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DIASPORE IN A PYROPHYLLITE DEPOSIT ON THE AVALON PENINSULA, NEWFOUNDLAND

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ABSTRACT

Small nodules of diaspore are present in a pyrophyllite deposit on the Avalon Peninsula of Newfoundland. The pyrophyllite forms a lens-like body in a northerly-trending belt of rhyolitic flows and pyroclastics (part of the Harbour Main Group), near the contact with a granitoid pluton (the Holyrood Batholith), both of Late Precambrian age.

The pyrophyllite rock consists of a very fine-grained assemblage of pyrophyllite, muscovite, and quartz in variable proportions, with small quantities of kaolinite. The diaspore nodules occurring sporadically throughout the deposit contain platy diaspore, interstitial pyrophyllite, small amounts of rutile and local coarse-grained barite. The nodules range in size from 1 cm to 10 cm. Microprobe analyses, optical data and unit-cell dimensions of the diaspore and pyrophyllite are given.

Alteration of the original rhyolite took place along shear zones acting as channels for acid solutions derived from the Holyrood pluton. The formation of the pyrophyllite lenses involved outward migration of silica and alkalies, leaving behind less mobile alumina; short-range transport of aluminum in solution is indicated by the diaspore nodules, by rare thin veins containing platy pyrophyllite, and by veins of massive pyrophyllite cutting silicified rhyolite at the termination of some shear zones. Published experimental work shows that the pyrophyllitization may have taken place in a temperature range of 260-280°C under a pressure of 2 kbar or less.

SOMMAIRE

De petits nodules de diaspore se trouvent dans un gîte de pyrophyllite, sur la péninsule d'Avalon, à Terre-Neuve. La pyrophyllite, de forme lenticulaire, apparaît dans une bande en direction à peu près N-S de coulées rhyolitiques et de roches pyroclastiques (appartenant au groupe de Harbour Main), près du contact avec un massif plutonique (batholithe de Holyrood), qui datent tous deux de la fin du Précambrien.

La roche à pyrophyllite consiste en un assemblage à grain très fin de pyrophyllite, muscovite et quartz, en proportions variables, avec un peu de kaolinite. Les nodules de diaspore se présentent sporadiquement dans tout le gîte et se composent de diaspore tabulaire, de pyrophyllite interstitielle, avec un peu de rutile et, localement, de barytine grenue. La taille des nodules varie de 1 à 10 cm. On donne, pour le diaspore et la pyrophyllite, l'analyse à la microsonde, les constantes optiques et les dimensions de la maille.

L'altération de la rhyolite originelle s'est produite le long de zones de cisaillement, voies d'accès pour les solutions acides provenant du pluton de Holyrood. La formation de pyrophyllite implique une migration de la silice et des alcalis, laissant derrière eux l'alumine moins mobile; un transport de l'alumine à courte distance est indiqué par les nodules de diaspore, par quelques filonnets contenant de la pyrophyllite tabulaire et par les filons de pyrophyllite massive qui traversent la rhyolite silicifiée là où prennent fin certaines zones de cisaillement. De travaux expérimentaux publiés, il ressort que la pyrophyllitisation se serait produite dans l'intervalle de température compris entre ~260 et ~280°C, sous une pression n'excédant pas 2 kbar.

(Traduit par la Rédaction)

INTRODUCTION

The only producing pyrophyllite mine in Canada lies on the Avalon Peninsula of Newfoundland, within a belt of Late Precambrian rocks of the Avalon Zone which extends intermittently along the southeastern flank of the Appalachian mobile belt from Newfoundland to South Carolina (Williams 1976).

The Avalon Peninsula is an elongate northerly-trending dome with a predominantly volcanic assemblage (the Harbour Main Group) in the core, overlain on the flanks by mainly sedimentary rocks of the Conception, Cabot and Musgravetown Groups. The Harbour Main volcanics in the central part of the peninsula are intruded by a shallow-level granitoid pluton, the Holyrood Batholith. The Late Precambrian rocks are folded, cut by numerous faults, and overlain unconformably by patches of Lower

Cambrian to Lower Ordovician sedimentary rocks (McCartney 1967; Rose 1952).

