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THE TANCO PEGMATITE AT BERNIC LAKE, MANITOBA.

VI. LITHIUM-RUBIDIUM-CESIUM MICAS

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ABSTRACT

Fourteen specimens of Li-Rb-Cs micas from four different zones in the Tanco pegmatite, Bernic Lake, Manitoba, and two similar micas from other localities have been examined in detail. X-ray powder photographs and diffractograms of all 16 specimens and single-crystal precession photographs of four of them show that twelve have the muscovite-type structure $2M_1$, one the structure type $1M$, and the remaining three consist of intimate mixtures of structures $2M_1$ and $1M$ with two having $2M_1 > 1M$ and one $1M > 2M_1$. For the 14 specimens from Tanco, the refractive indices in the plane of the cleavage β or β' vary from 1.560 to 1.593 and γ or γ' from 1.561 to 1.597. Complete chemical analyses on six of the specimens, and partial analyses of six others, all from Tanco, showed the following variations in weight percent: Li_2O 0.15 — 4.52, Rb_2O 1.50 — 3.93, Cs_2O 0.12 — 0.79; the six completely analyzed specimens showed F to vary from 0.64 to 6.10 weight percent. The analyses show that with increasing Li_2O there is a definite or suggested increase in Rb_2O, Cs_2O, SiO_2 and F, and a decrease in Al_2O_3. Seven specimens have $Li_2O < 0.56$, and the other five analyzed specimens have Li_2O between 3.12 and 4.52 weight percent. Nine specimens with $Li_2O \leq 3.38$ weight percent consist of structure type $2M_1$, and the remaining three specimens with Li_2O 3.50 — 4.52 weight percent consist of "mixed type" $2M_1 + 1M$. The cell dimensions show an almost negligible variation with change in composition. The refractive indices γ (or γ') decrease regularly with increasing Li_2O and F, the change probably being due mainly to the increase in F. The paragenesis of the micas from the Tanco pegmatite is discussed briefly.

INTRODUCTION

One of the several groups of minerals from the Li-Cs-Be-Ta pegmatite at the Tanco mine, Bernic Lake, Manitoba, that has been recently investigated in detail in the Mineralogical Laboratory of the Department of Earth Sciences, University of Manitoba, is the Li-Rb-Cs-bearing micas. This paper, taken mainly from an unpublished thesis by Rinaldi (1970), reports the results of this investigation.

The location, geology, and general mineralogy of the Tanco pegmatite, formerly called the Montgary and later the Chemalloy pegmatite, are described by Hutchinson (1959) and Wright (1963), and most recently in a paper accompanying this one, by Crouse and Černý (1972). More detailed descriptions of the minerals in this pegmatite are given by Nickel (1961), and companion papers to the present one describe in detail several of the other mineral groups. X-ray powder data for several mica specimens from this pegmatite are given by Bristol (1962).

The purposes of the present investigation were : (1) to define crystallographically and chemically, the different varieties of mica that occur in the Tanco pegmatite ; (2) to examine the variations of certain physical properties with Li, Rb, Cs and F contents in the Tanco micas as well as in others ; and (3) to provide, along with the parallel investigations of other mineral groups in the pegmatite, data necessary for a genetic interpretation of the Tanco pegmatite.

The most important published papers relating chemical composition and physical properties in Li-bearing Fe-poor micas are those by Levinson (1953) and Foster (1960), and one on Rb-bearing muscovites by Gordiyenko & Denisov (1964). Reference is made later to these and other papers.

MATERIAL AND EXPERIMENTAL METHODS

Material

The investigation reported here was carried out on 14 mica specimens from the Tanco mine; however only 12 samples were chemically analyzed. The two micas for which only physical properties were determined (RNB. 330 from zone (4) and R.15 from zone (5)) are not included in Table 1, which gives brief descriptions of each specimen, the zone from which each specimen was collected, and the source of each specimen. Fuller descriptions are given by Rinaldi (1970, pp. 7-11). The locations of the specimens collected by R. R. are shown on a location map by Rinaldi (1970, Fig. 1) but, because some of the locations have since been removed by the mining operation, the location map is not reproduced here. For most readers, the maps of the mine geology by Crouse & Černý (1972, Figs. 3 and 4), showing the different zones will provide a satisfactory enough correlation between specimen location and the mine geology.

Experimental methods

The experimental work carried out on these mica specimens was of three types. (1) Complete chemical analyses on six of the specimens and partial analyses on six others were carried out using standard methods. All the metallic elements except Si and Al were determined by atomic absorption ; these two elements were determined by x-ray fluorescence. Fluorine was determined by a photometric method described in a personal communication to P. Černý from P. Povondra & R. Valach (1969) in Prague. (2) X-ray powder diffraction photographs using 114.6 mm diameter cameras, and x-ray powder diffractograms, both taken with Cu$K\alpha$ radiation, were recorded for 14 specimens, and single-crystal precession photographs were taken of four of them. (3) The two refractive indices in the plane of the cleavage were measured for all 14 specimens.

EXPERIMENTAL RESULTS

X-ray diffraction

The 14 specimens yielded three different types of *x*-ray powder photographs : some characterictic of a $2M_1$ structure, some of a $1M$ structure, and the remainder of a mixture of these two. Fig. 1 reproduces powder photographs of the three types, the mixed type shown in Fig. 1b having

TABLE 1. DESCRIPTION OF SPECIMENS

Specimen Number	*Zone Number**	*Description*	*Source***
R. 1	(4)	Yellowish white muscovite in crystals 5-20 mm in size.	R.R.
R. 2	(4)	Greenish white muscovite in books or crystals ~ 10 mm across.	R.R.
R. 3	(4)	Violet mica in small (1-3 mm) curved crystals elongated along [001].	R.R.
R. 4	(4)	Greenish white muscovite in large (2-3 cm) crystals.	R.R.
R. 7	(9)	Massive violet lithian muscovite.	R.R.
R. 8	(6)	Fine-grained green muscovite.	R.R.
R. 9	(9)	Massive violet mica similar in appearance to R. 7	R.R.
R. 12	(5)	Flaky white muscovite which breaks readily into needle-shaped laths.	R.R.
R. 13	(5)	Greenish yellow medium-grained (~ 3 mm) muscovite.	R.R.
RBLM. 72	(6)	Yellowish green massive, muscovite containing relics of K-feldspar. Similar to R. 8.	R.R.
REN. 1	(4)	Curved violet mica in large columnar aggregates; sample no. 1 in Nickel (1961, p. 13).	E.H.N.
REN. 5	(9)	Fine-grained violet mica similar to R. 7 and R. 9; sample no. 5 in Nickel (1961, p. 13).	E.H.N.

