Chemical composition of fluorapatite and associated minerals from skarn near Gatineau, Quebec

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Abstract

Sixteen fluorapatite specimens from regional skarns in granulite terrain were associated with Al-zoned diopside $\pm$ scapolite $\pm$ actinolite $\pm$ calcite (+ rare phlogopite). Apatite was low in Ce (ave. 0.19% Ce_2O_3) and enriched in *LREE* relative to *HREE* (La/Yb = 31 to 74 in 4 specimens). Some specimens showed small negative Eu anomalies and some crystals were zoned in *REE*. SrO averaged 0.36%. The mineral contained some carbonate (ave. 0.5% CO_2 in 5 specimens), appreciable silica (ave. 0.5%), and variable sulphate (0.1 to 1.2% SO_3). Excess charge due to S^{6+} was largely compensated by Si^{4+}. Chlorine was minor and F accounted for 75–98% of the F, Cl and OH ions. Apatite from marble lacking amphiboles and pyroxenes has a similar chemical composition, but apatite from later carbonatite and fenite contains more Ce and Sr. Apatite from Gatineau fenite, Gatineau carbonatite and world-wide siliceous igneous rock generally contains less S. Apatite from Gatineau skarns normally contains more Cl and less S than that from phosphorite. Magnesian marble was silicated to skarn by reaction with siliceous gneiss. Phosphorus, *REE*, and Sr were removed from nearby rocks and transported in aqueous, carbonated solutions containing minor amounts of F, Cl and S at granulite-facies conditions. Apatite and calcite precipitation took place in skarns and marble during the Grenville (Proterozoic) orogeny.

KEYWORDS: skarn, fluorapatite, diopside, rare earth elements, strontium, chlorine, granulite metamorphism.

Introduction

FLUORAPATITE has been known from south-western Quebec and southeastern Ontario since 1831 (Ingall, 1831). From 1876 to 1894 it was an important economic commodity, supplying super-phosphate plants with high-grade apatite for the fertilizer industry from a multitude of small surface workings. The largest single deposit (the Emerald Mine, 40 km northeast of Ottawa) produced at least 35 000 tonnes of apatite (Spence, 1920).

Early analyses of fluorapatite from this area were given by Hoffmann (1879), Spence (1920), Walker and Parsons (1926), and Dadson (1933). Un-fortunately, considerable iron oxide, Al_2O_3, MgO and, in some cases, excessive CO_2 cast doubt on the purity of samples taken from localities known to have closely associated hematite, phlogopite and calcite. Rare earth elements and SrO were not recorded.

The origin of its host rock, variously termed skarn, pyroxenite, metapyroxenite, and calc-silicate rock, has been debated since the 1870s. The first publications, in support of either igneous or metamorphic derivations, are summarized by de Schmid (1912), Spence (1920) and Landes (1938). However later publications by Shaw (1960), Shaw *et al.* (1963), Kretz (1960, 1977) and Villeneuve (1977) provide formidable geochemical evidence for the metamorphic theory. Today few geologists would contest development via this path.

In the Gatineau area, presently being mapped by the author, field evidence favours metaso-matic development through reaction of aluminous, magnesian marble with siliceous gneisses during regional metamorphism. The major silicate minerals (diopside, scapolite, actinolite, phlogopite) are also present within the marble and transitions occur from the lime–silicate rock (which we will call skarn) into silicated marble, with the gradual disappearance of silicates, into relatively pure carbonate strata. The general close association of skarn and marble is undeniable (Fig. 1) and irrefut-able intrusive relationships and textures in the
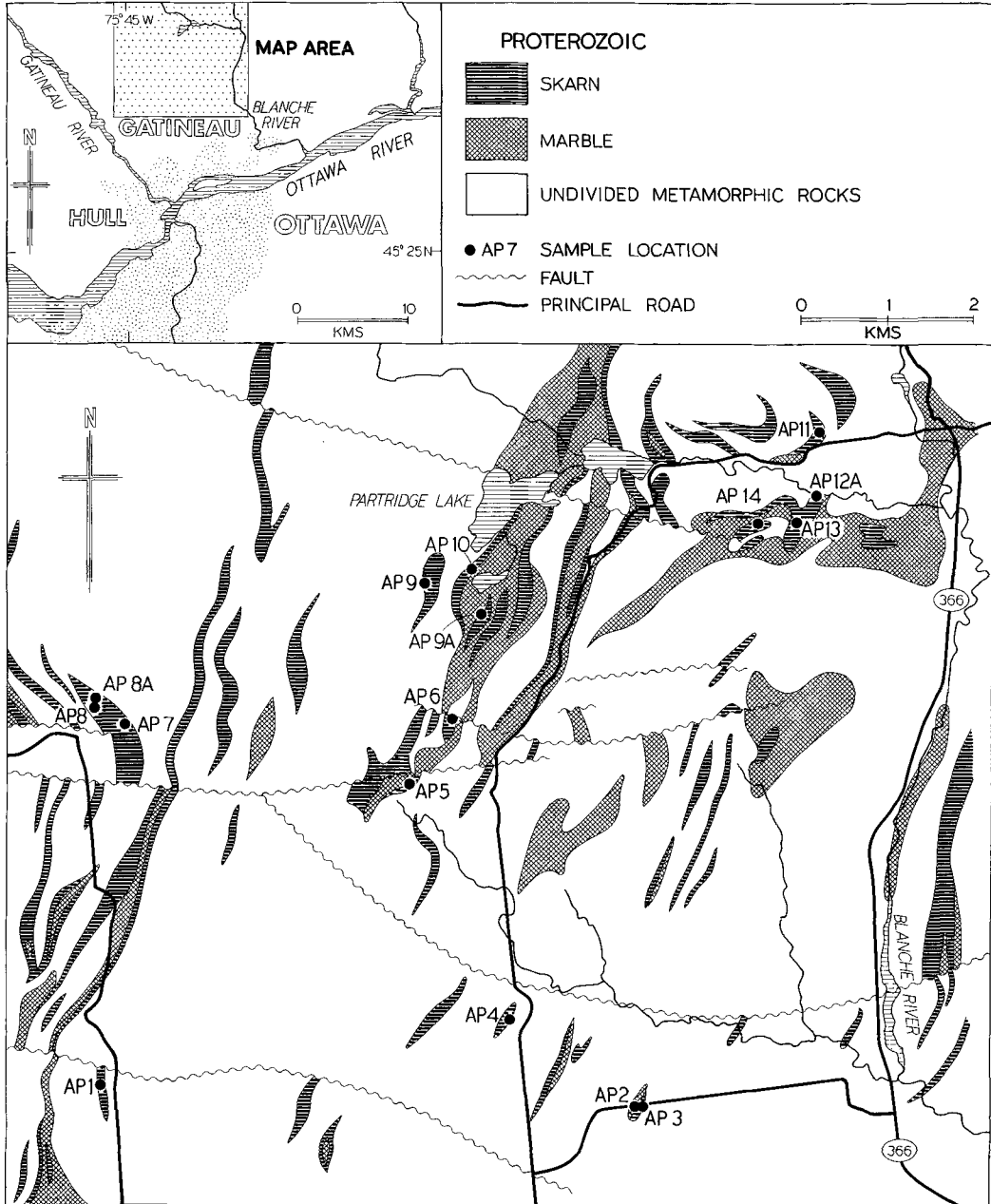
FIG. 1. Distribution of marble and skarn, and location of sample sites in the area north of Gatineau, Quebec. The site of AP 15 is 8.2 km SSE of the centre of the map, that of AP 17 is 8.6 km NNE of the centre.

skarn are entirely lacking. These features have been noted in similar rocks elsewhere, e.g. Slyudyanka, USSR (Khorzhinsky, 1947), Tranomaro, Madagascar (Moine et al., 1985), and North Burgess Township, Ontario (Currie, 1951). The rocks are therefore believed to be skarns, in the broad sense of the Metamorphic Nomenclature Committee of Canada (Shaw, 1957), which includes products of both regional and contact metamorphism, but while this general mode of origin has foundation,

details of metasomatic development, the role of P and F, and the ultimate origin of the apatite, are as much a mystery today as they were in 1920.

