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Europium-rich dark monazite – a potential new ore mineral for Alaska, USA?

Introduction

Mineralogical drainage surveys carried out between 2004 and 2008 in the Taylor Mountains quadrangle, Alaska, USA, have identified three areas containing a distinctive dark monazite in panned stream sediment concentrate samples. Similar dark monazite of unknown or limited extent has been observed in sediments from elsewhere in Alaska (12 occurrences), France, Taiwan, Zaire and Russia (Rosenblum & Mosier 1983). Systematic mineral identification of panned concentrate samples by R. Tripp led to the discovery of dark gray to black, pellet-like, crystalline monazite grains collected from streams draining sedimentary rocks in the eastern quarter of the quadrangle (Fig. 1). A few sites at the western edge of the quadrangle also contained dark monazite in concentrate samples.



Figure 1a: Location of the Taylor Mountains quadrangle, Alaska, USA with adjoining portions of Lake Clark and Dillingham quadrangles;

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The Taylor Mountains quadrangle is largely underlain by Cretaceous flysch sequences that overlap several older terranes (Miller et al. 2004). Much of the flysch in the quadrangle had previously been assigned to the Upper Cretaceous Kuskokwim Group; however, newer mapping may indicate that the flysch includes multiple, genetically unrelated units. Late Cretaceous to early Tertiary igneous rocks (Sleitat Mountain) intrude the flysch and this is where the central study area (TA559) of this report is located, as shown on Figure 1. Well-developed hornfels consisting of metasiltstone, metasediment, and metashale surrounds Sleitat Mountain. Cordierite, corundum and sillimanite were collectively found in concentrate from streams draining the hornfels around Sleitat Mountain and several other igneous intrusions. In contrast, andalusite and dark monazite were observed in almost every concentrate sample, suggestive of low-grade regional metamorphism of the flysch in the eastern quarter of the quadrangle.

Sample Collection and Laboratory Analysis Methods

Heavy-mineral concentrate samples collected primarily from first-order and second-order streams (as determined from the 1:63,360-scale quadrangle maps) consisted of numerous grab samples along a 9 to 30 m length of an active stream channel. The grab samples were composited into a single 3 to 4 kg sample and screened in the field with a 2.0 mm (10-mesh) stainless steel screen to remove the coarse material. The <2.0 mm fraction then was panned to remove lower-density material such as quartz, feldspar, organic material, and clay-size particles.

The concentrate samples were air-dried in the laboratory and then sieved through a 0.5 mm (35-mesh) screen. The <0.5 mm fraction was further separated using bromoform heavy liquid (2.89 g/cm³) to remove any remaining lighter minerals with specific gravities of less than 2.85 g/cm³. A Frantz Isodynamic Magnetic Separator was used to split the processed concentrate samples into two mineral fractions: a magnetic fraction containing magnetite, ilmenite, garnet,

amphibole, pyroxene, epidote, and other high-iron silicates and a relatively non-magnetic fraction containing most ore-related minerals including both dark and yellow monazite.

The non-magnetic fraction was scanned visually under a ring light-illuminated binocular microscope using up to 56X magnification. In most cases, the monazite grains could be identified from their physical properties, but X-ray diffraction and six-step semi-quantitative emission spectrography (Grimes & Marranzino 1968) were used to confirm the identification of monazite. This non-magnetic fraction commonly contained andalusite, apatite, cordierite, epidote, muscovite, rutile, sphene, tourmaline, and zircon. Some samples also contained gold, cinnabar, and corundum.

Schist and phyllitic rock, as well as numerous monazite grains, were initially examined with a JEOL 5800LV scanning electron microscope (SEM) to identify monazite in the rock and acquire information on the distribution of inclusions and possible chemical zoning. Next, twenty-five detrital monazite grains from TA559 concentrate were analyzed using a JEOL 8900 fully-automated five spectrometer electron probe microanalyzer operated at 20 keV accelerating voltage, 150 nA current (cup), and 5 µm beam diameter. The K_α x-ray lines were analyzed for Al, Si, and P, the L_α x-ray lines for Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Dy, and Er, and the M_α x-ray lines for Pb, Th, and U. The count times (peak plus background) were 1320 seconds for Al, Si, and Ca, 40 seconds for P and 200 seconds for the remaining elements. Linear background corrections were assumed for all elements. Interferences of off peaks on the analyzed peaks were corrected using the electron microprobe software Probe for Windows by Probe Software, Inc. The distribution of selected elements within the dark monazite was determined using wavelength dispersive spectrometry (WDS) by the JEOL-8900 electron microprobe.

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Distribution of detrital dark monazite

In total, 838 heavy mineral concentrate samples were collected from streams in the Taylor Mountains quadrangle over a period of four years. In addition, 14 samples were collected in the adjacent Lake Clark and Dillingham quadrangles (Fig. 1b). Of these, 229 samples (27%) contained dark monazite in varying amounts. Most dark monazite occurs in the eastern Taylor Mountains quadrangle with a western limit along the Nushagak River south of Butch Mountain and north-northeast to the northern border of the quadrangle. Dark monazite extends into the Lime Hills quadrangle to the northeast to the Mosquito River drainage in the Lake Clark quadrangle to the east and into the Dillingham quadrangle to the south. Two smaller areas of dark monazite in stream concentrates occur along the western edge of Taylor Mountains quadrangle.

