# LEUCOSPHENITE FROM MONT ST. HILAIRE, QUEBEC

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## ABSTRACT

Leucosphenite occurs as small, colourless to pale blue, elongated tabular crystals and wedge-shaped crystals in the pegmatites and thermally metamorphosed inclusions in the nepheline syenite, Mont St. Hilaire, Quebec. It is mainly associated with microcline, albite, aegirine, calcite, and quartz.

Leucosphenite crystals from Mont St. Hilaire display a variety of habits commonly with {100}, {010}, {001}, {110}, and rarely with {130}. The space group is $C2/m$; $a = 9.781$, $b = 16.854$, $c = 7.208$Å, $\beta = 93°16'$. Density $= 3.065(5)$ g/cm³. Hardness $= 6$-$6.5$. Cleavages {010} and {001} are fair. It is optically biaxial (+); $\alpha = 1.643$, $\beta = 1.657$, $\gamma = 1.681$, $2V = 76°$, in sodium light. The optical plane is normal to (010) and $Y \wedge c$ varies with a maximum $+9°$. It has marked dispersion with $r > v$.

Electron microprobe analysis gave $SiO_2$ 53.66; $Al_2O_3$ 0.11; $Nb_2O_5$ 0.77; $TiO_2$ 14.52; BaO 11.98; CaO 0.06; total Fe as FeO 0.04; $Na_2O$ 11.61; $K_2O$ 0.51; MgO, MnO, $ZrO_2$ and $Y_2O_3$ not found; total $= 99.86\%$ including 6.6% $B_2O_3$ determined by wet chemistry. The analysis recalculates to $Na_{4.11}K_{0.12}Ba_{0.86}Ca_{0.01}Ti_{2.00}Nb_{0.06}B_{2.08}Al_{0.02}Si_{9.81}O_{30}$.

## INTRODUCTION

Leucosphenite was originally described by Flink (1901) as a white wedge-shaped mineral occurring in the pegmatite veins in the Narsarsuk region, Greenland. Flink gave $BaNa_4(Ti_2O_2)(Si_2O_5)_5$ as its ideal formula. Leucosphenite was later found as a widespread mineral in the Green River formation in Northeastern Utah (Milton *et al.* 1954) and Wyoming (Milton 1957). On the basis of chemical analysis of the Utah mineral and spectrographic analysis of the type Greenland crystals, Milton and others proposed a formula, $CaBaNa_6BTi_3Si_9O_{29}$, including boron and calcium as essential components. Yefimov & Katayeava (1959) reported the occurrence of leucosphenite in the pegmatite veins of the Inaglina ultrabasic-alkaline massif, South Yakutia, USSR; they did not find calcium in the Yakutia leucosphenite but confirmed boron as an essential component and assigned the mineral a new formula, $Na_4BaTi_2B_2(SiO_3)_{10}$. This formula was later verified by crystal structure analysis (Shumyatskaya *et al.* 1968) and was adopted by Strunz in the fifth edition of his *Mineralogische Tabellen* (1970). Chao *et al.* (1967) reported a new occurrence of leucosphenite at Mont St. Hilaire, Quebec; the description

of this rare mineral from Mont St. Hilaire now forms the core of this paper.

## Occurrence

The general geology of the Mont St. Hilaire area has been described in detail by Dresser & Denis (1944) and summarized by Chao *et al.* (1967). Leucosphenite was found in small amounts in the pegmatites and in the wall-rock inclusions in the nepheline syenite of the Mont St. Hilaire pluton. The inclusions of shale interbedded with thin layered limestone were metamorphosed to greenish grey or grey hornfels and to siliceous marble. Well-formed, tabular, pale blue leucosphenite crystals up to 1 mm
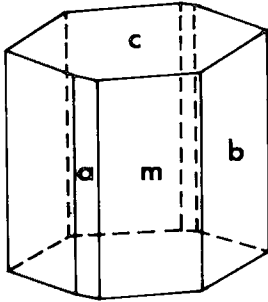


