

Origin of alunite- and jarosite-group minerals in the Mt. Leyshon epithermal gold deposit, northeast Queensland, Australia

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ABSTRACT

The origin of alunite-jarosite family minerals, found in surface samples from the Mt. Leyshon Au deposit, northeast Queensland, has been determined using electron microprobe and textural data. The base-metal contents of the alunite-group minerals provide significant additional information to help evaluate whether they formed during hypogene or weathering processes.

K- and Na-rich members of the alunite group $(K,Na)Al_3(SO_4)_2(OH)_6$ generally form as hydrothermal minerals in the acid-sulfate epithermal system at Mt. Leyshon. Because of their high-level formation they may be affected by subsequent weathering, during which they may incorporate Ag, Bi, Cu, and Pb derived from sulfides. Pb-rich members of the alunite group [plumbogummite, $PbAl_3H(PO_4)_2(OH)_6$, and hinsdalite, $PbAl_3(PO_4)(SO_4)(OH)_6$] that form during weathering derive components from both sulfides and apatite.

Jarosite-group minerals, $(K,Na)Fe_3(SO_4)_2(OH)_6$, that replace alunite-group minerals during weathering retain the Na-K ratio of the parent; substantial Al is also retained by the resultant jarosite. Jarosites that directly replace pyrite have low Al contents and Na-K ratios that differ from those of any alunite-group minerals present within the sample.

INTRODUCTION

Minerals in the alunite-jarosite family have the general formula $AB_3(XO_4)_2(OH)_6$ where A is a large cation (like K^+ , Na^+ , NH_4^+ , Pb^{2+} , and Ca^{2+}) held in 12-fold coordination. B sites are occupied mainly by trivalent Al or Fe for alunite and jarosite groups, respectively. Cu and Zn may also be minor but important components in the B sites. The (XO_4) sites are usually occupied by SO_4^{2-} , PO_4^{3-} , or AsO_4^{3-} .

Extensive solid solution occurs within all three sites (Scott, 1987) with that between alunite, $KAl_3(SO_4)_2(OH)_6$, and natroalunite, $NaAl_3(SO_4)_2(OH)_6$, and their Fe analogues, i.e., jarosite, $KFe_3(SO_4)_2(OH)_6$, and natrojarosite, $NaFe_3(SO_4)_2(OH)_6$, being particularly well studied experimentally (e.g., Parker, 1962; Brophy and Sheridan, 1965). Pb-rich members of the alunite-jarosite family generally have some trivalent anions present within the (XO_4) sites to balance the effect of the divalent cations in the A site, e.g., hinsdalite, $PbAl_3(PO_4)(SO_4)(OH)_6$.

Alunite-group minerals are often formed under acid oxidizing conditions in hypogene porphyry Cu and epithermal Au deposits (e.g., Knight, 1977). However such minerals may also form during weathering of sulfides in these types of deposits (e.g., Bladh, 1982). Thus in weathered environments the origin of alunite-group minerals may not be obvious.

Minerals in the alunite-jarosite family are well developed in outcropping volcanic breccias at the Mt. Leyshon Au deposit, 110 km southwest of Townsville, northeast Queensland. The breccias occur in a roughly circular (di-

ameter ~2 km) Permian volcanic complex at the boundary of the Ordovician-Devonian Ravenswood Granodiorite and metasediments of the Carboniferous-Ordovician Puddler Creek Formation (Fig. 1).

In outcrop the complex shows zones characterized by strong development of kaolinite, iron oxides, and alunite- and jarosite-group minerals within a broader iron-oxide-jarosite zone and peripheral chlorite-feldspar zone. Au occurs in outcrop in the alunite zone (Scott, 1988). Below the zone of weathering the mineralization consists of gold and electrum associated with pyrite and $Cu \pm Pb \pm Zn \pm Bi$ sulfides within pervasive sericitic alteration (Morrison et al., 1987).