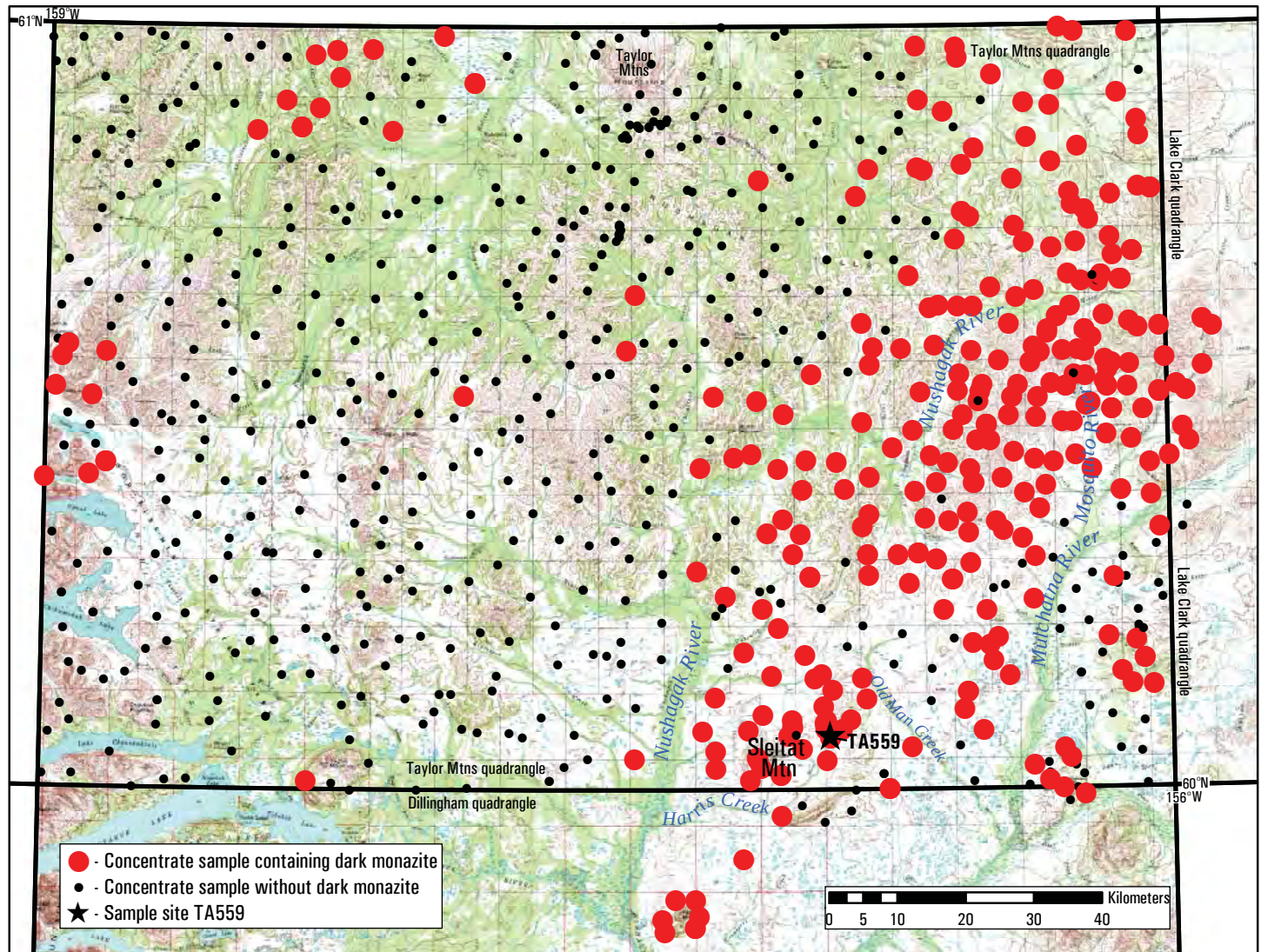


Figure 1b: Black dots indicate stream sediment concentrate locations that did not contain monazite. Red dots indicate locations where monazite was observed in the samples. The solid black star indicates site TA559 of the monazite study area in this report.

During the field season of 2008, a concerted effort was made to locate the source rocks of the dark monazite, in the southern part of the Taylor Mountains quadrangle, with particular emphasis on collecting rubble crop along the ridge extending northward from Sleitat Mountain towards Old Man Creek. Additional concentrate samples were collected from virtually every stream draining the ridge. All concentrate samples contained dark monazite and most contained gold colors and tiny flakes (Fig. 2). The likely lode source, gold-bearing quartz veins, is less than 1000 m from the sample location.

