

## LEUCOSPHENITE, AND ITS OCCURRENCE IN THE GREEN RIVER FORMATION OF UTAH AND WYOMING

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### ABSTRACT

The history of work on leucosphenite,  $\text{Na}_4\text{BaTi}_2\text{B}_2\text{Si}_{10}\text{O}_{30}$ , is reviewed. Its paragenesis in the Green River Formation in Utah and Wyoming is described and four other known occurrences in Greenland, Quebec, Eastern and Central Asia are briefly noted. Goniometric observations, optical properties, cell dimensions, and 2 new chemical analyses which confirm the formula are reported. Optical properties and cell dimensions of leucosphenite from many sources in the Green River Formation and from Mont St. Hilaire, Quebec, show little variation. Leucosphenite melts incongruently at about 800°C to a glass and  $\text{Ba}_2\text{TiO}(\text{Si}_2\text{O}_7)$ , fresnoite.

### HISTORY

Leucosphenite was first recognized as a distinct mineral in an alkalic pegmatite in the Narsarsuk region in Greenland and referred to as "petalite-like" by Gustav Flink (1898, p. 236). Three years later he named it and gave a comprehensive description (Flink, 1901, p. 137-146). Specimens have been preserved at the Mineralogisk Museum in Copenhagen, the British Museum of Natural History, London, and at Harvard University, Cambridge.

Half a century later Milton *et al.* (1953) reported the occurrence of leucosphenite as an authigenic mineral in sedimentary rocks of the Green River Formation (Eocene) in the Uinta Basin, northeastern Utah. Soon after, leucosphenite was also found in the Green River Formation in the Bridger Basin, southwestern Wyoming (Milton, 1957) and since has been noted at many points in this formation in both states.

Yefimov and Katayeava (1959) reported leucosphenite in pegmatite veins in an ultrabasic-alkaline massif in South Yakutia, U. S. S. R., and Dusmatov, *et al.* (1967) mention leucosphenite from the Alaiski massif, Turkestan, Central Asia. Most recently Chao *et al.* (1967) have found it in the nepheline syenite of Mont St. Hilaire, Quebec.

## ASSOCIATIONS

In the Narsarsuk region leucosphenite was found with elpidite, aegirine, epididymite, albite, and polyolithionite in a small pegmatite in syenite. The Yakutia leucosphenite occurs in two aegirine-feldspar veins in the Inaglina massif (Yefimov and Katayeva, 1959; Yefimov *et al.*, 1963, p. 156-157). Associated minerals include arfvedsonite, eudialyte, neptunite, lamprophyllite, and lorenzenite, mostly known also from the Narsarsuk syenite pegmatite though not mentioned specifically as associated with leucosphenite at the type locality. In the Turkestan occurrence leucosphenite is found with reedmergnerite, eudialyte, stillwellite, pyrochlore, calcite, and galena as accessories in pegmatites composed mainly of microcline, aegirine, albite, and quartz. At Mont St. Hilaire, leucosphenite is closely associated with albite, aegirine, narsarsukite, molybdenite, calcite, and fluorite.

Though these four occurrences of leucosphenite are by no means identical, they are all connected with alkalic igneous rocks and there is extensive correspondence among the associated rare minerals.

The occurrence of leucosphenite in the sedimentary Green River Formation in Utah and Wyoming is entirely different (Milton and Eugster, 1959; Bradley and Eugster, 1969). Even so, some of the other authigenic silicates in this formation (*e.g.*, albite and K-feldspar, riebeckite, acmite, Na-catapleite, labuntsovite, and elpidite) are typical of the alkaline igneous associations in which leucosphenite occurs elsewhere. However, in the Green River Formation the immediate associates of leucosphenite vary greatly in different localities. Nepheline has not been found, although hydrous sodium aluminosilicates (*e.g.*, analcime) are abundant and widespread. The Piceance Basin of the Green River Formation in Colorado is not known to contain any leucosphenite and differs mineralogically in other respects from the Green River Formation of Utah and Wyoming (Milton, 1971).

## MATERIAL

Leucosphenite has been found in the Green River Formation only in drill cores and cuttings despite much search of outcrops. In view of the stability of this mineral and the wide extent of outcrop of horizons which contain it at depth, this is surprising.

Materials available for this study include crystals and fragments from various localities in the Uinta Basin, Duchesne County, northeastern Utah, and crystals from the Bridger Basin, Sweetwater County, in southwestern Wyoming. The Utah material is normally turbid or obviously mixed with shale inclusions, but small portions of these crystals may be clear and uncontaminated and such were selected for analysis. In no case could enough be obtained from

a single crystal for a conventional wet analysis. In some poorly developed crystals (Figures 1 and 2) the matrix rock enclosed in the leucosphenite may be greater in amount than the leucosphenite itself. Figure 3 shows a thin section of typical Utah leucosphenite, with shortite, in the matrix rock.

#### LEUCOSPHENITE IN THE GREEN RIVER FORMATION

##### *Utah*

In reporting the discovery of leucosphenite in the Green River Formation in Utah, Milton *et al.* (1953) stated that "it has been found in several cores from oil wells in Duchesne and Uintah counties, and well cuttings indicate a vertical range of several thousand feet. It occurs in oil shale as well-formed crystals several millimeters in length, commonly associated with shortite,  $\text{Na}_2\text{Ca}_2(\text{CO}_3)_3$ . It also occurs in analcime-bearing layers in beds of the Green River Formation like those described by W. H. Bradley in 1930." The following year (Milton *et al.*, 1954) leucosphenite was described as occurring in the Green River Formation of Utah with the new minerals redmergnierite and eitelite and with searlesite, crocidolite, and shortite, in dolomitic shale. Six years later (Milton *et al.*, 1960a) leucosphenite

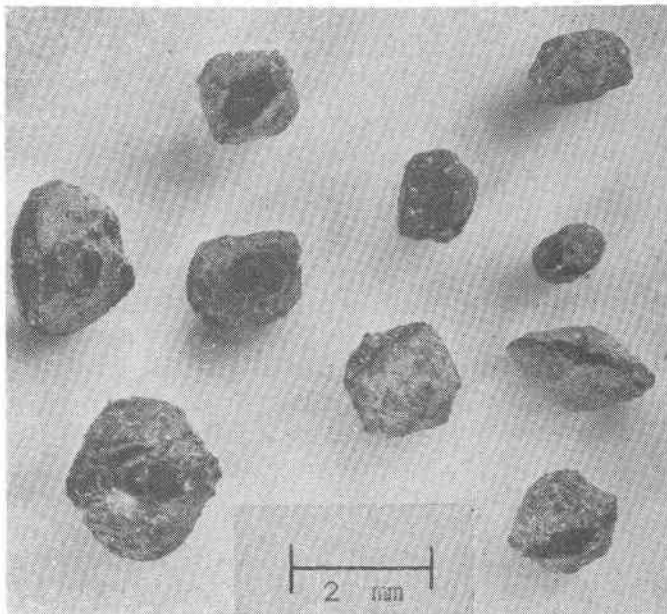


FIG. 1. Leucosphenite, Kermit Poulson well #1, 4500-4510 ft. Discoidal crystals heavily impregnated with brown shale matrix. Only an equatorial zone of prism faces is developed.