* Zones numbered as follows (Crouse & Černý 1971) :
(4) : lower intermediate : K-feldspar-albite-quartz-spodumene (-amblygonite) ;
(5) : upper intermediate : spodumene-quartz-amblygonite (-petalite) ;
(6) : central intermediate : K-feldspar-albite-quartz (-beryl-wodginite) ;
(9) : lepidolite : Li-muscovite.

** Source symbols as follows : R.R. — collected by Romano Rinaldi ; E.H.N. — provided by E. H. Nickel, Mines Branch, Ottawa.

$1M \gg 2M_1$. The powder diffractograms confirmed or clarified the nature of each sample as one of the three types $2M_1$, $1M$, or mixtures of the two. However, the diffractograms indicated that the mixed types may have widely different proportions of the two structure types, and different specimens showed $2M_1 \gg 1M$, $2M_1 > 1M$ and $1M \gg 2M_1$. Some representative diffractograms with distinctive reflections labelled are reproduced in Fig. 2. The structure types found for twelve of the specimens are given in Table 3 (with the chemical and optical results) ; the remaining two specimens, R.15 and RNB.330 were found to have structure $2M_1$ and $1M$ respectively.

Several prominent reflections in the powder patterns of the two pure structure types $2M_1$ and $1M$ as published by Yoder & Eugster (1955) are

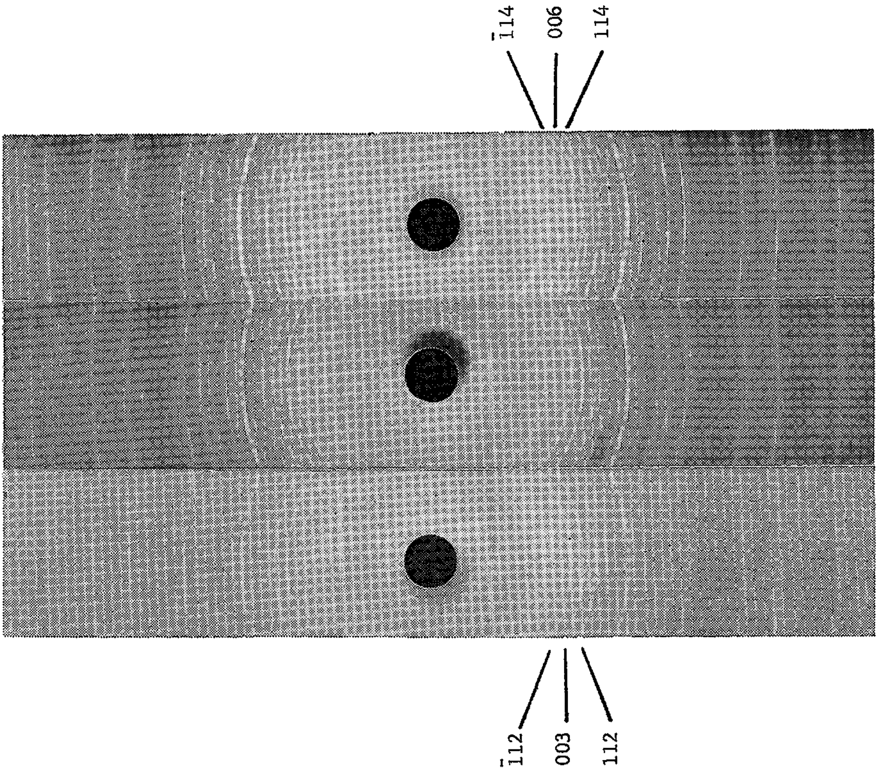

FIG. 1. X-ray powder photographs taken with Cu/Ni radiation and 114.6 mm diameter cameras, illustrating three of the different structure types. The indices for several distinctive reflections for the two pure structure types are shown.
a (Top) Type $2M_1$; specimen REN. 5.
b Type $1M \gg 2M_1$; specimen REN. 1.
c (Bottom) Type $1M$; specimen RNB. 330.

distinctive for each type, and hence useful in differentiating powder patterns of the two types and especially in interpreting patterns of mixed types. The data for these two distinguishing sets of reflections are given

