

Fluorite and its potential as an indicator mineral for carbonatite-related rare earth element deposits

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Abstract

Fluorite has potential as a proximal indicator mineral for the exploration of specialty metals, particularly rare earth elements and Nb in carbonatite-related deposits. Plots of Tb/La - Tb/Ca were generated using all available published data (worldwide) to compare REE concentrations in fluorite from carbonatite, peralkaline-alkaline, Mississippi Valley Type (MVT), and vein (sedimentary, igneous, and metamorphic hosted) deposits. Y-Yb plots distinguish between fluorites from carbonatite and MVT deposits. Fluorite from carbonatite-related and MVT deposits form compositional fields with minor overlap. Ongoing investigations include: microchemical analyses of fluorites from key Canadian and foreign deposits, diagram refinement by contouring, the production of probability plots, and experimentation with discrimination diagrams incorporating normalized REE slope calculations with the presence or absence of Ce and Eu anomalies.

Keywords: Fluorite, REE, exploration, discrimination diagrams, specialty metals

1. Introduction

A major objective of the Specialty Metal component of the TGI-4 is to develop and improve technologies and methods that can be applied during the exploration for buried REE and Nb ± Ta deposits. Specialty metals are found in a variety of geological settings including carbonatites and peralkaline intrusions (Simandl, 2012; Simandl et al., 2012). Although fluorite is commonly associated with these specialty metals deposits, it is also present in MVT deposits, Climax-type molybdenum deposits, and fluorite ± barite veins hosted by sedimentary, igneous, and metamorphic rocks. Fluorite can accept trace elements including REEs that substitute for Ca²⁺ in its crystal structure and fluorite crystals may contain inclusions of REE-bearing fluorocarbonates or other REE-bearing minerals. As a consequence, and due to its distinctive physical properties (colour, fluorescence, high density, low hardness, and cleavage), fluorite is a prospective proximal indicator mineral, particularly for carbonatite-related Rare Earth Element (REE) deposits. Herein we present Tb/La - Tb/Ca and Logarithmic Y - Yb plots based on a current, worldwide compilation of REE abundances reported in literature, demonstrating that these diagrams can discriminate between fluorite from carbonatites and MVT deposits.

2. Fluorite

Fluorite occurs in a variety of colours ranging from transparent and colourless to nearly opaque and black, but is commonly purple, green, blue, or yellow, and can exhibit colour zoning, (Trinkler et al. 2005, Staebler et al. 2006). Fluorite is typically fluorescent, and less commonly phosphorescent,

thermoluminescent, and triboluminescent. Colour in fluorite can be caused by a variety of factors including impurities, structural defects, and REE substitutions for Ca, whereas fluorescence is generally attributed to REE substitutions of divalent and trivalent cations for Ca, (Verbeek, 2006). Studies on crystals from the Bingham deposit (New Mexico) identified sector zoning in single crystals and demonstrated that trace element incorporation in fluorite is surface specific (Bosce and Rakovan, 2001).

Other distinguishing characteristics of fluorite are its crystal habit, hardness, and cleavage. Fluorite crystallizes as a cubic, face-centred lattice. Fluorine anions form square stacks forming eight-fold coordination sites for calcium. Ca cations are at the corners and centers of the cubic lattice (Nesse, 2000). Fluorite commonly forms cubes or octahedrons, less commonly dodecahedrons and, rarely, tetrahedrons, trapezohedrons, trisoctahedrons, hexoctahedrons, and botryoidal forms. In thin section, fluorite is isotropic and rarely displays weak, anomalous anisotropism. Twinning is typical in fluorite, following spinel-law twins that can form penetration and contact twins (Staebler et al., 2006). Fluorite generally occurs as broken fragments due to its perfect {111} cleavage in four directions (Nesse, 2000). Fluorite's softness combined with its tendency to cleave makes it an excellent proximal indicator of mineral deposits. Additionally, the density of fluorite can be as high as 3.6 g/cm³ when enriched in REEs (Staebler et al., 2006) approaching those of common specialty metal indicator minerals such as REE-bearing hydrated carbonates and fluorocarbonates (3.5 - 5.0 g/cm³) pyrochlore (4.2 - 6.4 g/cm³) columbite (5.3 - 7.3 g/cm³), and other indirect indicators such as barite (4.5 g/cm³),

celestite (3.9 - 4.0 g/cm³), and apatite (3.1 - 3.2 g/cm³).