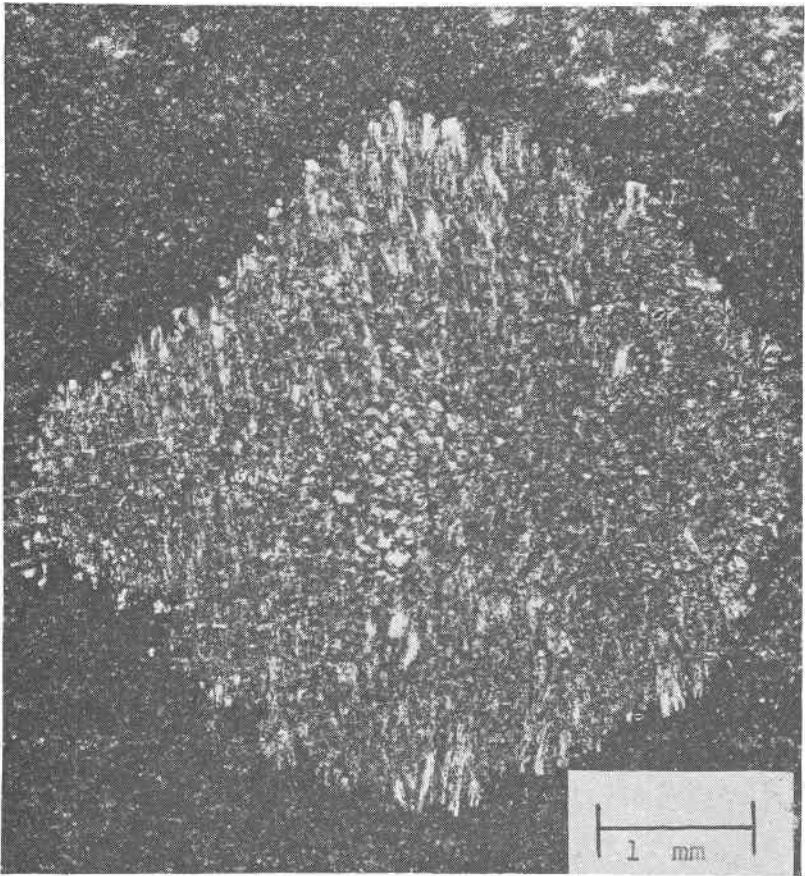


FIG. 2. Leucospheinite in thin section, Joseph Smith well #1, Uintah County, Utah, core #4, 4558-4595 ft. Note crystal growth out into matrix and vague composite outline.

was reported with reedmergnerite in six wells in Duchesne County. The host rock is mostly brown dolomitic rock except for black oil shale in the Joseph Smith No. 1 well. One of the six wells, the Sinclair Mortenson No. 1 well, is the site of the discovery of leucospheinite in the Green River Formation in brown oil shale (Milton, 1957, p. 142). Recently a seventh Duchesne County well, Continental Duchesne 638 No. 1 (Sec. 14, T4S, R5W), was found to contain a similar leucospheinite-reedmergnerite association. These seven wells are in an area comprising townships 3S and 4S north to south and ranges 1W to 5W east to west, mostly east of the town of Duchesne. The Mapco Shrine Hospital No. 1 well core, Sec. 10, T3S, R5W, shows much

reedmergnerite but no leucosphenite. Cuttings from the Kermit Poulson No. 1 well (one of the seven containing leucosphenite), type locality of eitelite, contained leucosphenite through a vertical range of almost 2000 feet.

A typical piece of core of dark rock containing leucosphenite is shown in Figure 4 (the attribution of locality may be erroneous). A tiny fragment of the matrix in this core is of such fine grain that it yields an X-ray powder pattern without grinding; dolomite is the main constituent. A thin section from the same core shows some detrital quartz grains which are large enough to yield spots on the X-ray pattern. Dolomite and calcite are exceedingly fine-grained and not separately identifiable. What appear to be grains of dark minerals at low magnification are clots of organic matter very unevenly distributed. Chips of the rock are slowly disintegrated by cold dilute HCl; the residue is mostly quartz. Other constituents include grains of muscovite and biotite plus probable albite, magnetite, and sphene. These are present in such small amount as to give no record in the

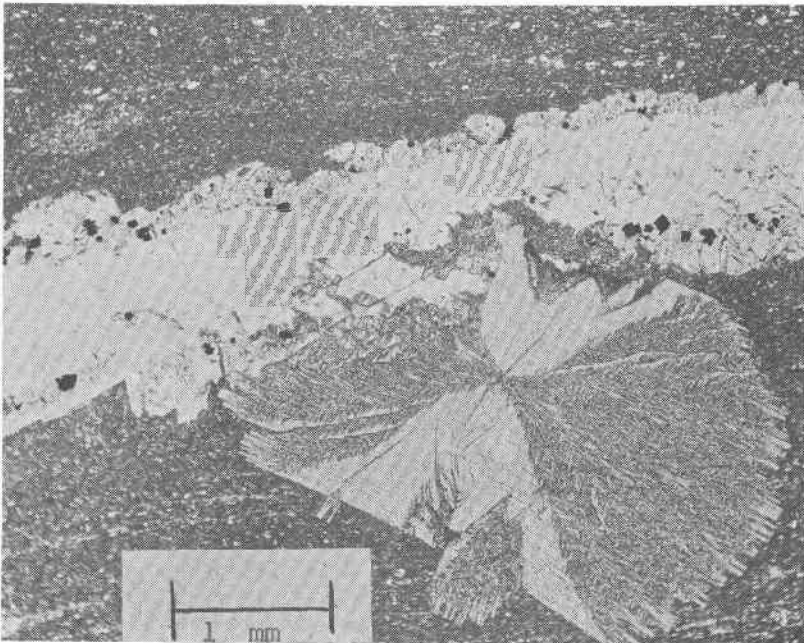


FIG. 3. Leucosphenite in thin section with shortite in dolomitic marlstone, Uinta Basin, Duchesne County, Utah. Note oriented matrix inclusions and terminations "feathering" out into the matrix.