The Avalon rocks have been subjected to a low grade of metamorphism, grading from prehnite-pumpellyite facies on the peninsula itself to greenschist facies to the west of the Isthmus of Avalon. In addition a diffuse thermal aureole, characterized by incipient development of actinolite in basic rocks, follows the contacts of the Holyrood pluton. The rocks within the contact aureole straddle the boundary between upper prehnite-pumpellyite and lower greenschist facies (Papezik 1974a).

Pyrophyllite Deposits

Along the northern part of its eastern contact, south of the settlement of Foxtrap on Conception Bay, the Holyrood pluton intrudes a northerly-trending belt of rhyolitic flows and pyroclastics with minor sediments. The acid rocks have been locally fractured and/or sheared, mainly in easterly to northeasterly direction, and altered along the shear zones to a very fine-grained assemblage of quartz, muscovite ("sericite") and pyrophyllite in widely different proportions. Several of the lens-like zones rich in pyrophyllite are of economic grade, and the largest of them has supported commercial production of pyrophyllite for more than 20 years.

The pyrophyllite deposits in the area have been known since the end of the nineteenth century, and have been described by Buddington (1916) and Vhay (1937); more recent detailed work has been reported by Keats (1970) and Papezik (1974b). The largest deposit, discovered in 1898, has been worked intermittently since 1903. The present mine (the "Oval Pit") was opened in 1956 by Newfoundland Minerals Ltd. It produces 30,000 to 40,000 short tons of pyrophyllite annually; the mineral is exported to the U.S.A. for use in the production of ceramic tile.

The mine is located on a ridge about 2.5 km southeast of the settlement of Foxtrap, Conception Bay (47°29′06″N, 52°57′12″W; Nat. Topogr. Ser. Map 1 N/7, Bay Bulls). It is an open pit about 500 m long, 400 m wide and 50 m deep, developed in three benches. The north wall of the pit lies in grey flow-banded spherulitic rhyolite with large brownish patches of finely divided hematite. The south wall consists of sediments ranging from coarse angular breccia with fragments up to 30 cm across, through conglomerate composed of 1-3 cm clasts to fine-grained sandstone. The sediments are in fault contact with the rhyolitic rocks, and may be derived in part by slumping and rapid erosion of a fault scarp. The pyrophyllitized zone, exposed over its full width in the pit, is about 200 m wide.

In addition to the main zone, other altered zones of some significance are found at Mine Hill, 0.75 km to the south of the Oval Pit, and at Dog Pond, 8 km south of the mine, at 47°26′14″N, 52°56′45″W.

TABLE 1. ROCKS FROM THE FOXTRAP PYROPHYLLITE MINE, AVALON PENINSULA, NEWFOUNDLAND

	1	2	3
SiO_2	73.28	63.89	19.60
TiO_2	-	0.13	0.48
Al_2O_3	16.00	30.0	67.62
Fe_2O_3	1.32*	0.11*	0.06
FeO	-	-	0.06
MnO	-	-	nil
MgO	0.16	0.07	nil
CaO	0.58	nil	nil
Na_2O	7.43	0.16	0.06
K_2O	0.70	0.95	0.15
P_2O_5	-	-	nil
L.O.I.	0.19	5.10	12.20
	99.66	100.41	100.23

* Total Fe as Fe_2O_3 - not determined

1. Flow-banded rhyolite, 0.8 km NE of the Foxtrap mine. Unaltered except for a probable alkali exchange; no pyrophyllite, only traces of sericite. Analysis by American Olean Tile Co., Lansdale, Pennsylvania (Keats 1970).
2. High-grade massive pyrophyllite rock, Foxtrap mine. (Anal.: A. Lee and H. Burke, Nfld. Dept. of Mines & Energy). Calculated mode: Pyrophyllite 89.6%, sericite 9.2%, diaspore 1.2%.
3. Diaspore nodule, Foxtrap mine. (Anal.: J.-L. Bouvier, Geol. Survey of Canada). Calculated mode: Diaspore 69.5%, pyrophyllite 28.0%, sericite 2.0%, rutile 0.5%.

