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Solid solution in, and classification of, gossan-derived members of the alunite-jarosite family, northwest Queensland, Australia

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ABSTRACT

Minerals of the alunite-jarosite family with the general formula $AB_3(XO_4)_2(OH)_6$ occur in gossans related to Pb-Zn mineralization in the Mount Isa region of northwest Queensland. Examination of 226 electron-microprobe analyses indicates that extensive solid solution occurs within the A, B and (XO_4) sites, with the variations in the A and (XO_4) sites often related.

Substitution of divalent cations for monovalent cations in the A sites of alunite or jarosite can be balanced by (1) replacing two monovalent cations by one divalent cation and leaving A-site vacancies, as in plumbojarosite, (2) incorporating divalent ions in the B sites, as in osarizawaite and beaverite, or (3) replacing divalent anions with trivalent anions, as in beudantite. A second trivalent anion can still maintain charge balance provided it is protonated, as in plumbogummite. Complex interaction of the mechanisms often occurs.

Complete solid solution between Al and Fe in B sites occurs at least in the more phosphate-rich alunite-jarosites. Nevertheless, because of the probable stability differences and ease of practical subdivision, separation into alunite and jarosite supergroups according to greater Al or Fe occupancy is considered appropriate.

A classification that prevents proliferation of named minerals of the alunite-jarosite family has been approved by the Commission on New Minerals and Mineral Names of the International Mineralogical Association.

INTRODUCTION

Minerals of the alunite-jarosite family have the general formula $AB_3(XO_4)_2(OH)_6$, where A is a large cation, such as Na^+, K^+, Ag^+, NH_4^+, H_3O^+, Ca^{2+}, Pb^{2+}, Ba^{2+}, Sr^{2+}, Ce^{3+}, and other rare earth elements in 12-fold coordination. B sites are occupied by the cations Al^{3+}, Fe^{3+}, Cu^{2+}, and Zn^{2+} in octahedral coordination. The anion $(XO_4)^{y-}$ may be SO_4^{2-}, PO_4^{3-}, AsO_4^{3-}, CO_3^{2-}, SbO_4^{3-}, CrO_4^{2-}, or SiO_4^{2-}. Al or Fe is the major occupant of the B sites, but with the plethora of possible combinations in the A and (XO_4) sites, the minerals are usually considered in three groups based on the nature of the anions (e.g., Palache et al., 1951; Strunz, 1978; Ramdohr and Strunz, 1978): (1) alunite group, with two divalent (XO_4) anions and usually monovalent A cations; (2) beudantite or woodhouseite group, with one divalent and one trivalent (XO_4) anion and usually divalent A cations; (3) plumbogummite, crandallite, or goyazite group, with two trivalent (XO_4) anions and either divalent or trivalent A cations.

As seen above, different names have been proposed for the same groups. In this paper the nomenclature of Palache et al. (1951) has been followed, with some slight modification.

The subdivisions above reflect the strong relationship between A and (XO_4) occupancy. Substitution of one trivalent for one divalent anion as in the beudantite group is accompanied by a concurrent change in the A-site cation from monovalent to divalent. Complete replacement by a trivalent anion is accompanied by substitution of a trivalent cation on the A site as in the case of florencite, $CeAl_3(PO_4)_2(OH)_6$. Alternatively, if the A-site cation remains divalent, protonation of one of the trivalent anions also results in a stable compound, as in the case of gorceixite, $BaAl_3H(PO_4)_2(OH)_6$ (Radoslovich, 1982). In this paper it is argued that these alternatives are sufficiently different to warrant separate classification as "florencite" and "plumbogummite" groups.

Although solid solution has been frequently reported in the A sites, similar substitution within the B and (XO_4) sites of minerals is poorly documented. This paper presents compositional data for 67 minerals of the alunite-jarosite family found in 46 gossan samples derived by oxidation of mid-Proterozoic Pb-Zn mineralization in northwest Queensland, Australia. It shows that phosphate-sulfate solid solution from $PO_4/SO_4 = 0$ to at least 1 is related to the occupancy of the A sites. Extensive Fe-Al substitution, especially in volcanogenic deposits to the east of Mount Isa, is also reported.

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SUBSTITUTION AND CLASSIFICATION—A REVIEW

In addition to monovalent ions, divalent Ca^{2+} and Pb^{2+} may substitute in the A sites. Two different modes of substitution are known: some A sites are left unoccupied to maintain the total A-site charge at one, as in minamiite, $(Na_{0.36}K_{0.10}Ca_{0.27})Al_3(SO_4)_2(OH)_6$ (Ossaka et al., 1982), or plumbojarosite, $Pb_{0.5}Fe_3(SO_4)_2(OH)_6$ (Szymanski, 1985), in which 27% and 50%, respectively, of the A sites are unoccupied. Alternatively, in beaverite, $Pb(Fe,Cu)_3(SO_4)_2(OH)_6$, and its Al analogue, osarizawaite, $Pb(Al,Cu)_3(SO_4)_2(OH)_6$, the charge balance is maintained by a combination of partial substitution of the divalent ions, Cu and Zn, for trivalent ions in the B sites and by some deficiency in the occupancy of these B sites (Jambor and Dutrizac, 1983).

Within the plumbogummite group, where A is generally divalent, the charge balance was originally assumed to be restored by protonation of one of the OH radicals, giving plumbogummite the formula $PbAl_3(PO_4)_2(OH)_5 \cdot H_2O$. However, structural work (Blount, 1974; Radoslovich, 1982) indicates that the extra proton is actually attached to one of the XO_4 anions so that the generalized formula is better written $AB_3(XO_4)(XO_3OH)(OH)_6$ or $AB_3H(XO_4)_2(OH)_6$. With one divalent and one trivalent anion, such minerals could be considered as members of, or a subgroup within, the beudantite group. However, the protonation of a trivalent anion modifies the P–O bond lengths to make its space group *Cm* rather than $R\bar{3}m$ (Radoslovich, 1982), thereby justifying retention of the classification "plumbogummite group." Members with a trivalent A-site cation and two "true" trivalent anions need to be distinguished from the "plumbogummite group" as defined above. Therefore they are designated members of the "florencite group."