Comparison of yellow and dark monazite

The characteristics of the more common yellow monazite are in sharp contrast to dark monazite. Yellow monazite (Fig. 3) is a light rare earth element (REE) phosphate mineral, typically a cerium (Ce), lanthanum (La), yttrium (Y) phosphate [(Ce,La,Y)(PO₄)]. Lanthanum is ordinarily present in a 1:1 ratio with Ce. Thorium oxide (ThO₂) can substitute for Ce and La in amounts up to 12 wt.%; Th-free monazite is rare. Uranium (U) has been occasionally reported (Palache et al. 1951) in amounts up to approximately 1

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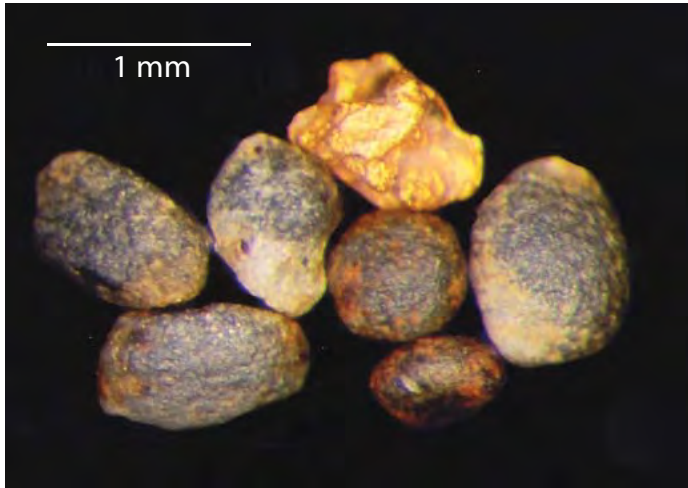


Figure 2. Photomicrograph of monazite grains together with gold. The gold from these samples is “fresh” looking with rough hackly surfaces and angular three-dimensional morphology, suggesting that the source is nearby.

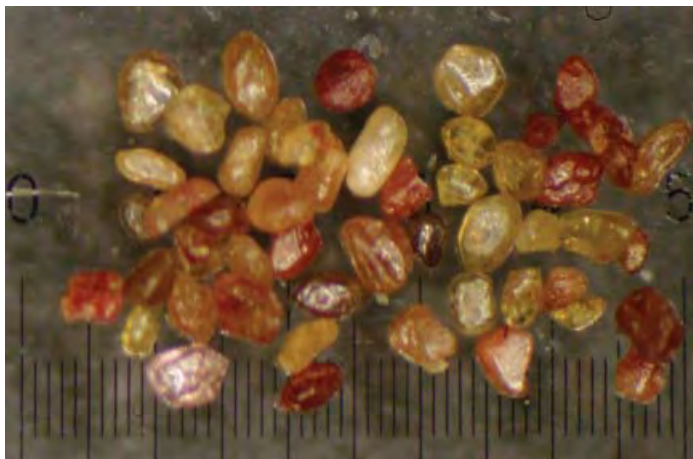


Figure 3. Photomicrograph of typical detrital “yellow” monazite grains showing color, shape, and luster. Yellow monazite colors range from yellow to reddish brown, greenish, to nearly white. Scale in photo is 0.1 mm per graduation.

wt.% U_3O_8 . The yellow variety occurs as monoclinic platelets to prismatic crystals that are commonly small, but sometimes large and coarse (Fig. 3). The physical properties of the yellow monazite include:

- one well-developed cleavage plane [100];
- well-marked parting plane [001];
- conchoidal to uneven fracture;
- hardness of 5 to 5½;
- varying specific gravity from 4.6 to 5.4 g/cm³, but mostly in the range of 5.0 to 5.2 g/cm³;
- luster can be variable, usually resinous or waxy, but may incline towards vitreous to adamantine;
- colors range from yellow to reddish brown, greenish, to nearly white;
- small crystals of yellow monazite range from sub-transparent to sub-translucent.

Yellow monazite is widely disseminated as an accessory

mineral in granitic and gneissic rocks. Monazite is of sufficient hardness to form considerable detrital deposits such as those found in gold sands of the southern Urals, Russia; in black sands of the Manajary basin, Madagascar; in sands of the Embabaan district, Swaziland, southern Africa; in tin placers of the Moolyella district, Western Australia; the west coast of Ceylon; and in the provinces of Bahia, Espirito Santo, Minas Gerais, and Rio de Janeiro, Brazil (Hedrick 2004). In the United States, considerable amounts of yellow monazite occur in the sands of Burke, Cleveland, Lincoln, McDowell, Polk and Rutherford Counties in North Carolina and the beach sands in Florida (Hedrick 2004). Since yellow monazite may contain appreciable amounts of Th and U, this variety has largely been eliminated from commercial use as a REE source because of concerns about radioactivity hazards.