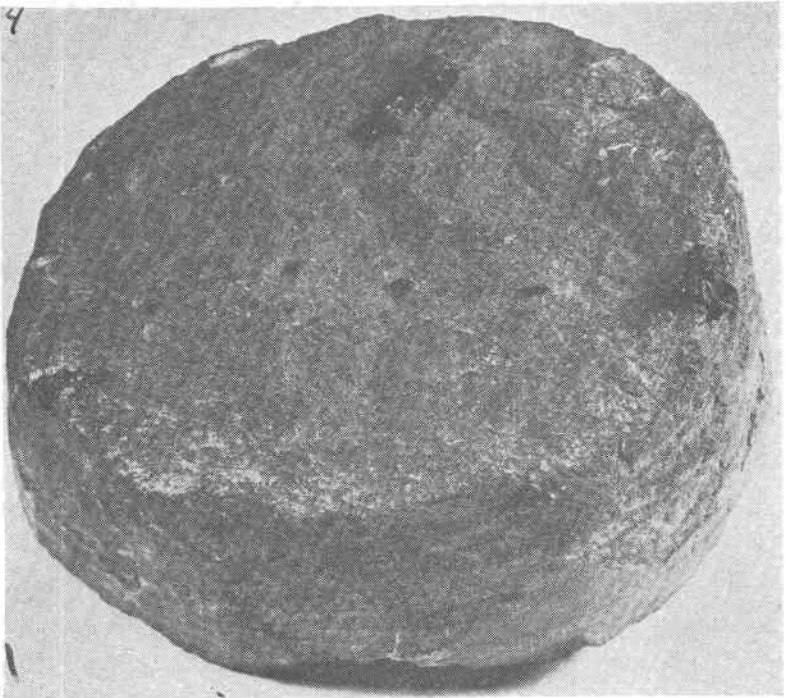


FIG. 4. Section of 4-inch drill core from O. L. Johnson #1 well, 4853 ft, Uinta Basin, Utah, showing large crystals of leucosphenite in dolomitic marlstone.

X-ray pattern. The specific gravity of this rock is 2.55(5) (Berman balance).

The matrix of leucosphenite from the Continental Duchesne No. 1 well was examined by means of a thin section of several pieces of cuttings, in immersion mounts, and by X-ray diffraction. Fine-grained calcite in rhombs and albite in crystals up to 0.1 mm, embedded in very fine dirty matrix, appear to be authigenic. Quartz in rough grains, mostly under 0.05 mm, looks detrital. Small, high index crystals are probably magnesioriebeckite and acmite. The magnesioriebeckite occurs with acmite in small clusters or sheaves of bluish needles attached to the surfaces of cuttings, apparently grown in cracks. Very thin, isolated, colorless, hexagonal flakes of detrital mica are far less abundant than the riebeckite. The dark, brownish, structureless groundmass is optically isotropic. A typical dark chip of the cuttings heated in a closed tube quickly turns black and in immersion crushed bits show black material in place of the brown groundmass.

The X-ray photograph of a bit of matrix detached from a crystal corresponding to the analyzed sample D in Table 5 shows the complete patterns of quartz and albite plus a few weak unidentified lines. In immersion this matrix is seen to be largely structureless dark brownish isotropic material of index near 1.54 in which are embedded grains of quartz and albite, the latter recognized by Professor R. L. Hay as authigenic.

Summarizing, the matrix in immediate contact with leucosphenite crystals in the Green River Formation in Utah varies markedly in mineral composition but is invariably fine grained. Authigenic albite is present in six of the eight samples of matrix tested and may be dominant. Quartz, probably mostly detrital, is a principal mineral in only two samples. Dolomite and calcite may occur together in the matrix and, judging from statements in the literature, dolomitic marlstone is the most common matrix rock of leucosphenite in the Green River Formation in the Uintah Basin. Shortite, nahcolite, redmergerite, searlesite, acmite, riebeckite, and pyrite are other minerals associated with leucosphenite in Utah.

#### *Wyoming.*

Excellent material can be obtained from the waste from the trona milling process. All of the plants in Sweetwater County except one



FIG. 5. Section of 2½-inch drill core from the Green River Formation, Wyoming, showing vein of shortite and loughlinite and two rosettes of leucosphenite enclosed in oil shale. Larger rosette (near arrow) is 1 cm in diameter.

roast the ore, which may affect the minerals in the residues. The Westvaco plant does not roast the ore; residues from it are the source of most of the Wyoming leucosphenite used in this study (see also Mrose *et al.*, 1961, p. 421).

Leucosphenite is listed by Bradley and Eugster (1969, p. 10) among "authigenic minerals found in the saline zone of the Wilkins Peak member of the Green River Formation in Wyoming." Milton *et al.* (1965) describe the occurrence of the mineral mckelveyite<sup>1</sup> as "associated with labuntsovite, leucosphenite, searlesite, and other typical minerals of the Green River Formation." Elsewhere (Milton *et al.*, 1960b, p. 178; Milton and Fahey, 1960, p. 162) leucosphenite is referred to as occurring "as rosettes, aggregates of thin plates, in close association with the trona beds" or "relatively abundant in microscopic crystals, with trona."

The largest rosette of leucosphenite in the drill core shown in Figure 5 is about 1 cm in diameter. Individual crystals within each rosette are no more than a few millimeters in maximum dimension and a few tenths of a millimeter thick. At the top of the core, not discernible in the picture, a few crystals belonging to a rosette almost completely embedded in the matrix protrude from the surface. It may be that the free rosettes from the Rogers well, one of which is shown in Figure 7D, correspond to those seen in the core.

