MONAZITE-(Sm), A NEW MEMBER OF THE MONAZITE GROUP FROM THE ANNIE CLAIM #3 GRANITIC PEGMATITE, SOUTHEASTERN MANITOBA

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ABSTRACT

Monazite-(Sm) was found in the Annie Claim #3 pod of lepidolite-subtype granitic pegmatite within the Greer Lake intrusion of pegmatitic leucogranite, in the Archean Bird River Subprovince of the Superior Province, in southeastern Manitoba. It occurs as tabular crystals ±0.4 mm in diameter, associated with manganocolumbite, quartz, albite and lithian muscovite. Monazite-(Sm) is yellowish, translucent, with a white streak, vitreous to greasy luster, and no observed fluorescence. One good cleavage is present, tenacity is brittle, and the fracture is uneven. Monazite-(Sm) has no observable pleochroism; α 1.768 (5), β 1.771(3), γ > 1.808(3); 2V(meas) 29(8)°, X = b, Z ∩ c = 9° (β obtuse). It is monoclinic, space group P21/n; a refinement from single-crystal and powder (Gandolfi) X-ray-diffraction data gave a 6.725(1), 6.739(6), b 6.936(1), 6.951(3), c 6.448(1), 6.462(3) Å, β 104.02(1)°, 104.03(4)°, V 291.8(1), 293.6(2) Å³, Z = 4; Dcalc derived from average chemical composition is 5.512 and 5.478 g/cm³. The strongest six lines of the (Gandolfi) X-ray-diffraction pattern [d in Å(hkl)] are: 4.647(5)(011), 4.164(8)(¯111), 3.492(4)(111,020), 3.065(10)(120), and 2.857(9)(¯112,012). The monazite-(Sm) contains, in wt.%, up to 14.29 Sm₂O₃, 13.48 Gd₂O₃ and 6.28 Nd₂O₃, and moderate percentages of the huttonite and brabantite components. The most Sm-rich composition gives (Sm0.197Gd0.179Ce0.148Th0.125Ca0.107Nd0.090La0.030Y0.030Pr0.023Tb0.017Zr0.017Dy0.016Pb0.016U0.002) > 1.808(3); 2X = 49.9(5); 6.936(1), 6.951(3), 6.448(1), 6.462(3) Å, β 104.02(1)°, 104.03(4)°, V 291.8(1), 293.6(2) Å³, Z = 4. The density calculated from the average chemical composition is 5.512 and 5.478 g/cm³. The middle-REE-dominant signature of the Annie Claim #3 monazite-(Sm) is shared with the broadly associated Y(Ta,Nb)O₄ mineral (formanite?) and dysprosian xenotime-(Y); this exotic pattern ofREE abundances is possibly generated by selective and differential complexing of REE in the granite-to-pegmatite sequence of solidification.

Keywords: monazite-(Sm), new mineral species, rare-earth elements, granitic pegmatite, Annie Claim #3, Manitoba, Canada.