Fluorite is primarily used as a flux in steelmaking, iron and steel casting, and other metallurgic processes such as primary aluminum production. Fluorite is also used in producing hydrofluoric acid, glass, enamels, cement, welding rod coatings, herbicides, Teflon, Freon, and other chlorofluorocarbons (Simandl 2009a; b; Staebler et al., 2006).

Information about fluorite occurrences in British Columbia are compiled in Pell (1992) and summarized by Simandl (2009b). Fluorite occurs as a gangue mineral in a number of REE deposits including carbonatites (e.g., Eldor, QC), peralkaline intrusions (e.g., Nechalacho, NWT; Strange Lake, QC and Labrador), REE occurrences of uncertain origin (e.g. Rock Canyon Creek, BC), carbonate-hosted zinc and lead-bearing hydrothermal deposits including MVTs (e.g., Cave in Rock, Illinois), epithermal precious metal deposits (Epinger and Closs, 1990), Climax-type molybdenum deposits (e.g. Climax, Colorado; Ludington and Plumlee, 2009), other greisens-granite-pegmatite-hosted metal-bearing deposits, and fluorite ± barite veins.

3. Historic fluorite discrimination diagrams

The REE chemistry of fluorite has been previously used to generate discrimination diagrams for deposit types. Schneider et al. (1975) and Möller et al. (1976) created logarithmic abundance ratio diagrams of REE and calcium in fluorite involving genetic terminology. These papers used atomic ratios of Tb/La and Tb/Ca to establish reference lines between genetic fields for “pegmatitic”, “hydrothermal”, and “sedimentary” fluorite. These boundaries were used by many authors (e.g. Ekambaram et al., 1986; Koç and Reçber, 2001; Sánchez et al., 2010; Schönenberger et al., 2007; Schwinn and Markl, 2005; Subías and Fernández-Nieto, 1995) to assign origin to specific deposits or fluorite districts. Gagnon et al. (2003) questioned universal validity of these diagrams. Tümenbayar (1996) developed a discrimination diagram based on logarithmic concentrations of Y versus Yb measured in ppm to classify Mongolian fluorite deposits into four categories. This diagram defined fields for epithermal, hydrothermal, pegmatitic, and magmatic fluorite deposits. Koç et al. (2003) modified the fields established by Tümenbayar (1996); however, the basis for this modification is not clear. Herein we use Tb/La - Tb/Ca and Y - Yb diagrams to plot all available fluorite data that can be assigned to carbonatite, peralkaline-alkaline, Mississippi Valley Type (MVT), and vein (sedimentary- igneous- and metamorphic-hosted) deposits.

4. Compilation diagrams

The database to construct Figures 1 to 3 was compiled from publications covering an array of deposit types worldwide. It contains more than 1000 entries of REE concentrations in fluorite. The deposit type for each entry was determined from the geological information in its source document; only entries clearly identified as carbonatite, peralkaline-alkaline, MVT, or vein type were used to construct the discrimination diagrams.

Most of the data represent analyses by inductively coupled plasma mass spectroscopy (ICP-MS) or neutron activation, although some laser ablation ICP-MS results are included.

Vein-type mineralization was subdivided according to its host rock (sedimentary, igneous, or metamorphic). Carbonatite data were also divided in Figure 2 to represent fluorite in carbonatite-related fluorite deposits and fluorite from carbonatites explored for REE or Nb. The compiled data were superimposed on the modified Tb/La - Tb/Ca diagram of Möller et al. (1976). Positions of the original Möller et al. (1976) reference lines dividing the diagram into “pegmatitic”, “hydrothermal”, and “sedimentary” fields were recalculated to ppm values for ease of use by the mineral exploration community and are shown on all Tb/La - Tb/Ca plots for reference.

