# TIN-BEARING SILICATES FROM SKARN IN THE CASSIAR DISTRICT, NORTHERN BRITISH COLUMBIA

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## ABSTRACT

Two skarns in the Cassiar District of northern British Columbia are characterized by the presence of up to 0.9% $SnO_2$ in a unique iron-rich silicate assemblage of andradite, epidote, ferroactinolite, and hedenbergite. No normal tin minerals are present, and indications are that tin is principally in the lattice of garnet and epidote, probably as $Sn^{4+}$ substituting for $Fe^{3+}$.

## INTRODUCTION

The occurrence of tin in skarn in northern British Columbia in the pass at Lat. 59°17 ½′ N., Long. 130°31′ W. (Fig. 1) was first noted by Watson & Mathews (1944, p. 43). By spectrographic analyses, they found 0.3% tin and 0.5% tin respectively in two samples of skarn consisting mainly of pyroxene and of garnet. They also found that other samples of similar material were barren or nearly so, and concluded that the tin was distributed in an irregular manner.

One of the writers (R.M.) examined the locality in 1963 and collected samples of the various types of skarn present in the area. Only two were found to contain more than 0.1% tin; other samples from the pass and surrounding areas (Fig. 2) contain several hundredths of one per cent tin. Of the two tin-rich skarns, one is very coarse green garnetite which is brownish on weathered surfaces, and the other is a dark pyroxenite in which clinopyroxene and amphibole are the predominant constituents. Spectrographic analyses of bulk samples of the two skarns gave 1.1 and 0.78% tin respectively, and it thus appears that the materials may correspond with the tin-rich samples of Watson & Mathews.

Preliminary study of thin sections and mineral separates of the tin-rich skarns revealed no ordinary tin materials, and electron microprobe analyses indicated that much of the tin was bound within the lattice of garnet. The occurrence of tin in garnet has been reported on several occasions, most recently by Hosking (1965), Dadak & Novak (1965), and Sharkawi & Dearman (1966). In the present study, an attempt has been made to establish the tin distribution within the various coexisting silicates in the skarns.