In an attempt to define the apatite composition more precisely and to solve some of the problems of origin, 18 fluorapatite-rich specimens were collected for research from a restricted area north of Gatineau, Quebec. Sixteen of these are located in Fig. 1 and two (AP 15 and AP 17) are just outside the map area. Sixteen of the specimens are from skarns but one (AP15) is from marble lacking lime silicates and one (AP12A) is from a calcite-richterite–fluorapatite-(ferri)phlogopite carbonatite. Specimendetails are given in Table 1. Included are township, range and lot number, which enable collation with de Schmid (1912), Spence (1920) and Hogarth (1981). This paper will emphasize the chemical mineralogy of the apatite-rich specimens but future research will involve fluid-inclusion and isotope studies.

Local geology. The geology of the Gatineau area has been described by Hogarth (1981) and Hogarth and Rushforth (1986) and the following summary is taken mainly from these works. Coarse-grained skarns are interlayered with magnesian marble, quartzite, biotite gneiss, hornblende gneiss, diopside($\pm$hypersthene) gneiss, and leucocratic (granite) gneiss. The whole assemblage attained granulite-facies conditions during regional metamorphism of the Grenville orogeny (thermal peak at *c.* 1100 Ma b.p.). Pyroxene geothermometry, using the equations of Kretz (1982) and muscovite–sillimanite geobarometry using the curves of Evans (1965) and Kerrick (1972), suggest temperatures of 670 °C and 7 kbar. Local melts resulted in granite pegmatite sills and dykes. All of these rocks were locally fenitized and cut by small carbonatites of late Grenvillian age (*c.* 900–1000 Ma b.p.). Post-metamorphic dykes (dolerite, basalt, syenite, minette, kersantite) are small, comparatively uncommon and bear no spatial relationship with the skarns. These rocks are cut by faults of the Ottawa–St. Lawrence fracture system, which has been periodically reactivated since Precambrian time.

The normally conformable skarns are locally squeezed into fractures, thereby acquiring a pseudo-igneous aspect. The dominant mineral is an iron-bearing, aluminous, diopsidic pyroxene but, in many skarns, calcite, tremolite–actinolite, apatite and, locally, perthitic microcline are common. Titanite and pyrite are ubiquitous but minor.

Within many skarns are layers, veins, lenses and irregular masses of apatite, phlogopite and pink calcite, combined in various proportions (the 'vein dykes' of Ellsworth, 1932). It is these bodies that supplied the 19th and 20th century miners with 'phosphate' and 'mica' (de Schmid, 1912; Spence, 1920) and provided all the skarn apatites of this study.

A notable feature of this apatite is its large grain size. Commonly crystals are centimetric but some (from localities of specimens AP 7, AP 9 and AP 14) are decimetric. Individual masses of roughly terminated to anhedral crystals ('massive apatite'; localities of AP 1, AP 2, AP 4, AP 5, AP 8, AP 8A, AP 9A and AP 10) may attain tonnes of relatively pure mineral. Sand-sized grains, aggregated into semi-coherent masses ('sugar apatite'; localities of AP 11(m) and AP 17), some containing apatite 'porphyroblasts' (AP 11(p)), are found along faults and within shear zones and may represent pre-existing apatite that was granulated during metamorphism. No evidence of secondary apatite was seen and post-Grenvillian mineralization within the skarns is apparently absent.

Chemical composition

Experimental procedure. Lithium borate glasses were analysed by R. Hartree (Univ. Ottawa) by X-ray fluorescence (XRF) using a rhodium tube operated at 60 kV and 40 mA. Analyses were standardized against Moroccan phosphorite BCR No. 32 (Serrini and Haemers, 1980; Colinet *et al.*, 1983). Rare earth elements (*REE*) were determined by J. Loop (Univ. Ottawa) by direct current plasma–atomic emission spectrophotometry (DCP). Some analyses were checked by C. Pride (formerly Univ. Ottawa) by neutron activation analysis (INAA).

Polished thin sections were analysed by T. N. Solberg (Virginia Polytechnic Institute) with an ARL-SEMQ, 9-spectrometer microprobe (EM) using procedures and standards similar to those described by Hogarth *et al.* (1985). A number of grains were analysed with automated traverses. Ti and Al, not recorded in Table 3, were sought by EM, but were below the detection limits (0.03 and 0.05 wt. % respectively) in all cases. U and Th were not checked, but scintillation counter measurements at the occurrences showed that apatite was virtually free of these elements.

Six samples, selected for CO_2 analysis, were carefully hand picked under the binocular microscope. Grains of 5 concentrates were water-clear, but grains of AP 12A were slightly milky. Carbon dioxide was evolved during acid digestion and determined by IR spectroscopy (precision $\pm 0.1\%$). In one sample (AP 17) Cl was checked by ion-exchange/chromatography (EM = 0.29; chromatography 0.291%). CO_2 and Cl determinations were made by P. Belanger for G. Hall (Geological Survey of Canada).

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Table 1. Information on localities.

Spec. No.	Township* Range/Lot	Brief description of apatite and skarn**	Mine	Production of apatite	Colour of apatite#	Reference
AP 1	H,X/4a	Massive Ap layer 20x1 m in Di-Act skarn	Barrett	1050T	br-rd	Spence (1920, p.83) Hogarth (1981, p.22)
AP 2	T,IV/20d	Massive Ap, irregular mass 15x8 m in Di skarn	St. Thomas	>200T	ch-br	
AP 3	T,IV/20d	Dissem. Ap in Fst-Scp skarn		none	aq	
AP 4	T,IV/23b	Massive Ap, lens 28x3 m in Di-Phl skarn	Gahagan	1200T	gy	Hogarth (1981, p.24)
AP 5	T,VI/24a	Massive Ap-Cal layer 30x2 m in Di-Phl skarn	Canada Phosphate	<200T	or	Hogarth (1981, p.25)
AP 6	T,VII/23b	Dissem. Ap in Fst-Scp-Cal skarn	Flynn	<100T	bl-gn	Hogarth (1981, p.25)
AP 7	H,XIII/2	Crystals Ap in Cal-Phl lens, Di-Scp skarn	Ward	15T	gn	Hogarth (1981, p.23)
AP 8	H,XIII/3b	Massive Ap, irreg. mass 9x5 m in Di-Phl skarn	O'Brien (S)	>200T	gn	de Schmid (1912, p.99)
AP 8A	H,XIII/3b	Massive Ap, irreg. mass 20x5 m in Di-Scp skarn	O'Brien (N)			
AP 9	T,VIII/23b	Crystals Ap in Cal lens 9x2 m, in Di-Scp skarn	Pennock	<100T	gn	
AP 9A	T,VIII/22	Massive Ap lens 20x2 m in Fst-Scp skarn	Wallingford Lake (S)	>200T	gn	Hogarth (1981, p.26)
AP 10	T,VIII/22	Massive Ap in Cal layer 30x1 m in Di-Scp skarn	Wallingford Lake (N)		br-rd	
AP 11	T,IX/14b	"Sugar Ap" lens 40x1 m in Di-Phl skarn	Perkins	>120T	bl-gn	Spence (1920,pp.91-2)
AP 12A	T,VIII/15a	Ap-Cal-Richterite-Phl carbonatite	Pioneer	>200T	gn-yw	Hogarth (1983)
AP 13	T,VIII/15b	Ap lens, 120x5 m in Di-Phl skarn	Phosphate King	8500T	pl	Spence (1920, pp.90-91)
AP 14	T,VIII/16a	Ap-Cal lens in Di-Phl skarn	Wallingford	4000T	gn	Spence (1920, p.91)
AP 15	T,II/16b	Ap crystals in marble layer		none	gn-wh	Hogarth (1983)
AP 17	T,XI/12b	"Sugar Ap" layer 1 m thick in Di skarn	Fidelity	1000T	gn-gy	Hogarth (1983)

