VARIATION OF SOME RARE EARTHS IN ALLANITE

JUDITH W. FRONDEL, Harvard University, Cambridge, Mass.

ABSTRACT

Using x-ray spectrographic techniques, 64 specimens of allanite from 51 localities were analyzed for Ca, Ce, La, Nd and Y. Ce is the chief rare-earth in allanite, and makes up usually at least one-half of the total rare-earths. Ce almost always predominates over La in atomic per cent, with the ratio of Ce to La generally between 2:1 and 3:1. Nd, always present, is a significant member of the Ce group, and occurs in amounts greater than those of Y. Y is lacking in many allanites and rarely occurs in large amounts. Allanite—like minerals having Y$_2$O$_3$ greater than 5.0 per cent may represent separate species, since x-ray diffraction patterns of such specimens are different from those of allanite. Except for these specimens of anomalous rare-earth composition, all heated specimens, in which order was restored, gave the allanite x-ray diffraction pattern. Heating increased the refractive indices of allanite.

SPECIMENS STUDIED

In investigating the rare-earth variation in allanites, a total of 64 specimens were studied. All of these specimens, with the exception of two which came from the mineral collection of Bryn Mawr College, were selected from the Harvard University mineral collection, and represent 51 well-known allanite localities all over the world (Table I).

X-RAY SPECTROGRAPHIC ANALYSIS

All specimens were analyzed with a Norelco broad-focus x-ray spectrograph, using a flow-counter with methane-argon gas mixture, a helium path and tungsten excitation. The unit was run at 50 KV and 45 ma. A chart-recorder was used and quantitative measurements were based on peak to background ratios. The samples were prepared for analysis in the following manner: A one-gram, powdered sample of each specimen was mixed with 15 grams of anhydrous borax [Na$_2$B$_4$O$_7$] glass and fused in a platinum crucible at 1000° C. for 3 hours. The fusion was cast in a steel ring-mold held at 450° C., and then was annealed at 250° C. for 12 hours. This dilution method of sample preparation was used to minimize matrix absorption effects. The case buttons provide permanent and easily storable samples.