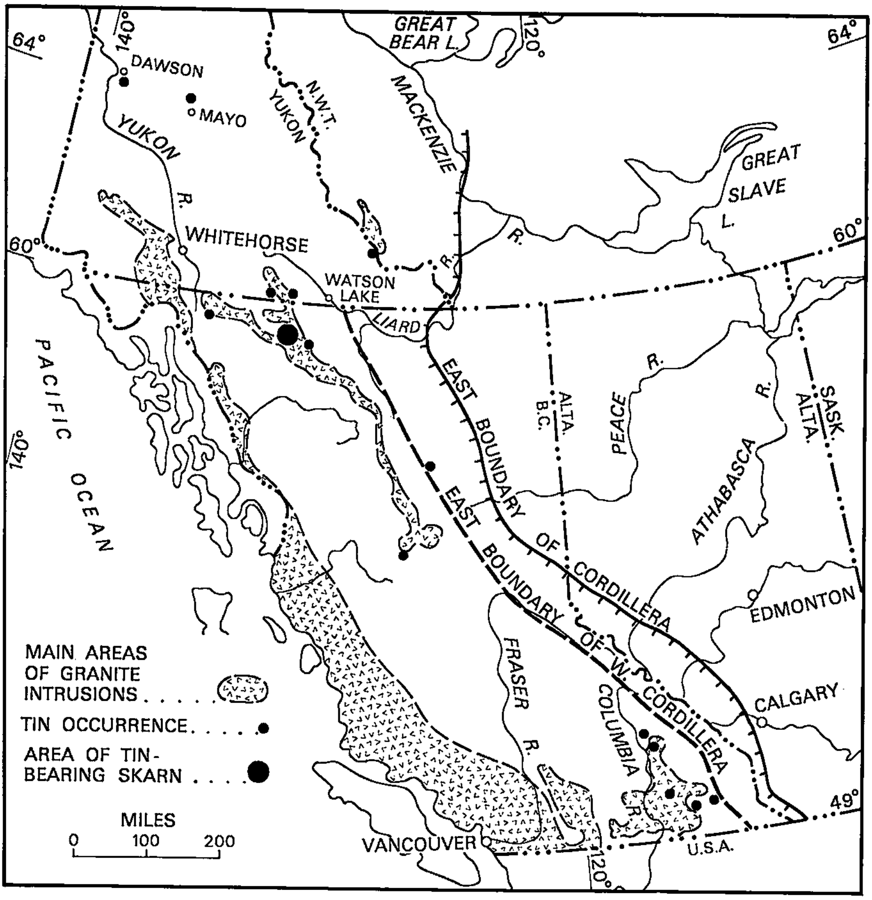

FIG. 1. Regional setting of Cassiar tin-bearing skarn.

## GEOLOGICAL SETTING

### *Regional*

The area (Fig. 1) lies within the eastern crystalline fringe belt of the Western Cordilleran region. This belt consists of Proterozoic and early Palaeozoic mainly metasedimentary rocks, in contrast with the Late Palaeozoic-Mesozoic volcanic-sedimentary assemblages farther west, and marks the eastern limit of the main zones of granitic intrusions. It includes several high-grade metamorphic complexes but the metamorphism, although regional in nature, is variable, and is quite low-grade in some places.

The belt contains all the beryllium, most of the known tin and tungsten, and many of the molybdenum occurrences of the Cordilleran region. It also contains most of the dominantly lead-zinc deposits. These mineral deposits are mainly confined to the vicinity of the two major eastern batholithic belts: the Nelson Batholith in the south, and the Cassiar-Omineca Batholith in the north.

### *Local*

The local geology is described in Watson & Mathews (1944). The bedded rocks (Fig. 2) comprise micaceous quartzites and argillaceous quartzites with occasional lenses of impure limestone, most of which has been recrystallized or converted to calc-silicate hornfels, and garnet, pyroxene, and vesuvianite skarns. The intrusive rocks are massive, medium-grained biotite granites. They contain a little muscovite in places, and large miarolytic cavities occur in the cirque east of Oblique Creek. Pegmatitic phases are few and small, but pink aplite is quite common and makes up much of the contact phase at the pass.

Little bedrock is actually exposed in the vicinity of the pass, most of the rock consisting of loose angular blocks of uncertain origin. However, on ascending the pass from the north, apparently southeastward-dipping

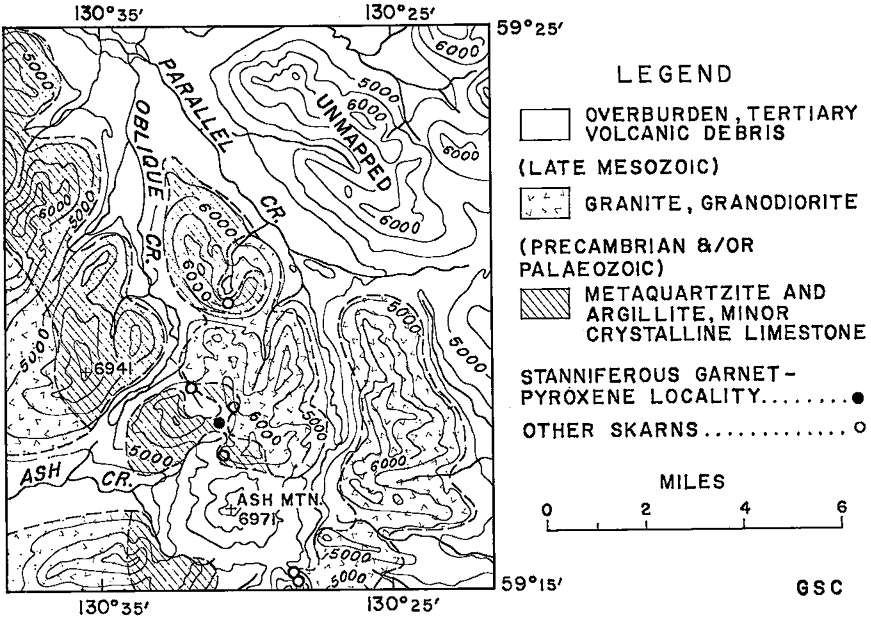

FIG. 2. Local geology of tin-bearing skarn locality.

crystalline limestone studded with small pink garnets is succeeded by skarn consisting chiefly of radiating sheaves of pale brownish green vesuvianite. Above this and just below the pass, the coarse tin-rich green garnetite occupies a limited area surrounded by broken rock and drift. The tin-rich pyroxenite occurs as a single boulder several hundred feet southwest of the pass.