Solid solution in the A sites is well documented within groups, e.g., among jarosite, natrojarosite, and hydronium jarosite $(K,Na,H_3O)Fe_3(SO_4)_2(OH)_6$ (Brophy and Sheridan, 1965). However, substitution involving different valencies in the A sites has also been documented between the endmembers florencite, $CeAl_3(PO_4)_2(OH)_6$, crandallite, $CaAl_3H(PO_4)_2(OH)_6$, gorceixite, $BaAl_3H(PO_4)_2(OH)_6$, and goyazite, $SrAl_3H(PO_4)_2(OH)_6$ (McKie, 1962). Here substitution of divalent Ca, Ba, or Sr for trivalent Ce could be balanced by either substitution of SO_4^{2-} for PO_4^{3-} or protonation of phosphate. Although McKie (1962) found some minor sulfate substitution, protonation of the phosphate appears to be the major factor in balancing the charge. However, in the system alunite $[(Na,K)Al_3(SO_4)_2(OH)_6]$–woodhouseite $[CaAl_3(PO_4)(SO_4)(OH)_6]$–goyazite $[SrAl_3H(PO_4)_2(OH)_6]$ (Wise, 1975), variable A-site valencies are balanced by SO_4-PO_4 intersubstitution. Similar large-scale intersubstitution of divalent and trivalent anions has recently been reported in the cases of schlossmacherite, $(H_3O_{0.63}Ca_{0.25}Na_{0.06}K_{0.04}Sr_{0.01}Ba_{0.01})(Al_{2.84}Cu_{0.06}Fe_{0.03})(SO_4)_{1.46}(AsO_4)_{0.54}(OH)_{5.46}$ (Schmetzer et al., 1980), and philipsbornite, $(Pb_{0.90}Mn_{0.03})(Al_{2.76}Zn_{0.09}Cu_{0.07}Fe_{0.05})H_{0.86}(AsO_4)_{1.57}(CrO_4)_{0.40}(SO_4)_{0.10}(OH)_{5.72}$ (Walenta et al., 1982), where intersubstitutions occur in A (and B) sites as well. Nevertheless, except for minor variation about the named stoichiometric endmembers, large-scale substitution in (XO_4) sites is not well documented.

Table 1. Ionic radii for A- and B-site cations and bond lengths for (XO_4) anions (in Å)

B sites		A sites				XO_4 sites*	
Al^{3+}	0.51	Na^+	0.97	H_3O^+	1.24	C–O	1.22
Fe^{3+}	0.64	Ca^{2+}	0.99	Ag^+	1.26	S–O	1.43
Ti^{4+}	0.68	La^{3+}	1.02	K^+	1.33	Si–O	1.50
Cu^{2+}	0.72	Ce^{3+}	1.03	Ba^{2+}	1.34	P–O	1.56
Zn^{2+}	0.74	Sr^{2+}	1.12	NH_4^+	1.43	As–O	1.78
		Pb^{2+}	1.20				

Note: Data from Weast (1968, p. F152–F157) and Botinelly (1976).
* Average X–O bond lengths.

Substantial solid solution between Al and Fe in B sites is also poorly documented, although intermediate compositions between alunite, $KAl_3(SO_4)_2(OH)_6$, and jarosite, $KFe_3(SO_4)_2(OH)_6$, do exist (Brophy et al., 1962). Similarly a series between osarizawaite, $Pb(Al,Cu)_3(SO_4)_2(OH)_6$, and beaverite, $Pb(Fe,Cu)_3(SO_4)_2(OH)_6$, is known (Paar et al., 1980), but in this case there is the added complication of divalent ion incorporation in B sites. Alunite may form under higher pH conditions than jarosite (Hladky and Slansky, 1981). Stability differences between otherwise similar Al- and Fe-rich members also occur generally (Hladky, pers. comm., 1982). Furthermore, because of the size difference between Al^{3+} and Fe^{3+} (Table 1), B-site occupancy influences a_0 and is readily differentiated by X-ray diffractometry and optical methods. Therefore Botinelly's (1976) suggestion of initial subdivision into separate Al- and Fe-rich series has much in its favor despite the degree of intersubstitution illustrated below.

In such a scheme, initial subdivision would be into the "alunite supergroup" or "jarosite supergroup" depending on the dominant B-site cation. Logically, the valency of the (XO_4) anions would then be used to further subdivide the series into groups as in Table 2. Three factors could be used to argue against the proposed classification, namely, possible confusion in nomenclature (i.e., "alunite" and "jarosite" refer both to a supergroup and a specific group within that supergroup), the comparative rarity of members of the lusungite group, and the absence of known Fe equivalents of the florencite group. However, these factors are outweighed by the practical considerations of the ease of the initial subdivision into Al- and Fe-rich series and the differences in formation conditions.

GEOLOGIC SETTING

Minerals of the alunite-jarosite family occur in gossans at Mount Isa (in the Bernborough, BSD, DDH Z625, and Black Star areas), Copalot, Mount Novit, Hilton, Lake Moondarra, Kamarga, Dugald River, Jolimont, Pegmont, and Fairmile (Fig. 1). Except for the last four prospects, the deposits are associated with Carpentarian dolomitic shale in a 40-km north-south belt centered on Mount Isa

Table 2. Classification of groups in the alunite-jarosite family

		B site	
A site	(XO_4) site (mol)	Alunite supergroup (Al > Fe)	Jarosite supergroup (Fe > Al)
A^+	$(XO_4)^{3-} \leq 0.5$	Alunite group Alunite Natroalunite Schlossmacherite Minamiite Osarizawaite	Jarosite group Jarosite Natrojarosite Hydronium jarosite Ammoniojarosite Argentojarosite Plumbojarosite Beaverite
A^{2+}	$0.5 < (XO_4)^{3-} < 1.5$	Hinsdalite group Hinsdalite Hidalgoite Weilerite Kemmlitzite Svanbergite Woodhouseite	Beudantite group Beaudantite Corkite
A^{2+}	$(XO_4)^{3-} \geq 1.5$	Plumbogummite group Plumbogummite Goyazite Gorceixite Crandallite Philipsbornite	Lusungite group Lusungite Dussertite Zairite
A^{3+}	$(XO_4)^{3-} \geq 1.5$	Florencite group Florencite ?Waylandite ?Eyelettersite	—

and at Kamarga 220 km to the northwest. The Dugald River deposit is 90 km northeast of Mount Isa in a generally higher-grade metamorphic terrane with slates rather than shales, but it shows similarities to the preceding deposits (Taylor and Scott, 1982, 1983). The mineralogy of gossans and ironstones from these shale- or slate-hosted deposits is almost invariably goethite, hematite, quartz, minerals of the alunite-jarosite family, muscovite, and kaolinite. Pb- and Zn-bearing manganese oxides occur at Mount Isa and Dugald River. Cerussite, anglesite, and malachite-rosasite, although occasionally found in outcrop, generally occur deeper in the gossan profile (Taylor and Scott, 1982).

