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An occurrence of Ba-rich micas from the Alaska Range

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ABSTRACT

Two compositionally different groups of Ba-rich micas are present within one rock sample from the contact aureole of the Middle Fork plutonic complex, northwest Alaska Range. The more Ba-rich group has pale green to colorless pleochroism, up to 18 wt% BaO, and the average formula $(Ba_{0.55}K_{0.40}Na_{0.03})(Mg_{2.57}Al_{0.16}Fe^{2+}_{0.23}Fe^{3+}_{0.04}Ti_{0.01})(Si_{2.24}Al_{1.76})O_{10.00}[(OH)_{1.30}F_{0.70}]$. The other is pale brown to colorless, contains up to 13 wt% BaO, and has an average formula of $(Ba_{0.40}K_{0.54}Na_{0.03})(Mg_{2.57}Al_{0.07}Fe^{2+}_{0.24}Ti_{0.10})(Si_{2.40}Al_{1.60})O_{10.00}[(OH)_{1.40}F_{0.60}]$. Optically the two are similar, with $0° < 2V_X < 5°$ and birefringences of 0.018 (green mica) and 0.022 (brown mica). Compositionally, they most closely approximate the brittle mica kinoshitalite, $(Ba_{0.58}K_{0.35}Na_{0.11}Ca_{0.01})(Mg_{2.06}Al_{0.22}Mn^{2+}_{0.52}Mn^{3+}_{0.21}Fe^{3+}_{0.05}Ti_{0.01})(Si_{2.05}Al_{1.95})O_{10.00}[(OH)_{1.61}F_{0.06}O_{0.33}]$. However, in comparison with reported kinoshitalites, the Middle Fork specimens are not manganiferous, are considerably richer in Mg and Fe, and have different optical properties.

GEOLOGIC SETTING AND PETROGRAPHY

The barian mica–bearing sample is from the southwest contact zone of the early Tertiary Middle Fork plutonic complex of the northwest Alaska Range (Solie, 1983), where hedenbergite + fayalite ± quartz syenite intrudes thinly bedded Paleozoic calc-phyllites and marbles. Thin syenitic dikes and sills are abundant within the contact zone. The profoundly recrystallized contact rocks from which the micaceous sample was taken retain crude sedimentary layering. The presence of wollastonite in samples from within 10 m of the mica-bearing specimen is indicative of high-temperature ($\geq 600°C$) contact metamorphism (Harker and Tuttle, 1956; Greenwood, 1967). The pressure of contact metamorphism is probably about 1–2 kbar, based on the estimated thickness of originally overlying strata (Bundtzen and Gilbert, 1983).

The green and white calc-silicate sample contains the barian micas, diopside, calcite (<0.2 wt% MgO), green spinel (MgO > FeO), hematite, and alkali feldspar. Grain size of all the phases is fine to medium with some mica grains as large as 1 mm in diameter. Spinel occurs as inclusions in barian micas and diopside, diopside and calcite occur as symplectic intergrowths in some layers, and a semi-opaque alteration product is prevalent in areas of the rock. Texturally, both green and brown barian micas appear to be in equilibrium. Green mica forms up to about ten modal percent of the sample, and brown mica, up to about two modal percent. Brown mica occurs as cores within some green mica grains, as rims around other green micas, and as monochromatic brown grains. In some cases, one side of a crystal is brown and the other side is green, with no apparent rimming relationship.

ANALYTICAL PROCEDURES

Electron-microprobe analyses were performed on (001) cleavage surfaces of mineral separates using the ARL-SEMQ nine-channel microprobe at Virginia Polytechnic Institute and State University. The data were reduced following the method of Bence and Albee (1968). Mineral separations were done using methylene iodide, were further cleaned by use of a magnetic separator, and then were hand-picked for purity. Spinel inclusions in micas were released by grinding and were subsequently magnetically removed. Fe^{2+} determinations were performed using Whipple's (1974) titration method, on 0.125 g of green mica separate and 0.019 g of brown mica separate. Precision of measurements is estimated to be better than ±0.1 wt% FeO, based on replicate analyses of a whole-rock sample and blanks. Ba concentrations in the whole rock were analyzed using X-ray fluorescence spectrometry on pelletized rock powders.