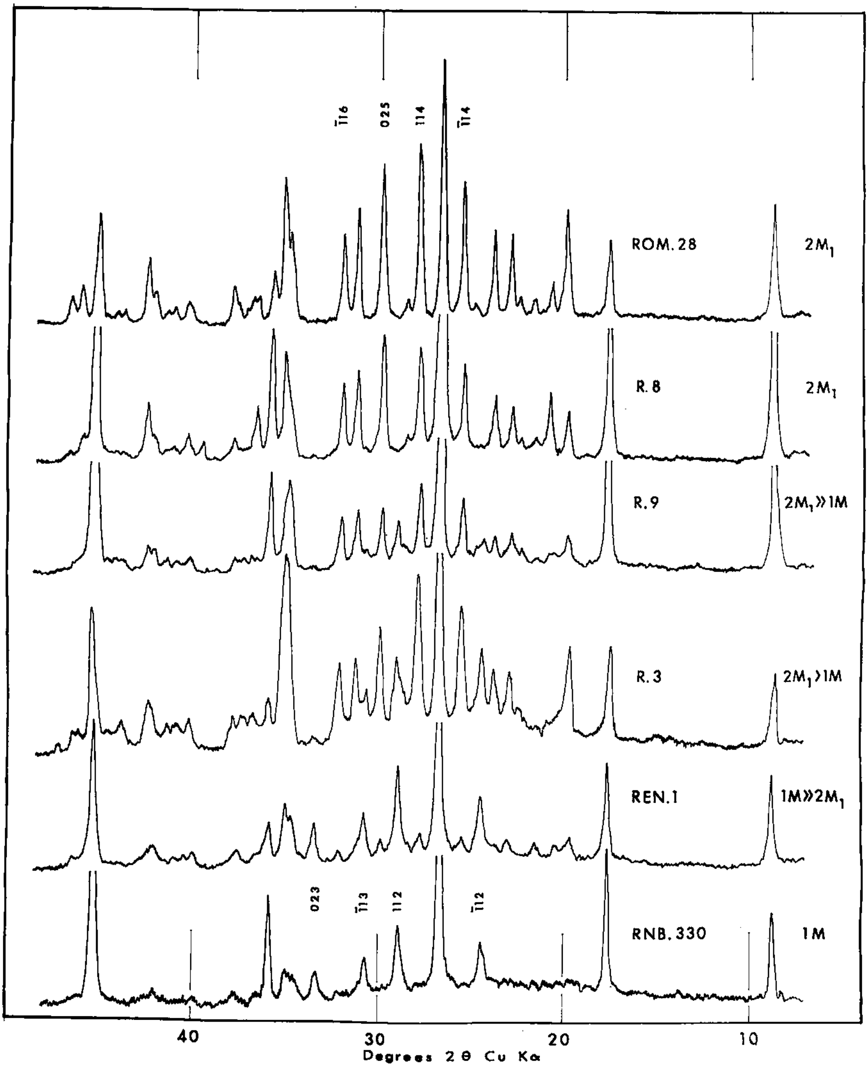

FIG. 2. X-ray powder diffractograms taken with Cu/Ni radiation of representatives of the different structures types. The indices for several distinctive reflections for the two pure structure types are shown.

in Table 2 and, as noted above, these reflections are marked on Figs. 1 and 2.

Using accurate spacings taken from highly resolved powder diffractograms calibrated with an internal standard (annealed synthetic CaF_2), and using moderately accurate cell dimensions for the two pure structure types obtained from single-crystal precession photographs (as described below), the powder patterns of all samples were indexed and more accurate cell dimensions were derived (for the one or the dominant structure type) using the computer program of Evans *et al.* (1963) as modified by D. E. Appleman. The indexed data for five representative specimens are given by Rinaldi (1970, Table 1), but they are not reproduced here because they are essentially the same as the data for the two pure structure types given by Yoder & Eugster (1955) and reproduced as Powder Diffraction File cards 7-32 ($2M_1$) and 7-25 ($1M$). The refined cell dimensions of the pure or dominant phase of the 16 specimens are given by Rinaldi (1970, Table 2), but because the dimensions for each of the two phases show little variation, only one example for each phase is given here :

	Sample No.	*a*Å	*b*Å	*c*Å	β
$2M_1$	R. 8	5.190 ± .003	9.006 ± .007	20.189 ± .003	95°47′ ± 02′
$1M$	REN. 1	5.224 ± .001	9.013 ± .002	10.205 ± .001	100°50′ ± 01′

The means and the standard deviations of the standard errors for all the individual cell dimensions are *a* 0.004 ± .004, *b* 0.011 ± .005, *c*. 0.006 ± .002 Å, β 3 ± 1′.

Single-crystal precession photographs of four representative specimens (Rinaldi 1970, pp. 22-32) confirmed the structure types as $2M_1$ for specimens R.7, R.9 and R.13, and $1M$ for specimen REN.1. On the basis of these results, we take all of the few specimens showing the $1M$-type powder

TABLE 2. DISTINCTIVE X-RAY POWDER REFLECTIONS FOR STRUCTURE TYPES $2M_1$ AND $1M$

(See Figs. 1 and 2)

$2M_1$ *Structure (Specimen R. 8)*				$1M$ *Structure (Specimen RNB. 330)*			
2θ° (CuKα)	*d*Å	*I*	*hkl*	2θ° (CuKα)	*d*Å	*I*	*hkl*
25.83	3.449	32	$\bar{1}14$	24.47	3.638	16	$\bar{1}12$
27.82	3.207	40	114	26.94	3.091	28	112
29.80	2.998	50	025	30.75	2.908	16	$\bar{1}13$
31.95	2.801	28	$\bar{1}16$	33.39	2.683	10	023

patterns as containing true 1*M* micas, and we do not consider the occurrence of the 3*T* structure type probable (this structure type cannot be distinguished from 1*M* by powder methods). The presence of $2M_2$ was not indicated in single-crystal or powder *x*-ray patterns. The single crystal *x*-ray photographs also confirmed the space group of $2M_1$ as $C2/c$ (or Cc) and that of 1*M* as $C2/m$ (or $C2$ or Cm). These photographs further confirmed the cell dimensions as those already established for the respective structure types by, for example, Yoder & Eugster (1955) ; the cell dimensions (Rinaldi 1970, p. 32) agree, with one exception, with those given above from the refined powder data.

Optics

A partial determination of the optical properties was carried out on the 14 specimens described in Table 1. The minimum and maximum indices of refraction in the plane of the cleavage, β or β' and γ or γ', were measured by the immersion method using sodium light (λ = 5890 Å). In order to make sure that each mica flake used for a determination was lying flat, interference figures were observed, and only those flakes yielding well-centred figures were used for refractive index measurements.

Optical examination has shown that the examined micas are mostly homogeneous on the scale of the optical microscope, the only exception being some flakes of the mixed-type specimens. No conspicuous inhomogeneity was observed as, *e.g.*, that observed in some pegmatitic Li-rich micas by Rimsaite (1968). Thus most of the mixed structural types are believed to be inhomogeneous on a submicroscopic *x*-ray scale, and the data presented here for these specimens should be regarded as characteristic for bulk compositions indicated by the "structure type" statement in Table 3.