Dark monazite, in contrast to yellow monazite, contains low levels of Th and U, but relatively high Eu values. Detrital dark monazite occurs in rounded to discoidal, inclusion-rich, xenoblastic forms, commonly with white sericitic “tails and rims” (Fig. 4). However, there is evidence of forms showing idioblastic features as well. Dark monazite shows

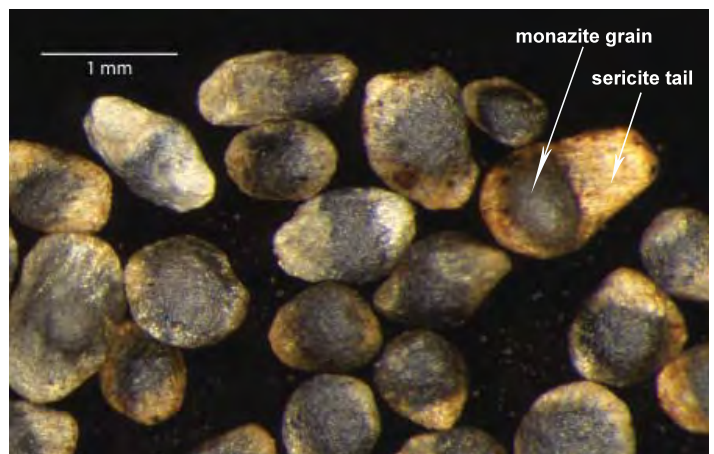


Figure 4. Photomicrograph of dark monazite grains with attached “tails and rims” of white sericite. Most stream sediment sample locations yield dark monazite with attached sericite, although in some areas, erosion was sufficient to totally remove the sericite. (Magnification 31X)

fracture and hardness similar to yellow monazite, but contrasts in specific gravity, luster, and color, with the following properties:

- well-developed cleavage (see Fig. 5);
- well-marked parting plane;
- specific gravity from 4.25 to 4.70 g/cm³ (Rosenblum & Mosier 1983);
- luster ranges from submetallic in the blackest of grains, glittery vitreous on tiny crystalline facets to variable resinous;
- color ranges from light gray on thin edges, to black and sometimes inclining towards beige.
- grains range translucent to opaque

The presence of microscopic clouds of amorphous carbon, detrital and authigenic rutile, and inclusions of silt-sized

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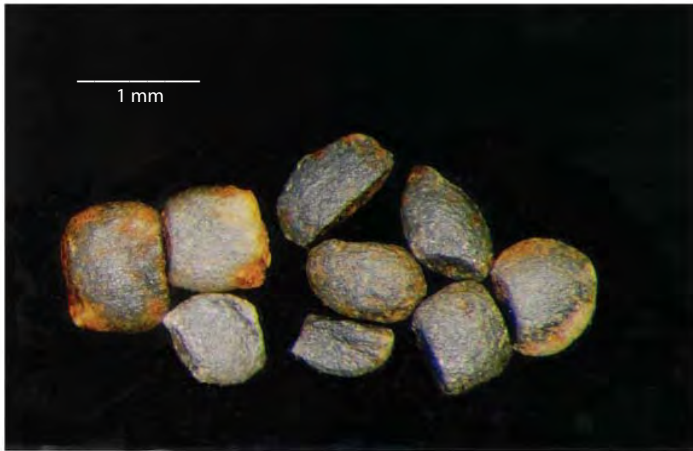


Figure 5. Photomicrograph of dark monazite grains showing well developed cleavage and parting surfaces similar to those of yellow monazite (magnification 31X).

detrital quartz, feldspar, chlorite, sericite, and apatite affect its specific gravity, luster and color. Grain morphology, which includes an apparent annular ring structure (produced by growth ridges), has microcrystalline faces reflecting the glittery vitreous luster (Fig. 6). The black spots were shown by X-ray diffraction to be amorphous and therefore are interpreted to be organic carbon inclusions.

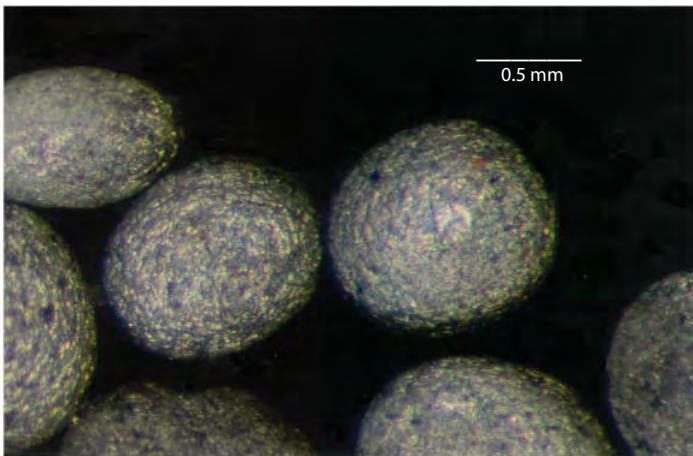


Figure 6. Photomicrograph of dark monazite grain morphology. Note the apparent annular ring structure produced by growth ridges and microcrystalline faces. The black spots are organic carbon inclusions. (Magnification 56X)

Scanning electron microscope and electron probe microanalysis of dark monazite

The primary stream sediment sample site (TA559, Fig. 1b) selected for this study is in the upper reaches of Harris Creek. A typical inclusion distribution within a monazite grain is quartz, albite, muscovite, chlorite, rutile and apatite (Fig. 7). Chemical zoning was not observed under the SEM, but later observations using the electron microprobe indicated distinct zoning.

