MONAZITE COLORED BY CARBON FROM DICKENS TOWNSHIP, NIPISSING DISTRICT, ONTARIO*

H. V. Ellsworth, *Canada Geological Survey.*

Some years ago there was considerable prospecting for feldspar and mica in the neighbourhood of Aylen Lake, and along the Parry Sound-Ottawa railway between Barrys Bay and Madawaska. Several pegmatite dikes were opened up, at least two of which produced feldspar for some time, and one dike on Aylen Lake is said to have yielded some 60 tons of muscovite. Radioactive minerals in small quantities frequently occur in this region, almost all the numerous dikes examined by the writer showing some indications of their presence. The monazite[1] described here was obtained from a dike on the claim of F. G. Armstrong of Pembroke, lot 9, conc. 13, Dickens Tp., probably on the north half of the lot. The dike strikes about N 60° E mag. along the side of a hill and is exposed for a length of 200 feet. The true width was not definitely ascertainable but it appeared to reach a maximum of 50 feet or more. To a large extent it consists of graphic intergrowth of quartz and microcline but patches of coarse crystal microcline and massive white quartz occur. Some good sized crystals of muscovite and biotite in less quantity occur, but as a rule the mica is so crumpled and cracked as to be valueless. Red garnets up to one inch in diameter are rather common in the feldspar, and a two inch crystal of molybdenite was seen. Small brilliant black crystals of a mineral resembling euxenite or samarskite are quite abundant in the feldspar in places, and occasional flat reddish brown crystals of monazite were seen. The crystal with which this paper is chiefly concerned was the largest specimen collected. It measured about three inches in one direction as indicated by the field sketch reproduced in Fig. 1. Unfortunately the mass was broken into fragments, some

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[1] Monazite is not uncommon in the pegmatites of the Precambrian Shield but is almost invariably greatly altered. This particular material was selected for study because it was the only transparent monazite ever obtained in Canada, so far as the writer is aware, and consequently because of its very slight alteration would be of unusual interest in connection with radioactive age determination. The presence of carbon was not suspected at first, the color naturally being ascribed to iron or manganese.

of which were lost by the chiselling necessary to remove it from the rock and it was not possible to reassemble it completely. Fig. 2 is a photograph showing the best fragment recovered. The peculiar crystal habit exhibited by this monazite apparently has not been recorded hitherto. The whole crystal growth appears to be very complex. The two apparently simple, more or less symmetrical, halves of the mass are themselves twinned along a diagonal line as shown by the diagonal fracture in Fig. 2, and an examination of other fragments (Fig. 4) indicates a still more complex structure due to repeated twinning and parallel growth. The wide end of the tapering mass bears well developed crystal faces and is more or less transparent for a distance of about an inch back from the faces. The remainder becomes dark colored, in places almost black, to-

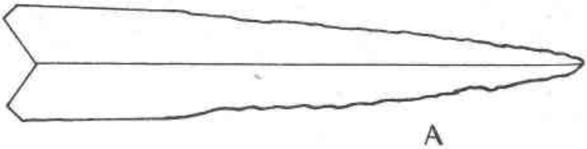

FIG. 1. Reproduction of field sketch showing outline of monazite crystal growth, natural size, as first seen embedded in pegmatite.

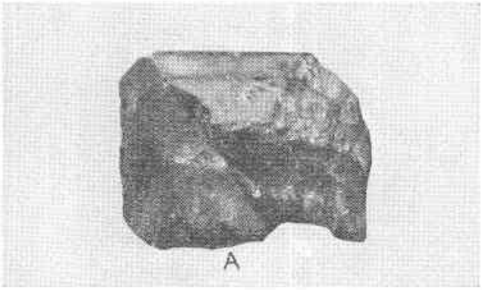

FIG. 2. Photograph showing part of monazite of Fig. 1, natural size after freeing from the rock. The front surface in the photo probably represents the top side of crystal growth seen in Fig. 1. The diagonal fracture from NW to SE seems to indicate a twinning plane between a parallel growth to the NE and a twin to the SW. The plane of fracture parallel to the surface of the paper thus appears to be common to a twin and a parallel growth. Unfortunately the part broken away from the SW front half, below the diagonal fracture, which would have given the key to the arrangement, was lost. Parallel growths are developed on the back of this mass.