Sommaire

Nous avons découvert la monazite-(Sm) dans la lentille de pegmatite granitique à lépidolite dite Annie Claim #3, au sein du massif intrusif du lac Greer, en la sous-province de Bird River de la Province du Supérieur, dans l’Archeen du sud-est du Manitoba. Elle se présente en cristaux tabulaires atteignant 0.4 mm de diamètre, associée à manganocolumbite, quartz, albite et muscovite lithian. Les cristaux de monazite-(Sm) sont jaunâtres, translucides, ayant une rayure blanche, un éclat vitreux à résineux, sans fluorescence. Nous décelons un bon clivage; la tenacité est cassante, et la fracture, inégale. La monazite-(Sm) ne montre aucun pleochroisme observable; α 1.768 (5), β 1.771(3), γ > 1.808(3); 2V(meas) 29(8)°, X = b, Z ∩ c = 9° (β obtuse). Elle est monoclinique, groupe spatial P21/n; un affinement des paramètres réticulaires à partir des données prélevées sur monocristal et sur poudre (méthode de Gandolfi) a donné a 6.725(1), 6.739(6), b 6.936(1), 6.951(3), c 6.448(1), 6.462(3) Å, β 104.02(1)°, 104.03(4)°, V 291.8(1), 293.6(2) Å³, Z = 4. La densité calculée à partir de la composition moyenne est 5.512 et 5.478 g/cm³. Les six raies les plus intenses du spectre de diffraction X (Gandolfi) [d en Å(hkl)] sont: 4.647(5)(011), 4.164(8)(¯111), 3.492(4)(111,020), 3.065(10)(120), et 2.857(9)(¯112,012). La monazite-(Sm) contient jusqu’à 14.29% Sm₂O₃, 13.48% Gd₂O₃ et 6.28% Nd₂O₃ (poids), et des proportions appréciables des composantes huttonite et brabantite. La composition la plus enrichie en Sm donne (Sm0.197Gd0.179Ce0.148Th0.125Ca0.107Nd0.090La0.030Y0.030Pr0.023Tb0.017Zr0.017Dy0.016Pb0.016U0.002) > 1.808(3); 2X = 49.9(5); 6.936(1), 6.951(3), 6.448(1), 6.462(3) Å, β 104.02(1)°, 104.03(4)°, V 291.8(1), 293.6(2) Å³, Z = 4. Un ajustement des teneurs en U et Th de la monazite-(Sm) à leurs teneurs originelles il y a 2.64 Ga améliore légèrement la stoechiométrie et prouve que tout le Pb est radiogénique. L’enrichissement de ce minéral en terres rares moyennes se voit aussi dans un minéral associé de composition Y(Ta,Nb)O₄ (formanite?) et le xénotème-(Y) enrichi en dysprosium.

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à Annie Claim #3. Cet enrichissement inhabituel pourrait bien refléter la complexation sélective et différentielle des terres rares au cours de la solidification progressive sur l’intervalle granite–pegmatite.

(Traduit par la Rédaction)

Mots-clés: monazite-(Sm), nouvelle espèce minérale, terres rares, pegmatite granitique, Annie Claim #3, Manitoba, Canada.

INTRODUCTION

There are three known members of the monazite group sensu stricto. They contain as dominant trivalent cations the three lightest, largest and geochemically most abundant rare-earth (REE) elements, La, Ce and Nd, in accord with their preference for the monazite structure (Ni et al. 1995). Monazite-(Ce) is by far the most widespread member, whereas monazite-(La) is rarely observed, and monazite-(Nd) seems to be known so far from only three localities (Gaines et al. 1997, Anthony et al. 2000).

Here we report on a new monazite-group mineral, the REE composition of which is distinctly shifted toward the middle REE (MREE): monazite-(Sm) in fact has Gd a close second in importance to Sm. Moreover, Tb is the third most abundant REE in terms of chondrite-normalized values. The new mineral is named in accordance with recommendations of the Commission on New Minerals and Mineral Names of the International Mineralogical Association for REE minerals. The species and the name were approved by the above Commission (IMA 2000-001). The polished section analyzed by electron microprobe (EMPA), the single crystal used for Gandolfi XRD are catalogued under #M7181 in the R.B. Ferguson Museum of Mineralogy, Department of Geological Sciences, University of Manitoba.

THE PARENT PEGMATITE

Monazite-(Sm) was found in the Annie Claim #3 granitic pegmatite, a zoned body of the lepidolite subtype located in the southwestern margin of its parent Greer Lake pegmatitic granite. This LCT-family granite and derived pegmatites, dated at 2640 ± 7 Ma (Baadsgaard & Černý 1993), were intruded in metasediments of the Lamprey Falls Formation of the Bird River Greenstone Belt, in the Bird River Subprovince of the western part of the Superior Province of the Canadian Shield; the geographic location is 1.2 km northwest of Greer Lake in southeastern Manitoba, close to Winnipeg River, at latitude 50°21'12"N and longitude 95°20'18"W. Černý et al. (1981) provided details concerning the regional geology.