No similar garnetite or pyroxenite was found in the ridges near the pass but tin-rich ferroactinolite and epidote, comparable to those in the pyroxenite, were found in samples collected from the skarn southeast of Ash Mountain in 1966. Other minerals found to contain several hundredths of 1% tin are a dark reddish and a white garnet, green clinopyroxene, and vesuvianite. Vesuvianite contains up to 0.025% beryllium. Very little scheelite or sulphides were found. The present study concerns only the garnetite and pyroxenite skarn samples collected at the pass in 1963.

## TIN-RICH GARNETITE

The tin-rich garnetite (MF 304.3) appears brown in hand specimen, with a rough, facetted surface of interlocking garnet crystals and finer interstitial material.

In thin section (Fig. 3, 4) it is seen to be composed of about 65% interlocking crystals of deep green garnet up to one inch across, and about 35% finer pale green to greenish brown clinopyroxene, with a little calcite, fluorite and graphite. The last two minerals are associated with the pyroxene rather than the garnet. The pyroxene is mainly in anhedral interstitial masses and in thin bands along the zone boundaries of the garnet crystals. It is non-pleochroic, partly twinned, with (+) $2V$ 60–65°, and $Z \wedge c$ up to 45°.

The garnet is finely banded in concentric zones parallel to the crystal outlines, the bands showing variable colour intensity and optical anisotropy. Scans across the zones with the electron probe showed that the darker green bands are richer in tin. Despite this variation, $x$-ray powder diffraction patterns are sharp in the back-reflection region. Using 114.6 mm cameras and Cu$K\alpha_1$ radiation, the (880), (12.2.2), and (14.4.2) reflections yield a cell edge of $12.064 \pm 0.002$ Å.

In addition to the heterogeneous tin distribution within the zoned crystals, one inclusion about 40 $\mu$ in diameter was observed during microprobe scans over an area of approximately 4 sq. cm. The presence of the inclusion, a tin-calcium-silicate, indicated a need for considerable caution in the preparation of the garnet sample purified for chemical analysis. In addition to crushing, sieving, and separation by gravity and magnetic techniques, the sample was hand picked and subsequently

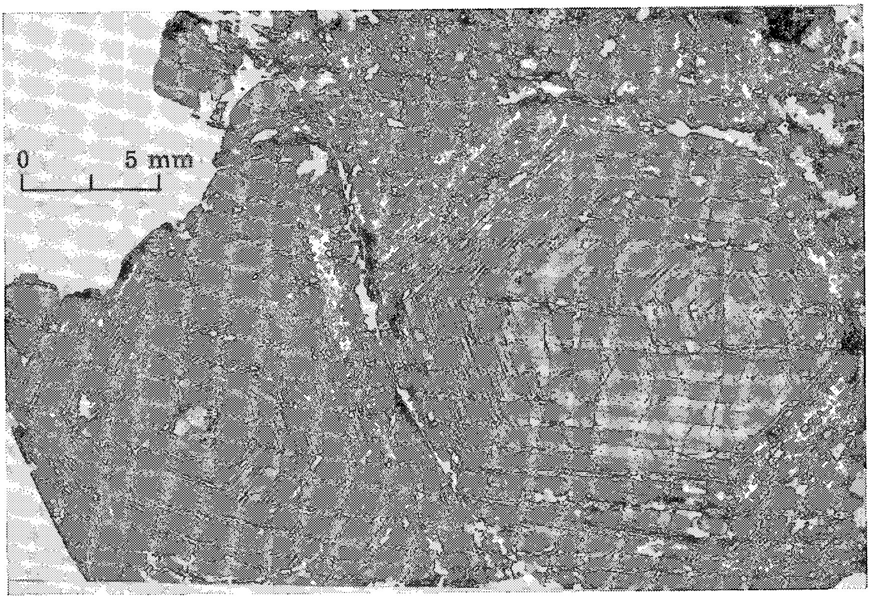

FIG. 3. Tin-rich garnetite, MF304.3, plain light ×4.