Because they have been observed only in the weathered top 60 m of the deposit, the alunite-jarosite minerals have been considered to be weathering products (Morrison et al., 1987). K-Ar dating of surficial alunite has suggested that such formation occurred 2–3 Ma (Bird et al., 1987). However, this study of the distribution of the alunite-jarosite minerals at Mt. Leyshon and their textural and compositional features suggests that many of the alunites originally had a hydrothermal origin.

SAMPLES AND METHODS

A total of 316 samples was collected from the surface of the deposit prior to the commencement of mining in 1986. Of these samples, 126 contain members of the alunite-jarosite family. Because alunite-group minerals were generally discolored by admixed jarosite, or goethite, or both, X-ray diffractometry was used to distinguish the

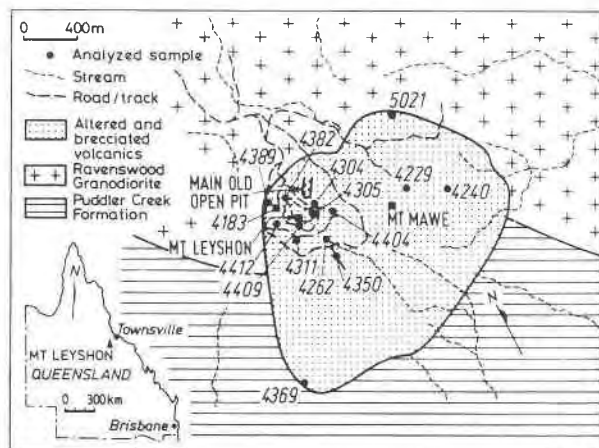


Fig. 1. Extent of alteration and brecciation in outcrop at Mt. Leyshon and location of samples analyzed.

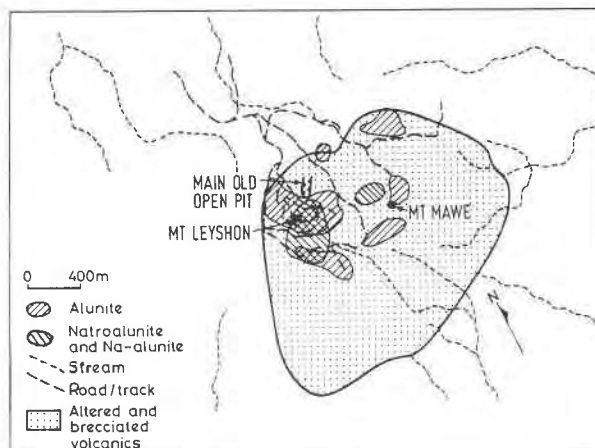


Fig. 2. Distribution of alunite-group minerals, Mt. Leyshon.

alunite and jarosite groups (cf. Scott, 1987). Fifty-nine samples (including 24 bearing alunite-jarosites) were analyzed chemically. On the basis of the mineralogical and chemical data, 184 electron microprobe analyses were made on 15 alunite-jarosite-bearing samples from the altered volcanics (Fig. 1).

Samples were analyzed using a Camebax electron microprobe operating at 20-kV accelerating voltage with a beam current of 10 nA. The beam was defocused to a diameter of 20 μm to avoid thermal damage during counting periods of 5 s. By arranging the counting sequence so that the thermally mobile major elements (Na, K, Al, and S) were counted first and less abundant elements later, consistent analytical results were obtained. No observable beam damage was done to the sample under these conditions. Oxide totals were generally lower than expected for stoichiometric alunite-jarosites owing to the porous nature of many samples or the possibility of unanalyzed components, e.g., NH_4^+ , H_3O^+ in A sites (Altaner et al., 1988; Ripmeester et al., 1986). Therefore, mineral compositions were calculated on the basis of 2 mol (XO_4) as recommended by Scott (1987).

OCCURRENCE OF ALUNITE-GROUP MINERALS

Members of the alunite group present in outcrop at Mt. Leyshon can be divided into K-, Na-, and Pb-rich varieties.