The subellipsoidal, subhorizontal body of the pegmatite is approximately 7 × 10 meters in size, with distinct concentric zoning (Masau 1999). The zones show an inward progression, changing in texture and mineral assemblage, exemplified by a border-to-core succession of five main types of mica (from muscovite to lithian muscovite to lepidolite), associated mainly with albite and quartz. Microcline perthite is sporadic in the outermost zones and absent from the intermediate and inner zones. Accessory minerals include spessartine, cesian beryl, cassiterite with exsolved zirconian–hafnian wolodiegin (Masau et al. 2000a), manganocolumbite, manganotantalite and rare primary wolodiegin, mircolite, uranpyrochlore, ferrotiolite, probable formanite-(Y), fluorapatite, zircon, coffinite, uraninite, dysprosian xenotime-(Y) (Masau et al. 2000b) and monazite-(Sm). The pegmatite and the parent pegmatitic granite do not contain any minerals of B or phosphates of Li, Al, Fe or Mn, and sulfides are extremely rare.

EXPERIMENTAL

Monazite-(Sm) was analyzed using a Cameca SX–50 electron microprobe in wavelength-dispersion mode, under the following conditions: operating voltage 20 kV, beam current 40 nA, beam size 3 μm. Counting times for peak and background determinations were 20 s and 10 s, respectively, for Si, P, Ca, Zr, Dy, Pb, Th and U; for the remaining elements, the values were 40 and 20 s, respectively. Monazite-(Sm) was analyzed using the following standards: SmPO4 (PKo, SmLβ) and other individual synthetic phosphates APO4 (LO for Y, La, Ce, Nd and Th, Lβ for Pr, Gd and Dy), zircon (SiKα, ZrLβ), mimetite (PbMβ), ThO2 (ThMα), UO2 (UMβ) and Wilberforce apatite (CaKα). Concentrations of Ho, Er and Tm [the last adjusted for overlap with Sm using the method of Åmli & Griffin (1975) and Nagy (1993)] were found to be below detection limits (0.03, 0.02 and 0.04 wt.%, respectively, at 100 nA and 100 s). Contents of Yb and Lu were at about their respective limits of detection under these conditions (0.02 and 0.05 wt.%); Na, K, Mn, Fe, Mn, Ba, Sr, As, S, F, Nb and Ta were not detected. The analytical data were reduced and corrected by the PAF method of Pouchou & Pichoir (1984, 1985). Chondritic REE values of Taylor & McLennan (1985) were used to normalize the REE contents.

Single-crystal data were collected on a Bruker automated four-circle diffractometer equipped with a CCD detector, using MoKα radiation with λ = 0.71073 Å. Unit-cell dimensions were refined from 106 diffraction maxima (I(σi) > 10) with 20 up to 60°. Powder data were recorded with a 114.6 mm Debye–Schererrer camera with Gandolfi attachment, using Ni-filtered CuKα radiation with λ = 1.5418 Å.
Monazite-(Sm)

Monazite-(Sm) was found to be closely aggregated with platy crystals of manganocolumbite in the inner intermediate zone of the parent pegmatite. The aggregate is associated with quartz, "cleavelandite" and curvilinear lithian muscovite. No other REE minerals are present in close proximity to monazite-(Sm), although some do occur elsewhere in the pegmatite, as mentioned above.

Platy subhedral crystals of monazite-(Sm) attain a maximum dimension of 0.4 mm (Fig. 1). They are brittle, yellowish and translucent to transparent, with vitreous to slightly greasy luster and white streak. Microscopic fragments show one good cleavage and, otherwise, uneven fracture-surfaces. No fluorescence was observed in UV light. Density and hardness could not be measured. However, density calculated from the average chemical composition is 5.512 and 5.478 g/cm³ for unit-cell dimensions derived from X-ray diffraction single-crystal and powder (Gandolfi) data, respectively. Optical properties measured at a wavelength of 589.9 nm are α 1.768(5), β 1.771(3), γ > 1.808(3) [no immersion liquid with n > 1.808 was available to the authors; γ calculated from α, β and 2V (measured) is 1.818]. The measured (+)2V is 29(8)°; the value calculated using 1.808 as a minimum for γ is 32°, X = b, Z ∞ c = 9° (β obtuse). Microscopic fragments of monazite-(Sm) are colorless.