The white veins in the core shown in Figure 5 consist of loughlinite (Fahey *et al.*, 1960) in chrysotile-like fibers up to 5 mm long and of coarsely granular shortite. Though both minerals may be present in one vein they tend to be separated. Shortite also occurs as isolated crystals or nests of crystals apart from the veins. There is some pyrite in the veins, up to 0.5 mm in diameter, a few perfect cubes being attached to loughlinite. Some much finer pyrite, 0.1 mm or less, is scattered through the brownish matrix. A chip of this matrix yielded without grinding, a powder pattern of muscovite and microcline with no trace of albite, quartz, calcite, or dolomite. The "kerogen" matrix is soft and wax-like, is unaffected by HCl, and is not readily soluble in most organic solvents. Heated gently in a closed tube, the chips are instantly blackened, but only on the surface. The index of refraction of the waxy substance is *ca.* 1.545. The density of matrix chips is 1.94(2) (Berman balance).

The wax-like material probably is similar to that described by Fahey (1962, p. 17-18) from the core of the John Hay Jr. well #1. He

<sup>1</sup> Recently "mckelveyite" from Westvaco's John Hay Trona mine was found to be in part a new polymorph, ewaldite (Donnay *et al.*, 1971).



reported the density of the mixed material to be 1.898, and the index of refraction of the organic material as 1.60. He calculated the density of the organic material to be 1.01.

### MORPHOLOGY AND SYMMETRY

Flink (1901) presented six crystal drawings of leucosphenite in his initial description, all corresponding to the symmetry of the monoclinic prismatic class, but made no statement as to the symmetry. All of Flink's drawings show crystals elongated parallel to  $a$  and one of these has been redrawn for Figure 6. Yefimov and Katayeva (1959) presented a single drawing of leucosphenite of equidimensional habit from Yakutia, here redrawn for Figure 6. This also looks monoclinic

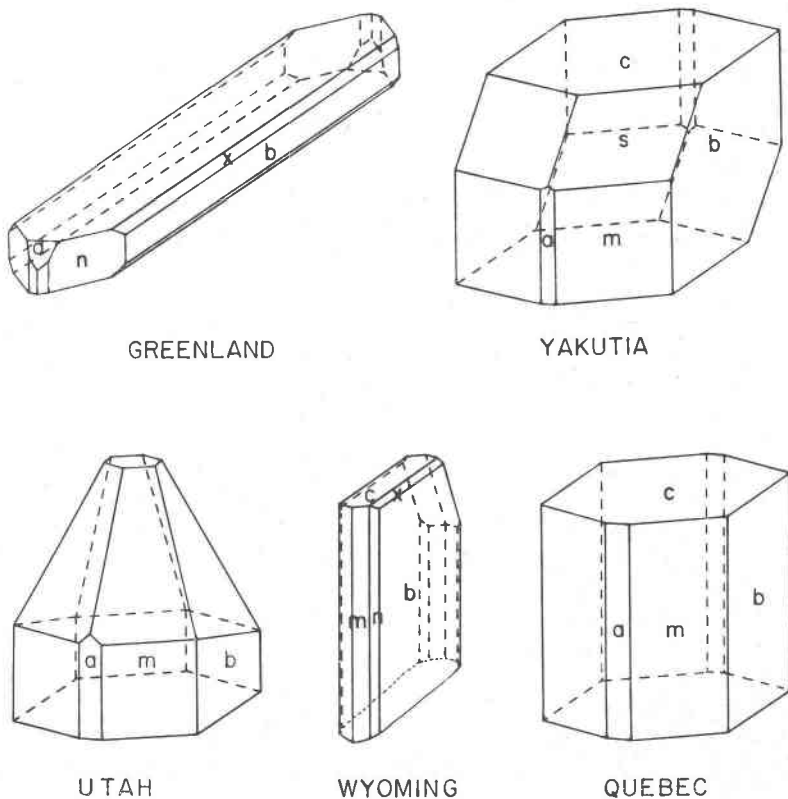


FIG. 6. Typical habits of leucosphenite from various sources. Indices are given in Tables 1 and 2. Unlabelled forms on Utah crystal are  $\{221\}$ ,  $\{\bar{2}21\}$ , and  $\{\bar{1}\cdot 24\cdot 4\}$ ; see comments in text. Axonometric drawings (Tunell, 1942) based on a common gnomonic projection. Angular coordinates of picture plane  $\varphi 70^\circ$ ,  $\rho 80^\circ$ .

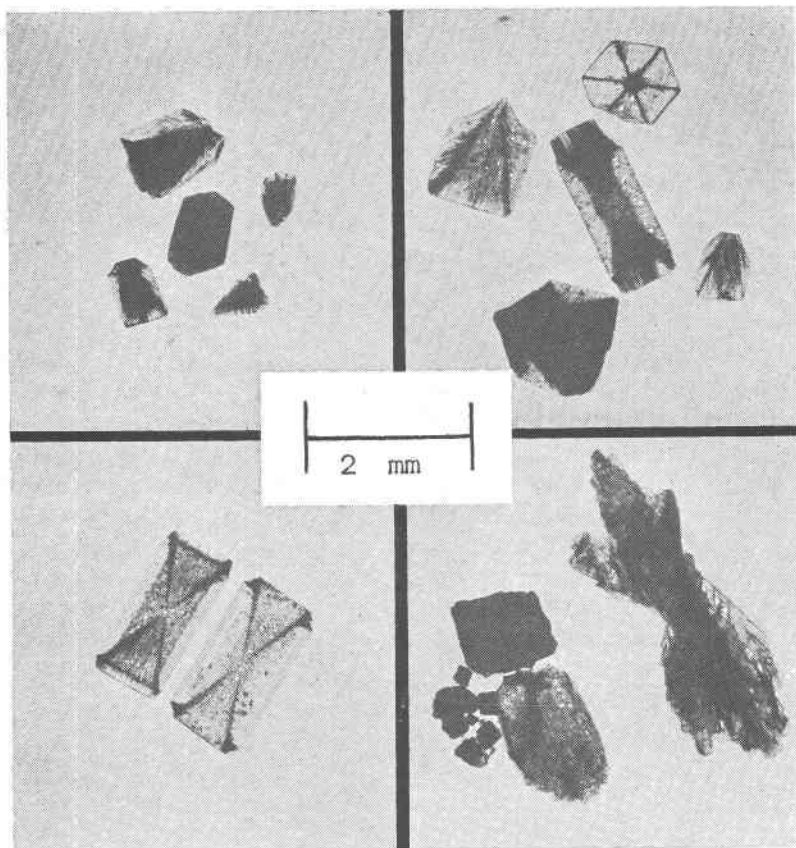


FIG. 7. Leucosphenite from the Green River Formation. Crystals immersed in liquid with  $n = 1.618$ .

