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COMPOSITIONAL VARIATIONS OF PUMPELLYITE ALONG THE WESTERN MARGIN OF THE QUEBEC APPALACHIANS*

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ABSTRACT

Pumpellyite is a widely occurring mineral along the western margin of the Quebec Appalachians. It is found in mafic rocks of both the external and internal tectonic domains of St-Julien & Hubert (1975). Compositions vary from iron-rich to the more normal aluminous variety. In some occurrences pumpellyite is vanadium-bearing. The progression from iron- to aluminum-rich composition can be correlated with increasing grade of metamorphism, in particular with increasing pressure, going east and south from the external domain into the internal domain. This correlation agrees well with a tectonic model that proposes increasingly deeper exposures toward the east and south.

Keywords: Quebec Appalachians, metamorphism, pumpellyite.

SOMMAIRE

On trouve la pumpellyite dans les métabasites des domaines externe et interne (St-Julien & Hubert 1975) le long de la bordure occidentale des Appalaches québécoises. Sa composition varie de ferrifère à alumineuse (normale); une variété vanadifère a été identifiée en quelques endroits. En allant du domaine externe, à l'ouest, au domaine interne à l'est et au sud, la progression en caractère alumineux montre une corrélation avec un métamorphisme prograde qui reflète surtout l'accroissement de pression. Cette corrélation s'accorde avec un modèle tectonique qui préconise, vers l'est et le sud, l'affleurement de parties de plus en plus profondes de la croûte terrestre.

(Traduit par la Rédaction)

Mots-clés: Appalaches québécoises, métamorphisme, pumpellyite.

INTRODUCTION

Recognized as a low-grade metamorphic index mineral and found around the world in a number of orogenic belts (Turner 1981), pumpellyite has only recently been noted in the Appalachians (Coombs *et al.* 1970, Zen 1974, Franks 1974, Papezik 1974, Richter & Roy 1976, Trzcienski *et al.* 1979). The occurrence of pumpellyite in various tectonic settings and its highly variable composition complicate an evaluation of its petrological significance (Zen 1974, Williams & Einarson 1976, Mével 1981). As a contribution to the understanding of pumpellyite paragenesis, a number of new pumpellyite localities (Fig. 1) have been documented and studied along the western margin of the Quebec Appalachians, within both the external and internal tectonic domains of St-Julien & Hubert (1975). Pumpellyite is found in a number of different mineral assemblages; its variable composition is related not only to the associated minerals, but also to the conditions of metamorphism.

PETROGRAPHY

Within the external domain, pumpellyite occurs in allochthonous, pre-Silurian metabasic extrusive and intrusive rocks; in the internal domain, it occurs in metavolcanic rocks mapped as part of the Caldwell Group and in metagabbros of the Asbestos Ophiolite (Fig. 1). The extrusive rocks containing pumpellyite are vesicular, brecciated, hyaloclastic, hyalo-ophitic to subophitic basalts. In most occurrences pumpellyite is found as replacements of plagioclase laths or as rosettes in glassy patches of the rock matrix (Fig. 2). Less commonly, pumpellyite is found as pseudomorphs or partial replacements of primary ferromagnesian minerals. The common habit of pumpellyite is that of rod-like or needle-like crystals; less commonly, it takes the form of a fibrous mass with rotary extinction between crossed nicols. Pleochroism varies from pea green to pistachio green, blue green or brownish green. Anomalous birefringence is also a diagnostic property. Once recognized, pumpellyite is easily distinguished

