volcanic rocks are present in scattered exposures. The Paleozoic rocks have undergone complex high-angle and low-angle faulting and are host for numerous small, precious- and base-metal vein and replacement deposits and two significant contact metamorphic base-metal deposits. The quartz monzonite stock contains many small mineralized fissures, tungsten-molybdenum pipes, and adularia + quartz + calcite veins containing minor amounts of beryllium. Chlorite replacement of igneous biotite within the quartz monzonite considered here is related to the adularia + quartz + calcite veins and increases in abundance near these veins. Chlorite also replaces biotite near contact metamorphic deposits and small veins within the stock. Samples for this study were collected from outcrops of the pluton.

#### ANALYTICAL PROCEDURES

Analyses of Si, Al, Ti, Mg, Fe, Mn, K, Ca, Na, Ba, Cl, and F in coexisting biotite and chlorite were obtained from polished thin sections with an ARL model EMX electron microprobe. Raw probe data were collected to a constant integrated beam-current-time product, and 10 spots 10  $\mu\text{m}$  in diameter were analyzed on each biotite or chlorite grain. Background-corrected probe data were corrected for matrix effects using the factors of Albee and Ray (1970). Standards used were kaersutite for Ca and Na, scapolite for Cl, fluorophlogopite for F, and analyzed biotite specimens for the remaining elements. Because of the matrix effect of (OH) in biotite and chlorite, an iterative computational procedure was used for the matrix correction. After a structural formula was computed from an initial estimate of mineral composition, the wt. % water was calculated from that structural formula, followed by recalculation of matrix effects, composition, and a new structural formula. Three iterations usually produced satisfactory convergence. This procedure resulted in the inclusion of water not determined in probe analysis in the matrix correction and an estimate of wt. % water consistent with mineral composition.

Chlorite separates for X-ray powder diffraction and wet chemical analysis were made from quartz monzonite in which practically all of the biotite had been altered to chlorite. The separation procedure involved grinding, sieving to 80  $\times$  115 mesh, magnetic separation, and jiggling on smooth paper. Chlorite and biotite separates with less than 5% impurities were analyzed for ferrous iron by the method of Peck (1964) and for total iron by atomic absorption spectrometry. Ferric iron was calculated by difference.

#### PETROGRAPHY

The Gold Hill stock varies from granite to granodiorite in composition and from fine-grained to coarse-grained porphyritic in texture. The dominant phenocryst is euhedral pink orthoclase up to 5 mm in length.

Groundmass minerals are quartz, plagioclase ( $\text{An}_0$  to  $\text{An}_{30}$ ), orthoclase, perthite, and minor microcline, biotite, and hornblende. Common accessory minerals are magnetite, zircon, sphene, and apatite. Two textural types of biotite are present in the quartz monzonite: subhedral, solitary biotite crystals and biotite aggregates with hornblende cores or within hornblende grains. Both textural types of biotite are altered to chlorite. More rarely, hornblende and plagioclase ( $\text{An}_{30}$ ) are altered to chlorite. Chlorite is commonly accompanied by rutile, calcite, and sphene and rarely by epidote, muscovite, quartz, and potassium feldspar. The biotite and chlorite analyses presented here are of subhedral, solitary biotite crystals and chlorite which pseudomorphically replaces subhedral biotite.

The most abundant opaque mineral is magnetite with hematite intergrowths. Chalcopyrite was observed in association with one chlorite + quartz + rutile pseudomorph of biotite, and coexisting bornite + chalcopyrite + covellite was noted as an inclusion in one magnetite grain. Pyrite inclusions in quartz and plagioclase were observed in all samples, but they are not abundant. Limonite pseudomorphs after pyrite with occasional remnants of pyrite were observed in most samples, but do not exceed 1% in any one sample. No petrographic evidence for significant volume change that accompanied the alteration of biotite to chlorite was observed.

#### MINERAL COMPOSITIONS

Compositions of biotite and chlorite replacing biotite are presented in Table 1 and Table 2. The compositions of coexisting chlorite and biotite are plotted in Figure 1 along with the compositions of biotite grains showing no chlorite alteration. Tie lines connect coexisting biotite and chlorite. Mole ratios of Mg/(Mg + Fe total) of the biotite range from 0.51 to 0.60 and are comparable to the compositions of igneous biotite in similar rocks elsewhere in the Basin and Range Province (Jacobs and Parry, 1976). The mole ratios of Mg/(Mg + Fe total) of chlorite range from 0.52 to 0.65; tetrahedral silicon varies from 2.77 to 3.08 atoms per half unit cell. Chlorite compositions plot in the pynochlorite-ripidolite field in the classification of Hey (1954) or in the brunsvigite field in the classification of Foster (1962). The chlorites contain much less Ti, F, Ba, and Cl than the coexisting biotites. The Al-Fe-Mg plot of Figure 1 shows that a chlorite may be either higher or lower in Mg than the coexisting biotite—Mg-enriched chlorite tends to occur with Mg-rich biotite. Three Fe-rich chlorites coexisting with lower Mg-biotites are plotted. Two chlorites contain about the same proportion of Mg as their biotite host but are enriched in Al.

The volume percentage of biotite grains replaced by chlorite is shown in Figure 1. High-Mg chlorite replaces 4–10 volume % (mean = 8%) of host biotite grains; high-Fe chlorite replace 48–61 volume % (mean =

Table 1. Microprobe analyses of biotite from the Gold Hill pluton, Utah.