In the upper reaches of Harris Creek, numerous grab samples of rubble crop were collected for the purpose of finding the dark monazite in source rocks. The bedrock in

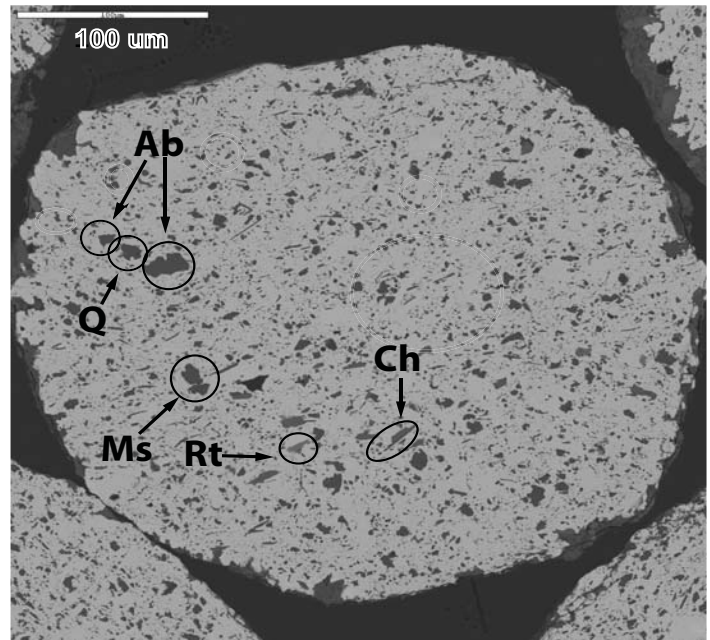


Figure 7. SEM photomicrograph showing typical inclusion distribution within a dark monazite grain from TA559 concentrate. The observed inclusions are quartz (Q), albite (Ab), muscovite (Ms), chlorite (Ch), and rutile (Rt).

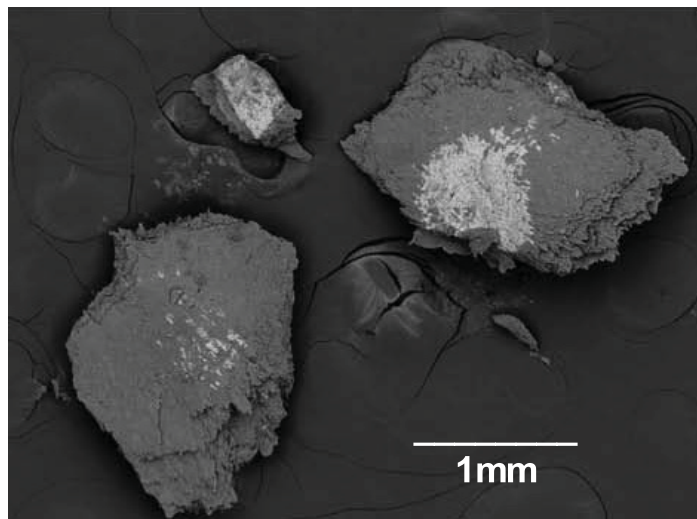
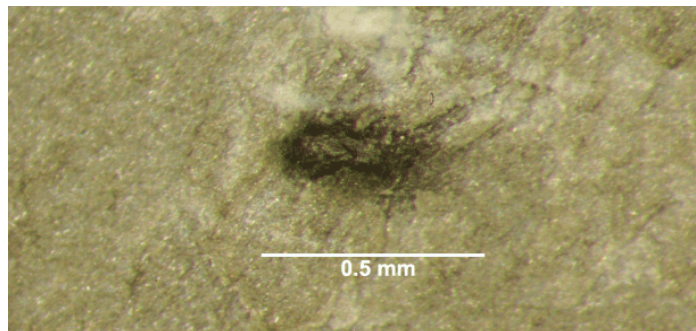
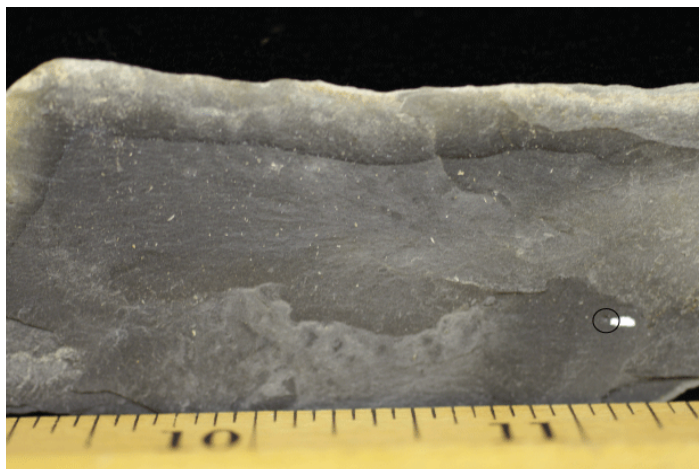
this area is composed of mid-Cretaceous clastic sedimentary rocks of the Kuskokwim Group. The common presence of sericite schist at the TA559 site suggests that the rocks have been subjected to low-grade metamorphism. The sericite schist contains numerous tiny black inclusions (Fig. 8a) which when viewed more closely are dark monazite grains, typically 0.5 to 1 mm in size (Fig. 8b). If one did not know to look for these grains, they could easily be overlooked. A grab sample was split and scanned by SEM backscatter to reveal the internal morphology of the monazite in the cleavable sericite schist (Fig. 8c). Examination of the halves (plus a smaller piece that broke off during the split) clearly show how monazite (the white colored material in the photo) grew between the schist cleavages showing it has both lateral growth along a cleavage plane as well as thickness across cleavage planes. Additional SEM scans performed on sericitic schists, metasiltstones, metagraywackes, and quartzites to determine which rock-types host the dark monazites suggest that sericitic schists host more occurrences of dark monazite than the other rock-types. The siltstone was also found to host monazite, mostly in the sericitic portions of the rock. Thus, monazite is present, but less commonly than in the schist.