FIG. 1. Crystal drawing of leucosphenite from Mont St. Hilaire, Quebec, showing $a\{100\}$, $b\{010\}$, $c\{001\}$ and $m\{110\}$.
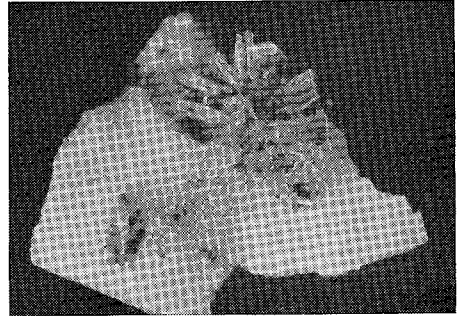


FIG. 2. Tabular leucosphenite crystals (1mm in length), elongated along $a$ and flattened on $\{010\}$.
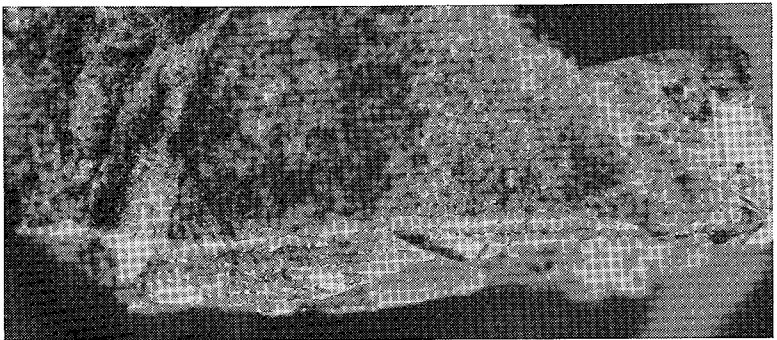


FIG. 3 (bottom). Wedge-shaped leucosphenite crystal bounded by $\{001\}$, $\{010\}$, $\{130\}$ and narrow $\{100\}$ (length of crystal = 3.5mm).

in length were found in pegmatite with microcline, albite, aegirine, sodalite and minor amounts of ramsayite, elpidite, polylithionite, ancylite, pyrrhotite and pyrite. An inclusion in the syenite contains leucosphenite as very small tabular crystals (average 0.2 mm) lining a small vug. The minerals surrounding the vug are microcline, albite, calcite, fibrous ramsayite, fine needles of aegirine and a hitherto undescribed mineral UK#6 (Chao *et al.* 1967). Leucosphenite was also found in the quartz veins in the hornfels as pseudohexagonal crystals up to 1 cm across, associated with lemon-yellow narsarsukite, purple fluorite, calcite, a recently described new mineral, carletonite (Chao 1971) and minor amounts of ancylite, aegirine, ramsayite, molybdenite and galena.

## CRYSTALLOGRAPHY

Leucosphenite crystals from Mont St. Hilaire display a variety of habits. Some crystals are elongated along *c*, bounded by {010}, {001} and {110}, in order of prominence (Fig. 1) ; {010} is sometimes the only prominent form and {100} is either absent or very small, resulting in a tabular habit. Some crystals are tabular but distinctly elongated along *a* with prominent {010} and small {001}, {110}, and {100} (Fig. 2). A few crystals are elongated along *a* but bounded by {010}, {001}, {100} and extremely well-developed {130}, giving rise to a wedge-shape habit (Fig. 3). For all crystals examined the forms {021}, and {221}, reported to be common on crystals from Greenland and USSR, are characteristically absent. The ($hk$0) faces on large crystals (up to 1 cm) from Mont St. Hilaire are often heavily striated along *c*. Small crystals, although not apparently striated, always gave multiple signals on the optical goniometer, rendering accurate measurements of the interfacial angles difficult.

Weissenberg and precession photographs of several single crystals were prepared with Cu$K\alpha$ and Mo$K\alpha$ radiations respectively. Systematic extinctions were found to be consistent with those required by space groups $C2$, $Cm$ and $C2/m$. Morphological studies of the Mount St. Hilaire leucosphenite revealed no symmetry lower than $2/m$. Crystal structure analysis by Shumyatskaya *et al.* (1968) indicated the space group to be $Cm$ but in a later refinement of the structure (Shumyatskaya *et al.* 1971) $C2/m$ was adopted as the correct space group.

