

## PETROLOGICAL AND GEOCHEMICAL CHARACTERISTICS OF THE PLEASANT RIDGE ZINNWALDITE – TOPAZ GRANITE, SOUTHERN NEW BRUNSWICK, AND COMPARISONS WITH OTHER TOPAZ-BEARING FELSIC ROCKS

RICHARD P. TAYLOR

Ottawa – Carleton Geoscience Centre, Department of Earth Sciences, Carleton University, Ottawa, Ontario K1S 5B6

### ABSTRACT

The Pleasant Ridge pluton and adjacent tin-mineralized cupola at Bonny River, in southern New Brunswick, were emplaced around 360 Ma, well after the cessation of tectonothermal activity associated with the Acadian Orogeny. Granites from the Pleasant Ridge and Bonny River plutons are fine- to medium-grained, equigranular to seriate textured, with essential quartz, alkali feldspar, plagioclase, lithian mica, and topaz. Accessory minerals include zircon, niobian rutile, columbite-tantalite, cassiterite, uraninite, monazite, and bastnäsite. The granites contain primary albite ( $An_{0-3}$ ) and are subsolvus in type. Both the topaz (17.6–19.4 wt.% F) and lithian mica (5.9–7.4 wt.% F) are F-rich. The lithian mica (2.84 wt.%  $Li_2O$ ) resembles zinnwaldite. The granites are weakly peraluminous ( $A/CNK = 1.0-1.2$ ) with normative ( $Qtz + Kfs + Ab$ ) contents greater than 96%, and in the  $Qtz-Ab-Or$  system with excess water they plot close to the minimum ( $T = 690^\circ C$ ) at 1 kbar with 1 wt.% added fluorine. The granites are fluorine-rich ( $F = 0.7$  to 1.0 wt.%,  $F/Cl > 70$ ), high-silica ( $SiO_2 > 75$  wt.%) types with extremely elevated contents of Li (703–1124 ppm), Rb (1444–1752 ppm), Y (79–97 ppm), Sn (9–16 ppm), Cs (28–53 ppm), Ta (11–28 ppm), W (3–27 ppm), and U (11–35 ppm), and depletions of  $TiO_2$  (<0.03 wt.%), MgO (<0.15 wt.%), CaO (<0.41 wt.%),  $P_2O_5$  (<0.01 wt.%), V, Cr, Co, Ni, Cu, and Sr (<10 ppm). Whole-rock  $\delta^{18}O$  values are nearly constant (8.1–8.6‰), whereas  $\delta D$  values have a broad range from –66 to –106‰ indicating that magmatic degassing likely occurred during emplacement. The Pleasant Ridge granite has many features in common with topaz granites from the Proterozoic (e.g., Eurajoki, Finland) and Phanerozoic (e.g., St. Austell, England); nevertheless it is distinct from some members of this group of intrusive felsic rocks in: (1) its lower  $P_2O_5$  (<0.1 wt.%) and  $Al_2O_3$  (<14.5 wt.%), and higher  $SiO_2$  (>73 wt.%) contents, and (2) its higher abundances of REE, Y, and Th. It is thus representative of a “low-P” subtype of topaz granite, which is geochemically distinct from its “high-P” ( $P_2O_5 > 0.4$  wt.%,  $Al_2O_3 > 14.5$  wt.%,  $SiO_2 < 73$  wt.%, very low REE, Y, and Th) counterparts (e.g., St. Austell).

**Keywords:** granite, topaz, fluorine, phosphorus, geochemistry, Pleasant Ridge pluton, New Brunswick.

### SOMMAIRE

Le pluton de Pleasant Ridge et la coupole minéralisée en étain de Bonny River, dans le sud du Nouveau-Brunswick, ont été mis en place il y a environ 360 million d'années, bien après la fin de l'activité tectonothermale de l'orogénèse acadienne. Ces granites présentent un grain fin à moyen et une texture équante à porphyroïde. Ce sont des granites renfermant essentiellement quartz, feldspath alcalin, plagioclase, mica lithique et topaze, avec zircon, magnétite, rutile niobifère, colombite-tantalite, cassitérite, uraninite, monazite et bastnäsite comme accessoires. Ils contiennent de l'albite primaire ( $An_{0-3}$ ) et sont donc de type subsolvus. La topaze (17,6 à 19,4% F) et le mica lithique (5,9 à 7,4% F) sont riches en fluor. Le mica lithique (2,84%  $Li_2O$ , 13,26% FeO) est proche du pôle zinnwaldite. Les granites sont moyennement hyperalumineux ( $A/CNK$  entre 1,0 et 1,2), avec un indice de différenciation ( $Qtz + Kfs + Ab$  normatifs) supérieur à 96. Dans le système  $Qtz-Ab-Or$  avec excès d'eau, ils se placent près du minimum thermique ( $T = 690^\circ C$ ) déterminé à 1 kilobar avec ajout de 1% de fluor. Géochimiquement, les granites sont riches en fluor (0,7 à 1,0% F,  $F/Cl > 70$ ) et en silice ( $SiO_2 > 75\%$  en poids), avec des teneurs extrêmement élevées en Li (703–1124 ppm), Rb (1444–1752 ppm), Y (79–97 ppm), Sn (9–16 ppm), Cs (28–53 ppm), Ta (11–28 ppm), W (3–27 ppm) et U (11–35 ppm). Ils sont très pauvres en  $TiO_2$  (<0,03%), MgO (<0,15%), CaO (<0,41%),  $P_2O_5$  (<0,01%), V, Cr, Co, Ni, Cu et Sr (<10 ppm). Les compositions isotopiques  $\delta^{18}O$  des roches totales montrent des variations limitées (8,1–8,6‰), alors que les rapports isotopiques  $\delta D$  varient largement, de –66 à –106‰ caractéristiques interprétées comme résultant d'un dégazage magmatique au cours de la mise en place. Le granite de Pleasant Ridge possède plusieurs caractères en commun avec d'autres granites à topaze mis en place aussi bien au cours du Protérozoïque (par exemple Eurajoki, Finlande) que du Phanérozoïque (par exemple St. Austell, Angleterre). Cependant, il se distingue de certains membres de ce groupe par (1) ses teneurs faibles en  $P_2O_5$  (<0,1%) et  $Al_2O_3$  (<14,5%), et élevées en  $SiO_2$  (>73%), et (2) ses teneurs plus élevées en terres rares, Y et Th. Il est ainsi représentatif d'un sous-type de granites à topaze “pauvre en phosphore”, qui se distingue géochimiquement du sous-type “riche en phosphore” ( $P_2O_5 > 0,4\%$ ,  $Al_2O_3 > 14,5\%$ ,  $SiO_2 < 73\%$ , à très faibles teneurs en terres rares, Y et Th), par exemple le granite de St. Austell.

(Traduit par la Rédaction)

**Mots-clés:** granite, topaze, fluor, phosphore, géochimie, pluton de Pleasant Ridge, Nouveau-Brunswick.

INTRODUCTION

GEOLOGICAL SETTING

Topaz granites are a unique igneous rock type that occurs only rarely in the geological record; their nature and origin are poorly understood. This study presents: (1) a comprehensive account of the petrological and geochemical characteristics of an occurrence of lithian mica - topaz granite in the Pleasant Ridge pluton in southern New Brunswick, and (2) a comparison of the characteristic features of the Pleasant Ridge topaz granite with those of topaz granites from localities in Europe and Asia.

A series of small, metaluminous to weakly peraluminous, granite plutons intrude Lower Paleozoic rocks adjacent to the northwest margin of the St. George batholith in southern New Brunswick (Fig. 1). The four largest (2 to 15 km<sup>2</sup>) of the bodies are designated, from west to east, the Tower Hill, Sorrel Ridge, Pleasant Ridge, and Beech Hill plutons (Fig. 2). These plutons have been collectively termed the "Beech Hill Series" by Dagger (1972). However, there exists a clear

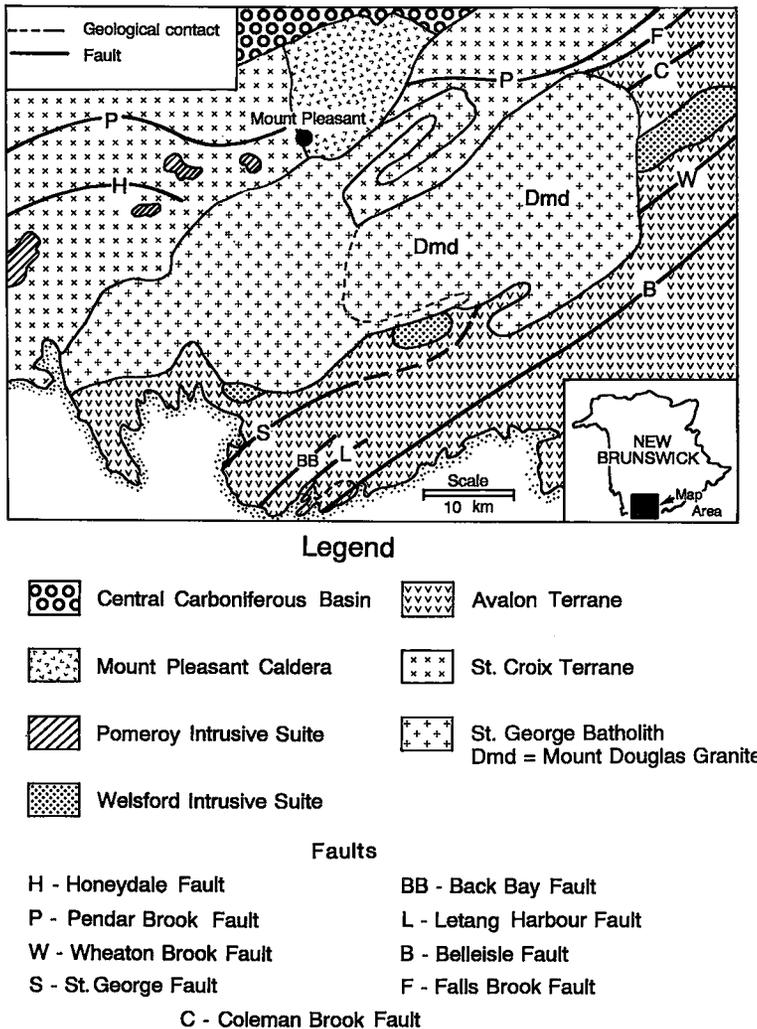


FIG. 1. Location map and tectonic setting of the Pomeroy Intrusive Suite, of which the Pleasant Ridge and Bonny River granites are members, and the adjacent Saint George batholith (after McLeod 1990).

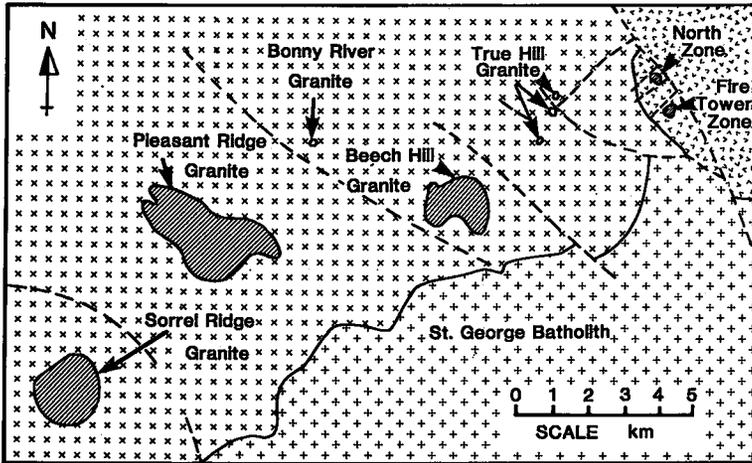


FIG. 2. Local geology of the Pleasant Ridge - Mount Pleasant area (after Ruitenbergh 1967, McLeod 1990), showing the locations of the granites that comprise the Pomeroy Intrusive Suite. Legend as in Figure 1.

division of ages within this group. The Tower Hill pluton is excluded because of its earlier age of emplacement, Silurian as opposed to Late Devonian for the more easterly plutons (Butt 1976, Taylor *et al.* 1985, Sinclair *et al.* 1988). Thus the terminology adopted in this paper follows that of McLeod (1990), who referred to the younger group of plutons, that includes the Pleasant Ridge granite, as the Pomeroy Intrusive Suite.

The plutons of the Pomeroy Intrusive Suite have been interpreted to represent a group of post-Acadian granites of epizonal to mesozonal character (Butt 1976, Ruitenbergh *et al.* 1977). They have been the focus of Sn-W exploration in recent years because of their proximity to the large deposits of W-Mo in the Fire Tower Zone (Kooiman *et al.* 1986) and Sn in the North Zone (Sinclair *et al.* 1988) at Mount Pleasant (Fig. 2). These mineral deposits are underlain by a suite of geochemically distinctive granites (Taylor *et al.* 1985, 1989, Kooiman *et al.* 1986). Two partially unroofed tin-mineralized cupolas have been discovered recently, as a result of mineral exploration at Bonny River and True Hill (Fig. 2). Tin-tungsten-bearing veins and associated greisen alteration are present in all representatives of the Pomeroy Intrusive Suite, and also occur in the Mount Douglas Intrusive Suite (McLeod 1990), which comprises the eastern half of the adjacent Saint George batholith (Fig. 1).

On the basis of gravity profiles, derived from geophysical data contained in Thomas & Willis (1989), and the results of recent geological mapping, McLeod (1990) concluded that: (1) the

Mount Douglas Granite was emplaced mainly into Lower Paleozoic sedimentary rocks, crystallized at very high crustal levels, and extends to a maximum depth of 7 km below the surface, and (2) the roughly contemporaneous Pomeroy Intrusive Suite, which is located along the northwestern margin of the main batholith (Figs. 1, 2), occupies a similar position and may represent a direct lateral extension of the batholith below the Paleozoic country rocks (McLeod 1990).

The country rock to the granites in the St. Stephen - Mount Pleasant area consists of Cambro-Ordovician quartzite and graphitic slate of the Cookson Formation, which are unconformably overlain by a sequence that includes: conglomerate of the Oak Bay Formation, turbiditic greywacke and slate of the Silurian Waweig and Digdeguash formations, and calcareous sandstone, slate and phyllite of the Upper Silurian - Lower Devonian Flume Ridge Formation. All of these lithologies have been affected by Acadian polyphase deformation (Ruitenbergh *et al.* 1977, Fyffe *et al.* 1981). From west to east, there is a gradual change in the level of emplacement of the granites; the Sorrel Ridge Granite (Fig. 2) intrudes Ordovician slate of the Cookson Formation and has produced a contact metamorphic aureole of cordierite-biotite grade, whereas all of the more easterly plutons were emplaced into phyllite and greywacke of the Silurian Waweig Formation and have a narrow aureole of biotite hornfels (Ruitenbergh 1967, Butt 1976). The intrusive rocks that host the Sn-W-Mo deposits, in the Fire Tower Zone and North Zone at Mount Pleasant (Figs. 1, 2), are subvolcanic in

character (Kooiman *et al.* 1986) and are located close to the southwestern margin of the Mount Pleasant caldera (Fig. 1), an area of gently dipping Late Devonian volcanic and sedimentary rocks termed the Piskahegan Group (McCutcheon 1986).

Although the field relationships permit some broad constraints to be placed upon the timing of granite emplacement (between Late Silurian and Mississippian), some uncertainty still exists regarding the absolute ages of emplacement and crystallization of the individual plutons. Butt (1976) presented an Rb–Sr isochron age of  $422 \pm 15$  Ma for the Tower Hill Stock; Rb–Sr isochron ages of  $349 \pm 11$  Ma (Butt 1976) and  $337 \pm 15$  Ma (Fyffe *et al.* 1981) have been reported for the Beech Hill Stock. No published geochronological data are currently available for the other plutons; however, the unpublished results of a program of  $^{40}\text{Ar}/^{39}\text{Ar}$  incremental heating experiments yield plateau ages of 361 and 362 Ma, respectively, for the Pleasant Ridge and Bonny River granites (D.A. Archibald, pers. comm.), confirming their emplacement in the Late Devonian. Slightly older  $^{40}\text{Ar}/^{39}\text{Ar}$  ages (*ca.* 367 Ma) have been obtained for biotite from the Mount Douglas Granite (McLeod *et al.* 1988) (Fig. 1).