*H = Hull Township, T = Templeton Township. **Mineral abbreviations: Act = actinolite, Ap = apatite, Cal = calcite, Di = diopside, Fst = fassaite, Phl = phlogopite, Scp = scapolite. #Colour in hand specimen; abbreviations: aq = aquamarine, bl-gn = blue-green, br-rd = brick-red, ch-br = chocolate-brown, gn = green, gn-gy = greenish grey, gn-wh = greenish white, gn-yw = greenish yellow, gy = grey, or = orange, pl = purple.

Ca' ions. In this paper Ca' will refer to all ions in the Ca sites (La, Ce ... K), P' to all ions in the P site (S, P, Si, C), and F' to all ions in the F site (F, Cl, OH). Chemical compositions of apatite will involve analyses of all skarn apatites of Table 1. Comparison will be made with apatite from metamorphic, igneous and sedimentary environments. Comparison will also be made with apatite from other skarn deposits and from nearby carbonatites and fenites.

Ca' ions, other than Ca, are not important. Strontium, *REE* and Na play a minor role. Their abundance overlaps the range of both igneous and metamorphic rocks (Table 4) although granitic rocks tend to have more Na. Strontium overlaps the range of apatite from Canadian radioactive and Slyudyanka skarns; Ce tends to lie between these skarn types. Fluorapatites from Gatineau carbonatites and fenites are very different, showing notable concentrations of Sr, Ce and Na and commonly displaying simple or rhythmically zoned Sr and Ce in crystals (Hogarth *et al.*, 1985). Apatite from marble (AP 15) has a composition intermediate between the skarns and carbonatite.

REE distributions in most whole-rock analyses (Table 2) may be attributable to apatite with possible modification by impurities, but in the virtually pure specimens AP 7 and AP 14 (examined in crushed grains and thin sections) the distributions may represent apatite alone. Similarly, no potential *REE* carriers were observed in the rather pure specimens AP 8 and AP 11 (apatite with very minor calcite). These four specimens were characterized by a variable *REE* content (ΣREE 690 to 4530 ppm), moderately high La/Yb ratios (31 to 74) and low to absent, negative Eu anomalies (δ Eu = 0.6 to 1.0). *REE* abundances are shown in Fig. 2. Variable *REE* content is also apparent in electron microprobe analyses (Table 3; Ce_2O_3 0.05 to 0.76%) and *LREE* dominance reflected in the very low Y_2O_3 values (max. 0.08%). *LREE* enrichment (compared to *HREE*) and negative Eu anomalies are also evident from data of apatite from regional diopsidic skarns of Aldan and Slyudyanka, USSR, in Khrostova (1962), Mikhailov and Mineev (1970) and Moskvitina and Smirnov (1977).

Fe, Mg and Mn are even less important. Cer-

Table 2. Analyses of apatite and apatite-rich rock*.

Spec. No. % Ap	AP 1 95	AP 2 90	AP 3 15	AP 4 75	AP 5 75	AP 6 20	AP 7 c. 100	AP 8 >98	AP 9 90	AP 10 >95	AP 11 >98	AP 12A 85	AP 13 >95	AP 14 c. 100	AP 15 15
SiO_2 %	2.35	2.70	40.87	11.56	1.91	38.66	1.17	0.79	1.00	1.54	1.01	0.25	2.27	0.72	0.59
TiO_2	0.02	0.00	0.50	0.00	0.01	0.48	0.00	0.00	0.00	0.01	0.00	0.01	0.00	0.00	0.00
Al_2O_3	0.35	0.05	6.05	0.06	0.05	5.58	0.02	0.13	0.03	0.45	0.03	0.06	0.02	0.03	0.02
FeO_T	0.94	0.44	3.95	0.13	1.30	2.91	0.08	0.05	0.06	0.43	0.07	0.12	0.13	0.06	0.53
MnO	0.03	0.00	0.12	0.00	0.02	0.07	0.01	0.00	0.00	0.00	0.02	0.01	0.01	0.01	0.28
MgO	0.97	0.86	10.30	0.11	0.11	10.89	0.11	0.09	0.15	0.34	0.10	0.15	0.23	0.19	0.02
SrO	0.32	0.33	0.08	0.23	0.46	0.07	0.34	0.32	0.45	0.15	0.25	2.34	0.24	0.26	0.08
BaO	0.02	0.01	0.01	0.01	2.06	0.02	0.00	0.00	0.01	0.00	0.00	0.16	0.00	0.01	0.01
CaO	51.2	49.6	26.4	45.5	49.7	29.3	52.8	52.3	51.6	53.15	55.5	53.45	53.4	53.9	44.1
Na_2O	0.0	0.0	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
K_2O	0.2	0.0	0.3	0.0	0.0	0.3	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0
P_2O_5	38.2	37.25	5.6	33.9	31.9	7.8	39.5	38.2	36.9	37.9	40.8	37.1	40.8	39.1	5.4
SO_{3T}	0.3	0.6	0.5	0.6	0.7	0.3	0.3	0.6	0.75	0.75	0.4	0.4	0.5	0.8	0.1
Zr ppm	75	75	450	50	100	140	75	70	100	60	50	430	60	60	<10
Zn	10	<10	190	<10	<10	50	<10	<10	<10	10	<10	<10	<10	<10	20
V	20	20	70	15	40	50	10	10	<10	20	10	<10	30	10	<10
Y	240	290	60	200	120	80	250	300	230	120	40	130	100	190	70
La	777	1010	62	785	545	174	633	972	596	480	152	1700	765	305	197
Ce	1590	2100	1156	1650	1100	422	1520	2110	1700	1120	325	4110	1890	672	460
Nd	799	942	50.7	714	623	213	845	1070	701	480	157	2130	780	335	228
Sm	124	187	4.9	135	82.5	26.0	122	152	93.6	69.4	25.5	257	78.0	59.4	33.0
Eu	21.4	22.5	1.5	17.1	12.7	2.6	17.7	22.9	10.3	16.0	4.1	41.2	[17.3]	15.6	4.95
Gd	66.0	108	[5.2]	[28.2]	39.8	5.2	65.8	98.4	55.9	34.5	10.4	63.2	34.4	30.3	21.0
Dy	54.6	58.1	5.3	38.5	26.6	2.1	49.2	64.0	33.5	24.5	8.4	25.8	20.9	18.9	11.4
Er	23.1	22.0	2.2	12.8	10.3	1.1	18.8	23.1	13.6	6.9	2.8	6.5	8.2	6.9	5.9
Yb	18.2	17.6	1.4	19.3	13.7	1.1	20.7	13.6	12.5	5.2	2.05	3.8	7.6	4.8	4.6
$\sum$ REE	3470	4470	290	3400	2450	850	3290	4530	3220	2240	690	8340	3600	1450	970
La/Yb	43	57	44	41	40	160	31	71	48	92	74	450	100	60	43
δEu	0.71	0.45	1.1	0.65	0.68	0.45	0.61	0.58	0.24	1.0	0.78	1.0	-	1.0	0.58