1 Mineralogical Contribution No. 413, Harvard University.
<table>
<thead>
<tr>
<th>Specimen No.</th>
<th>Catalogue No.</th>
<th>Locality</th>
<th>Brief Description of Hand Specimen</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1)</td>
<td>Chaffee, C-1</td>
<td>Chaffee Co., Colorado</td>
<td>Prisms in red feldspar. Pegmatite veins in quartz monzonite.</td>
</tr>
<tr>
<td>(2)</td>
<td>Everard, SA-1</td>
<td>Everard Range, South Australia</td>
<td>Black vitreous large crystal, oxidized surface. Pegmatite veins in granite.</td>
</tr>
<tr>
<td>(3)</td>
<td>Nutarami, G-1</td>
<td>Nutarmint, Avigeit, West Greenland</td>
<td>Black, vitreous, in feldspar in granite rocks containing hornblende zircon crystals and allanite.</td>
</tr>
<tr>
<td>(4)</td>
<td>Pacoima, P-1</td>
<td>Pacoima Canyon, Los Angeles Co., California</td>
<td>Large platy crystals, dull greasy. Pegmatite intrusions in gabbro.</td>
</tr>
<tr>
<td>(5)</td>
<td>37111</td>
<td>Monroe, Orange Co., New York</td>
<td>Large crystal fragment, vitreous black. In gneiss.</td>
</tr>
<tr>
<td>(6)</td>
<td>37121</td>
<td>Buoe, near Arendal, Norway</td>
<td>Nearly pure mass, vitreous black, some feldspar and veinlike black greasy inclusions. Pegmatite veins in gneiss.</td>
</tr>
<tr>
<td>(7)</td>
<td>37144</td>
<td>Baringer Hill, Llano Co., Texas</td>
<td>Rounded pure mass, 5&quot; X 4&quot; X 1&quot;, vitreous black. Pegmatite in granite.</td>
</tr>
<tr>
<td>(8)</td>
<td>37211</td>
<td>Skepholm, Sweden</td>
<td>Small crystals in granitic veins and gneisses.</td>
</tr>
<tr>
<td>(9)</td>
<td>37231</td>
<td>Miask, Urals</td>
<td>Fragment of large platy (?) crystals. Intergrown in miascite granite.</td>
</tr>
<tr>
<td>(10)</td>
<td>80512</td>
<td>Avigeit, Greenland</td>
<td>Vitreous black small crystals in granite rocks containing hornblende, zircon crystals and allanite.</td>
</tr>
<tr>
<td>(11)</td>
<td>84652</td>
<td>Arendal, Norway</td>
<td>Large conchooidal vitreous black fragment. Pegmatite veins in gneiss.</td>
</tr>
<tr>
<td>(13)</td>
<td>86089</td>
<td>Arendal, Norway</td>
<td>Large platy crystal. Pegmatite veins in gneiss.</td>
</tr>
<tr>
<td>(14)</td>
<td>86090</td>
<td>Modum, Norway</td>
<td>Columnar, in feldspar. In granite pegmatite.</td>
</tr>
<tr>
<td>(16)</td>
<td>91985</td>
<td>Vaarala, Finland</td>
<td>Black vitreous, in feldspar. Pegmatite in granite gneiss.</td>
</tr>
<tr>
<td>(17)</td>
<td>92933</td>
<td>Woburn, Massachusetts</td>
<td>Black vitreous, massive, with pink feldspar. Pegmatite veins in gneiss.</td>
</tr>
<tr>
<td>(18)</td>
<td>96020</td>
<td>Hybla, Ontario, Canada</td>
<td>Black vitreous fragment. In granite pegmatite.</td>
</tr>
<tr>
<td>(19)</td>
<td>97399</td>
<td>Lake Baikal, Russia</td>
<td>Platy crystals, oxidized surface. In mica-pegmatite.</td>
</tr>
<tr>
<td>(20)</td>
<td>98985</td>
<td>Near Eau Clair, Nipissing District, Ontario, Canada</td>
<td>Large platy crystal. In granite pegmatite.</td>
</tr>
</tbody>
</table>
### Table 1—(continued)