ward the thin edge of the wedge-shaped mass. The dark color has been found to be due to the presence of carbon in a state of very minute dissemination. In thin sections under the microscope the dark material does not appear to be different in any way from the transparent parts, except for the presence of some very minute dust like inclusions visible under high powers. That the dark color is due to carbon is indicated by the following observations: On ignition of nearly black fragments before the blowpipe in air the mineral changes to a cream or yellow color. Crushed dark fragments packed in the bottom of a hard glass closed tube and similarly heated almost to a white heat do not lose their dark color, and no evidence of a distillate of heavy hydrocarbons condensed on the inside of the tube can be seen under the microscope, after drying out the water over sulfuric acid. A slight whitish ring of sublimate is produced which is probably lead oxide. The colors of certain minerals such as smoky quartz, purple fluorite and certain dark red feldspars, which are probably due to atomic disturbances induced by radioactive radiation, are discharged by heating to about 500° even in the absence of air, so these tests alone show that there is

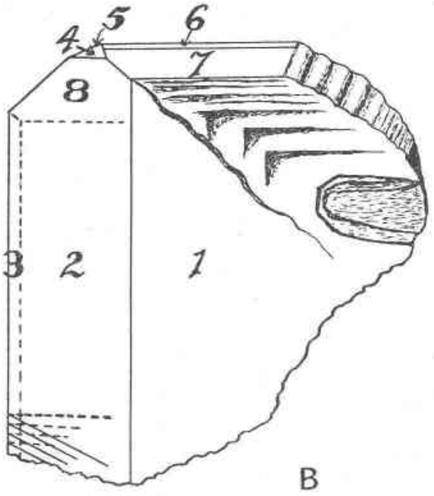

FIG. 3. Sketch showing details of crystal faces of Fig. 2, the diagonally fractured part of the mass being removed. Traces of what appears to be a good cleavage appear in the SW corner, the plane of which is at approximately 63° to 1 and the intersection on 1 normal to edges between 1, 2, and 3.

no relation between the dark color of the monazite and that of smoky quartz. The best positive evidence that carbon is present, however, is obtained on heating 5 or 10 grams of the powdered mineral with sulfuric acid. The pasty mass thus obtained is brownish black in color and when diluted with water a very distinct brownish coloration and a brownish to black scum or precipitate is obtained, which is not affected by HCl but is destroyed by heating with concentrated nitric acid, or persulfates, or by ignition. The sulfuric acid used when heated alone did not become colored in the least, and if the powdered mineral is ignited at a red heat in air before treatment with sulfuric acid no brown coloration or scum is produced. Finally several grams of the powdered mineral were ignited in a current of CO_2-free oxygen, observing all the precautions incident to a quantitative carbon determination, the emerging gases being passed through barium hydroxide solution with the result that a very distinct and unmistakable precipitate of barium carbonate was produced. This test of course does not necessarily prove that carbon as such is present, since rocks and minerals often contain CO_2 and CO as gas inclusions, occluded, or otherwise, but nevertheless it furnishes confirmatory evidence. Thus it appears that the dark color is certainly due to carbon—not hydro-

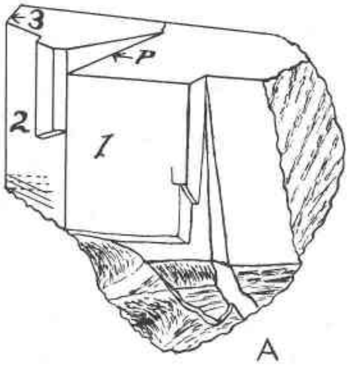

FIG. 4. Sketch showing another portion of the mass of Fig. 1, about natural size $P\wedge2=68^\circ$. Approximate polar angles $1\wedge2=40^\circ$, $2\wedge3=87^\circ$, P is a plane following the intersection of 1 and 2 along which the mass has separated into two parts. The top of the drawing represents the mass as though it were smoothly sectioned in order to give a better idea of the form; actually it is irregularly fractured in a way having no apparent crystallographic significance. A good cleavage as indicated is developed at the SW corner.

carbons—disseminated as extremely minute particles, some of which are possibly of atomic dimensions since reactions for carbon can be obtained even from the clear transparent material in thin sections of which not the slightest visible evidence of the presence of carbon can be detected, using magnifications up to 400 diameters and arc light illumination. Even selected fragments of the very finest perfectly transparent material which are normally a deep reddish brown color become almost colorless on ignition in air, exhibiting only a faint yellowish tinge. Thus there is reason to believe that the normal reddish brown color of the transparent portions is also due to carbon present as particles of such small size as to be invisible under the microscope. It might be mentioned that the alteration along cleavages and cracks shows up much more clearly after the mineral has been ignited, as an opaque whitish to yellow, or even reddish film, while the clear transparent areas remain unaffected except for the loss of color as previously noted. In spite of the marked reactions just described it is not likely that the carbon constitutes more than a few hundredths of a per cent of the dark material, at most, probably less. No quantitative determination was attempted on the sample analyzed which consisted of the best selected clear reddish brown material and contained much less carbon than the dark parts.