*REE by DCP; other constituents by XRF. δ Eu = Eu measured/Eu interpolated Sm + Gd. Numbers in square brackets indicate interpolated values.

tainly, part of the Fe in some previous analyses, and in the present XRF analyses, is due to hematite impurity. Intergranular red hematite in AP 1 and AP 10, and intragranular specularite in AP 2, account for the red and brown colours of hand specimens. Mn attains significant importance in AP 3 and may account for the striking blue colour in hand specimen.

Phosphoritic apatite is distinct from all other types in its very low Sr and Ce contents. Liu (1981) also notes that sedimentary apatite is characterized by appreciable Ba, an element prominent at the trace level (200–800 ppm) in high-grade phosphorite. However, these levels of Ba are below the detection limit of the probe.

P′ ions. Gatineau apatites tend to be high in Si, but similar contents have been recorded elsewhere from both igneous and metamorphic environments (Petrov and Zhuravel, 1971). Sulphur is also high but varies greatly from one specimen to the next and even within an individual crystal. Thus the core of AP 9 (variety AP 9(h), Table 3; birefringence 0.008) is enriched in S, along with Si, Ce and Na, relative to a narrow overgrowth (AP 9(1), Table 3; birefringence 0.002). The more highly birefringent core may reflect a P → C substitution (McConnell, 1973), C tending to balance the number of P′ ions, presently at a 2.2% deficit with respect to Ca′. The major variants of this specimen are plotted for step counts across part of a crystal in Fig. 3. A zoned crystal in AP 8A was also traversed but showed a much less pronounced chemical variation (Fig. 4) and no change in birefringence. The large variation within triplet analyses is thought to be caused mainly by approach of detection limits, but part
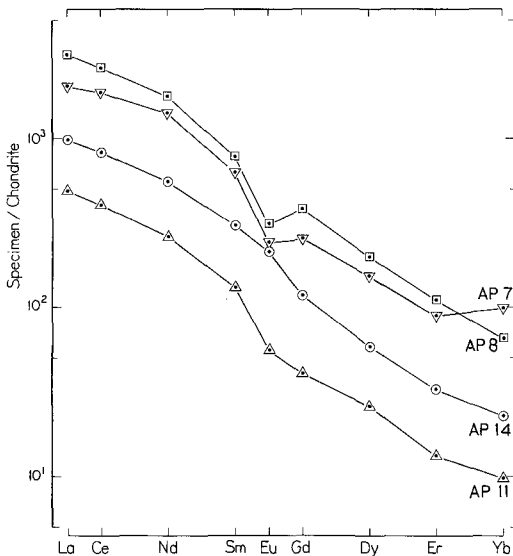

FIG. 2. Chondrite-normalized *REE* abundances in apatite from skarns near Gatineau, Quebec.

Table 3. Chemical composition* of apatite from Gatineau, Quebec.