<table>
<thead>
<tr>
<th>Specimen No.</th>
<th>Catalogue No.</th>
<th>Locality</th>
<th>Brief Description of Hand Specimen</th>
</tr>
</thead>
<tbody>
<tr>
<td>(21)</td>
<td>99264</td>
<td>Franklin, Sussex Co., New Jersey</td>
<td>Platy crystals in green feldspar, with feldspar and magnetite.</td>
</tr>
<tr>
<td>(22)</td>
<td>99811</td>
<td>Consolidated Spar Mine, Kingman, Arizona</td>
<td>Black, vitreous, columnar. In granite pegmatite.</td>
</tr>
<tr>
<td>(23)</td>
<td>102385</td>
<td>Lac à Baude, Normand Twp., Laviolette Co., Quebec, Canada</td>
<td>Small fragment, vitreous, black. In granite pegmatite.</td>
</tr>
<tr>
<td>(24)</td>
<td>102406</td>
<td>Ytterby, Washolm, Sweden</td>
<td>Columnar, vitreous, black. In granitic feldspar-rich differentiation of a black amphibolite.</td>
</tr>
<tr>
<td>(25)</td>
<td>102412</td>
<td>Lescombe property, Monmouth Twp., Haliburton Co., Ontario, Canada</td>
<td>From phlogopite mine, dull greasy type, very high Th (determined by qualitative run in x-ray spectrograph). From syenite pegmatite (?).</td>
</tr>
<tr>
<td>(26)</td>
<td>102414</td>
<td>White Face Mountain, Adirondacks, New York</td>
<td>Large fresh vitreous crystal (in anorthosite?)</td>
</tr>
<tr>
<td>(27)</td>
<td>102415</td>
<td>Midongy de l'Ouest, Madagascar</td>
<td>Fragment, vitreous black shiny. K-rich pegmatites in granites and schists.</td>
</tr>
<tr>
<td>(28)</td>
<td>102422</td>
<td>Boksput, South Africa</td>
<td>Fragment of large crystal, Greasy. Dull; high Th (determined by qualitative run in x-ray spectrograph). Pegmatite in granite-gneiss.</td>
</tr>
<tr>
<td>(29)</td>
<td>102423</td>
<td>Fryso 4, Iveland, Norway</td>
<td>Piece of large crystal, black vitreous. In granite pegmatite.</td>
</tr>
<tr>
<td>(30)</td>
<td>102429</td>
<td>Tvedestrand, Norway</td>
<td>Broken crystal. In granite pegmatite.</td>
</tr>
<tr>
<td>(31)</td>
<td>102449</td>
<td>Huddersfield Twp., V. 22, Pontiac Co., Quebec, Canada</td>
<td>Large bent platy crystals, with phlogopite, dull, greasy. In apatite-mica-pegmatite.</td>
</tr>
<tr>
<td>(33)</td>
<td>102471</td>
<td>Tutuslavi, Randu, Dutria, Bihar, India</td>
<td>Large mass. Pegmatites in mica-schists and gneisses.</td>
</tr>
<tr>
<td>(34)</td>
<td>102480</td>
<td>1½ miles northeast of Jamestown, Colorado</td>
<td>Large irregular mass. In granite pegmatite.</td>
</tr>
<tr>
<td>(35)</td>
<td>102499</td>
<td>Gole Mine, Murchison Twp., Nipissing Dist., Ontario, IV, 14, Canada</td>
<td>Large altered columnar crystals. In granite pegmatite.</td>
</tr>
<tr>
<td>(36)</td>
<td>102505</td>
<td>Vaarala, Finland</td>
<td>Thick tabular crystals, sub-vitreous, black. Pegmatite in granite gneiss.</td>
</tr>
<tr>
<td>(37)</td>
<td>102506</td>
<td>Dickens Twp., Nipissing Dist., Ontario, Canada (from between Barry's Bay and Malawalka?)</td>
<td>Platy crystals, with feldspar, mica, quartz. In granite pegmatite.</td>
</tr>
<tr>
<td>(38)</td>
<td>102508</td>
<td>La Marche Mine, Kensington Twp., Gatenaau Co., Quebec IV, 45, Canada</td>
<td>With spheine, calcite, feldspar, molybdenite. In apatite-mica-pegmatite.</td>
</tr>
<tr>
<td>Specimen No.</td>
<td>Catalogue No.</td>
<td>Locality</td>
<td>Brief Description of Hand Specimen</td>
</tr>
<tr>
<td>-------------</td>
<td>---------------</td>
<td>---------</td>