Based solely upon petrological criteria, Taylor *et al.* (1985) subdivided the plutons of the study area, and related granites from the area of Mount Pleasant (Figs. 1, 2), into four groups: (1) biotite–muscovite-bearing (Tower Hill), (2) biotite-bearing (Sorrel Ridge, Beech Hill, True Hill), (3) fluorite- and topaz-bearing (Mine North Zone intrusive bodies), and (4) lithian mica–topaz-bearing (Pleasant Ridge and Bonny River). These are considered to be the first reported occurrences of topaz granite in the northern Appalachians. Subsequently, more detailed studies of the North Zone at Mount Pleasant (Sinclair *et al.* 1988, Taylor *et al.* 1989), and the Pleasant Ridge and Bonny River plutons (this paper), have clearly demonstrated: (1) an intimate spatial association of important zones of hydrothermal tin mineralization with topaz granite in the North Zone at Mount Pleasant, and the Bonny River cupola, and (2) a Late Devonian age (*ca.* 360 Ma) of emplacement for topaz granite at both localities. A similar relationship has also been recognized at East Kemptville, in Nova Scotia, where a *ca.* 370 Ma topaz leucogranite body is host to a large deposit of tin that contains 56 million tonnes at 0.165% Sn (Kontak 1990). In combination, these occurrences indicate that an important metallogenic episode, involving the generation of felsic magmas that crystallized as topaz granite and of related tin deposits, occurred in the northern Appalachians in the Late Devonian.

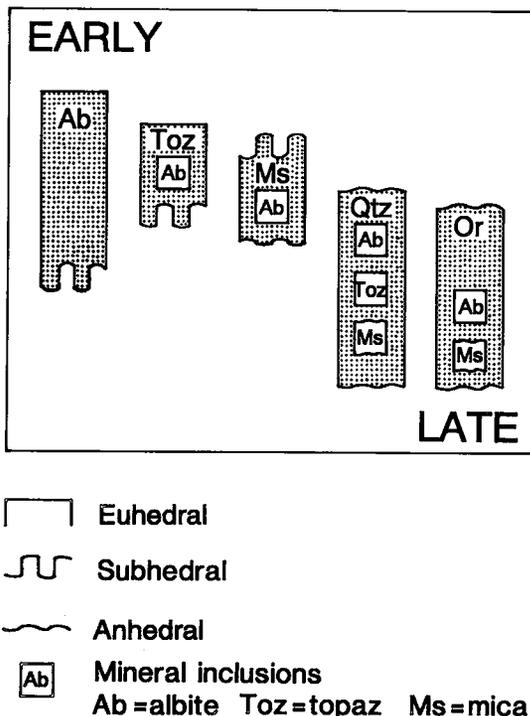
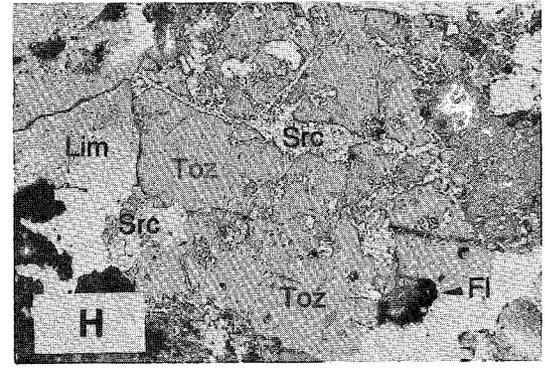
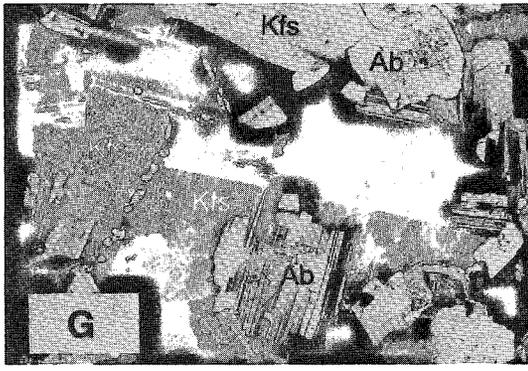
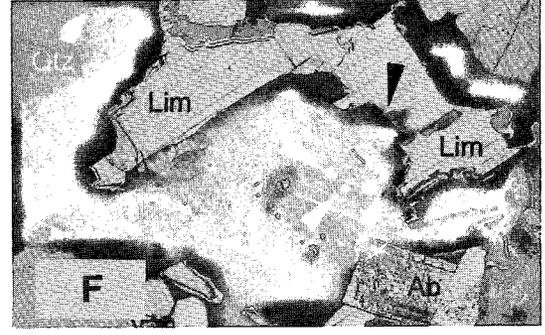
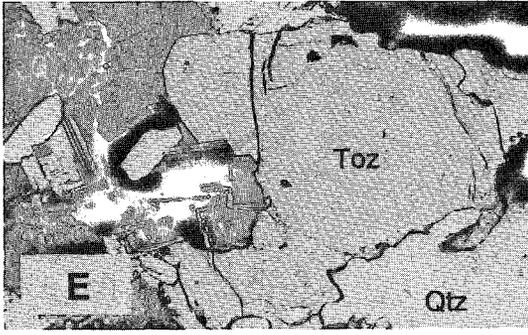
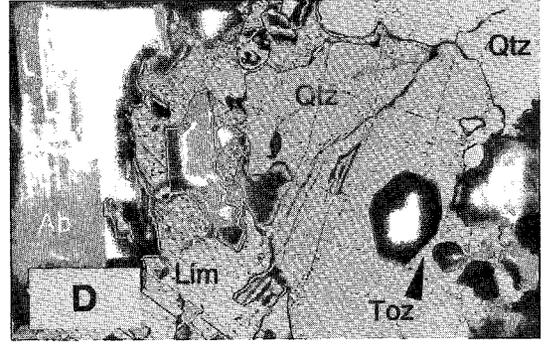
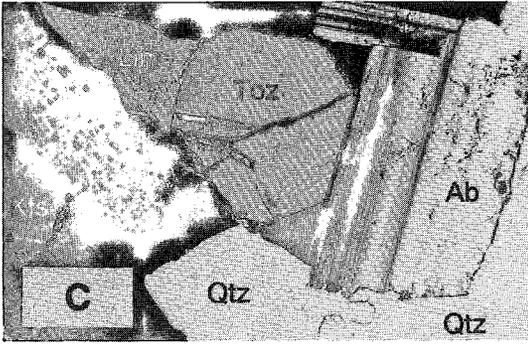
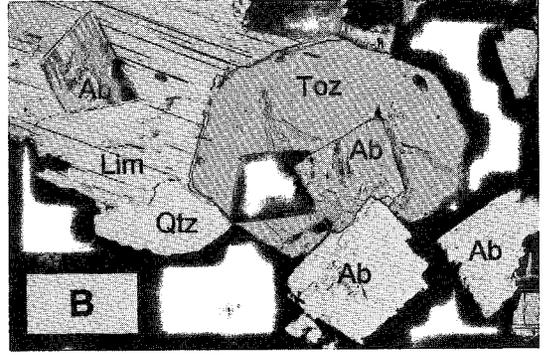
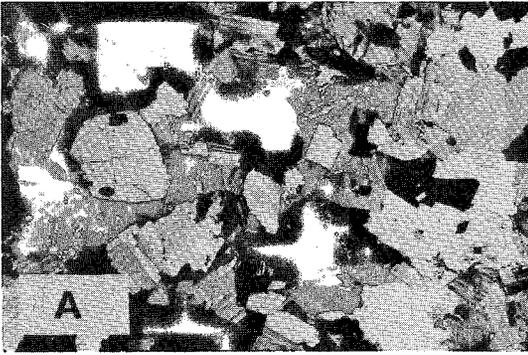


FIG. 3. Schematic sequence of crystallization for the Pleasant Ridge granite.

FIG. 4. Photomicrographs showing characteristic textural features of the essential and accessory minerals in the Pleasant Ridge granite (all taken using crossed nicols). The field of view is 6 mm in Fig. 4A, and 3 mm in Figs. 4B through 4H: (A) hypidiomorphic-granular to seriate texture; (B) euhedral inclusions of albite in early, euhedral topaz, and later subhedral lithian mica; (C) early, euhedral topaz; (D) euhedral topaz as inclusions (see arrow) in late, interstitial quartz; (E) topaz grain exhibiting a lateral change in growth habit from euhedral (early) to anhedral and embayed (late); (F) subhedral, interstitial lithian mica grains with several minute inclusions of accessory minerals with radioactive haloes (marked with arrows); (G) late, interstitial K-feldspar with inclusions of euhedral albite; (H) alteration of magmatic topaz to sericite + fluorite in altered topaz granite from the Bonny River cupola. Abbreviations are: Ab albite, Toz topaz, Lim lithian mica, Qtz quartz, Kfs K-feldspar, Src sericite, Fl fluorite.



## PETROGRAPHY AND MINERAL CHEMISTRY

From observations of surface outcrop and diamond drill-core, the Pleasant Ridge pluton (Lat. 45°26' N, Long. 66°49' W) and the nearby cupola at Bonny River (Fig. 2) appear to be composed of essentially one rock type, at the current level of erosion: a medium- to fine-grained, alkali feldspar granite (Streckeisen 1973). The rock is equigranular to seriate in texture, and grey-white in color. Xenoliths of igneous or sedimentary character have not been observed. The minerals present in the granite have been identified by optical microscopy, X-ray diffraction (XRD), and scanning electron microscopy with energy-dispersion analysis (SEM-ED). All quantitative data concerning mineral compositions have been produced by electron-microprobe analysis using wavelength-dispersion analysis (EMPA-WD), with the exception of the data for lithium, obtained by atomic absorption spectrometry (AAS), ferrous iron (by titration), and water (by combustion, with infrared detector, CIRD, or mercury manometry). Details of the operating conditions and analytical standards used for electron-microprobe analysis are contained in the footnotes to Tables 1, 2, and 3.

Essential minerals in the Pleasant Ridge granite are, in order of decreasing abundance, quartz, plagioclase and K-feldspar, present in subequal amounts, lithian mica, and topaz. On the basis of textural relationships, the granite contains primary albite. A schematic sequence of crystallization for the Pleasant Ridge granite is illustrated in Figure 3, which also serves to summarize the most important textural features. In deducing the sequence of crystallization, use has been made of inclusion relations, reaction relations, and the shape of individual grains. It has been assumed that small euhedral inclusions in the center of a larger grain nucleated no later than the enclosing grain, and that crystals growing suspended in a magma are likely to grow as euhedra, whereas during the later stages of magmatic crystallization, crystals will mutually interfere, forming anhedral grains and interstitial fabrics.

Albite was the first mineral to crystallize (*i.e.*, the liquidus phase) (Fig. 3). It typically occurs as individual fine- to medium-grained euhedral crystals (Fig. 4A) and as fine-grained euhedral inclusions in all of the other essential minerals (*e.g.*, in topaz and lithian mica, Fig. 4B). Electron-microprobe analyses of grains of albite from a variety of different textural settings (Table 1) indicate that its composition is quite uniform ( $An_{0-3}$ ). The presence of albite as a discrete magmatic mineral, together with K-feldspar, in the Pleasant Ridge granite, supports its designation as a subsolvus granite *s.s.* (Tuttle & Bowen 1958). The

TABLE 1. COMPOSITION OF ALKALI FELDSPAR (Kfs) AND PLAGIOCLASE (Pl) IN THE PLEASANT RIDGE GRANITE AND THE ST. AUSTELL PLUTON

Sample Texture	Pleasant Ridge granite											
	Kfs1 IA (N=4)	Kfs2 IA (N=3)	Kfs3 IS (N=6)	Kfs4 IA (N=3)	P17 SUJ (N=3)	P18 SUJ (N=3)	P19 SUJ (N=2)	P13 IEU (N=3)	P12 IEU (N=2)	P11 IEU (N=2)	P12 IEU (N=2)	
Concentrations in weight percent												
SiO <sub>2</sub>	64.81	65.02	64.87	64.49	67.93	67.49	67.72	67.66	67.87	67.81	67.51	
Al <sub>2</sub> O <sub>3</sub>	18.85	18.82	18.76	18.51	19.87	19.77	20.05	20.12	20.34	20.34	20.06	
CaO	0.00	0.00	0.00	0.00	0.25	0.52	0.28	0.00	0.13	0.12	0.32	
Na <sub>2</sub> O	0.84	1.44	1.58	0.48	11.59	11.41	11.58	11.77	11.72	11.72	11.53	
K <sub>2</sub> O	15.68	14.70	14.41	16.02	0.20	0.30	0.17	0.23	0.26	0.20	0.20	
P <sub>2</sub> O <sub>5</sub>	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
Total	100.16	100.08	99.81	99.50	99.44	99.49	100.38	99.76	100.32	99.82		
Number of ions on the basis of 32 (O)												
Si	11.935	11.940	11.856	11.968	11.939	11.881	11.806	11.867	11.848	11.861	11.861	
Al	4.091	4.086	4.076	4.049	4.076	4.104	4.282	4.157	4.184	4.164	4.164	
Ca	—	—	—	—	0.047	0.089	0.050	—	0.024	0.022	0.052	
Na	0.300	0.513	0.563	0.171	3.881	3.896	3.814	4.003	3.966	3.928	3.928	
K	3.679	3.444	3.388	3.792	0.046	0.067	0.040	0.052	0.058	0.043	0.043	
P	—	—	—	—	—	—	—	—	—	—	—	

Sample Texture	St. Austell pluton											
	Kfs1 IA (N=3)	Kfs2 IA (N=2)	Kfs3 IA (N=1)	Kfs4 IA (N=3)	P12 IA (N=3)	P13 IEU (N=3)	P14 IEU (N=3)	P15 IEU (N=3)	P12 IEU (N=4)			
Concentrations in weight percent												
SiO <sub>2</sub>	64.29	64.57	64.34	64.22	63.60	68.49	68.14	68.13	68.29	67.90		
Al <sub>2</sub> O <sub>3</sub>	18.80	18.82	18.70	18.53	18.84	18.73	19.67	20.03	19.84	19.77		
CaO	0.00	0.00	0.00	0.00	0.03	0.05	0.13	0.22	0.12	0.25		
Na <sub>2</sub> O	0.44	0.61	0.78	0.52	6.62	11.66	11.52	11.67	11.64	11.51		
K <sub>2</sub> O	15.96	15.77	15.41	15.83	15.77	0.21	0.17	0.13	0.14	0.18		
P <sub>2</sub> O <sub>5</sub>	0.36	0.54	0.39	0.48	0.63	0.28	0.24	0.36	0.21	0.43		
Total	99.85	100.31	99.82	99.38	99.29	100.42	99.87	100.54	100.04	99.44		
Number of ions on the basis of 32 (O)												
Si	11.870	11.861	11.891	11.901	11.820	11.914	11.918	11.843	11.823	11.832		
Al	4.092	4.075	4.073	4.046	4.094	4.044	4.054	4.103	4.042	4.097		
Ca	—	—	—	—	0.006	0.010	0.026	0.041	0.023	0.047		
Na	0.157	0.216	0.279	0.188	2.224	3.831	3.906	3.934	3.942	3.924		
K	3.758	3.695	3.633	3.695	0.379	0.047	0.038	0.028	0.031	0.041		
P	0.056	0.084	0.061	0.075	0.089	0.041	0.035	0.053	0.031	0.084		

Note. Analytical procedure: EMPA-WD, 15 kV, 20 nA, using a beam defocused to about 20 µm, and natural secondary (for Si, Al, K), albite (for Na), diopside (for Ca), and apatite (for P) standards. N = number of spot analyses. Abbreviations are: IA = interstitial, anhedral K-feldspar; IS = interstitial, subhedral K-feldspar; IEU = included, euhedral plagioclase; SUJ = subhedral, interstitial plagioclase.

