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Rare earths in the aeschynite–priorite series. The status of lyndochite[1]

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Summary. Data on the contents of individual rare earths are summarized for 24 members of the aeschynite–priorite series and 3 lyndochites. It is shown that the data are in accord with the assignment of lyndochite to the aeschynite–priorite series. The available X-ray data are not definitive, but indicate at least strong similarity of lyndochite to aeschynite; the classification of lyndochite as a variety of euxenite is excluded.

LYNDOCHITE was first described from Canada by Ellsworth (1927), who considered it, on the basis of morphological data, to be probably a variety of euxenite, unusually high in Nb, Th, and Ca, and low in U; this interpretation has been accepted in most reference works. Butler (1957) determined the individual rare earths in the type material and found that chemically lyndochite resembled aeschynite rather than euxenite. In 1962, Gorzhevskaya and Sidorenko described a new occurrence of lyndochite from China, giving a chemical analysis, including determination of the individual rare earths; their material was crystalline (non-metamict) and gave the X-ray pattern of aeschynite. Recently Horne and Butler (1965), evidently unaware of the work on lyndochite from China, described a 'second occurrence' of lyndochite from Kenya, including a new chemical analysis, determination of the individual rare earths, and X-ray study of the lyndochites from Canada and Kenya. They concluded that the X-ray powder patterns of these two lyndochites were very similar to one another (the sample from Canada contained a little euxenite), but failed to match those of any other similarly heated metamict titanoniobate, thus supporting the view that lyndochite should be classed as a distinct species. They further compared the

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TABLE 1. Atomic percentages of rare earths (excluding Yt) in aeschynite–priorite series. Σ = La + Ce + Pr

No.	1	2	3	4	4*a*	5	6	7	8	9	10	11	12	13
La	—	—	—	—	0·3	0·9	2·4	4·2	7·0	2·9	3·2	2·2	3·8	4·3
Ce	—	—	—	1·1	2·8	3·2	13·3	15·8	19·8	21·7	27·4	21·9	26·4	31·2
Pr	—	—	0·8	1·1	0·9	1·1	6·9	4·7	—	5·0	3·4	13·1	8·7	7·4
Nd	—	9·5	8·9	4·4	4·0	6·7	23·1	21·3	23·3	39·5	35·1	40·7	25·8	20·1
Sm	10·2	13·0	8·6	10·6	5·7	7·5	17·2	9·8	10·0	9·9	8·8	14·4	14·2	7·5
Eu	—	—	—	—	0·9	—	2·0	—	—	0·4	—	1·5	0·8	0·4
Gd	16·2	15·7	10·5	30·6	10·6	14·6	12·7	10·5	11·6	8·6	10·4	3·0	11·2	7·3
Tb	8·0	3·4	2·8	1·0	2·8	4·9	2·4	3·2	—	1·2	1·0	0·5	1·5	1·3
Dy	32·9	19·4	20·1	29·8	20·3	30·4	10·4	12·4	13·7	4·7	5·5	1·4	5·1	6·8
Ho	6·0	3·6	4·1	0·2	5·8	6·2	1·4	2·9	—	—	—	0·3	0·5	1·5
Er	19·7	13·6	16·4	9·7	21·9	12·4	5·1	5·8	7·2	2·2	2·0	0·5	1·2	5·8
Tm	0·9	—	—	5·8	5·1	1·5	0·7	—	—	—	0·8	0·1	—	0·9
Yb	6·1	19·8	25·1	5·6	15·8	9·7	2·0	9·4	7·4	3·5	2·1	0·3	0·6	4·7
Lu	—	2·0	2·7	0·1	3·1	0·9	0·4	—	—	0·4	0·3	0·1	0·2	0·8
Yt/(Yt+Ln) %	48·9	65·5	67·2	39·7	74·6	54·5	—	46·9	53·8	37·1	28·8	8·8	24·1	30·0
Σ	0	0	0·8	2·2	4·0	5·2	22·6	24·7	26·8	29·6	34·0	37·2	38·9	42·9
La to Nd	0	9·5	9·7	6·6	8·0	11·9	45·7	46·0	50·1	69·1	69·1	77·9	64·7	63·0
Sm to Ho	73·3	55·1	46·1	72·2	46·1	63·6	46·1	38·8	35·3	24·8	25·7	21·1	33·3	24·8
Er to Lu	26·7	35·4	44·2	21·2	45·9	24·5	8·2	15·2	14·6	6·1	5·2	1·0	2·0	12·2
La/Nd	—	—	—	—	0·08	0·13	0·10	0·20	0·30	0·07	0·09	0·05	0·15	0·21

