The Canadian Mineralogist
Vol. 40, pp. 671-678 (2002)

LAFLAMMEITE, $Pd_3Pb_2S_2$, A NEW PLATINUM-GROUP MINERAL SPECIES FROM THE PENIKAT LAYERED COMPLEX, FINLAND

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ABSTRACT

Laflammeite, $Pd_3Pb_2S_2$, is a new parkerite-type sulfide discovered in the Penikat layered complex, Finland. It occurs in altered pyroxenite in the Kirakkajuppura deposit in association with actinolite and clinochlore, accessory chromite, various platinum-group minerals [vysotskite, zvyagintsevite, cuprorhodsite – (ferrorhodsite) – malanite, laurite – erlichmanite, irarsite, keithconnite, unusual Pb–(Pd)-rich thiospinels of Cu, Fe, Rh and Ir, unnamed Pd_9PbO_{10} and $Rh(Ni,Fe,Cu)_2S_3$], Pd-rich gold, and minor base-metal sulfides (chalcopyrite, bornite and millerite). Laflammeite occurs as individual subhedral to euhedral crystals up to 0.3 mm in size, and in intergrowths with Pt–Ni-poor vysotskite. Laflammeite is opaque and cream-colored with a brownish tint in reflected light in air. The anisotropy is weak, from brownish gray to grayish brown. The bireflectance is weak; pleochroism was not observed. Reflectance percentages in air (and in oil) are, for R_1 and R_2, 45.3, 46.2 (31.55, 32.45) at 470 nm, 46.5, 47.55 (32.6, 33.75) at 546 nm, 47.7, 48.8 (33.9, 35.05) at 589 nm, and 49.6, 50.0 (36.0, 36.3) at 650 nm. Fine twins are present. Microhardness values VHN_{50} are 156 and 185; the average is 171 kg/mm^2. Cleavage is {010}, perfect. The average results of 26 wavelength-dispersion electron-microprobe analyses are Pd 39.16, Ir 1.08, Pb 52.01, S 7.90, total 100.15 wt.%, corresponding to $(Pd_{2.96}Ir_{0.05})_{\Sigma 3.01}Pb_{2.02}S_{1.98}$ (basis: 7 atoms per formula unit). The powder pattern was indexed on a monoclinic cell with *a* 11.521(11), *b* 8.294(10), *c* 8.321(6) Å, β 134.38(5)°, *V* 568.3(6) Å^3. The space group is probably *C*2/*m*, with *Z* = 4. D_{calc} is 9.41 g/cm^3. The strongest lines in the pattern [*d* in Å(I)(*hkl*)] are: 5.953(6)(001), 4.144(10)(020), 2.917(9)($\bar{2}$20), 2.413(8)(022), 2.365(7)(422) and 2.082(5)(040). Laflammeite likely crystallized from a hydrothermal fluid or a late-stage liquid in an environment poor in overall sulfur and rich in the *PGE*, Pb and volatiles.

Keywords: laflammeite, Pd–Pb sulfide, new mineral species, platinum-group mineral, platinum-group elements, lead, layered intrusion, mafic-ultramafic rocks, Kirakkajuppura, Penikat, Finland.

SOMMAIRE

La laflammeïte, $Pd_3Pb_2S_2$, nouvelle espèce minérale sulfurée de type parkerite, provient du complexe stratiforme de Penikat, en Finlande. On la trouve dans des pyroxénites altérées du gisement de Kirakkajuppura en association avec actinolite et clinochlore, chromite accessoire, une variété de minéraux du groupe du platine [vysotskite, zvyagintsévite, cuprorhodsite – (ferrorhodsite) – malanite, laurite – erlichmanite, irarsite, keithconnite, des thiospinelles de Cu, Fe, Rh et Ir enrichis en Pb–(Pd), Pd_9PbO_{10} et $Rh(Ni,Fe,Cu)_2S_3$ sans nom], or palladifère, et des quantités mineures de sulfures de métaux de base (chalcopyrite, bornite et millerite). La laflammeïte se présente en cristaux idiomorphes à sub-idiomorphes atteignant 0.3 mm de diamètre, et en intercroissances avec la vysotskite à faible teneur en Pt et Ni. C'est un minéral opaque de couleur crème avec une teinte brunâtre en lumière réfléchie dans l'air. L'anisotropie est faible, de gris brunâtre à brun grisâtre. La biréflectance est faible, et aucun pléochroïsme a été observé. La réflectance R_1 et R_2, dans l'air (et dans l'huile), exprimée en pourcentages, est 45.3, 46.2 (31.55, 32.45) à 470 nm, 46.5, 47.55 (32.6, 33.75) à 546 nm, 47.7, 48.8 (33.9, 35.05) à 589 nm, et 49.6, 50.0 (36.0, 36.3) à 650 nm. De fines macles sont présentes. Les valeurs de microdureté, VHN_{50}, sont 156 et 185, pour une moyenne de 171 kg/mm^2. Le clivage est {010}, parfait. La moyenne des résultats de 26 analyses obtenues avec une microsonde électronique en dispersion de longueurs d'ondes est: Pd 39.16, Ir 1.08, Pb 52.01, S 7.90, total 100.15% (poids), ce qui correspond à $(Pd_{2.96}Ir_{0.05})_{\Sigma 3.01}Pb_{2.02}S_{1.98}$ sur une base de sept atomes par unité formulaire. Le spectre de diffraction (méthode des poudres) est indexé sur une maille monoclinique,