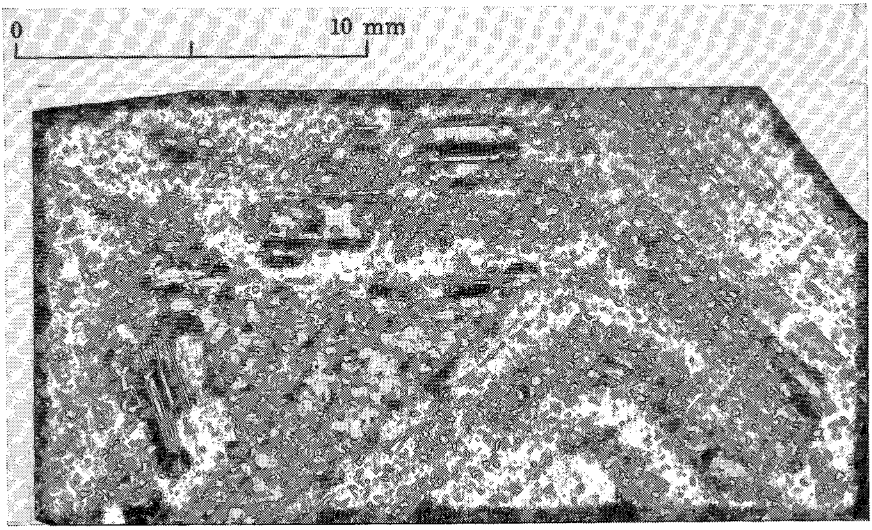

FIG. 4. Tin-rich garnetite, MF304.4, crossed polarizers ×39.

subjected to several HF and concentrated $HNO_3$ treatments. At an intermediate stage in these treatments, the material was ground by hand in a mortar and examined in immersion oil after each acid bath. The procedure was continued until the sample appeared completely purified. Unfortunately, much of the garnet was also consumed by the acids during this drastic procedure, and the final sample weighed only 450 mg. The results of the chemical analysis (Table 1) show that the garnet is unique in its andradite content (99.95), the remainder being 0.4 and 0.1 molecules of almandite and pyrope respectively. The tin content is 0.9% $SnO_2$.

The refractive index of the tin-bearing garnet is 1.88 $\pm$ $<0.01$, which is almost the same as that of synthetic andradite ($n = 1.887$) reported by Skinner (1956). On the other hand, the cell edge of synthetic andradite is 12.048 $\pm$ 0.001 Å (Skinner 1956), considerably smaller than the 12.064 Å obtained for the Cassiar mineral. The reason for this difference will be discussed in a later section. In conclusion, however, it would appear that virtually all the tin in the skarn is evidently present in solid solution in the garnet.

TABLE 1. CHEMICAL ANALYSIS OF GREEN TIN-BEARING GARNET FROM GARNETITE SKARN

(Analyst: J. L. Bouvier)

| | | No. of ions on the basis of 24 (O) | | |
|---|---|---|---|---|
| $SiO_2$ | 33.6 | Si | 5.69 | 6.00 |
| $Al_2O_3$ | 2.5 | Al | 0.31 | |
| $Fe_2O_3$ | 30.2 | Al | 0.19 | 4.04 |
| FeO | 1.8 | $Fe^{+3}$ | 3.85 | |
| MnO | 0.1 | Mg | 0.05 | 5.97 |
| MgO* | 0.2 | $Fe^{+2}$ | 0.25 | |
| CaO | 31.2 | Mn | 0.01 | |
| $H_2O$ | 0.2 | Ca | 5.66 | |
| $SnO_2$ | 0.9 | Sn | 0.12 | |
| | 100.7 | | | |

*By emission spectroscopy.