- A. (upper left) Crystals from Uinta Basin, Utah, the 3 larger ones from Continental-Duchesne #1, 2711 ft. Crystal in center viewed approximately parallel to  $c$ .
- B. (upper right) Crystals from Utah, selected from lot from which material was taken for analysis.
- C. (lower left) Crystals from Uinta Basin showing hourglass structure.
- D. (lower right) Left, broken sheaf, Wyoming. Right, typical sheaf (rosette) of crystals from Rogers well, Sweetwater County, Wyoming.

prismatic, but the authors likewise made no statement about symmetry. Determination of the structure of leucosphenite in the space group  $Cm$  by Shumyatskaya *et al.* (1968) established the monoclinic domatic symmetry. (However, see postscript.)

Five crystals from Mont St. Hilaire, ten from Sweetwater County, Wyoming, and about fifty from various localities in the Uinta Basin,

Utah, were measured by 2-circle reflecting goniometer for this study. In the lower part of Figure 6 typical habits of leucosphenite from each of these sources are shown. The material from St. Hilaire has a simple habit with few forms and no indication of symmetry lower than  $2/m$ . However, the material available was scanty and examination of more material may reveal more forms or other habits. Crystals from Wyoming are always broken and show terminations at only one end of  $c$ . However, they all have one feature in common;  $\{010\}$  is invariably the dominant form and the crystals are distinctly lath-like or platy. Crystals of leucosphenite from the Uinta Basin, Utah, show considerable diversity of habit, mostly rather different from the Greenland, Yakutia, and Wyoming habits. This variety of habits, shown in Figures 7A, 7B, and 7C, may be connected with the fact that the crystals are from many sources within the Basin, but even the crystals from a single core at a particular horizon, as those from the Continental Duchesne #1 core, at 2711 feet, are by no means uniform in habit.

The crystals shown in Figures 7A-C indicate the range in habit. All but two are shown with the direction of view parallel to  $b$ . Two or three of the crystals appear to have the symmetry  $2/m$  or even seem to be orthorhombic due to the close approach of  $\beta$  to  $90^\circ$ . Several crystals in Figures 7A and 7B show polar habit, being terminated by a large pedion, arbitrarily designated  $\{00\bar{1}\}$ , at one end and a combination of various domes,  $\{hkl\}$ ,  $\{\bar{h}kl\}$ , and  $\{0kl\}$ , with sometimes a small pedion,  $\{001\}$ , at the other end. The domes, other than  $\{hk0\}$  and  $\{\bar{h}k0\}$  are usually striated and irregular and mostly cannot be identified with certainty. However, they include many forms not previously reported for leucosphenite and not listed in Tables 1 and 2. Frequently steeper forms lie "above" those less steep giving rise to concave profiles seen on several of the pictured crystals. Generally the faces at the sides above  $\{010\}$  are not  $\{0kl\}$  faces but  $\{\bar{h}kl\}$  close to the  $[100]$  zone, *e.g.*,  $(1\cdot24\cdot4)$  close to  $(061)$ , as shown in Figure 6. Besides the small pedion faces at the tip, some crystals have a narrow step of  $\{001\}$  between the faces of the  $[001]$  zone and the inclined faces. This can be seen on the crystal at lower left in Figure 7A.

Whereas most leucosphenite crystals from Mont St. Hilaire are perfectly clear and those from Wyoming have few inclusions, all but the smallest crystals from the Uinta Basin enclose a great deal of organic matter, generally in sectors or sheets clearly related to the crystal growth, while other parts of the same crystal may be quite clear. Some typical patterns of inclusions are shown in Figures 7A-C. Crystals of polar habit commonly show a cone of inclusions spreading downward

from the small pedion at the tip, seen in several of the pictured crystals. This suggests growth downward from the tip. In other cases the inclusions may be related to edges or reentrant angles on the crystals. In the crystal with hexagonal outline (viewed along  $c$ ) at the upper right of Figure 7B, the inclusions are associated with  $[001]$  edges between faces of the forms  $\{110\}$ ,  $\{\bar{1}10\}$ , and  $\{010\}$ . In the case of the central crystal in this figure and, especially, the two crystals in Figure 7C, inclusions are connected with  $(001)$  and  $(00\bar{1})$  steps at both ends. One of these crystals has such steps all around, the other only adjacent to faces of the  $\{110\}$  and  $\{\bar{1}10\}$  forms. The photograph shows that the inclusions are more copious in the crystal with steps all around. Viewed along  $a$  these crystals show profiles similar to those in the views along  $b$  pictured in Figure 7C but with a more acute angle between the sheets of included matter.

Flink (1901, p. 142) described crystals of leucosphenite twinned on  $(001)$ ; he included a drawing of one such crystal with the composition plane  $(001)$ . No crystals comparable to the twin described by Flink have been encountered. However, there is a possibility that  $(001)$  twins exist among the Uinta Basin leucosphenites. On several of these crystals, it was found by 2-circle goniometer that faces on the back were out of alignment to about the extent to be expected in the case of  $(001)$  twinning with the composition surface irrational and normal to both  $(001)$  and  $(010)$ . However, accurate measurements were precluded by the roughness of the crystals. No optical or X-ray indications of twinning were observed.

Table 1 shows the forms reported or found on leucosphenite crystals from five regions. No distinction is made between  $\{hk0\}$  and  $\{\bar{h}k0\}$  forms, nor between  $\{001\}$  and  $\{00\bar{1}\}$  or  $\{0kl\}$  and  $\{0k\bar{l}\}$  as only some of the crystals from the Uinta Basin would allow this distinction. Even on these crystals  $\{110\}$  and  $\{\bar{1}10\}$  are usually equally developed and can be distinguished only if found in suitable combination with other forms. Flink emphasized the predominance of  $\{130\}$  over  $\{110\}$ . Though a few faces of  $\{130\}$  were recorded they are never conspicuous, whereas all faces of  $\{110\}$  and  $\{\bar{1}10\}$  were recorded on all of the better crystals from Wyoming, Utah, and Quebec.

None of the crystals measured was of such quality as to warrant the calculation of linear elements. Accordingly the elements given in Table 2 are based on cell dimensions determined for Utah leucosphenite. Linear elements given by Flink (1901, p. 138),  $a:b:c = 0.5813:1:0.8501$ ,  $\beta = 93^\circ 23'$ , and by Yefimov and Katayeava (1959, p. 896),  $a:b:c = 0.5783:1:0.8499$ ,  $\beta = 93^\circ 22'$ , correspond to a unit distance on  $c$  doubled relative to that chosen from the structure cell.