plagioclase from other topaz granites also is characterized by its very low An contents (*e.g.*, Eurajoki, Väkkärä granite:  $An_{0-5}$ , Haapala 1977; St. Austell:  $An_2$ , Manning & Hill 1990; Tregonning granite:  $An_{2-3}$ , Stone 1975; Meldon aplite:  $An_{1-5}$ , Chaudhry 1971) and its lack of compositional zoning. Topaz was the next mineral to crystallize in the Pleasant Ridge granite (Fig. 3). Early-formed crystals of topaz, like those of albite, occur both as fine- to medium-grained single euhedral crystals (Fig. 4C), and as fine-grained euhedral inclusions in lithian mica, quartz (Fig. 4D), and K-feldspar. However, topaz commonly also exhibits an anhedral habit, and it is not uncommon for single grains to show a combination of these habits (Fig. 4E). In the latter case, optically continuous single grains exhibit a lateral change in habit from euhedral to anhedral and poikilitic (Fig. 4E). In such cases, the contacts typically represent the grain boundaries between topaz and quartz. Both the euhedral and anhedral types of topaz in the Pleasant Ridge granite have high fluorine contents that range from 17.79 to 19.04 wt.% (Table 2), close to the theoretical maximum for the mineral (*i.e.*, 20.7 wt.%: Deer *et al.* 1966). Such high-F

**TABLE 2. COMPOSITION OF TOPAZ IN THE PLEASANT RIDGE GRANITE (PRG) AND IN TOPAZ GRANITE FROM THE EURAJOKI (EP), ST. AUSTELL (SAP), AND MOUNT PLEASANT (MP) PLUTONS**

	PRG/2 EEU (N=3)	PRG/5 EEU (N=6)	PRG/3 LA (N=2)	PRG/6 LA (N=4)	SAP/1 EEU (N=6)	SAP/2 SU (N=4)
Concentrations in weight percent						
SiO <sub>2</sub>	32.82	33.22	33.65	32.89	32.82	32.74
Al <sub>2</sub> O <sub>3</sub>	56.05	56.24	55.85	55.93	55.99	56.04
F	18.42	17.79	18.84	19.04	18.06	18.14
-O=F	-7.76	-7.49	-7.93	-8.02	-7.60	-7.64
Total	99.53	99.76	100.41	99.84	99.27	99.28
Number of ions on the basis of 24 (O,OH,F)						
Si	4.032	4.094	4.125	4.043	4.027	4.017
<sup>VI</sup> Al	—	—	—	—	—	—
<sup>IV</sup> Al	8.113	8.116	8.066	8.101	8.096	8.103
F	7.162	6.930	7.300	7.398	7.005	7.036
Concentrations in weight percent						
SiO <sub>2</sub>	32.92	32.64	32.63	32.33	32.73	32.48
Al <sub>2</sub> O <sub>3</sub>	56.08	55.66	55.72	56.21	56.13	56.21
F	18.13	18.82	18.66	19.29	19.19	19.78
-O=F	-7.63	-7.92	-7.86	-8.12	-8.08	-8.33
Total	99.50	99.20	99.15	99.71	99.97	100.14
Number of ions on the basis of 24 (O,OH,F)						
Si	4.046	3.995	3.994	3.969	4.027	3.986
<sup>VI</sup> Al	—	0.005	0.006	0.031	—	0.014
<sup>IV</sup> Al	8.121	8.022	8.031	8.101	8.136	8.114
F	7.043	7.261	7.220	7.487	7.462	7.673

Note. Analytical method: EMPA-WD, 15 kV, 20 nA, analytical standard used was fluoritopaz (Barton 1982). N = the number of spot analyses. Abbreviations are: EEU = early, euhedral topaz; IEU = included, euhedral topaz; SU = subhedral topaz; LA = late, anhedral topaz.

topaz is similar to that from other topaz granites, such as the non-megacrystic lithian mica granite (NMLG) phase of the St. Austell pluton, the Väkkärä granite phase of the Eurajoki pluton, and those of the North Zone at Mount Pleasant (Table 2). The H<sub>2</sub>O content of topaz in the Pleasant Ridge granite (measured by mercury manometry during hydrogen isotope analysis; Taylor, unpubl. data) is very low, with a mean value of H<sub>2</sub>O<sup>+</sup> (>130°C) of 0.13 wt.%. Such a value is very similar to the range of values recorded for topaz in topaz rhyolites (0.06 to 0.11 wt.%: Foord *et al.* 1991).

The mica is a lithium- and iron-bearing (Li<sub>2</sub>O,  $\bar{x}$  = 2.84 wt.%, N = 3, analyzed by AAS; FeO,  $\bar{x}$  = 13.40 wt.%, N = 8, analyzed by titration), fluorine-rich type (F up to 7.27 wt.%, Table 3). It exhibits many of the textural features of topaz, which is consistent with their close association in the sequence of crystallization (Fig. 3), although fine-grained euhedral inclusions of lithian mica have only been identified in quartz and K-feldspar. Early lithian mica occurs as individual fine- to medium-grained subhedral grains (Fig. 4F) or as

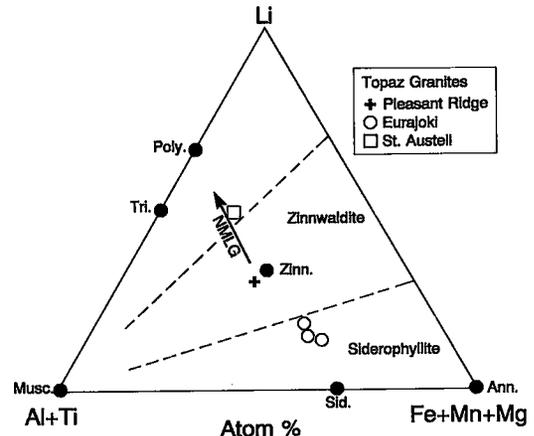


FIG. 5. Plot of occupancy of the octahedral sites in terms of Li - (Al + Ti) - (Fe + Mn + Mg) (atomic) after Henderson *et al.* (1989) for averaged microprobe data of Pleasant Ridge granite (PRG), non-megacrystic lithian mica granite (NMLG) of St. Austell pluton, and the Väkkärä granite of the Eurajoki pluton (Table 3), with Li determined by AAS and ferrous iron by titration. Abbreviations are: Poly polyolithionite, Tri trilithionite, Zinn zinnwaldite, Musc muscovite, Sid siderophyllite, Ann annite. Boundaries demarcating fields of lepidolite, zinnwaldite, and siderophyllite are from Stone *et al.* (1988).

fine-grained subhedral inclusions in quartz and K-feldspar (Fig. 4G). Later mica is typically fine-grained, anhedral, and occurs interstitially to quartz and feldspar. In chemical composition, the mica closely resembles zinnwaldite from the type locality (*cf.* Deer *et al.* 1971), and also the lithian micas of the "non-megacrystic Li-mica granite" (NMLG) phase of the St. Austell pluton (Henderson *et al.* 1989). In the Fe-bearing, lithian mica series described by Henderson *et al.* (1989), the main mechanism of substitution is <sup>VI</sup>Li + <sup>IV</sup>Si = <sup>VI</sup>(Fe<sup>2+</sup>) + <sup>IV</sup>Al, with the occupancy of octahedral sites being maintained at 6.0 atoms. The lithian micas from the NMLG phase of the St. Austell pluton thus define an evolutionary trend characterized, in terms of the occupancy of octahedral sites, by increasing Li and decreasing Fe to produce the mineral series zinnwaldite - lepidolite (Fig. 5). The lithian micas at Pleasant Ridge plot at the Fe-rich end of the NMLG trend, close to the composition of ideal zinnwaldite (Fig. 5).

The non-megacrystic lithian mica granite unit of the St. Austell pluton has been termed "topaz granite" by Manning & Hill (1990). It is a medium- to fine-grained rock characterized by the presence

TABLE 3. COMPOSITION OF MICA IN THE PLEASANT RIDGE GRANITE (PRG) AND IN TOPAZ GRANITE FROM THE ST. AUSTELL (SAP) AND EURAJOKI (EP) PLUTONS

Sample Texture	PRG/6 SU (N=4)	SAP/1 SU (N=4)	SAP/2 IA (N=5)	SAP/3 IA (N=3)	EP/1 -- (N=1)	EP/2 -- (N=1)	EP/3 -- (N=1)
Concentrations in weight percent							
SiO <sub>2</sub>	43.84	49.21	49.54	50.20	39.43	39.12	40.30
TiO <sub>2</sub>	0.21	0.26	0.26	0.25	0.73	0.59	0.64
Al <sub>2</sub> O <sub>3</sub>	21.86	20.34	20.36	19.66	19.43	20.80	20.13
Fe <sub>2</sub> O <sub>3</sub>	na	na	na	na	5.08	2.77	4.04
FeO	13.93	7.64	7.57	7.63	17.68	19.04	16.53
ZnO	0.00	0.04	0.03	0.07	0.44	0.42	0.40
MnO	1.40	0.72	0.73	0.75	0.91	1.00	0.95
MgO	0.00	0.16	0.16	0.18	0.11	0.08	0.10
CaO	0.02	0.04	0.04	0.02	0.18	0.06	0.08
Li <sub>2</sub> O	2.84	4.94	5.13	5.07	1.18	1.36	1.63
Na <sub>2</sub> O	0.19	0.22	0.22	0.16	0.27	0.22	0.18
K <sub>2</sub> O	9.84	9.91	9.90	10.06	9.14	9.30	9.22
Rb <sub>2</sub> O	0.60	1.08	1.15	1.15	0.76	0.73	0.90
F	7.27	7.74	7.43	7.70	4.30	4.83	4.90
H <sub>2</sub> O	0.55	0.63	0.59	0.88	1.84	0.88	1.55
-O=F	-3.06	-3.26	-3.13	-3.24	-1.81	-2.04	-2.06
Total	99.49	99.67	99.98	100.54	99.67	99.26	99.49
Number of ions on the basis of 24 (O,OH,F)							
Si	6.371	6.818	6.847	6.879	5.99	6.01	6.07
<sup>VI</sup> Al	1.629	1.182	1.153	1.121	2.01	1.99	1.93
<sup>IV</sup> Al	2.114	2.139	2.164	2.053	1.47	1.77	1.64
Ti	0.023	0.027	0.027	0.026	0.08	0.07	0.07
Fe <sup>3+</sup>	---	---	---	---	0.58	0.32	0.46
Fe <sup>2+</sup>	1.692	0.885	0.875	0.874	2.25	2.45	2.08
Zn	---	0.004	0.003	0.007	0.05	0.05	0.05
Mn	0.172	0.085	0.086	0.087	0.12	0.13	0.12
Mg	---	0.033	0.033	0.037	0.03	0.02	0.02
Ca	0.003	0.006	0.006	0.003	0.03	0.01	0.01
Li	1.659	2.762	2.851	2.793	0.72	0.84	0.99
Na	0.053	0.059	0.059	0.043	0.08	0.07	0.05
K	1.823	1.751	1.745	1.758	1.77	1.82	1.77
Rb	0.056	0.097	0.102	0.101	0.07	0.07	0.09
OH	0.532	0.582	0.544	0.804	1.86	1.00	1.56
F	3.339	3.390	3.247	3.336	2.07	2.35	2.33

Note. Analytical procedure: EMPA-WD, 15 kV, 20 nA, standard used was F-rich phlogopite. N = number of spot analyses. Li<sub>2</sub>O = lithium analysis of mineral separates by AAS. Abbreviations are: SU = subhedral mica; IA = interstitial, anhedral mica; na = not analyzed. Data on mica separates from Eurajoki are from Haapala (1977, Table 8, p.91).

of essential euhedral to subhedral topaz in addition to lithian mica, albite, orthoclase, and quartz. Plots of the occupancy of octahedral sites of primary lithian micas from the Pleasant Ridge and St. Austell plutons (Table 3), and the Eurajoki pluton (Haapala 1977), serve to illustrate the broad range of compositional variations found in lithian mica from topaz granites (Fig. 5): from lithian siderophyllite (Eurajoki) through zinnwaldite (Pleasant Ridge) to zinnwaldite → lepidolite (St. Austell). In addition to their high concentrations of lithium (up to 6.4 wt. % in lepidolite), mica in topaz granites is also notable for having high abundances of Mn, Rb and Cs (Table 3: this study, Chaudhry & Howie 1973, Du Bray *et al.* 1988, Haapala 1977, Henderson *et al.* 1989, Štemprok & Sulcek 1969).

Quartz and K-feldspar occur as medium-sized

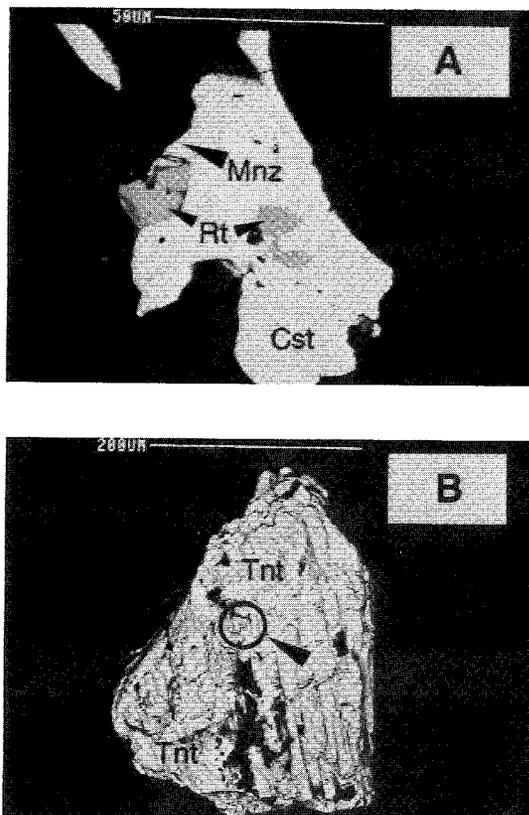


FIG. 6. Back-scattered electron photomicrographs of accessory minerals from the Pleasant Ridge granite: (A) monazite, cassiterite, and zoned niobian rutile in a complex mineral intergrowth hosted by a grain of lithian mica, from polished thin-section C82-5C; (B) large (>200 μm) grain of tantalite with very small (<10 μm) grains of cassiterite and uraninite adhering to its surface, from a heavy mineral separate (>3.3 g/cm<sup>3</sup>). The circle in (B) shows the location of the cassiterite grains illustrated in (C). Abbreviations are: Mnz monazite, Cst cassiterite, Rt rutile, and Tnt tantalite.

grains (up to 5 mm), whose anhedral shape and interstitial location support their late position in the sequence of crystallization (Fig. 3). Euhedral inclusions of any of the early-formed albite, topaz, and zinnwaldite are common in both minerals (Fig. 4). The quartz typically does not have undulose extinction or subgrain development or show any other evidence of strain. The K-feldspar commonly exhibits simple Carlsbad twinning and the development of microscopic string perthite (Fig. 4G), and has an average Or content of 86 mole % (Table 1). Occasional grains show microcline twinning.