Alunites [i.e., with compositions close to $\text{KAl}_3(\text{SO}_4)_2(\text{OH})_6$] are particularly well developed about Mt. Leyshon (Fig. 2) and are characterized by up to 10% Na in A sites and Fe up to 12% in B sites (Table 1). In some samples (4183, 4229, and 5021) alunite occurs as 100- μm blebs replacing potassium feldspar. Alunite from sample 4409 contains appreciable Ag and Bi and is coated by iron oxides. In samples 4305 and 4389 it occurs as veins up to 5 mm thick. Alunite veins in sample 4389 contain pyrite grains $\sim 100 \mu\text{m}$ across (Fig. 3). The alunite in sample 4305 is green and contains a relatively high Cu

content (Table 1) and 30 μm -sized spherules of Cu-rich goethite. Although jarosite-group minerals are also present in some of these alunite-bearing samples, no jarosite-group minerals are present in samples 4305, 4389, and 4409 (Table 1).

Sodian alunite (i.e., alunite with greater than 10% of A sites occupied by Na) and natroalunite have a more restricted distribution than alunite but are especially well developed on the southern flank of Mt. Leyshon (Fig. 2). Between 30 and 53% of their A sites are occupied by Na (Table 1). The Fe contents of sodian alunite blebs in 4382 are generally similar to those in alunites. However, the high Fe content of natroalunite from sample 4304 probably reflects its intergrowth with sodian jarosites and na-

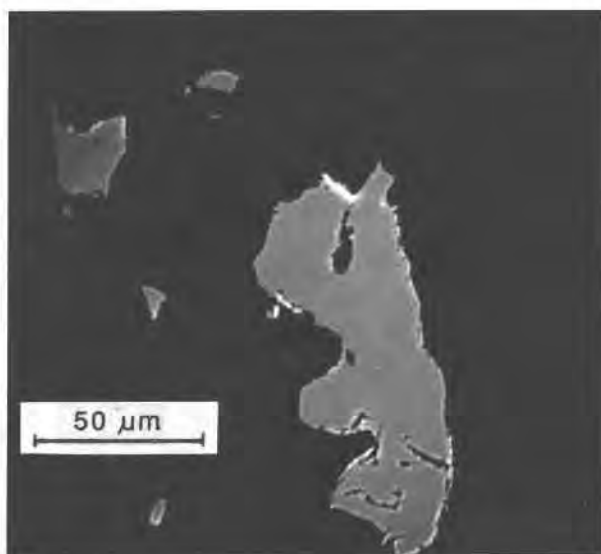


Fig. 3. Pyrite (gray) rimmed by copper sulfide (white) in alunite vein (black) (BSE image, sample 4389).

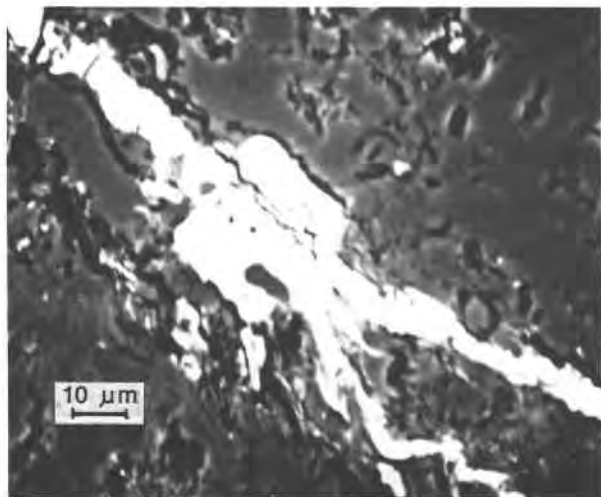


Fig. 4. Plumbogummite vein (white) cutting hematite cast after pyrite (BSE image, sample 4382).

trojarosite. Significant PO_4 occurs in the XO_4 sites in sodian alunites and natroalunite from 4382 and 4304.