	AP 1	AP 2	AP 3	AP 4	AP 5	AP 6	AP 9A	AP 9(h)	AP 9(1)	AP 9A	AP 10	AP 11(p)	AP 11(m)	AP 12A	AP 13	AP 15	AP 17
#n	3	8	5	13	7	3	9		6	6	7	18	9	7	7	8	7
La_2O_3	0.11(0.05)	0.09(0.05)	0.02(0.02)	0.06(0.03)	0.05(0.02)	0.09(0.03)	0.04(0.02)	0.36(0.04)	0.12(0.03)	0.03(0.02)	0.06(0.04)	0.06(0.03)	0.02(0.01)	0.12(0.04)	0.06(0.02)	0.08(0.04)	0.03(0.02)
Ce_2O_3	0.22(0.11)	0.20(0.08)	0.05(0.02)	0.15(0.08)	0.13(0.07)	0.21(0.02)	0.11(0.06)	0.76(0.29)	0.41(0.05)	0.07(0.02)	0.14(0.04)	0.16(0.08)	0.05(0.04)	0.34(0.04)	0.13(0.07)	0.25(0.12)	0.09(0.02)
Nd_2O_3	0.10(0.05)	0.08(0.04)	0.03(0.02)	0.08(0.01)	0.04(0.02)	0.13(0.03)	0.06(0.02)	0.27(0.08)	0.26(0.06)	0.02(0.02)	0.06(0.04)	0.06(0.04)	0.03(0.03)	0.16(0.03)	0.06(0.01)	0.15(0.07)	0.03(0.02)
Y_2O_3	0.02(0.02)	0.03(0.02)	0.03(0.01)	0.03(0.01)	0.00	0.03(0.02)	0.02(0.01)	0.06(0.02)	0.08(0.03)	0.01(0.01)	0.01(0.01)	0.06(0.04)	0.01(0.005)	0.02(0.02)	0.02(0.01)	0.02(0.01)	0.01(0.01)
Fe_2O_3	0.08(0.002)	0.08(0.006)	0.07(0.01)	0.08(0.003)	0.08(0.004)	0.05(0.04)	0.02(0.01)	0.08(0.02)	0.08(0.002)	0.01(0.006)	0.04(0.02)	0.08(0.004)	0.07(0.003)	0.09(0.004)	0.08(0.004)	0.06(0.005)	0.07(0.01)
MnO	0.01(0.005)	0.00	0.47(0.01)	0.01(0.01)	0.00	0.01(0.004)	0.00	0.01(0.01)	0.01(0.005)	0.01(0.01)	0.00	0.01(0.01)	0.01(0.005)	0.01(0.01)	0.01(0.01)	0.00	0.01(0.01)
BaO	0.01(0.01)	0.01(0.01)	0.00	0.00	0.00	0.00	0.01(0.01)	0.01(0.01)	0.01(0.01)	0.02(0.01)	0.00	0.00	0.02(0.01)	0.08(0.02)	0.00	0.00	0.01(0.01)
SrO	0.41(0.04)	0.41(0.03)	0.32(0.02)	0.31(0.05)	0.38(0.02)	0.28(0.04)	0.28(0.03)	0.61(0.09)	0.58(0.06)	0.19(0.02)	0.19(0.04)	0.26(0.03)	0.28(0.04)	2.92(0.17)	0.29(0.03)	0.70(0.04)	0.27(0.02)
MgO	0.08(0.001)	0.09(0.003)	0.06(0.002)	0.09(0.003)	0.09(0.003)	0.07(0.001)	0.08(0.01)	0.09(0.01)	0.09(0.003)	0.06(0.002)	0.06(0.002)	0.07(0.004)	0.07(0.004)	0.09(0.002)	0.09(0.005)	0.07(0.004)	0.09(0.005)
CaO	54.45(0.22)	53.81(0.27)	55.40(0.34)	55.20(0.31)	55.45(0.38)	54.84(0.81)	55.28(0.24)	53.93(0.16)	54.48(0.27)	54.77(0.50)	54.68(0.55)	55.59(0.23)	55.19(0.23)	52.33(0.19)	55.09(0.30)	54.66(0.20)	54.37(0.21)
Na_2O	0.07(0.01)	0.07(0.01)	0.01(0.005)	0.06(0.02)	0.04(0.01)	0.32(0.02)	0.01(0.01)	0.29(0.06)	0.10(0.03)	0.05(0.01)	0.03(0.01)	0.04(0.01)	0.02(0.01)	0.19(0.01)	0.12(0.03)	0.09(0.03)	0.01(0.01)
K_2O	0.02(0.001)	0.02(0.001)	0.02(0.001)	0.02(0.001)	0.02(0.001)	0.01(0.001)	0.02(0.001)	0.02(0.001)	0.02(0.002)	0.01(0.001)	0.01(0.001)	0.03(0.001)	0.02(0.01)	0.02(0.002)	0.02(0.002)	0.01(0.002)	0.02(0.001)
SiO_2	0.14(0.08)	0.42(0.16)	0.18(0.04)	0.28(0.11)	0.22(0.05)	0.73(0.04)	0.18(0.05)	0.73(0.02)	0.32(0.05)	0.35(0.05)	1.05(0.10)	0.46(0.06)	0.10(0.07)	0.02(0.004)	0.13(0.07)	0.33(0.10)	0.25(0.07)
P_2O_5	42.05(0.20)	40.14(0.70)	41.00(0.39)	40.63(0.50)	40.49(0.30)	41.14(0.41)	41.78(0.40)	39.61(0.45)	41.19(0.24)	41.48(0.26)	37.89(0.11)	40.53(0.56)	40.91(0.56)	40.09(0.31)	41.11(0.37)	41.33(0.58)	40.78(0.48)
SO_3	0.20(0.04)	0.35(0.08)	0.22(0.02)	0.22(0.08)	0.20(0.05)	0.32(0.08)	0.15(0.05)	0.82(0.09)	0.10(0.04)	0.50(0.07)	1.18(0.08)	0.57(0.07)	0.17(0.07)	0.19(0.03)	1.23(0.03)	0.34(0.10)	0.26(0.06)
CO_2		0.2		0.55							1.2			1.6	0.3		0.2
F	3.12(0.09)	3.45(0.27)	3.72(0.03)	3.43(0.19)	3.33(0.29)	3.23(0.26)	3.27(0.20)	3.31(0.26)	3.36(0.23)	3.61(0.08)	3.31(0.04)	3.23(0.11)	3.32(0.17)	3.57(0.06)	2.85(0.15)	3.00(0.16)	2.98(0.16)
H_2O	0.13(0.01)	0.28(0.03)	0.04(0.002)	0.01(0.005)	0.15	0.07(0.007)	0.06(0.02)	0.00	0.00	0.01(0.004)	0.16	0.58(0.03)	0.51(0.04)	0.12	0.26(0.03)	0.22(0.01)	0.29(0.03)
$-O{\equiv}F,Cl$	0.28	0.04		0.15		0.26	0.22	0.17	0.14	0.08		0.08	0.04		0.39	0.30	0.27
Total	100.15	98.21	100.06	99.37	99.27	100.91	100.26	99.70	99.97	99.77	98.89	100.34	99.43	100.46	100.55	100.30	98.72

Ions per 6 (Si + P + S + C)

	AP 1	AP 2	AP 3	AP 4	AP 5	AP 6	AP 9A	AP 9(h)	AP 9(1)	AP 9A	AP 10	AP 11(p)	AP 11(m)	AP 12A	AP 13	AP 15	AP 17
La	0.007	0.006	0.001	0.004	0.003	0.006	0.002	0.020	0.008	0.002	0.004	0.004	0.001	0.007	0.004	0.005	0.002
Ce	0.013	0.013	0.003	0.009	0.008	0.013	0.007	0.043	0.026	0.003	0.008	0.010	0.003	0.021	0.008	0.015	0.006
Nd	0.006	0.005	0.002	0.006	0.002	0.008	0.004	0.017	0.016	0.001	0.003	0.004	0.002	0.009	0.004	0.009	0.002
Y	0.001	0.003	0.003	0.003	0.000	0.003	0.002	0.005	0.007	0.001	0.001	0.002	0.001	0.002	0.000	0.002	0.001
Fe	0.011	0.011	0.010	0.011	0.012	0.007	0.011	0.012	0.011	0.006	0.005	0.011	0.010	0.012	0.011	0.008	0.010
Mn	0.001	0.000	0.007	0.001	0.000	0.001	0.000	0.001	0.001	0.001	0.000	0.001	0.001	0.001	0.001	0.000	0.001
Ba	0.001	0.001	0.000	0.000	0.000	0.000	0.001	0.001	0.001	0.001	0.000	0.000	0.001	0.005	0.000	0.000	0.001
Sr	0.040	0.041	0.032	0.030	0.038	0.027	0.027	0.061	0.057	0.018	0.019	0.026	0.028	0.280	0.028	0.068	0.027
Mg	0.020	0.023	0.015	0.023	0.023	0.017	0.020	0.023	0.023	0.015	0.015	0.018	0.018	0.022	0.022	0.018	0.023
Ca	9.753	9.901	10.159	9.970	10.288	9.746	9.965	9.940	9.931	9.823	9.711	10.152	10.177	9.271	9.831	9.877	9.918
Na	0.023	0.023	0.003	0.020	0.013	0.103	0.003	0.097	0.033	0.016	0.008	0.013	0.007	0.061	0.039	0.029	0.003
K	0.004	0.007	0.004	0.004	0.004	0.002	0.004	0.004	0.004	0.002	0.002	0.007	0.004	0.004	0.004	0.002	0.004
Σ	9.88	10.02	10.24	10.08	10.39	9.933	10.05	10.23	10.12	9.89	9.78	10.25	10.25	9.70	9.95	10.03	10.00
Si	0.023	0.072	0.031	0.047	0.038	0.121	0.030	0.126	0.054	0.059	0.175	0.078	0.017	0.003	0.022	0.056	0.043
P	5.952	5.836	5.941	5.798	5.936	5.777	5.951	5.769	5.933	5.879	5.407	5.849	5.961	5.612	5.881	5.901	5.878
S	0.025	0.045	0.028	0.028	0.026	0.102	0.019	0.106	0.013	0.063	0.147	0.073	0.022	0.024	0.029	0.043	0.033
C		0.047		0.127							0.272			0.361	0.068		0.046
Σ	6	6	6	6	6	6	6	6	6	6	6	6	6	6	6	6	6
F	1.650	1.874	2.014	1.829	1.824	1.694	1.740	1.801	1.808	1.911	1.732	1.741	1.807	1.867	1.501	1.600	1.505
Cl	0.037	0.081	0.012	0.003	0.000	0.020	0.017	0.000	0.000	0.003	0.055	0.168	0.149	0.000	0.073	0.063	0.084
OH	0.313	0.045	0.000	0.168	0.176	0.286	0.243	0.199	0.192	0.086	0.202	0.091	0.044	0.133	0.426	0.337	0.211
Σ	2	2	2.03	2	2	2	2	2	2	2	2	2	2	2	2	2	2