The chemical composition of monazite-(Sm) is shown in Table 1. The average composition of the fragments used for XRD and optical study (Table 1, #1) yields a formula with Sm distinctly dominant over Gd, Ce, Th and Ca, with subordinate to minor contents of other REE. The dominance of Sm and Gd over other cations is even better expressed in the most Sm-enriched composition (Table 1, #2): (Sm0.197 Gd0.179 Ce0.148 Th0.125 Ca0.107 Nd0.090 La0.030 Y0.030 Pr0.023 Tb0.017 Zr0.017 Dy0.016 Pb0.016 U0.002)0.997 (P0.963 Si0.044)1.007 O4. However, note that this extreme composition was not obtained on any of the grains extracted from the polished section for XRD work and determination of physical properties.

Slight improvement of stoichiometry and charge balance is achieved by calculation of the original U and Th contents of the monazite-(Sm) as they were at the time of crystallization 2.64 Ga ago (Table 1, #1* and 2*). The virtually identical atomic content of present-day Pb (0.023 and 0.016 in #1 and 2) with those of decayed U + Th (0.023 and 0.018; 1* less 1 and 2* less 2, respectively).
respectively) shows that all Pb is radiogenic, and that any potential loss of Pb from the monazite structure has been negligible.

The current content of Th is substantial, but it gives only 4 to 10 mole % of the huttonite component ThSiO₄ and 10 to 14 mole % of the brabantite component CaTh(PO₄)₂, when split between the available Si and Ca, respectively. The stoichiometry of the brabantite and huttonite components in solid solution is confirmed by the good alignment of atomic Si versus (Th + Zr + U – Ca) in Figure 2A, although slightly shifted in favor of the latter elements. Good analytical totals and stoichiometry calculated on an anhydrous basis indicate that any potential role of a tetrahedral array of (OH) groups must be negligible, if any, in both the current and original compositions. Figure 2B shows that the A/(P + Si) ratio equals 1 within the limits of analytical error.

Unit-cell dimensions were derived from single-crystal and powder data: a = 6.725(1), 6.739(3), b = 6.936(1), 6.951(3), c = 6.448(1), 6.462(3) Å, β = 104.02(1)°, 104.03(4)°, V = 291.8(1), 293.6(2) Å³, Z = 4. The XRD powder data are shown in Table 2. Systematically absent diffraction-maxima in the single-crystal and powder data are consistent with space group P2₁/n. Both the unit-cell dimensions and the powder data closely match those available for other monazite-group minerals (e.g., Ni et al. 1995). However, the substantial contents of Th and Ca do not permit direct comparison with cell-parameter values calculated on the basis of occupancy by the REE alone (Ni et al. 1995).

**GEOCHEMICAL CONSIDERATIONS**

As noted in the introduction, dominance of La or Nd is rather rare in monazite-group minerals, although both these elements are commonly the second and third most abundant REE in monazite-(Ce). The extent of enrichment in Sm (and Gd) shown in Figure 3 for monazite-(Sm) has not so far been reported. The highest Sm₂O₃ contents in monazite-(Sm) are shown in Table 3 for monazite-(Sm) and compared to other monazite-group minerals.

