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On the occurrence of niobium–tantalum and other rare-element minerals in the Meldon aplite, Devonshire

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ABSTRACT. A number of rare-element minerals from the Meldon aplite are described, e.g. native arsenic and antimony, arsenolite, stibiconite, and various niobium–tantalum minerals. Some of the minerals are new to the occurrence and some may be new to the British Isles. Particular attention has been paid to the Nb–Ta minerals and a large number of electron-microprobe analyses for manganocolumbite, manganotantalite, and microlite are presented. Examination of numerous, minute crystals and fragments of columbite–tantalite revealed a complex oscillatory zoning, indicating a slow rate of cooling and perhaps incomplete miscibility of the isomorphous columbite–tantalite series.

THE Meldon aplite, near Okehampton, Devon (grid ref. SX 573 927), is a highly mineralized dyke in places up to 20–25 m wide and some 3 km long emplaced in the metamorphic aureole north-west of the main Dartmoor granite (Worth, 1920). This lithium- and fluorine-rich aplite, together with associated pegmatite veinlets and lenses, is of considerable geochemical interest from its unique assemblage of rare accessory minerals found only in a few complex lithium pegmatites.

In recent years Chaudhry and Howie (1969, 1970, 1973, 1976) have given admirable accounts of the Meldon axinite, topaz, Li–Al micas and lithian tourmaline. Chaudhry (1971) has described the feldspars, and in addition, Chaudhry and Mahmoud (1979) have presented a statistical study of the major minerals occurring in the aplite.

After Dearman and Claringbull's description of bavenite in 1960, Kingsbury (1966) recorded a remarkable list of minerals new to Meldon, amongst them, amblygonite, montebrasite, spodumene, pollucite, and a series of beryllium minerals such as beryl, chrysoberyl, beryllonite, eudidymite, milarite, and exceptionally rare rhodizite.

The present paper is based on new material collected from the Meldon aplite by one of us (OvK) in 1980, and deals with a re-examination of columbite–tantalite (von Knorring, 1951) and associated microlite, observed in the aplite and pegmatite veinlets. In the course of the study some of the rarer minerals described by Kingsbury have been checked and their association noted. In addition, native arsenic and antimony with arsenolite, stibiconite, löllingite, fersmite, herderite, and autunite, believed to be new to the occurrence, are also described.

The aplite. The full extent of the Meldon aplite is not known, as exposures are poor, but Worth (1920) traced the main dyke for over 3 km and located also other, smaller parallel aplitic veins to the south-east of the main occurrence. The aplite is trending NE–SW, dips at approximately 50° SE and may form a much larger intrusion at depth. It is, on the whole, a light coloured, fine-grained (0.15–0.30 mm) rock containing numerous inclusions of the surrounding calcareous shales which are extensively metasomatized at the contacts to fine-grained flinty skarns (diopside, tremolite, axinite, quartz, etc.).

Commonly, distinct streakiness is seen in the aplite due to accumulation of coarser minerals, such as apatite, tourmaline, lithian mica, or fluorite, especially close and often parallel to the contacts with the country rock. The main mineral constituents of the aplite are albite, potash-feldspar, lithian-micas, and quartz in varying amounts with accessory elbaite tourmaline, topaz, apatite, fluorite, and occasionally petalite. There are also smaller amounts of amblygonite–montebrasite, various beryllium minerals, columbite–tantalite and microlite.

Chaudhry (1971) and Chaudhry and Howie (1973) distinguished three types of aplite based on colour and mineralogy as follows:

(*a*) 'Blue aplite.' A bluish, marginally chilled facies which approaches albitite in composition and consists of albite and quartz with minor amounts of apatite, tourmaline, orthoclase, and colourless lithian mica.

(*b*) 'White aplite.' This rock carries albite, quartz, and colourless to light-pink lithian mica in order of abundance.

(*c*) 'Brown aplite.' This is a coarser, metasomatized variety with larger amounts of pink to brown lithian mica and more quartz than the other types and may also contain additional elbaite, topaz, fluorite, and apatite.