Chemical composition of detrital dark monazite

The results of the electron microprobe analyses are given in Table 1. None of the analyses showed Pb contents above the detection limit of 200 ppm. For this reason, no attempt was made to date these monazites.

Thorium oxide concentrations of the dark monazite range from 0.02 to 1.75 wt.%, with the average being 0.61

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Figures 8. Photographs and photomicrograph of in situ dark monazite in phyllitic schist. (a) photograph of schist showing in situ monazite, scale is in inches. The monazite grain studied in detail is located just to the left of the white spot painted on the rock and enclosed by the circle. (b) close-up of the in situ dark monazite, approximately 1 mm in length, in sericite schist (light color). (c) SEM backscatter image of the schist split along a cleavage plane containing the dark monazite (bright material) showing it has both lateral growth along a cleavage plane as well as thickness across cleavage planes.

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Grain	Al	Si	Ca	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Dy	Er	Pb	Th	U	P	O	Sum
1	bd	1.37	0.10	0.55	7.11	24.20	3.60	15.42	2.97	0.56	1.17	0.18	bd	bd	0.78	0.03	13.3	28.4	99.7
2	bd	bd	0.06	0.43	4.91	22.00	3.93	20.02	4.49	0.62	1.52	0.21	bd	bd	0.15	0.01	13.7	27.6	99.7
3	bd	bd	0.12	0.48	6.60	25.00	3.95	16.79	2.99	0.52	1.12	0.18	bd	bd	0.96	0.01	13.7	27.7	100.1
4	0.02	0.01	0.15	0.49	5.32	22.20	3.91	18.66	3.75	0.72	1.32	0.13	bd	bd	1.11	0.02	13.6	27.4	98.8
5	bd	bd	0.03	0.30	13.83	30.90	3.05	9.91	0.89	bd	0.16	0.12	bd	bd	0.18	bd	13.9	28.1	101.4
6	bd	bd	0.12	0.64	9.75	26.90	3.24	13.03	2.16	0.36	0.85	0.22	bd	bd	0.73	0.03	13.8	27.8	99.6
7	0.10	1.73	0.12	0.46	6.32	22.80	3.45	15.70	3.23	0.60	1.28	0.15	bd	bd	1.12	0.02	13.0	28.2	98.3
8	bd	bd	0.08	0.45	6.93	26.10	3.98	17.27	2.73	0.34	0.93	0.15	bd	bd	0.32	0.01	13.8	27.9	101.0
9	bd	bd	0.04	0.29	10.52	29.90	3.66	12.95	1.29	0.20	0.49	0.11	bd	bd	0.10	0.01	13.6	28.0	101.2
10	bd	bd	0.08	0.49	6.59	25.40	3.94	17.45	3.05	0.35	1.05	0.19	bd	bd	0.78	0.02	13.7	27.8	100.9
11	bd	bd	0.03	0.37	10.20	29.00	3.57	13.50	1.96	0.36	0.72	0.14	bd	bd	0.16	0.02	13.8	28.1	101.9
12	bd	bd	0.12	0.49	10.95	28.20	3.32	11.81	1.49	0.23	0.66	0.09	bd	bd	1.12	0.03	13.6	27.6	99.7
13	bd	bd	0.05	0.46	8.26	27.60	3.73	15.27	2.39	0.31	0.78	0.17	bd	bd	0.20	0.02	13.9	28.0	101.1
14	bd	bd	0.10	0.60	5.32	22.50	3.90	19.81	4.19	0.61	1.41	0.23	bd	bd	0.56	0.01	13.8	27.9	100.9
15	bd	bd	0.01	0.32	11.22	30.50	3.60	12.63	1.48	0.23	0.59	0.11	bd	bd	0.04	bd	13.8	28.1	102.6
16	0.01	1.51	0.14	0.55	6.99	25.20	3.79	15.95	2.87	0.59	1.16	0.20	bd	bd	0.83	0.01	13.3	28.9	102.0
17	bd	bd	0.16	0.58	6.99	25.40	3.57	15.98	2.88	0.71	1.05	0.21	bd	bd	1.75	0.02	13.8	27.8	100.9
18	bd	bd	0.09	0.50	7.81	26.90	3.64	15.49	2.71	0.31	0.93	0.19	bd	bd	0.75	0.02	13.7	27.8	100.8
19	bd	0.05	0.12	0.35	11.47	28.70	3.29	11.29	1.37	0.10	0.54	0.10	bd	bd	1.27	bd	13.8	27.9	100.4
20	bd	bd	0.02	0.24	9.59	29.20	3.62	13.58	2.02	0.21	0.63	0.10	bd	bd	0.14	bd	13.8	27.9	101.1
21	bd	bd	0.10	0.35	10.49	30.00	3.47	12.29	1.43	0.16	0.51	0.12	bd	bd	0.75	bd	14.0	28.3	102.0
22	0.10	bd	0.04	0.56	9.77	29.20	3.71	13.66	1.86	0.34	0.85	0.24	bd	bd	0.02	0.03	13.9	28.4	102.7
23	bd	bd	0.05	0.29	7.71	27.30	3.92	15.81	2.48	0.32	0.88	0.12	bd	bd	0.69	0.01	13.8	28.0	101.4
24	bd	bd	0.04	0.26	13.17	30.50	3.14	10.41	1.04	0.05	0.27	0.07	bd	bd	0.54	0.01	13.8	28.0	101.3
25	bd	bd	0.11	0.70	4.88	21.60	3.85	20.53	5.04	0.61	1.72	0.27	bd	bd	0.18	0.03	13.8	27.8	101.1
AVG	0.06	0.93	0.08	0.45	8.51	26.70	3.63	15.01	2.51	0.39	0.90	0.16	bd	bd	0.61	0.02	13.7	28.0	101.7
1σ	0.05	0.83	0.04	0.13	2.55	3.00	0.27	2.94	1.08	0.20	0.38	0.05	bd	bd	0.46	0.01	0.0	0.3	1.1

