



Noble gases (Ar, Kr, Xe) and halogens (Cl, Br, I) in fluid inclusions from the Athabasca Basin (Canada): Implications for unconformity-related U deposits



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ABSTRACT

The formation of unconformity-related uranium deposits in the Proterozoic Athabasca Basin (Canada) involved basin-scale circulation of U-bearing brines during high-grade diagenesis (150–200 °C) at ~1.6–1.5 Ga. The UO₂ ores occur both sides of the unconformity and are associated with extensive brecciation and illite–sudoite–dravite alteration. Quartz and dolomite cementing veins and breccias are associated with alteration and mineralisation and contain a fairly uniform population of fluid inclusions characterised by variable Na:Ca and salinities of 25–35 wt.% salts and high U concentrations of up to 600 ppm U. In order to further constrain the origin of these U-rich brines, we analysed the naturally occurring isotopes of Ar, Kr and Xe, together with halogens (Cl, Br and I), K, Ca and U in irradiated quartz and dolomite samples containing representative fluid inclusions. This was achieved by the noble gas method for halogen measurement (extended ⁴⁰Ar–³⁹Ar methodology) using a combination of noble gas extraction techniques.

The fluid inclusions opened by crushing quartz and dolomite samples in vacuum have similar molar Br/Cl ratios of 5.8×10^{-3} to 10.4×10^{-3} , and molar I/Cl ratios of 1.8×10^{-6} to 8.2×10^{-6} . These compositions lie over the top half of the modern-day seawater evaporation trajectory, consistent with the fluids deriving the bulk of their salinity by subaerial evaporation of seawater, beyond the point of halite saturation. The I/Cl ratios are much lower than is typical of fluids that have interacted with I-rich organic matter present in many sedimentary basins or fluid inclusions found in Mississippi Valley type (MVT) Pb–Zn ore deposits. This is significant because provided the U-rich fluid inclusions are representative of the ore-stage fluids, the low I/Cl ratios of the fluid inclusions do not favour fluid interaction with organic matter (or hydrocarbons), as a major process for localising U mineralisation.

The majority of samples contain fluid inclusions with age-corrected ⁴⁰Ar/³⁶Ar of between the modern atmospheric value of ~300 and 450. These values are considered representative of the fluid's initial composition and are typical of upper crustal sedimentary formation waters. The fluid inclusions non-radiogenic ⁸⁴Kr/³⁶Ar and ¹²⁹Xe/³⁶Ar ratios are slightly enriched in ¹²⁹Xe relative to air and the fluid inclusions are estimated to contain $0.5\text{--}17.3 \times 10^{-10} \text{ mol g}^{-1} \text{ } ^{36}\text{Ar}$ which is up to twenty times the ³⁶Ar concentration of air-saturated seawater. The data are interpreted to reflect acquisition of atmospheric noble gases from sedimentary rocks and suggest acquisition of radiogenic ⁴⁰Ar within K-rich basement rocks, that would have been an important source of excess ⁴⁰Ar, was limited by temperatures of less than 200 °C.

Taken together the halogen and noble gas composition of the U-bearing fluid inclusions are strongly controlled by subaerial evaporation and subsequent interaction with sedimentary rocks, showing that low temperature evaporitic brines dominated the mineralising system. Mineralisation is unlikely to have been triggered by fluid interaction with organic matter, or mixing with voluminous basement-derived fluids; however, the data do not completely preclude a role for volumetrically minor fluid or gas phases introduced by deep-seated basement faults preferentially located at the sites of mineralisation.

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1. Introduction

Spectacularly high grade and large tonnage deposition of uraninite (up to 200 kt of U at 20% U on average at McArthur River) has occurred in sedimentary rocks close to the basement unconformity of the Paleoproterozoic Athabasca Basin (Saskatchewan, Canada) (Jefferson et al., 2007; Kyser and Cuney, 2008). These occurrences attest to protracted basin-scale circulation of U-bearing fluids through sedimentary and basement rocks concurrent with sediment diagenesis at temperatures of 150–200 °C (e.g. Hoeve and Sibbald, 1978; Kotzer and Kyser, 1995; Derome et al., 2005; Boiron et al., 2010; Richard et al., 2012).

This style of “unconformity-related” uranium mineralisation is unique to a restricted number of Proterozoic basins including the Athabasca and Thelon basins of Canada (e.g. Renac et al., 2002; Beyer et al., 2011) and the Kombolgie Basin of Northern Australia (Derome et al., 2003a, 2007; Polito et al., 2011). However, comparable processes may have been responsible for the famous Oklo U deposits in the Franceville Basin (Gabon), which on account of their high grades and slightly greater age formed natural nuclear reactors at ~2 Ga (e.g. Shukolyukov et al., 1976; Mathieu et al., 2000; Meshik et al., 2004; Boiron et al., 2010). The occurrence of these deposits may be restricted because their genesis is facilitated by the presence of a U-rich basement (10–100 ppm U), such as underlies many Paleoproterozoic terranes (Cuney, 2010). Furthermore, high porosity, kilometre-thick quartz sandstones, that are common in the basal section of the mineralised basins, acted as essential reservoirs for voluminous saline fluids, that enabled efficient fluid convection, suitable for transport of oxidised U(VI) from basement and/or sedimentary sources to the sites of deposition (Kyser et al., 2000).