Chemistry

Complete chemical analyses were carried out on six of the specimens, and partial analyses for Fe, Mg, Li, Na, K, Rb and Cs on another six specimens. The chemical analyses are given along with the structure types and refractive indices in Table 3, as are the numbers of atoms on the basis of 20(O) + 4(F + OH) for six completely analyzed specimens. It is important to note that the analyses for H_2O (+ and −) were not taken into account in recasting the analyses to give the numbers of atoms. The reason the results were treated in this way is because the authors regard the H_2O analyses as unreliable. The H_2O + and the H_2O − analyses on a given sample were done at different times many weeks apart; it was only after the analyses were completed and the material destroyed, we realized that, because of the strong tendency of finely-powdered mica to absorb water from the atmosphere, rigid control should have been exercised over the H_2O- and H_2O+ analyses. In calculating the numbers of atoms, we have, therefore, used ideal numbers of anions, combining the determined F with sufficient OH to make $(F + OH) = 4$, and then combining this with $20(O)$ to make total anions $20(O) + 4(F + OH)$. The numbers of cations deduced in this way appear reasonable when compared with those given by, for example, Foster (1960).

As Table 3 shows, two of our analyses are on specimens (REN. 1 and REN. 5) which had been analyzed previously by Nickel (1961), and we give Nickel's results with our own in Table 3. We cannot account for some of the striking differences in the results obtained from the two different laboratories on the same specimen. Some of these differences may be due to the fact that, although for a given specimen the two analyses were carried out on material from the same hand specimen, the actual powder used was different. It seems unlikely, however, that this could account for some of the largest and most important differences, namely Rb_2O of 3.18% and 5.10% for REN. 5, and F of 6.10% and 1.51% for REN. 1 and of 4.75% and 1.44% for REN. 5. Regarding the Rb results, other Rb analyses done in our Chemical Laboratory suggest that our Rb data are reliable. Regarding the gross differences in the F contents obtained by the two different laboratories on both specimens, we think that our much higher F contents are more like those to be expected for micas with our Li_2O contents, judging by compilations of analyses such as those in Foster (1960, Table 6, p. 142) and in Deer, Howie & Zussman (1962, Table 16, p. 88). We feel this to be so despite Nickel's more satisfactory totals. On the other hand, analytical errors could be responsible for the irregular behaviour of Si and Al (specimens R. 7, R. 9, and, partially, REN. 1 ; Fig. 3) ; in this case, Nickel's data fit better than ours the expected changes in Si and Al with Li. In general, although some of our analyses may have appreciable error, both the numbers of atoms given in Table 3, and the variations of physical properties with alkali contents described in the next section, suggest that our results (excluding H_2O) are reasonably accurate throughout.

DISCUSSION

Chemical composition

We have noted in the preceding section that the contents of the different metallic elements and of F given by our analyses for our specimens are in general agreement with those given in published compilations of

TABLE 3. CHEMICAL AND OPTICAL RESULTS AND STRUCTURE TYPES

The specimens are listed in order of increasing Li_2O contents.

Sample No.	R. 13	RBLM.72	R. 12	R. 8	R. 2	R. 1	R. 4	R. 7	REN. 5		R. 9	R. 3	REN. 1	
Structure Type	$2M_1$	$2M_1$	$2M_1$	$2M_1$	$2M_1$	$2M_1$	$2M_1$	$2M_1$	$2M_1$		$2M_1 > 1M$	$2M_1 \gg 1M$	$1M \gg 1M_1$	
										Nickel*				Nickel*
SiO_2	n.d.	n.d.	n.d.	43.20	n.d.	n.d.	n.d.	47.80	44.85	(48.00)	47.50	45.25	45.75	(51.65)
Al_2O_3	n.d.	n.d.	n.d.	34.36	n.d.	n.d.	n.d.	28.64	26.64	(28.31)	28.64	25.90	28.84	(26.65)
FeO (Total Fe)	0.41	0.31	0.45	0.32	0.50	0.59	0.51	0.06	0.05	(0.26)	0.03	0.07	0.08	(0.06)
MnO	n.d.	n.d.	n.d.	0.07	n.d.	n.d.	n.d.	0.20	0.38	(0.50)	0.22	0.21	0.42	(0.44)
MgO	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.02	(2.08)	<0.01	0.37	0.00	(0.90)
Li_2O	0.15	0.21	0.32	0.37	0.42	0.54	0.56	3.12	3.38	(3.42)	3.50	3.97	4.52	(4.28)
Na_2O	0.33	0.24	0.82	0.22	0.92	0.95	0.85	0.19	0.19	(0.17)	0.19	0.41	0.32	(0.40)
K_2O	9.75	11.48	9.42	10.12	9.39	9.00	9.20	9.98	9.43	(8.61)	9.45	9.12	9.00	(8.68)
Rb_2O	1.50	1.56	1.81	1.69	2.05	2.12	2.24	2.91	3.18	(5.10)	3.59	3.46	3.93	(4.47)
Cs_2O	0.24	0.20	0.12	0.21	0.21	0.21	0.12	0.50	0.55	(n.d.)	0.71	0.79	0.79	(n.d.)
F	n.d.	n.d.	n.d.	0.64	n.d.	n.d.	n.d.	2.73	4.75	(1.44)	4.08	4.18	6.10	(1.51)
H_2O^+	n.d.	n.d.	n.d.	6.00	n.d.	n.d.	n.d.	5.60	4.84	(2.74)	4.38	6.84	2.17	(2.13)
H_2O^-	n.d.	n.d.	n.d.	0.80	n.d.	n.d.	n.d.	0.48	1.60	(—)	0.21	0.73	3.51	(—)
Subtotal				98.01				102.22	99.86	(100.63)	102.51	101.30	105.43	(101.17)
O = 2F				—0.27				—1.15	—2.00	(—0.61)	—1.72	—1.76	—2.57	(—0.64)
TOTAL				97.74				101.07	97.86	(100.02)	100.79	99.54	102.86	(100.53)

Analysts: R. Hill of the Chemical Analytical Laboratory in the authors' Department and R. Rinaldi for all analyses except those of F which were done by K. Ramlal, head of our Analytical Laboratory, and by Mr. Hill.