COMPOSITION AND OPTICAL PROPERTIES

Green mica

The pale green mica has an average compositional formula of $(Ba_{0.55}K_{0.40}Na_{0.03})(Mg_{2.57}Al_{0.16}Fe_{0.27}Ti_{0.01})(Si_{2.24}Al_{1.76})O_{10.00}[(OH)_{1.30}F_{0.70}]$, based on nine electron-microprobe analyses of individual grains (Table 1). Included in the microprobe analytical scheme were Cr, Zn, Ni, P, Sr, Rb, Mn, and Cl. All were close to or below analytical detection limits. A Fe^{2+} determination on the bulk mica separate yielded 3.46 wt% FeO; that is, 86% of the average total Fe by weight is Fe^{2+}.

The pleochroism of the green mica is weak, with a formula of X = colorless, $Y = Z$ = pale green. The optic sign is (−), and $2V_X$ is between 0° and 5°. Refractive indices are $\alpha = 1.598(2)$ and $\beta \cong \gamma = 1.615(2)$. Crystals commonly have pseudohexagonal form and a perfect (001) cleavage.

Brown mica

The brown mica has a lower Ba concentration than the green variety, with the average compositional formula of $(Ba_{0.40}K_{0.54}Na_{0.03})(Mg_{2.57}Al_{0.07}Fe_{0.24}Ti_{0.10})(Si_{2.40}Al_{1.60})O_{10.00}[(OH)_{1.40}F_{0.60}]$, based on nine electron-microprobe analyses (Table 2). Titration of the bulk mineral separate yield-

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TABLE 1. Representative green Ba-rich mica compositions

	1	2	3	4	5	6	7	8	9
SiO_2	27.66	27.74	27.88	28.00	28.27	28.12	28.09	27.90	28.15
TiO_2	0.20	0.20	0.18	0.13	0.15	0.14	0.17	0.19	0.21
Al_2O_3	20.13	20.71	20.22	20.41	20.12	19.95	20.17	20.14	20.19
FeO*	4.29	4.13	3.81	4.20	4.03	3.63	4.40	4.23	3.50
MnO	0.15	0.17	0.17	0.16	0.15	0.13	0.18	0.18	0.12
MgO	21.22	21.78	21.45	21.29	21.73	21.81	21.20	21.30	22.02
CaO	0.09	0.08	0.08	0.08	0.08	0.09	0.08	0.09	0.07
BaO	17.59	17.24	17.40	17.01	17.73	18.24	17.32	16.99	18.06
Na_2O	0.13	0.11	0.15	0.16	0.20	0.14	0.17	0.18	0.12
K_2O	3.95	3.96	4.03	3.88	3.95	3.92	3.97	4.03	3.80
F	2.62	2.82	2.69	2.76	3.01	3.11	2.66	2.37	2.82
H_2O	2.50	2.46	2.49	2.47	2.38	2.32	2.51	2.63	2.46
Sum	99.43	100.21	99.42	99.39	100.53	100.29	99.80	99.23	100.33
Si	4.465	4.421	4.483	4.490	4.503	4.502	4.504	4.489	4.488
^{IV}Al	3.535	3.579	3.517	3.510	3.497	3.498	3.496	3.511	3.512
^{VI}Al	0.294	0.310	0.314	0.348	0.279	0.265	0.315	0.308	0.282
Ti	0.024	0.024	0.022	0.016	0.018	0.017	0.020	0.023	0.025
Fe	0.579	0.550	0.512	0.563	0.537	0.486	0.590	0.569	0.467
Mn	0.021	0.023	0.023	0.022	0.020	0.018	0.024	0.025	0.016
Mg	5.106	5.173	5.141	5.089	5.159	5.204	5.067	5.108	5.233
Ca	0.016	0.014	0.014	0.014	0.014	0.015	0.014	0.016	0.012
Na	0.041	0.034	0.047	0.050	0.062	0.043	0.053	0.056	0.037
K	0.813	0.805	0.826	0.794	0.803	0.800	0.812	0.827	0.773
Ba	1.113	1.077	1.096	1.069	1.107	1.144	1.088	1.071	1.128
F	1.338	1.421	1.368	1.400	1.516	1.575	1.349	1.206	1.422
H	2.692	2.615	2.671	2.642	2.529	2.478	2.685	2.823	2.617