The indices given by these authors can be transformed to those in Table 2 by the matrix  $100/010/00\frac{1}{2}$ .

## PHYSICAL PROPERTIES

Flink (1901, p. 144) reported the hardness of leucosphenite to be 6.5, whereas Yefimov and Katayeva (1959, Table 2) reported 6.1

Table 1. Forms recorded on leucosphenite crystals from different regions.

Forms	Greenland	Yakutia	Utah *	Wyoming *	Quebec *
<u>a</u> 100	x	x	x	x	x
<u>b</u> 010	X	X	X	X	X
<u>c</u> 001	X	X	X **	x	X
<u>x</u> 021	x	x		x	
<u>d</u> 201	x		<u>x</u>		
<u>m</u> 110	x	X	X	x	X
<u>n</u> 130	X	x	<u>x</u>	<u>x</u>	
<u>s</u> 111	x	X			
$\bar{1}11$			x		
<u>p</u> $\bar{2}21$	x		x	x	
221			x		
<u>q</u> 263	<u>x</u>				
<u>r</u> $4 \cdot 12 \cdot 3$	<u>x</u>				
<u>hkl</u>			X #		
<u>Ok1</u>			X ##		

Letters are those assigned by Flink (1901) but indices are based on structure cell. See text.

\* Only forms surely identified on two or more crystals are listed.

X major form; x minor form; x rare form.

\*\* On many crystals from Utah the pedions  $\{001\}$  and  $\{00\bar{1}\}$  were recognized as distinct forms. No such distinction had been made by other observers, nor was it possible on crystals from Wyoming or Quebec. See text.

# Crystals from most of the Utah samples show  $\{hkl\}$  and  $\{\bar{h}kl\}$  forms other than those for which indices are listed. They are generally "steeper" forms not indexed with certainty and in many cases are dominant forms. See photographs in figs. 7A-B and text.

## Crystals from Utah generally show "steep"  $\{Ok1\}$  forms with indices not surely determinable but in the range ca. 031 to 091, or  $\{\bar{h}kl\}$  forms close to these represented schematically in the drawing in fig. 6 by  $\bar{1} \cdot 24 \cdot 4$ .

and Chao and Watkinson (1972) 6-6.5. Flink reported only {010} cleavage, but Yefimov and Katayeva noted "important" {010} and {001} cleavage. In connection with the goniometric measurements an attempt was made to distinguish cleavage surfaces. In agreement with the Russian authors, (001) as well as (010) were repeatedly recorded as cleavage surfaces on crystals from various sources and no other cleavage was seen. The {001} cleavage was most frequently noted on the lath-like crystals from Wyoming.

The specific gravity reported by Flink for Greenland leucosphenite was 3.05, whereas Yefimov and Katayeva reported 3.089 for Yakutia leucosphenite, close to 3.103, the value calculated from its cell dimensions and the ideal composition. The specific gravity of leucosphenite from the Westvaco Trona mine, Wyoming, was reported by Mary Mrose in 1958-9 to be 3.050(5). Due to the small size of most crystals and the prevalence of inclusions in larger crystals, Berman balance determinations were feasible only on groups of small crystals, using the basket technique. Such a determination on ten selected clean

Table 2. Leucosphenite angle table. Monoclinic; domatic --  $\underline{m}$ .

		$\underline{a}:\underline{b}:\underline{c} = 0.5818:1:0.4281$		$\beta$ 93° 23'	$\underline{p}_0:\underline{q}_0:\underline{r}_0 = 0.7358:0.4274:1$		
		$\underline{x}_2:\underline{p}_2:\underline{q}_2 = 2.3400:1.7216:1$		86° 37'	$\underline{p}_0'$ 0.7371,	$\underline{q}_0'$ 0.4281,	$\underline{x}_0'$ 0.0591
Forms	$\phi$	$\rho$	$\phi_2$	$\rho_2 = B$	C	A	
$\underline{c}$ 001	00° 00'	3° 23'	86° 37'	90° 00'	.. ..	86° 37'	
$\underline{b}$ 010	90 00	90 00	.. ..	0 00	90 00	90 00	
$\underline{a}$ 100	90 00	90 00	0 00	90 00	86 37	.. ..	
$\underline{n}$ 130	29 51	90 00	0 00	29 51	88 19	60 09	
$\underline{m}$ 110	59 51	90 00	0 00	59 51	87 04½	30 09	
$\underline{x}$ 021	3 57	40 38	86 37	49 29	40 31	87 26	
$\underline{d}$ 201	90 00	56 53	33 07	90 00	53 30	33 07	
$\underline{s}$ 111	61 44	42 07	51 28½	71 30	39 07	53 47	
$\bar{1}11$	57 44	38 43	128 08	70 30	41 40	131 56	
$\underline{p}$ $\bar{2}21$	-58 49	58 50	144 45	63 42	61 43	137 04	
221	60 49	60 20	35 7	64 56	57 24	40 40	
$\underline{q}$ 263	32 44	45 30	61 10	53 08	43 45	67 18	
$\underline{r}$ 4·12·3	31 28	63 20	43 50	40 20	61 45	62 11	

Elements based on cell dimensions determined for leucosphenite from Utah,  $\underline{a}$  9.783,  $\underline{b}$  16.814,  $\underline{c}$  7.198 Å,  $\beta$  93° 23'.

crystals of aggregate weight 5.17 mg yielded the value 3.04(4). A slightly better result, 3.06(2), was obtained from several determinations by a variant of the suspension method described by Bloss (1961, p. 63-4).

#### OPTICAL PROPERTIES

Flink (1901, p. 142) reported that leucosphenite is "white inclining to grayish blue" and Yefimov and Katayeava (1959) say that it "is light-blue in color, or, rarely, colorless; sometimes, in large masses, the color changes to a dirty light-blue." Small crystals from Quebec, Wyoming, and Utah are all colorless. Larger crystals from Utah may be brown to black due to abundant inclusions but even these generally have clean colorless parts.

Although Flink (1901, p. 142-144) determined the optical properties of leucosphenite with great care, using the prism method and reporting indices to the fourth decimal place for "red," "yellow," and "green" light, there is an inconsistency in his results noted by Larsen (1921, p. 100). Larsen redetermined the optical properties and from his report that "X is parallel to the elongation" it is clear that the Greenland crystals which he examined must have been elongated parallel to *a* as described by Flink and here pictured in Figure 6, though the tables (Larsen 1921, p. 221 and Larsen and Berman, 1934, p. 210) contain the notation "Mon. elong *c*."