Accessory minerals in the Pleasant Ridge granite account for a very small proportion (<0.01 vol.%)

of the rock; however, their diversity and unusual compositions make their distributions highly significant. Common accessory minerals (identified by SEM-ED analysis) include zircon, a niobium-rich variety of rutile, columbite-tantalite, cassiterite, and monazite; bastnäsite and uraninite occur more rarely. The accessories typically occur as small inclusions (<100  $\mu\text{m}$ ; Fig. 6) in zinnwaldite, where their presence may be highlighted by the existence of a radioactive halo (Fig. 4F); less commonly, they are found intergrown with quartz and topaz. Other than their diversity, what is notable about many of the accessory minerals is the complex nature of their intergrowths, and their very fine grain-size

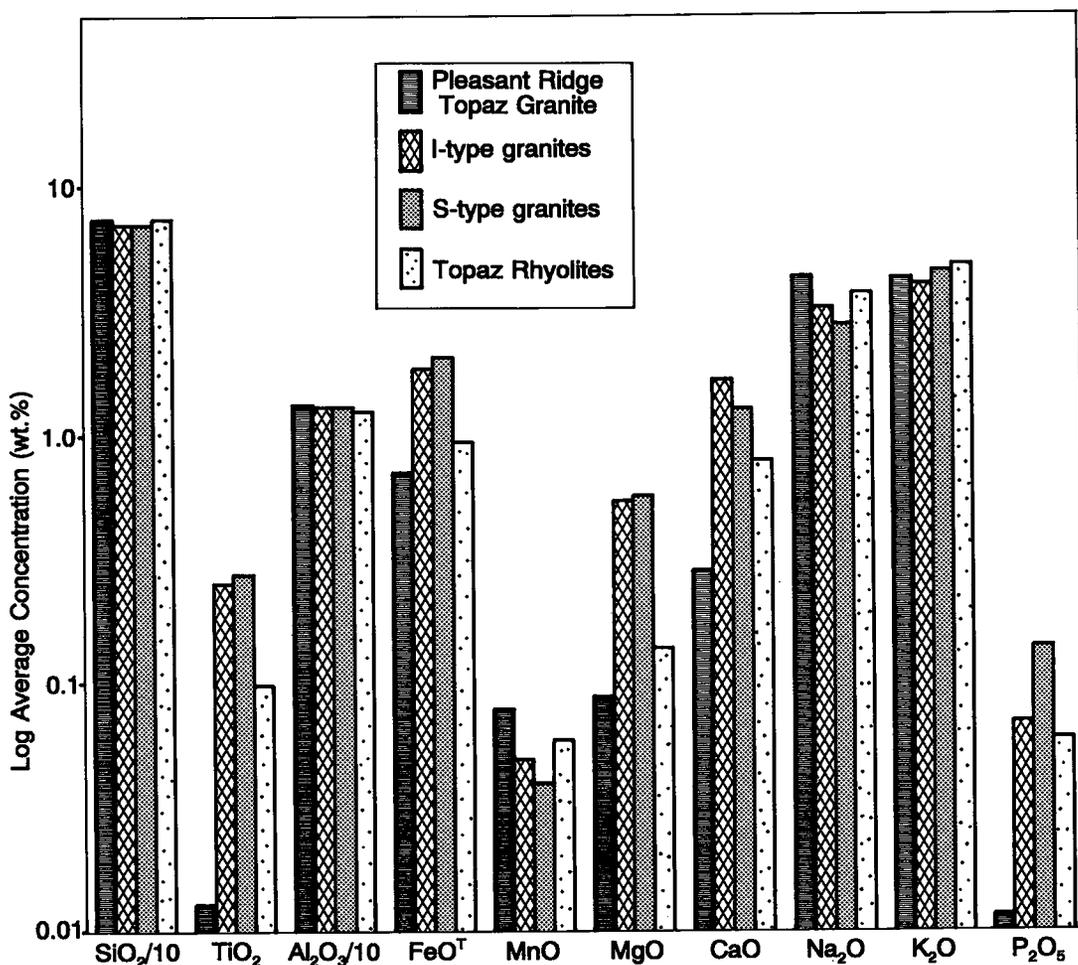


FIG. 7. Histograms of the average major-element abundances (in wt.%) of topaz granite from the Pleasant Ridge pluton (Table 4), felsic I- and S-type granites (data from Whalen *et al.* 1987), and topaz rhyolites (data from Christiansen *et al.* 1984).

**TABLE 4. WHOLE-ROCK GEOCHEMICAL DATA ON THE PLEASANT RIDGE  
AND BONNY RIVER GRANITES AND OTHER TOPAZ GRANITES**

Sample Pluton	82-4A PRG	82-4B PRG	82-4C PRG	82-5A PRG	82-5B PRG	82-5C PRG	4-151 BRG	2273 SA	1160 SA	1091 SA	256/73 EG	149/70 EG	MP-05 MP
Major element concentrations in weight percent													
SiO <sub>2</sub>	75.84	75.06	75.01	75.11	75.64	75.11	74.89	72.71	72.47	71.46	74.94	74.83	75.96
TiO <sub>2</sub>	0.02	0.01	0.01	0.02	0.01	0.01	0.03	0.04	0.04	0.03	0.02	0.02	0.03
Al <sub>2</sub> O <sub>3</sub>	13.39	13.46	13.77	13.79	13.26	13.52	13.94	15.51	15.23	16.00	13.77	14.06	13.12
Fe <sub>2</sub> O <sub>3</sub> <sup>T</sup>	0.79	0.79	0.80	0.70	0.82	0.82	1.03	0.61	0.65	1.04	1.07	1.17	1.44
MnO	0.08	0.09	0.09	0.07	0.08	0.07	0.09	0.08	0.08	0.09	0.05	0.04	0.07
MgO	0.08	0.05	0.11	0.09	0.07	0.15	0.06	0.19	0.18	0.18	0.09	0.06	0.05
CaO	0.26	0.30	0.26	0.32	0.26	0.32	0.41	0.36	0.34	0.36	0.68	0.64	0.36
Li <sub>2</sub> O	0.20	0.22	0.24	0.19	0.20	0.20	0.15	0.31	0.36	0.51	0.05	0.10	0.11
Na <sub>2</sub> O	4.43	4.25	4.36	4.59	4.14	4.40	4.20	4.64	3.29	3.78	3.65	3.67	4.08
K <sub>2</sub> O	4.34	4.24	4.26	4.27	4.32	4.25	4.50	4.07	4.98	4.04	4.78	4.34	4.38
Rb <sub>2</sub> O	0.18	0.18	0.19	0.16	0.18	0.16	0.16	0.15	0.16	0.22	0.12	0.11	0.13
H <sub>2</sub> O <sup>T</sup>	0.1	0.2	0.2	0.2	0.2	0.2	0.3	0.3	0.6	0.5	0.4	0.5	0.2
F	0.70	0.98	0.91	0.70	0.76	0.69	0.74	1.05	1.21	1.55	1.06	0.90	0.59
P <sub>2</sub> O <sub>5</sub>	0.01	0.00	0.00	0.00	0.01	0.01	0.00	0.54	0.51	0.73	0.01	0.01	0.00
CO <sub>2</sub>	0.0	0.1	0.1	0.0	0.1	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.1
S	0.01	0.00	0.01	0.01	0.00	0.01	0.00	0.01	0.01	0.00	0.01	0.01	0.00
-O=F	-0.29	-0.41	-0.38	-0.29	-0.32	-0.29	-0.31	-0.44	-0.51	-0.65	-0.45	-0.38	-0.25
Total	100.14	99.52	99.94	99.93	99.73	99.73	100.19	100.13	99.76	99.84	100.25	100.08	100.37
A/CNK	1.08	1.11	1.12	1.08	1.11	1.09	1.11	1.22	1.33	1.42	1.11	1.18	1.08
%C	0.96	1.59	1.77	1.05	1.58	1.37	1.43	3.53	4.54	5.56	1.39	2.21	1.67
D.I.	96.64	96.21	96.09	95.77	96.29	96.04	94.29	97.82	97.86	97.27	97.62	97.63	97.39
Trace element abundances in parts per million													
Li	922	1015	1124	889	947	908	703	1429	1686	2385	240	480	499
Rb	1608	1686	1762	1444	1619	1463	1471	1403	1507	1997	1058	963	1210
Cs	45	49	53	44	49	52	28	103	107	68	7.6	5.5	21
Sr	<5	6	<5	<5	5	<5	7	52	15	20	28	<5	<5
Ba	169	329	126	129	141	98	144	75	91	67	155	129	124
Sc	9.6	11	10	10	10	10	5.7	2.5	2.5	4.1	10	8.9	2.6
Cu	3	3	3	3	2	3	na	na	na	na	na	na	3
Zn	20	15	18	14	14	52	15	34	35	92	159	175	101
Y	85	82	79	94	83	97	121	5	6	<5	71	63	157
La	27	29	28	33	34	29	28	1.9	1.8	1.0	39	28	50
Ce	75	83	81	91	95	86	82	2.9	2.6	1.0	90	60	126
Pr	11	11	11	12	12	12	na	na	na	na	11	6.6	17
Nd	33	35	34	38	36	36	33	na	1.2	0.60	36	20	53
Sm	11	11	11	12	12	12	10	0.30	0.40	0.20	9.6	5.1	15
Eu	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	0.03	0.18	0.18	0.11	0.20	0.05	<0.05
Gd	11	12	11	12	12	13	na	na	na	na	9.0	5.0	15
Tb	2.8	2.8	2.6	2.9	2.8	3.0	2.7	0.05	0.10	0.05	2.0	1.4	3.1
Dy	21	20	20	22	20	24	19	0.46	0.41	0.20	15	11	23
Ho	4.7	4.6	4.3	4.7	4.5	5.2	4.3	0.32	0.24	0.18	3.4	2.9	5.2
Er	17	16	16	18	16	18	na	na	na	na	12	11	18
Tm	3.3	3.3	3.2	3.4	3.3	3.6	2.2	na	na	na	2.1	2.2	3.4
Yb	26	24	23	27	24	28	21	0.25	0.23	0.22	17	18	25
Lu	4.2	3.6	3.6	4.0	3.5	4.3	3.1	0.03	0.04	0.03	2.8	3.0	3.7
Zr	67	60	65	68	61	72	81	15	15	14	58	30	114
Nb	41	34	50	43	26	39	45	45	45	57	60	61	51
Mo	4	4	3	2	3	6	na	na	na	na	na	na	10
Sn	15	13	10	7	16	9	16	5	9	4	36	100	21
Sb	0.2	0.2	0.2	0.3	0.4	0.4	0.1	0.1	0.1	0.1	<0.1	<0.1	0.2
Hf	5.9	6.9	6.0	6.4	5.3	5.7	7.6	1.1	1.0	1.1	6.2	5.0	8.9
Ta	11	16	18	13	13	15	28	22	19	25	23	28	19
W	15	26	24	21	14	17	3.4	41	25	39	na	na	7.0
Th	30	31	29	31	31	29	19	2.9	2.7	2.3	28	19	47
U	26	32	35	11	25	18	16	13	7.4	7.2	7.6	3.6	30

**Note.** Abbreviations are: PRG = Pleasant Ridge granite; BRG = Bonny River granite; SAP = St. Austell pluton; EP = Eurajoki pluton; MP = Mount Pleasant; na = not analyzed; Fe<sub>2</sub>O<sub>3</sub><sup>T</sup> = total Fe as Fe<sub>2</sub>O<sub>3</sub>; H<sub>2</sub>O<sup>T</sup> = total H<sub>2</sub>O; A/CNK = molar Al<sub>2</sub>O<sub>3</sub> / (CaO + Na<sub>2</sub>O + K<sub>2</sub>O); %C = % normative (CIPW) corundum calculated on a F-free basis; D.I. = Differentiation Index. The following elements were routinely analyzed for, but were not present in concentrations greater than those of the analytical limit of detection: Cl (<50 ppm), V (<5 ppm), Cr (<10 ppm), Ni (<5 ppm) and Co (<1.0 ppm).

and mode of occurrence (Fig. 6). Such characteristics present a number of problems, most particularly recognition by optical microscopy. Nevertheless, similarly diverse assemblages of accessory minerals have been described in other topaz granites: for example, zircon, cassiterite, bastnäsite and monazite are common, and niobian rutile, columbite, thorite, xenotime, magnetite, ilmenite, and molybdenite are sparse accessories in the Vakkärä granite phase of the Eurajoki pluton (Haapala 1977).

Fluorite and sericite, although present in minor amounts, clearly owe their origin to hydrothermal processes. Typically they occur together, as replacements of primary topaz and lithian mica (Fig. 4H).

### LITHOGEOCHEMISTRY

Major- and trace-element data for the Pleasant Ridge and Bonny River granites are presented in Table 4, together with comparative chemical data for representative topaz-bearing granites from the Eurajoki pluton, Finland, the St. Austell pluton, England, and the nearby Mount Pleasant area (Fig. 2). Data for the international standard rocks used as reference materials are given in the Appendix, which also contains full details of the sampling and analytical methods used in this study. The oxygen and hydrogen isotope data, together with whole-rock  $H_2O$  concentrations determined by mercury manometry, are presented in Table 5. All of the isotopic data are reported in ‰ relative to the Standard Mean Ocean Water (SMOW) reference standard.

### Major elements

The samples of topaz granite from the Pleasant Ridge and Bonny River granites have a very restricted range of compositions: they are all high-silica ( $SiO_2 > 75$  wt.%), weakly peraluminous ( $A/CNK = 1.07-1.12$ ), fluorine-rich rocks ( $F > 0.69$  wt.%), with low concentrations of  $TiO_2$  ( $< 0.03$  wt.%),  $MgO$  ( $< 0.15$  wt.%),  $CaO$  ( $< 0.41$  wt.%) and  $P_2O_5$  ( $< 0.01$  wt.%) relative to other types of granite (Fig. 7). Low  $TiO_2$ ,  $MgO$ ,  $CaO$  and  $P_2O_5$  are reflected by the albitic nature of the plagioclase (Table 1), the lithium-rich character of the mica (*i.e.*, zinnwaldite; Table 3), and the absence of apatite as a common accessory mineral. Although niobian rutile and monazite are present as accessory minerals, their abundances are so small ( $< 0.01$  vol.%) as to have a negligible effect upon the  $TiO_2$  and  $P_2O_5$  contents, respectively, of the bulk rock.  $MnO$  contents are high (0.07–0.09 wt.%) in comparison to other granites (Fig. 7), a feature that results from the Mn-rich nature of the lithian mica present (Table 3). The granite samples have concentrations of total iron that are low (total Fe as  $FeO^T$  is less than 0.95 wt.%) compared to other types of granite (Fig. 7), and reflect the leucocratic character of the granite. Alkali contents are high, with  $Na_2O$  contents ranging from 4.14 to 4.59 wt.%, and  $(Na_2O + K_2O)$  values from 8.46 to 8.86 wt.%; unlike many common types of granite, the Pleasant Ridge granites have  $Na_2O$  in excess of  $K_2O$ , with molar Na/K ratios ranging from 1.42 to 1.63.

The low  $CaO$  contents, high contents of  $Na_2O$ , and high Na/K ratios are reflected in the normative

TABLE 5. OXYGEN AND HYDROGEN ISOTOPIC ANALYSES OF TOPAZ GRANITES

Sample	Lithology	$\delta^{18}O$ (‰)	$\delta D$ (‰)	$H_2O$ (wt.%)
Pleasant Ridge pluton				
B2-4A	Topaz granite with zinnwaldite	8.6	na	na
B2-4B	Topaz granite with zinnwaldite	8.1	-66	0.14
B2-4C	Topaz granite with zinnwaldite	8.1	-105	0.15
B2-5A	Topaz granite with zinnwaldite	8.2	-72	0.18
B2-5B	Topaz granite with zinnwaldite	8.4	-84	0.18
B2-5C	Topaz granite with zinnwaldite	8.1	-85	0.19
Bonny River cupola				
4-151	Topaz granite with zinnwaldite (weak deuteric alteration)	6.8	-106	0.30
Mount Pleasant North Zone				
MP-05	Topaz granite with siderophyllite	8.2	-92	0.28
St. Austell pluton				
2273	Topaz granite with lepidolite	10.6	-44	0.21
1180	Topaz granite with lepidolite	11.1	-74	0.52
1091	Topaz granite with lepidolite	11.8	-100	0.39
Eurajoki pluton				
14773	Topaz granite with siderophyllite	9.6	-70	0.55
25673	Topaz granite with lepidolite	9.8	-95	0.36

Note.  $\delta^{18}O$  and  $\delta D$  values are reported in per mil (‰) relative to SMOW.  $H_2O$  concentrations measured by mercury manometry; na = not analyzed.