5. Results

Carbonatite-related fluorite data define a field that is aligned along, but lies mainly below, the pegmatite-hydrothermal reference line (Fig. 1a). The field of fluorite from MVT deposits plots on both sides of the hydrothermal-sedimentary reference line and the highest density of samples straddles this boundary (Fig. 1b). Peralkaline-alkaline related fluorite data are scattered, but with the exception of one sample, all are above the hydrothermal-sedimentary reference line (Fig. 1c). Samples from fluorite-bearing veins hosted by sedimentary deposits plot entirely below the pegmatite-hydrothermal reference line (Fig. 1d). Data from fluorite veins hosted by igneous rocks (Fig. 1e) overlap those hosted by sedimentary rock, but extend farther below the hydrothermal-sedimentary reference line. Fluorite veins hosted by metamorphic rocks (Fig. 1f) define a field that overlaps the hydrothermal and sedimentary fields, with the main concentration of samples elongated and parallel to the hydrothermal-sedimentary reference line. Figures 2 and 3 depict distinct fields for fluorite from carbonatite and MVT deposits for Tb/La - Tb/Ca and Y - Yb diagrams respectively.

6. Discussion

Distinct compositional fields were produced representing REE signatures in fluorite for carbonatite and MVT deposits (Figs. 2, 3).

Fluorite from peralkaline-alkaline settings define a broad field that lies above the sedimentary-hydrothermal reference line (Fig. 1c); and comparison between Figs. 1a and 1c demonstrate a compositional overlap between fluorites from carbonatite and peralkaline-alkaline settings.

Data from fluorite-bearing veins (Figs. 1d-f) scatter across the central parts of the Tb/La - Tb/Ca diagrams, but do not extend as far into the low Tb/La and high Tb/Ca areas as the carbonatite and peralkaline-alkaline data (Figs. 1a, c). Similarly the vein data do not extend as far into the high Tb/La and low Tb/Ca parts of the diagram as the MVT fluorite compositions. The extensive compositional overlap between fluorite-bearing veins cutting sedimentary, igneous and metamorphic rocks (Figs. 1d-f) indicates that chemistry of the local host rock is not the main factor controlling REE concentration in fluorite.