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a 11.521(11), *b* 8.294(10), *c* 8.321(6) Å, β 134.38(5)°, *V* 568.3(6) $Å^3$. Le groupe spatial est tout probablement *C*2/*m*, avec *Z* = 4. D_{calc} est 9.41 g/cm^3. Les six raies les plus intenses du spectre [*d* en Å(I)(*hkl*)] sont: 5.953(6)(001), 4.144(10)(020), 2.917(9)$(\bar{2}20)$, 2.413(8)(022), 2.365(7)(422) et 2.082(5)(040). La laflammeïte aurait cristallisé à partir d'une phase fluide hydrothermale ou d'un liquide tardif dans un milieu à faible teneur en soufre mais à la fois enrichi en plomb, les éléments du groupe du platine et des composants volatils.

(Traduit par la Rédaction)

Mots-clés: laflammeïte, sulfure de Pd–Pb, nouvelle espèce minérale, minéral du groupe du platine, élément du groupe du platine, plomb, complex stratiforme, roches mafiques et ultramafiques, Kirakkajuppura, Penikat, Finlande.

INTRODUCTION

Recently, we reported the occurrence of a previously unknown chalcogenide, $Pd_3Pb_2S_2$, from the Kirakkajuppura platinum-group-element (*PGE*) deposit in the Early Proterozoic Penikat layered complex, Finland (Barkov *et al.* 1999). Our aim here is to describe the properties of this Pd–Pb sulfide, which is structurally related to parkerite, $Ni_3Bi_2S_2$.

We have named the new Pd–Pb sulfide *laflammeite* after J.H. Gilles Laflamme (b. 1947), of the Mining and Mineral Sciences Laboratories, Canada Centre for Mineral and Energy Technology (CANMET), Ottawa, Canada, in recognition of his important contributions to the characterization of platinum-group minerals. The mineral and its name were approved prior to publication by the Commission on New Minerals and Mineral Names, IMA (no. 2000–014). The type material is deposited at the Canadian Museum of Nature, Ottawa, Canada (CMNMC 83195).

OCCURRENCE AND ASSOCIATED MINERALS

The Kirakkajuppura *PGE* deposit (Fig. 1) is associated with the sulfide-poor Sompujärvi *PGE* reef. It is located near the interface between ultramafic and gabbroic cumulates, close to the contact with the country rocks at the northern end of the intrusion (Alapieti & Lahtinen 1986, Halkoaho *et al.* 1990, Halkoaho 1993). This deposit locally contains unusually high concentrations of the *PGE* (possibly surpassing 0.5 kg/t total *PGE*, mainly Pd), observed in altered mafic-ultramafic rocks, predominantly pyroxenite which typically contains ≤200 ppm S and 0.1–3 wt.% Cr, and thus is nearly sulfide-free. The *PGE* mineralization is associated with actinolite and clinochlore, accessory chromite, minor base-metal sulfides (*BMS*: chalcopyrite, bornite, millerite, and secondary chalcocite), and an unnamed Pb–V oxide ($Pb^{2+}{}_4V^{5+}{}_2O_9$), a rare accessory (Barkov *et al.* 1999).

The principal platinum-group minerals (*PGM*) at Kirakkajuppura are Pt–Ni-poor vysotskite, zvyagintsevite, an unnamed Pd–Pb oxide ($Pd^{2+}{}_9Pb^{2+}O_{10}$) formed at the expense of zvyagintsevite (Barkov *et al.* 1999), and various *PGE*-rich thiospinels, members of cuprorhodsite–ferrorhodsite and cuprorhodsite–malanite series (Barkov *et al.* 2000). Nearly all the sulfur in the *PGE*-rich, *BMS*-poor samples is accounted by the *PGE*-rich sulfides, mainly vysotskite, which is clearly the predominant *PGM* (Barkov *et al.* 1999).