Jolimont is a Zn prospect, 160 km southeast of Mount

Table 3. Compositions and names of selected minerals of the alunite-jarosite family, northwest Queensland

Location	Sample	No. of analyses	Composition*	Name
Pegmont	50619	2	$Pb_{0.91}(Al_{2.44}Fe_{0.27}Zn_{0.01})(PO_4)_{1.93}(SO_4)_{0.07}(OH)_{4.04}$	Plumbogummite
Pegmont	50619	1	$Pb_{0.91}(Al_{2.01}Fe_{0.58}Zn_{0.01})(PO_4)_{1.57}(SO_4)_{0.43}(OH)_{4.03}Cl_{0.11}$	Ferrian plumbogummite
Pegmont	50619	1	$Pb_{0.91}(Fe_{1.85}Al_{0.90})(PO_4)_{1.96}(SO_4)_{0.04}(OH)_{4.13}$	Aluminian lusungite
Pegmont	50628A	8	$Pb_{0.97}(Al_{1.95}Fe_{0.98})(PO_4)_{1.38}(SO_4)_{0.62}(OH)_{5.35}$	Ferrian hinsdalite
Pegmont	50628A	2**	$Pb_{1.10}(Fe_{2.11}Al_{0.89})(PO_4)_{1.12}(SO_4)_{0.88}(OH)_{6.08}$	Aluminian corkite
Fairmile	58594	5	$Pb_{0.92}Ca_{0.01}(Fe_{2.48}Al_{0.32})(PO_4)_{1.28}(SO_4)_{0.72}(OH)_{4.18}$	Corkite
Fairmile	58612	2	$Pb_{0.92}K_{0.04}(Fe_{2.66}Al_{0.24})(SO_4)_{1.29}(PO_4)_{0.71}(OH)_{5.87}$	Corkite
Jolimont	50636	4†	$Sr_{0.44}Ca_{0.27}Ba_{0.13}Ce_{0.10}K_{0.05}La_{0.02}(Al_{2.99}Zn_{0.01})(PO_4)_{1.71}(SO_4)_{0.29}(OH)_{5.37}$	Calcian goyazite
Dugald River	50568	4	$K_{0.56}Pb_{0.15}(Fe_{2.64}Zn_{0.19}Cu_{0.02})(SO_4)_{2.00}(OH)_{5.16}Cl_{0.04}$	Plumbian jarosite
Dugald River	50569	3**	$Pb_{0.87}K_{0.14}(Al_{2.45}Fe_{0.55})(SO_4)_{1.22}(PO_4)_{0.66}(AsO_4)_{0.12}(OH)_{6.12}$	Ferrian hinsdalite
Kamarga	63541	5†	$Pb_{0.82}Ca_{0.12}Ba_{0.07}(Al_{2.68}Zn_{0.22}Cu_{0.10})(SO_4)_{1.01}(PO_4)_{0.99}(OH)_{5.83}Cl_{0.10}$	Calcian hinsdalite
Bernborough	58241	7**	$Pb_{0.84}K_{0.05}(Al_{2.58}Zn_{0.28}Fe_{0.07}Cu_{0.07})(SO_4)_{1.48}(PO_4)_{0.49}(AsO_4)_{0.03}(OH)_{5.86}$	Hinsdalite
Bernborough	58244	3†	$Pb_{0.49}K_{0.36}Ca_{0.02}(Al_{2.74}Zn_{0.18}Cu_{0.08})(SO_4)_{1.67}(PO_4)_{0.27}(AsO_4)_{0.06}(OH)_{5.73}$	Plumbian alunite
BSD	58230	6**	$K_{0.47}Pb_{0.17}Ca_{0.06}(Al_{2.60}Fe_{0.17}Cu_{0.07}Zn_{0.06})(SO_4)_{1.74}(PO_4)_{0.26}(OH)_{5.54}$	Plumbian alunite
Copalot	66169A	3**	$Pb_{0.94}K_{0.04}(Al_{2.15}Zn_{0.42}Fe_{0.41}Cu_{0.02})(SO_4)_{1.08}(PO_4)_{0.92}(OH)_{5.53}Cl_{0.03}$	Hinsdalite
Black Star	58349	3†	$K_{0.71}Pb_{0.39}Ca_{0.03}(Al_{2.24}Zn_{0.62}Cu_{0.14})(SO_4)_{1.92}(PO_4)_{0.08}(OH)_{5.67}Cl_{0.04}$	Zincian alunite
DDH Z625	59439	1†	$K_{0.73}Sr_{0.23}Pb_{0.07}Ca_{0.04}Ba_{0.03}(Al_{2.85}Cu_{0.10}Zn_{0.05})(SO_4)_{1.69}(PO_4)_{0.31}(OH)_{5.93}Cl_{0.09}$	Strontian alunite
DDH Z625	59465	4**	$K_{0.82}Pb_{0.08}Ca_{0.04}(Al_{2.99}Fe_{0.01})(SO_4)_{1.79}(PO_4)_{0.21}(OH)_{5.82}Cl_{0.03}$	Plumbian alunite
Hilton	50377	4**	$Pb_{0.51}K_{0.38}Ba_{0.01}(Al_{2.65}Fe_{0.27}Zn_{0.05}Cu_{0.03})(SO_4)_{1.51}(PO_4)_{0.49}(OH)_{5.85}$	Plumbian alunite
Hilton	50382	6**	$K_{0.67}Pb_{0.10}(Al_{2.61}Fe_{0.35}Cu_{0.02}Zn_{0.02})(SO_4)_{1.87}(PO_4)_{0.13}(OH)_{5.67}Cl_{0.03}$	Plumbian alunite
Mount Novit	54529	5†	$Pb_{0.72}K_{0.11}Ca_{0.05}Ba_{0.03}(Al_{2.78}Zn_{0.16}Cu_{0.06})(SO_4)_{1.48}(PO_4)_{0.52}(OH)_{5.93}Cl_{0.06}$	Potassic hinsdalite
Mount Novit	58305	7**	$Pb_{0.93}K_{0.06}Ag_{0.02}(Al_{2.04}Fe_{0.84}Cu_{0.05}Ti_{0.04}Zn_{0.03})(PO_4)_{0.99}(SO_4)_{0.82}(AsO_4)_{0.20}(OH)_{5.69}Cl_{0.08}$	Ferrian hinsdalite
Lake Moondarra	54559	6†	$Ce_{0.29}K_{0.29}Ca_{0.22}La_{0.17}Sr_{0.10}Nd_{0.05}(Al_{2.94}Zn_{0.06})(PO_4)_{1.89}(SO_4)_{0.11}(OH)_{5.47}Cl_{0.04}$	Potassic florencite

* H^+ attached to (PO_4) ions not shown.

** Fe content reduced (see text).

† Fe and Al contents reduced (see text).