* Analyses of specimens REN. 1 and REN. 5 given by Nickel (1961, Table 4) as his analyses numbers 1 and 5.

Numbers of Atoms on the basis of 20(O)+4(F+OH)

Note: The calculation of these numbers of atoms was done without using the H_2O contents given above, which the authors regard as unreliable (see text); the calculations were done assuming a total O^{-2} equivalent of 22, and sufficient OH^- to make (F + OH) = 4.

Sample No.	R.8		R.7		REN.5		R.9		R.3		REN.1	
Si	6.114	8.000	6.261	8.000	6.522	8.000	6.515	8.000	6.526	8.000	6.312	8.000
^{IV}Al	1.886		1.739		1.478		1.485		1.474		1.688	
^{VI}Al	3.884	4.098	3.205	5.073	3.086	5.116	3.144	5.104	2.927	5.342	3.000	5.565
Fe^{+2}	0.037		0.007		0.006		0.003		0.009		0.009	
Mn^{+2}	0.008		0.025		0.046		0.026		0.026		0.049	
Mg	-		-		0.004		-		0.079		-	
Li	0.209		1.836		1.974		1.931		2.301		2.507	
Na	0.060	2.050	0.053	2.222	0.053	2.132	0.049	2.058	0.113	2.158	0.085	2.063
K	1.826		1.865		1.749		1.653		1.677		1.584	
Rb	0.152		0.274		0.296		0.315		0.320		0.348	
Cs	0.012		0.030		0.034		0.041		0.048		0.046	
F	0.286	4.000	1.264	4.000	2.184	4.000	1.770	4.000	1.905	4.000	2.661	4.000
OH	3.714		2.736		1.816		2.230		2.095		1.339	

Optical Properties

Sodium D line, $\lambda = 5890\ \mathring{A}$

Estimated error of refractive indices given to four figures is 0.001.

Sample No.	R.13	RBLM.72	R.12	R.8	R.2	R.1	R.4	R.7	REN.5	R.9	R.3	REN.1
β or β'	1.589	1.591	1.589	1.591	1.593	1.588	1.589	1.576	1.572	1.572	1.574	1.560
γ or γ'	1.591	1.597	1.593	1.595	1.595	1.590	1.591	1.578	1.573	1.573	1.575	1.561

analyses of Li-bearing micas by, for example, Foster (1960) and Deer *et al.* (1962). Specifically, it can be noted that our results show the same trends indicated by the results of others, namely, that with increasing Li_2O, there is in general increasing Rb_2O, Cs_2O, and F (Fig. 3). As noted above, the irregular variations of Al_2O_3 and SiO_2 with Li_2O, also shown in Fig. 3, are possibly influenced by analytical errors. Fig. 3 also reveals that our specimens have either low $Li_2O < \sim 1/2\%$, or high, with $Li_2O > \sim 3\%$. Foster's (1960) compilation of a much larger number of Li-mica analyses

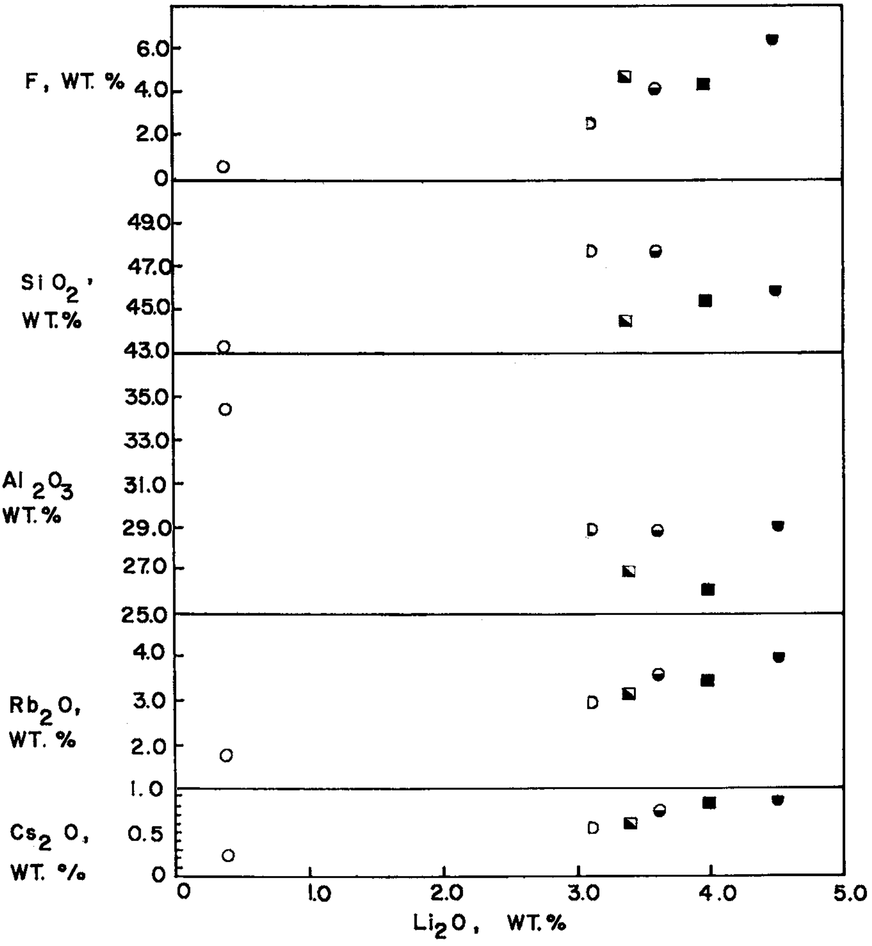

FIG. 3. Variations with Li_2O contents of Rb_2O, Cs_2O, Al_2O_3, SiO_2 and F. The specimen numbers corresponding to the different symbols are given in the box in Fig. 6.

shows relatively few specimens with Li_2O contents between ~ 1% and ~ 3½%.