The other *PGM* and *PGE*-rich phases observed in the deposit are laflammeite, laurite–erlichmanite, irarsite, keithconnite, a Pb-rich phase $[Pd_{20}(Te_6Pb)_7]$ that likely is related to keithconnite, unnamed Rh(Ni,Fe,Cu)$_2S_3$ and (Pd,Pt)Cu, and native gold, which contains up to 9 wt.% Pd (Barkov *et al.*, unpubl. data). In addition, we observed a series of unusual Pb–(Pd)-rich thiospinels (or compounds related to thiospinels) of Cu, Fe, Rh and Ir, which will be described in a separate publication.

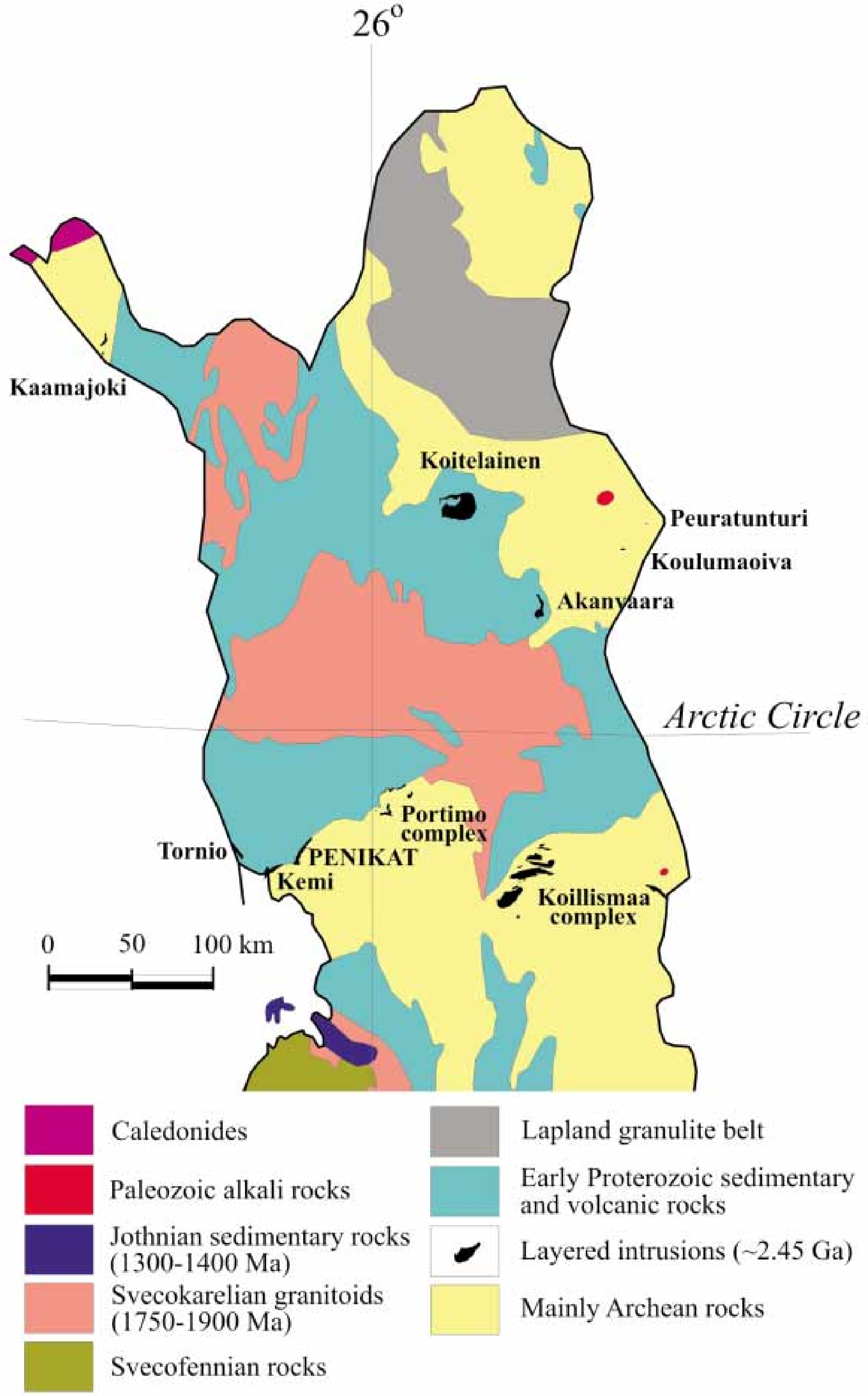
PROPERTIES OF LAFLAMMEITE

Appearance and physical properties

Commonly, laflammeite occurs as individual subhedral platelets up to 150 μm in the longest dimension (Figs. 2A, B), and in intergrowths with members of the vysotskite–braggite series (Figs. 3A, B). The largest crystal of laflammeite, which is subhedral, reaches 0.3 mm in size (Fig. 4).

Laflammeite is opaque. The color of the powder is dark gray and metallic looking. Fine twins are present in the crystals. The cleavage, inferred to be {010}, is perfect. The mineral seems to be quite brittle. Microhardness values of laflammeite (two indentations on two grains: VHN_{50}) are 156 and 185, the average is 171 kg/mm^2, which corresponds to a Mohs hardness of