<td>----------------------------------</td>
</tr>
<tr>
<td>(39)</td>
<td>102512</td>
<td>Lyndoch Twp., V. 23, Renfrew Co., Ontario, Canada</td>
<td>Platy crystals in granite pegmatite.</td>
</tr>
<tr>
<td>(40)</td>
<td>102516</td>
<td>La Marche Mine No. 2, Kensington Twp., Gateneau Co., Quebec IV, 45, Canada</td>
<td>Thin platy crystals (1 cm x 1 mm) and massive, with molybdenite, biotite, calcite. In apatite-mica-pegmatite.</td>
</tr>
<tr>
<td>(41)</td>
<td>102517</td>
<td>Lyndoch Twp., Renfrew Co., Ontario, Canada</td>
<td>Platy crystals in quartz, feldspar. In granite pegmatite.</td>
</tr>
<tr>
<td>(42)</td>
<td>102522</td>
<td>Macdonald Mine, Hybla, Ontario, Canada</td>
<td>Very large rough mass, shiny, subvitreous. In granite pegmatite.</td>
</tr>
<tr>
<td>(44)</td>
<td>102534</td>
<td>Cardiff Twp., Ontario, Canada</td>
<td>Platy or lenticular small crystals in granite pegmatite.</td>
</tr>
<tr>
<td>(45)</td>
<td>102565</td>
<td>Argenteuil Co., Quebec, Canada &quot;Old Sigelino property near Calumet,&quot; &quot;Pleinville II, 27 (?)&quot;</td>
<td>With ilmenite and micro-monazite. In granite pegmatite.</td>
</tr>
<tr>
<td>(46)</td>
<td>102573</td>
<td>Clapham Twp., IV, 28, Pontiac Co., Quebec, Canada</td>
<td>Large platy mass, vitreous, greasy, feldspar, mica, with diopside, scapolite, phlogopite. Vein in pyroxenite (?)</td>
</tr>
<tr>
<td>(48)</td>
<td>102599</td>
<td>Macdonald Mine, Hybla, Ontario, Canada</td>
<td>1-foot mass, dull greasy, pyrite inclusions. In granite pegmatite.</td>
</tr>
<tr>
<td>(49)</td>
<td>102600</td>
<td>Cardiff Twp., Haliburton Co., Ontario XX, 7, Canada</td>
<td>Large platy crystals, with tangerite (?), red feldspar, calcite. In granite pegmatite.</td>
</tr>
<tr>
<td>(51)</td>
<td>103886</td>
<td>Woburn, Massachusetts (A.C. Lane spec.)</td>
<td>In feldspar, with epidote. Pegmatite in gneiss.</td>
</tr>
<tr>
<td>(52)</td>
<td>105680</td>
<td>Aquarius Range, Mohave Co., Arizona</td>
<td>Black dull greasy fragment of large crystal, Ga, Pb, Be in spec. In granite pegmatite.</td>
</tr>
<tr>
<td>(53)</td>
<td>106172</td>
<td>Woodland Park, Colorado</td>
<td>Large crystal fragment, vitreous black. In granite pegmatite.</td>
</tr>
<tr>
<td>(54)</td>
<td>106180</td>
<td>Trout Creek Dist., Chaffee Co., Colorado</td>
<td>Columnar crystal, dull, greasy. Pegmatite in quartz monzonite.</td>
</tr>
<tr>
<td>(55)</td>
<td>106184</td>
<td>Amherst Co., Virginia</td>
<td>Large crystal, dull greasy, oxidized on surface. In altered feldspar in pegmatite.</td>
</tr>
</tbody>
</table>
Method of analysis. First, a standard “allanite mix” (Table II) was made from oxides weighed out in the proper molar proportions. The “average allanite” composition was computed from 61 representative analyses reported in the literature. The small content of alkalies, water, and of other accessory constituents was disregarded. Rare-earth standards were prepared by adding to this bulk mixture Ce₂O₃, La₂O₃, Nd₂O₃ and Y₂O₃ in varying molecular proportions and totaling 19.15 weight per cent. Each sample then was mixed with borax glass to give a weight ratio of 1:15 and was fused and cast. These buttons not only served as preliminary standards, but were used continually throughout the investigation to check the stability and response of the x-ray spectrograph.