Change of structure type with chemical composition

Levinson (1953) has correlated the structure types of Li-bearing micas with Li_2O content, and it is useful to compare our correlation with the one observed. If one notes that his "normal" muscovite and his 1-layer monoclinic lepidolite are now designated structure types $2M_1$ and $1M$ respectively, and that his 3-layer hexagonal lepidolite and his 6-layer monoclinic lepidolite have been shown by Smith & Yoder (1956) to be equivalent to structure types $3T$ and $2M_2$ respectively, then Levinson's correlation between structure type and Li_2O content and our corresponding correlation taken from Table 3 and summarized in Fig. 4, can be compared as follows:

Levinson (1953)		*This Paper*	
Wt.% Li_2O	*Name & structure type*	*Wt.% Li_2O*	*Name & structure type*
$< \sim 3.3$	lithian muscovite, $2M_1$	< 3.50	lithian muscovite, $2M_1$
3.4-4.0	transitional structure, $2M_2+2M_1$	3.50-3.97	mixed type, $2M_1 + 1M$, $2M_1 > 1M$
4.0-5.1	lepidolite, $2M_2$	4.52	mixed type, $2M_1 + 1M$, $1M > 2M_1$
> 5.1	lepidolite, $1M$	(> 4.52	no specimens examined)

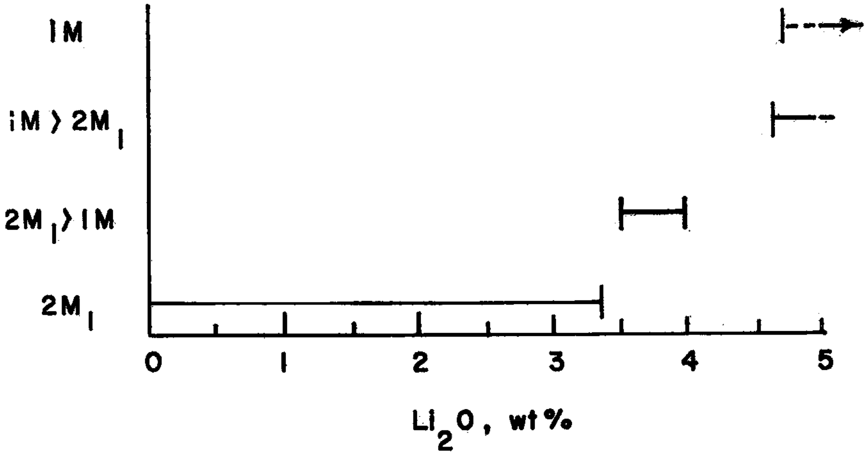

FIG. 4. Schematic representation of the relationship between Li_2O content and structure type.

Our results confirm Levinson's that the upper limit in Li_2O content for lithian muscovite with the structure type $2M_1$ is about 3½ wt. %. Our results also indicate that the transition at this amount of Li_2O is to a two-phase mica, Levinson's "transitional structure" and what we prefer to call "mixed type." However, Levinson found a $2M_2$ phase in these mixtures and in the 4.0 – 5.1 wt. % Li_2O range, whereas in our specimens the 1M structure appears. According to Levinson, this latter type is characteristic of lepidolites with > 5.1 wt. % Li_2O. Deviations from Levinson's scheme are also known from other studies (*e.g.* Berggren 1940 : *1M* with 4.35 wt. % Li_2O ; Lundblad 1942 : *1M* with 4.55 wt. % Li_2O ; Staněk 1965 : *1M* with 4.23 wt. % Li_2O ; Heinrich 1967 : $2M_2$ with 5.26, and a *1M* + $2M_2$ mixture with 4.26 wt. % Li_2O ; Černý *et al.* 1970 : *1M* < $2M_2$ mixtures with 5.42 and 5.72, and $2M_1$ with 5.91 wt. % Li_2O). These results suggest that the structures of the Li-rich lepidolites are determined not only by the Li-contents but also by the overall chemical composition and/or the conditions of crystallization (as proposed by Munoz (1968) on the basis of experimental work).

As to the nature of the transition from a one-phase lithian muscovite $2M_1$ at about 3½% Li_2O to a one-phase lepidolite *1M* or $2M_2$ at 4-4½% Li_2O, our conclusions agree with those of Levinson that in this composition range the Li will tend to form a Li-rich true lepidolite and a Li-poor (lithian) muscovite either intimately intergrown or as separate grains. This is strongly suggested not only by Levinson's and our X-ray powder diffraction records of individual samples, but also by the fact that one of our specimens, RNB.330 (not chemically analyzed), showed from powder diffractograms of three different portions, one portion as $2M_1$ only, a second as $2M_1$ » *1M*, and a third as *1M* only.

Variation of cell dimensions with chemical composition

One might expect that with larger Li^{1+} replacing smaller Al^{3+}, and with larger Rb^{1+} and Cs^{1+} replacing smaller K^{1+}, there would be a corresponding increase in some or all of the cell dimensions. However, as we have noted earlier in the section describing our *x*-ray diffraction results, the variations in cell dimensions we obtained for each of the two structure types, $2M_1$ and *1M*, are small. The probable explanation for this is that suggested by S. W. Bailey (personal communication 1970) : the octahedral substitution of Li^{1+} for Al^{3+} and the simultaneous tetrahedral substitution of Si^{4+} for Al^{3+} provide a better fit of the larger tetrahedral sheet onto the smaller octahedral sheet, and reduces the tetrahedral rotation necessary to adjust for the dimensional misfit in Li-poor micas. The decreased tetrahedral rotation increases the size of the interlayer cavities containing K^{1+}, and the larger Rb^{1+} and Cs^{1+} may enter these sites without noticeable expansion of the structure. On the other hand, Gordiyenko & Denisov (1964) have shown that Rb^{1+} and Cs^{1+} considerably increase the *c* dimension in true muscovites in which the substitution of rare alkalies for K^{1+} is the only chemical change and there is no other crystallochemical adjustment. This observation is compatible with Bailey's interpretation of these structures.