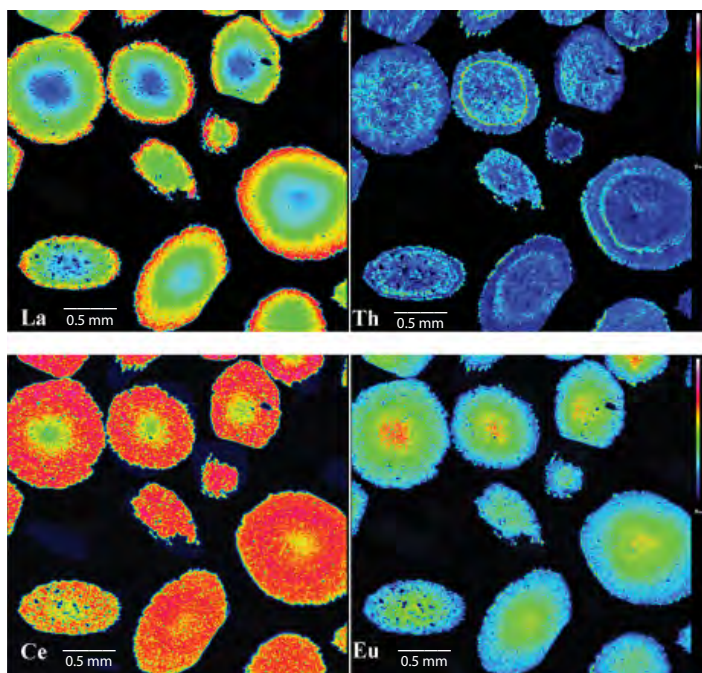
* bd - below detection limit

Table 1. Electron microprobe chemical analyses of 25 detrital dark monazite grains from TA559 concentrate showing the average REE concentrations and confirming the high Eu content. All values are in wt. %.

wt.%. Uranium oxide concentrations are mostly below 0.03 wt.%. Conversely, Eu_2O_3 contents are very high, ranging from 0.05 to 0.72 wt.%, the average being 0.39 wt.%. Neodymium (Nd) concentration, which is closely associated with Eu content at the cores of the dark monazite, ranges from 9.91 and 20.53 wt.% Nd_2O_3 , with an average of 15.01 wt.%. Lastly, Ce_2O_3 concentrations range from 21.6 to 30.9 wt.%, with an average of 26.7 wt.%.

X-ray mapping of Eu content in these detrital dark monazite grains (Fig. 9) shows they are highly zoned, with the Eu concentrated in the cores and decreasing substantially outward toward the rims. Conversely, La content is low in the grain cores and increases sharply at the rims. Cerium concentration is more chemically homogenous throughout the grains, usually with the cores only slightly reduced in content as shown by the limited range in color from green to red

Figure 9. Electron microprobe photomicrograph of element distributions within detrital dark monazite grains. Colors indicate variation in elemental concentration, where dark colors (blue) are the lowest concentration and bright colors (white) are the highest concentration as shown by the color bar at the right of the figure. Monazite grains are zoned with Eu at the core and La at the rim.