The standard buttons were run in the x-ray spectrograph for Ce, La, Nd and Y, and the 55 specimens, prepared as buttons, also were run for these elements. Four of the 55 specimens were selected for conventional wet-method chemical analysis. Complete analyses were made on two specimens, one lacking Y and the other exceptionally high in Y. Partial analyses were made on two specimens representing the average Ce and La occurrence in allanite (Table III).
In the standard allanite buttons the per cent of CaO was constant, 9.96 weight per cent. Results for CaO in the two wet-method chemical analyses (42 and 35, Table III) were taken and, together with the standard buttons, formed the basis for a standard graph for Ca. The 55 allanite buttons were run then also for Ca. Comparisons of wet-method chemical analyses and x-ray fluorescence analyses on the same sample are made in Table IV. The results of all the x-ray spectrographic analyses are tabulated in Table V. Results were recalculated on the basis of the total rare-earths determined—100 per cent (Table VI).

**Table II. Standard “Allanite Mix”**

<table>
<thead>
<tr>
<th>Oxide</th>
<th>Weight Per Cent</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>31.81</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>15.84</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>5.88</td>
</tr>
<tr>
<td>FeO</td>
<td>9.00</td>
</tr>
<tr>
<td>MnO</td>
<td>1.52</td>
</tr>
<tr>
<td>CaO</td>
<td>9.96</td>
</tr>
<tr>
<td>MgO</td>
<td>0.67</td>
</tr>
<tr>
<td>ThO₂</td>
<td>1.18</td>
</tr>
<tr>
<td>UO₂</td>
<td>0.30</td>
</tr>
<tr>
<td>TiO₂</td>
<td>0.77</td>
</tr>
<tr>
<td>H₂O</td>
<td>3.92</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>80.85</strong></td>
</tr>
</tbody>
</table>

Added to this standard mix were Ce₂O₃, La₂O₃, Nd₂O₃ and Y₂O₃, in varying molecular proportions and totaling 19.15 weight per cent.

**Analysis on powders.** The four specimens analyzed by wet chemical methods were run also as powders in the x-ray spectrograph. Care was taken to have the samples uniformly ground and packed in the sample-holder. Graphs were plotted from the results and a comparison was made with the results on the same specimens run as buttons. The amount of error in the analyses on powder, due probably to element interference, was from less than 1 to 2.5 per cent for Y, from 6 to 20 per cent for Nd, from 3 to 5 per cent for Ce, and from less than 1 to 8 per cent for La. Though at first thought this may seem considerable, yet it would appear practical to run powders instead of borax fusions whenever only general orders of magnitude are needed or when time and apparatus for making the fusions are not available. With this consideration, nine specimens, for which borax fusions had not been made, were analyzed as powders only (Tables VII and VIII).
TABLE III. ANALYSES MADE BY CONVENTIONAL WET METHOD

A. Partial Analyses

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>(9)</th>
<th>(27)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ce, La oxides</td>
<td>19.84</td>
<td>20.34</td>
</tr>
<tr>
<td>Y, Nd oxides</td>
<td>0.21</td>
<td>0.32</td>
</tr>
<tr>
<td>H₂O (+)</td>
<td>3.45</td>
<td>2.79</td>
</tr>
<tr>
<td>H₂O (−)</td>
<td>nil</td>
<td>nil</td>
</tr>
</tbody>
</table>

B. Complete Analyses

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>(42)</th>
<th>(35)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>32.29</td>
<td>33.16</td>
</tr>
<tr>
<td>TiO₂</td>
<td>0.06</td>
<td>0.24</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>18.19</td>
<td>18.69</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>6.53</td>
<td>4.36</td>
</tr>
<tr>
<td>FeO</td>
<td>7.98</td>
<td>9.30</td>
</tr>
<tr>
<td>MgO</td>
<td>0.78</td>
<td>0.46</td>
</tr>
<tr>
<td>CaO+SrO</td>
<td>14.70</td>
<td>12.46</td>
</tr>
<tr>
<td>ThO₂</td>
<td>0.41</td>
<td>0.33</td>
</tr>
<tr>
<td>Σ Ce, La oxides</td>
<td>15.00</td>
<td>7.60</td>
</tr>
<tr>
<td>Σ Y, Nd oxides</td>
<td>1.49</td>
<td>7.89</td>
</tr>
<tr>
<td>MnO</td>
<td>0.30</td>
<td>2.68</td>
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<tr>
<td>P₂O₅</td>
<td>0.21</td>
<td>0.13</td>
</tr>
<tr>
<td>H₂O (+)</td>
<td>1.97</td>
<td>2.47</td>
</tr>
<tr>
<td>Na₂O</td>
<td>.005</td>
<td>.022</td>
</tr>
<tr>
<td>K₂O</td>
<td>nil</td>
<td>nil</td>
</tr>
<tr>
<td>SO₂</td>
<td>tr.</td>
<td>nil</td>
</tr>
<tr>
<td>U₂O₈</td>
<td>n.d.</td>
<td>n.d.</td>
</tr>
<tr>
<td>PbO</td>
<td>tr.</td>
<td>tr.</td>
</tr>
<tr>
<td>Total</td>
<td>99.91</td>
<td>99.79</td>
</tr>
<tr>
<td>Ce₂O₃</td>
<td>6.72</td>
<td>1.54</td>
</tr>
</tbody>
</table>

Trace elements found spectrographically: Ga, Cu, Sn, Ni, Cr, Co, Mo, Zr, V, Bi, Be and Sc.