Pb-rich members of the alunite group are also phosphate-rich (Table 1), with compositions varying between plumbogummite, $\text{PbAl}_3\text{H}(\text{PO}_4)_2(\text{OH})_6$, and hinsdalite, $\text{PbAl}_3(\text{SO}_4)(\text{PO}_4)(\text{OH})_6$, and have been identified within the alunite zones at Mt. Leyshon and northeast of Mt. Mawe. These Pb-rich minerals contain significant Ca and Zn and generally are present as veins or blebs closely associated with iron oxides (Fig. 4). The plumbogummite and hinsdalite in sample 4409 have Bi contents up to 700 ppm (Table 1).

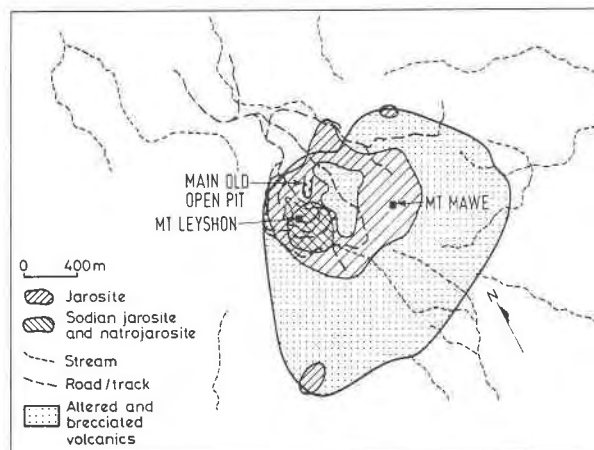


Fig. 5. Distribution of jarosite-group minerals, Mt. Leyshon.

OCCURRENCE OF JAROSITE-GROUP MINERALS

Jarosite-group minerals have a more extensive distribution than alunite-group minerals within the weathered rocks of the volcanic complex at Mt. Leyshon (Fig. 5).

Jarosite [i.e., $\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$] contains up to 13% Na in A sites with 40 to 64% of such sites being occupied by Na in sodian jarosite and natrojarosite, $\text{NaFe}_3(\text{SO}_4)_2(\text{OH})_6$ (Table 2). The jarosite-group minerals frequently occur admixed with muscovite and goethite or replacing alunite (Fig. 6). When jarosite replaces alunite, it retains the Na-K ratio of the original alunite, e.g., both alunites and jarosites are Na-poor in sample 4229 and Na-rich in sample 4304 (Tables 1 and 2). Thus the distribution of Na-rich jarosites

TABLE 1. Compositions of alunite-group minerals at Mt. Leyshon

Sample	No. of analyses	A Site					B Site				XO ₄ Site		Trace-element contents			
		K	Na	Ba	Pb	Ca	Al	Fe	Cu	Zn	SO ₄	PO ₄	Pb	Cu	Zn	
(wt%)																
(a) Alunite																
4183*	5	1.00	0.02	0.01	0	—	2.48	0.35	0.01	0	1.98	0.02	<0.01	0.10	0.04	
4229*	9	0.86	0.10	0.01	0	—	2.59	0.34	0.01	0	1.98	0.02	<0.01	0.07	0.05	
4305	7	0.98	0.02	0.01	0	—	2.63	0.14	0.05	0	2.00	0	<0.01	0.76	0.02	
4350*	7	0.94	0.01	0.01	0	0	2.33	0.32	0.01	0	1.99	0.01	<0.01	0.07	<0.01	
4389	6	0.94	0.08	0	0	—	2.67	0.10	0.01	0	1.99	0.01	<0.01	0.12	0.05	
4409	8	0.91	0.06	0	0.02	—**	2.79	0.23	0.01	0	1.97	0.03	0.77	0.07	0.05	
5021*	8	0.88	0.03	0.01	0.02	0	2.90	0.08	0	0	1.99	0.01	0.64	0.02	0.02	
(b) Natroalunite and sodian alunite																
4304*	5	0.29	0.53	0	0	—	1.67	1.32	0	0	1.92	0.08	<0.01	0.02	0.02	
4382*	8	0.51	0.30	0.01	0	—	2.64	0.27	0.01	0	1.90	0.10	<0.01	0.07	<0.01	
4382*	2	0.41	0.38	0	0	—	2.74	0.26	0.02	0	1.91	0.09	<0.01	0.08	<0.01	
(c) Plumbogummite and hinsdalite																
4382*	10	0.03	0.01	0.03	0.73	0.22	2.78	0.10	0.01	0	0.11	1.89	—	0.07	0.05	
4404*	5	0.07	0	0.01	0.87	—	2.88	0.09	0.01	0.02	0.47	1.53	—	0.03	0.19	
4404*	1	0.40	0.03	0.02	0.50	—	2.70	0.27	0.01	0.02	1.09	0.91	—	0.07	0.19	
4409	6	0.06	0.01	0.01	0.82	0.09†	2.87	0.12	0	0.02	0.16	1.84	—	<0.01	0.18	
4409	6	0.18	0	0.01	0.69	0.11†	2.87	0.14	0	0.02	0.42	1.58	—	<0.01	0.18	
4409	4	0.31	0.01	0.01	0.52	0.10†	2.88	0.14	0	0.02	0.69	1.31	—	<0.01	0.16	