about 3½. The indentations are fractured and slightly distorted, and they produced stepped slippage along the linear cleavage. More indentations were impossible because of the characteristic cleavage of the mineral. Parkerite (1.3 wt.% Pb) from Noril'sk has a somewhat lower microhardness (119–138 kg/mm^2: Genkin *et al.*

FIG. 1. Generalized geological map of Finland, showing the location of the Penikat layered complex, which hosts the Kirakkajuppura PGE deposit.


26°
Kaamajoki
Koitelainen
Peuratunturi
Koulumaoiva
Akanvaara
Arctic Circle
Portimo complex
Tornio
PENIKAT
Kemi
Koillismaa complex
0
50
100 km
Caledonides
Paleozoic alkali rocks
Jothnian sedimentary rocks (1300-1400 Ma)
Svecokarelian granitoids (1750-1900 Ma)
Svecofennian rocks
Lapland granulite belt
Early Proterozoic sedimentary and volcanic rocks
Layered intrusions (~2.45 Ga)
Mainly Archean rocks

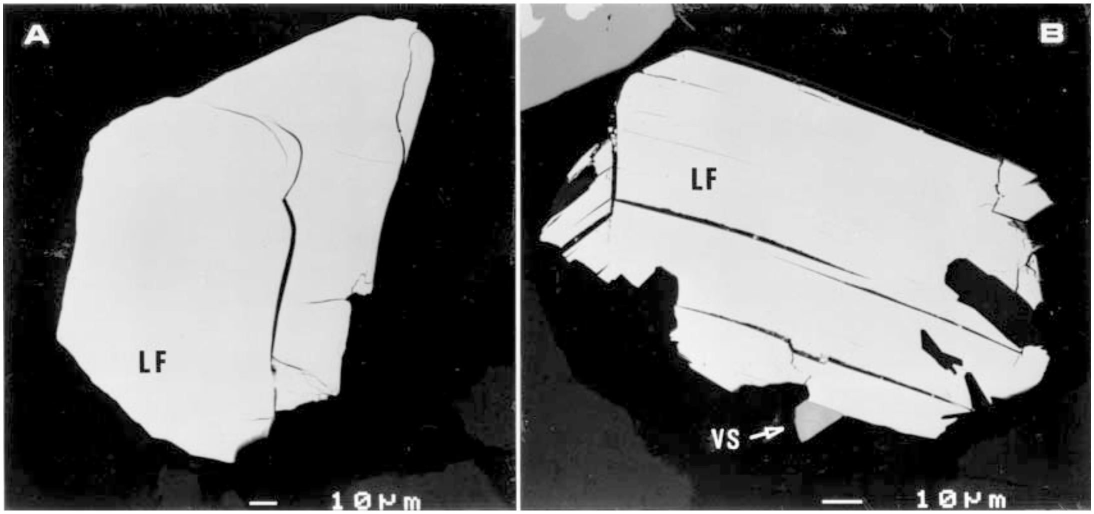


FIG. 2. Subhedral crystals of laflammeite (LF) from the Kirakkajuppura *PGE* deposit, Penikat layered complex. A tiny grain attached to the crystal shown in Figure 1B is vysotskite (VS): $(Pd_{0.86-0.90}Pt_{0.14-0.11}Ni_{0.03})_{\Sigma 1.04}S_{0.96}$. The surrounding material is epoxy (black). Back-scattered electron images. The scale bar is 10 µm in each case.