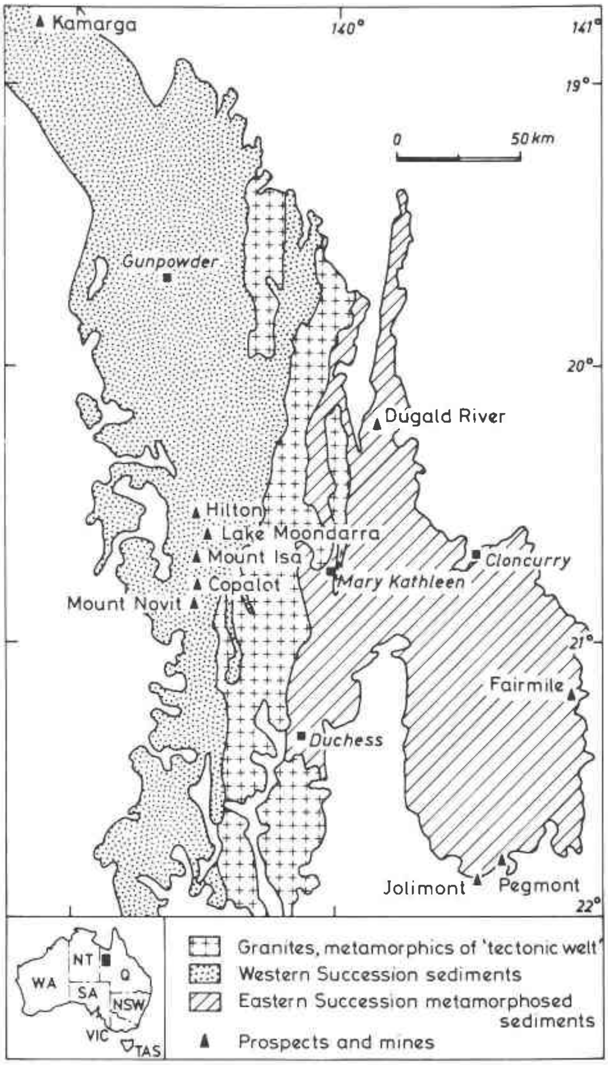

Fig. 1. Proterozoic outcrop of the Mount Isa inlier, showing Pb-Zn deposits and prospects sampled in this study.

Isa (Fig. 1), with outcropping samples consisting of weathered phyllites cut by kaolinite veins. Much of the Zn occurs in goethite, and the alunite-jarosite minerals are Sr-rich. The Pegmont and Fairmile deposits are volcanogenic and are associated with banded iron formations (BIF) in amphibolite-facies metamorphic rocks south of Cloncurry (Fig. 1). Surface samples are often stained black by manganese oxides and consist of assemblages of variable amounts of goethite, alunite-jarosites, graphite, hematite, quartz, pyromorphite, manganese oxides, spessartite, and kaolinite.

METHODS

Analytical details

Elemental compositions were determined using the 0–10-keV spectral range of the Link energy-dispersive X-ray system attached to a Microscan V wavelength-dispersive microprobe. Beam current was 5×10^{-9} A, and the voltage 20 kV. These conditions enabled determination of elemental contents down to 0.1–0.2 wt%. Owing to the possibility of overlap of the $SK\alpha$ and $PbM\alpha$ radiation on the energy-dispersive system, the wavelength-dispersive system was used to determine Pb and SO_3 down to 0.05 and 0.2 wt%, respectively. Samples from Pegmont and Fairmile were analyzed exclusively by wavelength-dispersive X-ray spectrometry before the Link system was available. Therefore, analyses of minerals of the alunite-jarosite family in these samples may not necessarily be complete.

Table 4. Compositions (wt%) of minerals of the alunite-jarosite family contaminated by iron oxides

	Location Sample no.	Hilton 50377		Mount Novit 54530		Mount Novit 54535	
	No. of analyses	4	2	6	2	2	1
	SiO_2	3.29	4.54	0.41	0.52	0.61	1.48
	Al_2O_3	21.33	19.96	22.75	21.39	28.31	25.52
	Fe_2O_3	15.98	21.51	10.13	15.04	3.78	10.82
	K_2O	2.86	2.73	1.82	1.47	1.82	1.53
	CaO	<0.12	<0.12	—	—	1.44	1.25
	BaO	<0.30	—	0.83	0.83	2.70	2.76
	SrO	—	—	—	—	0.41	0.33
	CuO	0.39	0.38	0.44	0.46	0.46	0.64
	ZnO	0.68	0.78	0.42	0.43	0.56	0.20
	PbO	18.15	15.05	24.20	23.17	19.11	17.18
	SO_3	19.13	18.72	17.74	16.63	17.08	15.19
	P_2O_5	5.52	4.67	9.02	8.32	11.72	10.61
	H_2O_{diff}	12.65	11.65	12.25	11.73	12.00	12.52
	Structural formula based on $2(SO_4,PO_4)$, $3(Al,Fe,Cu,Zn)$						
A	Pb	0.51	0.45	0.62	0.64	0.45	0.45
	K	0.38	0.39	0.22	0.19	0.20	0.20
	Ca	0	0	—	—	0.14	0.13
	Ba	0	—	0.03	0.03	0.09	0.10
	Sr	—	—	—	—	0.02	0.02
B	Al	2.65	2.62	2.56	2.59	2.94	2.94
	Fe	0.27*	0.29*	0.38*	0.35*	0*	0*
	Cu	0.03	0.03	0.03	0.03	0.03	0.05
	Zn	0.05	0.06	0.03	0.03	0.03	0.01
XO_4	SO_4	1.51	1.56	1.27	1.28	1.13	1.12
	PO_4	0.49	0.44	0.73	0.72	0.87	0.88

Note: Recalculated on the basis of the method described in the text.
* Fe content reduced so that (Al + Fe + Cu + Zn) = 3 mol.

Many of the alunite-jarosites occur as composite grains with iron oxide, quartz, muscovite, and kaolinite inclusions. The SiO_2 content of the latter three contaminants betrays their presence, but in the goethite- and hematite-rich gossans, the extent of any iron oxide contamination within the alunite-jarosite analyses is not always obvious because of possible Al-Fe substitution within the mineral itself. Thus, when compositions are reported in the following sections, analyses with the lowest Fe contents are taken as being the most pure, and values for several individual analyses have been averaged.

Data treatment

A total of 226 analyses has been grouped into 52 alunite supergroup and 15 jarosite supergroup averages (23 representative average analyses are quoted in Table 3). However, even for these analyses, much of the Fe content (and sometimes all of it, plus some of the Al) was superfluous to the requirements of the structural formula, $AB_3(XO_4)_2(OH)_6$. As the XO_4 anions are the major structural units in alunite-jarosites, excess Fe was regarded as being due to contamination, and mineral compositions were calculated on the basis of 2 mol (XO_4) and up to 3 mol B per formula unit. If the total B content exceeded 3 mol, the trivalent content

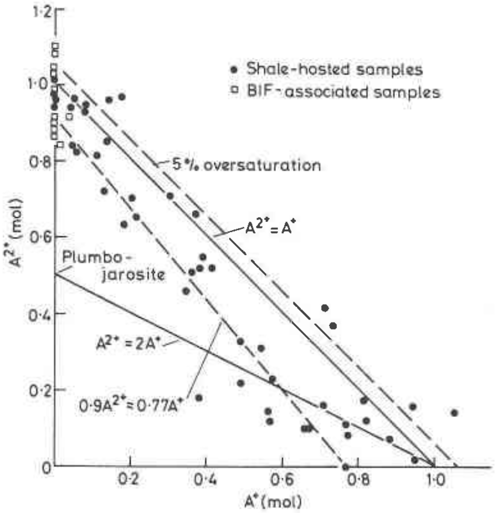

Fig. 2. Monovalent relative to divalent occupancy of A sites in samples from northwest Queensland.