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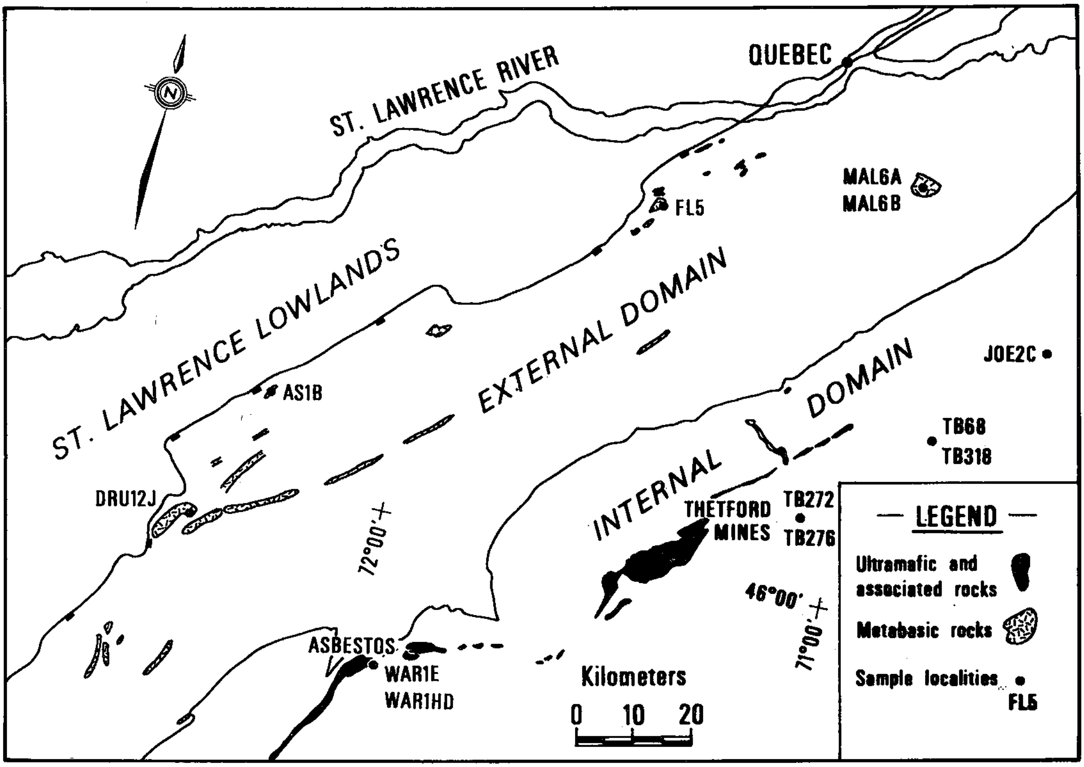

FIG. 1. Tectonic subdivision (after St-Julien & Hubert 1975) and sample localities of analyzed pumpellyite specimens along the western margin of the Quebec Appalachians.

TABLE 1. METAMORPHIC MINERALS IN PUMPELLYITE-BEARING ASSEMBLAGES

Sample	Albite	Quartz	Sphene	Epidote	Calcite	Amphibole	Chlorite	Prehnite	White mica	Opaque
TB272	X	X	X	X	X	X	X		X	X
TB276	X		X	X		X	X		X	
TB68	X	X	X	X			X			
TB318	X	X	X	X			X		X	X
MAL6A	X	X		X			X		X	X
MAL6B	X	X	X	X	X		X		?	X
FL5	X	X	X				X			X
AS1B	X	X			X		X			
JOE2C	X	X		X		?	X			
PD11	X	X	X				X			X
DRU12J	X	X			X		X			X
WAR1HD	X	X	X		X	X		X		
WAR1E	X	X	X			X	X		X	

TABLE 2. REPRESENTATIVE MICROPROBE ANALYSES OF PUMPELLYITES FROM THE WESTERN MARGIN OF THE QUEBEC APPALACHIANS SHOWING THE INTERSAMPLE AND INTRASAMPLE VARIATIONS FOUND