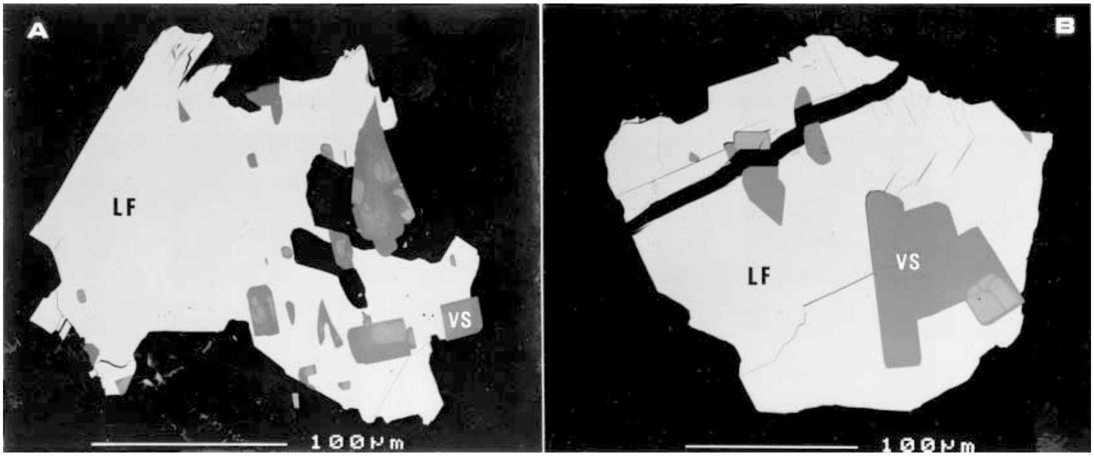


FIG. 3. Intergrowths of laflammeite (LF) with complexly zoned vysotskite (VS). The composition of the vysotskite varies from $(Pd_{0.99}Ni_{0.05}Pt_{<0.01})_{\Sigma 1.04}S_{0.96}$ to $(Pd_{0.81}Pt_{0.19}Ni_{0.04})_{\Sigma 1.04}S_{0.96}$. The surrounding material is epoxy (black). Back-scattered electron images. The scale bar is 100 µm in each case.

1981). The density of laflammeite could not be measured because of the small grain-size.

Optical properties

Laflammeite is cream-colored with a brownish tint in reflected light in air. Internal reflections were not observed. Its anisotropy is weak, from brownish gray to grayish brown. The bireflectance is weak, and pleochroism was not observed. Reflectance and color values are listed in Tables 1 and 2.

The optical spectra of laflammeite are distinct from those of rhodplumsite (Fig. 5), a rhodium–lead sulfide that displays a similar stoichiometry ($Rh_3Pb_2S_2$) but crystallizes with a shandite-type structure and has a strong anisotropy and bireflectance (Genkin *et al.* 1983).

The composition and formula

The composition of five grains of laflammeite is quite uniform. Mainly, it contains Pd, Pb and S. In addition, the presence of a minor concentration of Ir (~1 wt.%, *i.e.,* 0.05 Ir atoms per formula unit, *apfu*) is suggested by our electron-microprobe data (Table 3). This feature is quite unusual for a Pd-based mineral. Bismuth was sought (wavelength-dispersion spectroscopy), but not detected. This finding contrasts with the association of Bi and Pb established in parkerite–(shandite) from Noril'sk (up to 12 wt.% Pb, located in the Bi site: Ponomarenko *et al.* 1987). The mean composition of laflammeite (Table 3) corresponds to the ideal formula $Pd_3Pb_2S_2$ [*cf.* parkerite: $Ni_3(Bi,Pb)_2S_2$, monoclinic; shandite: $Ni_3Pb_2S_2$, trigonal, pseudocubic].

Many phases of the type $M_3M_2X_2$ and having the shandite or parkerite structure have been synthesized (*e.g.*, Range *et al.* 1983, Natarajan *et al.* 1988). A Mössbauer study of one of these compounds, $Ni_3Sn_2S_2$, which has a shandite-type structure (Range *et al.* 1997), reveals that the valence state of Ni and Sn is 0 and 2+, respectively (Gutlich *et al.* 1999). There are bonding interactions between Ni and S and between Ni and Sn, whereas no evidence of bonding was found among Ni atoms in synthetic $Ni^0{}_3Sn^{2+}{}_2S^{2-}{}_2$. By analogy, bonding interactions between adjacent Pd atoms may be absent in the structure of laflammeite, so that the formal charges of the participating elements could be $Pd^0{}_3Pb^{2+}{}_2S^{2-}{}_2$.