The association of carbon and hydrocarbons with radioactive minerals is not unusual in the pegmatites of Ontario and Quebec studied by the writer. The present case, however, is interesting in that we have here a transparent mineral which is definitely proved to be colored by carbon.

CRYSTALLOGRAPHY

A small crystal one quarter inch in diameter (Figs. 5, 6) was measured on the two-circle goniometer in various orientations. A projection using the large front face (Fig. 5) as base came out in fairly good agreement with published data (Goldschmidt), indicating the presence of the faces: $c(001)$, $a(\bar{1}00)$, $m(\bar{1}\bar{1}0)$, $X(\bar{1}01)$, $V(\bar{1}\bar{1}1)$ and $*(0\bar{3}2)$. However, as may be seen from the photograph the faces of even this small crystal are too distorted and imperfect to permit accurate measurement. The large crystal (Figs. 1, 2, 3) appears to be similar in habit to the small one. Measurements with a contact goniometer on the faces shown in the drawing (Fig. 3) gave the following approximate values for polar angles: $1 \wedge 2 = 57°$,

$2\wedge 3=87^\circ$, $2\wedge 4=39^\circ$, $1\wedge 7=47^\circ$, $1\wedge 6=63^\circ$, $5\wedge 6=57^\circ$, (5 is very narrow, rough and rounded), $7\wedge 8=92^\circ$. On the small crystal faces (001) and $(0\bar{3}2)$ apparently correspond to 1 and 7, respectively, on the large crystal, and (001) and $(\bar{1}01)$ agree with 1 and 2 to within 3 or 4 degrees, but the measurements seem to be sufficiently accurate in this case to demonstrate that this difference of 3 or 4 degrees actually exists. Both crystals are wedge shaped, the angle between 1 and the corresponding face behind being about 2 or 3 degrees.

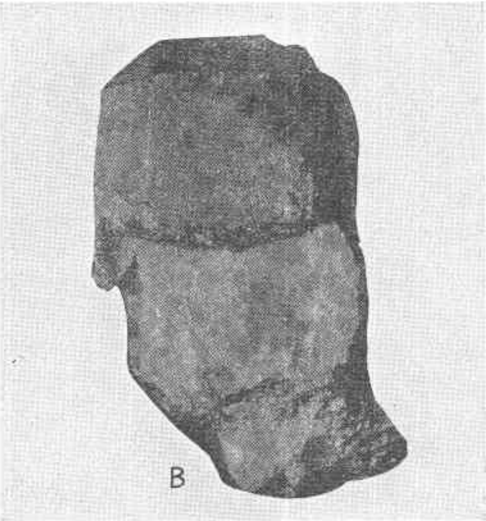

FIG. 5. Photograph of small, simple monazite crystal (same locality) measured on goniometer, actual size $\frac{1}{4}$ inch diameter. The large front face is believed to be *c*(001), face at the left (032), at NW corner *m*$(\bar{1}\bar{1}0)$ and *V*$(\bar{1}\bar{1}1)$, top *a*$(\bar{1}00)$ and *X*$(\bar{1}01)$. Lower part of photo shows mounting wax holding crystals.

The writer's thanks are due Dr. Eugene Poitevin for collaboration in attempting to work out the crystallography of this mineral.

OPTICAL PROPERTIES IN RELATION TO HOMOGENEITY

Both the transparent and dark colored portions of the crystal were examined in representative thin sections to discover, if possible, evidences of alteration or the presence of inclusions. If, as has sometimes been supposed, the thorium is present in the form of thorite as inclusions or intergrowths, the thorite should make up

9.09 per cent of the whole material (7.32 ThO_2+1.77 SiO_2), if the full amount of silica were present. Actually, however, only 1.54 per cent of silica was found by analysis. Thorite is always isotropic and should be easily detected between crossed nicols, but no indications of isotropic inclusions could be seen. A prominent cleavage or parting is well shown in all the sections as a series of roughly parallel rather closely spaced lines of cracks. Along the edges of these major fractures the clear monazite has altered to a cloudy whitish material scarcely noticeable with strong illumination but very distinctly visible when the right conditions are obtained. Just as in the case of many titano-columbate minerals examined by the writer this alteration could easily account for the silica present without assuming the presence of thorite. The mineral also shows a rather definite parting or rough cleavage across the major cleavages and in addition a still more minute system of irregular minor fractures may be seen under high powers with certain conditions of illumination. The alteration seems to vary directly with the size of the cleavage cracks or fractures, being scarcely apparent along the more minute fractures. Apart from the whitish alteration product visible chiefly along the major fractures, the only indication of a possible lack of homogeneity is a variation in the interference colors producing a mottled effect of red and blue. All the sections were apparently a little thick and the predominant color over the greater part of the section was a pale permanganate pink, probably