	FL5-4	TB272-26	TB276-10	TB68-3S	TB318-9	PD11-3	PD11-4	MAL6B-3	MAL6B-9	MAL6A-5	MAL6A-6	DRU12J-4	WAR1E-1	WAR1HD-15	AS1B-2	AS1B-4	JOE2C-10
SiO_2	37.91	36.85	38.18	37.56	37.26	35.95	37.68	38.63	37.05	37.79	38.69	36.41	37.90	37.74	36.76	34.77	36.39
V_2O_3	0.0	0.0	0.0	0.0	0.0	1.79	1.91	0.57	0.43	0.23	0.34	0.0	0.0	0.0	0.0	0.0	0.25
Al_2O_3	22.01	20.97	25.36	23.85	24.31	13.56	21.29	19.35	18.96	20.83	18.89	21.20	22.43	25.71	18.96	11.94	20.71
FeO^*	8.53	8.95	3.65	6.84	4.79	19.12	10.23	10.32	12.37	9.51	11.52	8.28	5.88	3.06	10.87	20.66	10.22
MnO	0.0	0.0	0.20	0.21	0.66	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.05	0.09	0.0	0.0	0.0
MgO	2.53	1.77	1.97	0.87	2.69	0.73	0.88	2.11	2.02	2.61	2.27	2.25	3.35	2.85	3.29	1.36	2.49
CaO	20.84	22.40	22.53	23.56	22.39	21.98	21.98	22.31	22.44	22.60	21.56	24.89	22.43	23.07	20.70	20.48	21.97
Total	91.82	90.94	91.89	92.86	92.10	93.13	93.97	93.29	93.20	93.57	93.27	93.03	92.08	92.53	90.58	89.21	92.03
	Atomic proportions based upon cations = 16.00																
Si	6.25	6.16	6.20	6.12	6.05	6.14	6.19	6.34	6.11	6.14	6.36	5.92	6.16	6.06	6.17	6.18	6.02
V	0.0	0.0	0.0	0.0	0.0	0.20	0.21	0.06	0.05	0.03	0.04	0.0	0.0	0.0	0.0	0.0	0.03
Al	4.28	4.13	4.85	4.59	4.66	2.73	4.12	3.74	3.69	3.99	3.66	4.07	4.30	4.89	3.75	2.50	4.04
Fe^*	1.18	1.25	0.50	0.93	0.65	2.73	1.41	1.42	1.70	1.29	1.58	1.13	0.80	0.41	1.53	3.07	1.41
Mn	0.0	0.0	0.03	0.03	0.09	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.01	0.01	0.0	0.0	0.0
Mg	0.62	0.44	0.48	0.21	0.65	0.19	0.22	0.52	0.50	0.63	0.56	0.55	0.81	0.68	0.82	0.36	0.61
Ca	3.68	4.01	3.92	4.12	3.90	4.02	3.89	3.92	3.97	3.93	3.80	4.34	3.91	3.97	3.72	3.90	3.89

FeO^* and Fe^* represent total iron in the microprobe pumpellyite analyses.