## TIN-RICH PYROXENITE

The tin-rich pyroxenite is dark greenish brown in hand specimen. Most of the brown colour is due to the presence of secondary iron oxides along grain boundaries, cleavages, microscopic fractures, and on exposed grain surfaces. The principal constituents of the pyroxenite skarn are clinopyroxene, amphibole, epidote, garnet, calcite and quartz. These form compact masses, but many of the silicates are also present as euhedral crystals which project into cavities less than half a centimeter in diameter.

Under the binocular microscope nearly all the unaltered clinopyroxene is of the light to medium green colour generally characteristic of diopside. Amphibole occurs typically as masses consisting of aggregates of black, lustrous sheaves. Individual fibers projecting into cavities are dark green and transparent. Epidote tends to occur as black to smoky-coloured individual subhedral grains averaging about 2 mm in diameter. Garnet has a somewhat similar occurrence but is readily distinguished because of its pale brown colour.

Thin sections of the pyroxenite skarn consist mainly of the pale greenish clinopyroxene, epidote, and dark green amphibole. Most of the pyroxene is present as coarse subhedral grains up to 6–7 mm in length. The mineral is weakly pleochroic from colourless to pale green, optically positive with $2V$ about 60°, and has extinction angles up to 45°. It is thus quite similar to that in the garnetite. Optical spectrographic analysis of the clinopyroxene gave major Ca and Fe with only 0.15% Al and 0.55% Mg, thus placing the mineral in the hedenbergite-ferrohedenbergite series. Because of the low Mg and Al contents, the proportion of Fe and Ca can be approximately determined from the cell dimensions using the method outlined by Brown (1960) and modified by Viswanathan (1966). The reflections 200, 020, 220, $\bar{2}21$, $\bar{3}11$, 330, $\bar{3}31$, $\bar{4}21$, 041 and 150 were measured by diffractometer and on Guinier films, using internal standards in both. Least squares treatment of the measured $d$-spacings gave $a = 9.847$, $b = 9.025$, $c = 5.228$ Å, $\beta = 104.62°$, $a \sin \beta = 9.528$. These place the composition of the Cassiar mineral near the hedenbergite corner of the diopside-hedenbergite-clinoenstatite-clinoferrosilite quadrilateral. The $a \sin \beta$ value (9.528) is comparable to Viswanathan's (1966) assigned value of 9.530 for the theoretical hedenbergite end-member.

*Epidote* is abundant in some thin sections and almost absent in others. Although it is commonly euhedral to subhedral in hand specimens because of its occurrence adjacent to cavities, in thin section the epidote is typically anhedral and weakly pleochroic from colourless to pale greenish yellow. The $\beta$ and $\gamma$ refractive indices are 1.729 and 1.754 respectively, both $\pm 0.005$. A chemical analysis of hand-picked crystal fragments is given in Table 2. The resultant numbers of ions (and their distribution) were calculated on the anhydrous basis of 25 (O) in order to correspond to the analyses and $x$-ray determinative data for epidotes as given by Myer (1966). The analysis of the Cassiar epidote shows two unusual features—a tin content of 0.8% $SnO_2$, and an FeO content of 1.5%. The latter yields $Fe^{+2}/Fe^{+3} = 0.165$, whereas Myer (1965, 1966) found this ratio to be a maximum of 0.08 in epidotes.