Note: No significant AsO_4 was found. Compositions are based on the general formula $\text{AB}_3(\text{XO}_4)_2(\text{OH})_6$ with XO_4 normalized to 2.

* Jarosite-group minerals also present in these samples.

** Ag = 470, Bi = 220 ppm also present.

† Bi = 700 ppm also present.

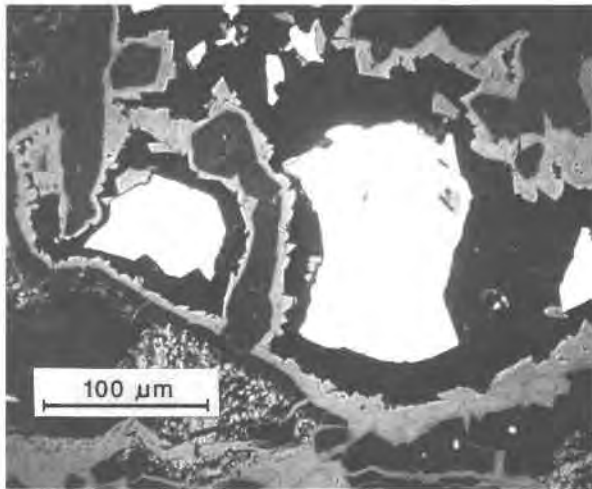


Fig. 6. Weathering of pyrite (white) leading to the development of voids (black) and jarosite (gray) replacement of alunite (dark gray) (BSE image, sample 4229).

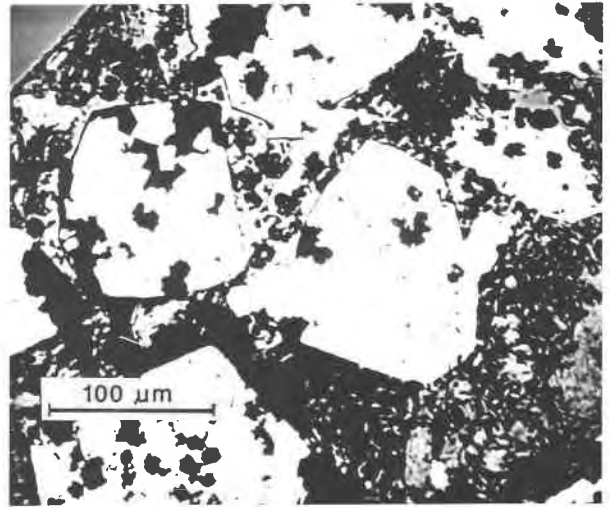


Fig. 7. Jarosite (white) casts after pyrite. Muscovite (gray) and quartz (black) (BSE image, sample 4311).

corresponds closely with the distribution of Na-rich alunites at Mt. Leyshon (cf. Figs. 2 and 5). However, where there is no physical contact between alunite and jarosite, the Na-K ratios may be quite dissimilar (as in samples 4350 and 4382, Tables 1 and 2).