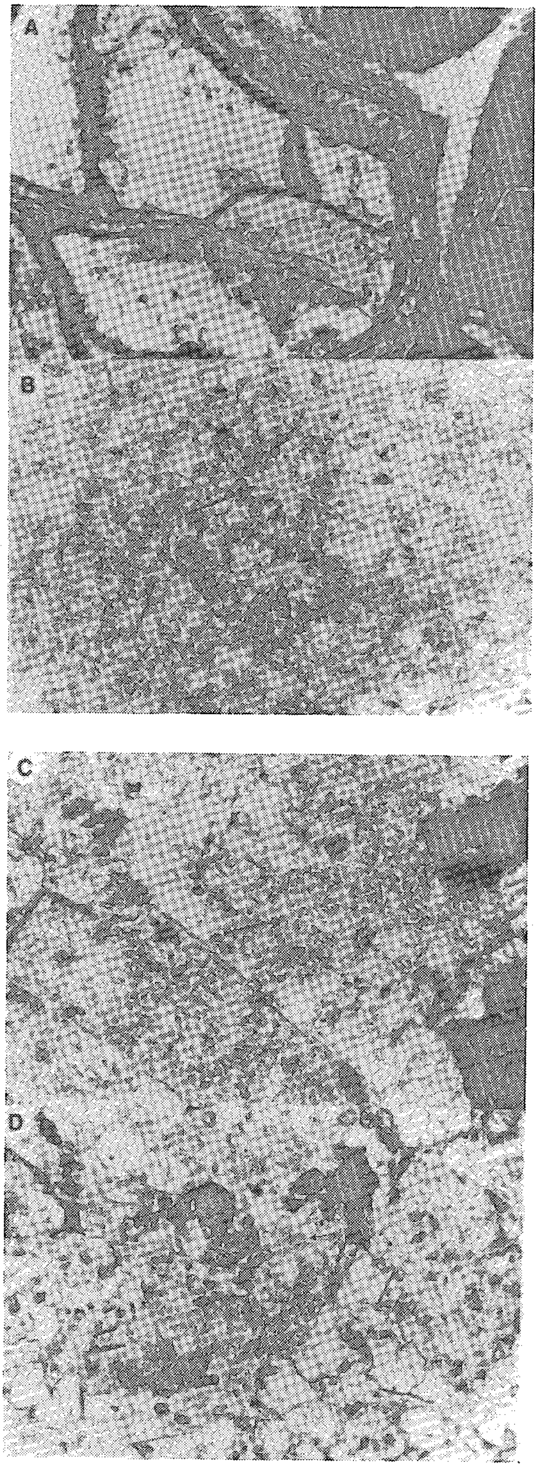

FIG. 2. Photomicrographs of representative habits of pumpellyite from the western margin of the Quebec Appalachians. Typical habit within each sample is pointed to by an arrow. A) Sample JOE2C containing highly pleochroic pumpellyite in a hyaloclastic rock. Dark regions are Fe-rich glass. Field of view is 2 mm. B) Sample DRU12J containing pumpellyite rosettes in a quartz–calcite matrix. Field of view is 1 mm wide. C) Fine grained vanadium-bearing pumpellyite from the Port Daniel Complex (Ayrton 1967) (sample PD11) with albite, quartz and opaque mineral. Field of view is 0.5 mm wide. D) Fine, tabular pumpellyite from sample FL5 associated with pyroxene, biotite, albite, chlorite, quartz and opaque mineral. Field of view is 1.0 mm wide. E) Nonpleochroic tabular pumpellyite from the Asbestos Ophiolite (sample WAR1HD) in association with quartz, albite, sphene and prehnite. Field of view is 1.0 mm wide. F) Rod-like pleochroic pumpellyite (sample MAL6B) in association with quartz, albite, sphene, epidote and opaque mineral. Field of view is 1.0 mm wide.

from epidote and actinolite. The most common original igneous minerals, plagioclase and clinopyroxene, remain as phenocrysts in the rock, although the plagioclase has been albitized. The matrix of many of the metabasites is commonly partially devitrified glass with or without calcite and microlites. Most samples investigated exhibit no signs of penetrative deformation. Septechlorite and iddingsite are present as pseudomorphs after olivine in several samples.

The intrusive rocks of the external domain are alkalic diabase sills and dykes retaining original kaersutite and biotite. The replacement habit and pleochroism of pumpellyite from these rocks are similar to pumpellyite from the extrusive rocks of both domains. In contrast, pumpellyite in metagabbros of the internal domain has different characteristics. These metagabbros are coarse grained equigranular rocks, with up to 50% of their original igneous minerals preserved as clinopyroxene, brown hornblende and albitized plagioclase. In these rocks, pumpellyite in the form of tabular, nonpleochroic crystals is commonly found within a prehnite or quartz matrix. The birefringence of this pumpellyite is moderate to pale mottled orange or pinkish yellow. It is more difficult to identify than the highly pleochroic variety. Honey yellow epidote with a habit similar to that of pumpellyite is also present. In general, all pumpellyite-bearing rocks retain much of their igneous character and show little or no signs of penetrative deformation. The metamorphic minerals found in these rocks (in addition to pumpellyite), which are present in highly variable proportions, are: quartz, epidote, sphene, calcite, amphibole, prehnite, albite, chlorite, white mica and opaque minerals.

PUMPELLYITE CHEMISTRY

Microprobe analyses were performed on a number of pumpellyite grains from selected rock samples representing different assemblages of metamorphic minerals (Table 1) found along the western margin of the Quebec Appalachians. All analyses were carried out on a MAC 400 microprobe at 15 kV and a sample current of 20 nanoampere as measured on benitoite using well-analyzed mineral standards. Wavelength-dispersion analyses were corrected for matrix effects by the Bence–Albee method (Bence & Albee 1968); energy-dispersion analyses were done by Quantex-Ray (KEVEX Corporation).

Representative analytical results (Table 2) indicate that the crystals of pumpellyite vary greatly in their chemical composition not only from rock to rock but within a single sample (see data for PD11–3,4, MAL6A–5, 6 and MAL6B–3,9 Table 2). Microprobe analyses present two inherent problems: 1) H_2O is not analyzed, such that all totals will be less than 100%; 2) ferrous and ferric iron are indistinguishable one from the other, thus necessitating treatment of Fe as total iron.