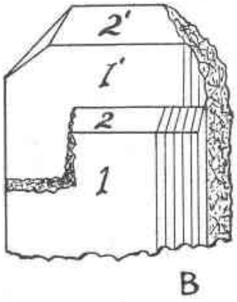

FIG. 6. Sketch of opposite side of crystal shown in Fig. 5, showing parallel growth. Faces 2 and 2' are rough but apparently parallel. Approx. polar angles $1\wedge 2$, $1'\wedge 2'=47°$. A good cleavage is developed on the R. H. side of the front individual as indicated by the parallel lines. This was measured on the two circle goniometer and found to be 104° 45' to face 1.

representing the second order, while the thinnest edges were pale yellow, succeeded higher up by the red of the first order. The permanganate pink is mottled with blue spots apparently of the first order. The blue spots may possibly represent areas in which the mineral has a lower birefringence, or they may represent lower spots where the abrasive perhaps has ground softer altered areas to a greater extent than the parts represented by permanganate pink, or these spots may merely be places where the abrasive has torn off larger flakes of the mineral than usual. Viewed obliquely under a binocular microscope the surfaces of the sections appear rather rough and pitted, lending weight to the last explanation as being the most probable. The blue areas do not appear to be related in any way to the major cleavages. It appears certain, however, that thorite is not present and that the silica can be accounted for by the alteration along the major cleavage cracks.

Summary

The mineral is reddish brown to black, transparent to opaque, anisotropic. Shows one good cleavage (?) and a parting normal to the cleavage. Lustre vitreous, fracture subconchoidal to uneven, H=5.5, Sp. Gr.=5.270. In wedge shaped complex crystal growths.

The sample for analysis was carefully selected from the best transparent material. Lead, uranium and thorium were determined on 5 and 10 gram lots. The analysis yielded results as follows:

Monazite, Dickens Township, Nipissing District, Ontario

PbO (Pb=0.31)	0.33
U_3O_8 (U=0.27)	0.32
ThO_2 (Th=6.43×0.38=2.44 U equiv.)	7.32
Ce_2O_3	22.63
$(La, Di)_2O_3$	34.63
$(Yt, Er)_2O_3$	4.66
CaO	0.35
MgO	0.02
Fe_2O_3	0.08
MnO	Not detected
Al_2O_3, etc.	<0.10
P_2O_5	27.89
SiO_2	1.54
$H_2O-110°$	0.06
$H_2O+110°$	0.34
He, etc.	Not determined
C	Traces present
	100.27

$$\frac{\text{Pb}}{\text{U}+0.38\ \text{Th}}=0.11$$

Sp. Gr. = 5.270 at 20.65°

Monazite is a particularly interesting mineral considered as a geological age indicator and in connection with the theories of radioactive disintegration because, according to most of the published analyses, it contains thorium but no uranium. Minerals in which thorium is present unaccompanied by uranium are rare, if indeed any such exist. It seems likely that in the older analyses of monazite small amounts of uranium actually present may have been overlooked or at least not determined. Thorium minerals compared with uranium minerals usually yield low age results and various theories have been proposed to account for this. At one time it was thought that possibly the lead is unstable, or that there may be another end product besides lead, but the tendency at present is to ascribe the low lead ratios of thorium minerals to loss of lead due to alteration and leaching. Holmes has advanced a plausible explanation of the discrepancies by supposing that the lead of uraninite is present in the mineral as uranate while in thorium minerals more particularly thorites and thorianites, it must be present as oxide or other compound, and that the lead uranate is less soluble and less liable to be leached away than the lead compound occurring in thorium minerals. On the other hand, lead phosphate which might be formed in monazite, is a very insoluble compound. The writer has shown that complex U, Th, titano-columbate minerals which contain appreciable silica due to alteration yield low lead ratios, while those with little or none are in accord with uraninite. It is interesting to note that almost all monazite analyses show a silica content of one per cent or more, and it has been supposed that the silica could be accounted for along with the Th content by assuming thorite to be present as inclusions or intergrowths. But, as thin sections of the Dickens township monazite containing 7 per cent ThO_2 failed to yield any evidence of the presence of thorite, it seems more reasonable to believe that the silica found in monazite is due to replacement, involving loss of lead just as appears to be the case with the titano-columbate minerals. There is also, of course, in all cases where the lead ratio is too low the possibility that leaching may have removed not only lead but also radium and mesothorium, or other members of the radioelements. If this were so it could be detected by determining radium and mesothorium by the emanation method and calculating whether they were present in amounts required by the normal equilibrium ratios to correspond with the amount of U and Th found by analysis.