TABLE 1. REFLECTANCE VALUES OF LAFLAMMEITE FROM PENIKAT, FINLAND

λ nm	R_1 % (air)	R_2 % (air)	R_1 % (oil)	R_2 % (oil)
400	44.0	45.2	30.5	31.9
420	44.55	45.8	30.95	32.25
440	44.9	46.0	31.4	32.55
460	45.2	46.1	31.5	32.55
470	45.3	46.2	31.55	32.45
480	45.35	46.2	31.55	32.5
500	45.6	46.4	31.75	32.6
520	45.9	46.8	32.1	33.05
540	46.3	47.2	32.45	33.5
546	46.5	47.55	32.6	33.75
560	46.7	47.75	32.9	34.1
580	47.4	48.5	33.5	34.7
589	47.7	48.8	33.9	35.05
600	48.0	49.05	34.2	35.25
620	48.6	49.4	34.85	35.75
640	49.3	49.85	35.55	36.2
650	49.6	50.0	36.0	36.3
660	49.85	50.0	36.25	36.45
680	50.55	50.15	36.8	36.6
700	51.3	50.4	37.55	36.7

The spectra were obtained with a Zeiss Axiotron MPM80 spectrophotometer. The reflectance standard used was WTiC, and the oil used was $n = 1.515$. The standard wavelengths (COM) are underlined.

TABLE 2. COLOR VALUES OF LAFLAMMEITE FROM PENIKAT, FINLAND

	Illuminant *C*				Illuminant *A*			
	R_1	R_2	$^{im}R_1$	$^{im}R_2$	R_1	R_2	$^{im}R_1$	$^{im}R_2$
x	0.317	0.316	0.319	0.319	0.454	0.454	0.457	0.456
y	0.320	0.320	0.321	0.321	0.408	0.408	0.407	0.407
Y %	46.9	47.8	33.0	34.1	47.2	48.2	33.4	34.5
λ_d	584	584	586	586	593	592	616	616
P_e %	2.9	2.8	3.7	3.5	4.5	4.4	3.7	3.3

Note: Reflectance values in air and in oil are given in Table 1.

X-ray powder-diffraction data and the relationship with known species and phases

Single-crystal X-ray studies could not be carried out because of the finely twinned nature of the crystals available for study (as revealed in an attempted study by a precession single-crystal method). Thus, laflammeite was studied by X-ray powder diffraction, using 114.6 mm Debye–Scherrer camera with a Gandolfi attachment and Cu*K*α radiation.

The pattern obtained (Table 4) indicates that laflammeite is possibly isostructural with parkerite (Brower *et al.* 1974), on the basis of the overall similarity of the powder patterns. It also is related to a family of synthetic parkerite-type sulfides such as $Pd_3Bi_2S_2$ (PDF 38–1279: *a* 11.745, *b* 8.312, *c* 8.306 Å, β 134.99°) and $Rh_3Bi_2S_2$ (PDF 38–1281: *a* 11.271, *b* 8.371, *c* 7.933 Å, β 133.29°), which all have monoclinic symmetry, space group *C*2/*m* and $Z = 4$ (Range *et al.* 1983). A broad line at 4.781 Å ($I = 2$) and a few very weak lines ($I < 1$) observed in the laflammeite pattern are ascribed to a minor admixture of an unknown material.

TABLE 3. COMPOSITION OF LAFLAMMEITE, $Pd_3Pb_2S_2$, FROM THE PENIKAT COMPLEX, FINLAND

	1	2	3		1	2	3
n	43	26					
Pd wt.%	39.77	39.16	40.02	Pd *apfu*	2.97	2.96	3.00
Ir	n.a.	1.08	–	Ir	–	0.05	–
				Σ	2.97	3.01	3.00
Pb	52.98	52.01	51.94	Pb	2.03	2.02	2.00
S	8.07	7.90	8.04	S	2.00	1.98	2.00
Total	100.82	100.15	100.00				