The chemical formula for pumpellyite proposed by Passaglia & Gottardi (1973) and slightly modified by Coombs *et al* (1976) is: $W_4X_2Y_4Z_6O_{20+x}(OH)_{8-x}$, where W = Ca,Mn, X = (Mg, Fe^{2+},Mn)$_{2+x}$(Fe^{3+},Al)$_x$, Y = Fe^{3+}, Al, Z = Si. Since there are 16 cations in the ideal formula, all analytical results have been formulated on this basis (Table 2). Calcium and silicon vary slightly from their ideal values of four and six, respectively, whereas Mg, Fe and Al vary considerably, as clearly portrayed on a triangular plot (Fig. 3) based upon atomic proportions of Mg, Al and Fe* ($Fe^{2+} + Fe^{3+}$). The pure Fe^{2+}, Fe^{3+} species is julgoldite (Moore 1971).

Not shown in Figure 3 is the variable vanadium content of pumpellyite. Four different rocks contain pumpellyite with detectable vanadium (Table 2). The most vanadium-rich pumpellyite comes from a basalt associated with the Port Daniel Complex in the Gaspé area (see Ayrton 1967 for the geological location). The substitution of V^{3+} for Al^{3+} or Fe^{3+} can be expected for crystal-chemistry reasons. This substitution has also been observed in ardennite (Allman & Donnay 1971), which is crystallographically similar to pumpellyite. It appears that low-grade metamorphism leads to a preferential concentration of the available vanadium of the host rock into pumpellyite. To our knowledge, these are the first reported occurrences of V-bearing pumpellyite.

Examination of Figure 3 indicates that Mg consistently falls between 5 and 15% of the cations in the X and Y crystallographic positions, and that the major variation involves Fe* and Al. Also, the majority of compositions lie (Fig. 3) on the Mg-poor side of the julgoldite – Mg–Al-pumpellyite line. The linear antipathetic relationship between Fe* and Al (Fig. 4) indicates that these two cations consistently occupy from 5 to 5.5 of 6 cation sites available to them. The fact that most compositions fall below or near 5.5 (Fig. 4) implies that the remaining site-occupancy is taken up by Mg. The results of our analyses, together with those of Coombs *et al.* (1976) and Liou (1979), show that extensive solid solution exists in the pumpellyite–julgoldite series, and that there may be a miscibility gap between julgoldite and pumpellyite at Al $<$ 2.0 (Fig. 4). The slight deficiency of Ca in the W site is probably filled by Fe^{2+}, as suggested by Allman & Donnay (1971). Liou (1979) suggested that because of the Fe*–Al antipathetic relationship, most of the iron is probably Fe^{3+}. From Figure 3, however, we see that only below the line Fe-pumpellyite – Mg-pumpellyite need iron be Fe^{3+} in order to fill the deficient Al^{3+} site. Above this line there is sufficient Al, *i.e.*, four cations, to retain charge balance. This argument is not totally resolvable at this point, however: H_2O