	20-1	20-3	20-4	20-5	20-6	20-12	20-13	20-15	21-7	21-13	21-14	21-15	22-1	22-2	23-8	24-2	24-5	24-6	24-7
SiO <sub>2</sub>	37.0	38.2	37.8	38.1	38.1	37.5	37.7	38.6	37.9	38.0	38.2	38.2	37.5	37.5	38.5	36.4	36.3	37.2	36.3
TiO <sub>2</sub>	4.1	3.7	3.8	3.9	3.9	3.6	3.9	3.6	4.0	3.2	3.2	3.6	2.9	2.6	3.8	3.8	3.9	2.9	3.7
Al <sub>2</sub> O <sub>3</sub>	12.6	12.1	12.4	12.7	12.6	12.5	12.5	12.4	13.0	13.3	13.2	12.2	13.7	13.9	12.2	13.2	13.0	13.5	13.5
FeO <sup>1</sup>	16.9	17.2	17.2	16.8	17.6	17.5	17.3	17.2	17.1	16.9	17.1	16.9	18.3	18.8	17.0	17.9	17.8	17.1	18.2
Fe <sub>2</sub> O <sub>3</sub> <sup>1</sup>	0.54	0.55	0.55	0.54	0.56	0.56	0.55	0.55	0.55	0.54	0.55	0.54	0.58	0.60	0.54	0.57	0.57	0.55	0.58
MnO	0.32	0.29	0.28	0.36	0.28	0.30	0.32	0.29	0.30	0.30	0.30	0.30	0.30	0.30	0.20	0.30	0.30	0.30	0.30
MgO	13.3	13.2	13.7	12.9	13.0	13.5	13.0	13.3	13.9	14.5	14.7	14.0	11.8	11.4	13.4	13.5	13.1	13.6	13.0
CaO	0.18	0.17	0.17	0.11	0.09	0.11	0.09	0.12	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15	0.15
K <sub>2</sub> O	9.7	9.2	9.6	9.4	9.4	9.4	9.4	9.5	9.0	9.0	8.5	9.0	9.7	9.7	9.5	9.8	9.5	9.9	9.7
Na <sub>2</sub> O	0.13	0.13	0.12	0.12	0.16	0.18	0.16	0.16	0.16	0.16	0.16	0.16	0.16	0.16	0.16	0.16	0.16	0.16	0.16
BaO	0.27	0.34	0.22	0.28	0.33	0.25	0.24	0.35	0.25	0.25	0.25	0.25	0.25	0.25	0.34	0.16	0.38	0.13	0.25
Cl	0.18	0.17	0.15	0.18	0.18	0.19	0.17	0.19	0.19	0.19	0.19	0.19	0.19	0.19	0.19	0.19	0.19	0.19	0.19
F	0.82	0.79	0.83	0.80	0.79	0.82	0.75	0.85	0.82	0.82	0.81	0.82	0.81	0.81	0.71	0.97	0.98	0.93	0.92
H <sub>2</sub> O <sup>2</sup>	3.5	3.6	3.5	3.6	3.5	3.5	3.6	3.5	3.5	3.6	3.6	3.5	3.5	3.5	3.6	3.4	3.4	3.5	3.4
F:Cl = O	0.38	0.37	0.39	0.38	0.37	0.39	0.36	0.40	0.39	0.39	0.39	0.39	0.38	0.38	0.34	0.45	0.45	0.44	0.43
Total	99.2	99.3	99.9	99.4	100	99.5	99.3	100	100	100	101	99.4	99.5	99.5	100	100	99.3	99.7	99.9

	Number of ions based on 10 oxygens and 2 (OH, Cl, F)																			
Si	2.83	2.90	2.86	2.87	2.87	2.86	2.87	2.84	2.84	2.84	2.85	2.89	2.87	2.87	2.91	2.77	2.79	2.83	2.77	
Al (IV)	1.14	1.09	1.10	1.11	1.12	1.12	1.12	1.09	1.15	1.16	1.15	1.09	1.13	1.13	1.09	1.18	1.18	1.17	1.21	
Al (VI)	0.00	0.00	0.00	0.03	0.00	0.00	0.01	0.01	0.00	0.01	0.01	0.00	0.11	0.13	0.00	0.00	0.00	0.04	0.00	
Ti	0.24	0.21	0.22	0.22	0.22	0.21	0.22	0.21	0.23	0.18	0.18	0.20	0.17	0.15	0.22	0.22	0.23	0.17	0.21	
Fe <sup>3+</sup>	1.08	1.10	1.09	1.06	1.11	1.12	1.10	1.09	1.08	1.06	1.06	1.07	1.17	1.21	1.07	1.14	1.14	1.09	1.16	
Fe <sup>2+</sup>	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.04	0.03	0.03	0.03	0.03	0.03	
Mn	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	
Mg	1.52	1.50	1.55	1.46	1.47	1.53	1.48	1.49	1.55	1.62	1.64	1.58	1.34	1.30	1.51	1.54	1.50	1.54	1.48	
Ca	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	
Na	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	
K	0.95	0.89	0.93	0.91	0.90	0.92	0.91	0.86	0.86	0.86	0.81	0.87	0.95	0.94	0.91	0.96	0.93	0.96	0.95	
Ba	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	
Cl	0.02	0.02	0.02	0.02	0.02	0.03	0.02	0.03	0.03	0.02	0.02	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	
F	0.20	0.19	0.20	0.19	0.19	0.20	0.18	0.20	0.20	0.19	0.19	0.20	0.20	0.20	0.17	0.23	0.24	0.22	0.22	
OH	1.78	1.79	1.78	1.79	1.79	1.78	1.80	1.77	1.78	1.78	1.78	1.78	1.78	1.78	1.81	1.74	1.74	1.75	1.75	

<sup>1</sup> Total iron measured as FeO; Fe<sub>2</sub>O<sub>3</sub> calculated from wet chemical analyses of mineral separates.

<sup>2</sup> Wt. % H<sub>2</sub>O calculated from the structural formula.

Table 2. Microprobe analyses of chlorite from the Gold Hill pluton, Utah.