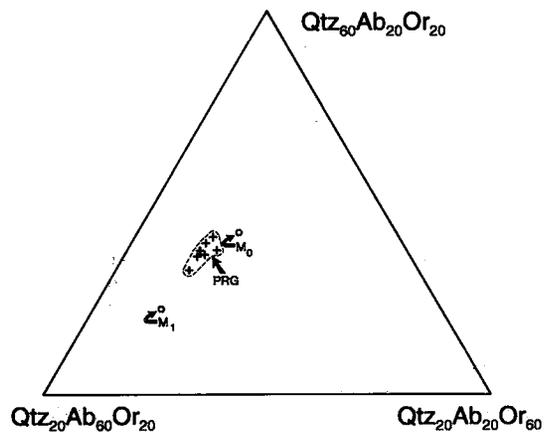


Fig. 8. Qtz-Ab-Or plot of Pleasant Ridge and Bonny River topaz granites (crosses), showing the minima for the system with 0 ( $M_0$ ) and 1 wt.% added fluorine ( $M_1$ ) and excess water at 1 kbar (data for minimum compositions are from Manning 1981).

compositions of the topaz granites. The granites plot in the Qtz-Or-Ab system between the minimum for fluorine-free conditions and the minimum with 1 wt.% fluorine added at 1 kilobar under water-saturated conditions (Fig. 8). These minima correspond to temperatures of 730°C (Tuttle & Bowen 1958) and 690°C (Manning 1981), respectively. However, care must be taken in assigning such temperatures of crystallization for the Pleasant Ridge granite. Although there are a number of geological (see section on Geological Setting) and geochemical criteria (see section on Stable isotope compositions) that support crystallization of the topaz granite magma under water-saturated conditions at a shallow crustal level, the confining pressure cannot be precisely constrained. In addition, exsolution of an aqueous fluid during granite emplacement will result in the partitioning of some of the fluorine into the exsolved fluid phase (*cf.* Webster & Holloway 1990), resulting in an underestimation of the original F content of the magma by analysis of the crystalline rock.

Although they have many features in common with the topaz-bearing granite of the Eurajoki pluton and Mount Pleasant, samples of Pleasant Ridge topaz granite differ from topaz granites of the St. Austell pluton. In particular, the samples of topaz granite from the St. Austell pluton are characterized by lower SiO<sub>2</sub>, higher Al<sub>2</sub>O<sub>3</sub>, and higher P<sub>2</sub>O<sub>5</sub> concentrations (Table 4).

### Volatiles

Concentrations of total H<sub>2</sub>O (<0.3 wt.%), CO<sub>2</sub> (<0.1 wt.%), chlorine (<100 ppm), and sulfur (<0.2 wt.%) in the Pleasant Ridge topaz granite are very low, and contrast markedly with the fluorine concentrations, which are very high (0.69 to 0.98 wt.%). Whether the low abundances of H<sub>2</sub>O, CO<sub>2</sub>, Cl, and S are representative of the chemistry of original magma, or are an artefact of magmatic degassing during granite emplacement (*cf.* Taylor *et al.* 1983, Taylor 1988), is uncertain. Although the low solubility of CO<sub>2</sub> in peraluminous magmas has been demonstrated experimentally (Holloway 1976), the other volatiles under consideration all have moderate to high solubilities in metaluminous to peraluminous granitic magmas (*cf.* Burnham 1979, Clemens 1984, Webster & Holloway 1990). Both Cl and S are, however, partitioned strongly into the exsolved aqueous fluid during magmatic degassing at shallow crustal levels (<2 kbar: Holland 1972, Burnham & Ohmoto 1980). In the case of Cl, Webster & Holloway (1990) reported a variation in D<sub>Cl</sub> of 9.8 to 46.2 (where D<sub>Cl</sub> = ppmw of Cl in the fluid / ppmw of Cl in the melt) for water-saturated haplogranitic

melt at pressures of approximately 2 kbar and a temperature of approximately 800°C.

In contrast to the other volatile species, fluorine partitions selectively into granitic magma rather than into the exsolved aqueous fluid phase (Manning 1981, Webster & Holloway 1990), although some F is always lost to the vapor phase during degassing; Webster & Holloway (1990) reported a variation in D<sub>F</sub> of 0.22 to 0.37 for haplogranitic melt with <2 wt.% F at P = 2 kbar and T = 795°C. High concentrations of F are thus to be expected in volatile-rich granites even if magmatic degassing has occurred. Although Turekian & Wedepohl (1961) proposed an average value of 850 ppm F for low-Ca granites, Bailey (1977) reported that F contents in pantellerites and alkali granites of varying alkalinity typically range from 1,000 to 3,000 ppm; in individual plutons of peralkaline albite-riebeckite granite, abundances of up to 0.94 wt.% F have been recorded (the Kaffo Valley granite, Nigeria; Kinnaird *et al.* 1985). In addition to their high F contents, peralkaline granites and rhyolites also typically have high concentrations of chlorine: for example, 0.07 to 0.18 wt.% Cl in granites from the Liruei Complex (Bowden & Turner 1974) and 0.37 to 0.78 wt.% in pantellerites (Bowden 1974). The only metaluminous to peraluminous rocks with F concentrations similar to those in the Pleasant Ridge topaz granite are ongonites (topaz-bearing quartz keratophyres with up to 2.01 wt.% F: Kovalenko & Kovalenko 1976), topaz rhyolites (up to 1.3 wt.% F in vitrophyres: Christiansen *et al.* 1984) and, of course, topaz-bearing granites from other locations, such as St. Austell (Table 4). All of these rocks are notable in having low to very low contents of Cl (<0.14 wt.% for glassy topaz rhyolites: Christiansen *et al.* 1986, <250 ppm for topaz granites: this study, Manning & Hill 1990) and high to very high F/Cl ratios (3 to 10 for topaz rhyolite glasses, 40 to 70 for topaz granites) in contrast to their peralkaline counterparts (F/Cl <3: Christiansen *et al.* 1983).

### Trace elements

The trace-element contents of the Pleasant Ridge and Bonny River granites are unique in many respects, regardless of whether they are examined in a local or a regional context. Even though the contiguous biotite granites of the Pomeroy Intrusive Suite (Sorrel Ridge, Beech Hill, and True Hill granites; Fig. 2) and the Mount Douglas Granite (McLeod *et al.* 1988) have unusually high (*e.g.*, Rb) or low (*e.g.*, Sr) concentrations of many trace elements, which demonstrate their A-type affinities (Taylor *et al.* 1985, McLeod *et al.* 1988, Whalen 1986), they do not exhibit levels of

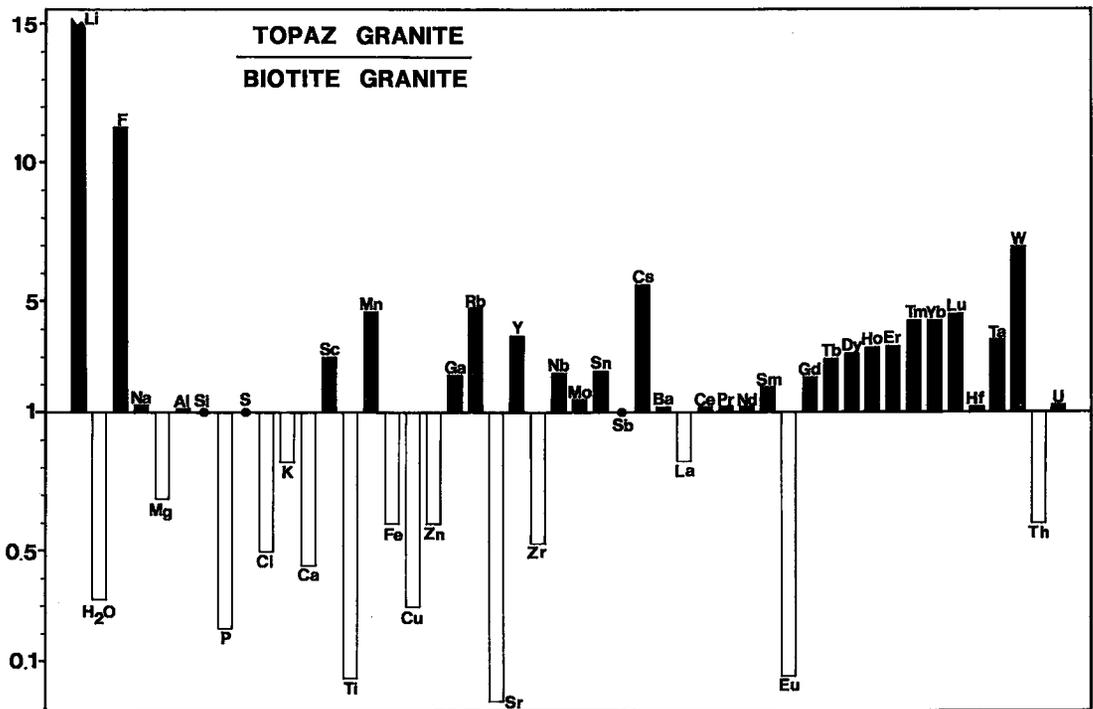


FIG. 9. Enrichment factors for 48 elements in the Pleasant Ridge topaz granite (Table 4) compared to a sample of biotite granite from the contiguous Sorrel Ridge pluton (Fig. 2; Taylor, unpubl. data).

enrichment or depletion of the magnitude found in the topaz granite in the Pleasant Ridge and Bonny River plutons (Fig. 9).

By comparison with even the most highly fractionated examples of I- and S-type granite with similar contents of SiO<sub>2</sub> (cf. Sawka *et al.* 1990), the Pleasant Ridge topaz granite is enriched in: the alkali metals Li (up to 1124 ppm), Rb (up to 1752 ppm), and Cs (up to 53 ppm); and the high-valence cations Y (up to 97 ppm), Nb (up to 50 ppm), Sn (up to 16 ppm), Ta (up to 18 ppm), W (up to 26 ppm), and U (up to 35 ppm). Conversely, they have low to very low abundances of the alkaline earth metals Sr (<10 ppm) and Ba (<169 ppm), V, Co, Ni, Cu, Mo (all are <10 ppm), Cr (<20 ppm), Zn (<52 ppm), and Zr (<72 ppm). Many of these enrichments and depletions are extreme: for example, using the mean abundances for the six samples of Pleasant Ridge granite in Table 4, the contents of Rb, Y, and U are from two to seven times as great as those of aluminous A-type granites, and the depletions in Sr, Ba, Zn, and Zr are from two to four times lower (comparative data from Collins *et al.* 1982). The concentrations of Rb, Cs, and Ta are from two to nine times greater

than those of F-rich I-type granite, and from two to five times greater than those of volatile-rich S-type granite (comparative data from Sawka *et al.* 1990). Concentrations of Sc (10 ppm) and Th (30 ppm) in the Pleasant Ridge topaz granite are similar to those recorded for aluminous A-type granites.

For the trace elements that show striking enrichments, the major host minerals are: the lithian mica (*i.e.*, zinnwaldite) for Li, Rb, and Cs (Table 3), niobian rutile for Nb and Ta, columbite-tantalite for Ta, Nb, W, and Sc, cassiterite for Sn, and uraninite for U. In addition, Sn also is concentrated in the niobian rutile (EPMA-WD gives 63.4–79.9 wt.% TiO<sub>2</sub>, 11.8–7.8 wt.% Nb<sub>2</sub>O<sub>5</sub>, 6.7–9.7 wt.% FeO, 1.5–5.3 wt.% Ta<sub>2</sub>O<sub>5</sub>, 1.3–1.8 wt.% SnO<sub>2</sub>). The analysis of monazite and bastnäsite, by SEM-ED, indicates that these accessories are the major carriers of Th and LREE. Mica from other topaz granites is typically lithium-rich, and characteristically also is enriched in both Rb and Cs (Haapala 1977, Henderson *et al.* 1989). Niobian rutile, columbite-tantalite, and cassiterite also have been identified in topaz granite from: St. Austell, Eurajoki (Taylor, unpubl. data), Yichun, China (Pollard & Taylor 1991), and

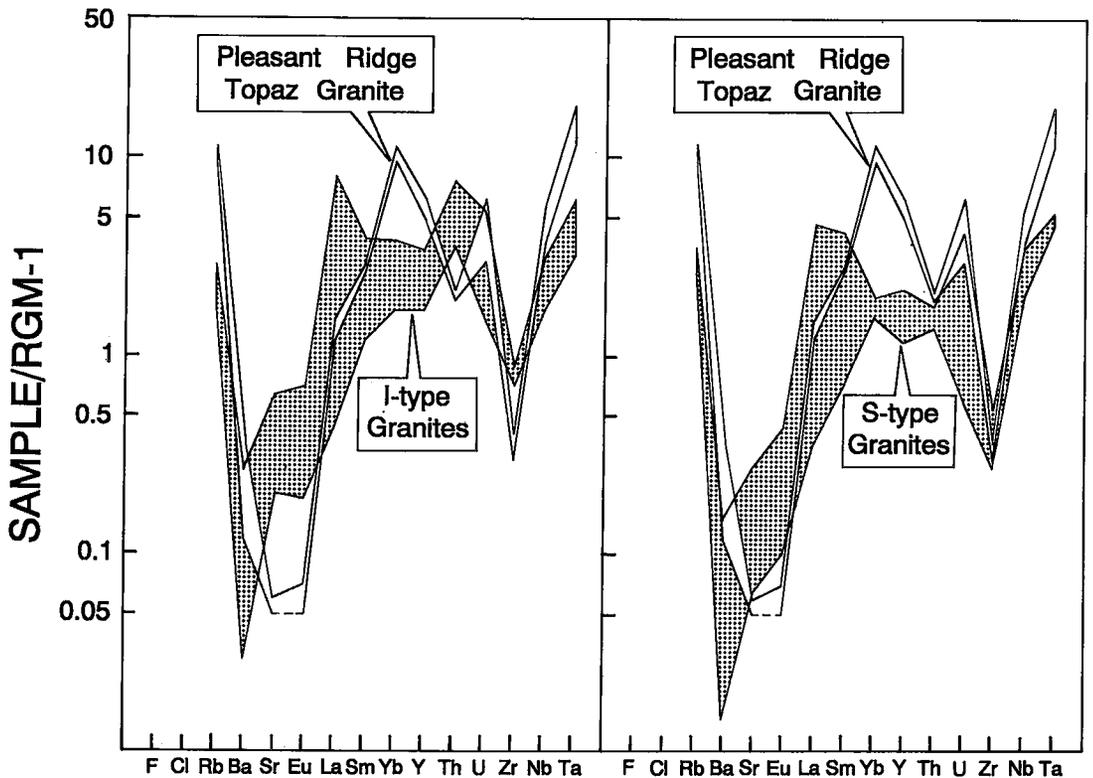


FIG. 10. Concentrations of F plus selected trace-elements in topaz granites from Pleasant Ridge (Table 4) compared to those of representative examples of highly fractionated I-type (WTG 26) and S-type (WTG 9) granites (data from Sawka *et al.* 1990, Table 3; no F and Cl data reported). The values used for normalization pertain to RGM-1, a rhyolite obsidian from Medicine Lake volcano, California, and U.S. Geological Survey geochemical reference (Govindaraju 1984); the elements are listed in order of their relative field-strength, increasing to the right (after Christiansen *et al.* 1986).

Cinovec, Czechoslovakia (Stemprok & Šulcek 1969).

Some of the compositional features of the Pleasant Ridge granite, other topaz-bearing felsic rocks, and of I- and S-type granites are compared, in terms of their trace element contents, in Figures 10 and 11. Such illustrations show the relative concentrations of many elements in a single sample or group of closely related samples. All concentrations were normalized to those of U.S. Geological Survey geochemical reference sample Rhyolite RGM-1, and the elements in Figures 10 and 11 are listed in order of increasing field-strength (following the procedure of Christiansen *et al.* 1986). The striking enrichments of Rb, Nb, and Ta and the relative depletions of Sr, Eu, and Zr in the Pleasant Ridge topaz granite, in comparison to other more widely distributed types of granite, are readily apparent from an inspection of Figure 10. It is

noteworthy that these specific enrichments and depletions are features common to the topaz-bearing granites from the Eurajoki and St. Austell plutons (Fig. 11), and are also distinguishing features of the group of subvolcanic, fluorine-rich igneous rocks termed "ongonites" (Fig. 12).