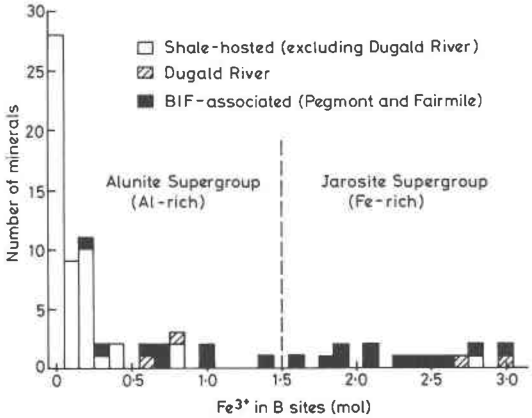

Fig. 3. Histogram of distribution of Fe^{3+} (mol) in B sites in samples from northwest Queensland.

was reduced by first excluding Fe and then, if necessary, Al. The number of OH radicals present is simply the number required to achieve electrostatic neutrality in the formula. However, because of the unknown extent of protonation of PO_4^{3-}, especially in double phosphates, the OH^- content may often be artificially low.

The validity of this method of data treatment is shown by the fact that similar compositions were obtained when data from a single sample that showed a range of Fe contents were treated in this way (Table 4). Nevertheless, analyses of highly contaminated alunite-jarosites are more likely to be in error than those for purer samples and consequently have not been used in calculating the compositions presented here. The abundances of the components of the alunite-jarosite minerals are given in moles per formula unit unless otherwise indicated.

RESULTS AND DISCUSSION

A-site occupation

K and Pb are the most abundant constituents in the A sites of the alunite-jarosite minerals of northwest Queensland (Table 3). When the A sites are partly occupied by divalent cations rather than solely by monovalent cations, total occupancy of the A sites generally falls well below the line representing perfect one for one ($A^{2+} = A^{+}$) substitution (Fig. 2). The line of best fit suggests that two types of substitution occur. Initially charge is conserved by a divalent cation substituting for two monovalent cations, i.e., $A^{2+} = 2A^{+}$, as in Pb^{2+} substitution for $2K^{+}$ in going from jarosite to plumbojarosite. This process dominates the region up to 0.2-mol A^{2+}, at which level 20% of the A sites are vacant (Fig. 2). Above this level of divalent substitution, the data points reflect substitution in the approximate proportions $A^{2+} = 0.86A^{+}$ (i.e., 0.9 $A^{2+} = 0.77$ A^{+}, Fig. 2), resulting in up to 14% vacancies in A sites.

In alunite-jarosite minerals, occupancy of A sites is generally greater than 70% (Fig. 2) but, as complete filling is uncommon (Botinelly, 1976), the overfilling by >5% in eight samples appears abnormal. Because of the "looseness" of the structure (Botinelly, 1976), it is possible that "excess" A-site cations are adsorbed onto the alunite-jarosite structure. Alternatively, some of the surplus divalent ions could be accommodated in B sites as in osarizawaite-beaverite (Jambor and Dutrizac, 1983). Cu^{2+} and Zn^{2+}, which are only 15% larger than Fe^{3+} (Table 1), have been so accommodated (Table 3). However, the other divalent cations in the alunite-jarosite type minerals are at least 50% larger than Fe^{3+} (Table 1) and are unlikely to be incorporated in B sites. As the method of calculation in this study does not preclude the possibility of unanalyzed components in (XO_4) sites, artificial overfilling of the A sites is also possible. The most likely additional component in this study is CO_3^{2-}, which is only slightly smaller than SO_4^{2-} anions (Table 1) and has been recorded in substantial amounts in plumbogummite (Fortsch, 1967) and in lesser amounts in alunites (Simpson, 1951; Keller et al., 1967). Therefore, the effect of not analyzing for components like CO_3^{2-} probably causes the "excess" of A-site cations, but additional work is required to resolve the problem completely.

B-site occupation

In the shale-hosted deposits at Kamarga, Dugald River, Jolimont, and about Mount Isa, all but three of the minerals of the alunite-jarosite family are members of the alunite supergroup (Fig. 3). Some of the analyses with $Fe^{3+} < 0.05$ mol could be artifacts reflecting the method of calculation. However, even if all of the analyzed Fe were present in B sites, only nine additional samples would be added to the jarosite supergroup.

As the possibility of complete solid solution between Al and Fe in the trivalent B sites exists (Brophy et al., 1962), it is desirable to indicate further the relative proportions of Al and Fe. Thus alunite supergroup members with $0.5 < Fe^{3+} < 1.5$ mol are referred to as "ferrian" and jarosite supergroup members with $0.5 < Al^{3+} < 1.5$ mol as "aluminian" (see Table 3).

The majority of Fe-rich alunite-jarosite family minerals

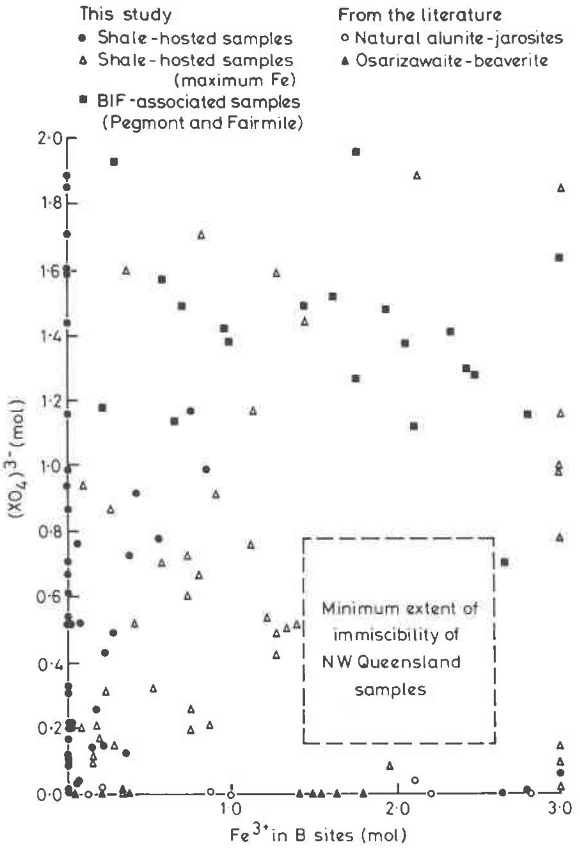

Fig. 4. Trivalent anions in XO$_4$ sites relative to Fe^{3+} in B sites for northwest Queensland and other natural samples. Data for alunite-jarosites from Simpson (1948; 1951, p. 661) and Brophy et al. (1962). Data for osarizawaite-beaverite from Kasymov (1958), Van Tassel (1958), Taguchi (1961), Morris (1962), Taguchi et al. (1972), Cortelezzi (1977), and Paar et al. (1980).