1 Analyst, Jun Ito.

Results of x-ray spectrographic analysis. In all but two samples, (35 and 50), Ce is the predominant rare-earth. In no sample is Ce₂O₃ less than La₂O₃ and the ratio of Ce₂O₃ to La₂O₃ almost always is between 2 to 1 and 3 to 1. In all except 5 of the samples Y₂O₃ is less than Nd₂O₃. X-ray diffraction patterns1 obtained on two (35 and 50) of these exceptional speci-

1 See section on x-ray diffraction analysis.
mens indicate that they are not allanite, though their chemistry (Table III; both 35 and 50 came from the same mine) is much like that of allanite. Hence it is suggested that where $Y_2O_3$ is greater than 5.0 per cent, the material is not allanite, yet may be related closely to it.\(^2\) Though $Y_2O_3$ is not present or is less than one per cent of the total analysis in

\(^2\) The relationship or identity of these specimens with lomboardite of Neumann and Nilssen (1962) is the subject for a further investigation.

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Wet Method</th>
<th>X-Ray Spectrography</th>
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<tr>
<td></td>
<td>Oxides</td>
<td>Wt. Per Cent</td>
</tr>
<tr>
<td>(9)</td>
<td>Ce+La Oxides</td>
<td>19.84</td>
</tr>
<tr>
<td></td>
<td>Y+Nd Oxides</td>
<td>.21</td>
</tr>
<tr>
<td></td>
<td>CeO$_2$</td>
<td>not determined separately</td>
</tr>
<tr>
<td></td>
<td>La$_2$O$_3$</td>
<td>not determined separately</td>
</tr>
<tr>
<td></td>
<td>$\Sigma$RE$_2$O$_3$ determined</td>
<td>20.05</td>
</tr>
<tr>
<td>(27)</td>
<td>Ce+La Oxides</td>
<td>20.34</td>
</tr>
<tr>
<td></td>
<td>Y+Nd Oxides</td>
<td>.32</td>
</tr>
<tr>
<td></td>
<td>CeO$_2$</td>
<td>not determined separately</td>
</tr>
<tr>
<td></td>
<td>La$_2$O$_3$</td>
<td>not determined separately</td>
</tr>
<tr>
<td></td>
<td>$\Sigma$RE$_2$O$_3$ determined</td>
<td>20.60</td>
</tr>
<tr>
<td>(42)</td>
<td>Ce+La Oxides</td>
<td>15.00</td>
</tr>
<tr>
<td></td>
<td>Y+Nd Oxides</td>
<td>1.49</td>
</tr>
<tr>
<td></td>
<td>CeO$_2$</td>
<td>6.72</td>
</tr>
<tr>
<td></td>
<td>La$_2$O$_3$</td>
<td>8.28 (by difference)</td>
</tr>
<tr>
<td></td>
<td>$\Sigma$RE$_2$O$_3$ determined</td>
<td>16.49</td>
</tr>
<tr>
<td>(35)</td>
<td>Ce+La Oxides</td>
<td>7.60</td>
</tr>
<tr>
<td></td>
<td>Y+Nd Oxides</td>
<td>7.89</td>
</tr>
<tr>
<td></td>
<td>CeO$_2$</td>
<td>1.54</td>
</tr>
<tr>
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<td>La$_2$O$_3$</td>
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Table V. X-ray Spectrographic Analyses
(arranged in order of decreasing CeO₂)

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about three fourths of the specimens, no specimen was lacking in Nd, and Nd$_2$O$_3$ virtually always is more than one per cent of the total analysis.