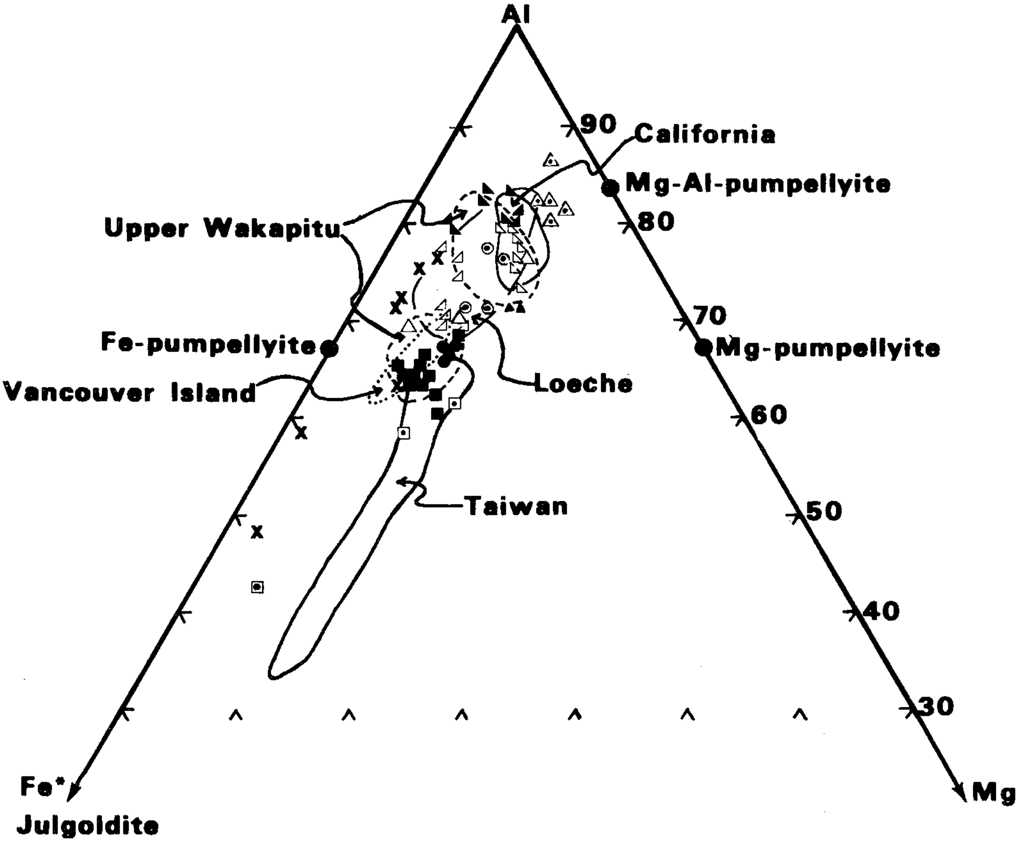

FIG. 3. Compositional variations of pumpellyites from the western margin of the Quebec Appalachians in terms of Mg–Al–Fe*, where Fe* is total iron. Pumpellyite compositional fields are from Vancouver Island (Surdam 1969), California (Ernst *et al.* 1970), Upper Wakatipu (Kawachi 1975), Loeche, Switzerland (Coombs *et al.* 1976) and Taiwan (Liou 1979). Symbols: ◢ sample TB68, ◿ sample TB272, ◣ sample TB276, ◺ sample TB318, △ sample FL5, × sample PD11, ■ sample MAL6, ⊙ DRU12J, ▲ sample WAR1E, ◬ sample WAR1HD, ⊡ sample AS1B. • sample JOE2C.

has not been determined, thus leaving the OH:O ratio unknown. The high iron contents ($Fe^{2+} + Fe^{3+}$) correlate directly with the highly pleochroic nature of the pumpellyite.

DISCUSSION

Pumpellyite compositions from the western margin of the Quebec Appalachians resemble those (Fig. 3) from a variety of metamorphic terrains (Surdam 1969, Ernst *et al.* 1970, Glassley 1975, Kawachi 1975, Coombs *et al.* 1976, Liou 1979) and presumably reflect variations of metamorphic conditions. An attempted correlation of bulk-rock chemistry and pumpellyite composition has met with little success (Schiffman & Liou 1980) and receives no support from our rocks; examples of pumpellyite of similar composition occur in basaltic rocks that vary from alkalic to tholeiitic in character.

In an experimental study of Mg–Al-pumpellyite, Schiffman & Liou (1980) showed that pumpellyite stability is a function of pressure and temperature. Their experimental data impose some constraints on the P–T conditions of formation in the Quebec pumpellyite-bearing rocks. At pressures less than 10 kbar, Mg–Al-pumpellyite is not stable above 400°C. Since the Quebec examples are not as magnesian nor as aluminous, and since the Mg–Al-pumpellyite breakdown-assemblage clinozoisite + grossular + chlorite + quartz is not observed, 400°C is a maximum possible temperature for any of the Quebec localities.