No.	14	15	16	17	18	19	20	21	22	23	24	25	26
La	7·2	8·1	12·6	14·0	14·7	14·7	17·1	17·9	20·2	15·7	21·8	11·6	19·0
Ce	36·9	45·1	41·3	45·0	43·8	47·6	46·7	46·0	47·1	51·5	49·7	58·3	53·5
Pr	10·7	5·4	7·6	8·6	9·3	7·3	7·4	8·9	6·2	7·0	4·2	7·8	7·5
Nd	29·0	24·9	22·0	26·1	25·5	22·3	20·7	17·6	24·5	16·2	19·4	13·0	18·4
Sm	6·7	4·9	4·7	3·6	3·0	3·0	2·6	2·8	1·0	2·2	1·6	1·9	0·2
Eu	0·6	—	0·6	0·5	0·4	0·1	0·3	0·3	0·1	0·2	—	0·2	—
Gd	3·5	3·2	3·4	0·2	1·2	1·7	1·7	2·4	0·4	2·8	3·3	2·6	0·7
Tb	0·4	0·8*	0·5	0·2	0·2	0·2	0·3	0·3	—	0·4	—	0·2	—
Dy	2·2	2·7	2·9	1·1	1·1	1·6	1·2	2·0	0·4	1·9	—	2·1	0·4
Ho	0·3	0·5	0·5	0·1	0·3	0·4	0·2	0·5	—	0·4	—	0·4	0·1
Er	0·8	2·8	1·9	0·5	0·3	0·8	0·7	0·5	0·1	0·6	—	0·9	0·2
Tm	0·1	0·2	0·3	—	—	—	0·1	—	—	0·1	—	0·1	—
Yb	1·3	1·4	1·5	0·1	0·2	0·3	0·9	0·8	—	0·8	—	0·8	—
Lu	0·3		0·2	—	—	—	0·1	—	—	0·2	—	0·1	—
Yt/(Yt+Ln) %	19·6	12·4*	14·1	5·9	4·4	14·1	7·2	4·2	—	14·2	1·9†	—	9·9
Σ	54·8	58·6	61·5	67·6	67·8	69·6	71·2	72·8	73·5	74·2	75·7	77·7	80·0
La–Nd	83·8	83·5	83·5	93·7	93·3	91·9	91·9	90·4	98·0	90·4	95·1	90·7	98·4
Sm–Ho	13·7	12·1	12·6	5·7	6·2	7·0	6·3	8·3	1·9	7·9	4·9	7·4	1·4
Er–Lu	2·5	4·4	3·9	0·6	0·5	1·1	1·8	1·3	0·1	1·7	—	1·9	0·2
La/Nd	0·25	0·33	0·57	0·54	0·58	0·66	0·83	1·02	0·83	0·97	1·12	0·89	1·03

* Yt + Tb cited; calculated assuming Yt/Tb = 10.

† Yt + Dy + Ho calculated as Yt.

KEY TO TABLE I

1 and 5. Priorite from albitized granites, eastern Siberia, Kostin et al. (1960)

2 and 3. Blomstrandine from granite pegmatites, Norway: 2, from Kabuland, 3, from Rasvag; Hongslo and Langmyrh (1960)

4. Blomstrandine from granite pegmatite, Aktau massif, western Uzbekistan; Khamrabaev and Azimov (1964); accuracy stated to be ±50 per cent.

4*a*. Blomstrandine from pegmatite of alkalic granite, 'European USSR'; Luncs (1965)

6. Aeschynite from alkali syenite pegmatite, Tatarka, Yenisei Range; Semenov and Barinskii (1958).

7. Aeschynite from syenite pegmatite, Siberia; Semenov (1963, p. 82)

8. Aeschynite from pegmatitic albitized nepheline mariupolite, Balyktygkhan massif, eastern Tuva; Pavlenko, Vainshtein, and Turanskaya (1959)

9. Lyndochite from pegmatites and quartz veins in gneisses, Kenya; Horne and Butler (1965)

10. Lyndochite from granite pegmatite, Lyndoch Township, Ontario; Butler (1957)

11. Aeschynite from hydrothermalite of granosyenite, Mongolia; Semenov (1963, p. 82)

12. Tantal-aeschynite from granitic pegmatite, Siberia; Kornetova, Aleksandrov, and Kazakova (1963)

13. Yttrium-aeschynite from Ilmen Mts., Urals; Makarochkin, Es'kova, and Gonibesova (1959)

14. Sinicite, from granitic pegmatites, Ulashan, Inner Mongolia; Semenov (1963, p. 82)

15. Aeschynite from greisen; Podol'skii et al. (1965)

16. Thoro-aeschynite, from microcline veinlets in the contact zone of biotite-syenite and gneiss, Vishnevye Mts., Urals; Es'kova, Zhabin, and Mukhitdinov (1964) (calculated taking Yt_2O_3 9·0; given as 8·0–10·0 with the statement that it was obtained by difference, although the sum with Yt_2O_3 9·0 is 90·9 per cent!)