	20-1	20-3	20-4	20-6	21-13	21-14	22-1	22-2	24-5	24-6	24-7
SiO <sub>2</sub>	29.6	30.1	28.8	30.3	29.0	29.1	26.6	26.6	26.6	28.3	26.5
TiO <sub>2</sub>	0.22	0.21	0.19	0.25	0.10	0.07	0.05	0.07	0.13	0.10	0.21
Al <sub>2</sub> O <sub>3</sub>	16.3	16.2	17.1	15.9	17.7	18.4	19.5	19.4	18.2	16.8	18.1
FeO <sup>1</sup>	18.5	18.5	18.5	17.8	17.0	16.8	21.6	21.9	23.5	22.0	22.8
Fe <sub>2</sub> O <sub>3</sub> <sup>1</sup>	3.0	3.0	3.0	2.9	2.7	2.7	3.5	3.5	3.8	3.5	3.7
MnO	0.44	0.40	0.42	0.39	0.40	0.40	0.40	0.40	0.25	0.25	0.25
MgO	19.6	19.1	19.0	20.0	20.6	20.1	15.7	15.4	16.4	17.1	16.3
CaO	0.23	0.26	0.26	0.17	0.20	0.20	0.20	0.20	0.20	0.20	0.20
K <sub>2</sub> O	0.28	0.02	0.02	0.08	0.18	0.12	0.12	0.13	0.22	0.68	0.43
Na <sub>2</sub> O	0.03	0.01	0.01	0.05	0.03	0.03	0.03	0.03	0.03	0.03	0.03
BaO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cl	0.02	0.01	0.01	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
F	0.21	0.19	0.19	0.31	0.25	0.25	0.24	0.24	0.16	0.19	0.24
H <sub>2</sub> O <sup>2</sup>	11.7	11.7	11.7	11.7	11.7	11.8	11.4	11.4	11.3	11.4	11.3
F, Cl = O	0.09	0.11	0.08	0.13	0.11	0.11	0.11	0.11	0.07	0.08	0.10
Total	100	99.7	99.1	99.7	99.8	99.9	99.3	99.2	101	100	100
Number of ions based on 10 oxygens and 8 (OH, F, Cl)											
Si	3.02	3.08	2.96	3.08	2.94	2.94	2.79	2.79	2.77	2.93	2.78
Al(IV)	0.98	0.92	1.04	0.92	1.06	1.06	1.21	1.21	1.23	1.07	1.22
Al(VI)	0.98	1.03	1.04	0.99	1.05	1.13	1.20	1.19	1.01	0.98	1.01
Ti	0.02	0.02	0.02	0.02	0.01	0.01	0.00	0.01	0.01	0.01	0.02
Fe <sup>2+</sup>	1.58	1.58	1.59	1.52	1.44	1.42	1.89	1.92	2.05	1.91	2.00
Fe <sup>3+</sup>	0.23	0.23	0.23	0.22	0.21	0.21	0.27	0.28	0.30	0.28	0.29
Mn	0.04	0.04	0.04	0.03	0.03	0.03	0.04	0.04	0.02	0.02	0.02
Mg	2.99	2.90	2.92	3.03	3.11	3.03	2.45	2.41	2.55	2.64	2.54
Ca	0.03	0.03	0.03	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
Na	0.01	0.00	0.00	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
K	0.04	0.00	0.00	0.01	0.02	0.02	0.02	0.02	0.03	0.09	0.06
Ba	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cl	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
F	0.07	0.08	0.06	0.10	0.08	0.08	0.08	0.08	0.05	0.06	0.08
OH	7.93	7.92	7.94	7.90	7.92	7.92	7.92	7.92	7.95	7.94	7.92

<sup>1</sup> Total iron measured as FeO; Fe<sub>2</sub>O<sub>3</sub> calculated from wet chemical analyses of mineral separates.

<sup>2</sup> Wt. % H<sub>2</sub>O calculated from the structural formula.

54%) of host biotite grains. The Mg, Fe, and Al content of the chlorites varies systematically with volume-percent replacement of the biotites as shown by the dotted arrows on Figure 1.

The petrographic evidence for the conservation of volume in the replacement of biotite by chlorite is shown in Figure 2. Chlorite replaces biotite along cleavage lamellae (Figure 2A) with no distortion of the lamellae or increased porosity. No petrographic evidence was noted to indicate that the apatite and zircon inclusions participate in the alteration reaction. Biotite in Figure 2B, however, has been replaced by chlorite and sphene with no apparent change in the total volume of solids.

### FLUID INCLUSIONS

The temperature limits for the biotite to chlorite reaction were estimated from the homogenization temperatures of fluid inclusions. Igneous quartz grains in the rock commonly contain abundant trains of secondary fluid inclusions along healed fractures. Altered bio-

tite is commonly in contact with one or more of these quartz grains, and, in places, trains of fluid inclusions lead to the chloritized portions of the biotite. The close spatial association of the fluid inclusions and the altered biotite suggests that the fluids in the inclusions are responsible for the biotite alteration and that the filling temperature of the fluid inclusions is the temperature of the biotite alteration. The fluid inclusions are less than 15 μm in size, irregular, and contain a vapor bubble which is less than 5% of the volume of the inclusion and no daughter minerals. Homogenization temperatures measured with a Roman Science Model 3 heating stage ranged from 171°C to 251°C with a mean value of 216°C. Geologic reconstruction of the stratigraphic section suggests a thickness of cover of approximately 2 km. The estimated maximum pressures are 200 bars for hydrostatic conditions and 500 bars for lithostatic conditions. The maximum pressure correction of the filling temperatures would be +20° to +40°C (Potter, 1977). No pressure correction was applied to the filling temperatures measured in this study.