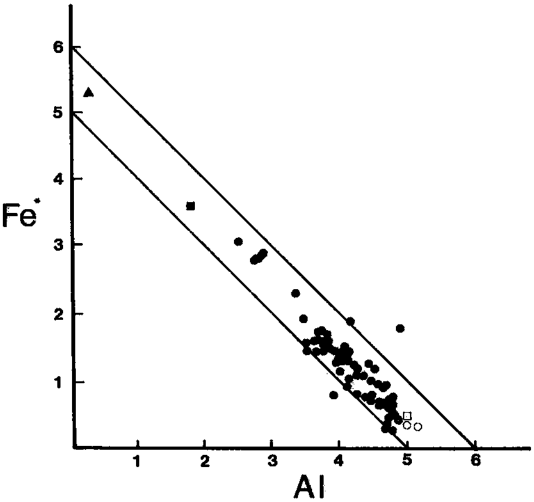

FIG. 4. Inverse relationship between Al and Fe*, where Fe* is total iron, for pumpellyites (●) from the western margin of the Quebec Appalachians; symbols: ▲ natural julgoldite (Moore 1971), ■ most Fe-rich pumpellyite of Liou (1979), □ and ○ most Al-rich pumpellyites of Kawachi (1975) and Coombs *et al.* (1976), respectively.

A compilation of extant pumpellyite compositions (Fig. 3) from several different pressure-regimes from around the world indicates that there is a correlation between estimated pressure of formation and pumpellyite composition. The highest pressure-regime from which pumpellyite has been analyzed is that associated with the glaucophane schists of California (Ernst *et al.* 1970). These examples of pumpellyite, amongst the most aluminous reported in the literature, come from an area where pressure is estimated to be about 7 kbar. Coombs *et al.* (1976) reported similar compositions, which were estimated to have formed at about 370°C and a pressure of "several kilobars". Additional detailed studies of pumpellyite (Surdam 1969, Kawachi 1975, Liou 1979) also show (Fig. 3) that, for inferred lower-pressure environments, the total Fe-content of pumpellyite increases. Passaglia & Gottardi (1973) noted that with increasing Fe-content in pumpellyite, the unit-cell volume also increased. This observation also suggests that Fe-content of pumpellyite and pressure may be inversely related.

In Quebec, the most iron-rich pumpellyite crystals generally occur in the western part of the external domain (St-Julien & Hubert 1975), where metamorphic conditions are estimated to be <200°C and <2 kbar, based on the fact that natural gas is found at depth in the area (C. Hubert, pers. comm., 1981). East and south of this area, the coexistence of pumpellyite, chlorite, epidote, actinolite and quartz indicates metamorphic conditions near those for the reaction (Nitsch 1971): pumpellyite + chlorite + quartz → epidote + actinolite + H_2O, which, at moderate pressures (2 to 4 kbar), would indicate a temperature of about 325°C (Schiffman & Liou 1980).

The most magnesian and aluminous pumpellyite compositions come from the Asbestos Ophiolite; they suggest pressures of about 7 kbar, based upon similar pumpellyite compositions described from California (Ernst *et al.* 1970). This pressure agrees with an estimated pressure of about 6 kbar suggested for a Na–Ca-amphibole-bearing metavolcanic suite (Trzcienski 1976) not far to the west of Asbestos, at Richmond, Quebec. Observed assemblages of pumpellyite + prehnite and prehnite + tremolite + clinozoisite + chlorite at Asbestos also limit the temperature to a maximum between 350°C and 375°C (see Fig. 10, Schiffman & Liou 1980).

The pumpellyite compositions across the western margin of the Quebec Appalachians suggest increasing P–T conditions going from northwest to southeast, which is in agreement with the tectonic model of an ever-deepening tectonic section exposed in this direction (St-Julien & Hubert 1975).

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REFERENCES

ALLMAN, R. & DONNAY, G. (1971): Structural relations between pumpellyite and ardennite. *Acta Cryst.* B27, 1871-1875.

AYRTON, W.G. (1967): Chandler–Port Daniel Area. *Quebec Dep. Nat. Res. G.R.* 120.

BENCE, A.E. & ALBEE, A.L. (1968): Empirical correction factors for the electron microanalysis of silicates and oxides. *J. Geol.* 76, 382-403.

COOMBS, D.S., HORODYSKI, R.J. & NAYLOR, R.S. (1970): Occurrence of prehnite–pumpellyite facies metamorphism in northern Maine. *Amer. J. Sci.* 268, 142-156.

