ARTICLE IN PRESS

Chemical Geology xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Chemical Geology

journal homepage: www.elsevier.com/locate/chemgeo

Characterizing fluids associated with the McArthur River U deposit, Canada, based on tourmaline trace element and stable (B, H) isotope compositions

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ARTICLE INFO

Keywords:
Magnesio-foitite
Hydrothermal activity
REE
Boron isotopes
LA-ICPMS
SHRIMP
Hydrogen isotopes
P2 fault

ABSTRACT

The origin and nature of fluids responsible for U deposits of the Athabasca Basin have been in debate. This study presents the trace element abundances and stable isotope ratios (D/H and $^{11}B/^{10}B$) of well characterized tourmaline samples determined by in situ analytical techniques, in order to evaluate the nature of fluids during the mineralization of the McArthur River U deposit, the world's largest high-grade U deposit. Magnesio-foitite, an alkali-deficient Mg-rich tourmaline, is abundant along the 13-km long P2 fault, a reverse structure that controls the location of the McArthur River deposit. Magnesio-foitite contains variable concentrations of rare earth elements (REEs, up to 46 ppm in total) and Y (up to 35 ppm). Chondrite normalized patterns indicate low light REEs (LREEs) relative to heavy REEs (HREEs) ($Ce_N < Y_N$, Y as a proxy for HREEs) and slight negative Eu anomalies, similar to those of uraninite from the deposit. The data combined with textural evidence suggest that magnesio-foitite and uraninite co-crystallized with LREE-rich aluminum phosphate sulfate minerals. Relatively high REE contents of magnesio-foitite in the ore zone indicates the incursion of a REE-rich basement fluid. Low values of δD (− 98 to − 41‰) for magnesio-foitite suggests that the mineralizing fluid originated from groundwater. Variably high values of $\delta^{11}B$ (+ 13.1 to + 23.2‰) are explained by the dissolution of B from carbonate or evaporitic rocks and preferential removal of ^{10}B by the crystallization of illite and kaolinite. The results of this study support the ascent of a REE-rich basement fluid during mineralization and extensive modification of basinal fluids through the crystallization of clay minerals.

1. Introduction

Tourmaline is a robust mineral in evaluating the composition and origin of hydrothermal fluids as it can accommodate a wide range of elements. Trace element abundances in tourmaline provide insights into hydrothermal and magmatic processes (e.g., Jolliff et al., 1987; Taylor et al., 1999; Slack and Trumbull, 2011; Čopjaková et al., 2013, 2015). For example, tourmaline composition can be used to evaluate the evolution of melts and the co-crystallization of phases in pegmatite (Čopjaková et al., 2013).

Tourmaline occurs as an accessory mineral in unconformity-type U deposits of the Athabasca Basin in northern Saskatchewan, Canada. Unconformity-type U deposits account for > 33% of the world's U resources (Jefferson et al., 2007) and the Athabasca Basin hosts very large high-grade U deposits, including the world-class McArthur River deposit. These deposits form by the reduction of U^{6+}-bearing oxidized fluids at the unconformity between sedimentary basins and underlying crystalline basement rocks. However, the source and nature of mineralizing fluids is debated. The prevailing model for Athabascan deposits suggests mixing of oxidizing, highly saline (25–35 wt% NaCl equiv.), basinal water with a reduced fluid derived from the basement or produced through the interaction of basinal fluids with the basement (e.g., Hoeve and Sibbald, 1978, Bray and Spooner, 1988, Kotzer and Kyser, 1995, Fayek and Kyser, 1997). The high salinity of the basinal fluid is attributed to the dissolution of evaporites (e.g., Bray and Spooner, 1988; Kotzer and Kyser, 1995) or the evaporation of seawater (e.g., Richard et al., 2010, 2012, 2013).