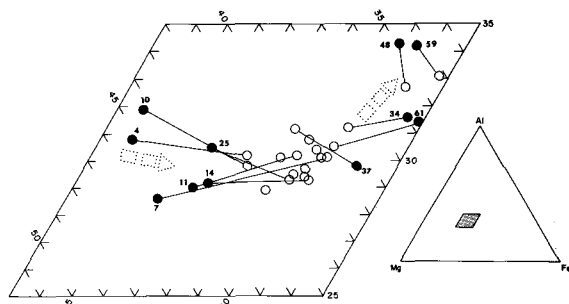
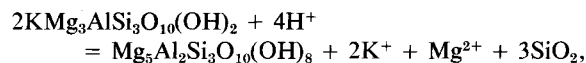


Figure 1. Compositions of coexisting biotite and chlorite from the Gold Hill pluton, Utah, in terms of atomic proportions of Fe, Mg, and Al. Tie lines connect coexisting biotite (open circles) and chlorite (solid circles). Dotted arrows show the trend of increasing volume percent biotite replaced by chlorite, and bold numbers are the volume percent biotite replaced by chlorite.

### CHEMICAL MASS TRANSFER

Igneous biotite in the Gold Hill rocks reacted irreversibly with a hydrothermal solution, and chlorite was precipitated from the solution replacing the biotite. A first approximation of irreversible mass transfer can be made from a balanced chemical reaction for the conversion of biotite to chlorite. Conserving one or more components among the solid phases in a reaction is a convenient method of deducing reaction coefficients and approximating chemical mass transfer (Helgeson, 1968). The calculated activities of Al species in many hydrothermal systems are very low (Helgeson, 1970), and large relative changes in their activities have a negligible effect on the mass transfer among solids in the system (Helgeson *et al.*, 1969). Al may be considered as conserved among the solid phases biotite and chlorite. Quartz is also present in the rocks so that conserving silica among solid phases in the reaction will cause negligible errors in estimating the irreversible mass transfer (Helgeson, 1968). The chemical reaction for the hydrothermal replacement of biotite by chlorite in which Al and silica are conserved among the solid phases is:



where the magnesium end members phlogopite and clinocllore represent biotite and chlorite. The reaction as written consumes  $\text{H}^+$  from solution and produces  $\text{K}^+$  and  $\text{Mg}^{2+}$  as phlogopite is consumed and clinocllore + quartz are produced.

An alternate means of deducing reaction coefficients for the hydrothermal replacement of biotite by chlorite is suggested by the petrographic observation shown in Figure 2A, i.e., biotite is replaced by an equivalent volume of chlorite. The molar volumes of phlogopite and clinocllore are  $149.66 \text{ cm}^3/\text{mole}$  and  $207.11 \text{ cm}^3/\text{mole}$ ,

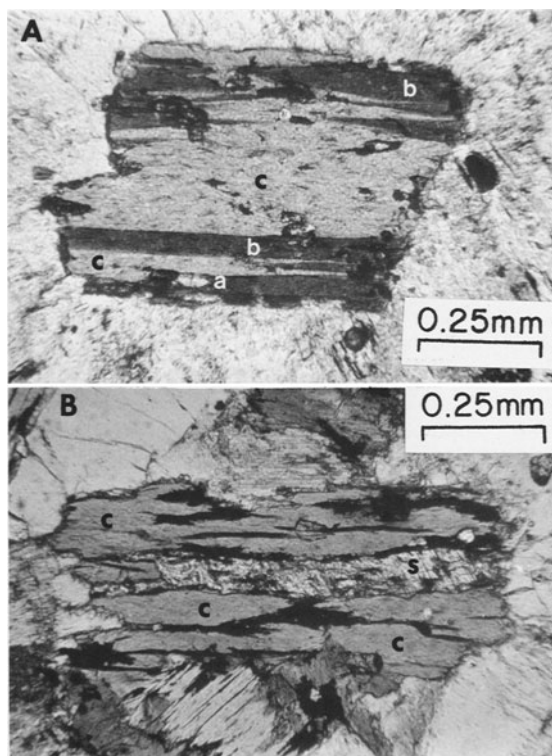
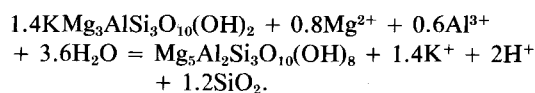


Figure 2. Photomicrographs of chlorite replacements of biotite in plane light. (A) Chlorite (c) and relict biotite (b) with accessory apatite (a). (B) Chlorite (c) and sphene (s).

respectively (Helgeson *et al.*, 1978). The chemical reaction in which phlogopite is replaced by an equal volume of clinocllore is (rounding reaction coefficients to two significant figures):



The reaction coefficients for phlogopite and clinocllore are fixed by volume constraints, the reaction coefficients for  $\text{Mg}^{2+}$ ,  $\text{Al}^{3+}$ , and  $\text{K}^+$  are fixed by mass balance, the coefficient for  $\text{H}^+$  is fixed by charge balance, and the coefficients for  $\text{H}_2\text{O}$  and  $\text{SiO}_2$  are fixed by mass balance. The reaction as written produces  $\text{H}^+$  and  $\text{K}^+$  and consumes  $\text{Mg}^{2+}$  and  $\text{Al}^{3+}$  as phlogopite is replaced by an equal volume of clinocllore. The Al-conservative reaction consumes  $\text{H}^+$  with a volume decrease of  $92.21 \text{ cm}^3/\text{mole}$  of chlorite or 31% of the original phlogopite volume. The volume change can be compensated for by increased porosity, by precipitation of other product minerals, or by changes in rock volume by adjustment on fractures.