**TABLE 1: REPRESENTATIVE COMPOSITIONS OF MONAZITE-(Sm)**

| P₂O₅ | 27.48 | 27.39 | 27.48 | 27.39 | P | 0.942 | 0.903 | 0.948 | 0.951
| SiO₂ | 15.3 | 14.9 | 15.3 | 14.9 | Si | 0.622 | 0.646 | 0.626 | 0.646
| ZₐO₅ | 6.38 | 6.32 | 6.38 | 6.32 | Zₐ | 0.328 | 0.328 | 0.326 | 0.328
| ThO₂ | 10.64 | 10.64 | 10.64 | 10.64 | Th | 0.390 | 0.390 | 0.390 | 0.390
| La₂O₃ | 4.02 | 4.02 | 4.02 | 4.02 | La | 0.126 | 0.126 | 0.126 | 0.126
| Ce₂O₃ | 14.00 | 14.00 | 14.00 | 14.00 | Ce | 0.540 | 0.540 | 0.540 | 0.540
| Pr₂O₅ | 1.99 | 1.99 | 1.99 | 1.99 | Pr | 0.097 | 0.097 | 0.097 | 0.097
| Nd₂O₃ | 6.63 | 6.63 | 6.63 | 6.63 | Nd | 0.228 | 0.228 | 0.228 | 0.228
| Sm₂O₃ | 13.06 | 13.06 | 13.06 | 13.06 | Sm | 0.489 | 0.489 | 0.489 | 0.489
| Gd₂O₅ | 12.68 | 12.68 | 12.68 | 12.68 | Gd | 0.402 | 0.402 | 0.402 | 0.402
| Tb₂O₅ | 10.58 | 10.58 | 10.58 | 10.58 | Tb | 0.350 | 0.350 | 0.350 | 0.350
| Dy₂O₃ | 6.18 | 6.18 | 6.18 | 6.18 | Dy | 0.191 | 0.191 | 0.191 | 0.191
| Ho₂O₅ | 5.62 | 5.62 | 5.62 | 5.62 | Ho | 0.176 | 0.176 | 0.176 | 0.176
| Er₂O₃ | 3.81 | 3.81 | 3.81 | 3.81 | Er | 0.117 | 0.117 | 0.117 | 0.117
| Tm₂O₃ | 2.75 | 2.75 | 2.75 | 2.75 | Tm | 0.070 | 0.070 | 0.070 | 0.070
| Yb₂O₃ | 2.14 | 2.14 | 2.14 | 2.14 | Yb | 0.068 | 0.068 | 0.068 | 0.068
| Lu₂O₃ | 1.65 | 1.65 | 1.65 | 1.65 | Lu | 0.050 | 0.050 | 0.050 | 0.050
| Ce₂O₃ | 2.75 | 2.75 | 2.75 | 2.75 | Ce | 0.117 | 0.117 | 0.117 | 0.117
| CoO | 2.14 | 2.14 | 2.14 | 2.14 | Co | 0.068 | 0.068 | 0.068 | 0.068
| Y₂O₃ | 1.65 | 1.65 | 1.65 | 1.65 | Y₂ | 0.050 | 0.050 | 0.050 | 0.050
| Total | 100.01 | 100.01 | 100.01 | 100.01 | Total | 100.01 | 100.01 | 100.01 | 100.01

- 1 Sr% average of 3 grains used for calculating physical, optical, and crystal data
- Composition 1 and 2 are adjusted for 2.96 GPa of hydrostatic pressure on 2, 4, and 6-atm. U and Th; atomic contents normalized to 4 oxygen atoms.
- Not detected: Na, K, Mg, Sr, Ca, Eu, Nd, Tb, Dy, Er, Tm, Yb, Lu.
content noted to date is 9.7 wt.% (accompanied by 4.3 wt.% Gd$_2$O$_3$), associated with extreme Nd in monazite-(Nd) from its type locality, where it occurs as a hydrothermal mineral in Alpine veins; Graeser & Schwander (1987), Demartin et al. (1991). High Sm and Gd also were reported in an incompletely analyzed sample from Chernaya Salma in Karelia (Vainshtein et al. 1956, quoted in Neumann et al. 1966). Otherwise, Sm invariably is subordinate to minor with respect to the major REE, as is Gd. This is abundantly documented in general reviews of monazite (e.g., Neumann et al. 1966, Fleischer & Altschuler 1969), as well as in statistically extensive studies of monazite from a variety of specific petrological environments: granitic suites, granitic pegmatites, tin deposits, Alpine fissures, sediments and metamorphic rocks (e.g., Lee & Bastron 1967, Donnot et al. 1973, Petrak & Owens 1975, Burnotte et al. 1989, Wark & Miller 1993, Watt 1995, Förster & Rheede 1995, Franz et al. 1996, van Emden et al. 1997, Förster 1998).