#### Rare-earth elements

Total rare-earth-element ( $\Sigma 14$  REE) contents for the individual samples of Pleasant Ridge granite range from 247 to 281 ppm (Table 4). Such total abundances are quite typical of those of granitic rocks of the continental crust (*cf.* Cullers & Graf 1984). The Pleasant Ridge topaz granite exhibits a flattened, gullwing-shaped chondrite-normalized REE pattern ( $0.67 < La/Lu_N < 1.01$ ) with a deep negative europium anomaly ( $Eu/Eu^* < 0.01$ ) (Fig. 13). The large negative Eu anomaly requires that

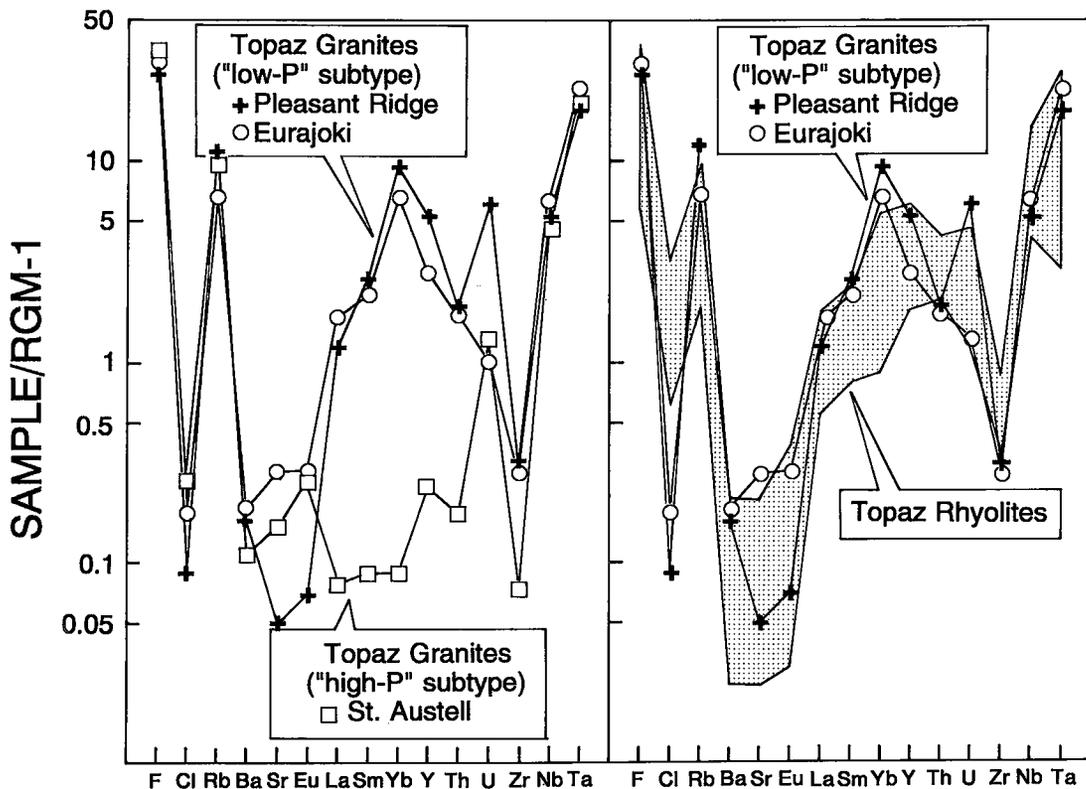


FIG. 11. Concentrations of F plus selected trace-elements in "low-P" topaz granites from the Pleasant Ridge pluton (sample 82-4C, Table 4) and the Eurajoki pluton (sample 256/73, Table 4), and "high-P" topaz granite from the St. Austell pluton (sample 1160, Table 4). The values used for normalization pertain to RGM-1, and the field for topaz rhyolites is from Christiansen *et al.* (1986, Fig. 41).

much feldspar was involved in the mineral-melt equilibria during melting or crystallization (Cullers & Graf 1984). Similarly, the low  $La/Lu_N$  ratio requires the presence during melting or crystallization of one or more *LREE*-enriched accessory minerals. Monazite, which occurs in trace amounts in all of samples of the Pleasant Ridge granite, is the most likely host for the *LREE* (*cf.* Charoy 1986, Congdon & Nash 1991), and as little as 0.05% monazite is required to account for all of *LREE* in the Pleasant Ridge granite.

Although the depletion of light rare-earth elements in felsic magmas is not uncommon (Miller & Mittlefehldt 1982), the high concentrations of the heaviest *REE* (up to 28 ppm Yb and 4.3 ppm Lu: Table 4), and very low  $La/Lu_N$  ratio in the Pleasant Ridge granite (Fig. 13), are unusual. Uraninite, where present as an accessory mineral in granitic rocks, is known to concentrate the *HREE*,

specifically Yb and Lu (Fryer & Taylor 1987), and is thus a possible host for these elements. However, the extremely low abundance of uraninite in the Pleasant Ridge granite precludes its role as the only carrier of the heaviest *REE*. Thus, it is likely that an as yet unidentified accessory mineral, such as xenotime, thorite, or fergusonite, is the major reservoir of the *HREE* in the Pleasant Ridge topaz granite. Xenotime and thorite both have been identified by Haapala (1977) as accessory minerals in samples of Eurajoki topaz granite, whose *REE* abundances and distribution are almost identical to those of the Pleasant Ridge granite (Fig. 13). In addition, thorite and fergusonite both occur as accessories in topaz rhyolite from the Honeycomb Hills, Utah (Congdon & Nash 1991).

The *REE* content and distribution in topaz granite from the St. Austell pluton (Fig. 13) are quite distinct from those that characterize the topaz

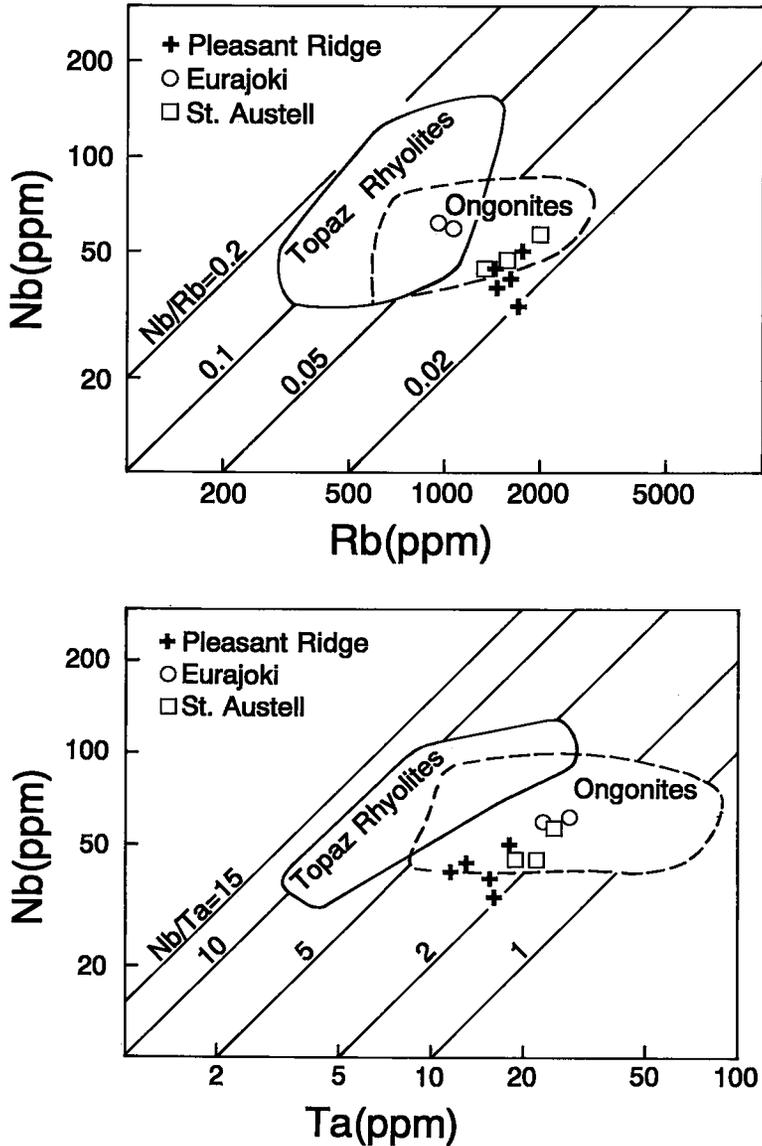


FIG. 12. Rb-Nb-Ta concentrations in topaz granites from Pleasant Ridge, St. Austell, and Eurajoki (Table 4). Fields for topaz rhyolites and ongonites from Christiansen *et al.* (1986).

granites in the Pleasant Ridge and Eurajoki plutons (Fig. 13), and also that of topaz rhyolite (Fig. 14). The total rare-earth-element content ( $\Sigma 10REE$ ) of the St. Austell topaz granite is typically less than 10 ppm (Table 4), with a La/Lu<sub>N</sub> ratio of 3.5 to 6.6 (Fig. 13). This very significant depletion in the

total REE content also is accompanied by depletions of both Y (<10 ppm) and Th (<5 ppm) in the St. Austell topaz granite, which again are in marked contrast to the abundances of these elements in topaz granite in the Pleasant Ridge and Eurajoki plutons (Table 4, Fig. 11).

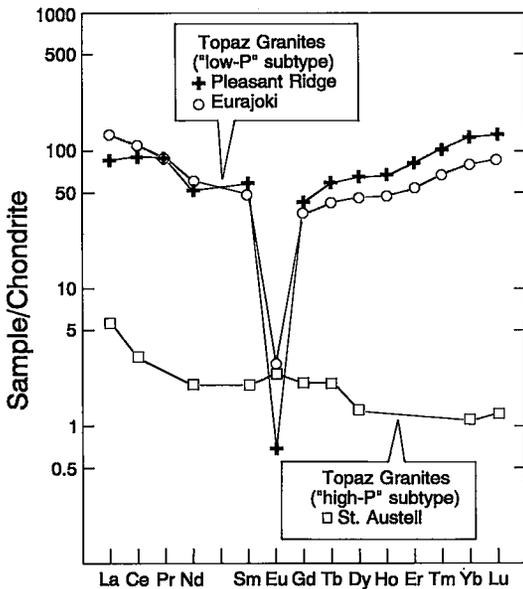


FIG. 13. Chondrite-normalized rare-earth-element plot of samples of topaz granite from the Pleasant Ridge (sample C82-4C, Table 4), Eurajoki (sample 256/73, Table 4), and St. Austell (sample 1160, Table 4) plutons. The chondritic values of Boynton (1984) were used for the normalization.

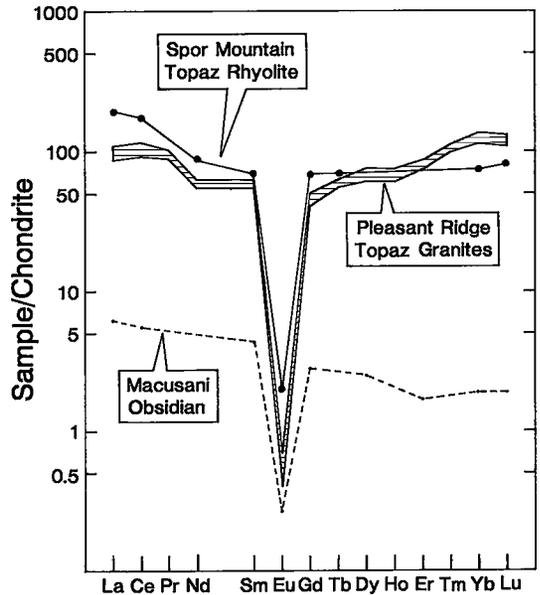


FIG. 14. Chondrite-normalized rare-earth-element plot of the field of values of the Pleasant Ridge granite (Table 4), and examples of topaz rhyolite (data from Christiansen *et al.* 1986) and Macusani glass (data from Pichavant *et al.* 1988). The chondritic values of Boynton (1984) were used for the normalization.

### Ratios of oxygen and hydrogen isotopes

Results of oxygen and hydrogen isotope analyses of whole-rock samples of the Pleasant Ridge and Bonny River granites are presented in Table 5, together with comparative data for samples of topaz granite from the Eurajoki pluton in Finland, the St. Austell pluton in England, and the nearby Mount Pleasant area (Fig. 2). The oxygen isotope values for the topaz granite samples from the Pleasant Ridge pluton are fairly uniform, with  $\delta^{18}\text{O}$  values that range from 8.1 to 8.6‰. In contrast, the granite sample from the tin-mineralized Bonny River cupola, which has suffered postmagmatic hydrothermal alteration (with the partial breakdown of magmatic topaz and lithian mica to form sericite and fluorite; Fig. 4H), has a lower  $\delta^{18}\text{O}$  value of 6.8‰. Hydrogen isotope compositions for samples of the Pleasant Ridge granite vary markedly (-66 to -105‰) and have a much wider range than the corresponding  $\delta^{18}\text{O}$  compositions (Fig. 15).

Studies of the stable isotope characteristics of classic I- and S-type granite suites (O'Neil & Chappell 1977, O'Neil *et al.* 1977) have demonstrated that a clear distinction can be made

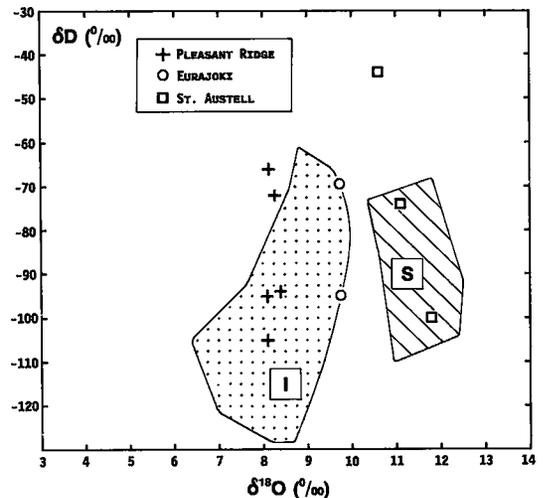


FIG. 15. Values of  $\delta^{18}\text{O}$  (‰) versus  $\delta\text{D}$  (‰) for topaz granites from Pleasant Ridge, St. Austell, and Eurajoki (Table 4). The fields for I- and S-type granites are from O'Neil *et al.* (1977).

regarding the nature of the protolith from the measured oxygen isotope composition. S-type granites (sedimentary protolith) have consistently higher oxygen isotope values (e.g.,  $\delta^{18}\text{O}$  between 10.4 and 12.5‰ in the New England Batholith, between 9.9 and 10.5‰ in the Berridale Batholith, Australia) than the spatially related I-type granites (igneous protolith), whose  $\delta^{18}\text{O}$  values range from 7.7 to 9.9‰ (New England) and from 7.9 to 9.4‰ (Berridale Batholith). Such a distinction, with a dividing line of  $\delta^{18}\text{O}$  of around 10‰ results from the enrichment in  $^{18}\text{O}$  of sediments during processes of surface weathering (Taylor 1968, Savin & Epstein 1970). The  $\delta^{18}\text{O}$  values of the Pleasant Ridge topaz granite (8.1 to 8.6‰) are thus consistent with partial melting of a source dominated by metaigneous rocks. An examination of the oxygen isotope compositions of other topaz granites (Table 5) indicates that samples from Eurajoki and Mount Pleasant also have  $\delta^{18}\text{O}$  values that are typically less than 10‰, but that samples from St. Austell are characterized by higher  $\delta^{18}\text{O}$  values (10.6–11.8‰). The higher  $\delta^{18}\text{O}$  values of the St. Austell topaz granites are consistent, both with their more strongly peraluminous character (Table 4), and also with a source in which there was an important component of pelitic sedimentary material (cf. Miller 1985).