Equilibrium stability relations in the system  $\text{K}_2\text{O}-\text{MgO}-\text{Al}_2\text{O}_3-\text{H}_2\text{O}$  at 100 bars and  $200^\circ\text{C}$  are shown in

Figure 3. Volume- and Al-conservative reactions were used to construct Figure 3A and Figure 3B, respectively. Equilibrium constants were computed using thermodynamic data from the computer program SUPCRT (Helgeson *et al.*, 1978). The volume-conservative equilibria shown in Figure 3A are critically dependent on the activity ratio  $\text{Al}^{3+}/(\text{H}^+)^3$  in solution. The value used for  $\log \text{Al}^{3+}/(\text{H}^+)^3 = 1.6$  was chosen to show the stability relations for constant-volume reactions with values of the solution parameters  $\log a_{\text{K}^+}/a_{\text{H}^+}$  and  $\log a_{\text{Mg}^{2+}}/(a_{\text{H}^+})^2$  similar to those for the Al-conservative reactions.

Systematic variations in the chlorite composition as more biotite is replaced suggest that the chlorite changes composition in response to changes in the solution composition as the chemical reaction between the aqueous phase and the biotite proceeds. The solution compositions which result from the reaction of phlogopite with an initial solution to produce clinocllore were estimated and plotted in Figure 3A and 3B as solution reaction paths. An arbitrary composition of the initial solution was chosen in the stability field of chlorite. New solution compositions which would result from the dissolution of successive small amounts of phlogopite, and the precipitation of small amounts of clinocllore were estimated using the calculation procedures of Helgeson *et al.* (1970). Solution reaction paths resulting from volume- and Al-conservative reactions are shown as large arrows in Figure 3A and 3B, respectively. All of the solution species were not considered, therefore the reaction paths shown in Figure 3 are approximations only. The major difference between the reaction paths shown in Figure 3A and 3B is in the fate of the hydrogen ion: the volume-conservative reaction (Figure 3A) produces  $\text{H}^+$ , whereas the Al-conservative reaction (Figure 3B) consumes  $\text{H}^+$ . The reaction path shown in Figure 3A yields clinocllore + muscovite or clinocllore + muscovite + phlogopite, assemblages that are common in hydrothermal systems (Schwarz, 1958), however, muscovite is not a common alteration product of biotite in the Gold Hill, Utah, samples. The Al-conservative reaction path shown in Figure 3B yields clinocllore + phlogopite as solutions saturated with clinocllore continue to react with phlogopite.

Phlogopite and clinocllore are not involved in the hydrothermal reactions at Gold Hill. The end-member compositions have been diluted with other components, dominantly Fe, which lower the activities of phlogopite in the biotite and clinocllore in the chlorite. Solutions in equilibrium with the most- and least-magnesian chlorite at Gold Hill are shown on Figure 3B as contours of the ratio of the mole fraction of clinocllore  $[\text{X Mg}_5\text{Al}_2\text{Si}_3\text{O}_{10}(\text{OH})_2]$  to mole fraction of phlogopite squared  $[\text{X KMg}_3\text{AlSi}_3\text{O}_{10}(\text{OH})_2]^2$ . An ionic-site mixing model was used to calculate the mole fraction of each end member (Powell, 1978), and the mole fraction was assumed to be equal to the activity.

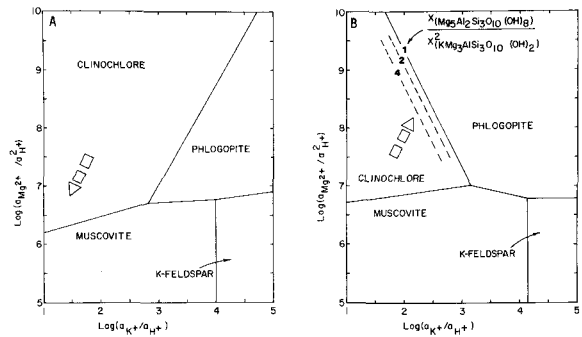


Figure 3. Calculated stability relations of chlorite, phlogopite, muscovite, and K-feldspar at 200°C and 100 bars for volume-conservative reactions (A) and Al-conservative reactions (B). Dissolved silica is fixed at quartz saturation and activity of water = 1. Equilibrium constants are calculated with computer program SUPCRT (Helgeson *et al.*, 1978). Arrows indicate hypothetical reaction paths for the alteration of biotite to chlorite. Mineral composition contours are constructed using an ionic site-mixing model.

The mole fraction of clinocllore in the chlorite varies from 0.09 for sample 21-13 (the most magnesian chlorite) to 0.03 for samples 22 and 24 (the least magnesian chlorites). The mole fraction of phlogopite varies from 0.11 to 0.13 and averages 0.12.  $X$  clinocllore/ $X^2$  phlogopite varies from 5.3 for the highest magnesian chlorite to 2.6 for the low-magnesium chlorites of sample 24. The higher magnesian chlorites form from solutions with lower  $\log a_{\text{K}^+}/a_{\text{H}^+}$  values for a given  $\log a_{\text{Mg}^{2+}}/(a_{\text{H}^+})^2$  than the lower magnesian chlorites. The reaction path shown in Figure 3B would thus cross the contour for  $X$  clinocllore/ $X^2$  phlogopite = 4 followed by intersection of the reaction path with the contour for  $X$  clinocllore/ $X^2$  phlogopite = 2 as  $\text{H}^+$  is consumed. The chlorite compositions thus record a portion of the solution reaction path. The clinocllore component of the chlorite decreases as the composition of the solution approaches the stability field of phlogopite. This hypothetical reaction path approximates the systematic variation in chlorite composition observed in the Gold Hill samples as successively more biotite is altered to chlorite. The trend in the chlorite compositions suggested by the reaction path of Figure 3B is also shown as a trend in the chlorite compositions in Figure 1 and is the basis for the conclusion that the alteration of biotite to chlorite is Al-conservative rather than volume-conservative, as suggested by petrographic observations. Volume changes accompanying the alteration must therefore be accommodated by readjustment of the surrounding mineral grains along fractures and by volume changes produced by the introduced minerals such as the sphene shown in Figure 2B.