**Table VII. Analyses Made on Powder Samples**
(arranged according to decreasing Ce$_2$O$_3$)

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**Table VIII. Per Cent of Rare Earths in RE$_2$O$_3$ Determined in Powder Samples**
(arranged according to decreasing Ce$_2$O$_3$)

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**Some Analytical Data from the Literature Contrasted with the X-ray Spectrographic Data in the Present Study**

Hasegawa (1960) presented 19 new Japanese allanite analyses made by standard chemical methods and these were compiled together with 31 analyses found in the mineralogical literature. From these data Hasegawa drew histograms showing the weight per cent of various oxides versus the frequency of their occurrence. In Fig. 1, these histograms are contrasted with some drawn up from the x-ray spectrographic data in the present investigation. Discrepancy occurs mainly with the histograms for La$_2$O$_3$ and Ce$_2$O$_3$. This is due, no doubt, to the fact that Hasegawa did not consider Nd$_2$O$_3$ separately. Very probably, this oxide is included in the per cents for La$_2$O$_3$, and the values for La$_2$O$_3$ appear greater than they actually are.

Though in the present investigation the limits of total rare-earths determined (Table V) are from 15.4 per cent to 27.4 per cent of the total analysis, allanites with as little as 2.44 per cent rare earths$^1$ or as much as

$^1$ From some data supplied by Michael Fleischer of the U. S. Geological Survey.
Consequently the frequently made suggestion that a complete series exists between epidote [general formula, \( \text{Ca}_2(\text{Fe}^{3+},\text{Al})_8\text{Si}_5\text{O}_{12}(\text{OH}) \)] and allanite [general formula, \( (\text{Ca}_{2-n},\text{Ce}_n)_{2}(\text{Fe}^{2+},\text{Fe}^{3+1-n})_2(\text{Fe}^{3+},\text{Al}_{2-n})_2\text{Si}_5\text{O}_{12}(\text{OH}) \)] appears valid.

**X-ray Diffraction Study**

X-ray diffraction studies were made on all of the specimens using a Norelco diffractometer with chart recorder. Because allanite occurs largely in a metamict state, the majority of the specimens gave no x-ray diffraction patterns on unheated samples. Some unheated samples yielded good patterns (Table IX and Fig. 2). Small pure lumps of almost one half of the 64 specimens were selected and heated in a nitrogen atmosphere at approximately 650° C. for 2 to 3 hours. A few samples were heated still longer, several for as much as three days, in an attempt to restore order to the metamict material. The specimens to be heated were selected to represent the variation of the rare earths determined. Of these
Heated samples, 14 recrystallized and gave allanite x-ray diffraction patterns. Two other specimens (35 and 50, both from the same mine) recrystallized after heating for three days, but gave x-ray diffraction patterns somewhat different from allanite. (Though the patterns of these two specimens resemble each other and may represent a new species, the verification of this suggestion is left for a separate investigation). All samples, heated and unheated, were ground to a very fine powder and run for an x-ray diffraction pattern. Subsequent regrinding of the samples was found, in general, to improve the quality of the patterns. Hence, each sample was reground and rerun until no further changes or improvement of the pattern could be obtained.

The results of this x-ray study are shown in Figures 2, 3 and 4, and Table IX. The patterns were indexed according to Ueda (1957) and measured. $b$ was calculated from (020). Since no morphological measurements were obtained, the $\beta$ angle could not be determined and, therefore $a$ and $c$ could not be calculated. No relationship was noted between the variation in rare earths and the variation in the d spacings. Moreover,

1 See footnote re: lombaardite of Neumann and Nilsen (1962).
such a relationship could be fortuitous, since variations in the rare earths probably are compensated for by variations in other elements (e.g. not only the rare earths substituting for Ca, but Al substituting for Fe$^{3+}$) in the allanite structure.

Some difference in $d$ spacings was noted between unheated and heated samples of the same specimen (Table IX and Fig. 4). Restoration of order, by heating, to the metamict allanite causes a decrease in the $d$ spacings. This observation is in agreement with the results of Ueda and Korigawa (1954) and Ueda (1957) who carried out detailed studies on the metamict state of allanite.

The degree of metamictization did not appear to be connected with any variation of the rare earths determined in the present study. It is more likely that the age of the allanite and the small amounts of U and Th generally present determine its metamict state.