Plots of the individual cell dimensions *a*, *b* or *c* against any one of Li_2O, Rb_2O or Cs_2O, all of which increase with each other as shown in Fig. 3, show a suggestion of increasing cell dimension with increasing oxide. Because the effect in each case is so poorly defined, we are not including these plots here although Rinaldi (1970) does show plots of *a* and *b* against Rb_2O contents as his Figs. 11a and 11b. The monoclinic angle β in our $2M_1$ micas shows a change, but not a regular variation, with Li_2O content (Fig. 5). This figure also shows that the β angles fall into two groups, one larger with $\beta \sim 95°44' \pm 4'$ for the micas with $Li_2O < \sim ½\%$, and one smaller with $\beta \sim 95°33' \pm 3'$ for the micas with $Li_2O > \sim 3\%$. Only one aspect of the cell dimensions, the cell volume, shows a reasonably regular variation with Li_2O content (Fig. 5). This Figure shows that the cell volumes are essentially constant for those micas with $Li_2O < \sim ½\%$, but that they increase fairly regularly with increasing Li_2O for those micas with $Li_2O > \sim 3\%$. Fig. 5 also suggests a grouping of the plotted points of Li_2O *vs.* cell volume into two subparallel lines, for which we have no explanation. This feature may be fortuitous in which case it would presumably disappear if more data were available.

Variation of refractive indices with chemical composition

Plots of the refractive indices γ or γ' against Li_2O contents for the 12 partially and completely analyzed specimens, and of these refractive indices against F contents for the six completely analyzed specimens, using the data in Table 3, are shown in Fig. 6. Both plots show well the previously known relationship that these refractive indices decrease fairly regularly with increasing amounts of these elements. The "clustering" of the refractive indices in the Li_2O plot results mainly from the fact noted earlier under Chemical Composition that Li_2O composition between ½ and 3% are not represented in our specimens.

Because Rb_2O, Cs_2O and SiO_2 also increase with increasing Li_2O (Fig. 3), plots of the refractive indices against these three elements are similar to those shown for Li_2O and F in Fig. 6, but they are not reproduced here. Similarly, because Al_2O_3 decreases with increasing Li_2O (Fig. 3), the refractive indices increase with increasing Al_2O_3.

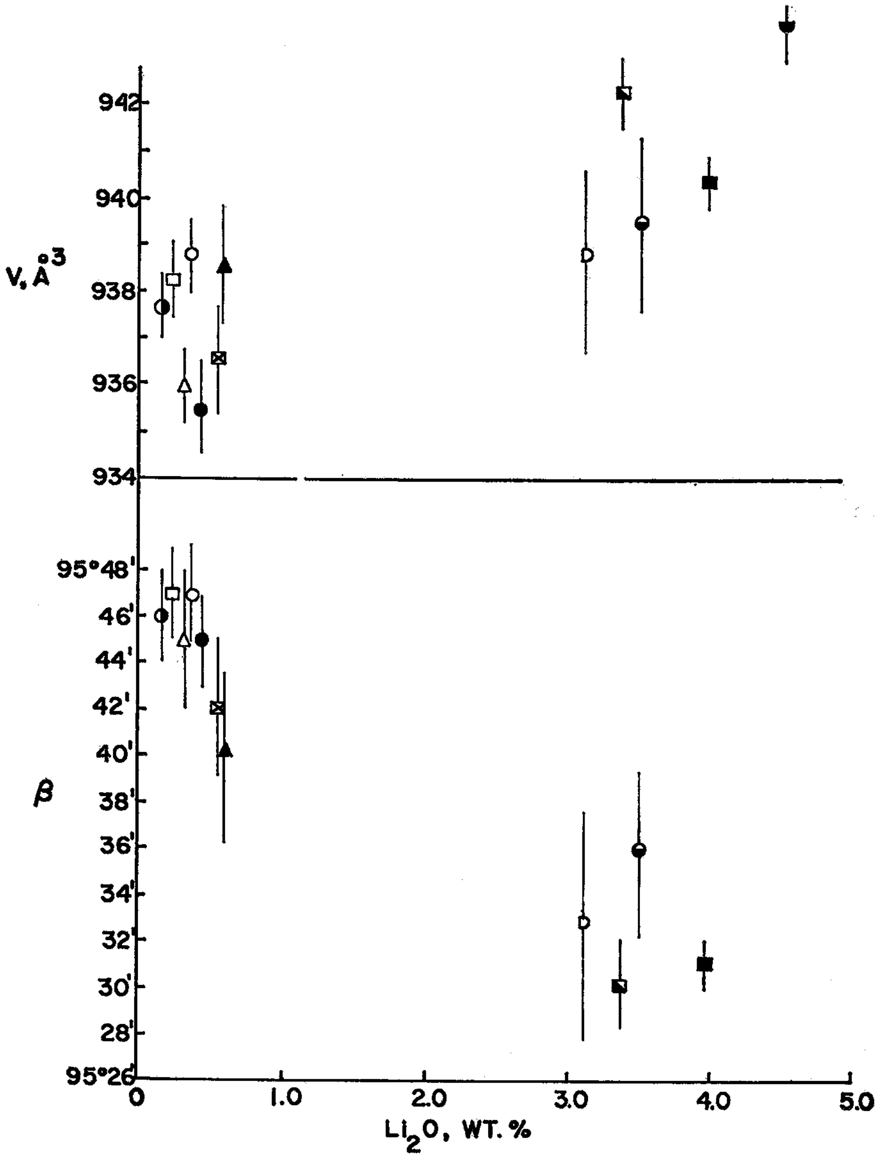

FIG. 5. Variation with Li_2O content of the monoclinic angle β in $2M_1$ structure types, and of the cell volume V in $Å^3$ for all specimens. The specimen numbers corresponding to the different symbols are given in the box in Fig. 6.