The chemical mass transfer due to the replacement of biotite by chlorite may be estimated from the data of

Table 3. Representative mass balance for Al- and volume-conservative biotite to chlorite reactions for sample 20-1 (Tables 1 and 2).<sup>1</sup>

	Al-conservation reaction (1.72 biotite + ions = chlorite + ions)				Volume-conservation reaction (1.41 biotite + ions = chlorite + ions)					
	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)
K		0.95		0.04	1.59		0.95		0.04	1.30
Na		0.02		0.01	0.02		0.02		0.01	0.02
Ca		0.02		0.03			0.02		0.03	
Ba		0.01		0.00	0.02		0.01		0.00	0.01
Mg		0.52	0.38	2.99			1.52	0.84	2.99	
Mn		0.02	0.01	0.04			0.02	0.01	0.04	
Fe <sup>2+</sup>		1.08		1.58	0.28		1.08	0.06	1.58	
Fe <sup>3+</sup>	1.72	×	0.03	+	0.18	=	0.23	+	1.41	×
Ti		0.24		0.02	0.39		0.24		0.02	0.32
Si		2.83		3.02	1.85		2.93		3.02	0.92
Al		1.14		1.96			1.14	0.35	1.96	
F		0.20		0.07	0.27		0.20		0.07	0.21
Cl		0.02		0.00	0.03		0.02		0.00	0.03
H			2.15							1.06
H <sub>2</sub> O			1.36					3.24		

## Mass transfer per cubic meter of rock containing 5% biotite completely altered to chlorite

Moles added		Moles lost		Moles added		Moles lost	
Mg	74	K	311	Mg	200	K	309
Fe <sup>3+</sup>	35	Fe <sup>2+</sup>	54	Fe <sup>3+</sup>	45	Ti	76
H <sup>+</sup>	420	Ti	76	Fe <sup>2+</sup>	14	F	50
Mn	2	F	53	Al	83	Cl	7
Chlorite	195	Cl	6	Chlorite	238	H	252
		Biotite	336			Biotite	336

<sup>1</sup> See text for explanation of calculations.

Tables 1 and 2. Mass transfer for the replacement of biotite 20-1 by chlorite 20-1 is shown in Table 3 assuming Al is conserved between biotite and chlorite. For comparison purposes, the mass transfer for the reaction conserving volume is also presented in Table 3.

The estimation of the chemical mass transfer and volume changes in the replacement of biotite by chlorite requires an estimate of the molar volumes of biotite and chlorite which are a function of composition. Hewitt and Wones (1975) found the Fe<sup>2+</sup>-Mg<sup>2+</sup> substitution to have the greatest influence on the cell size of biotite. Variations due to a limited amount of octahedral Al<sup>3+</sup> proxying for Fe and Mg were found to be only 25% as large. The effects of Fe<sup>3+</sup> and Ti<sup>4+</sup> content and F<sup>-</sup> and Cl<sup>-</sup> substitution for OH<sup>-</sup> are significant but data are not available to evaluate their influence. Because the synthetic biotite study of Hewitt and Wones (1975) did not involve Fe<sup>3+</sup>, Ti<sup>4+</sup>, F<sup>-</sup>, or Cl<sup>-</sup>, a linear regression of natural biotite data taken from the literature was chosen to predict the molar volumes:

$$V = 150.6 - 3.214[\text{Mg}/(\text{Mg} + \text{Fe total} + \text{Ti} + \text{Mn})]$$

where V = molar volume in cm<sup>3</sup>. The octahedral Al was subtracted in order to compare the natural mineral data to that of synthetic Fe<sup>2+</sup>-Mg-Al-biotite (Hewitt and Wones, 1975). Natural biotites have significantly small-

er molar volumes than synthetic biotites, perhaps as a result of the Fe<sup>3+</sup>, F, and Cl contained in the natural minerals.

McOnie *et al.* (1975) included molar volume as a function of composition of synthetic chlorites in their study of the stability of Fe<sup>2+</sup>-Mg<sup>2+</sup>-chlorites. Comparison of their regression curve with molar volumes of natural chlorites indicates that natural chlorites have slightly smaller molar volumes than synthetic chlorites. The natural chlorite data were used to estimate the molar volumes in this study:

$$V = 213.3 - 4.909[\text{Mg}/(\text{Mg} + \text{Fe total} + \text{Ti} + \text{Mn})]$$

The basis for estimating chemical mass transfer is a balanced chemical reaction. Chemical reactions are shown in Table 3 for the replacement of biotite 20-1 by chlorite 20-1 assuming that Al or volume is conserved. The reaction coefficient for the 1.72 biotite (column 1, Table 3) was chosen to provide an equivalent amount of Al in reactant biotite and one mole of product chlorite. Column 2 in Table 3 is the half unit-cell composition of biotite 20-1 from Table 1. Column 3 represents the reaction coefficients for ions which must be added to produce chlorite 20-1; column 4 is the composition of chlorite 20-1 from Table 2, and column 5 represents the reaction coefficients of ions that must be removed

Table 4. Mass transfer per cubic meter of rock containing 5% biotite altered to chlorite for each biotite-chlorite pair from Tables 1 and 2.<sup>1</sup>

Sample number	Chlorite volume % replacement	Moles of constituents gained or lost								
		K	Mg	Fe <sup>2+</sup>	Fe <sup>3+</sup>	Ti	F	Cl	H <sup>+</sup>	V (cm <sup>3</sup> )
20-1	11	34	-8	6	-4	8	6	1	-46	99
20-3	14	42	-6	10	-5	9	7	1	-68	1500
20-4	25	78	0.4	21	-7	18	14	2	-24	3200
20-6	7	21	-7	5	-2	5	3	0.4	-27	610
21-13	4	11	-1	4	-1	2	2	0.3	-20	430
21-14	10	27	-1	10	-3	6	5	0.7	-56	1300
22-1	48	151	-12	32	-17	27	26	5	-222	6500
22-2	59	185	-6	40	-21	29	31	6	-275	7600
24-5	61	188	32	13	-26	46	44	6	-347	7800
24-6	37	113	-3	-5	-17	21	23	4	-104	3000
24-7	34	105	11	8	-14	23	20	4	-176	3900