The rocks within the main altered zone consist of a very fine-grained assemblage of pyrophyllite, muscovite ("sericite") and quartz (Table 1). Feldspar is completely absent, and kaolinite is present only in trace amounts. The proportions of the three main minerals vary within wide limits. Pyrophyllite-quartz rocks with little muscovite are common; monomineralic domains are relatively small, and restricted to lenses of massive pyrophyllite and a few quartz veins. Scanning electron microscope (Cambridge Scientific Instruments "Stereoscan" Mark 2A, operated by Dr. V. C. Barber, Dept. of Biology, Memorial Univ.) shows that individual flakes of pyrophyllite and muscovite are about 10-40 μm across; irregular patches of cryptocrystalline silica reach 0.1 mm or more in size (Fig. 1). The massive pyrophyllite rock is pale greenish yellow and translucent, with a waxy lustre. It has an attractive appearance when cut and polished, and is suitable for carving.

Diaspore

Some of the high-grade pyrophyllite lenses contain sparsely distributed rounded nodules rich in diaspore, AlO(OH). The diaspore nodules, light purplish grey in color, range in size from 1 cm to about 10 cm, only rarely attaining larger dimensions. A chemical analysis of a typical diaspore nodule is given in Table 1.

The diaspore nodules are generally fine-grained at the margins, and are commonly surrounded by a diffuse halo of finely disseminated diaspore in nearly pure massive pyrophyllite (Fig. 2). Inwards from the margins, the fine-grained cloudy diaspore gives way to diaspore rosettes, intergrown with increasingly coarse pyrophyllite. The diaspore crystals are platy, up to 2-3 mm long and usually less than 0.1 mm thick (Fig. 3). Some of the diaspore nodules contain irregular patches, from microscopic dimensions to several centimeters long, of white to buff-colored barite (sp. gr. 4.44±0.03). In addition, very small (0.1-0.2 mm) crystals of rutile are common in the nodules; locally they are concentrated along the contacts of the barite patches. In some of the smaller barite domains, the intersecting network of diaspore plates is continuous throughout the barite, but pyrophyllite is absent (Fig. 4).

Electron microprobe analyses of diaspore and pyrophyllite and optical data for both minerals (determined by standard immersion and U-stage methods) are given in Table 2. The analyses are

TABLE 2. DIASPORE AND PYROPHYLLITE, FOXTRAP, NEWFOUNDLAND. MICROPROBE ANALYSES AND OPTICAL DATA

	1	2	3	4
SiO_2	0.07		67.10	66.70
TiO_2	0.18		0.04	
Al_2O_3	84.32	84.98	28.42	28.30
Fe_2O_3*	0.12		0.25	
MnO	nil		nil	
MgO	nil		0.02	
CaO	nil		0.01	
Na_2O	0.01		0.07	
K_2O	0.05		0.06	
	84.75		95.97	
H_2O**	15.02	15.02	5.00	5.00
	99.77	100.00	100.97	100.00
n_α	1.700 ± 0.002		1.530 (calc.)	
n_β	1.715 ± 0.002		1.584 ± 0.002	
n_γ	1.740 ± 0.002		1.598 ± 0.002	
2V	(+) 84° ± 1°		(-) 54° ± 1°	

*Total Fe as Fe_2O_3 **Stoichiometric H_2O

1. Platy diaspore crystal; diaspore nodule in pyrophyllite.
2. AlO(OH).
3. Pyrophyllite intergrown with diaspore; diaspore nodule.
4. $Al_2Si_4O_{10}(OH)_2$.

Analyst: D.B. Clarke, Dalhousie University, Halifax. (MnO by A. Thompson, Memorial University).