Note: Number of cations on the basis of 24 anions. H_2O was not analyzed but is added into total oxides based on the calculated mineral formula.
* Total Fe reported as FeO.

ed a Fe^{2+} content of 3.76 wt%. This is greater than the average total Fe from the nine microprobe analyses, though some individual grains contain >3.76 wt% total Fe. Thus, within the error of the Fe determination, essentially all of the Fe is Fe^{2+}. Trace elements included in the microprobe analyses resulted in near or below analytical detection limits for Cr, Zn, Ni, P, Sr, Rb, Mn, and Cl. The pleochroic scheme of the brown micas is X = colorless, $Y = Z$ = pale brown. The $2V_X$ ranges from 0° to 5°, the optic sign is (−), and the refractive indices are $\alpha = 1.592(2)$,and $\beta \cong \gamma = 1.614(2)$.

DISCUSSION

The Ba-rich micas of this occurrence are compositionally and optically different from Ba-rich micas reported in nephelinites from Hawaii (Mansker et al., 1979), in pyroxene leucitites from the Alban Hills, Italy (Thompson, 1977), in Mn-rich contact rocks from the Noda-Tamagawa mine, Iwate Prefecture, Japan (Yoshii et al., 1973b), and from Kyoto Prefecture, Japan (Yoshii et al., 1973a). The latter two occurrences have been described as kinoshitalite. Yoshii and Maeda (1975) concluded from their study of physical and optical properties of the Ba-rich manganoan micas from Japan that the boundary between manganoan phlogopite and kinoshitalite is near the ratio Ba:K = 1:2; that is, kinoshitalite contains Ba ≥ 0.5K. Using this criterion, both the green and brown micas of this study can be considered as kinoshitalite, since Ba:K = 1.37:1 for the green micas and 0.74:1 for the brown. However, when compared to the kinoshitalite from Japan (Yoshii et al., 1973a, 1973b; Yoshii and Maeda, 1975; Kato et al., 1979; Guggenheim and Kato, 1984), the Middle Fork micas are notably Mn deficient and correspondingly higher in both Fe_{total} and MgO. Also, the Middle Fork specimens are richer in F. Thus, the description of kinoshitalite as the barian analogue of manganoan phlogopite (Yoshii et al., 1973b) is unsuitable when applied to the Middle Fork micas. They do, however, approximate the definition of kinoshitalite (Yoshii et al., 1973a) as being the magnesian analogue of anandite $[BaFe_3(Si_3Fe^{3+})O_{10}(OH)S]$ (Guggenheim, 1984).

The differences in optical properties among the various barian micas is apparent in Table 3. The $2V_X$ of the measured Japanese specimens is 23° to 25°. It is 0° to 5° in the Middle Fork samples. This is probably due to substitutions of Mg, Ti, and Fe for Mn in the octahedral sites of the Middle Fork micas. Color differences probably can be attributed to this substitution as well. The Japanese kinoshitalites are light yellowish brown (Yoshii and Maeda, 1975) or colorless (Yoshii et al., 1973a), whereas of the Middle Fork samples the less Ba-rich mica is light brown (not yellowish), and the more Ba-rich mica is a pale light green. These idiochromatic colors, caused by transition-metal constituents, are a result of crystal-field transitions (Nassau, 1978).

The differences between the two Middle Fork micas are the different Ba:K ratios, the higher Ti content of the brown micas, and the greater Fe_{total} and Fe^{3+} contents of the green micas. It is probably the differences in Fe and Ti that lead to the color differences between the two compositions. Single grains in which both compositions (and colors) are present commonly have a sharp color interface.