The Al content of the jarosite-group minerals may be quite high, especially when they replace alunites (e.g., sample 4304). However, where jarosites directly replace pyrite (Fig. 7), Al within B sites is low (sample 4311, Table 2). No Pb-rich jarosite-group minerals have been found (Table 2).

DISCUSSION

In the advanced argillic alteration zone of an epithermal Au deposit, alunite, kaolinite, and gold are associated. Such acid-sulfate epithermal deposits also tend to be Cu-rich, to contain bismuthinite, and to have alunite replacing potassium feldspar (Hayba et al., 1986; Stoffregen, 1987). These features all occur at Mt. Leyshon (e.g., Morrison et al., 1987), although previous workers have considered the alunite and kaolinite to be derived by weathering.

TABLE 2. Compositions of jarosite-group minerals at Mt. Leyshon

Sample	No. of analyses	A site					B site				XO ₄ site			Zone*
		K	Na	Ba	Pb	Ca	Fe	Al	Cu	Zn	SO ₄	PO ₄	AsO ₄	
(a) Jarosite														
4229	3	0.72	0.13	0	0	—	2.77	0.16	0	0	1.99	0.01	0	A
4229	3	0.78	0.08	0	0	—	2.35	0.68	0	0.01	1.97	0.03	0	A
4240	6	0.80	0.06	0.10	0.04	—	2.02	0.93	0.04	0.01	1.59	0.38	0.03	J
4350	9	0.82	0.09	0	0	0	2.58	0.36	0	0	1.99	0.01	0	A
4369	8	0.90	0.01	0.01	0.01	0.01	2.85	0.13	0	0	1.90	0.10	0	J
4382	5	0.83	0.01	0	0	—	2.67	0.23	0.02	0.01	1.98	0.02	0	A
4412	5	0.89	0.08	0.01	0	0	2.84	0.16	0	0.01	1.92	0.08	0	J
4412	10	0.93	0.03	0.01	0	0	2.69	0.30	0	0.01	1.89	0.11	0	J
(b) Natrojarosite and sodian jarosite														
4262	5	0.52	0.40	0	0	0	2.82	0.12	0	0	1.91	0.08	0.01	A
4262	5	0.52	0.45	0	0	0	2.87	0.07	0	0.01	1.98	0.02	0	A
4304	3	0.47	0.42	0	0	—	2.67	0.34	0	0.01	1.97	0.03	0	A
4304	1	0.33	0.47	0	0	—**	2.58	0.59	0	0	1.91	0.08	0	A
4304	2	0.30	0.48	0	0	—	1.99	0.92	0	0	1.95	0.05	0	A
4304	6	0.28	0.51	0	0	—	1.79	1.32	0	0	1.93	0.07	0	A
4311	6	0.51	0.48	0	0	0	2.96	0.04	0	0	1.97	0.03	0	A
4311	3	0.49	0.52	0	0	0	2.98	0.02	0	0	1.97	0.03	0	A
4311	5	0.40	0.57	0	0	0	2.98	0.02	0	0	1.97	0.03	0	A
4311	2	0.31	0.64	0	0	0	2.99	0.01	0	0	1.97	0.03	0	A

Note: Compositions are based on the general formula $AB_3(XO_4)_2(OH)_6$ with XO₄ normalized to 2.

* A = Alunite- and jarosite-group minerals present. J = Jarosite-group minerals only.

** 0.01 Ag in this analysis.

Outcrop from the highly altered and weathered area centered on Mt. Leyshon, where Au is also present (Scott, 1988), contains alunite intimately associated with goethite, but jarosite is absent, e.g., samples 4305, 4409 (Table 1, Fig. 1). The work of Bladh (1982) indicates that such assemblages reflect oxidation of pyrite under quite Al-rich conditions and are likely to occur during the weathering of advanced argillic alteration assemblages. A K-Ar age of ~2–3 Ma is reported for the analysis of two such samples (Bird et al., 1987). However when one of these samples was dated, using the Rb-Sr method, a Permian age (260 Ma) was obtained (Whitford et al., 1988). If both these determinations are correct, this feature is best explained by weathering of original hypogene alunite during which the K-Ar systematics are affected but the Rb-Sr systematics are not.