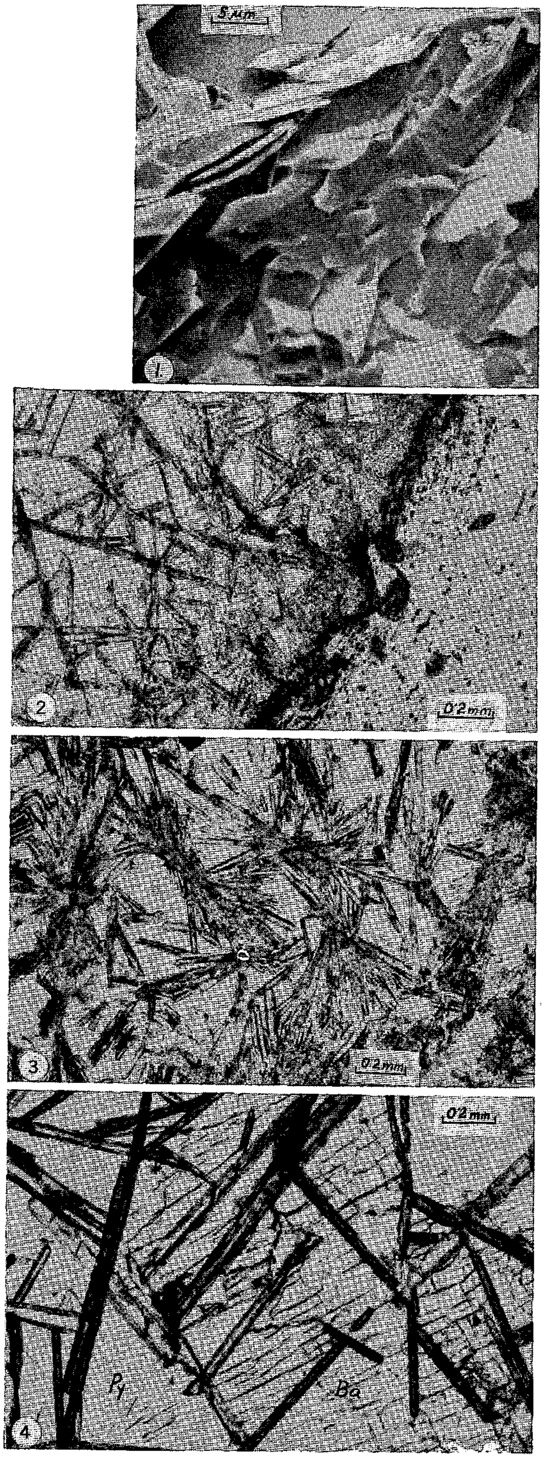

FIG. 1. Scanning electron microscope image of pyrophyllite and sericite flakes in a massive pyrophyllite rock.
2. Contact of a diaspore-pyrophyllite nodule with massive pyrophyllite.
3. Diaspore rosettes in a diaspore-pyrophyllite nodule.
4. Barite in a diaspore-pyrophyllite nodule.
(All illustrated specimens are from the Foxtrap pyrophyllite mine).

very close to stoichiometric compositions; elements not present in theoretical formulae amount to less than 0.5% in both cases. X-ray diffraction patterns of both minerals agree closely with those in the Powder Diffraction File and in published literature (Brindley & Wardle 1970). Cell dimensions were calculated using a computer program of Appleman *et al.* (1972), modified by R. G. Cawthorn and D. Press of Memorial University; they differ only slightly from published data (Table 3).

TABLE 3. UNIT-CELL DIMENSIONS OF DIASPORE AND PYROPHYLLITE FROM FOXTRAP, NEWFOUNDLAND

	1	2	3	4
a	4.399±0.006Å	4.396Å	5.167±0.002Å	5.172Å
b	9.414±0.006	9.426	8.950±0.003	8.958
c	2.849±0.005	2.844	18.645±0.005	18.676
β	90°		99°56'±02'	100.0°
V	117.98±0.44	117.84*	849.40±0.43	852.1*

*calculated from published data.

1. Diaspore, Foxtrap, Newfoundland. (This study).
2. Diaspore, Springfield, Mass. JCPDS card 5-355.
3. Pyrophyllite, Foxtrap, Newfoundland. (This study).
4. Pyrophyllite (monoclinic); Honami, Japan (Brindley & Wardle 1970).

The occurrence of diaspore in pyrophyllite deposits is common enough to be the rule rather than the exception. It has been reported from North Carolina (Zen 1961, Stuckey 1967); Japan (e.g. Watanabe 1951; Iwao & Udagawa 1969), India (Misra & Sood 1947; Prakash *et al.* 1970), Morocco (Leblanc 1970), and other localities. In Canada, diaspore and pyrophyllite occur as interstitial minerals in quartzite of the Early Proterozoic Lorraine Formation north of Lake Huron (Chandler *et al.* 1969); no other Canadian occurrences are known to the authors. At many of the reported localities, the diaspore-pyrophyllite rocks contain variable amounts of "acid-indicator" minerals such as alunite, topaz, fluorite, zunyite, dumortierite and apatite. None of these has been identified so far in the Foxtrap deposit; only the barite, not reported elsewhere in this association, provides an indication of the chemistry of the mineralizing solutions.