<sup>1</sup> Assume Al is conserved. Moles lost to solution are indicated by positive values; those added to the rock as negative values.

in solution. Volume is conserved in the replacement of biotite 20-1 by chlorite 20-1 if the reaction coefficient for biotite is 1.41, shown in column 6, Table 3. Column 7 is the biotite composition from Table 1, column 8 shows the reaction coefficients for ions which must be added in the volume-conservative reaction, column 9 is the composition of chlorite 20-1 from Table 2, and column 10 shows reaction coefficients for ions that must be removed in solution.

Quartz monzonite containing 5 volume% biotite contains 336 moles of biotite per m<sup>3</sup> of rock. Chemical gains and losses if all of this biotite were converted to chlorite conserving aluminum or volume are shown in Table 3. The number of moles of Mg which must be added to convert all of the biotite in one m<sup>3</sup> of rock to chlorite is calculated as follows: 0.38 moles of Mg must be added (Table 3, column 3) to 1.72 moles of biotite to produce one mole of chlorite; therefore 74 moles of Mg must be added to convert 336 moles of biotite to chlorite (336/1.72 × 0.38). Similar calculations are repeated for each constituent gained or lost in an Al-conservative or a volume-conservative reaction.

Mass transfer per m<sup>3</sup> of rock containing 5% biotite that is partly altered to chlorite is presented in Table 4 for Al conservation in each of the biotite-chlorite pairs in Tables 1 and 2. Calculations for Table 4 are similar to those for Table 3 except that the observed percentage of chlorite replacement of biotite was used instead of estimating mass transfer if 100% of the biotite were replaced as in Table 3. The mass transfer of Mg varies from 12 moles added per m<sup>3</sup> of rock to 32 moles removed in solution depending on biotite and chlorite compositions and extent of replacement. Masses of other constituents removed per m<sup>3</sup> of rock during the alteration of biotite are 11 to 188 moles of K, 2 to 46 moles of Ti, 2 to 44 moles of F, and 0.3 to 6 moles of Cl. Masses of the constituents added to the rock include 20 to 347 moles of H<sup>+</sup> and 1 to 26 moles of Fe<sup>3+</sup>. The

volume decrease as a result of the alteration of biotite is 430 cm<sup>3</sup>/m<sup>3</sup> of rock for the least altered sample to 7800 cm<sup>3</sup>/m<sup>3</sup> for the most altered. The alteration of biotite is a likely source of some K in the adularia + quartz + calcite veins, and nearby sphene, rutile, and anatase account for the Ti released. Cl and F may have remained in solution.

## CONCLUSIONS

Chlorite replacing biotite in the Gold Hill, Utah, pluton is of hydrothermal origin. Secondary fluid inclusions in nearby quartz suggest that the alteration of biotite took place at near 200°C. The Mg content of chlorite decreases systematically as more biotite is replaced. A comparison of the chlorite compositions with hypothetical reaction paths on solution species-activity diagrams in which either Al or volume are conserved suggests that the alteration of biotite to chlorite conserves Al and consumes H<sup>+</sup>. The decrease in volume of 430 to 7800 cm<sup>3</sup>/m<sup>3</sup> of rock must be accommodated by adjustment on fractures without producing petrographically discernable distortion of the chlorite. F, Cl, Fe<sup>2+</sup>, and K are lost to solution, and Mg, Fe<sup>3+</sup>, and Mn are added from solution to form chlorite.

## ACKNOWLEDGMENTS

J. D. Hazelton and W. P. Nash guided the use of the electron microprobe. G. H. Ballantyne developed the Bence-Albee correction program for hydrous minerals, J. R. Hindman assisted with the X-ray diffraction studies, and T. Butkus assisted with the sample collecting. G. H. Ballantyne, L. G. Schultz, D. D. Eberl, and T. Cerling provided thoughtful reviews.

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(Received 23 February 1981; accepted 6 May 1981)

**Резюме**—Гидротермальный хлорит заменяет вулканический биотит в кварцевом монзоните из Золотой Горы, Юта. Химический состав сосуществующих биотита и хлорита, определенный при помощи электронной микросонды и влажных химических методов, использовался для определения переноса химической массы во время процесса перемены. Молярное соотношение Mg/(Mg + Fe) изменяется от 0,52 до 0,65 в хлорите и от 0,51 до 0,60 в материнском биотите. Содержание Mg в хлорите систематически уменьшалось при увеличении объемного процента замененного биотита. Температуры гомогенизации жидких инклюзий в близких кварцевых микровенах указывают на то, что хлоритовые перемены происходили при температуре приблизительно 200°C.

Текстурные соотношения показывают, что процесс перемены биотита в хлорит изохорический, но сравнение минерального состава и минеральных скоплений с фазовыми диаграммами, в которых Al или объем сохраняются в твердых фазах, указывает на то, что состав хлорита лучше объясняется как функция прогресса реакции в Al-сохраняющейся системе. Состав хлорита изменяется в соответствии с изменениями состава раствора, получаемого растворением последующих малых количеств биотита. Характерное равновесие массы для перемены всего биотита в хлорит в 1 м<sup>3</sup> породы, содержащем 336 молей биотита, указывает на то, что 74 моля Mg, 35 молей Fe<sup>3+</sup>, 420 молей H<sup>+</sup>, и 2 моля Mn добавляются к породе, и что 311 молей K, 54 моля Fe<sup>2+</sup>, 76 молей Ti, 53 моля F, и 6 молей Cl переходят в раствор. Перенос массы для частично измененного биотита равен 11 до 188 молям K, 2 до 46 молям Ti, 2 до 44 молям F, и 0,3 до 6 молям Cl, удаленным из одного кубического метра породы, и 1 до 26 молям Fe<sup>3+</sup> и 20 до 347 молям H<sup>+</sup> добавленным к нему. Перенос массы Mg изменяется от 12 молей добавленных до 32 молей удаленных из кубического метра породы в зависимости от минерального состава и степени замены. [E.C.]