Sample 4389 contains pyrite within an alunite vein (Fig. 3). The alunite has a $\delta^{34}\text{S}$ value = 9.8‰, whereas the coexisting pyrite has $\delta^{34}\text{S}$ = 8.8‰ (A. S. Andrew, personal communication, 1987). Bird et al. (1989) regard this as indicating that the alunite is secondary, i.e., derived from weathering sulfides (cf. Jensen et al., 1971). However because this alunite also has an old Rb-Sr age (D. J. Whitford, personal communication, 1987) the more likely explanation is that the alunite is formed by oxidation of H_2S above the water table (cf. Hayba et al., 1986) and is hypogene.

In addition, base-metal contents of alunites also appear to differ depending upon the extent of exposure to weathering. Those alunites least likely to have been affected by weathering (i.e., those still bearing sulfides, e.g., 4229 and 4389) tend to have base-metal contents <0.2 wt% whereas weathering-affected alunites (i.e., those with intimately associated goethite, e.g., 4305 and 4409) have higher base-metal contents (>0.6 wt%, Table 1). The actual base metals present may be either Cu or Pb \pm Bi \pm Ag, suggesting contributions from Cu-bearing sulfides or Pb-Bi-bearing sulfosalts, respectively. Both types of sulfides are present in unweathered rocks at depth (Morrison et al., 1987).

The low base-metal contents of natroalunite and sodian alunite in 4304 and 4382 suggest that these occurrences are also hypogene. Furthermore, Na-rich members of the alunite group can only form from solutions with very high Na-K ratios (Parker, 1962); such solutions are unlikely to be extensively developed during weathering of the muscovite-rich rocks in the Au-rich zone at Mt. Leyshon. However, high Na-K ratios are retained when alunite is altered to jarosite during weathering (see below).

The origin of plumbogummite and hinsdalite is not so easily evaluated. Stoffregen and Alpers (1987) have found P-rich members of the alunite group that formed by replacement of apatite during advanced argillic alteration at the Summitville epithermal Au deposit. Similarly, the presence of Ca in the P-rich alunite-group minerals at Mt. Leyshon may suggest that apatite was a significant factor in their genesis. However, the significant Bi, Cu, and Zn contents of these Pb-rich minerals (Table 1) may also imply incorporation of base metals derived from weathering

sulfides. Thus Pb- and P-rich alunite-group minerals may have originally formed as Ca-rich members of the alunite group (e.g., woodhouseite as at Summitville; Stoffregen and Alpers, 1987) with subsequent weathering leading to the incorporation of Pb and other base metals. Alternatively, plumbogummite and hinsdalite may have formed directly by acid attack on apatite during weathering of Pb-rich sulfides. Because such sulfides are intimately associated with Au at Mt. Leyshon (Morrison et al., 1987), either hypothesis satisfactorily accounts for the restriction of the lead phosphates to the alunite zone.

Jarosite-group minerals replace alunites in some cases (Fig. 6) with simple one-for-one substitution of Fe for Al, and the retention of some Al and the Na-K ratio of the original alunite. In contrast, jarosite formed by replacement of pyrite (Fig. 7) has a low Al content.

CONCLUSIONS

K- and Na-rich alunite-group minerals originally formed as hydrothermal alteration minerals at Mt. Leyshon. Because of their formation close to the original surface, i.e., above the water table, they were susceptible to modification during subsequent weathering. During such weathering they may have incorporated Ag, Bi, Cu, and Pb derived from the breakdown of sulfides.

Pb- and P-rich alunite-group mineral formation may have required the destruction of both apatite and Pb-rich sulfide. Such specialized conditions may be reflected by the restricted distribution of plumbogummite and hinsdalite at Mt. Leyshon.

Jarosite-group minerals at Mt. Leyshon formed either by replacement of alunites, retaining the Na-K ratio of the parental alunite and generally some Al, or directly from pyrite, resulting in only a low Al content.

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