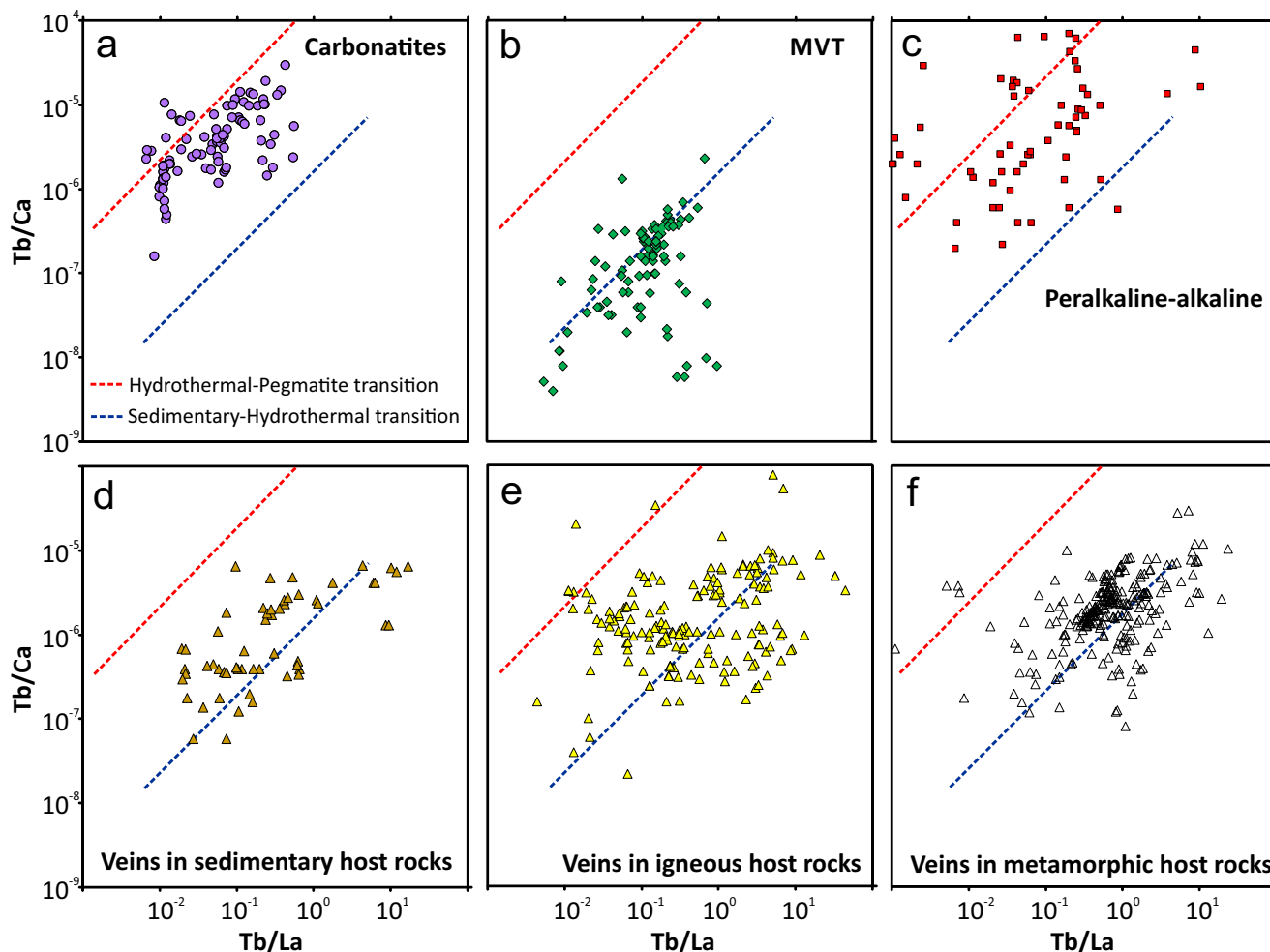


Fig. 1. Tb/La-Tb/Ca plots for fluorite (modified from Möller et al., 1976) with ratios calculated directly from Tb, La, and Ca concentrations in ppm. **a)** Carbonatite-related fluorite (from: Alvin et al., 2003; Bühn et al., 2003; Palmer and Williams-Jones, 1994; Santos et al., 1996; Xu et al., 2012). **b)** Mississippi Valley Type (MVT) fluorite (from: Bau et al., 2003; Hill et al., 2000; Levesse et al., 2006; Naldrett et al., 1987; Schneider et al., 1975; Souissi et al., 2010). **c)** peralkaline-alkaline rock-related fluorite (from: Gagnon et al., 2003; Hill et al., 2000; Koç et al., 2003; Minuzzi et al., 2008; Pekov et al., 2009; Schönerberger et al., 2008). **d)** Sedimentary rock-hosted fluorite-bearing veins (from: Hill et al., 2000; Koç and Reçber, 2001; Schwinn and Markl, 2005). **e)** Igneous rock-hosted fluorite-bearing veins (from: Ackerman, 2005; Castorina et al., 2008; Czaja et al., 2012; Dill et al., 2011; Hill et al., 2000; Levesse et al., 2006; Lüders et al., 2008; Monecke et al., 2002; Pinto-Coelho et al., 1999; Sallet et al., 2005; Schwinn and Markl, 2005). **f)** Metamorphic rock-hosted fluorite-bearing veins (from: Castorina et al., 2008; Cheilietz et al., 2010; Ekambaram et al., 1985; Lüders et al., 2008; Monecke et al., 2000; Schwinn and Markl, 2005). The red and blue dashed lines are the recalculated boundaries between the hydrothermal and pegmatite, and sedimentary and hydrothermal fields as defined by Möller et al. (1976).

A review of the relationships between the temperature of homogenization and composition of fluids relative to results of LA-ICPMS data on fluorite from the same samples might determine if the distribution of the data on the Tb/La - Tb/Ca diagrams reflects a trend of increasing fluorite crystallization temperatures moving from the lower right (high Tb/La and low Tb/Ca) to the upper left (low Tb/La and high Tb/Ca). Fluid-rock interaction is important at elevated temperatures as there is a general trend of increased solubility of REEs with increasing temperature (Wood, 1990; Williams-Jones et al., 2012). The increasing solubility is related to REE speciation, with F complexes predominant relative to Cl complexes in

experimental studies up to 250°C. Sulfate, carbonate, and phosphate complexes may also be important (Williams-Jones et al. 2012), but have not been investigated experimentally. Another important geochemical factor influencing REE transport and deposition is pH with decreasing acidity (and chlorinity) driving REEs out of solution (Williams-Jones et al., 2012).