TABLE 2. Representative brown mica compositions

	1	2	3	4	5	6	7	8	9
SiO_2	31.36	30.96	31.32	30.79	30.90	30.78	31.10	31.36	30.36
TiO_2	1.38	1.36	1.47	1.47	2.00	2.06	1.89	1.99	2.08
Al_2O_3	18.07	18.01	18.40	18.43	18.22	17.87	18.11	18.00	18.14
FeO*	3.12	3.14	3.15	3.96	4.09	3.47	4.16	3.66	3.61
MnO	0.11	0.11	0.13	0.14	0.17	0.10	0.13	0.14	0.18
MgO	22.66	22.61	22.78	21.86	21.90	22.16	21.88	21.91	22.00
CaO	0.06	0.07	0.07	0.06	0.08	0.06	0.07	0.08	0.07
BaO	13.08	12.92	13.13	12.79	13.07	12.91	12.22	12.90	13.38
Na_2O	0.21	0.17	0.21	0.19	0.23	0.23	0.19	0.18	0.12
K_2O	5.48	5.40	5.33	5.39	5.37	5.38	5.53	5.46	5.12
F	2.54	2.38	2.17	2.42	2.49	2.25	2.68	2.31	2.52
H_2O	2.70	2.74	2.89	2.73	2.72	2.80	2.63	2.81	2.67
Sum	99.70	98.87	100.14	99.21	100.19	99.12	99.46	99.83	99.19
Si	4.851	4.829	4.819	4.798	4.782	4.799	4.820	4.849	4.748
^{IV}Al	3.149	3.171	3.181	3.202	3.218	3.201	3.180	3.151	3.252
^{VI}Al	0.146	0.139	0.155	0.182	0.104	0.081	0.128	0.129	0.090
Ti	0.161	0.160	0.170	0.172	0.233	0.242	0.220	0.231	0.245
Fe	0.404	0.410	0.405	0.516	0.529	0.452	0.539	0.473	0.472
Mn	0.014	0.015	0.017	0.018	0.022	0.013	0.017	0.018	0.024
Mg	5.225	5.256	5.224	5.077	5.051	5.149	5.054	5.050	5.128
Ca	0.010	0.012	0.012	0.010	0.013	0.010	0.012	0.013	0.012
Na	0.063	0.051	0.063	0.057	0.069	0.070	0.057	0.054	0.036
K	1.081	1.074	1.046	1.071	1.060	1.070	1.093	1.077	1.021
Ba	0.793	0.790	0.792	0.781	0.792	0.789	0.742	0.782	0.820
F	1.243	1.174	1.056	1.193	1.219	1.109	1.314	1.130	1.246
H	2.786	2.851	2.966	2.838	2.808	2.912	2.719	2.899	2.785

Note: Number of cations on the basis of 24 anions. H_2O was not analyzed but is added into total oxides based on the calculated mineral formula.
* Total Fe reported as FeO.

Electron-microprobe analyses at this intersection yield intermediate compositions, probably as a result of averaging the two compositions under the electron beam, rather than a graded zonation between compositions. Within each composition, Ba distribution is homogeneous. Substitutions between the two groups appear to include the coupled substitution proposed by Mansker et al. (1979) and Yoshii et al. (1973b): $Ba^{2+} + {}^{IV}Al^{3+} = K^+ + Si^{4+}$. The correlation coefficient for this substitution is −0.992, and charge balance is attained by the substitution of Al for Si in the tetrahedral sites. In contrast to the Hawaiian micas, however, Ti^{4+} concentration is higher in the less Ba-rich Middle Fork micas, with a correspondingly lower Al^{3+} and Fe^{3+} content in the octahedral sites.

The lack of gradational composition change between the two Middle Fork micas is suggestive of a possible solvus relationship between them. However, it is not clear from the available data which of the compositional variables would be the controlling factor in such a relationship, if it exists. The different chemistries could simply reflect growth in small-scale local environments of different bulk and/or fluid composition. The correlation of F content with Ba content in the micas (Fig. 1) suggests that fluid composition may have been a controlling factor in the mica compositions.

Contrary to the suggestion that high pressure is requisite for high-Ba content in micas (Shmakin, 1984), the Middle Fork micas probably formed at moderately low pressures (1–2 kbar). However, it does seem probable that high temperature is important in the formation of these Ba-rich micas. Indeed, Hatch et al. (1957) found the F analogue of kinoshitalite, $BaMg_3Al_2Si_2O_{10}F_2$, to have the highest melting temperature (about 1450°C) of any of their synthetically produced fluorine micas.