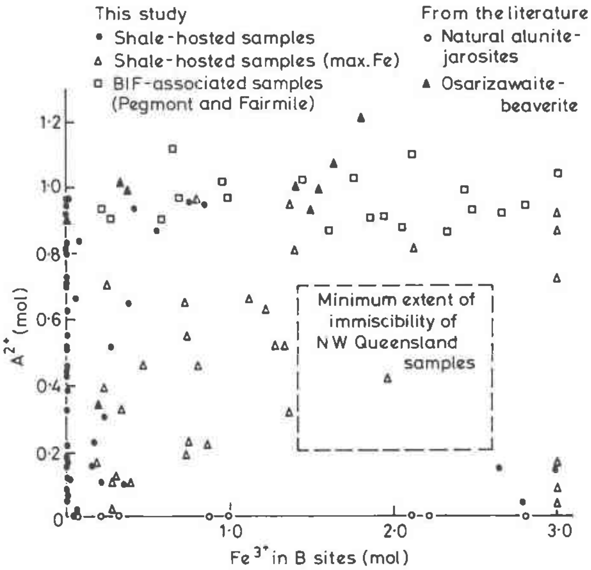

Fig. 5. Divalent cations in A sites relative to Fe^{3+} in B sites, showing extent of solid solution for northwest Queensland and other natural samples (from the literature as in Fig. 4).

occur at Dugald River, Pegmont, and Fairmile (Fig. 3), i.e., there are far more ferrian alunite-jarosites in prospects to the east of Mount Isa than in the shale- and slate-hosted deposits to the west. At Pegmont and Fairmile, the influence of pyromorphite, Pb$_5$(PO$_4$)$_3$Cl, on alunite-jarosite formation (Taylor and Scott, 1982) and the larger grain sizes of alunite-jarosites reduce the effects of possible contamination by aluminosilicates and iron oxides. Here the maximum amount of "excess Fe" excluded from the B sites is only 0.2 mol (compared with >3 mol in some shale-hosted samples), and B sites are often incompletely filled (Table 3). As the extent of Al-Fe substitution in these samples is not subject to any possible uncertainty (as in the alunite-jarosites from the shale-hosted deposits), the possibility of complete solid solution in B sites is demonstrated (Fig. 3).

Complete solid solution in B sites has been observed in sulfate-rich members of the alunite-jarosite family, e.g., alunite and jarosite (Brophy et al., 1962) and osarizawaite and beaverite. However, when the B-site occupancy of the northwest Queensland alunite-jarosites is considered relative to valencies in (XO$_4$) sites, complete solid solution occurs only where (XO$_4$)$^{3-}$ > 1.1 mol (Fig. 4). Similarly, A sites must be more than 80% filled by divalent cations to show continuous Al-Fe substitution (Fig. 5). Where (XO$_4$)$^{3-}$ < 1.1 mol and A^{2+} < 0.8 mol, the extent of miscibility of the northwest Queensland samples may be extended by considering all or part of the Fe to be in the B sites. Even then, a substantial gap remains where 0.15 < (XO$_4$)$^{3-}$ < 0.8 and 0.2 < A^{2+} < 0.7. Only sample 58349 (Black Star), which has (Zn + Cu) = 0.76 mol (Table 3), falls within the gap (Fig. 5), but even this sample would plot outside the gap if Zn and Cu were considered as Fe. Taken together, Figures 4 and 5 indicate that Al-Fe substitution is best developed within the alunite, hinsdalite, plumbogummite, and lusungite groups as well as between osarizawaite and beaverite. Further work is required to determine whether the compositional gaps in the jarosite and beudantite groups are real or merely sampling artifacts.

Cu and Zn consistently occur in the B sites (Table 3). In osarizawaite and beaverite, one-third of the B sites may be occupied by Cu, with subordinate Zn substitution (Taguchi, 1961; Morris, 1962; Taguchi et al., 1972; Cortelezzi, 1977). More than 2% and up to 7.6% Zn (by weight) occurs in seven shale-hosted minerals of the alunite-jarosite family in northwest Queensland (Table 3). Such levels are equivalent to 5 to 21% occupancies of the B sites and are up to double the 9% occupancies previously reported in natural materials (Livingstone and Cogger, 1966). Although Cu is preferentially incorporated relative to Zn, the work of Jambor and Dutrizac (1983) demonstrates the possibility of even higher Zn contents than the 3.8% they reported.

In northwest Queensland, more than 30% of the analyses from shale-hosted deposits have (Zn + Cu) > 0.1

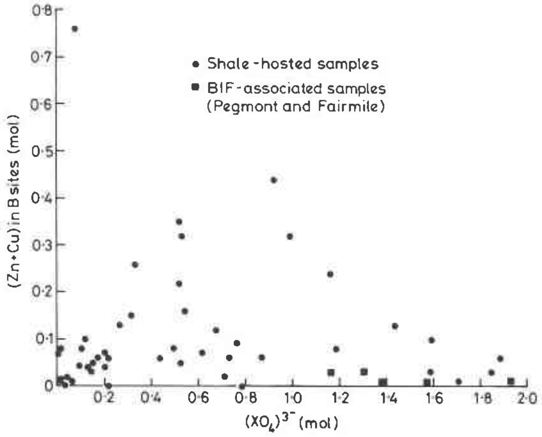

Fig. 6. (Zn + Cu) content of B sites relative to trivalent anion content of XO_4 sites in samples from northwest Queensland.

mol (Fig. 6). Where $0.5 < (XO_4)^{3-} < 1.5$ mol, 50% of the samples have $(Zn + Cu) > 0.1$ mol. However, where $(XO_4)^{3-} > 1.5$ mol, (Zn + Cu) contents are low (Fig. 6); indeed the maximum Zn and Cu contents recorded in the literature for phosphate-rich alunite-jarosites are 0.08 mol in philipsbornite (Walenta et al., 1982) and 0.07 mol in plumbogummite (Slansky, 1977), respectively. Thus, substantial Zn and Cu substitution in B sites occurs preferentially in sulfate-rich family members. The (Zn + Cu) content also tends to increase with Pb content, as in the artificially prepared beaverite–plumbojarosite–hydronium jarosite system (Jambor and Dutrizac, 1983).

(XO_4)-site occupation

Progressive substitution of trivalent for divalent anions is accompanied by a concurrent change from monovalent to divalent cations in A sites, i.e., $A^+B_3(XO_4)_2^{2-}(OH)_6 \rightarrow A^{2+}B_3(XO_4)^{3-}(XO_4)^{2-}(OH)_6$ (Fig. 7). In the alunite and jarosite groups, there is a cluster of analyses with $(XO_4)^{3-}$ and A^{2+} contents from zero to 0.25 mol. Except for the Zn- or Cu-bearing samples, above this level there is a gap until $(XO_4)^{3-}$ contents are about 0.45 mol. This gap corresponds to the region of greatest deficiency in A-site occupancy (Fig. 2). As no literature values have been found to fill the gap (Fig. 8), it may possibly represent a real compositional break.