The Y - Yb diagram (Fig. 3), represents the most straightforward method for the exploration industry to distinguish between fluorite grains from carbonatite and MVT settings in stream sediments and till. Consideration of additional elements, besides REEs, will probably be required

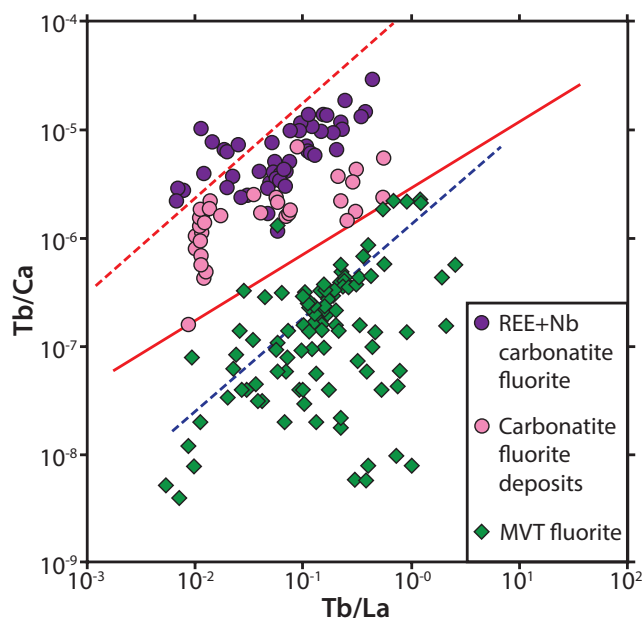


Fig. 2. Tb/La-Tb/Ca diagram showing compositional data for fluorite from carbonatite-related and MVT deposits. The solid red line defines the boundary between MVT-related fluorite and carbonatite-related fluorite. The carbonatite data are bicolored, representing fluorite found in carbonatite-related fluorite deposits and fluorite from carbonatites explored for REE or Nb. Information sources and reference lines as in Figures 1a and 1b.

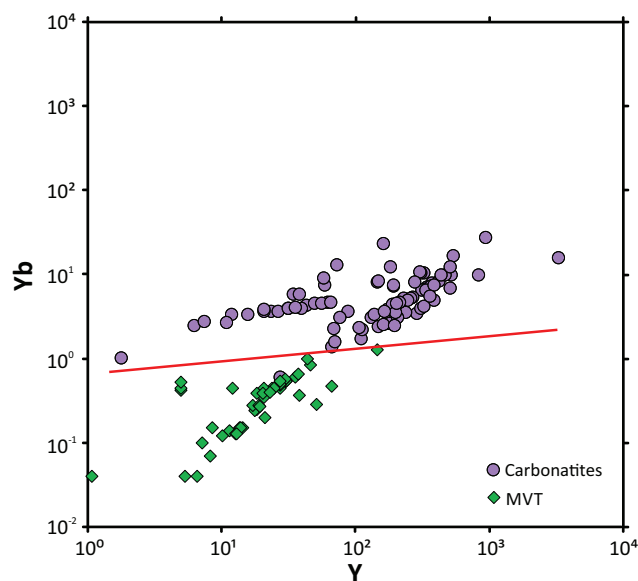


Fig. 3. Y-Yb plot for fluorite from carbonatite and MVT deposits. The carbonatite-related fluorites define a trend with a shallow positive slope and have higher concentrations of Yb than fluorites from MVT deposits with the exception of one sample. The solid red line defines the boundary between MVT-related fluorite and carbonatite-related fluorite. Information sources and reference lines as in Figures 1a and 1b.

to produce simple, user-friendly, fluorite-based, discrimination diagrams for indicator mineral studies.

7. Conclusion

Fluorite is a potential proximal indicator mineral for carbonatite-related specialty metal mineralization. Tb/La - Tb/Ca and Y - Yb diagrams may be used to distinguish between fluorite from carbonatite-related and MVT deposits; however, significant overlap exists between compositional fields of fluorites from sedimentary, igneous, and metamorphic rock hosted vein deposits. This indicates that REE concentration in fluorite is largely independent of the composition of the host rock. There is also significant overlap between compositions of fluorites from carbonatites, peralkaline-alkaline intrusions, and veins. Further refinements of these preliminary diagrams through data contouring and the production of probability plots are required. Effort is underway to analyze fluorite from key Canadian and foreign deposits using EMP and LA-ICPMS, and to create a complementary set of discrimination diagrams incorporating slopes of chondrite-normalized plots and Ce and Eu anomalies. Consideration of additional elements, besides REEs, will be investigated to produce user-friendly, fluorite-based, discrimination diagrams for indicator mineral studies.

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