Origin of the Diaspore-Pyrophyllite Assemblage

As all gradations from prophyllite-rich rock through quartz-muscovite schists to unaltered rhyolite can be seen within a relatively small area, there can be little doubt that the Al-rich rocks were produced by an alteration of rhyolite flows and pyroclastics, involving the introduction of an aqueous fluid. The sporadic occurrence of a sulfate in the Al-rich assemblage shows that the fluids involved in the alteration were acid, and the restriction of the altered zones to the vicinity of the Holyrood pluton indicates that the fluids either originated in, or were activated by, the granitoid pluton, probably during or shortly after its intrusion.

Comparison of the chemical composition of a relatively unaltered rhyolite, high-grade pyrophyllite rock and a diaspore nodule (Table 1) shows that the alteration of the original rhyolite involved mainly the removal of large quantities of silica and alkalies and smaller amounts of Ca, Mg and Fe. The sharp increase in the Al content of the pyrophyllite- and diaspore-bearing rocks is the result of residual concentration of alumina by the leaching of the more mobile components, an "enrichment by subtraction" (Althaus 1969, p. 105), and does not reflect metasomatic addition of Al from an external source.

According to a recent experimental work (Tsuzuki & Mizutani 1971), the pyrophyllitization of the rhyolitic rocks may have proceeded in several stages. Following the sericitization of the original alkali feldspar in the presence of aqueous fluids (a process which can release both alkalies and silica to the solution), the "sericite" was first altered to kaolinite in the presence of an acid, releasing further alkali ions:

$$\underset{\text{sericite}}{2\ KAl_2(AlSi_3O_{10})(OH)_2} + 3\ H_2O + 2\ H^+ \rightarrow \underset{\text{kaolinite}}{3\ Al_2Si_2O_5(OH)_4} + 2\ K^+$$

This was then followed by:

$$\underset{\text{kaolinite}}{Al_2Si_2O_5(OH)_4} + 2\ H_4SiO_4 \rightarrow \underset{\text{pyrophyllite}}{Al_2Si_4O_{10}(OH)_2} + 5\ H_2O$$

Experiments of Tsuzuki & Mizutani (1971) show the presence of a narrow zone of kaolinite between sericite and pyrophyllite, which migrates outward as alteration proceeds. A distinct kaolinite zone has not yet been detected in the Foxtrap pyrophyllite mine, but extensive silicification and sericitization is common in the area surrounding the pyrophyllite lenses. The internal structure of the main pyrophyllite zone in the Oval Pit is too complex and too obscured by mining operations to yield reliable data, but good evidence for successive alteration zones was found in a narrow (15 m) shear zone at the Dog Pond locality, where an aluminous (pyrophyllite-rich) core is flanked by a zone high in sericite (an "alkali front") followed by