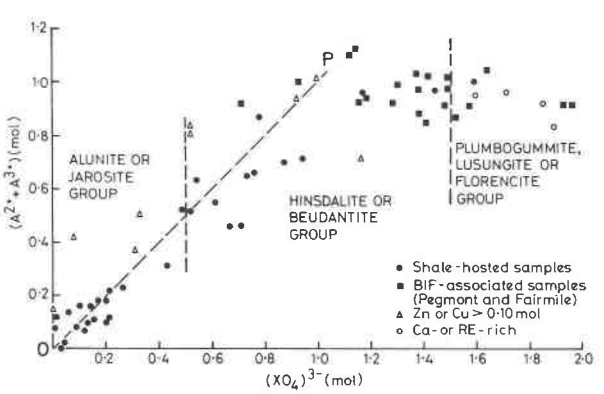

Fig. 7. Divalent and trivalent cations in A sites relative to trivalent anions in XO_4 sites in samples from northwest Queensland. Line 0–*P* represents perfect substitution $[A^{2+} + (XO_4)^{3-}]$ for $[A^+ + (XO_4)^{2-}]$ as in alunite → hinsdalite.

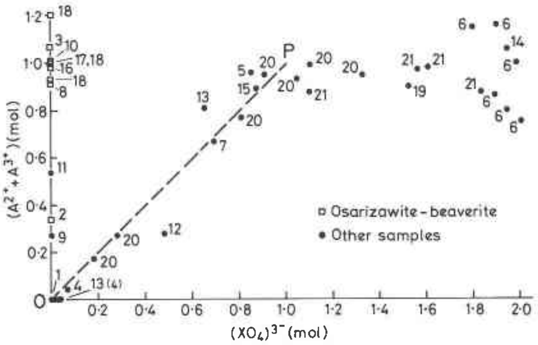

Fig. 8. A-site occupancy relative to (XO_4)-site occupancy for selected alunite-jarosite family minerals [from literature values, recalculated to $2(XO_4)$]. Source of data: 1, Brophy et al. (1962); 2, Cortelezzi (1977); 3, Kasymov (1958); 4, Keller et al. (1967); 5, Livingstone and Cogger (1966); 6, McKie (1962); 7, Melnikov et al. (1969); 8, Morris (1962); 9, Ossaka et al. (1982); 10, Paar et al. (1980); 11, Palache et al. (1951, p. 569); 12, Schmetzer et al. (1980); 13, Simpson (1938; 1948, p. 63; 1951, p. 661); 14, Slansky (1977); 15, Smith et al. (1953); 16, Taguchi (1961); 17, Taguchi et al. (1972); 18, Van Tassel (1958); 19, Walenta et al. (1982); 20, Wise (1975); and 21, Scott (unpub. data).

Only one of the northwest Queensland analyses that plot significantly (>0.1 mol) above the 1:1 $(A^{2+} + A^{3+})$ vs. $(XO_4)^{3-}$ line, 0–P, is not Zn- or Cu-rich (Fig. 7). (This sample, 50612 Fairmile, is the most sulfate-rich of the Fairmile and Pegmont samples but was not analyzed for Zn or Cu.) Osarizawaite and beaverite, with extensive Cu and Zn substitution in the B sites, plot well above the 1:1 trend (Fig. 8), but occupancy of the A sites of these samples averages only 92%. Therefore, their electrostatic neutrality is apparently maintained by a combination of vacancies in A sites and substitution of divalent Cu and Zn in B sites, as in the beaverite-plumbojarosite system (Jambor and Dutrizac, 1983). In the northwest Queensland samples, however, these two mechanisms are superimposed upon the substitution of $[A^{2+} + (XO_4)^{3-}]$ for $[A^+ + (XO_4)^{2-}]$ within the alunite, jarosite, hinsdalite, and beudantite groups.

Analyses plotting well below the 1:1 line in Figure 7 reflect partial occupation of A sites as a significant mechanism for maintaining electrostatic neutrality, although not quite as significant as in schlossmacherite (point 12, Fig. 8). Many of the northwest Queensland analyses, including those with $(PO_4)^{3-} > 1$ mol, have only minor Cu and Zn contents so that ionic neutrality is maintained mostly by a combination of the A-site vacancy and substitution of $[A^{2+} + (XO_4)^{3-}]$ for $[A^+ + (XO_4)^{2-}]$.

Further trivalent anion substitution occurs either with associated trivalent A-site substitution, $A^{2+}B_3(XO_4)^{3-}(XO_4)^{2-}(OH)_6 \rightarrow A^{3+}B_3(XO_4)_2^{3-}(OH)_6$, or, where the A site remains occupied by divalent cations, by protonation of one of the trivalent anions, $A^{2+}B_3H(XO_4)_2^{3-}(OH)_6$. In Figure 7, $(PO_4)^{3-}$ and $(PO_3OH)^{2-}$ are not distinguished, but the absence of samples with monovalent A and $(PO_4)/(SO_4) > 1$ (i.e., along the abscissa) implies that $(PO_3OH)^{2-}$ only occurs where phosphate species are the major anions in the (XO_4) sites, i.e., in the "double phosphates" of the plumbogummite, lusungite, and, possibly, florencite groups. When $(PO_4) > 1$ mol, considering about half the total phosphate as $(PO_3OH)^{2-}$ would also have the desired result of elevating the low (OH) contents of plumbogummite and lusungite samples to more normal levels, e.g., at Pegmont (50619), (OH) contents would be raised from 4.04, 4.03, and 4.13 mol (Table 3) to 5.00, 4.81, and 5.11 mol, respectively.

Where $(XO_4)^{3-} > 1$ mol, divalent cations usually occupy more than 90% of the A sites but, except for solid solution in the Ca-Sr-Ba-REE phosphates at Jolimont and Lake Moondarra and between plumbogummite and lusungite at Pegmont and Fairmile, double phosphates [i.e., $(PO_4) > 1.5$ mol] are not common in northwest Queensland (Fig. 7). As seen above, only the REE-rich members like florencite, $CeAl_3(PO_4)_2(OH)_6$, are true double phosphates with the potassic variety (54559) from Lake Moondarra (Table 3) being the sole example found during this study. Most apparent double phosphates are, in fact, members of the plumbogummite and lusungite groups where up to 50% of the $(PO_4)^{3-}$ occurs as $(PO_3OH)^{2-}$.

Minerals of the alunite-jarosite family from the shale-hosted deposits of northwest Queensland contain low phosphate contents, but those from Pegmont and Fairmile usually have $(PO_4) > 1$ mol (Fig. 7). The latter group are from a phosphatic-volcanogenic association (Taylor and Scott, 1982) where S content is relatively low, but even in that environment, double phosphates are not common.