———, NAKAMURA, Y. & VUAGNAT, M. (1976): Pumpellyite–actinolite facies schists of the Taveyanne Formation near Loèche, Valais, Switzerland. *J. Petrology* 17, 440-471.

ERNST, W.G., SEKI, Y., ONUKI, H. & GILBERT, M.C. (1970): Comparative study of low-grade metamorphism in the California Coast Ranges and the outer metamorphic belt of Japan. *Geol. Soc. Amer. Mem.* 124.

FRANKS, S.G. (1974): Prehnite–pumpellyite facies metamorphism of the New Bay Formation, Exploits Zone, Newfoundland. *Can. Mineral.* 12, 456-462.

GLASSLEY, W.E. (1975): Low variance phase relationships in a prehnite–pumpellyite facies terrain. *Lithos* 8, 69-76.

KAWACHI, Y. (1975): Pumpellyite–actinolite and contiguous facies metamorphism in part of Upper Wakatipu district, South Island, New Zealand. *N.Z. J. Geol. Geophys.* 18, 401-441.

LIOU, J.G. (1979): Zeolite facies metamorphism of basaltic rocks from the East Taiwan ophiolite. *Amer. Mineral.* 64, 1-14.

MÉVEL, C. (1981): Occurrence of pumpellyite in hydrothermally altered basalts from the Vema fracture zone (Mid-Atlantic Ridge). *Contr. Mineral. Petrology* 76, 386-393.

MOORE, P.B. (1971): Julgoldite, the Fe^{+2}–Fe^{+3} dominant pumpellyite. A new mineral from Långban, Sweden. *Lithos* 4, 93-99.

NITSCH, K.-H. (1971): Stabilitätsbeziehungen von Prehnit- und Pumpellyit-haltigen Paragenesen. *Contr. Mineral. Petrology* 30, 240-260.

PAPEZIK, V.S. (1974): Prehnite–pumpellyite facies metamorphism of Late Precambrian rocks of the Avalon Peninsula, Newfoundland. *Can. Mineral.* 12, 463-468.

PASSAGLIA, E. & GOTTARDI, G. (1973): Crystal chemistry and nomenclature of pumpellyites and julgoldites. *Can. Mineral.* 12, 219-223.

RICHTER, D.A. & ROY, D.C. (1976): Prehnite–pumpellyite facies metamorphism in central Aroostook County, Maine. *Geol. Soc. Amer. Mem.* 146, 239-261.

SCHIFFMAN, P. & LIOU, JUHN G. (1980): Synthesis and stability relations of Mg–Al pumpellyite, $Ca_4Al_5MgSi_6O_{21}(OH)_7$. *J. Petrology* 21, 441-474.

ST-JULIEN, P. & HUBERT, C. (1975): Evolution of the Taconian Orogen in the Quebec Appalachians. *Amer. J. Sci.* 275-A, 337-362.

SURDAM, R.C. (1969): Electron microprobe study of prehnite and pumpellyite from the Karmutsen Group, Vancouver Island, British Columbia. *Amer. Mineral.* 54, 256-266.

TRZCIENSKI, W.E., JR. (1976): Crossitic amphibole and its possible tectonic significance in the Richmond area, southeastern Quebec. *Can. J. Earth Sci.* 13, 711-714.

———, CHAMPIGNY, N. & BIRKETT, T.C. (1979): Pumpellyite paragenesis: Asbestos to the Gaspé, Quebec. *Geol. Assoc. Can./Mineral. Assoc. Can. Program Abstr.* 4, 83.

TURNER, F.J. (1981): *Metamorphic Petrology – Mineralogical, Field, and Tectonic Aspects* (2nd ed.). McGraw-Hill, New York.

WILLIAMS, H. & EINARSON, G.W. (1976): Discussion of "Prehnite- and pumpellyite-bearing mineral assemblages, west side of the Appalachian metamorphic belt, Pennsylvania to Newfoundland" by E-an Zen. *J. Petrology* 17, 135-136.

ZEN, E-AN (1974): Prehnite- and pumpellyite-bearing mineral assemblages, west side of the Appalachian metamorphic belt, Pennsylvania to Newfoundland. *J. Petrology* 15, 197-242.

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