Columns 1 and 2: laflammeite; column 3: $Pd_3Pb_2S_2$. The results of wavelength-dispersion analyses, WDS (in weight %), which led to composition 1, were acquired with a JEOL JXA-8900 instrument (20 kV, 20 nA). The following X-ray lines and standards were used: Pd*L*α (pure Pd and synthetic Pd_3HgTe_3), Pb*L*α and S*K*α (PbS). n.a.: not analyzed. Composition 2 is the average of two sets of WDS electron-microprobe analyses. They were carried out with two JEOL instruments, which were operated at an acceleration voltage of 20 kV and beam current of 20 nA and 15 nA, respectively. The following X-ray lines and standards were used: Pd*L*α (pure Pd), Pb*L*α (PbS and PbTe), Ir*L*α (pure Ir) and S*K*α (PbS and pyrite). The amounts of Pt, Rh, Ru, Os, Fe, Ni, Co, Bi are ≤0.15 wt.%. Sn and As were sought, but not detected. The atomic proportions are based on seven atoms per formula unit (*apfu*). *n*: number of analyses made.

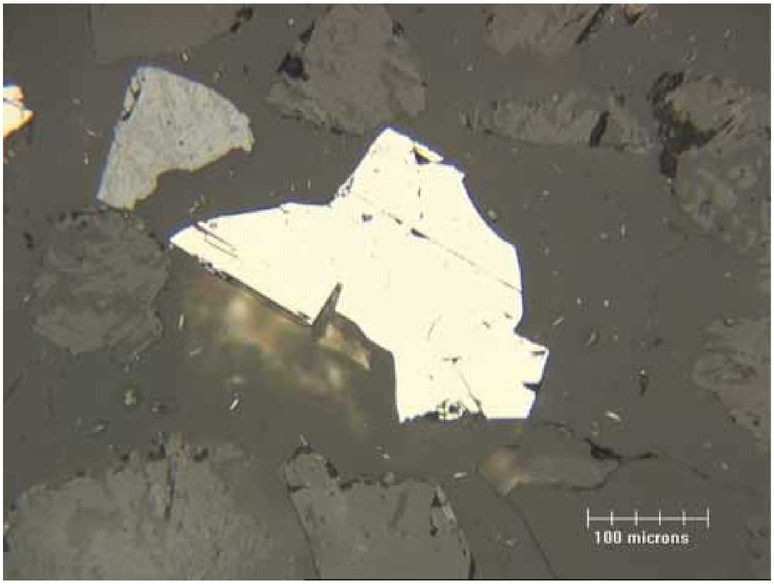


FIG. 4. A digital image of a large subhedral crystal of laflammeite. The surrounding material is epoxy. Scale bar is 100 μm.

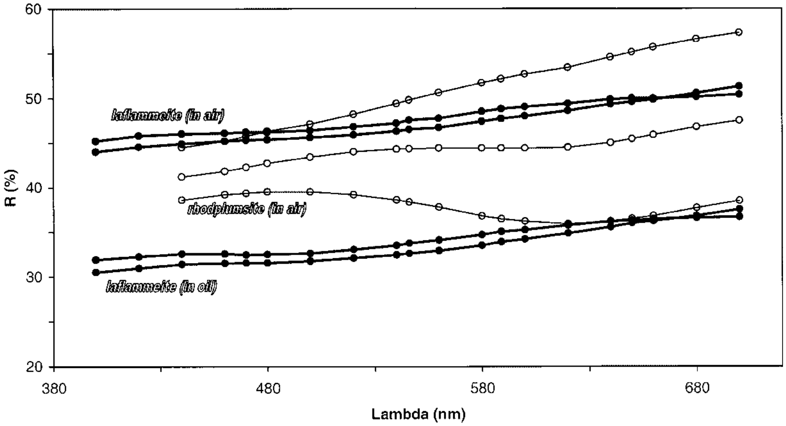


FIG. 5. Comparison of reflectance data for laflammeite (filled symbol) and rhodplumsite, $Rh_3Pb_2S_2$, from the Urals (open symbol: these three reflectance curves measured in air vary with optical orientation of rhodplumsite (Genkin *et al.* 1983). The reflectance of laflammeite was measured in air and in oil; values (R%) are plotted *versus* wavelength λ in nm.

Initially, the eight most intense diffraction maxima in the powder pattern of laflammeite were indexed by analogy with the related phases: $Pd_3Bi_2S_2$, $Rh_3Bi_2S_2$ and $Pd_3Bi_2Se_2$ (Range *et al.* 1983). On the basis of that successful cell-parameter refinement, two or three more lines were indexed, whose predicted position matched observed peaks; the *hkl* of those peaks matched observed *hkl* of peaks in the reference patterns. The assignments were done in a stepwise fashion. Our data extended beyond range of observations of the related phases; in all cases, we assigned *hkl* values of such peaks to predicted position of peaks consistent with inferred symmetry of the mineral.