The pegmatite veins. In his account of the Meldon aplite Worth (1920) mentioned that the aplite 'is traversed by veins of coarser material in which the normal minerals with the exception of topaz are repeated in larger form, some feldspars measuring over an inch in length'. McLintock (1923) noted a number of pegmatitic veins with varying mineralogical composition, among them veins carrying the rare petalite, and greisen-type veins rich in lithian mica and fluorite, and veinlets with axinite, prehnite, and fluorite.

Chaudhry and Howie (1973) recognized two major types of pegmatitic veins. In one type the veins are composed of orthoclase, quartz, Li–Al micas, albite and elbaite, whilst in the second, micaceous type, the minerals are orthoclase, Li–Al mica, quartz, topaz, and petalite. The most common veins, especially in the old aplite quarry, are those described by Chaudhry and Howie (1973), although the two types may overlap in their mineralogical composition, e.g. petalite is frequently seen with elbaite. In addition, narrow veinlets composed of microcline, albite, axinite, prehnite, fluorite, and calcite, etc., were observed elsewhere in the aplite.

In the following, some of the major pegmatitic minerals and accessories noted during the present study will be described in some detail.

Orthoclase is one of the major constituents in the pegmatitic veins forming distinct, often twinned crystals up to 5 cm long and 1 cm wide, concentrated in the central parts of the veins. It is interesting to note that orthoclase is exceptionally rare in granite pegmatites where the principle potassium feldspar is commonly microcline. Of a large number of African lithium pegmatites examined in the past (OvK), the mineral has been noted in only two deposits, in a particular petalite pegmatite within the Erongo aureole, near Karibib, Namibia, where orthoclase was intimately intergrown with petalite, and in the exotic Manjaka rubellite–kunzite–rhodizite pegmatite in the Sahatany Valley of Madagascar.

The K-feldspar in the narrow, axinite-bearing veins, however, appears to be microcline. Chaudhry (1971) reports low obliquity values from the K-feldspar and suggests that this is due to slow cooling at low temperature. From a geochemical point of view, the feldspars show significant concentrations of Rb and Cs (Chaudhry, 1971) as compared with pegmatitic feldspars from a non-lithian environment in southern Norway (Taylor *et al.*, 1960).

Albite is the main mineral in the aplitic phase and only a small amount of the mineral was noted in the examined rubellite-rich veinlets. However, the content of albite is considerably higher in the narrow axinite-bearing veinlets.

Rare accessory minerals. Native arsenic was observed in the form of brownish incrustations and silvery impregnation on quartz within small cavities lined with lithian mica flakes, closely associated with arsenolite, pink and green elbaite, and orthoclase. The associated aplite is rich in fluorite, yellow-green tourmaline, and blue manganoan apatite.

Although native arsenic is of rare occurrence in pegmatites it has been recorded from Dolcoath, near Camborne, together with cobalt and bismuth minerals (Greg and Lettsom, 1858). It has also been previously noted as an exsolution product in allemontite in the lithium pegmatites at Varuträsk, northern Sweden (Quensel, 1956), and at the Odd West pegmatite, south-eastern Manitoba, Canada (Černý and Harris, 1973).

An electron probe examination of the Meldon arsenic showed arsenian antimony in discrete crystals with hexagonal outlines, set in the arsenic matrix (fig. 1). A small amount of löllingite was also noted with this association.

Stibiconite was noted as a yellowish incrustation, filling small cavities.

Beryllonite occurs in whitish grains some 2 mm across and is mostly enclosed in elbaite with some lepidolite and quartz. It is fibrous and silky in appearance and is closely associated with pollucite, orthoclase, petalite, and lepidolite.

Herderite, forming an intergrowth with beryllonite, was noted in the aplitic matrix close to a pegmatite veinlet.

Amblygonite and montebrasite have been recorded from Meldon by Kingsbury (1966) and identified and discussed by Moss *et al.* (1969). During the present investigation the minerals were noted in three distinct pegmatitic veins. Amblygonite, the fluorine-rich end-member, is confined to the petalite- and rubellite-rich veins, whereas montebrasite is less widespread and is associated with grey lithian mica and orthoclase, and with microcline- and green tourmaline-bearing veinlets. Both minerals occur in centimetre-sized, irregular grains of grey colour, commonly replacing other minerals. The minerals are difficult to see and distinguish, with the naked eye, from the ubiquitous feldspar which they resemble. Amblygonite, like orthoclase, is of rare occurrence in lithium pegmatites; montebrasite is more usually found.