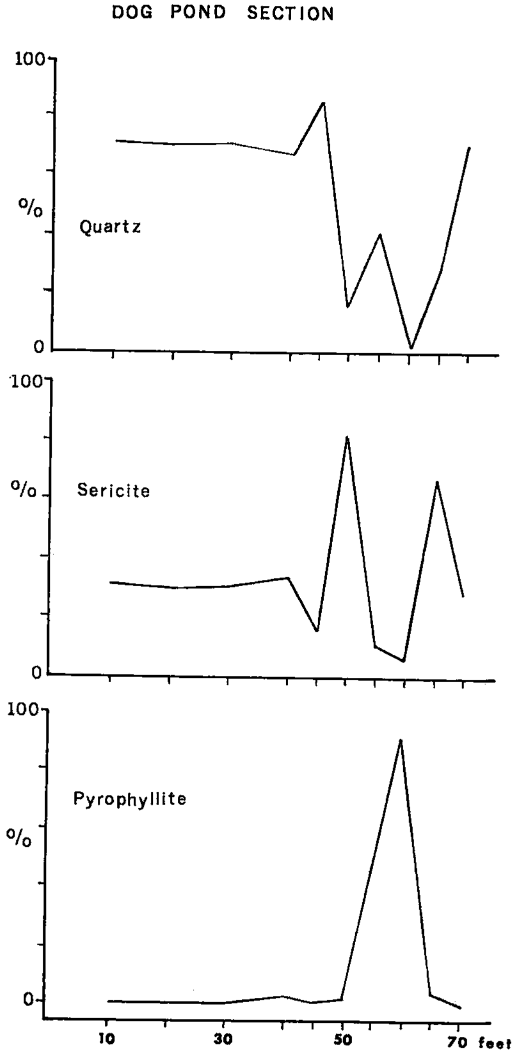

FIG. 5. Variation in mineral proportions across a narrow altered zone at Dog Pond. (Determined quantitatively from X-ray diffractograms). From Keats (1970).

a zone of intense silicification (Fig. 5). At least some of the silica moved in the colloidal state, as shown by the patches of what is now crypto- to microcrystalline quartz in the pyrophyllite rock.

(It has been pointed out to us by a reviewer that the evidence presented here is equally consistent with a simpler reaction:

$$2\ KAl_2(AlSi_3)O_{10}(OH)_2 + 6\ SiO_2 + 2\ H^+ \rightarrow 3\ Al_2Si_4O_{10}(OH)_2 + 2\ K^+$$

In view of the small amounts of kaolinite detected in some samples by XRD, the two-step process, documented experimentally, seems more realistic to us. Detailed sampling and XRD analysis of the pit walls from recognizable rhyolite to the strongly altered pyrophyllite zone is now in progress; this will either prove or disprove the existence of a distinct zone enriched in kaolinite. Until then, the direct sericite → pyrophyllite reaction must be considered as a possible alternative).

Further indication of the leaching of metal ions from the original rock and their diffusion through, and eventual concentration in, the rocks surrounding the altered zone is given by several large dark brownish patches prominent on the north wall of the Oval Pit. These patches, extending over the whole depth of the pit, are caused by a concentration of iron (as finely disseminated hematite) in the flow-banded rhyolite; this iron was clearly leached from the light yellowish pyrophyllite-sericite-quartz rock in the central part of the pit. A few thin (1 cm) veinlets of specular hematite have been found in some of these iron-rich patches.

Although it seems probable that no aluminum was added to the rock from external sources, the role of aluminum in the alteration process could not have been entirely passive. The process of leaching and large-scale removal of major chemical constituents in the presence of abundant water must necessarily involve a partial to complete disruption of the crystal lattice of the original minerals, differential movement of ions through pore fluids and fluids circulating in open fractures, and the crystallization of new minerals from the less mobile constituents. Even if this process, at any given point, were of very short duration, aluminum would have an opportunity to enter into solution. Such Al-rich solutions may have been responsible for the formation of a network of narrow (1-3 cm) veins of pure, massive, fine-grained pyrophyllite intersecting silicified rhyolite near the termination of a major shear zone on Mine Hill, 0.75 km south of the Oval Pit (Papezik 1974b), thin (1-15 mm) veinlets in iron-stained rhyolite near the bottom of the north wall of the Oval Pit which contain a core of massive pyrophyllite bordered near the walls by pyrophyllite flakes up to 3 mm across, and probably of the diaspore-pyrophyllite-barite nodules themselves.

P-T CONDITIONS OF FORMATION

The systems Al_2O_3–H_2O and Al_2O_3–SiO_2–H_2O have been studied by several investigators (Ervin & Osborn 1951; Matsushima *et al.* 1967; Althaus 1969; Haas & Holdaway 1973; Day 1974). In addition, the studies of Althaus (1966) and Tsuzuki & Mizutani (1971) provide specific data on the stability limits of pyrophyllite in acid environments. Thus the limits of temperature for the formation of the pyrophyllite-diaspore deposit at Foxtrap can be estimated, if a reliable estimate of pressure can be made independently.