The unit-cell parameters of laflammeite, refined from the powder data, are *a* 11.521(11), *b* 8.294(10), *c* 8.321(6) Å, β 134.38(5)°, *V* 568.3(6) Å^3. The space group is inferred to be *C*2/*m* and *Z* = 4, by analogy with the other parkerite-type phases. The *a*:*b*:*c* ratio calculated from the unit-cell parameters is 1.389:1:1.003, and the calculated density of laflammeite is 9.41 g/cm^3.

THE ENVIRONMENT OF FORMATION

The Kirakkajuppura deposit of the Penikat complex differs from other *PGE* deposits in layered intrusions by the presence of very high levels of the *PGE* in a hydrothermally altered rock that is very poor in sulfur. The low concentration of S in the environment is consistent with the high (Pd + Pb) / S value, 2.5 (atomic basis), observed in laflammeite.

The *PGM* in the Kirakkajuppura deposit are associated with Mg-rich hydrous silicates. Laflammeite displays an intergrowth relationship with the Pt-poor vysotskite (Figs. 3A, B). A low temperature of formation of the vysotskite, a phase that accounts for most of the sulfur in the *BMS*-poor (or *BMS*-free) rock, is implied by textural data (Barkov *et al.* 1999), and is consistent with the available experimental data (Cabri *et al.* 1978).

A number of uncommon *PGM* developed in the altered pyroxenite at Kirakkajuppura are enriched in Pb: zvyagintsevite, Pb–(Pd)-rich thiospinels of Cu, Fe, Rh and Ir, Pb-rich keithconnite, and unnamed Pd_9PbO_{10}, all are associated with laflammeite, and this assemblage is indicative of a relative enrichment in Pb in the environment. The enrichment in Pb at a late stage of hydrothermal alteration is clearly indicated by the development of chain-like veinlets of zvyagintsevite among the hydrous silicates in this rock (Barkov *et al.* 1999). Laflammeite likely formed from a hydrothermal fluid (or a late-stage liquid) in a micro-environment poor in sulfur and rich in Pd, Pb and volatiles.

TABLE 4. X-RAY POWDER-DIFFRACTION DATA FOR LAFLAMMEITE FROM PENIKAT, FINLAND

I	d_{meas}	d_{calc}	*hkl*	I	d_{meas}	d_{calc}	*hkl*
$\underline{6}$	5.953	5.947	001	1	1.711	1.712	$\bar{2}24$
$\underline{10}$	4.144	4.147	020	1	1.676	1.676	$\bar{6}24$
4	3.379	3.402	021	<1	1.612	1.612	$\bar{5}31$
1	2.985	2.973	002	<1	1.524	1.522	312
$\underline{9}$	2.917	2.922	$\bar{2}20$	3	1.461	1.461	$\bar{4}40$
2	2.603	2.602	$\bar{4}03$	<1	1.431	1.433	043
$\underline{8}$	2.413	2.416	022	<1	1.388	1.388	242
7	2.365	2.366	$\bar{4}22$	<1	1.367	1.369	$\bar{6}42$
<1	2.201	2.192	$\bar{4}21$	1	1.263	1.263	$\bar{4}26$
5	2.082	2.073	040	2B	1.242	1.241	$\bar{8}26$
2	2.052	2.059	400	<1	1.209	1.208	044
<1	1.882	1.880	$\bar{2}04$	<1	1.184	1.184	$\bar{2}45$

Note: B broad line. Values of d_{meas} and d_{calc} are expressed in Å. Intensities of the strongest four lines are underlined.

ACKNOWLEDGEMENTS

Our study was supported by research grants from the Natural Sciences and Engineering Research Council of Canada and from the Academy of Finland. This paper is dedicated to Dr. L.J. Cabri, J.H.G. Laflamme's close colleague and the team leader over the years, on the occasion of his retirement. We are grateful to M.A. Cooper, of the University of Manitoba, Winnipeg, for providing the X-ray powder-diffraction pattern, and for his attempts to carry out a single-crystal X-ray study of laflammeite. We also thank G. Poirier (McGill University) for his expert assistance with electron-microprobe analyses, and J.H.G. Laflamme and L.J. Cabri for the color digital image of laflammeite. Helpful comments of K. Kojonen, an anonymous referee, J.A. Mandarino, and Associate Editor N.J. Cook improved this paper.

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Received September 8, 2000, revised manuscript accepted July 15, 2001.