The Ba in the micas could have had a magmatic source, or it may originally have been present in the metasedimentary country rock. Within the adjacent Middle Fork plutonic complex, syenites, gabbros, and late hornblende-biotite granites have whole-rock Ba contents >1000 ppm. An alkali feldspar from the syenite has 0.33 wt% BaO; alkali feldspars from monzodiorites and a quartz monzonite have BaO up to 0.91 wt%. The adjacent syenite appears to have been a dry magma; there is no evidence from miarolitic cavities or pegmatites of Ba concentration in any late-stage magmatic fluids. If Ba were mobilized from the pluton into the surrounding contact aureole, it had to be dissolved and transported by hydrothermal fluids

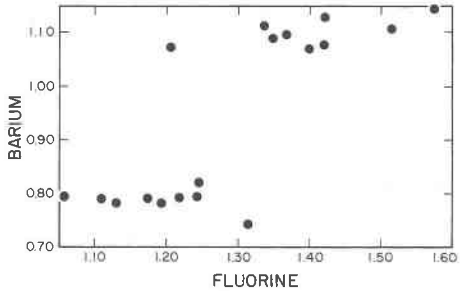

Fig. 1. Atomic contents of Ba plotted against F, based on 24 anions. The more Ba-rich green micas are more F-rich than the brown micas, with little overlap in compositions.

TABLE 3. Optical properties of Middle Fork Ba-rich micas in comparison with reported kinoshitalites from Japan

	Green Ba-rich mica*	Brown Ba-rich mica*	Kinoshitalite**	Kinoshitalite**	Kinoshitalite†
α	1.598 (2)	1.592 (2)	1.621	1.619	1.615 (2)
β	1.616 (2)	1.614 (2)	1.639	1.633	1.630 (2)
γ	1.616 (2)	1.614 (2)	1.641	1.635	1.630 (2)
$\gamma - \alpha$	0.018	0.022	0.020	0.016	0.015
$2V_x$	0°	0–5°	25°	23°	very small
Absorption	$X < Y = Z$	$X < Y = Z$	$X < Y \cong Z$	$X < Y \cong Z$	colorless

* This study.
** Iwate Prefecture, Japan (Yoshii and Maeda, 1975).
† Kyoto Prefecture, Japan (Yoshii et al., 1973a).

derived from the metasedimentary rocks. Because the partition coefficient of Ba between alkali feldspar (the dominant Ba-bearing mineral in the plutonic rocks) and hydrothermal solution at 700–800°C strongly favors the feldspar (Carron and Lagache, 1980), it seems unlikely that the plutonic rocks were the source. Furthermore, in the presumably CO_2-rich fluids generated during contact metamorphism of calcareous metasedimentary rocks, the solubility of $BaCO_3$ decreases with increasing temperature (Holland and Malinin, 1979). Therefore, Ba mobilization is commonly considered to be negligible during metamorphism (Barbey and Cuney, 1982) and even during anatexis (Lee and Doering, 1974). It seems most likely that the source of Ba was within the metasedimentary rocks of the contact zone. Country rocks surrounding the pluton that have not been recrystallized by contact metamorphism have been analyzed and contain up to about 2000 ppm Ba, thus providing a viable source for the Ba.

CONCLUSION

Two different compositions of Ba-rich micas from the Alaska Range are present in apparent equilibrium in contact-metamorphosed calcareous metasedimentary rocks. Though most closely approximating the Ba-Mg-rich brittle mica kinoshitalite, the end-member composition of which has never been observed in nature, the specimens reported herein both have significantly different compositions and optical properties from previously reported barian micas. The Middle Fork samples have lower Mn, higher Mg, smaller $2V_X$, and a different color than other known kinoshitalites. Petrologically, they seem to have formed at fairly high temperatures (≥600°C), but not necessarily at high pressure. The Ba was probably present within the recrystalling metasedimentary rocks and became incorporated into the micas as a function of very localized control of whole-rock and fluid composition during contact metamorphism.

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