The UO₂ ores in the Athabasca Basin are closely associated with intense illite–sудоite–dravite alteration in both basin and basement rocks and multiple generations of quartz and dolomite that form sandstone silicifications as well as veins and breccia cements (e.g. Hoeve and Sibbald, 1978; Pagel et al., 1980; Kotzer and Kyser, 1995; Derome et al., 2005). Similar high salinity brine inclusions have been found in sandstone silicifications, quartz and dolomite vein and breccia cements as well as secondary fluid inclusions planes in basement magmatic and metamorphic quartz from both mineralised and barren altered areas and have been implicated as the dominant fluid involved at the time of alteration and mineralisation by several workers (e.g. Pagel et al., 1980; Kotzer and Kyser, 1995; Derome et al., 2005; Mercadier et al., 2010; Richard et al., 2010, 2012).

The origin of the brines and the fluid/rock interactions they underwent have been assessed by a variety of techniques applied to fluid inclusions and their host minerals (quartz and dolomite) in a number of deposits including those studied here. Microthermometry, LA-ICP-MS and synchrotron-SRF and XANES analyses show that the brines have temperatures of 150–200 °C, salinities of 25–35 wt.% salts, variable Na:Ca ratios with NaCl-rich (Na > Ca > Mg > K) and CaCl₂-rich (Ca > Mg > Na > K) end-members, and that the fluid inclusions contain up to 600 ppm U(VI) (e.g. Pagel et al., 1980; Kotzer and Kyser, 1995; Derome et al., 2005; Richard et al., 2010, 2012, 2013a). Quartz and dolomite vein and breccia cements have been investigated isotopically for their O isotopic composition and these data show that quartz- and dolomite-forming fluids (trapped as fluid inclusions) are isotopically similar to the fluids responsible for deposit scale illite–sудоite–dravite alteration (Richard et al., 2013b). Furthermore, the fluid inclusions halogen (Cl, Br) composition (obtained by crush-leach ion chromatography and LA-ICP-MS analysis of individual fluid inclusions); the Cl isotope composition of fluid inclusion leachates; and the B isotope composition of dravite have been investigated. These studies favour subaerial evaporation of seawater

as the dominant mechanism for generating the fluids high salinities (Richard et al., 2011; Leisen et al., 2012; Mercadier et al., 2012).

Despite these powerful constraints on the nature of the ore fluids, there remain several open questions, that will be addressed by the current study which extends the analysis of halogens in the fluid inclusions, to include I, and combines the measurements of halogens with noble gases. Firstly, it is debatable if NaCl-rich and CaCl₂-rich brines represent distinct fluids, derived for example by evaporation of seawater to different degrees, or if the Ca–Na–Mg–K variation simply reflects variable fluid–rock interactions. Secondly, the basement rocks represent a potential U source (Hecht and Cuney, 2000; Mercadier et al., 2013), but the extent to which basinal fluids were convected into the basement and equilibrated with basement rocks is poorly constrained. Thirdly, the mechanism for the reduction of U(VI) to U(IV) and subsequent precipitation of UO₂ is still contentious. Fluid–rock interaction, fluid–organic matter interaction and mixing with deeply sourced fluids are alternatively invoked (e.g. Hoeve and Sibbald, 1978; Wilson and Kyser, 1987; Alexandre and Kyser, 2006).

The noble gases can provide new insights on fluid interaction with basement rocks, or the presence of deeply sourced fluid components because old K-rich basement rocks, such as the 1.8 Ga to Archaean age rocks underlying the Athabasca Basin (Card et al., 2007) would be expected to have high ⁴⁰Ar/³⁶Ar ratios (e.g. Kendrick et al., 2011a,b). In contrast, the relatively K-poor overlying sediments (deposited from 1.76 Ga; Ramaekers et al., 2007) that contained air saturated waters and trapped atmospheric noble gas components (e.g. Podosek et al., 1980) are more likely to preserve low ⁴⁰Ar/³⁶Ar values. Iodine is an essential element for life and its incorporation into organic matter means that fluid I/Cl ratios are a sensitive indicator for fluid interaction with organic matter (Kendrick et al., 2011c). The measurement of noble gases (Ar, Xe, Kr) and halogens (Cl, Br, I) by extended ⁴⁰Ar/³⁹Ar methodology provides simultaneous analysis of K, Ca and U (Kendrick, 2012), meaning noble gas and halogen signatures can be closely related to the fluids U and cation content (indicative of fluid–rock interaction). Finally, determination of Cl and Br, with I, by this methodology, provides an independent confirmation of ion chromatographic and LA-ICP-MS Br and Cl analyses (Richard et al., 2011; Leisen et al., 2012) and additional plots that include I for assessing the extent to which the data ‘fit’ the suggested fluid origin by seawater evaporation.

2. Geological setting, sampling and fluid inclusions

2.1. The Archaean to Paleoproterozoic basement

The Athabasca Basin overlies basement rocks of the Archaean to Paleoproterozoic Western Churchill Province which is further divided into the Rae Subprovince in the west and the Hearne Subprovince in the east by the Northeast-trending Snowbird tectonic zone (Hoffman, 1990; Card et al., 2007) (Fig. 1). The basement lithologies comprise Archaean gneisses, Paleoproterozoic metapelites and mafic to felsic intrusions, that were all strongly metamorphosed during the ~2.0–1.9 Ga Thelon–Talston orogeny in the west, and the ~1.9–1.8 Ga Trans-Hudson orogeny in the east (Chiarenzelli et al., 1998; Annesley et al., 2005; Card et al., 2007). Most of the unconformity-related uranium deposits in the Athabasca Basin are located in the vicinity of the Wollaston–Mudjatik transition zone (WMTZ) that separates the Wollaston and Mudjatik lithostructural domains of the Hearne Subprovince (Annesley et al., 2005; Fig. 1).

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