In the present study all optical determinations were made on single crystals or fragments using an apparatus similar to that described by Jones (1960). Indices were determined at two wavelengths, using interference filters and Cargille liquids checked by an Abbé refractometer reading directly to 0.0005, always corrected for temperature variation. Optic angle measurements were made directly on crystals mounted to rotate on *Y* and read to  $\pm\frac{1}{2}^\circ$ .

Optical properties of Utah leucosphenite corresponding to the analyzed sample are:  $\alpha$  1.644 (1.655),  $\beta$  1.6595 (1.6705),  $\gamma$  1.687 (1.701),  $2V\gamma$  75°,  $Z = b$ ,  $Y_{Ac} = 0^\circ$ ,  $r > v$ . Indices given first are for Na light, those in brackets for wavelength 486 nm. The range of variation of  $\beta$  for Green River leucosphenites is only 0.002.  $2V$  varies only a few degrees, probably within the limits of error. The total range of variation of  $\beta$  from the lowest (1.657), Quebec, to the highest, Yakutia, is only 0.007. All observers agree on the sense of dispersion of the optic axes; only Chao and Watkinson (1972) called it "crossed". Reported values of  $Y_{Ac}$  vary from  $-3^\circ 21'$  (Greenland) to  $+9^\circ$  (Quebec). Utah leucosphenites have an optical orientation similar to that reported by Flink and but little variation within each crystal. The relatively large crystals shown

in Figure 7C are exceptional in having uniform extinction parallel to  $c$ . The Wyoming leucosphenites, on the other hand, commonly show variable orientation within one crystal by as much as  $5^\circ$  or more.

#### SPACE GROUP AND CELL DIMENSIONS

The structure found by Shumyatskaya *et al.* (1968) is in the space group  $Cm$ . No statements as to the space group or the diffraction symbol for leucosphenite had previously been published. In the course of the present work, single crystal X-ray observations were made on 3 crystals from Mont St. Hilaire, 5 from the Westvaco mine, Wyoming, and 9 from several localities in Utah. No systematic absences other than those required by a  $C$  lattice were found.

Cell dimensions reported by previous observers, given in Table 3A, show little variation. In the present work all cell dimensions (Table 3B) have been determined from doubly quartz-calibrated Weissenberg patterns (Pabst, 1957) made with Cu radiation. Determinations were made only from high-angle reflections permitting independent measurements of the  $\alpha_1$  and  $\alpha_2$  components, 1.54051 and 1.54433 Å. The two or three constants obtainable from each film were calculated by least squares from the best high-angle reflections, up to six being used in each case if available. The results were tested by calculation of the standard error for each film. Precession patterns were used for a check on  $\beta$  by direct measurement, but many trials showed that the precision of lattice constants obtained by the method described far exceeds that obtainable from precession patterns. For convenient comparison standard errors were calculated in terms of percentage. For the films used in the final results this varied from about 0.06 to about 0.006 percent.

#### POWDER PATTERN

An unindexed X-ray powder diffraction pattern of leucosphenite from Narsarsuk was presented graphically by Neumann *et al.* (1957) and unindexed patterns of both Greenland and Yakutia leucosphenite were tabulated by Yefimov and Katayeava (1959); Chao *et al.* (1967, p. 122) also tabulated the spacings and intensities of the six strongest lines for leucosphenite from Mont St. Hilaire. The graphical pattern and the Yakutia pattern have been incorporated into the ASTM file on cards 18-712 and 15-13 respectively, "indexed by LGB." Lines not attributable to leucosphenite appear on both patterns and the indexing, though skillfully done, involves indices forbidden by  $C$ -centering.

Powder patterns of Utah leucosphenite are in complete agreement



Table 3. Cell dimensions of leucosphenites

A. Dimensions reported by other observers.						
Locality		$\underline{a}$	$\underline{b}$	$\underline{c}$	$\beta$	volume
Narsarsuk, Greenland	Strunz (1957, p. 285)	9.76	16.69	7.10	93° 23'	1154.54 Å <sup>3</sup>
Yakutia, U.S.S.R.	Shumyatskaya <u>et</u> <u>al.</u> , (1971) ##	9.799	16.840	7.199	93 22	1185.89
Mont St. Hilaire, Quebec	Chao & Watkinson (1972, Table 2)	9.781Å <sup>o</sup>	16.854	7.208	93 16(1)	1186.31
-----						
B. Dimensions determined in this study.						
Trona mine, Wyoming		9.788Å	16.826	7.198	93° 27'	1183.31
Uinta Basin, Utah	crystal used in early work, 1953	9.800	16.826	7.191	93 20	1183.75
" "	crystal from lot shown in fig. 7AB	9.783	16.814	7.198	93 23	1181.95
" "	crystal with those shown in fig. 7C	9.796	16.843	7.199	93 23	1185.73
" "	Continental Duchesne # 1 well	9.789	16.818	7.191	93 23	1181.80

## cited in postscript.

with the tabulation of an indexed powder pattern of the St. Hilaire leucosphenite by Chao and Watkinson (1972, Table 1). A powder pattern of leucosphenite from the Westvaco Trona mine<sup>1</sup>), Sweetwater County, Wyoming, USGS film 13525, kindly provided by Mary Mrose, matches exactly the films made from Utah leucosphenite.

#### COMPOSITION AND FORMULA

Table 4 shows the hypothetical cell content of leucosphenite corresponding to the formulas proposed by various workers based on the assumption of two Ba per cell. The Flink formula was based on a faulty analysis in which B<sub>2</sub>O<sub>3</sub> was not recognized and substantial ZrO<sub>2</sub> (perhaps from admixed elpidite) was reported. This was corrected in the next formula, but it too was based on a questionable analysis showing CaO, possibly from carbonate admixture. Strunz' formula probably is based on the preliminary report by Milton *et al.*

<sup>1</sup>The John Hay well, referred to above, and the Trona mine, are both parts of the Intermountain Chemical Co. (a subsidiary of the Food Machinery Corporation) trona mine at Westvaco, near Green River, Wyoming.

(1953). The formula proposed by the Russian authors has been confirmed by the structure determination by Shumyatskaya *et al.* (1968). Flink's formula conforms to this by simply adding  $B_2O_3$ .