*Analyses by electron microprobe except for CO_2 (by acid evolution/IR detection). H_2O determined by calculation. Oxygen equivalents of F, Cl deducted in the totals. #n = number of analyses averaged. Numbers in parentheses are standard deviations for several closely spaced analyses.
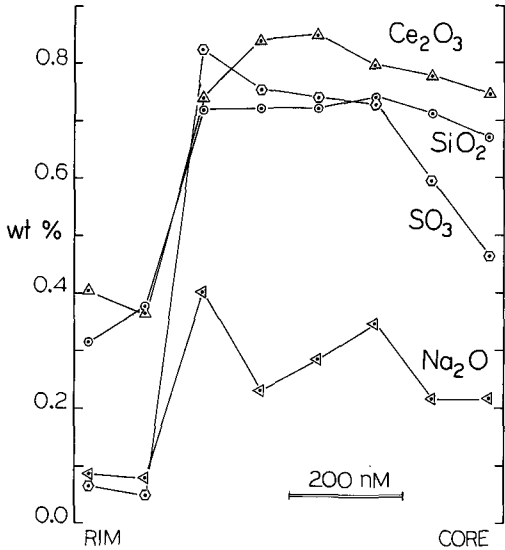
FIG. 3. Automated microprobe traverse from the overgrowth ('rim') into the core of an apatite crystal in AP 9.

may be real variations along the traverses. Like Si, S in apatite does not seem to be distinctive for metamorphic or igneous rocks, but phosphorites seem to be particularly sulphate-rich, and siliceous igneous rocks, sulphate-poor (Table 4). The ions S^{6+} and Si^{4+} are present at approximately the same concentration in the high-sulphate apatites (anal. AP 6, AP 9(h) and AP 10, Table 3), suggesting major substitution of the type $P^{5+}+P^{5+} \rightarrow S^{6+}+Si^{4+}$.

Carbonate, generally considered to belong in the P group (McConnell, 1973), is consistently present in small amounts. This anion seems to characterize apatite from all environments considered in Table 4, being highest in phosphorite and lowest in
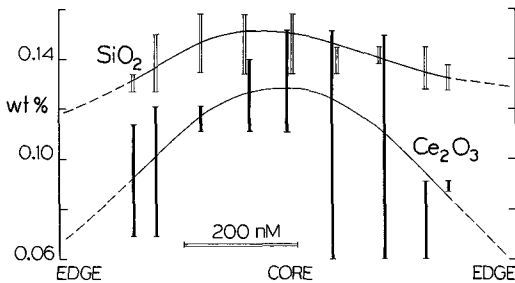
FIG. 4. Zoning in an apatite crystal from AP 8A. Stepcounted, microprobe analyses. Integrations were in threes, except on the flanks where they were in pairs of analyses.

siliceous igneous rocks. It has been suggested that C is coordinated with 3 O and 1 F or OH (replacing O), the PO_4 and $CO_3(F, OH)$ ions having identical charge. This hypothesis explains excess F and OH in many carbonate-apatites (such as AP 3, Table 3; Borneman-Starynkevich and Belov, 1953).

F' ions. Fluorine, a characteristic element in apatite from all environments, fills 75–98% (ave. 87%) of the F-site in apatite from the Gatineau skarns. Overfilling of F in this site, common in apatite from Gatineau fenite and from phosphorite, was rare in these specimens.

Chlorine averages 0.16%. It attains 0.58% (8.2% of F' ions) in porphyroblasts (AP 11(p)) and 0.51% (7.1% of F' ions) in the granoblastic matrix (AP 11(m)) of specimen AP 11. In contrast, skarn apatite from 'Elmsley, Ontario' contains 0.88% Cl (Dadson, 1933), from Bob's Lake, Ontario, 6.2% Cl (Hounslow and Chao, 1970), and from Kragero, Norway, 1.40–6.24% Cl (Morton and Catanzaro, 1964). Apparently a miscibility gap exists from near the composition of Elmsley apatite (Cl 12% of F' ions) to near that of the Bob's Lake apatite (Cl 91% of F' ions; Taborszky, 1972; Hounslow and Chao, 1970). Chlorine in the radioactive skarns, local carbonatites and local fenites, is almost absent.

Chlorine in apatite is regarded as a sensitive indicator of the genetic type of occurrence by Liu (1981), who gives F:Cl ratios of 180:1(wt) for sedimentary apatite, 17.5:1 for igneous apatite and 32–8.6:1 for pegmatitic apatite. These values are in rough agreement with those of Table 4 and exclude Gatineau skarn apatite from the phosphorite type (averaging $\frac{1}{5}$ the Cl in apatite from Gatineau skarns).

It is also apparent that apatite from both igneous and metamorphic rocks varies considerably in Cl content (Table 4). In metamorphic rocks this can be ascribed to the composition of the protolith and conditions of formation. In igneous rocks the F:Cl ratio is controlled by associated minerals and stage of crystallization during differentiation (Fershtater et al., 1986).

Associated silicates. Specimens were collected from the apatite bodies themselves and most were nearly free of minerals from the surrounding silicate skarns. Phlogopite, commonly associated with apatite, was not present in the polished thin sections, although it was the principal economic product at sample sites AP 13 and AP 14, during reworking of the mines in the present century. Phlogopite associated with apatite in skarns 20 km to the west, contained 0.8 to 9.5% FeO_T and 1.2 to 3.6% F (Hogarth, unpublished). Feldspars, common in skarns elsewhere in the region, were lacking in skarns at the sample sites.

The composition of pyroxene in polished thin

Table 4. Abundance of selected components of fluorapatite from various rocks and localities.

wt% range / wt% average* (No. of analyses)