The preliminary cell parameters obtained from the single crystal photographs were refined by a least-squares method (unpublished program by A. W. Hounslow & G. Y. Chao) using x-ray powder diffraction data

TABLE 1. X-RAY POWDER DIFFRACTION PATTERN OF LEUCOSPHENITE, MONT ST. HILAIRE (114.6 mm camera, CuKα radiation (λ = 1.5418Å), Si standard, visual intensities)

| $I_p$ | $d_{obs.}$ (Å) | $d_{calc.}$ (Å) | hkl | $I_w$ | $I_p$ | $d_{obs.}$ (Å) | $d_{calc.}$ (Å) | hkl | $I_w$ |
|---|---|---|---|---|---|---|---|---|---|
| 90 | 8.45 | 8.4495 | 110 | s | 40 | 2.813 | 2.8165 | 330 | s |
| | | 8.4271 | 020 | s | | | 2.8090 | 060 | s |
| | | 7.1966 | 001 | vw | | | 2.8086 | $22\bar{2}$ | m |
| | | 5.6171 | $\bar{1}11$ | vw | 40 | 2.732 | 2.7363 | $0\bar{4}2$ | vs |
| | | 5.4726 | 021 | vw | | | 2.6750 | 222 | m |
| | | 5.3501 | 111 | vw | | | 2.6678 | $\bar{3}31$ | w |
| 20 | 4.870 | 4.8826 | 200 | m | 10 | 2.616 | 2.6168 | 061 | ms |
| | | 4.8697 | 130 | s | 5 | 2.577 | 2.5799 | 331 | m |
| 100 | 4.222 | 4.2247 | 220 | vs | 10 | 2.460 | 2.4588 | $\bar{3}12$ | m |
| | | 4.2135 | 040 | s | | | 2.4413 | 400 | s |
| | | 4.1519 | $\bar{2}01$ | vw | 30 | 2.434 | 2.4348 | 260 | vs |
| 20 | 4.092 | 4.0874 | $\bar{1}31$ | ms | | | 2.4324 | $\bar{2}42$ | mw |
| 10 | 3.980 | 3.9809 | 131 | m | | | 2.4078 | $\bar{1}52$ | abs |
| 10 | 3.936 | 3.9376 | 201 | ms | | | 2.3989 | 003 | abs |
| 20 | 3.728 | 3.7244 | $\bar{2}21$ | s | | | 2.3638 | 152 | vw |
| | | 3.6361 | 041 | w | | | 2.3531 | $\bar{4}01$ | abs |
| | | 3.5983 | 002 | m | | | 2.3449 | 420 | w |
| 30 | 3.569 | 3.5674 | 221 | s | | | 2.3441 | 242 | w |
| 70 | 3.371 | 3.3710 | $\bar{1}12$ | vs | | | 2.3381 | $\bar{1}13$ | vw |
| 50 | 3.306 | 3.3093 | 022 | s | | | 2.3377 | 170 | ms |
| 30 | 3.250 | 3.2533 | 112 | s | 5 | 2.327 | 2.3266 | $\bar{2}61$ | w |
| | | 3.1960 | 310 | w | | | 2.3258 | 312 | m |
| 30 | 3.189 | 3.1899 | 240 | s | <5 | 2.306 | 2.3072 | 023 | m |
| | | 3.1863 | 150 | s | 40 | 2.280 | 2.2868 | 261 | vs |
| | | 2.9835 | $\bar{3}11$ | vw | | | 2.2783 | 113 | m |
| 30 | 2.982 | 2.9789 | $\bar{2}02$ | s | | | 2.2729 | $\bar{3}32$ | ms |
| | | 2.9574 | $\bar{2}41$ | s | | | 2.2729 | 401 | s |
| | | 2.9341 | $\bar{1}32$ | vw | | | 2.2664 | $\bar{4}21$ | w |
| 10 | 2.930 | 2.9338 | 151 | ms | 5 | 2.255 | 2.2540 | $\bar{3}51$ | m |
| 60 | 2.891 | 2.8937 | 151 | s | 5 | 2.231 | 2.2323 | $\bar{1}71$ | ms |
| | | 2.8769 | 241 | vs | 5 | 2.216 | 2.2145 | 171 | ms |
| 10 | 2.855 | 2.8622 | 311 | ms | | | 2.2142 | 062 | vw |
| | | 2.8554 | 132 | ms | 5 | 2.203 | 2.2033 | $\bar{2}03$ | vw |
| | | 2.8209 | 202 | s | | | 2.2003 | 351 | m |

$I_p = I$ powder; $I_w = I$ Weissenberg.