Several papers in the past 20 years have examined the geochemistry of tourmaline associated with U deposits of the Athabasca Basin, in order to determine the source and nature of fluids (e.g., Kotzer and Kyser, 1995; Mercadier et al., 2012; Adlakha and Hattori, 2016). The tourmaline was originally reported as dravite (e.g., Hoeve and Sibbald, 1978; Kotzer and Kyser, 1995), but later studies show that it is alkali-deficient and Mg-rich, similar to magnesio-foitite in composition (e.g., Rosenberg and Foit, 2006; Ng et al., 2013; Adlakha and Hattori, 2016). Adlakha and Hattori (2016) observed that the tourmaline is similar in

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http://dx.doi.org/10.1016/j.chemgeo.2017.06.030
Received 4 December 2015; Received in revised form 19 June 2017; Accepted 22 June 2017
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composition along ore-bearing faults and suggested that the basement structures provided the conduits for U mineralizing fluids. Kotzer and Kyser (1995) suggested that the fluids were mixtures of basinal and basement fluids at ~200 °C, based on O and H isotope compositions of tourmaline. The authors suggested Mg was supplied by basement fluids and B from basinal fluids that leached B from clays and evaporites of the Athabasca Basin. In contrast, Mercadier et al. (2012) suggested that evaporated seawater supplied Mg and B for tourmaline based on high $\delta^{11}B$ values of magnesio-foitite.

Although the major element abundance and stable isotope compositions (B, O, H) of tourmaline have been investigated in previous studies, its trace element abundance has not yet been reported. In order to evaluate the nature and source of fluids responsible for U mineralization, this paper reports the abundance of trace elements, as well as B and H isotope compositions, for well-characterized tourmaline samples in proximity, and distal, to the McArthur River U deposit. For comparison, the trace element abundances and B isotope compositions of pre-Athabasca hydrothermal tourmaline, oxy-dravite, were also determined and presented in this paper.

2. Geological context

Unconformity-type U deposits within the Athabasca Basin are spatially associated with the unconformity between the Athabasca Group sandstones and underlying crystalline basement rocks of the Rae and Hearne provinces (Fig. 1). The Hearne Province in the area comprises four domains: Peter Lake, Virgin River, Mudjatik and Wollaston. Most U deposits, including the McArthur River deposit, occur within, or overlying, the metasedimentary rocks of the western Wollaston Domain near the Archean granitoid gneisses of the Mudjatik Domain. The Wollaston Domain is a NE-striking fold-thrust belt. It comprises an Archean granitoid basement and overlying Paleoproterozoic metasedimentary rocks of the Wollaston Group (Lewry and Sibbald, 1977). The Wollaston Group includes a series of graphitic and non-graphitic pelite, semi-pelite and paragneiss interlayered with minor calc-silicate, arkose and quartzite (McGill et al., 1993). These rocks are locally intruded by pegmatite lenses which formed by anatexis of the metasedimentary rocks during the Trans-Hudson Orogeny (~1.8 Ga; e.g., Annesley et al., 2005).

Sedimentation of the Athabasca Basin initiated after 1.75 Ga (Kyser et al., 2000) and continued until approximately 1.54 Ga (Creaser and Stasiuk, 2007). Basal sedimentary rocks of the Athabasca Group consist of fluvial conglomerate and sandstone of the Read (Fig. 1) and Fair Point Formations (Ramaekers, 1990). These units are overlain by fluvial to shallow marine quartz arenite, siltstones and phosphatic mudstone of the Manitou Falls, Lazenby Lake, Wolverine Point, Locker Lake, Otherside and Tuma Lake Formations. Capping the western portion of the basin are fine sandstones and shales of the Douglas Formation, and stromatolite and dolostone of the Carswell Formation (Fig. 1). The thickest preserved portion of the basin is 1500 m; however, fluid inclusion studies by Pagel et al. (1980) suggest that the basin may have reached 5 to 7 km in thickness.

The least altered basement rocks in proximity to the McArthur River deposit consist mainly of garnet ± cordierite paragneiss and graphitic pelite, which are intruded by granitic pegmatite of centimetres to a few metres in width (McGill et al., 1993). A quartzite unit occurs in proximity to the McArthur River deposit. The quartzite formed a paleotopographic high during the deposition of Athabasca sandstones, as the unit is resistive against weathering and erosion. The Athabasca Group in this area is approximately 500 m thick and comprises four quartz-rich sandstone units: the basal Read Formation (previously the Manitou Falls Formation A), and the three overlying members of the Manitou Falls Formation, including the conglomeratic Bird (MFb), sandy Collins (MFc) and uppermost clay-intraclast-bearing Dunlop (MFd) members