Using essentially the same procedure as outlined by Myer (1965), the cell dimensions obtained for the Cassiar epidote are $a = 8.889$, $b = 5.632$, $c = 10.15$ Å, $\beta = 115°24'$, $V = 459.0$ Å$^3$. The chemical composition of normal epidote may be expressed as $Ca_2(Al_{1-x}, Fe_x^{+3})_3Si_3O_{12}(OH)$, where $x$ varies over the approximate range 0–0.40. Myer (1966) has shown that

TABLE 2. CHEMICAL ANALYSIS OF CASSIAR EPIDOTE
(Analyst: J. L. Bouvier)

| | | No. of ions on the basis of 25 (O) | | |
|---|---|---|---|---|
| $SiO_2$ | 38.2 | Si | | 6.005 |
| $Al_2O_3$ | 24.9 | Al | 4.610 | 5.987 |
| | | $Fe^{3+}$ | 1.194 | |
| $Fe_2O_3$ | 10.1 | Mg | 0.070 | |
| | | Mn | 0.013 | |
| FeO | 1.5 | Sn | 0.100 | |
| $SnO_2$ | 0.8 | Ca | 3.904 | 4.101 |
| | | $Fe^{+2}$ | 0.197 | |
| MnO | 0.1 | | | |
| MgO* | 0.3 | | | |
| CaO | 23.2 | | | |
| $H_2O$ | 1.9 | | | |
| | 101.0 | | | |

*By emission spectroscopy.

the unit cell parameters of epidote are a function of $Fe_x$ composition; using his determinative curves, the compositional relationships given in Table 3 were derived for the Cassiar epidote. The cell volume and chemical analysis yield $Fe_x$ compositions of 0.21 and 0.20 respectively. This agreement suggests that the anomalously large value for $b$ and the accompanying low values for $a$ and $c$ may not be fortuitous, but may indicate lattice distortion arising from the substitution of tin.

*Amphibole* in thin sections is present as sheaves of dark green needles, chiefly along the grain boundaries and cleavage planes of the pyroxene grains and in random patches within them. It is pleochroic in light to dark green colours, the latter having a bluish tint. It is negative with $2V$ about 50°, $Z \wedge c = 12° \pm 2°, \gamma = 1.694$.

$X$-ray diffraction patterns and optical spectrographic analyses of the amphibole indicate that it is ferroactinolite containing 2.7% Al, 0.59% Mg, and 0.37% Mn. The unit cell dimensions were calculated from 114.6 mm $x$-ray powder photographs, and Guinier films, supplemented by a zero-level precession photograph of the $a^*$ $c^*$ net. The calculated dimensions after least squares treatment of the $x$-ray powder data are: $a = 9.97$, $b = 18.21$, $c = 5.32$ Å, $\beta = 104.53°$, $V = 935$ Å$^3$. For synthetic ferroactinolite, Ernst (1966) obtained $a = 9.97 \pm 0.01$, $b = 18.34 \pm 0.02$, $c = 5.30 \pm 0.01$ Å, $\beta = 104.5 \pm 0.1°$, $V = 938$ Å$^3$. The agreement among the cell dimensions of the natural and synthetic minerals is thus relatively good. The smaller $b$ values of the Cassiar ferroactinolite may be a reflection of the mineral's alumina content, but any further discussion of the matter would be fraught with too many ambiguities.

*Garnet* in the pyroxenite skarn is quite unlike that in the garnetite. In

TABLE 3. RELATIONSHIP OF $Fe_x$ COMPOSITION AND UNIT CELL PARAMETERS OF CASSIAR EPIDOTE

| Parameter | $Fe_x$ composition |
|---|---|
| $a$ | 0.18 |
| $b$ | 0.25 |
| $V$ (Å)$^3$ | 0.21 |
| $\Delta 2\theta$ Cu$K\alpha_1$($020_{ep} - 111_{Si}$) | 0.25 |

the pyroxenite the garnet is pale brown, not zoned, and completely isotropic. Aggregates of subhedral crystals averaging 1–2 mm in diameter were commonly observed in hand specimens. Spectrographic analyses of hand picked material indicate that the mineral is a calcium-iron garnet containing 3.0% Al and 0.17% Mg; the measured cell edge of 12.014 ± 0.005 Å confirms that it is an andradite.

The remainder of the pyroxenite skarn mineralization is *quartz*, *calcite*, and *sphene*. The first two are abundant in the skarn, occurring principally as anhedral material filling interstices in the pyroxene-epidote-garnet assemblage. Only very small amounts of sphene are present. The mineral occurs as small (averaging 1 mm) rosette-like clusters of white crystals on pyroxene and amphibole. Identification of the sphene was made by *x*-ray powder patterns and confirmed by *x*-ray spectroscopy.