$vs$ = very strong, $s$ = strong, $ms$ = medium to strong, $m$ = medium, $wm$ = weak to medium, $w$ = weak, $vw$ = very weak; abs = absent.

(Table 1). The indexing of the powder pattern was based on comparison of the observed and calculated $d$-values, using single crystal photographs as a guide. The refined values and their estimated standard deviations are incorporated in Table 2.

The axial ratios given by Flink (1901), 0.5813 : 1 : 0.8591, and by Yefimov & Katayeava (1959), 0.5783 : 1 : 0.8499, correspond to a unit

TABLE 1. X-RAY POWDER DIFFRACTION PATTERN OF LEUCOSPHENITE, MONT ST. HILAIRE (114.6 mm camera, CuKα radiation ($\lambda = 1.5418$Å), Si standard, visual intensities)

| $I_p$ | $d_{obs.}$(Å) | $d_{calc.}$(Å) | $hkl$ | $I_w$ | $I_p$ | $d_{obs.}$(Å) | $d_{calc.}$(Å) | $hkl$ | $I_w$ |
|---|---|---|---|---|---|---|---|---|---|
| | | 2.1944 | 421 | w | 5 | 1.899 | 1.9004 | $\bar{5}11$ | m |
| 5 | 2.169 | 2.1766 | $\bar{1}33$ | m | | | 1.8996 | 531 | wm |
| | | 2.1666 | 332 | s | | | 1.8854 | $\bar{3}71$ | w |
| <5 | 2.130 | 2.1316 | $\bar{2}23$ | wm | <5 | 1.882 | 1.8839 | 243 | m |
| | | 2.1281 | 133 | wm | | | 1.8787 | $\bar{2}81$ | wm |
| | | 2.1124 | 440 | wm | | | 1.8724 | $\bar{3}33$ | w |
| | | 2.1068 | 080 | s | | | 1.8690 | 313 | m |
| 20 | 2.105 | 2.1061 | 203 | s | | | 1.8622 | $\bar{4}42$ | vw |
| | | 2.0847 | 043 | s | | | 1.8576 | 281 | m |
| <5 | 2.077 | 2.0759 | $\bar{4}02$ | m | <5 | 1.849 | 1.8536 | 371 | m |
| | | 2.0544 | $\bar{4}41$ | vw | | | 1.8471 | 511 | m |
| 10 | 2.045 | 2.0437 | $\bar{2}62$ | ms | 10 | 1.822 | | | |
| | | 2.0432 | 223 | vw | 20 | 1.799 | | | |
| 10 | 2.018 | 2.0219 | 081 | s | <5 | 1.784 | | | |
| | | 2.0157 | $\bar{4}22$ | m | <5 | 1.765 | | | |
| | | 2.0004 | $\bar{3}52$ | w | 20 | 1.735 | | | |
| | | 2.0004 | 441 | vw | 5 | 1.703 | | | |
| | | 1.9905 | 262 | w | 20 | 1.684 | | | |
| | | 1.9727 | $\bar{1}72$ | m | 10 | 1.664 | | | |
| 10 | 1.972 | 1.9723 | $\bar{3}13$ | m | 10 | 1.628 | | | |
| | | 1.9688 | 402 | ms | 5 | 1.610 | | | |
| <5 | 1.952 | 1.9525 | $\bar{2}43$ | m | 20 | 1.594 | | | |
| | | 1.9482 | 172 | vw | <5 | 1.580 | | | |
| | | 1.9401 | 510 | m | 5 | 1.562 | | | |
| | | 1.9357 | 370 | m | 10 | 1.544 | | | |
| 10 | 1.933 | 1.9344 | 280 | vw | ·10 | 1.527 | | | |
| | | 1.9338 | $\bar{1}53$ | m | 10 | 1.515 | | | |
| | | 1.9268 | 352 | vw | | many more lines | | | |
| | | 1.9172 | 422 | wm | | | | | |

distance on $c$ doubled relative to that chosen from the structural cell. The indices given by these authors can be transformed to those referred to in this paper by the matrix $100/010/00\frac{1}{2}$.