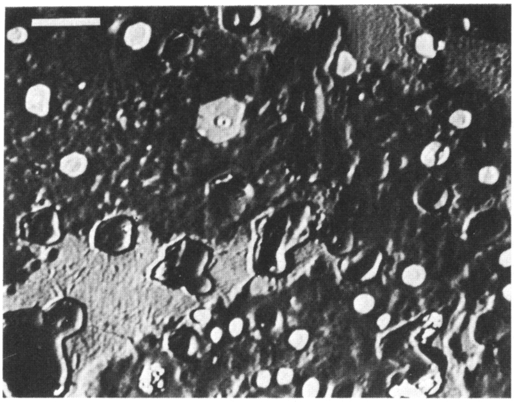
Autunite was observed in discrete greenish-yellow platy crystals in small cavities.

Beryl is occasionally noted in veinlets rich in cleavelandite and a grey lithian mica. It is of irregular habit, bluish and glassy in appearance, up to 2 cm across and is often accompanied by montebrasite.

Pollucite has been observed in the petalite-rich pegmatitic pods. The fresh saccharoidal mineral is seen in triangular outlines about 1 cm across bordering on orthoclase, quartz, and lepidolite; it is considerably lighter in colour than orthoclase. An altered, nodular form of pollucite which is in part replaced by a clay mineral, fluorite, quartz, and lithian mica is of a more common occurrence forming aggregates up to 2 cm across.

The niobium-tantalum minerals. Columbite-tantalite, manganotantalite, and microlite are characteristic accessory minerals of lithium pegmatites (von Knorring, 1970, 1974; Steiger and von Knorring, 1975; and von Knorring and Fadipe 1981).

A small amount of columbite was observed in the aplite and pegmatitic veinlets at Meldon (von Knorring, 1951). At that time, however, only a semi-quantitative spectrographic analysis was made, showing a preponderance of Nb over Ta. When examining the present specimens, a larger



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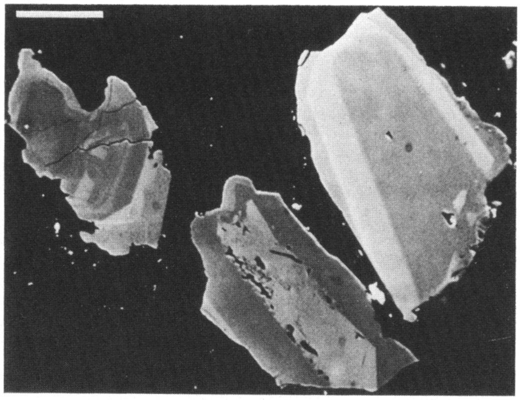

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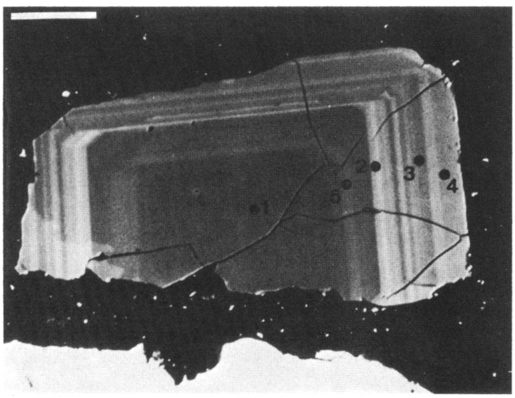

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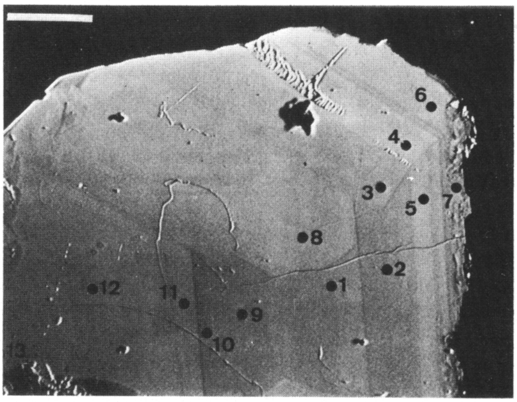

