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Radiolytic (H_2, O_2) and other trace gases (CO_2, CH_4, C_2H_6, N_2) in fluid inclusions from unconformity-related U deposits

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Abstract

Fluid inclusions from quartz and dolomite veins from five unconformity-related uranium deposits (Athabasca basin, Canada) have been analyzed by Raman spectrometry in order to identify trace gases in their vapor phase at room temperature. About 80% of fluid inclusions have detectable gases. The most common gases are H_2, O_2, CO_2, CH_4, C_2H_6 and N_2. So-called “NaCl-rich” and “$CaCl_2$-rich” brine inclusions have similar gas contents. U-bearing fluid inclusions have similar gas compositions when compared to U-absent fluid inclusions. Radiolysis (i.e. production of H_2 and O_2 from H_2O when fluids have been in contact with previously deposited UO_2) and fluid-rock interactions are the most probable origins for H_2, O_2 and CO_2, CH_4, C_2H_6 and N_2 respectively. The relative abundance of radiolytic O_2 is related to distance to ore and could be used as vectors towards mineralization. Other trace gases may be indicative of ore-forming processes, specifically of fluid-rock interaction and UO_2 deposition.

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Keywords: Raman; fluid inclusion; uranium deposit; gas; radiolysis; Athabasca basin

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1. Introduction

Trace gases in fluid inclusions have been widely documented around high-grade unconformity-related U deposits in the Athabasca basin (Canada). H_2 and O_2 have been interpreted as resulting from water radiolysis in contact with UO_2 ore[1-3]. Differential migration of H_2 and O_2 has been demonstrated for the McArthur River deposit, as fluid inclusions located at more than 10m from high-grade ore show H_2 while O_2 is not detected[3]. Other trace gases (CO_2, CH_4, C_2H_6, N_2) have a more debatable origin; water-rock interaction in graphite-bearing meta-pelites being the most frequently favored hypothesis[4-7]. Most authors acknowledge the critical importance of radiolytic and other trace

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gases as potential prospecting tools and as evidence of fluid-rock and fluid-gas interaction possibly at the origin of U^{VI} reduction in the ore-forming brines and UO_2 deposition[3,7,8]. Here, I provide new gas compositions of the vapor phase of fluid inclusions at room temperature for four U deposits (Shea Creek, Rabbit Lake, Eagle Point, P-Patch), using samples located at more than 10m from high-grade ore, as a complement to previously published data on McArthur River[3]. In addition, I provide gas composition for fluid inclusions from McArthur River in which U concentrations have been determined by LA-ICPMS[9,10]. The new data provide for the first time an overview of gas compositions in fluid inclusions from U deposits throughout the Athabasca basin.

2. Geological background, sampling and methods

The <1.75 Ga Athabasca basin unconformably overlies a 2.0-1.8 Ga basement. The current maximum thickness of the sedimentary cover (mostly sandstones and siltstones) is ~1.5 km and could have originally been as much as ~5 km. The U deposits are generally located near the basement/cover interface, and structurally controlled by sub-vertical faults rooted in graphite-rich basement meta-pelites. UO_2 ores have been dated to between 1.6 and 1.4 Ga and successive late episodes of mineralization, and/or recrystallisation occurred until ~0.7 Ga[11]. Sandstone silicification, and quartz and dolomite veins are spatially and temporally associated with the main clay alteration minerals and UO_2 ores. Hence, they are likely to host fluid inclusions that are relicts of the mineralizing fluids. The veins crosscut various lithologies such as meta-pelites and pegmatoids as well as basin sandstones. Veins were sampled from 4 deposits (Shea Creek, Rabbit Lake, Eagle Point, P-Patch) from various parts of the basin, in relatively fresh to extensively altered rocks. Prior to this study, fluid inclusions have been characterized petrographically and by microthermometry and similar fluid inclusions in the same samples were analysed destructively by LA-ICPMS[12]. Trace gases composition of fluid inclusions from McArthur River have been published previously[3] and corresponding published U concentrations in fluid inclusions determined by LA-ICPMS are presented here[9,10]. Primary and pseudosecondary fluid inclusions show two types of 100-200°C basinal brines of evaporated-seawater origin, showing evidence for mixing: a “NaCl-rich brine” end-member (Cl>Na>Ca>Mg>K) and a “$CaCl_2$-rich brine” end-member (Cl>Ca≈Mg>Na>K)[9,12-14].

Gas species in the vapor phase of fluid inclusions were determined at room temperature with a Labram Raman microspectrometer equipped with a Edge filter, a holographic grating with 1800 grooves per millimetre and a liquid nitrogen cooled CCD detector at GeoRessources (Nancy, France)[15]. The exciting radiation at 514.5nm provided by an ionized Argon laser (Spectra physics) was focused on the vapor phase of the fluid inclusions using a X80 objective (Olympus). Using this setup, neither limits of detection nor absolute concentrations of trace gases, can be determined. However the relative proportions of the different gas species at room temperature can be estimated qualitatively[15] (Fig. 1a,b).

3. Results and discussion

Eighty one fluid inclusions were analyzed (Shea Creek: N = 28, Rabbit Lake: N = 17, Eagle Point: N = 16 and P-Patch: N = 20) and about 20% show no detectable gas in their vapor phase. H_2 is the most frequently detected gas (in ca. 60-80% of gas-bearing inclusions), followed by CH_4 (ca. 35-80%), N_2 (ca. 20-60%) and CO_2 (ca. 5-50%) (Fig. 1d). C_2H_6 was detected only at Eagle Point (ca. 30%) and O_2 was never detected (Fig. 1c). CO_2 is remarkably frequent at Rabbit Lake (ca. 50%) compared to the other deposits (5-10%). There is no significant difference of gas compositions between fluid inclusions classified as “NaCl-rich brines” and “$CaCl_2$-rich brines”. When H_2 and C_2H_6 are taken separately (as H_2 is thought to originate from radiolysis and C_2H_6 is minor), fluid inclusions are dominated by CH_4-N_2 (CH_4 being predominant in most cases). A minority of fluid inclusions have detectable CO_2 together with CH_4-N_2 or CO_2 only (Fig. 1e). Fluid inclusions from McArthur River, classified as “NaCl-rich brines” and “$CaCl_2$-rich brines”, in which U concentration was detected and quantified by LA-ICPMS commonly have H_2 and O_2, with rarer CH_4 and C_2H_6, and no CO_2 and N_2 (Table 1). Fluid inclusions from Shea Creek, Rabbit Lake, Eagle Point and P-Patch differ from those from McArthur River in that they frequently contain N_2 and are devoid of O_2 (Fig. 1d)[3]. Alteration of NH_4-bearing feldspar and biotite in the basement and formation of NH_4-bearing micas in the clay alteration haloes are probably the dominant control on the N_2 concentration in the fluid inclusions[6]. The difference of N_2 composition between McArthur River and the other deposits may be attributed to subtle changes in biotite, feldspar and mica compositions that would require further investigation. The absence O_2 in fluid inclusions from the other deposits compared to those of McArthur River is attributable to the distance between samples and high-grade