**Resümee**—Hydrothormaler Chlorit ersetzt Biotit im Quarz-Monzonit von Gold Hill, Utah. Die chemische Zusammensetzung von koexistierendem Biotit und Chlorit, die mittels Mikrosonde und naßchemischen Methoden untersucht wurde, wurde verwendet, um den chemischen Stofftransport während des Umwandlungsprozesses zu berechnen. Das Mol-Verhältnis  $Mg/(Mg + Fe)$  variiert von 0,52 bis 0,65 im Chlorit und von 0,51 bis 0,60 im Ausgangsbiotit. Der Mg-Gehalt des Chlorit nimmt systematisch mit zunehmendem volumenprozentuellen Ersatz des Biotit ab. Die Homogenisierungstemperaturen der fluiden Einschlüsse von benachbarten Quarz-Mikroadern deuten darauf hin, daß die chloritische Umwandlung bei etwa 200°C stattgefunden hat.

Texturelle Beziehungen lassen darauf schließen, daß die Umwandlung Biotit in Chlorit isovolumetrisch ist. Ein Vergleich der Mineralzusammensetzungen und der Mineralvergesellschaftungen mit Phasendiagrammen, in denen das Al oder das Volumen zwischen festen Phasen konstant ist, deutet jedoch darauf hin, daß die Chloritzusammensetzungen am besten als eine Funktion des Reaktionsfortschrittes in einem System mit konstantem Al erklärt werden können. Die Chloritzusammensetzung variiert je nach den Veränderungen der Lösungszusammensetzung, die durch die allmähliche Auflösung von kleinen Biotitanteilen erzeugt wird. Eine repräsentative Massenbilanz für die Umwandlung des gesamten Biotit, der in 1 m<sup>3</sup> Gestein enthalten ist und 336 Mol Biotit enthält, in Chlorit deutet darauf hin, daß 74 Mol Mg, 35 Mol Fe<sup>3+</sup>, 420 Mol H<sup>+</sup>, und 2 Mol Mn dem Gestein zugeführt werden, während 311 Mol K, 54 Mol Fe<sup>2+</sup>, 76 Mol Ti, 53 Mol F, und 6 Mol Cl in Lösung gehen. Der Massentransport für teilweise umgewandelten Biotit bedeutet eine Abfuhr von 11 bis 188 Mol K, 2 bis 46 Mol Ti, 2 bis 44 Mol F, und 0,3 bis 6 Mol Cl und eine Zufuhr von 1 bis 26 Mol Fe<sup>3+</sup> und 20 bis 347 Mol H<sup>+</sup> pro m<sup>3</sup>. Der Massentransport von Mg variiert von 12 Mol Zufuhr bis 32 Mol Abfuhr pro m<sup>3</sup> Gestein je nach Mineralzusammensetzung und Ausmaß der Umwandlung. [U.W.]

**Résumé**—La chlorite hydrothermale remplace la biotite ignée dans la monzonite de quartz du Gold Hill, Utah. Les compositions chimiques de biotite et de chlorite coexistantes, déterminées par microprobe à électrons et par des méthodes chimiques mouillées, ont été utilisées pour évaluer le transfert en masse chimique pendant le procédé d'altération. La proportion de mole  $Mg/(Mg + Fe)$  varie de 0,52 à 0,65 dans la chlorite, et de 0,51 à 0,60 dans la biotite mère. Le contenu en Mg de la chlorite décroît systématiquement en proportion avec un accroissement du pourcentage de volume de remplacement de la biotite. Les températures d'homogénéisation d'inclusions fluides dans des micro-veines de quartz proches indiquent que l'altération chloritique s'est passée à approximativement 200°C.

Les relations texturales suggèrent que l'altération de biotite en chlorite est isovolumétrique, mais une comparaison de compositions minérales et d'assemblages minéraux avec des diagrammes de phase dans lesquels Al ou le volume sont conservés parmi des phases solides suggère que les compositions de chlorite sont expliquées le mieux en fonction de progrès de réaction dans un système Al-conservatif. La composition de la chlorite change en réponse à des changements de composition de solution produits par la dissolution de petites quantités successives de biotite. L'équilibre de masse représentatif de l'altération de toute la biotite en chlorite dans 1 m<sup>3</sup> de roche contenant 336 moles de biotite indique que 74 moles de Mg, 35 moles de Fe<sup>3+</sup>, 420 moles d'H<sup>+</sup>, et 2 moles de Mn sont ajoutées à la roche, et que 311 moles de K, 54 moles de Fe<sup>2+</sup>, 76 moles de Ti, 53 moles de F et 6 moles de Cl sont perdues en solution. Le transfert en masse pour la biotite partiellement altérée est tel que 11 à 188 moles de K, 2 à 46 moles de Ti, 2 à 44 moles de F et 0,3 à 6 moles de Cl sont retirées par un mètre cube de roche, et 1 à 26 moles de Fe<sup>3+</sup> et 20 à 347 moles d'H<sup>+</sup> y sont ajoutées. Le transfert en masse de Mg varie de 12 moles ajoutées à 32 moles retirées par mètre cube de roche, dépendant de la composition minérale et de la quantité de remplacement. [D.J.]