## PHYSICAL AND OPTICAL PROPERTIES

Leucosphenite from Mont St. Hilaire is colourless to very pale blue with a vitreous luster, transparent in small crystals and translucent in large ones. The average of three determinations of density at 23°C, using a Berman balance and toluene, gave $D = 3.065(5)$ g/cm³. Hardness on the Moh's scale is 6-6.5. Cleavage {010} and {001} are fair. Fracture is conchoidal.

The refractive indices were measured by immersion at 23°C on a flat stage with a sodium light source. Single crystals with known orientation

TABLE 2. PHYSICAL AND OPTICAL PROPERTIES OF LEUCOSPHENITE

|  | Mont St. Hilaire Quebec | Yakutia, USSR Yefimov & Katayeava (1959) | Narsarsuk, Greenland Flink (1901) [1] |
|---|---|---|---|
| $\alpha$ | 1.643 | 1.6485 | 1.6445 |
| $\beta$ | 1.657 | 1.6640 | 1.6609 |
| $\gamma$ | 1.681 | 1.6910 | 1.6878 |
| $\gamma - \alpha$ | 0.038 | 0.0425 | 0.0433 |
| $2V$ | 76° | 78° | 77°4′ (calc.) |
| Optical Sign | (+) | (+) | (+) |
| Dispersion | $r > v$ (crossed) | $r > v$ | $r > v$ |
| Orientation | $Z = b$ | $Z = b$ | $Z = b$ |
|  | $Y \wedge c = +9°$ (max.) | Parallel extinction | $Y \wedge c = -3°21′$ |
| Sign of Elongation | (±) | (−) | (−) |
| Hardness | 6-6.5 | 6.1 | 6.5 |
| Density (g/cm³) | 3.065(5) | 3.089 | 3.05 |
| $a$(Å) | 9.781(1) | 9.79 | 9.76 |
| $b$(Å) | 16.854(2) | 16.84 | 16.69 |
| $c$(Å) | 7.208(1) | 7.20 | 7.10 |
| $\beta$ | 93°16(1)′ | 93°22′ | 93°23′ |

[1] Refractive indices are for "yellow" light. Cell parameters from Strunz (1970, p. 431).

from $x$-ray diffraction or optical goniometry were used. All Cargille liquids used were checked by an Abbe refractometer. The uncertainty of the measurements was estimated to be less than ± 0.001. The optic angle was determined by Kamb's method (Kamb 1958) with a large uncertainty of ± 5°, due to the poor resolution of the isogyres. The results of this study are given in Table 2 where they are compared with properties of leucosphenite from Greenland and USSR. The refractive indices of the Mount St. Hilaire mineral are considerably lower than those of the Yakutia and Greenland minerals, particularly for β and γ. The extinction angle $Y \wedge c$ of the Mont St. Hilaire mineral is variable over a large range from crystal to crystal, and sometimes even within the same crystal.

### CHEMICAL ANALYSIS AND FORMULA

Leucosphenite was analyzed using the Cambridge MK 5 electron microprobe for the following elements using the standards shown in parentheses : Fe (metallic iron) ; Mg (synthetic phlogopite) ; Ca (calcite) ; Si, Al and K (microcline ); Zr (synthetic zirconia) ; Na (jadeite) ; Nb (metallic niobium) ; Y (synthetic yttrium iron garnet) ; Ba (synthetic barium ferrite) ; Ti (ilmenite) ; Mn (metallic manganese). Analyses were made at 15 KV accelerating voltage (20 KV for Y and Ba) and a sample current of about 50 nanoamps (about 30 for Na and Zr) using a finely focussed beam. Ten- or twenty-second counts were collected and the polished grain mount was moved about 10 microns before another of the approximately eight sets of counts was taken for core and margin of each of six grains. Ten-second counts for Na and Zr were collected while the specimen was translated at 10 μm per minute because the specimen current and counting rate for NaKα-radiation decreased slightly using a stationary spot.