In contrast, the variations of  $\delta\text{D}$  values in felsic plutonic rocks reflect the influence of other than source controls; these include magmatic degassing and exchange with isotopically light meteoric water (cf. Taylor 1988). Although the hydrogen isotopic composition of "deep-seated igneous rocks" has been estimated to vary between -60 and -85‰ (Taylor 1977), it has been clearly demonstrated (Nabelek *et al.* 1983, Taylor *et al.* 1983) that such a range can be extended to values as negative as -110‰ by magmatic degassing during ascent and emplacement. In their study of the stable isotope systematics of the Notch Peak granitic stock, Utah, Nabelek *et al.* (1983) documented a wide range of  $\delta\text{D}$  (-55 to -110‰) values in whole-rock samples with an almost constant  $\delta^{18}\text{O}$  composition (ca. 9.3‰). They attributed these variations to magmatic degassing, with the different responses of the two isotopic ratios to the operation of this process resulting from the much larger isotopic fractionation for D/H, versus that of  $^{18}\text{O}/^{16}\text{O}$ , between coexisting aluminosilicate melt and aqueous vapor phase (cf. Taylor 1988). When taken together with the petrographic data, which demonstrate the lack of pervasive mineral replacement and metasomatism (Fig. 4), the oxygen and hydrogen isotope compositions of the whole-rock samples of the Pleasant Ridge topaz granite are those that would be expected to result from magmatic

degassing during the emplacement of an epizonal pluton.

Comparison of the hydrogen isotopic compositions of the Pleasant Ridge granite with those of F-rich granites in other plutons shows that a similarly broad range of  $\delta\text{D}$  values (-44 to -100‰) also occurs in the topaz granites of the St. Austell pluton (Table 5) and at Yichun in China (Taylor & Pollard, unpubl. data). Collectively, the hydrogen isotopic data suggest that magmatic degassing may be an integral feature of the emplacement of these F-rich, topaz-bearing granites.

## DISCUSSION

The results of a number of recent experimental studies have demonstrated that topaz is a stable mineral in granitic melts over quite a large range of P and T conditions (Anfilogov *et al.* 1973, Kovalenko 1977, Weidner & Martin 1987, Webster *et al.* 1989). In particular, experimental data presented by Weidner & Martin (1987) for a study of the phase equilibria of a fluorine-rich leucogranite (of similar bulk composition to that of the Pleasant Ridge pluton) provide an excellent means of assessing the origin of the various minerals, interpreted here to be magmatic in character, present in the Pleasant Ridge granite. The data of Weidner & Martin (1987) indicate that for water-saturated conditions: (1) topaz, muscovite, and fluorite occur as magmatic minerals in accessory amounts together with plagioclase, K-feldspar, and quartz; (2) plagioclase ( $\text{An}_1$ ) is on the liquidus at low pressure (1 kbar), topaz is on the liquidus at higher pressure (4 kbar), and at intermediate pressure (2 kbar), all crystalline phases (albite, K-feldspar, quartz, topaz, muscovite, and fluorite) dissolve over a very narrow temperature interval of around 25°C; (3) at pressures of 1 to 4 kbar, liquidus temperatures are typically well below 750°C; (4) muscovite (exact composition not specified) is stable up to temperatures of 680–700°C in the pressure range from 1 to 4 kbar, a fact that Weidner and Martin attributed to the F-rich nature of the mica; and (5) highly evolved, F-rich granitic melts will exhibit subsolvus features unless emplaced at very shallow depths (<1.5 kbar).

The petrographic and chemical data presented here are consistent with a magmatic origin for the bulk of the albite, topaz, and lithian mica present in the Pleasant Ridge granite. Textural relations support the early crystallization of albite (as the liquidus mineral) of homogeneous character ( $\text{An}_{0-3}$ ; Table 1), and its inclusion in the various other later magmatic minerals (Figs. 3, 4). Such a mineral paragenesis (Fig. 3) is comparable to the sequence of crystallization recorded at pressures of 1 to 2

kbar in the experiments of Weidner & Martin (1987). The lithian mica in the Pleasant Ridge granite is characterized by its very high F content and  $F/(F+OH)$  ratios (Table 3), which results in its stability at high temperatures (Munoz 1971, 1984), and presence as a primary mineral in F-rich magmas (*cf.* Henderson *et al.* 1989). A shallow level of emplacement for the Pleasant Ridge and Bonny River granites (<2 kbar), which is consistent with the regional geological and geophysical setting of the Pomeroy Intrusive Suite (Fig. 2) as a whole (McLeod 1990, Thomas & Willis 1989), also would enhance the possibility of an initially water-under-saturated magma reaching saturation (Burnham 1979). The large range of hydrogen isotope compositions, but almost constant oxygen isotope composition of the Pleasant Ridge granite (Table 5), provide support for magmatic degassing during emplacement (*cf.* Nabelek *et al.* 1983).

Although the exact timing of degassing is difficult to assess in terms of the history of crystallization of the Pleasant Ridge pluton, the ubiquitous occurrence of all of the important accessories as mineral inclusions in subhedral lithian mica (Fig. 4F), which crystallized early (Fig. 3), is likely significant. As Webster (1990) has pointed out, the timing of exsolution of the aqueous fluid with respect to crystallization is critical because if (Cl,F)-enriched fluids are not released until most of the magma has crystallized, then a large proportion of the rare alkali (Li, Rb, Cs) and lithophile metals (Nb, Ta, Sn, W, U) may be sequestered into phases such as lithian mica (enriched in Li, Rb, and Cs), niobian rutile (enriched in Nb, Ta, and Sn), columbite-tantalite (enriched in Nb, Ta, and W), cassiterite, and uraninite. These accessories are homogeneously distributed in the Pleasant Ridge granite and appear to have crystallized early from the topaz-bearing felsic magma. As a consequence, the distribution of these elements likely approximates their distribution in the parental F-rich magma.

The unusual mineralogical character of the Pleasant Ridge topaz granite is reflected in its chemical composition, which is quite distinct from that of the most highly fractionated examples of I- and S-type granite, and aluminous A-type granite. Thus, the Pleasant Ridge topaz granite is highly enriched in fluorine, the alkali metals (Li, Rb, Cs), and the lithophile elements (Nb, Ta, Sn, and W), and has marked depletions of Cl, Ti, Fe, Mg, Ca, P, and Sr (Table 4, Fig. 10), features that are common also to the topaz granites from the Eurajoki and St. Austell plutons (Table 4). It is clear that topaz granites can, as a group, be distinguished from all of the other widely distributed types of granite by a variety of geochemical

criteria, the most notable of which is their very high F content.

However, also it is apparent from the results of this study that there exists a natural dichotomy within this unique group of F-rich felsic rocks, such that "low-P" and "high-P" subtypes of topaz granite can be distinguished (Fig. 16): topaz granites from the Pleasant Ridge and Eurajoki plutons belong to the "low-P" subtype ( $P_2O_5 < 0.1$  wt.%,  $Al_2O_3 < 14.5$  wt.%,  $SiO_2 > 73$  wt.%), and those from the St. Austell pluton belong to the "high-P" subtype ( $P_2O_5 > 0.4$  wt.%,  $Al_2O_3 > 14.5$  wt.%,  $SiO_2 < 73$  wt.%). Such a division reflects the more strongly peraluminous character of "high-P" topaz granites ( $1.2 < A/CNK < 1.5$ ), in contrast to their "low-P" counterparts, and also highlights the significant difference in the distribution of Y, REE, and Th that exists between the two subtypes (Table 6). The chemical compositions of topaz granites from other localities lend support to the subdivision proposed here, with granites from Fawwarah, Saudi Arabia (Du Bray 1984), Mount Pleasant, New Brunswick (Taylor *et al.* 1985, 1989), and the Erzgebirge in Germany and Czechoslovakia (Štemprok & Šulcek 1969, Tischendorf & Förster 1990) as representatives of the "low-P" subtype, and those from Tregonning, Cornwall (Exley & Stone 1982), Meldon, Devonshire (Chaudhry & Howie 1973), Beauvoir, France (Cuney & Autran 1987), and Yichun, China (Pollard & Taylor 1991) being characteristic of the "high-P" subtype (Table 6).

For the "high-P" subtype of topaz granite (Table 6), the whole-rock concentrations of  $P_2O_5$  that are present (St. Austell: 0.51–0.73 wt.%, Table 4; Tregonning: 0.44–0.73 wt.%, Exley & Stone 1982; Yichun: 0.51–0.62 wt.%, Pollard & Taylor 1991) require that phosphorus be present in minerals other than the phosphates. Kontak & Strong (1988) and London *et al.* (1990) recognized that the feldspars may represent a principal reservoir of phosphorus in P-rich peraluminous leucogranites and pegmatites. For example, Kontak & Strong (1988) detected greater than 1 wt.%  $P_2O_5$  in feldspars from the South Mountain batholith in Nova Scotia; in a preliminary study of P in alkali feldspars of rare-element granitic pegmatites, London *et al.* (1990, p. 771) reported concentrations that "...range from below detection limit to 1.20 wt.%  $P_2O_5$ , with approximately 60% of the data indicating greater than 0.30 wt.%". Electron-microprobe data for feldspar in samples from the St. Austell pluton (Table 1) indicate that  $P_2O_5$  concentrations in plagioclase (albite) and K-feldspar attain 0.43 and 0.63 wt.%, respectively, in this example of a "high-P" type of topaz granite. Given the modal abundance of albite (35–40%) and K-feldspar (20–25%) in the St. Austell topaz

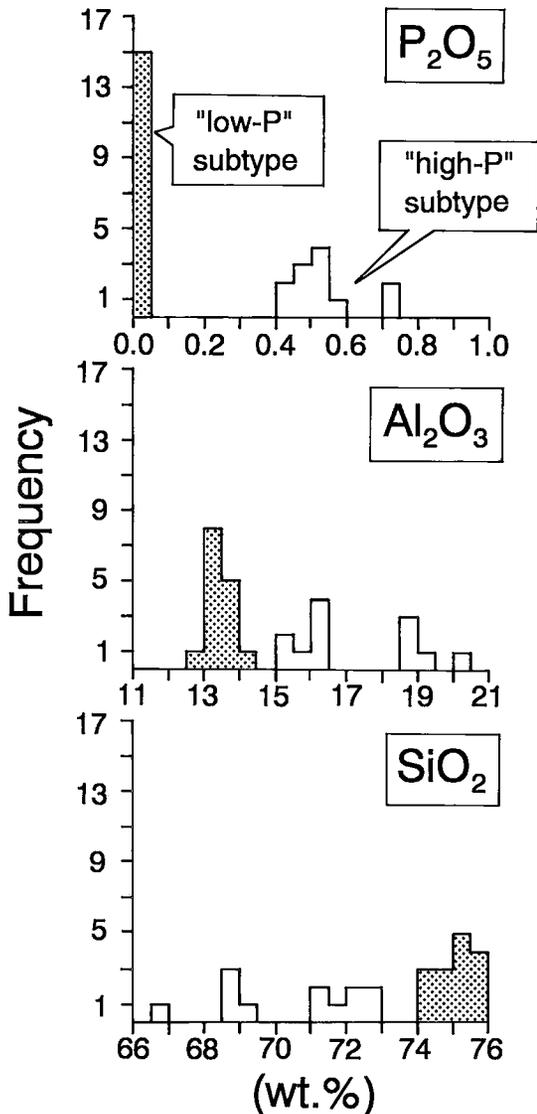


FIG. 16. Histograms of  $P_2O_5$ ,  $Al_2O_3$ , and  $SiO_2$  concentrations (in wt.%) in topaz granites: "low-P" types from Pleasant Ridge (Table 4), Eurajoki (Table 4), Mt. Pleasant (Table 4) and Fawwarah (Du Bray 1984); "high-P" types from St. Austell (Table 4), Tregonning and Meldon (Exley & Stone 1982), and Yichun (Pollard & Taylor 1991). The vertical scale shows the frequency of each value.

granite, such levels of phosphorus enrichment in the two feldspars can account for up to 50% of the total budget of phosphorus in the rock.

The summary presented in Table 6 is intended only to aid in the recognition of the magmatic characteristics of topaz granite, and its "low-P" and "high-P" subtypes. Although it incorporates data from as many of the known occurrences of topaz granite as possible, Table 6 should not be viewed as a panacea for understanding the nature of this unique group of igneous rocks. As a *caveat emptor* to the reader, it should be noted that the current petrological data-base for topaz granites is far from adequate to permit a rigorous analysis of their character and petrogenesis. Thus, with the acquisition of new data for topaz granites, particularly for occurrences in Russia, Mongolia, and China, the fabric of Table 6 will undoubtedly be subject to modification. In addition, although every effort has been made to include only data for "fresh" samples of topaz granite in Table 6, it is evident that high-temperature (deuteric) alteration is a common feature in many other occurrences of topaz granite. Such postmagmatic alteration typically involves the complete replacement of magmatic fluortopaz and lithian mica by fluorite and sericite (Fig. 4H), the sericitization of primary albite and K-feldspar, and the destruction of igneous textures (*cf.* Manning & Exley 1984). Also, it results in the large-scale redistribution of the alkalis (Li, Na, Rb, Cs), the alkaline earths (Ca, Sr), and other elements, and the modification of oxygen isotope compositions (*cf.* Fouillac & Rossi 1991).

Topaz rhyolites share many of the same chemical characteristics as the "low-P" subtype of topaz granite described here (Figs. 7, 11, 14). Although most topaz rhyolites contain only topaz crystallized during postmagmatic vapor-phase alteration (Christiansen *et al.* 1986), the rhyolitic lavas of the Honeycomb Hills, Utah have fluortopaz as a primary phenocryst phase (Congdon & Nash 1991). These unusual lavas, of Cenozoic age, are the product of a single eruptive cycle. In addition to topaz, the Honeycomb Hills lavas contain quartz, sanidine, albite ( $An_6$ ), fluorsiderophyllite, and fluorite as phenocrysts, and zircon, thorite, columbite, fergusonite, and monazite as magmatic accessories (Congdon & Nash 1991). A whole-rock composition for the Honeycomb Hills rhyolite ( $SiO_2 = 73.3$  wt.%,  $TiO_2 = 0.01$  wt.%,  $Al_2O_3 = 14.0$  wt.%,  $FeO^T = 0.8$  wt.%,  $MnO = 0.07$  wt.%,  $MgO < 0.01$  wt.%,  $CaO = 0.42$  wt.%,  $Na_2O = 4.59$  wt.%,  $K_2O = 4.44$  wt.%,  $P_2O_5 < 0.01$  wt.%,  $F = 0.61$  wt.%,  $Cl = 0.10$  wt.%, and maximum  $Li = 344$  ppm,  $Rb = 1960$  ppm,  $Cs = 78$  ppm,  $Nb = 81$  ppm,  $Ta = 44$  ppm,  $W = 34$  ppm, and  $Sn = 33$  ppm: Congdon & Nash 1991) corresponds closely to that of typical "low-P" topaz granite (Table 6), and provides corroboration for the existence of natural F-rich magmas of this charac-