## TIN DISTRIBUTION IN THE SKARNS

Except for the small inclusion detected by microprobe analysis of the zoned green garnet making up the bulk of the garnetite skarn, no other tin minerals have been identified in either the pyroxenite or garnetite. There is always a need for caution in the interpretation of whether an element such as tin is present as a discrete mineral of microscopic or submicroscopic size, or whether the element is indeed bound in the lattice of the host mineral. The relatively large amount of tin present together with the absence of inclusions in the well-purified garnet from the garnetite skarn would under normal circumstances strongly suggest that the element is in the garnet lattice. Confirmation has, in this case, been obtained by means of microprobe analyses. Equivalent treatment of the silicates in the pyroxenite skarn was not possible and it is therefore more realistic to regard the tin distribution, given below, in terms of mineral associations without specification as to how the tin occurs.

### *Sample preparation and analyses*

Garnet, epidote, pyroxene, and amphibole from the pyroxenite skarn were hand picked under a 50× binocular microscope. With the exception of amphibole, almost all the mineral fragments selected were well crystallized grains exposed in cavities. Intergrowth material was thus largely avoided. The samples were crushed by hand in a small agate mortar and repicked through several successive crushings until only clear transparent material remained. Individual samples were then ground in alcohol and subsequently analyzed under the supervision of W. H. Champ using the optical spectrographic techniques as outlined by Champ (1964). Several additional pyroxene and amphibole samples from various localities (Table 4) were also examined for comparative purposes. These "standards" consisted of material which was selected and cleaned by H. R. Steacy from specimens available in the National Mineral Collection, Systematic Reference Series.

In addition to the well-purified samples, two concentrates of amphibole and one of quartz were prepared by crushing, sieving, and heavy liquid separation. The quartz concentrate contained a substantial amount of impurities, but these were largely neglected as the purpose was to observe

Table 4. Tin Contents of Amphiboles and Pyroxenes as compared to equivalent determinations in Cassiar Skarn Minerals

(Optical spectrographic analyses by W. H. Champ, K. A. Church, and W. F. White)

| | Mineral | Locality | Sn, ppm |
|---|---|---|---|
| *Amphiboles* | edenite | Grenville Tp., Ont. | 1 |
| | hornblende | Bancroft, Ont. | 6 |
| | hornblende | Cardiff Tp., Ont. | 26 |
| | hornblende | Wakefield Tp., Que. | 1 |
| | tremolite (nephrite) | New Zealand | 1 |
| | ferrohastingsite | Hastings Co., Ont. | 4 |
| | actinolite | Ross Tp., Ont. | 1 |
| | ferroactinolite | pyroxenite skarn, Cassiar dist., B.C. | 2,700 |
| *Pyroxenes* | aegerine | Mt. St. Hilaire, Que. | 32 |
| | aegerine | Kola Peninsula | 1 |
| | augite | Prague, Czechoslovakia | 2 |
| | augite | Montreal, Hochelaga Co., Que. | 2 |
| | diopside | Portland, Ottawa Co., Ont. | 1 |
| | diopside | Cardiff, Haliburton Co., Ont. | 0 |
| | hedenbergite | pyroxenite skarn, Cassiar dist., B.C. | <70* |
| | | | Sn, % |
| *Epidote* | epidote | pyroxenite skarn Cassiar dist., B.C. | 1–2%† |
| *Garnet* | andradite (green) | garnetite skarn, Cassiar | 1–2%‡ |
| | andradite (brown) | pyroxenite skarn, Cassiar | 0.43% |
| *Concentrates* | amphibole A-1 | pyroxenite skarn, Cassiar | 0.25% |
| | amphibole A-2 | pyroxenite skarn, Cassiar | 0.28% |
| | quartz | pyroxenite skarn, Cassiar | 0.032% |

*The lower limit of detection because of the relative small amount of pure sample available.

†See chemical analysis, Table 1.