**Some Optical Data**

Optical measurements were made on only a few of the specimens. In view of the many varying and compensating elements in allanite, and the variable and often large content of non-essential water, a detailed optical study seemed futile. The effect of heating on the specimens was of chief interest. In all of the specimens studied optically, the indices of refraction were increased by heating (Table X).
Fig. 2. X-ray diffraction patterns of some unheated allanites.
Fig. 3. X-ray diffraction patterns of some heated allanites.
Fig. 4. Contrast of x-ray diffraction patterns of three allanite specimens before and after heating.
## Table X. Some Optical Data

(all \(n\) values ± .003)

<table>
<thead>
<tr>
<th></th>
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<th></th>
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</thead>
<tbody>
<tr>
<td>Remarks</td>
<td>Faintly pleochroic, low birefringence</td>
<td>Low birefringence</td>
<td>Deep olive-green, transparent on thin edges</td>
<td>Dark brown to black, almost opaque except on thin edges which are olive-brown</td>
<td>Olive-brown</td>
<td>Low birefringence</td>
<td>Transparent on thin edges, olive-green, a few grains faintly anisotropic</td>
<td></td>
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<tr>
<td>Mean (n)</td>
<td>1.670</td>
<td>1.687</td>
<td>1.750</td>
<td>1.720</td>
<td>1.750 almost colorless</td>
<td>1.755 yellow-green</td>
<td>1.760 olive-green</td>
<td>1.73</td>
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<tr>
<td>(\alpha)</td>
<td>1.710 pale yellow-green</td>
<td>1.710</td>
<td>1.715</td>
<td>1.720 yellow-green</td>
<td>1.723</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td>(+) Moderate</td>
</tr>
<tr>
<td>(\beta)</td>
<td>1.713 deeper yellow-green</td>
<td>1.715</td>
<td>1.723</td>
<td>(+) Very large</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td>(-) Moderate</td>
</tr>
<tr>
<td>(\gamma)</td>
<td>1.720 yellow-green</td>
<td>1.723</td>
<td>(++) Large</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td></td>
</tr>
<tr>
<td>2(V)</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td>Isotropic</td>
<td></td>
</tr>
<tr>
<td>Remarks</td>
<td>Transparent, olive-green, fine-grained inclusions gone</td>
<td>Pleochroic, low birefringence</td>
<td>Very fine-grained, low birefringence, pleochroic</td>
<td>Pleochroic</td>
<td>Strongly pleochroic</td>
<td>Faintly pleochroic</td>
<td>Increased birefringence</td>
<td>Faintly pleochroic</td>
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<tr>
<td>Mean (n)</td>
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<td>1.740 almost colorless</td>
<td>1.745 almost colorless</td>
<td>1.745 pale yellow-green</td>
<td>1.755 light brown</td>
<td>1.765 almost colorless</td>
<td>?</td>
<td>?</td>
</tr>
<tr>
<td>(\alpha)</td>
<td>1.740 almost colorless</td>
<td>1.745 almost colorless</td>
<td>1.745 pale yellow-green</td>
<td>1.755 light brown</td>
<td>1.765 almost colorless</td>
<td>?</td>
<td>?</td>
<td></td>
</tr>
<tr>
<td>(\beta)</td>
<td>1.745 yellow-green</td>
<td>1.753 yellow-green</td>
<td>1.755 deep yellow-brown</td>
<td>1.780 orange-brown</td>
<td>1.770 pale olive-brown</td>
<td>1.776 yellow-green</td>
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<tr>
<td>(\gamma)</td>
<td>1.752 deep yellow-brown</td>
<td>1.755 olive-brown</td>
<td>1.760 olive-green</td>
<td>1.785 deep brown</td>
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<td>1.783 orange-brown</td>
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<tr>
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<td>(+) Very large</td>
<td>?</td>
<td>(-) Large</td>
<td>(-) Large</td>
<td>(-) Large</td>
<td>(+) Moderate</td>
<td>?</td>
</tr>
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</table>
RARE EARTHS IN ALLANITE

ACKNOWLEDGEMENTS

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REFERENCES


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