**TABLE 6. SUMMARY OF THE CHARACTERISTIC FEATURES OF THE TWO SUBTYPES OF TOPAZ GRANITE**

	Low-P subtype	High-P subtype
Age and location	Middle Proterozoic* (ca. 1570 Ma: Eurajoki, Finland). Late Proterozoic (ca. 590 Ma: Fawwarah, Kingdom of Saudi Arabia). Late Devonian (ca. 360 Ma: Pleasant Ridge Mount Pleasant, New Brunswick, Canada). Pennsylvanian/Early Permian (ca. 305-295 Ma: Cinovec, Czechoslovakia; Altenberg and Zinnwald, Germany). Late Cretaceous (ca. 70 Ma: Kougarak, Alaska, U.S.A.).	Late Devonian (ca. 370 Ma: East Kemptville, Nova Scotia, Canada). Pennsylvanian/Early Permian (ca. 280 Ma: St. Austell, Tregonning, and Meldon in S.W. England; Beauvoir, France). Early Cretaceous (ca. 130 Ma: Yichun, China).
Essential minerals	Quartz, K-feldspar, albite ( $An_{0-9}$ ), F-rich topaz, and lithian mica (mica compositions range from lithian siderophyllite through zinnwaldite to lepidolite).	
Accessory minerals	Accessories common to both types include zircon, niobian rutile, columbite-tantalite, cassiterite, monazite, and sulfides.	
	Phosphates are very rare: mainly monazite.	Abundant phosphates: apatite, monazite, amblygonite-montebrazite.
Major elements	Common features of both types are: the very low abundances of $TiO_2$ (<0.1 wt.%), MgO (<0.2 wt.%), and CaO (<0.5 wt.%); the low content of $FeO^T$ (<1.0 wt.%); and the high concentrations of $Na_2O$ (>4.0 wt.%) and high Na/K ratios (1.0-2.0, but up to 3.6).	
	Low $P_2O_5$ (<0.10 wt.%), higher $SiO_2$ (>73 wt.%), lower $Al_2O_3$ (12-14.5 wt.%); weakly peraluminous (A/CNK = 1.0-1.2).	High $P_2O_5$ (>0.40 wt.%), lower $SiO_2$ (68-73 wt.%), higher $Al_2O_3$ (14.5-20 wt.%); strongly peraluminous (A/CNK = 1.2-1.5).
Volatiles	Common features are the very high concentrations of F (>0.4 wt.%, up to 2.5 wt.%), and the very low abundances of Cl (<250 ppm) and $CO_2$ (<0.2 wt.%).	
Minor and trace elements	Common features are the very high concentrations of the alkalis: Li (typically 250-2600 ppm, but up to 7500 ppm), Rb (typically 800-2400 ppm, but up to 4100 ppm), and Cs (typically 20-150 ppm, but up to 700 ppm); and the lithophile elements: Nb (30-120 ppm), Ta (10-60 ppm, but up to 250 ppm), W (10-50 ppm), and Sn (10-300 ppm). Also the low to very low abundances of V, Cr, Co, Ni, Cu, Sr, Zr, Mo, and Ba.	
Rare earth elements, Y, and Th	Total REE contents of 150 to 350 ppm; flat to gullwing-shaped REE patterns ( $La/Lu_N = 0.6$ to 1.5) with deep negative Eu anomalies ( $Eu/Eu^+ < 0.1$ ). High Y (60-160 ppm), and moderate to high Th contents (20-50 ppm).	Total REE contents of less than 10 ppm; slight enrichment of LREE ( $La/Lu_N > 1.5$ ); moderate negative Eu anomaly ( $Eu/Eu^+ < 0.2$ ). Low Y (<15 ppm), and low Th (<10 ppm) abundances.
Stable isotopes	Whole-rock $\delta D$ compositions of both types vary over an extended range from -40 to -110‰.	
	Whole-rock $\delta^{18}O$ compositions are lower and range from 7.5 to ca. 10.0‰.	Whole-rock $\delta^{18}O$ compositions are higher and range from ca. 10.0 to 13.5‰.

**Note:** \*Ages of boundaries in Time Scale after Palmer (1983). Sources of data are: Bray & Spooner (1983); Chaudhry (1971); Darbyshire & Shepherd (1985); Du Bray *et al.* (1988); Exley & Stone (1982); Haapala (1977); Henderson *et al.* (1989); Kontak (1990); Manning & Hill (1990); Pollard & Taylor (1991); Puchner (1986); Rossi *et al.* (1987); Schwartz (1992); Sheppard (1977); Štemprok & Šulcek (1969); Stone (1975); Stone & George (1988); Stone & Exley (1983); Stone *et al.* (1988); Taylor (this study); Taylor *et al.* (1985, 1989); Tischendorf & Förster (1990); von Knorring & Condliffe (1984); and unpublished data of the author.

ter. Although a volcanic analogue of the "high-P" subtype of topaz granite is not as evident, the compositional similarities between Macusani glass (Pichavant *et al.* 1988) and samples of "high-P" topaz granite are striking (Figs. 13, 14).

### CONCLUSIONS

(1) The small plutons at Pleasant Ridge and Bonny River appear, in their upper portions, to be composed entirely of alkali feldspar granite with essential quartz, K-feldspar, albite ( $\text{An}_{0.3}$ ), lithian mica (*var.* zinnwaldite), and topaz. Thus, they are examples of subsolvus granite (Tuttle & Bowen 1958). As such, they represent one of only a very small group of known occurrences in the northern Appalachians of the rare igneous rock type, *topaz granite*, others being located in the North Zone at Mount Pleasant in New Brunswick (Taylor *et al.* 1985, 1989), and at East Kemptville in Nova Scotia (Kontak *et al.* 1988, Kontak 1990).

(2) Two features of these occurrences of topaz granite are worthy of note. First, the available geochronological data indicate that granite emplacement occurred between 370 and 360 Ma. Second, significant zones of hydrothermal tin mineralization are closely related, both spatially and genetically, to the bodies of topaz granite. Together, these features indicate that an important metallogenic episode, involving the emplacement of topaz-bearing felsic magmas and the genesis of tin deposits, occurred in the northern Appalachians in the Late Devonian.

(3) The petrographic and chemical data for the Pleasant Ridge granite are consistent with a magmatic origin for the bulk of the albite, topaz, and lithian mica. Textural relations support the early crystallization of albite and topaz, and the subsequent inclusion of both minerals in later lithian mica, quartz, and K-feldspar. Such a mineral paragenesis is comparable to the sequence of crystallization recorded at pressures of 1 to 2 kbar in the experiments of Weidner & Martin (1987).

(4) The topaz granites from the Pleasant Ridge and Bonny River plutons are fluorine-rich, high-silica types with extremely elevated contents of Li, Rb, Cs, Y, Nb, Ta, Sn, W, and U, whose chemical compositions are distinct from those of the most highly fractionated examples of I- and S-type granite, and also from those of aluminous A-type granite.

(5) Whole-rock  $\delta^{18}\text{O}$  values of the Pleasant Ridge topaz granite are nearly constant (8.1–8.6‰), whereas its  $\delta\text{D}$  values have a broad range, from -66 to -105‰, features indicative of a source dominated by metaigneous rocks, and also of the occurrence of magmatic degassing during emplace-

ment. Similarly large ranges of  $\delta\text{D}$  value in topaz granite from other localities suggest that degassing may be, in general, an integral part of the emplacement of these F-rich felsic magmas. The oxygen isotope compositions of "high-P" topaz granite (*e.g.*, St. Austell) are higher than those of their "low-P" counterparts, exemplified here by the Pleasant Ridge topaz granite. The higher  $\delta^{18}\text{O}$  values (typically greater than 10‰), in combination with the more strongly peraluminous character, indicate the presence of an important component of pelitic metasediment in the source of the "high-P" subtype of topaz granite.

(6) The Pleasant Ridge and Bonny River topaz granites have many features in common with topaz granites from Proterozoic (*e.g.*, Eurajoki, Finland) and Phanerozoic (*e.g.*, St. Austell, England) lithotectonic settings. However, also it is apparent that there exists a natural dichotomy within this unique group of F-rich felsic rocks, such that "low-P" and "high-P" subtypes of topaz granite can be distinguished. Topaz granites from Pleasant Ridge, Mount Pleasant, and Eurajoki belong to the "low-P" subtype ( $\text{P}_2\text{O}_5 < 0.1$  wt.%,  $\text{Al}_2\text{O}_3 < 14.5$  wt.%,  $\text{SiO}_2 > 73$  wt.%), and those from St. Austell and a number of other localities (*e.g.*, Yichun, China: Pollard & Taylor 1991) are representatives of the "high-P" subtype ( $\text{P}_2\text{O}_5 > 0.4$  wt.%,  $\text{Al}_2\text{O}_3 > 14.5$  wt.%,  $\text{SiO}_2 < 73$  wt.%). Such a subdivision reflects the more strongly peraluminous character of "high-P" topaz granite.

(7) Topaz rhyolites share many of the same chemical features as the "low-P" subtype of topaz granite (typified by the Pleasant Ridge granite) described here. In particular, the rhyolitic lavas of the Honeycomb Hills, Utah, which have a phenocryst assemblage of quartz, sanidine, albite, fluortopaz, and fluorsiderophyllite (Congdon & Nash 1991) and an almost identical whole-rock composition, may represent the eruptive equivalent of "low-P" topaz granite.

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## APPENDIX: ANALYTICAL PROCEDURES

Representative whole-rock samples were collected from diamond drill-core. Samples were selected from long sections of core (>20 m) displaying homogeneity, and a low density of fractures and hydrothermal alteration or mineralization. Sample preparation was undertaken in such a way as to produce approximately 50 g of a homogeneous powder (<150 mesh), and to minimize contamination during crushing and grinding (e.g., use of diamond-impregnated and agate-lined grinding media; cf. Hickson & Juras 1986); the levels of contamination were monitored regularly using quartz blanks.

A variety of analytical techniques were used: wavelength-dispersion X-ray-fluorescence spectrometry using duplicate glass discs (XRF/GD: major elements plus V, Cr, Ni, Zn, Rb, Sr, Y, Zr, Nb, and Ba; Ottawa-Carleton Geoscience Centre), produced by the fusion of sample with a  $\text{Li}_2\text{B}_4\text{O}_7$  flux; instrumental neutron-activation analysis (INAA: Sc, Co, Sb, Cs, 10 REE, Hf, Ta, W, Th, and U; Ecole Polytechnique) using -150 mesh rock powder; inductively coupled plasma-source spectrometry (ICP: Y plus 14 REE: Geoscience Labs, Ontario Geological Survey), and atomic absorption spectrometry (AAS: Li, Cu, Mo; Ottawa-Carleton Geoscience Centre) after a stepwise acid digestion ( $\text{HF-HClO}_4\text{-HCl}$ ); titration (WCT: Ottawa-Carleton Geoscience Centre) for the determination of ferrous iron (Wilson's method); ion-selective electrodes (ISE: Bondar-Clegg, Ottawa) for F and Cl; and combustion - infrared detection (C/IRD: Geological Survey of Canada) for total  $\text{H}_2\text{O}$  and  $\text{CO}_2$ . Wherever possible, the analysis of elements contained in minerals that are resistant to acid attack (e.g., cassiterite, tantalite: cf. Potts 1987) was undertaken in such a way as to avoid the need for acid digestion; for example, the analysis of samples for tin was performed by wavelength-dispersion X-ray-fluorescence spectrometry using 5-g pressed-powder pellets (XRF/PP: Bondar-Clegg, Ottawa). Data for international reference standard materials used during this study, together with the accepted values for these standards (cf. Abbey 1983), are contained in Table A1.

Some of the elements sought in this study are subject to significant overlap interferences in instrumental analysis. For example, with X-ray-fluorescence analysis, a first-order interference involving the overlap of the  $\text{YK}\alpha$  peak by  $\text{RbK}\beta$  occurs (Potts 1987). Consequently, other analytical techniques were utilized (for example, ICP spectrometry for Y) for such elements.

Oxygen and hydrogen isotope compositions of whole-rock samples were determined using standard procedures.  $\text{BrF}_3$  (Clayton & Mayeda 1963) and  $\text{ClF}_3$  (Borthwick & Harmon 1982) were used for the extraction of oxygen, which was subsequently converted to  $\text{CO}_2$  for analysis. Hydrogen was released from rock powder melted *in vacuo* in a Pt crucible, quantitatively collected

TABLE A1. GEOCHEMICAL COMPOSITIONS OF INTERNATIONAL REFERENCE STANDARD MATERIALS ANALYZED DURING THIS STUDY

	RGM-1/USGSIII (XRF/GD) This study (N=15)	REF1	G-2/USGSII (XRF/GD) This study (N=13)	REF1
Major element concentrations in weight percent				
$\text{SiO}_2$	73.01 (0.23)	73.47	69.52 (0.31)	69.22
$\text{TiO}_2$	0.28 (0.02)	0.27	0.50 (0.03)	0.48
$\text{Al}_2\text{O}_3$	13.66 (0.06)	13.80	15.37 (0.07)	15.40
$\text{Fe}_2\text{O}_3$	1.84 (0.02)	1.89	2.69 (0.02)	2.69
MnO	0.04 (0.00)	0.04	0.03 (0.00)	0.03
MgO	0.25 (0.11)	0.28	0.77 (0.08)	0.75
CaO	1.17 (0.01)	1.15	1.91 (0.02)	1.96
$\text{Na}_2\text{O}$	4.12 (0.12)	4.12	4.15 (0.14)	4.06
$\text{K}_2\text{O}$	4.31 (0.02)	4.35	4.47 (0.02)	4.46
$\text{P}_2\text{O}_5$	0.03 (0.01)	0.057	0.13 (0.01)	0.13
Trace element abundances in parts per million				
Rb	155 (3.3)	1557	172 (6)	170
Sr	104 (2.0)	100	496 (14)	489
Ba	848 (33)	800	1836 (48)	1900
Zr	3 (11)	36	92 (6)	84
Zn	214 (2)	200	328 (4)	300
Nb	5 (2)	9.47	8 (1)	13
Ferrous iron, $\text{H}_2\text{O}$ and $\text{CO}_2$ concentrations in weight percent				
FeO	8.67 (0.20)	8.63	$\text{H}_2\text{O}$ 0.43	0.43
	3.73 (0.21)	3.62	$\text{CO}_2$ 0.15	0.15
Trace element abundances in parts per million				
Cs	0.82 (0.13)	0.8	na	---
Sc	24.4 (0.51)	22	na	---
Co	58.9 (1.3)	61	na	---
Y	na	nr	25.2 (0.8)	28.2
La	81.4 (1.2)	82	15.5 (0.3)	16.7
Ce	151.7 (4.4)	152	38.8 (0.8)	41
Pr	na	nr	5.88 (0.16)	5.6
Nd	66.9 (2.5)	70	25.3 (0.8)	24
Sm	12.2 (0.29)	12	6.38 (0.21)	6.1
Eu	3.6 (0.13)	3.6	2.15 (0.10)	2.0
Gd	na	nr	6.39 (0.17)	7
Tb	1.34 (0.08)	1.3	0.88 (0.04)	1.0
Dy	6.7 (0.59)	nr	5.43 (0.20)	4.8
Ho	1.16 (0.11)	nr	1.03 (0.03)	0.94
Er	na	nr	2.64 (0.11)	2
Tm	0.28 (0.06)	nr	0.33 (0.02)	0.31
Yb	1.73 (0.06)	1.8	2.02 (0.06)	2.1
Lu	0.25 (0.03)	0.24	0.29 (0.01)	0.32
Sb	0.29 (0.05)	nr	na	---
Hf	5.4 (0.18)	5.4	na	---
Ta	6.0 (0.2)	5.5	na	---
W	31.8 (2.1)	29	na	---
Th	10.9 (0.4)	11	na	---
U	2.4 (0.14)	2.4	na	---

Note. The concentrations are the mean values of a given number of analyses (N) and the value in parentheses is the value of one standard deviation. Abbreviations are: XRF/GD = X-ray fluorescence spectrometry using glass disc; AAS = Atomic absorption spectrometry; INAA = Instrumental neutron activation analysis; ICP = inductively coupled plasma spectrometry; na = not analyzed; nr = not reported. REF1 = Abbey (1983); REF2 = Govenderaju (1984); REF3 = Gladney *et al.* (1983). The ICP data were produced by the Ontario Geological Survey Geoscience Laboratories (Doherty, pers. comm.).

as  $\text{H}_2\text{O}$ , and reacted with a uranium furnace at  $T = 800^\circ\text{C}$  to yield hydrogen for isotopic analysis (Bigeleisen *et al.* 1952, Friedman 1953). All of the isotopic data are reported, in ‰ relative to the Standard Mean Ocean Water (SMOW) reference standard (O'Neil 1986). During the course of this study, NBS 28 yielded a  $\delta^{18}\text{O}$  of 9.6‰ and NBS 30, a  $\delta\text{D}$  of -65‰.