Although the refractive indices vary with changing amounts of all of these elements, it seems likely that, with the known marked variation of the refractive indices with F : (OH) ratio in minerals such as topaz, the

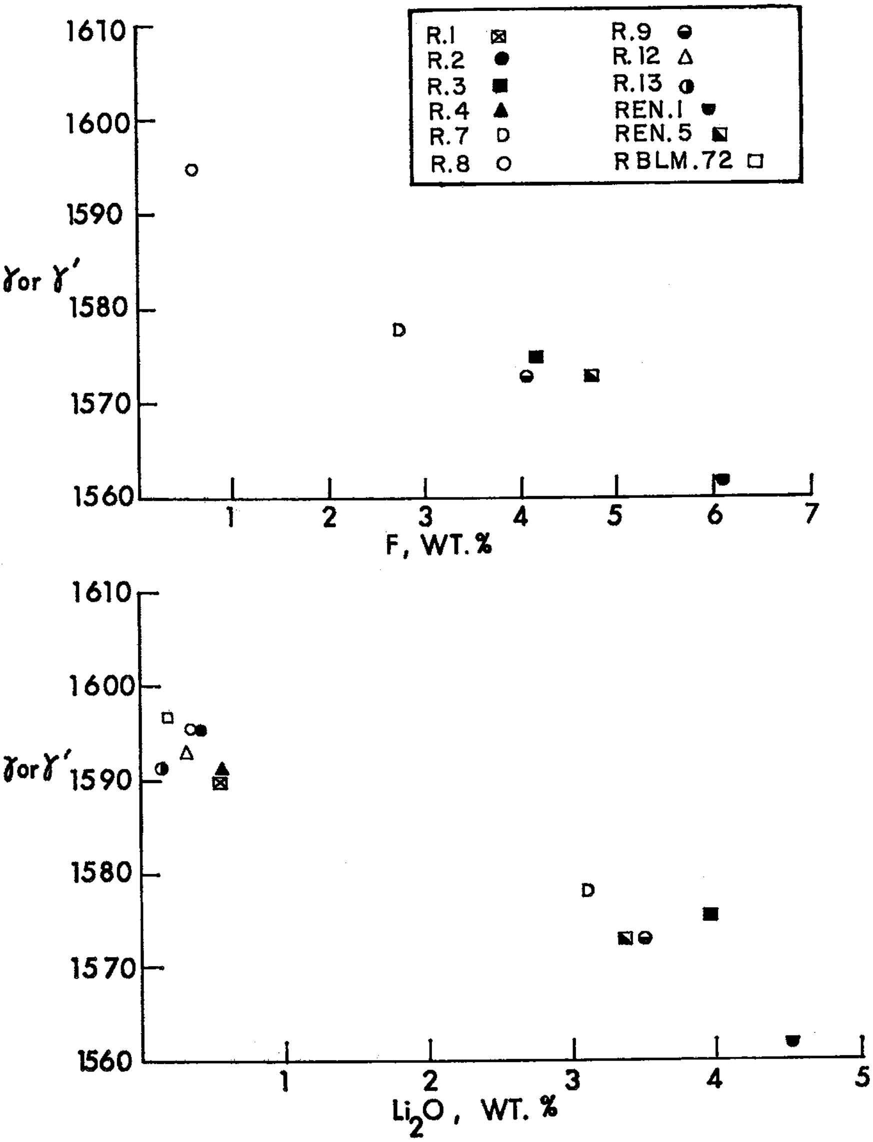

Fig. 6. Variations with Li_2O and F of γ or γ'. The specimen numbers corresponding to the different symbols are given in the box.

main contributor to the change in refractive indices in the Li-micas is probably the change in the F : (OH) ratio.

Paragenesis of the Li-micas in the Tanco pegmatite

The data obtained in the present study are not sufficient for a complete assessment of the petrological characteristics of the micas in the Tanco pegmatite ; nevertheless, they provide some interesting information on the paragenesis in which they occur.

In the lower intermediate zone (4) (Crouse & Černý 1972, and our Table 1) the chemical compositions and physical properties of the micas vary within wide limits (specimens R. 1, R. 2, R. 3, R. 4 and REN. 1). This can be expected in a zone which is typically inhomogeneous, consisting of patchy aggregates of widely different mineral associations. Purplish micas in this zone are always Li-rich and include some true lepidolites, whereas other colour varieties are Li-poor. The close association of micas with widely differing compositions within a single "morphological unit" reminds one of the zoning in amblygonite-montebrasite (Černá *et al.* 1972), which is most strikingly developed in this zone.

The only micas characteristic of the upper intermediate zone (5) (specimens R. 12 and R. 13) occur in its uppermost parts in close proximity to the pollucite zone (8). These greenish and white muscovites are very poor in Li, and their Rb and Cs contents are among the lowest found in any Tanco mica, despite their close association with pollucite.

The central intermediate zone (6) carries fine-grained green muscovite (specimens R. 8 and RBLM. 72). Their compositions are almost identical with those of green muscovites from zone (5), except for lower Na contents ; in zone (5) the Na-richer muscovites are associated with albite whereas in zone (6) the Na-poorer muscovites have apparently originated by replacement of microcline-perthite. The low Li-contents of these latter type minerals are not surprising in zone (6) because it is generally Li-poor ; the Rb and Cs contents of these micas were probably inherited from the replaced K-feldspar.

The fine-grained purple micas of the metasomatic lepidolite zone (9) (specimens R. 7, R. 9 and REN. 5) display the highest average Li-contents for micas from all zones of the Tanco pegmatite. In addition, the Li-contents seem to be quite constant in the different samples (3-3.5 wt. % Li_2O). However, most of the mica consists of lithian muscovite, and true lepidolite seems to be exceptional ; the designation "lepidolite zone" is retained in the descriptions of the mine geology only for convenience and conciseness. The maximum concentration of Li in the micas of this late metasomatic zone (9), formed at the expense of feldspars of zone (6), corresponds to the general observation that the Li-content of micas within a single Li-bearing pegmatite increases from the earliest types towards the latest-formed varieties.