17. Aeschynite from arfvedsonite–calcite–quartz veins, Vishnevye Mts., Urals; Zhabin, Aleksandrov, Kazakova, and Feklichev (1962)

18. Niobo-aeschynite from Vishnevye Mts., Urals; Zhabin, Mukhitdinov, and Kazakova (1960)

19. Aeschynite from alkali syenite pegmatite, Ilmen Mts., Urals; Semenov and Barinskii (1958)

20 to 22 Aeschynites from Vishnevye Mts., Urals; 20, from nepheline–feldspar pegmatitic veins; 21, from microcline veins at the contact of the intrusive; 22, anisotropic, non-metamict, from albitite in fenite; Es'kova, Zhabin, and Mukhitdinov (1964)

23. Aeschynite from alkalic syenite pegmatite, Vishnevye Mts.; Semenov and Barinskii (1958)

24. Lyndochite from China, associated with xenotime, clinozoisite, albite-oligoclase, and phlogopite; Gorzhevskaya and Sidorenko (1962)

25. Aeschynite from Ilmen Mts., Urals; Makarochkin, Es'kova, and Gonibesova (1959)

26. Alumo-aeschynite from quartz veins in fenites, Vishnevye Mts., Urals; Es'kova, Zhabin, and Mukhitdinov (1964)

Seven additional analyses of priorite (labelled euxenite) from metasomatic albite-syenite, Aksug massif, Tuva, given by Pavlenko *et al.* (1959), have been omitted because the light lanthanides were not determined; these fall between samples 1 and 2 on fig. 2.

distribution of the rare earths in these two lyndochites with those given in the literature for three aeschynites; lyndochites have much higher contents of yttrium than these three aeschynites.

Their discussion, however, takes account of only a small part (5 of 27 analyses) of the data available on the aeschynite–priorite series. This is one of the few series known in which the composition ranges from cerium-dominant (aeschynite) to yttrium-dominant (priorite).[1] Furthermore, Horne and Butler do not discuss the relation of the composition of the rare earths to the geological environment from which the samples came. The importance of the geological setting has been repeatedly discussed and was recently summarized (Fleischer, 1965). It seems desirable, therefore, to present a summary of the data for the aeschynite–priorite series and for lyndochite.

Chemistry of the aeschynite–priorite [aeschynite–(Ce)–aeschynite–(Yt)] series

There are many ways in which the variation of the composition of the lanthanide elements (Ln) plus yttrium can be presented. In table I, all available analyses have been recalculated so that the sum of the atomic percentages of the lanthanides, *excluding yttrium*, equals 100. This is necessary because comparison of the atomic percentages of a given lanthanide would otherwise be misleading for those analyses in which yttrium was not determined. The table also lists the atomic ratio Yt/(Yt+Ln),[2] the ratio La/Nd, and the sums (atomic percentages) of $\Sigma = (La+Ce+Pr)$, and of La to Nd, Sm to Ho, and Er to Lu, all of which have been used in various methods of classification and discussion of the rare earths. The analyses are arranged in increasing order of Σ.

Table II gives the contents of the principal elements in these minerals; no data were available for those given in table I but omitted from table II. It is readily apparent from table II that the variation in individual rare earths shown so strikingly in table I does not correlate with the total rare-earth content, with the contents of thorium, uranium, titanium, niobium, or tantalum, or with the ratio Ti/(Nb+Ta).

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[1] A. A. Levinson, A system of nomenclature for rare-earths minerals, in press, Amer. Min., proposes to rename the series as follows: aeschynite–(Ce) in place of aeschynite; aeschynite–(Yt) in place of priorite; and aeschynite–(Nd) for the previously unnamed variety represented by analysis 11 of tables I and II. This nomenclature has been approved by the Commission on New Minerals and Mineral Names, I.M.A. In this paper, the names given are those used in the papers cited.

[2] This is not directly comparable with the atomic percentages given for the lanthanide elements.