What may be the reason for the unique enrichment of the Annie Claim #3 monazite-(Sm) in Sm and Gd? Crystal-chemical factors would favor Ce > La > Nd over and above the MREE and HREE. Partitioning of these three elements into competing minerals cannot be invoked, as the associated minerals are not prominent REE carriers. The close similarity of ionic radii of Th, Sm and Gd, invoked by Förster et al. (2000) to explain the enrichment of Sm and Gd in huttonite, cannot be responsible, as monazite is routinely enriched in Th, but its concentration is not correlated with the content of Sm or Gd.

A significant clue to the puzzle is the abundance of the MREE in the other REE minerals in the Annie Claim #3 pegmatite: xenotime-(Y) is strongly enriched in Dy, Gd and Tb (±4.88, 7.59 and 3.31 wt.% oxides, respectively; Masau et al. 2000b), and a mineral with composition of Y(Ta,Nb)O$_3$ [probable formanite-(Y)] contains, in wt.%, 9.05 Gd$_2$O$_3$, 7.18 Dy$_2$O$_3$ and 3.22 Sm$_2$O$_3$ (unpublished data of the authors). These two minerals also defy the crystal-chemical preferences of their structures, and they occur isolated from any potentially competing phases.

The enrichment of all three minerals in MREE strongly suggests that this signature was a general feature of the parent pegmatite-forming melt from which the Annie Claim #3 pod solidified. The magma, residual after crystallization of a sizeable body of REE-poor leucogranitic parent, was significantly enriched in H$_2$O, F, Li (and probably also CO$_2$, the abundance of which is documented in late fluid inclusions). Such a volatile-rich environment of a highly hydrous melt could have been conducive not only to probable promotion of the tetrad effect (Masau et al. 2000b; see also Fig. 3), but to complexing of the REE as early as during the solidification of the parent granite. Complexing, selective in terms

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* Calculated with CardufK program. ** Crystallographic parameters (Nd-fonn). $d_0$ is the observed spacing, $d_{	ext{apo}}$ is the calculated spacing, all data are for untedded sole. Note the prominence of Sm and Gd in both diagrams, and the high value of chordite-normalized Tb.
of individual REE and differential in terms of complex stabilities, proposed by Gramaccioli & Pezzotta (2000; see also Pezzotta et al. 1999, Gramaccioli et al. 1999) could have generated the MREE-enriched signature of the residual pegmatite. Despite the recent progress in studies devoted to REE complexing at relatively low temperatures (e.g., Wood 1990, 1996), experiments under magmatic conditions bordering to supercritical are desirable to address problems such as that posed by our MREE-dominant minerals. Moreover, granitic pegmatites are notorious for disequilibrium crystallization, the REE minerals are rare and isolated from each other in the pegmatite examined, and the analytical data are incomplete for the discussion of the possible role of the tetrad effect. Consequently, any further discussion of the possible role of the tetrad effect versus the F-controlled enhancement or dispersal of different REE in fluid and solid phases (C.M. Gramaccioli, pers. commun., 2001) seems to be futile at present.

Minerals of the monazite and xenotime groups are very scarce in, and rarely recovered in any quantities from, highly fractionated granitic pegmatites rich in Li, Rb, Cs, F, B and P (mainly the lepidolite, spodumene and petalite subtypes of rare-element pegmatites of Černý 1991). Any finds of these phases from such environments should be thoroughly examined, as their REE patterns may help to shed light on fractionation of these elements in highly evolved, complex and ligand-rich environments. REE abundances in accessory minerals, which currently seem to represent only a few exotic anomalies, may prove to be typical of specific categories of pegmatite and the conditions of their derivation and consolidation.

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