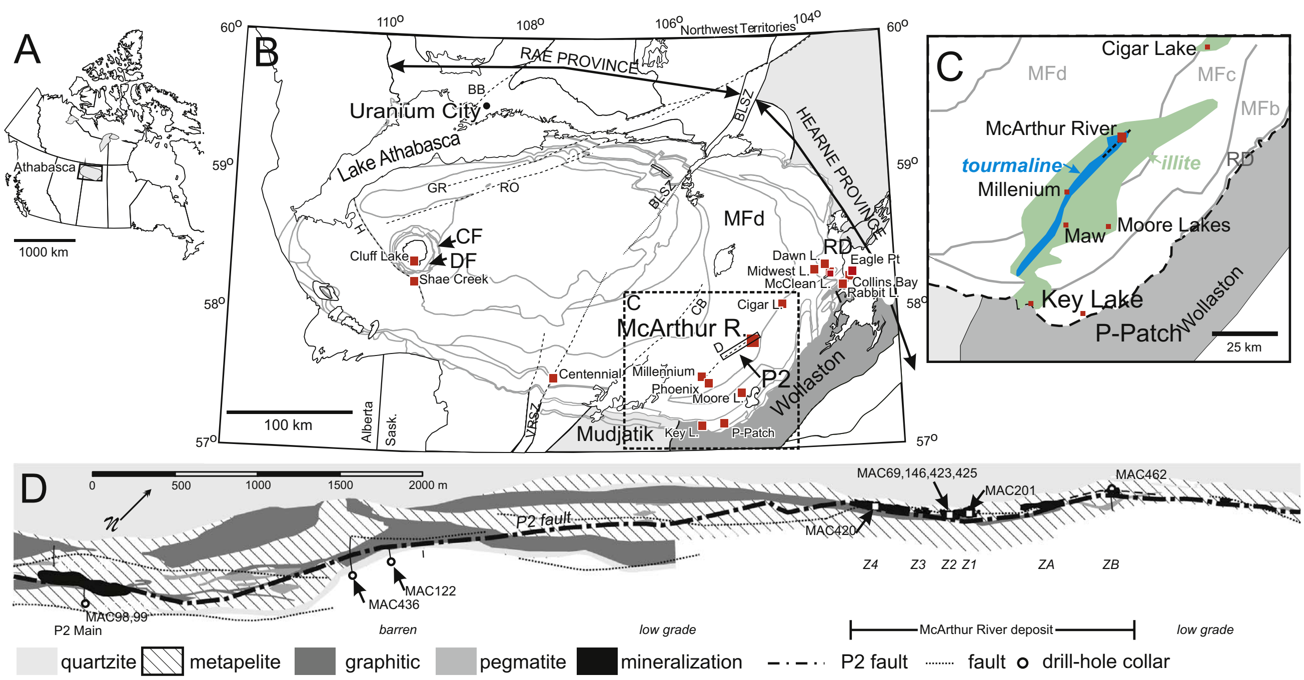

Fig. 1. A: An image of Canada shows the location of the Athabasca Basin. **B**) A map showing the location of the major U deposits and structures in the Athabasca Basin (modified from Jefferson et al., 2007 and Adlakha and Hattori, 2015, 2016). The units of the Athabasca Basin pertinent to this study are labelled: the Read Formation (RD) sandstone, the Dunlop Member of the Manitou Falls Formation (MFd), the Douglas Formation (DF), and the Carswell Formation (CF). The south-eastern margin of the Athabasca Basin is underlain by the Wollaston (dark grey) and Mudjatik Domains (light grey) of the basement. The locations of maps C and D of this figure are also indicated. Major shear zones: BB = Black Bay, BLSZ = Black Lake, CB = Cable Bay, GR = Grease River, H = Harrison, RO = Robbilard, VRSZ = Virgin River. **C**) A regional map in the eastern Athabasca Basin showing the locations of illite-rich (green) and tourmaline-rich (blue) zones, which envelope the major U deposits (data from Earle et al., 1999, and map modified from Jefferson et al., 2007). MFb and MFc = the Bird and Collins members of the Manitou Falls Formation, respectively. **D**) A plan view map showing the basement lithology at the unconformity and sample locations. Drillholes collared at surface are represented by open circles. Drillholes collared underground (i.e., within the McArthur River mine) are indicated by open squares. Modified from Adlakha and Hattori (2015, 2016). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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