Rock type, locality	SrO	Ce$_2$O$_3$**	Na$_2$O	SO$_3$	F	Cl	CO$_2$	Reference
Skarn, Gatineau	0.19-0.58 (16) / 0.34	0.05-0.41 (16) / 0.15	0-0.32 (16) / 0.05	0.10-1.18 (16) / 0.31	2.85-3.72 (13) / 3.31	0-0.51 (13) / 0.16	0.2-1.2 (5) / 0.5	This paper
Radioactive skarn, Canada	0.18-0.28 (3) / 0.24	0.38-1.23 (3) / 0.86	0.18-0.28 (2) / 0.23	0.03-0.72 (2) / 0.37	3.30-4.0 (4) / 3.7	0-0.02 (3) / 0.01	0.57-2.10 (4) / 1.07	#
Skarn, Slyudyanka	0.08-0.29 (2) / 0.18	0.003-0.01 (2) / 0.006	0.27	0.63	2.88	0.12	0.48	Cruft (1966, anal. 14), Bulakh (1984)
Marble, Gatineau-AP 15	0.70	0.25	0.09	0.29	3.00	0.22		This paper
Carbonatite, Gatineau	1.82-5.04 (9) / 4.06	0.34-1.72 (9) / 1.10	0.19-0.74 (9) / 0.52	0.06-0.20 (9) / 0.14	3.15-3.80 (9) / 3.52	0-0.01 (9) / 0.00		# #
Fenite, Gatineau	1.58-6.76 (9) / 4.17	0.72-1.71 (9) / 0.96	0.30-0.56 (9) / 0.41	0.05-0.17 (9) / 0.13	3.31-4.15 (9) / 3.79	0-0.02 (9) / 0.01	1.6	Hogarth (unpublished)
Metamorphic rock, world-wide	0-1.6 (25) / 0.34	0.01-1.63 (25) / 0.36						Cruft (1966)
Metamorphic rock, world-wide	0.02-0.36 (10) / 0.18	0.02-1.52 (9) / 0.33	0.04-0.98 (10) / 0.27	0.06-2.58 (22) / 0.79	0.10-3.72 (24) / 2.60	0.03-3.50 (17) / 0.71	0.26-1.05 (6) / 0.62	Petrov & Zhuravel (1971)
Siliceous igneous rock, world wide	0.02-1.60 (8) / 0.28	0.16-0.92 (8) / 0.43	0.13-0.45 (6) / 0.28	0.06-0.16 (5) / 0.09	2.06-3.72 (10) / 3.11	0.07-0.22 (5) / 0.18	0.26	Petrov & Zhuravel (1971)
Granite pegmatite, South Dakota	0-0.54 (19) / 0.09	0.01-0.11 (19) / 0.02	0.01-0.54 (19) / 0.11		1.2-4.0 (19) / 3.3	0.01-0.50 (18) / 0.08	0.01-0.95 (16) / 0.14	Jensen (1984)
Igneous rock, world-wide	0.004-8.70 (21) / 0.17	0-1.60 (21) / 0.08						Cruft (1966)
Phosphorite, world-wide	0.01-0.09 (12) / 0.03	0.002-0.19 (15) / 0.035	0.11-1.65 (30) / 0.53	0.42-2.32 (27) / 1.06	2.90-4.93 (26) / 3.79	0-0.06 (9) / 0.03	0.32-5.91 (26) / 3.03	Cruft (1966)

*Averages of world-wide metamorphic and igneous rocks of Cruft (1966) are geometric means, other averages are arithmetic means. **Ce$_2$O$_3$ has been assumed
†) REE$_2$O$_3$ where total rare earths are given without subdivision, as with Gulbrandsen et al. (1966); Petrov and Zhuravel (1971) and Bulakh (1984).
#Radioactive skarn, Canada includes apatite from the Silver Crater (Basin) property, Bancroft, Ontario (Walker and Parsons, 1926; Gulbrandsen et al., 1966)
and the Yates mine, Sandy Creek, Quebec (Cruft et al., 1965; Trzcienski et al., 1974). ##Carbonatite, Gatineau includes 6 analyses of primary apatite
(Hogarth et al., 1985, anal. A-F), specimen AP 12A, and 2 unpublished analyses. Phosphorite world-wide includes analyses in McConnell (1973), Gulbrandsen
(1966), McClellan and Lehr (1969), Russell and Trueman (1971), de Keyser and Cook (1972), Birch (1977), Bliskovskii (1979), Jamil et al. (1979), Altschuler
(1980), Lucas and Prévôt (1981), El-Kammar et al. (1983).

sections AP 1, AP 2, AP 3, AP 6 and AP 9A is shown in Table 5. It is diopside, inclining to fassaite in specimens AP 3, AP 6, and AP 8A. Zonation is of two types: Al-enriched rims (AP 2) and Al-depleted rims (AP 3, AP 6). The major features of this zonation are illustrated in Figs. 5 and 6, and the composition of a particularly fassaitic, unzoned grain (AP 6(a)) is given in Table 5. Grey-to-brown pyroxene seems particularly aluminous. Apatite postdates pyroxene and is contemporaneous or, in some cases, later than calcite.
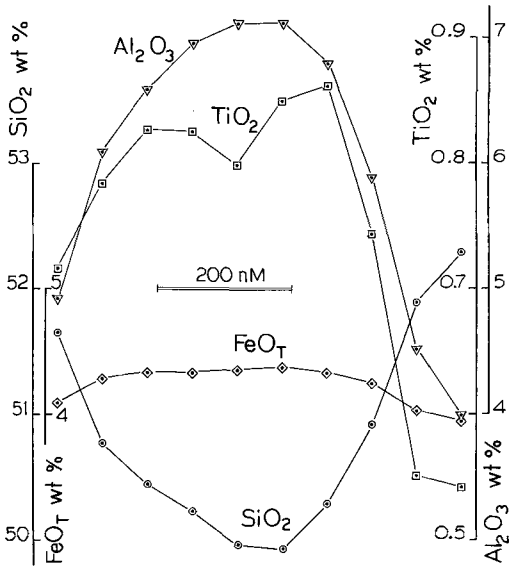
FIG. 5. Automated microprobe traverse across a zoned diopside crystal in AP 12 showing rims enriched in Fe, Al, and Ti, but depleted in Si.

Fassaite containing 7.6% Al_2O_3, 2.28% FeO and 1.35% Fe_2O_3 in skarn 30 km to the northeast was noted by V. S. Papezik (pers. comm., August 1980). Fassaites with up to 15.8% Al_2O_3 were analysed from radioactive skarns from Madagascar (Moine et al., 1985), but diopside from skarn at Sandy Creek, Quebec, was low in Al (0.4 to 1.3% Al_2O_3; Lafontaine, 1980).

Coarse-grained, bluish actinolite is abundant in AP 1 and may be secondary after diopside. Aluminous pyroxenes appear to be immune from this type of alteration. Yellowish white, sulphatic scapolite (Me 71) is a major component of AP 2. Scapolite was also present in hand specimens from other localities (Table 1) but did not appear in the polished thin sections.
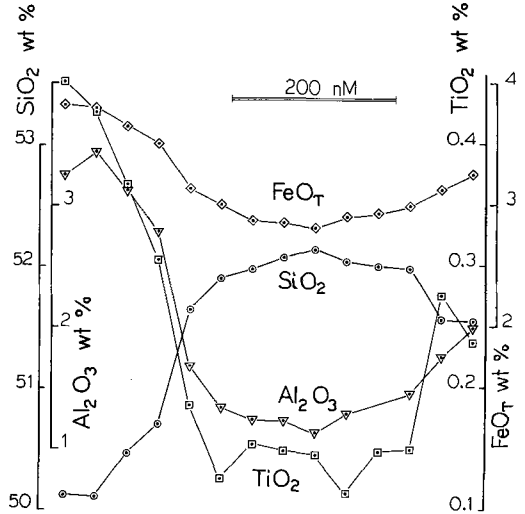
FIG. 6. Automated microprobe traverse across a zoned diopside–fassaite crystal in AP 6, showing rims enriched in Si but depleted in Fe, Al, and Ti.

Origin

The Gatineau skarns are characterized by the following paragenetic sequence:

clinopyroxene ≈ scapolite → phlogopite ≈ actinolite → calcite ≈ fluorapatite.
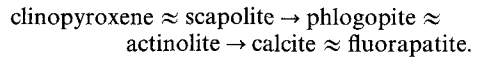
The required Si, Al, Na, and K in the silicate assemblage were probably derived from nearby siliceous, pelitic rocks during metamorphism. Mg and Ca had a ready source in the 'limestone'. The process is similar to the bimetasomatic exchange proposed by Korzhinskiy (1947) and the metamorphic remobilization of constituents proposed by Shaw et al. (1963).