A, B, AND (XO_4) SUBSTITUTION MODES—A SUMMARY

Substitutions of A^{2+} for A^+ and $(XO_4)^{3-}$ for $(XO_4)^{2-}$ are not independent. In Pb-Zn gossans of northwest Queensland, one Pb^{2+} cation may substitute for two K^+ cations in the A sites of alunite, $KAl_3(SO_4)_2(OH)_6$. Theoretically, such replacement could be extended to produce an Al analogue of plumbojarosite with the formula $Pb_{0.5}\square_{0.5}Al_3(SO_4)_2(OH)_6$, but in practice only 40% of the K^+ is lost in this manner, giving the A-site composition $K_{0.6}Pb_{0.2}\square_{0.2}$. In this compound, half of the resultant empty A sites (i.e., 10% of total A sites) are gradually filled as seven Pb^{2+} replace each remaining six K^+ ions, giving the A-site composition $Pb_{0.9}\square_{0.1}$. Concurrent with the A-site changes, $(PO_4)^{3-}$ replaces $(SO_4)^{2-}$ to produce $Pb_{0.9}Al_3(PO_4)(SO_4)(OH)_6$, i.e., hinsdalite, with 10% vacancies in the A sites.

Complete substitution of $(SO_4)^{2-}$ by $(PO_4)^{3-}$ gives plumbogummite, $PbAl_3H(PO_4)_2(OH)_6$. As one of the $(PO_4)^{3-}$ radicals is actually divalent $(PO_3OH)^{2-}$ (cf. Radoslovich, 1982), no further change in A sites occurs, and divalent occupancy of the A sites is often only 90%. However in florencite, $CeAl_3(PO_4)_2(OH)_6$, the trivalent charge in the A site balances the presence of a second truly trivalent phosphate radical. Therefore, it is suggested that the REE-bearing double phosphates should be distinguished from the plumbogummite group (as defined above) as the "florencite group."

Substitution of Pb^{2+} for K^+ in alunite can also be balanced by replacing up to one-third of the trivalent cations in B sites by divalent Cu and Zn, as in osarizawaite and beaverite, $Pb(Al,Fe,Cu)_3(SO_4)_2(OH)_6$. Some minerals of the alunite, jarosite, and hinsdalite groups from northwest Queensland display substantial development of this trend, plus partial occupancy of A sites (as in plumbojarosite) superimposed on the substitution of (Pb^{2+} and PO_4^{3-}) for (K^+ and SO_4^{2-}). Zn rather than Cu is the dominant divalent ion, with up to 0.62-mol (or 7.6 wt%) Zn occurring at Black Star, Mount Isa. These observations suggest that B sites can be at least partly occupied by Cu and Zn in sulfate-rich types.

Because of the method of calculation of the formulae, substitution of Fe for Al could have been artificially minimized, but even under these conditions, up to 0.84-mol Fe (58305, Mount Novit; Table 3), representing 28% occupation of B sites, occurs in shale-hosted alunites (Fig. 3). However, at Pegmont and Fairmile, where the effect of possible errors in B-site occupancy is minimal, Fe-Al intersubstitution is extensive. Therefore, the compositions of alunite-jarosites from northwest Queensland indicate that complete Fe-Al solid solution may occur in B sites, especially in the alunite, hinsdalite, plumbogummite, and lusungite groups.

RECOMMENDATIONS FOR CLASSIFICATION

To prevent a proliferation of names for minerals of the alunite-jarosite family, the following rules for classification and systematic naming are proposed for minerals with the general formula $AB_3(XO_4)_2(OH)_6$: (1) subdivision into alunite and jarosite supergroups based on dominant Al or Fe occupation of the B sites, respectively (Table 2); (2) $(XO_4)^{3-} < 0.5$ mol—alunite or jarosite groups; (3) $0.5 < (XO_4)^{3-} < 1.5$ mol—hinsdalite or beudantite groups; (4) $(XO_4)^{3-} > 1.5$ mol—(a) plumbogummite or lusungite groups if A-site occupancy is dominantly divalent or (b) florencite group if the A site is primarily occupied by trivalent cations; (5) if a minor B-site element exceeds 0.5 mol, appropriate adjectival modifiers are useful, e.g., ferrian, aluminian, zincian, etc.; and (6) inclusion of more than 0.05 mol of a minor cation in the A sites justifies an appropriate adjectival modifier to indicate the most abundant of the minor cations; however, if a modifier is applied under rule 5, rule 6 should not apply.

Because rules 1 to 4 implicitly define limits for already established minerals, the approval of the IMA Commission on New Minerals and Mineral Names has been obtained.

The concept of naming according to the proportions of endmembers, as used by McKie (1962), is not recommended because with multiple species in the A, B, and (XO_4) sites, the names of many of the alunite-jarosites reported above would involve more than six endmembers. In addition, when there are several anions in the A sites and mixed cations in the (XO_4) sites, ambiguities can arise in determining endmembers. Furthermore, because of named phosphate-sulfates and double phosphates for elements like Ca, Pb, and Sr, the description would often not be unique.

Under these rules the only minerals of the alunite-jarosite family apparently not specifically provided for, are those with only 50% of the A sites occupied, e.g., plumbojarosite, or substantial amounts of divalent ions in B sites, e.g., the osarizawaite-beaverite series. However, under rules 1 and 2, these examples are merely abnormal members of the alunite or jarosite groups; where necessary, Cu and Zn should be considered equivalent to Fe.

NEW MINERALS

No evidence for the existence of $Pb_{0.5}Al_3(SO_4)_2(OH)_6$, the Al analogue of plumbojarosite, was found, but plumbian alunite with compositions close to the ideal formula $Pb_{0.5}K_{0.5}Al_3(SO_4)_{1.5}(PO_4)_{0.5}(OH)_6$ does occur (50377, Hilton; 58244, Bernborough; Table 3), with the K and (PO_4) contents essential for electrostatic neutrality. Although minerals with 75% divalent and 25% trivalent (XO_4) site occupancy or vice versa have been defined previously (e.g., schlossmacherite and philipsbornite; points 12 and 19, Fig. 8), they were defined for their H_3O-Al-SO_4 and Pb-Al-AsO_4 contents, respectively (Schmetzer et al., 1980; Walenta et al., 1982). Plumbian alunite can be considered an intermediate between alunite, $KAl_3(SO_4)_2(OH)_6$, and hinsdalite, $PbAl_3(SO_4)(PO_4)(OH)_6$, however, its Pb, Al, and SO_4 contents could justify its classification as the Pb analogue of alunite and hence a new mineral. Similarly, the aluminian lusungite (50619, Pegmont, Table 3) has a formula close to $PbFe_3H(PO_4)_2(OH)_6$, the Fe analogue of plumbogummite, and also qualifies as a new mineral. Unfortunately, the compositions of these minerals change over very small distances, thus preventing adequate characterization of their properties.

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