Recent and new analyses of leucosphenite are assembled in Table 5 together with cell contents calculated on the basis of the reported specific gravities and the cell dimensions given in Table 3. The results confirm the formula assigned by Yefimov and Katayeva (1959). Taking into account the spectrographic results for the Green River leucosphenites, it is clear that there is but little variation in the minor substituents except for the fact that potassium seems to be less abundant ( $< 0.2$  percent K) in leucosphenite formed in the highly sodic lacustrine environment of the Green River Eocene.

#### HEAT TREATMENT OF LEUCOSPHENITE

Yefimov and Katayeva (1959) reported an endothermic effect at  $930^\circ C$  on a heating curve of leucosphenite, which they interpreted as corresponding to the melting point.

The effect of heating leucosphenite in air was tested on several small clean fragments of Uinta Basin material. Each fragment was examined optically in immersion before heat treatment and optically as well as by X-ray diffraction after each stage of the treatment. Heating was carried out in small, loosely covered, Pt crucibles in two series in several stages covering the temperature range from  $540^\circ$  to  $860^\circ C$  calibrated by the melting point of pure NaCl,  $801^\circ C$ . In this way it was established that leucosphenite melts incongruently at about  $800^\circ C$  with the formation of  $Ba_2TiO(Si_2O_7)$  (fresnoite), the colorless, rectangular crystals of which are readily recognizable in the glass of

Table 4. Formulas proposed for leucosphenite and corresponding ideal cell contents

Flink (1901)	Milton <i>et al.</i> (1953)	Strunz (1957)	Yefimov & Katayeva (1959)	
$Na_4BaTi_2(Si_{10}O_{27})$	$CaBaNa_3BTi_3Si_9O_{29}^*$	$Ba(Na,Ca)_4Ti_3(BO_3 Si_8O_{24})$	$Na_4BaTi_2B_2(SiO_3)_{10}$	
Cell content corresponding to two Ba or two formula units in each case.				
O	54	58	54	60
Si	20	18	16	20
Ti	4	6	6	4
B	-	2	2	4
Ba	2	2	2	2
Na	8	(Na+Ca) 8	(Na+Ca) 8	8

\* This formula was also used by Milton & Eugster (1959) and by Milton *et al.* (1960); however, in the meantime Milton (1957, p. 138) had used yet another formula.

Table 5. Chemical analyses and cell contents of leucosphenite.

	A Yakutia	B Quebec	C Wyoming	D Utah	
SiO <sub>2</sub>	54.3	53.66	55.9	55.0	
TiO <sub>2</sub>	13.92	14.52	15.5	15.2	
ZrO <sub>2</sub>	not found	not found			
Nb <sub>2</sub> O <sub>5</sub>	0.1	0.77			
Al <sub>2</sub> O <sub>3</sub>	not found	0.11			
Fe <sub>2</sub> O <sub>3</sub>	0.28	0.04 *			
B <sub>2</sub> O <sub>3</sub>	6.36	6.6	6.7	6.7	
MgO	0.13	not found			
MnO	trace	not found			
CaO	not found	0.06	not reported #	0.1 ###	
SrO	0.03	---			
BaO	13.00	11.98	12.7	13.0	
Na <sub>2</sub> O	10.70	11.61	10.7	10.0	
K <sub>2</sub> O	0.79	0.51	#	##	
H <sub>2</sub> O	not found	---			
sum	99.63	99.86	101.5	99.9	
sp. gr.	3.089	3.065	3.05	3.06	
Si	20,003	19.57	20.24	19.98	Ideal 20
Ti	3.85	3.98 (Nb 0.13)	4.16	4.15	4
Fe	0.08	0.01			
B	4.037	4.15	4.12	4.19	4
Mg	0.08				
Ba	1.88	1.71 (Ca 0.02)	1.77	1.85	2
Na	7.64	8.21	7.36	7.04	8
K	0.37	0.24			
O	59.90	59.80	59.32	59.86	60

- A. Yefimov & Katayeva (1959), analyst, Z. T. Katayeva.
- B. Chao & Watkinson (1972); Electron microprobe analysis by D.H. Watkinson; B<sub>2</sub>O<sub>3</sub> by B. Ingram, U.S.G.S.; \* total Fe as FeO.
- C. Electron microprobe analysis by E.J. Dwornik and H.J. Rose, U.S.G.S., Washington; B<sub>2</sub>O<sub>3</sub>, chemical determination by Blanche Ingram, U.S.G.S., Washington. Semiquantitative spectrographic analysis by W. B. Crandell, U.S.G.S., showed:- Al 0.015%, Fe .15, Mg .03, Mn .0002, Nb .03, Sr .001, Zr .07; K not detected (<0.2%); # Ca 0.07%.
- D. Electron microprobe analysis by E.J. Dwornik and H.J. Rose, U.S.G.S., Washington; B<sub>2</sub>O<sub>3</sub>, chemical determination by Blanche Ingram, U.S.G.S., Washington. Semiquantitative spectrographic analysis by J. L. Harris, U.S.G.S., showed:- Fe 0.2%, Mg .03%, Mn 10 ppm, Nb 500 ppm, Sr 150 ppm, Zr 1000 ppm; K not detected (<0.2%); ## Ca .15%.

index close to 1.615 surrounding remnants of unchanged leucosphenite. Ba<sub>2</sub>TiO(Si<sub>2</sub>O<sub>7</sub>) was identified by its complete powder pattern<sup>1</sup> which additionally showed two lines attributable to cristobalite.

<sup>1</sup>A previously measured line was found to have been omitted from the published pattern of fresnoite in Alfors *et al.* (1965) and ASTM card 18-197. The data for the line are: *I*, 20; *d* (meas), 1.920; *d* (calc), 1.921; *hkl*, 411. Moreover, *I* for *d* (meas) 1.870 should be 25.

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## POSTSCRIPT

After completion of this work another Russian paper (Shumyatskaya, *et al.* 1971) came to our attention. The principal results of the preliminary report on the structure of leucosphenite (Shumyatskaya *et al.*, 1968) are confirmed, but the space group is now given as  $C2/m$ . This is chosen on the basis of a negative result of a test for piezoelectricity. Presence or absence of a center of symmetry is connected with ordering of B and Si. For a model with partial ordering ( $C2/m$ )  $R = 13.5$ ; for a model with complete ordering ( $Cm$ )  $R = 13.3$ .

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