The later, post-silicate stage involves the introduction of calcite and apatite to the skarns, normally near their exterior, in places at the crests of minor folds (also observed by Wynne-Edwards, 1967, in the Westport area, 125 km SW), and commonly filling open spaces. No single, obvious source has been identified for P, REE, Sr, F, Cl and S. Gabbro is absent, syenites, carbonatites and fenites are later, granite pegmatites are impoverished in apatite (without a single crystal of apatite identified to date). Likewise, marine sedimentary apatite is ruled out, its REE distribution as well as Sr and CO_2 content would necessitate complete reconstruction of the mineral. The suggestion is here made that the necessary ingredients were derived from rocks nearby under conditions of granulite-facies metamorphism. Under these conditions, F, REE and Sr are known to be depleted in

Table 5. Chemical composition of pyroxene, amphibole and scapolite from skarns.

| | pyroxene | | | | | | | | amphibole | scapolite |
	AP 1	AP 2(r)	AP 2(c)	AP 3	AP 6(r)	AP 6(c)	AP 6(a)	AP 9A	AP 1	AP 9A
n	2	2	4	5	3	2	2	2	2	3
SiO_2	54.49	52.52	52.05	49.66	51.94	49.94	48.98	50.63	56.29	46.27
TiO_2	0.06	0.34	0.14	0.75	0.60	0.92	0.60	0.78	0.10	0.01
Al_2O_3	0.86	3.11	1.22	7.21	4.48	7.11	8.15	5.28	0.55	27.40
Fe_2O_3	1.37	2.47	3.18	2.25	2.25	2.71	1.75	1.72	4.79	0.00
FeO	3.63	1.42	0.00	1.58	1.99	1.92	1.92	3.51	2.82	0.00
MnO	0.14	0.11	0.14	0.08	0.09	0.09	0.06	0.18	0.22	0.00
MgO	15.57	15.67	16.61	13.72	15.01	13.77	14.04	14.13	19.55	0.02
CaO	24.49	25.28	24.91	25.57	24.57	24.30	23.66	23.06	9.23	16.02
SrO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.34
BaO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.03
Na_2O	0.50	0.27	0.26	0.25	0.50	0.60	0.41	0.61	2.09	3.71
K_2O	0.01	0.00	0.01	0.01	0.00	0.00	0.00	0.00	0.94	0.26
Cl	---	---	---	---	---	---	---	---	0.01	0.48
F	---	---	---	---	---	---	---	---	0.75	0.00
SO_3	---	---	---	---	---	---	---	---	0.00	1.32
H_2O	---	---	---	---	---	---	---	---	1.77	---
Total	101.12	101.19	98.52	101.08	101.43	101.26	99.56	99.90	98.80	95.75

Ions per formula

Pyroxene:

	AP 1	AP 2(r)	AP 2(c)	AP 3	AP 6(r)	AP 6(c)	AP 6(a)	AP 9A
Si	1.979	1.900	1.927	1.804	1.875	1.810	1.796	1.862
Al	0.021	0.100	0.053	0.196	0.125	0.190	0.204	0.138
Σ	2.00	2.00	1.98	2.00	2.00	2.00	2.00	2.00
Al	0.016	0.033	0.000	0.112	0.066	0.114	0.148	0.091
Ti	0.002	0.009	0.004	0.020	0.016	0.22	0.017	0.022
Fe^{3+}	0.037	0.067	0.089	0.062	0.061	0.074	0.048	0.048
Fe^{2+}	0.110	0.042	0.000	0.048	0.060	0.058	0.059	0.108
Mn	0.004	0.003	0.004	0.003	0.003	0.003	0.002	0.005
Mg	0.843	0.845	0.917	0.742	0.808	0.744	0.767	0.775
Σ	1.01	1.00	1.01	0.99	1.01	1.01	1.04	1.05
Ca	0.953	0.980	0.988	0.995	0.951	0.944	0.930	0.909
Na	0.035	0.019	0.018	0.018	0.035	0.042	0.028	0.043
K	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000
Σ	0.99	1.00	1.01	1.01	0.99	0.99	0.96	0.95

Amphibole AP 1:

Si	7.926
Al	0.074
Σ	8.00
Al	0.018
Ti	0.010
Fe^{3+}	0.508
Fe^{2+}	0.333
Mn	0.027
Mg	4.104
Σ	5.00
Ca	1.393
Na	0.571
Σ	1.96
Na	0.000
K	0.169
Σ	0.17
F	0.334
Cl	0.003
OH	1.663
Σ	2.00

Scapolite AP 9A:

Si	7.064
Al	4.936
Σ	12.00
Ti	0.001
Fe^{3+}	0.000
Fe^{2+}	0.000
Mn	0.000
Mg	0.004
Ca	2.622
Sr	0.031
Ba	0.002
Na	1.100
Σ	3.76
S	0.152
Cl	0.125
F	0.000
OH,Cl	0.723
Σ	1.00

Microprobe analyses. Oxygen equivalents of F, Cl deducted in totals. Fe_2O_3, FeO, H_2O calculated from formula content. Formulae calculated as pyroxene with 4 cations and 12 charges, amphibole with 13(Z + C) cations and 46 charges, scapolite with 12 Z cations. n = number of analyses averaged.

pelitic and calcareous rocks (Rösler *et al.*, 1977; Beuge *et al.*, 1978; Muecke *et al.*, 1979; Levitskii and Petrova, 1982).

Appreciable Cl is present in scapolite and some apatite. Scapolite, particularly in the marialite–mizzonite range, is an effective trap for Cl (Deer *et al.*, 1963) and it is notable that, in occurrences where the mineral was observed (sites of AP 6, AP 7, AP 8A, AP 9, AP 9A), Cl was effectively scavenged, leaving little for apatite. Like F, Cl may have been derived from the wall rock.

Mineral compositions suggest appreciable H_2O, CO_2 and minor F, S and Cl. Assuming granulite metamorphic conditions (670 °C, 7 kbar P_T), fugacities calculated by the method of Yardley (1985) and Korzhinskiy (1981) and using f_{H_2O} of Burnham *et al.* (1969) are as follows: $f_{H_2O} = 5800$ bars, $f_{HCl} = 0.14$ bars, $f_{HF} = 0.37$ bars for AP 4; $f_{H_2O} = 5800$ bars, $f_{HCl} = 9.0$ bars, $f_{HF} = 0.42$ bars for AP 11(p). These

fugacities must be considered qualitative in view of uncertainties introduced by analytical method and positioning of F and OH in the apatite structure.

Regardless of the agents of solution and transportation, Eu^{2+} would be expected to adhere tenaciously to feldspar in the source rocks during metamorphism or be removed en route to the skarns, thus accounting for the negative Eu anomalies. This mechanism has been used to explain Eu anomalies in metamorphic rocks of Scotland (Muecke *et al.*, 1979).

Apatite from calc–silicate-free marble (AP 15) closely resembles skarn apatite. Both have similar Ce, S, Si, F and Cl contents. Sr in AP 15, while higher than that of skarn apatite, is close to AP 9 and certainly well below that of AP 12A and other carbonatitic apatites. Lack of associated diopside and amphibole suggests that silicate chemistry is not an essential agent for precipitation. On the

basis of chemical composition alone, there seems to be no reason to construct a separate genetic model for this type.

Acknowledgements

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