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FIGS. 1-4. Backscattered electron images of minerals from the Meldon aplite. FIG. 1. Hexagonal to globular antimony (white to pale grey) in a matrix of native arsenic and arsenic oxides. Scale bar 10 μm. FIG. 2. Strongly zoned columbite-tantalite grains. Scale bar 40 μm. FIG. 3. Oscillatory zonation in a mangano columbite-tantalite grain. Positions of analysis points are indicated, numbers correspond with analyses in Table Ib. Scale bar 30 μm. FIG. 4. Zoned mangano-columbite grain from a pegmatite veinlet. Positions of analysis points are indicated. Numbers correspond with analyses in Table II. Scale bar 60 μm.

Table I. Chemical composition of columbite-tantalite minerals from Meldon Aplite

	Table Ia	Table Ib 1	2	3	4	5
Nb_2O_5	15.49 - 62.48	53.99	29.75	42.80	43.02	41.85
Ta_2O_5	16.31 - 66.91	25.94	52.92	36.92	38.57	36.24
WO_3	0.75 - 3.68	1.21	1.81	1.73	1.41	1.73
TiO_2	0.01 - 0.63	0.43	0.45	0.43	0.35	0.43
SnO_2	0.00 - 0.04	0.00	0.00	0.00	0.00	0.00
MnO	10.82 - 17.26	17.07	10.82	15.80	15.99	15.78
FeO	0.02 - 4.93	1.41	4.93	1.61	0.02	3.67
Total		100.05	100.68	98.87	99.36	99.70
		structural formula on the basis of 6 oxygens				
		1.527	0.950	1.291	1.296	1.256
		0.441	1.016	0.662	0.699	0.654
		0.020	0.033	0.030	0.024	0.030
		0.020	0.024	0.022	0.017	0.022
		0.000	0.000	0.000	0.000	0.000
		0.905	0.647	0.893	0.878	0.887
		0.074	0.291	0.090	0.011	0.204

Table Ia: maximum and minimum values of the major elements in 64 analyses of columbite-tantalite grains.

Table Ib: chemical analyses of an oscillatory zoned mangano columbite-tantalite grain from the Meldon Aplite (see Fig. 3).

All analyses quoted in this paper were made using a wavelength dispersive Jeol JXA-50A microprobe (accelerating voltage 20kV beam current 30 nanoamps. Pure oxides were used as standards for Ti and Sn, rhodonite as Mn standard, pure metals for the remaining elements).

Table II. Chemical Composition of Mangano-Columbite from Pegmatitic Veinlets in Meldon Aplite

	13	12	11	10	9	8	1	2	3	4	5	6	7
Nb_2O_5	44.21	43.42	40.39	44.57	42.24	34.41	34.28	37.98	37.03	32.35	34.18	33.75	35.43
Ta_2O_5	37.56	38.40	42.06	38.17	39.98	48.06	46.40	43.43	46.06	50.62	48.57	49.14	46.77
WO_3	1.79	1.71	1.08	1.24	1.57	2.16	2.45	1.37	1.40	1.33	1.23	1.22	0.95
TiO_2	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
SnO_2	0.02	0.02	0.02	0.02	0.02	0.04	0.04	0.03	0.03	0.03	0.04	0.04	0.04
MnO	15.51	15.98	16.04	15.77	15.86	14.90	16.29	16.17	15.74	15.52	15.47	15.46	15.93
FeO	0.13	0.13	0.17	0.11	0.12	0.16	0.20	0.17	0.20	0.18	0.14	0.08	0.13
Total	99.24	99.68	99.78	99.90	99.81	99.75	99.68	99.12	100.48	100.05	99.65	99.71	99.27
structural formula to 6 oxygens													
Nb	1.330	1.306	1.233	1.332	1.277	1.085	1.078	1.178	1.144	1.028	1.079	1.068	1.114
Ta	0.680	0.694	0.772	0.686	0.727	0.911	0.878	0.811	0.856	0.968	0.923	0.935	0.885
W	0.031	0.029	0.019	0.021	0.027	0.039	0.044	0.024	0.025	0.024	0.022	0.022	0.017
Ti	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Sn	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
Mn	0.874	0.900	0.917	0.883	0.898	0.880	0.960	0.941	0.911	0.924	0.915	0.916	0.938
Fe	0.007	0.007	0.004	0.006	0.007	0.009	0.012	0.010	0.011	0.011	0.008	0.005	0.007