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on the elemental map. Elevated Th contents occur with the highest La concentrations as narrow enriched zones in only a few of the monazite grains and satellite spots in the rims of other grains

Chemical composition of in situ dark monazite

SEM spectral imaging was collected on dark monazite imbedded in sericitic schist. A backscattered electron image of one of these monazite growths, together with elemental X-ray maps of Ce, La, and Eu concentrations, is given in Figure 10. As seen here, the monazite appears as complex idioblastic intergrowths with mica and quartz rather than a well-formed separate discoidal grain. The element maps suggest homogeneous distributions of Ce and Eu, but subtle differences in La distribution across the growth; note the darker yellow colors at the outer edge of the growth compared to the brighter yellow in the interior. Unfortunately, no samples of in situ dark monazite could be polished to a flat surface suitable for microprobe analysis. At this time, there are insufficient data to show whether the in-situ monazite grows to become discoidal grains such as those that are found in the stream sediments.

Occurrences of detrital gold

Angular to sub-rounded detrital gold (Fig. 2) was found with dark monazite in virtually every concentrate sample from streams draining the northeast-trending ridge from

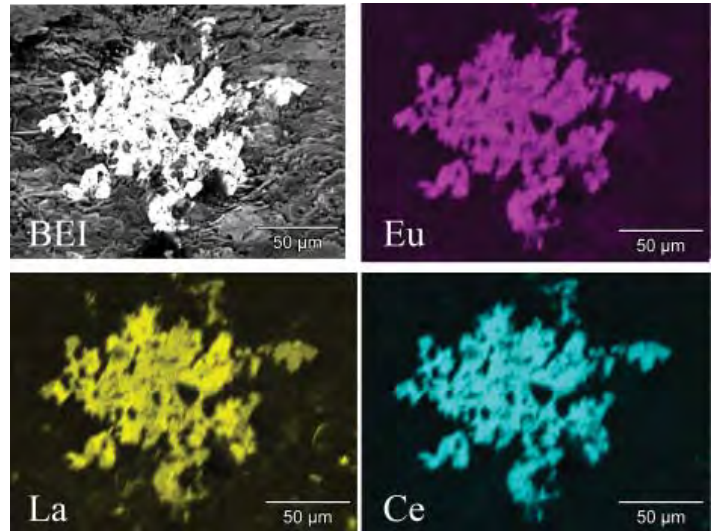


Figure 10. SEM backscatter image (BEI) of in situ dark monazite in sericite schist together with element distribution maps for Eu, La, and Ce. REE distribution is relatively homogeneous across the monazite growth. For each element (color) the dark regions represent lower concentration than bright regions. There is a slight decrease in La concentration at the outer boundaries, especially noticeable at the upper right area of monazite.

Sleitat Mountain. The gold may come from quartz veins, associated with the Sleitat Mountain stock, which cuts the mid-Cretaceous clastic sedimentary rocks of the Kuskokwim Group. Five to ten ounces of gold was recovered from a wolframite-bearing pegmatite vein nearby (Mertie 1938). A magnetic survey of Taylor Mountains quadrangle which included the Sleitat Mountain area shows a possible cupola to the northeast of Sleitat Mountain (available at: A Website for the Distribution of Data, World Wide Web: <http://pubs.usgs.gov/ds/2006/224/> and http://pubs.usgs.gov/ds/2006/224/Data/USGS_TaylorMtsMagFinalReport.pdf). The 2008 sampling traverse conducted across this ridge revealed igneous rubble crop at that location.

Conclusions

The relatively high Eu concentration, with generally very low contents of U and Th in dark monazite, from the Sleitat Mountain area and elsewhere in the Taylor Mountains quadrangle points to the desirability of this variety of monazite becoming a potential source for REE. Europium oxide has gained considerable commercial interest (Haxel et al. 2002; Hedrick 2004). Utilized principally as a phosphate activator, Eu-activated yttrium (Y) compounds produce red color in television, computer monitor, and light emitting diodes (LED). Certain alloys of Eu are used in the petroleum industry, such as cracking catalysts in refineries. Currently, Eu is separated from bastnaesite $[(\text{Ce}, \text{La})(\text{CO}_3)\text{F}]$ ore found in China and California, and the ionic clays of southern China; however, all contain only small concentrations of Eu. Europium is difficult to separate from the other REE, especially in the presence of U and Th, making it one of the most expensive REE. The United States once was self-sufficient in

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the production of REEs, but in the last 20 years has become dependent largely upon imports from China (Bayan Obo deposit and lateritic deposits of tropical southern China).

The thickness of the bedrock at Sleitat Mountain that contains the dark monazite is not known at this time. However, the heavy mineral concentrate surveys indicate the lateral distribution of the dark monazite to be widespread (Fig. 1b). The costs of mining the dark monazite possibly could be offset by the recovery of gold. The Th content of the dark monazite is low and the paucity of Th-rich yellow monazite suggests that radioactive hazards may be of little economic consequence. The importance of ensuring domestic resources of REE, and particularly the scarcity of national Eu resources, suggests that this area of the Taylor Mountains quadrangle would be an important region to evaluate as a potentially significant source of Eu.

Acknowledgements

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