‡See chemical analysis, Table 2.

whether tin showed a relative increase or decrease. The amphibole concentrates, on the other hand, were of greater than 95% purity, principally because of the ease of separation of the mineral.

### *Results*

The analyses given in Table 4 show that tin is not evenly distributed among the principal silicates of the pyroxenite skarn, but is concentrated primarily in epidote and andradite. The tin contents of the epidote and green andradite are above the working range of the trace element spectrographic method used and hence the cited values were reported only as estimates.

Assuming that most of the tin occurs within the lattice of the silicates, substitutions can be considered in terms of the ideal formulae given below:

| | |
|---|---|
| hedenbergite | $CaFe''Si_2O_6$ |
| ferroactinolite | $Ca_2Fe''_5Si_8O_{22}(OH)$ |
| epidote | $Ca_2Fe'''Al_2Si_3O_{12}(OH)$ |
| andradite | $Ca_3Fe'''Si_3O_{12}$ |

The pertinent elements common to all the above minerals are thus Ca, Fe, and Si. Substitution by tin may occur as both $Sn^{2+}$ and $Sn^{4+}$, the respective ionic radii being 0.93 and 0.71 Å (Ahrens). The geochemical studies of Hamaguchi *et al.* (1964) have shown that tin is impoverished in basic magmatic rocks, and hence while substitution of $Sn^{2+}$ for $Ca^{2+}$ (0.99 Å) is possible, enrichment of tin in residual magmas suggests that such a replacement does not take place. In the Cassiar minerals, the more likely possibilities are substitution of $Sn^{4+}$ (0.71 Å) for $Fe^{3+}$ (0.64 Å), or substitution of the anionic complex $(SnO_4)^{4-}$ for $(SiO_4)^{4-}$. It is noteworthy that most tin-rich garnets contain ferric iron as a principal constituent, and in the Cassiar skarns the greatest tin enrichment occurs in the two minerals which have a substantial ferric iron component (andradite and epidote). Published analyses of hedenbergite and ferroactinolite commonly show up to several per cent $Fe_2O_3$, particularly in the latter mineral. An assumed low ferric iron content in the amphibole could thus account for the relatively low tin content present in the mineral.

Having thus concluded that tin is most likely present as $Sn^{4+}$ substituting for $Fe^{3+}$, scans with the electron microprobe were made across the zoned green tin-bearing andradite. Although calcium content did not vary, both iron and tin showed fluctuations across the zones. The gross counts obtained for the two elements at 42 different spots showed that as tin increased, iron diminished. Although the substitution of tin for iron has thus been confirmed, the exact nature of this replacement is not known. Substitution of $Sn^{2+}$ (0.93 Å) for $Fe^{2+}$ (0.74 Å) could account for the presence of some of the tin, but the analyses (Table 4) show a strong sympathetic correlation between tin and minerals rich in ferric iron. $Sn^{4+}$ (0.71 Å) has a favourable similarity in size to $Fe^{3+}$ (0.64 Å), but has the disadvantage of requiring a compensatory substitution to achieve charge balance. As there are no indications of univalent cations such as alkalis being present, charge balance could be achieved by an unusual coupled substitution such as $Sn^{4+} + Fe^{2+} \rightarrow 2Fe^{3+}$.

## CONCLUSIONS

The skarns occurring in the Cassiar district are characterized both by the presence of tin and by the presence of an extremely iron-rich silicate assemblage. Most of the tin is associated with minerals rich in ferric iron, that is, andradite and epidote. For the most part, tin probably occurs within the lattices of these minerals as $Sn^{4+}$ substituted for $Fe^{3+}$.

Contrary to common expectation, it would appear that the entry of tin into some silicates of the pyroxenite skarn occurred at relatively modest temperatures. Ernst (1966) has shown that the maximum high-temperature stability limit of ferroactinolite is only 543° C at 3000 bars fluid pressure. Although the presence of alumina in the assemblage and in the Cassiar amphibole would probably extend the ferroactinolite stability limits, formation of this skarn mineral at rather high temperatures would appear to be unlikely.

## ACKNOWLEDGMENTS

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