See fig. 4 for positions of analyses

TABLE III. Partial analyses of microlite grains from the Meldon Aplite

	1	2	3	4	5	6	7
Nb_2O_5	2.77	1.98	1.14	7.83	1.44	1.56	5.52
Ta_2O_5	77.73	78.52	79.76	72.29	80.82	66.48	73.90
TiO_2	0.00	0.02	0.04	0.00	0.00	0.00	0.00
MnO	0.01	0.05	0.06	0.01	0.10	0.04	0.04
FeO	0.04	0.05	0.04	0.03	0.00	0.00	0.00
CaO	8.80	8.66	9.06	7.19	8.27	10.01	9.34
Na_2O	7.08	7.02	6.90	5.13	6.79	2.24	7.00
Total	96.43	96.30	97.00	92.48	97.42	80.33	95.80

variety of minute columbite–tantalite and numerous, octahedral multicoloured crystals of microlite were seen. The columbite consists of perfectly formed rectangular, or pointed prismatic crystals from 0.02 to 1.0 mm long. The colour is generally black, but translucent brown and reddish varieties were occasionally encountered, particularly in the petalite-lepidolite-bearing veinlets and with purple apatite.

Electron microprobe examination of numerous crystals and fragments of columbite–tantalite revealed a complex oscillatory zonation (figs. 2–4). In the back-scattered electron images, atomic number contrast shows the relatively Ta-rich zones in lighter shades of grey than the Nb-rich zones. Analysis has revealed that the Nb and Ta content of the various zones is highly variable, but that there is a progressive enrichment in Ta from core to margin (Tables I and II). In the manganocolumbites the overall Nb_2O_5 content varied from 62.5 to 15.5 wt. %, with a concomitant variation of 16.3 to 66.9 % in Ta_2O_5. The largest single variation in discrete grains was 62.5 % Nb_2O_5 in the centre and 30.7 % at the margin; correspondingly the Ta_2O_5 varied from 16.9 to 51.3 %.

Barsanov *et al.* (1971) have observed similar oscillatory zoning in columbite–tantalite and relate this phenomenon to incomplete miscibility of the isomorphous columbite–tantalite series, resulting in a rhythmic distribution of zones with a variable Nb : Ta ratio. Manganotantalites from a number of African lithium pegmatites sometimes display a simple macrozoning, visible to the naked eye. In this case a light-coloured Ta-rich marginal zone is seen, indicating enrichment of Ta relative to Nb during the last stage of pegmatitic crystallization (von Knorring and Fadipe, 1981). The difference between Ta in the core and rim may be as much as 30 %, close to the figure observed in the Meldon specimens.

Microlite. A characteristic feature for many lithium pegmatites is their tantalum mineralization. A closer examination of the Meldon hand specimens and heavy mineral concentrates showed indeed, apart from columbite–tantalite, microlite in minute yellow, grey, and brownish octahedral crystals. In hand specimens they are seen confined to the pegmatitic veinlets rich in lithian mica, elbaite, and fluorite, but microlite was also noted in the aplite and in the narrow axinite-bearing veinlets.

The largest crystals observed were only some 0.8 mm across, partially intergrown with elbaite, lithian mica, and quartz. Because of the small size, multiplicity of colours, and dispersed nature, microlite may be completely camouflaged in the host matrix and therefore difficult to detect macroscopically. The chemistry of microlite is highly variable but in most cases the Ta is high, usually over 70 % Ta_2O_5 (Eid and von Knorring, 1976). Primary microlite is susceptible to alteration processes and may be re-worked to secondary microlite or other Nb–Ta minerals. These features are seen in the Meldon microlite; in one instance a larger microlite grain was partially replaced by the rare mineral fersmite.

Microprobe examination of a number of microlite fragments (Table III) showed a high but variable Ta content. Marginal alteration rims and replacement veinlets are usually poorer in Ta which is considered to be replaced by uranium.

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