The Holyrood granitoid pluton, the probable source of the aqueous fluids, is a shallow-level pluton with many roof-pendants, indicating that only the upper part of the intrusive body has been uncovered by erosion. Hughes (1971) presents evidence that at least parts of the pluton had been formed under a pressure of 1.75 kbar but argues that this represents "magmatic overpressure", and that the depth of crystallization of the granite was considerably less than the 6 km implied by this figure. Strong & Minatidis (1975) suggest a pressure of 2-5 kbar for the formation of the Holyrood plutonic series, but agree with Hughes (1971) that field evidence favors a shallow level of crystallization. Thus it seems reasonable to conclude that the pyrophyllitization of the rhyolitic rocks intruded by the pluton took place under a pressure of about 2 kbar or less. As all published equilibrium curves for pyrophyllite and diaspore are steep in the region of 2 kbar and above, even an error of 1-2 kbar would not greatly influence the temperature estimate.

The lower stability limit of pyrophyllite is given by the reaction:

$$\underset{\text{kaolinite}}{Al_2Si_2O_5(OH)_4} + 2\ SiO_2 \rightarrow \underset{\text{pyrophyllite}}{Al_2Si_4O_{10}(OH)_2} + H_2O$$

The equilibrium curve at 2 kbar in the presence of pure aqueous fluid has been placed by several authors in the region between 345°C and 380°C (Althaus 1969; Thompson 1970; Matsushima *et al.* 1967), although some investigators (Reed & Hemley 1966; Day 1976) place it as low as 300°C.

The upper stability limit in this relatively low-pressure region is given by:

$$\underset{\text{pyrophyllite}}{Al_2Si_4O_{10}(OH)_2} \rightarrow \underset{\text{andalusite}}{Al_2SiO_5} + 3\ SiO_2 + H_2O$$

Again at 2 kbar, the equilibrium temperature is given by Althaus (1969) at 487°C, but considerably lower temperatures have been proposed by others (380°C – Haas & Holdaway 1973; 365°C – Day 1976).

However, in the presence of strong acids, the stability field of pyrophyllite is shifted to much lower temperatures. According to Althaus (1966), the lower stability limit is decreased to 275°C in a 5*N* HCl solution and to 270°C in a 10*N* H_2SO_4 solution, and the upper limit is lowered to 400°C in 5*N* HCl. These data are supported by the work of Tsuzuki & Mizutani (1971), who found that in acid solutions sericite is altered to kaolinite and then to pyrophyllite at temperatures above 255°C; below this temperature, pyrophyllite does not form and kaolinite is the final product. Although Thompson (1970) did not observe similar effects in 4*N* HCl, the excellent agreement between the data of Althaus (1966) and Tsuzuki & Mizutani (1971) supports the results of these authors.

No similar data exist on the stability of diaspore in acid solutions, but experimental studies involving pure aqueous phase (e.g. Ervin & Osborn 1951; Kennedy 1959; Matsushima *et al.* 1967; Haas 1972; Day 1976) agree that the equilibrium curve for the reaction diaspore → corundum+water lies near 400°C at 2 kbar. The lower stability limit is complicated by the problem of metastable existence of boehmite (Kennedy 1959), but at pressures of about 2 kbar, the stability field of diaspore has been extended downwards to 300°C by Ervin & Osborn (1951) and to approximately 150°C by Kennedy (1959). Thus the stability fields of pyrophyllite and diaspore overlap throughout the whole temperature range.

As andalusite is absent from the Foxtrap pyrophyllite deposit but traces of kaolinite have been detected by XRD, the Foxtrap mineral assemblage lies probably in the lower part of the stability field of pyrophyllite in the presence of acid solutions. Assuming the pressure of 2 kbar or less discussed above, it may be concluded that the alteration of the rhyolite took place in a temperature range of approximately 260°–280°C, and that the temperature very likely did not exceed 300°C.

It has been stated earlier that rocks adjacent to the contacts of the granitoid pluton have been affected by low-grade thermal metamorphism characterized by an incipient development of actinolite in basic rocks. The formation of actinolite at low pressures takes place at 350° – 360°C (Nitsch 1971). However, actinolite is absent from the few basic rocks in the vicinity of the pyrophyllite deposit. The rocks, in which primary textures are well-preserved and penetrative deformation is completely lacking, show only a non-diagnostic assemblage of albite-chlorite-sphene±quartz± hematite. Although this assemblage by itself would not be significant, taken in connection with other data presented here it may indicate that the temperatures necessary for the development of actinolite were not reached in this area.

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