The $x$-ray data were computer-corrected for generation (atomic number), absorption and fluorescence effects using the program written by Rucklidge (1967). The average analysis is given in Table 3 with $B_2O_3$ determined chemically by B. Ingram on a 200 mg hand-picked sample. The variation between core and margin and among grains was approximately equal to the standard deviation of counts for the series of about 8 counts at any one site. Thus zoning is not apparent from microprobe analysis. The analyses of the Yakutia and the Greenland leucosphenites are also listed in Table 3 although the latter is apparently in error in omission of $B_2O_3$ as was pointed out by Milton et al. (1954).

Formula of the Mont St. Hilaire leucosphenite calculated on the basis of 30 oxygen atoms per formula (Table 3) deviates only slightly from

TABLE 3. CHEMICAL ANALYSES OF LEUCOSPHENITE

| | 1 | 2 | 3 | 4 |
|---|---|---|---|---|
| $SiO_2$ | 53.66 | 54.3 | 56.94 | 54.25 |
| $Al_2O_3$ | 0.11 | not found | — | — |
| $B_2O_3$ | 6.6 | 6.36 | — | 6.29 |
| $Fe_2O_3$ | — | 0.28 | — | — |
| $Y_2O_3$ | not found | — | — | — |
| $Nb_2O_5$ | 0.77 | 0.1 | — | — |
| $TiO_2$ | 14.52 | 13.92 | 13.20 | 14.43 |
| $BaO$ | 11.98 | 13.00 | 13.75 | 13.84 |
| $CaO$ | 0.06 | not found | — | — |
| $SrO$ | — | 0.03 | — | — |
| $FeO$ | 0.04 | — | — | — |
| $MgO$ | not found | 0.15 | — | — |
| $MnO$ | not found | — | — | — |
| $Na_2O$ | 11.61 | 10.70 | 11.14 | 11.19 |
| $K_2O$ | 0.51 | 0.79 | 0.56 | — |
| $ZrO_2$ | not found | — | 3.50 | — |
| $H_2O$ | — | not found | 0.31 | — |
| Total | 99.86 | 99.53 | 99.40 | 100.00 |

*Atomic Ratios to 30 Oxygen Atoms*

| | | | | |
|---|---|---|---|---|
| Si | 9.81 | 10.03 | | 10.00 |
| Al | 0.02 | — | | — |
| B | 2.08 | 2.03 | | 2.00 |
| $Fe^{3+}$ | — | 0.04 | | — |
| Nb | 0.06 | 0.001 | | — |
| Ti | 2.00 | 1.93 | | 2.00 |
| Ba | 0.86 | 0.94 | | 1.00 |
| Ca | 0.01 | — | | — |
| Sr | — | 0.002 | | — |
| $Fe^{2+}$ | 0.005 | — | | — |
| Mg | — | 0.03 | | — |
| Na | 4.11 | 3.83 | | 4.00 |
| K | 0.12 | 0.18 | | — |
| $D_{calc.}$ | 3.078 | 3.092 | — | — |
| $D_{obs.}$ | 3.065(5) | 3.089 | 3.05 | — |

1. This study. Electron microprobe analysis by DHW; $B_2O_3$ by B. Ingram, U.S.G.S.; Total Fe as FeO.
2. Yefimov & Katayeava (1959).
3. Flink (1901).
4. $Na_4BaTi_2B_2Si_{10}O_{30}$.

the ideal formula $Na_4BaTi_2B_2Si_{10}O_{30}$ *. Density, calculated with the cell parameters obtained in this study and $Z = 2$, is 3.078 $g/cm^3$, in good agreement with the measured value 3.065(5).

The Mont St. Hilaire leucosphenite contains slightly lower Si and Ba but higher B and (Na, K) than does the ideal composition, indicating partial substitution of Si by B and Ba by (Na, K). The balance of charges is probably satisfied by coupled substitution of Ti and Nb.

The high $ZrO_2$ content (3.50%) in the analysis of the Greenland leucosphenite is probably due to contamination by elpidite which was reported by Flink (1901) as closely associated with the Greenland leucosphenite.

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* The structural formula given by Shumyatskaya *et al.* (1971) is $Na_8Ba_2Ti_4O_4$ $[Si_{16}(Si,B)_8O_{56}]$. The parentheses emphasize the disorder of Si and B in only one of the three eight-fold tetrahedral sites.

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