TABLE II. Chemical composition of aeschynites, priorites, and lyndochites (the sample numbers correspond to those in table I)

No.	1	2	3	4	4*a*	5	9	10	11	12	13
RE_2O_3	36·55	24·90	27·71	25·60	23·84	33·20	21·35	21·05	32·36	22·66	18·78
CaO	0·39	1·7	1·0	—	2·68	—	4·4	(3)·	2·50	4·01	2·53
ThO_2	1·85	5·80	5·78	0·08	0·6	2·75	10·13	10·77	3·70	1·31	20·34
U_3O_8	1·08	4·40*	3·67†	6·25‡	9·15	0·68	2·8	0·4	—	none	2·94
TiO_2	21·77	31·71	35·20	47·67	26·51	26·10	22·0	19·1	30·10	18·22	25·71
Nb_2O_5	34·37	17·72	14·26	25·60	23·94	27·62	32·50§	42·5§	17·64	17·87	24·23
Ta_2O_5	0·41	4·29	2·59	3·98	2·30	0·22			3·30	32·13	0·48

No.	14	15	16	17	18	20	21	24	25	26
RE_2O_3	25·57	26·95	15·94	31·93	28·17	29·30	28·91	27·03	25·16	18·89
CaO	1·20	2·24	2·73	2·60	4·82	2·73	—	5·34	2·65	7·15
ThO_2	8·29	4·96	29·56	0·75	2·52	13·06	12·35	3·75	17·42	1·83
U_3O_8	5·98‖	4·87	0·95	—	—	0·06	—	0·08‖	—	—
TiO_2	24·96	20·09	29·55	22·53	18·73	26·65	23·18	17·10	23·79	10·76
Nb_2O_5	20·85	27·62	16·15	38·70	41·41	23·59	29·60	35·90	25·35	45·48
Ta_2O_5	2·75	6·20	0·55	—	—	0·26	1·93	none	0·94	—

* UO_2 4·32, UO_3 0·08. † UO_2 1·27, UO_3 2·40. ‡ UO_2. § ratio Nb/Ta about 10. ‖ UO_3

It is desirable to plot the analyses in table I in some way that shows serial variation. Commonly the percentages of the individual lanthanides are plotted against ionic radii or atomic number, but it is difficult to see relationships in 27 such plots. Trial of many functions has shown that plots of Σ (= La+Ce+Pr) against the ratio Yt/(Yt+Ln) or against the ratio La/Nd give a spread of points that shows marked grouping of samples according to their geological environment. One might argue that this is a consequence of the separation of the rare-earth elements during geological processes; for the present purpose the graphs are used simply as empirical representations of serial variation.

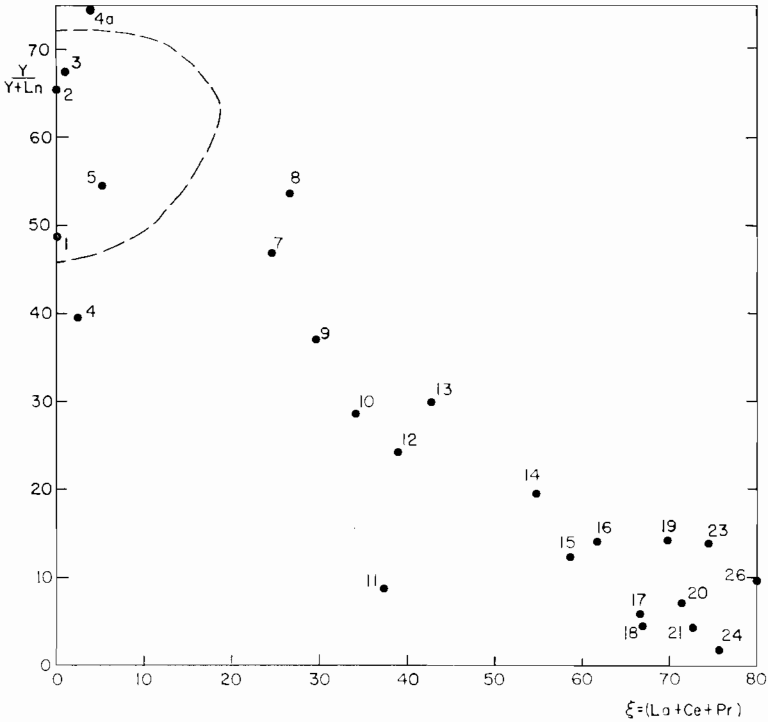

FIG. 1. Plot of Σ (= La+Ce+Pr) *vs.* the ratio 100Yt/(Yt+Ln). The dotted line encloses the field into which nearly all analyses of euxenites (34) fall.

Fig. 1 is a plot of Σ (= La+Ce+Pr) against the ratio 100Yt/(Yt+Ln). Nearly the whole series from yttrium-free to light-lanthanide-free is represented. As might be expected, the minerals from granitic pegmatites are enriched in yttrium and those from alkalic rocks are enriched in the light lanthanides. However, some minerals from granitic pegmatites, including lyndochites nos. 9 and 10, contain more of the light lanthanides, and some of the minerals from contact zones of alkalic intrusives contain more yttrium than might have been expected.

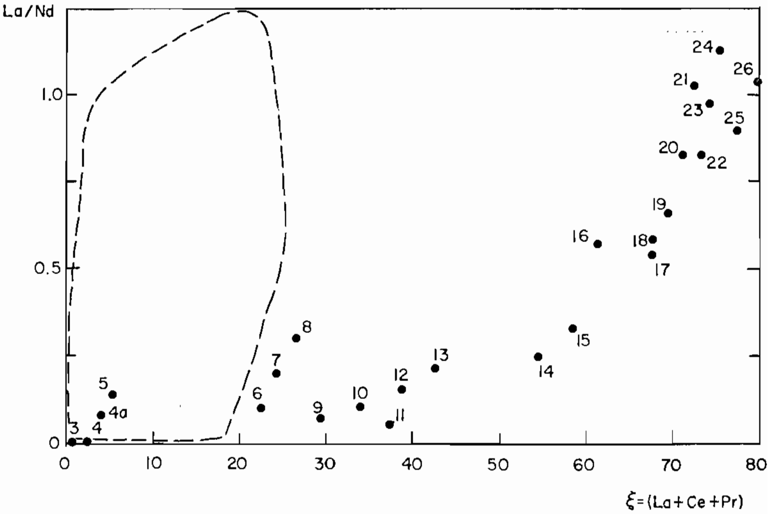

FIG. 2. Plot of Σ (= La+Ce+Pr) *vs.* the ratio La/Nd. The dotted line encloses the field into which nearly all analyses of euxenites (39) fall.

Fig. 2 is a plot of Σ against the ratio La/Nd. This also shows a serial variation that correlates fairly well with the variation in mode of occurrence. In view of the difficulty of the analyses, the deviations from a single curve seem not too great. It will be noted that in both plots the three lyndochites (nos. 9, 10, 24) are in no way unique members of the series. Horne and Butler commented on the similarity of the Tatarka aeschynite (no. 6) to lyndochites nos. 9 and 10; also similar to nos. 9 and 10 are aeschynites nos. 7–8, and 12–13, but lyndochite no. 24, very similar to lyndochites nos. 9 and 10 in major element content (table II), differs greatly from them in the rare-earth composition.

Horne and Butler pointed out that the composition of the rare earths in euxenite is quite different from that in aeschynite and lyndochite, but similar to that of priorite. The areas into which all but a very few of the available analyses of euxenite fall have been outlined by dotted lines in figs. 1 and 2. Nearly all of these are from granitic pegmatites.

In summary, then, the composition of the rare earths in the three lyndochites analysed is in accord with the classification of lyndochite as a variety of aeschynite. Its classification on morphological grounds as a variety of euxenite can now be excluded definitely on the basis of both composition and X-ray data. There remains the problem of the X-ray data by Horne and Butler (1965) on lyndochites nos. 9 and 10, which they believe to indicate that lyndochite is a distinct species. It should be recalled that these samples were metamict and were identified by the X-ray patterns obtained after heating the samples. Horne and Butler warned of the dangers inherent in this procedure and this warning is emphasized by recent studies of the thermal behaviour of euxenite and aeschynite (Komkov and co-workers, 1962, 1963*a*, *b*; Seifert and Beck, 1965). They found that natural aeschynites, as well as synthesized compounds of type $LnNbTiO_6$ with the structure of aeschynite, were converted to dimorphous compounds having the structure of euxenite at temperatures of 700–1100° when heated in air, or at somewhat lower temperatures when heated under hydrothermal conditions. The results differ in detail, but agree in general; further complications are caused by the formation of cubic oxide phases.

The X-ray patterns given by Horne and Butler for the two lyndochites are very similar to those in the literature for aeschynite, which also show considerable differences among themselves. It seems possible that these differences might be caused by differences in the degree of metamict decomposition of the samples before heating or by differences in the conditions under which the samples were heated to restore their crystallinity. A definite decision will require further careful study, but I believe that lyndochite should be tentatively classified as a thorian neodymian aeschynite.

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