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Geochemistry and chemical dating of uraninite in the Samarkiya area, central Rajasthan, northwestern India – Implication for geochemical and temporal evolution of uranium mineralization

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ABSTRACT

In the Samarkiya area, located at the central part of the Aravalli-Delhi Fold Belt (ADFB), uranium mineralization is hosted both by the basement Mangalwar Complex and the overlying supracrustal rocks of the Pur-Banera belt. The present study aims to appraise the geochemical and temporal evolution of uranium mineralization from the basement and the adjoining supracrustals in the Samarkiya area integrating textural features, geochemistry, and in situ U-Th-Pb_{Total} dating of uraninite.

Uraninite occurs as inclusions in the major rock forming minerals, viz. plagioclase, quartz, biotite, and chlorite. Based on the shape, location in the host mineral (well inside/at the grain boundary/along or connected to micro-cracks etc.) and association with other secondary uranium minerals, uraninites are classified into different groups, which are compositionally distinct, barring some exceptions. Integrating texture, geochemistry and in situ electron probe dating we propose that in addition to an old event at ~1.88 Ga in the basement rocks, there are two major events of uraninite formation at ~1.24–1.20 Ga and ~1.01–0.96 Ga in both the basement and supracrustal rocks. Although none of the pristine, unaltered uraninites that formed during the above mentioned events contain significant intrinsic minor or rare earth elements, the basement uraninites are consistently much enriched in thorium compared to those from the supracrustal. Based on the compositions, we propose that the basement uraninites formed from a high temperature magmatic/metamorphic fluid, whereas those in the supracrustal rocks crystallized from a low temperature, presumably oxidized fluid. Back-scattered electron images, X-ray elemental mapping of selected elements and EPMA spot analysis of large uraninite grains (both from the basement and the supracrustals) collectively demonstrate that subsequent to the major mineralizing event at ~1.24–1.20 Ga, the mineralized rocks were subjected to fluid-mediated alteration, which resulted in $\sum$REE + Y- and Si (Ca)-enrichment of existing ~1.24–1.20 Ga uraninites in the basement and supracrustal rocks, respectively. We cannot constrain the exact timing of this alteration event. However, as this event altered the ~1.24–1.20 Ga uraninites and as spot ages of the altered grains yield ages largely between ~1.24 and 0.96 Ga, it is reasonable to place this event between the second and third stages of uranium mineralization/mobilization at ~1.20 Ga and ~1.01 Ga, respectively. The last event that took place at ~1.01–0.96 Ga most likely represent an episode of recrystallization/alteration of existing uraninite leading to complete Pb-loss and resetting of the isotopic clock. However, we do not entirely reject the possibility of neo-mineralization.

The discrete events deciphered from uraninite in the Samarkiya area can also be broadly linked to some major magmatic-metamorphic events, identified from other independent studies, in the ADFB. For example, the earliest ~1.88 Ga event displayed by basement uraninite is most likely related to a pervasive magmatic-metamorphic event (~1.86–1.82 Ga) that affected the basement, whereas the last/latest event ~1.01–0.96 Ga can be linked to a pervasive metamorphic event that affected perhaps the entire ADFB. This last episode can also be linked to the tectono-thermal event related to the Rodinian amalgamation.

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The ~1.24–1.20 Ga event appears to be somewhat enigmatic in the context of well-known geological events in the area. However, based on some very recently published data, we interpret this to be a post-peak metamorphic (~1.37–1.35 Ga) hydrothermal event or even a new metamorphic event, hitherto unknown.

1. Introduction

The Proterozoic unconformity-related uranium deposits (e.g., Athabasca Basin, Canada; Kyser et al., 2000) constitute the third largest reasonably assured uranium resources of the world (IAEA, 2016). These very high grade (e.g., ~20% U_3O_8 at McArther deposit; Jefferson et al., 2007; Mercadier et al., 2013) large tonnage deposits, particularly those of Canada, collectively top the lowest cost category of the reasonably assured resources (IAEA, 2016). Consequently, Proterozoic unconformities are extensively explored as potential environs for uranium mineralization. Such deposits presently constitute ~9.95% of India's reasonably assured resources (IAEA, 2016). Some of the deposits associated with the Proterozoic rocks in India include those located in the Cuddapah basin (Rao et al., 2010; Deb and Pal, 2015; and references therein) and in the Singhbhum Shear Zone (Pal and Rhede, 2013) (Fig. 1a). The ~700 km long NNE-SSW trending Aravalli-Delhi Fold Belt (ADFB, Fig. 1a and b), being the largest Proterozoic belt in the northwestern India, has attracted the interest of the Atomic Minerals Directorate for Exploration and Research (AMDER), an organization responsible for U exploration in India. Subsequently, significant radioactive anomalies were reported from the metasedimentary rocks of the ADFB during the last two decades (viz., Umra and Rohil deposits near Udaipur and Jaipur, respectively in Rajasthan; Fig. 1b). In addition, >200 potential sites of anomalous radioactivity have been identified in and around the ~320 km long zone of albitite-microcli nite-pyroxinite, also referred to as the "Albitite line" (Ray, 1990) (Fig. 1b), in Rajasthan and Haryana (Singh et al., 2013).

The central part of the ADFB hosts the polymetamorphic terrain of the Banded Gneissic Complex (BGC; Gupta, 1934; Heron, 1953; Fig. 1b), which forms the basement to the supracrustal rocks of the Pur-Banera basin (PB; Fig. 1b), where the present investigation is carried out. In 2005, the AMDER discovered surface radioactive anomaly from the Samarkiya area, within the PB (Fig. 1c) and thereby considered the area as a potential target for uranium exploration. Subsequent petrographic and X-ray diffraction (XRD) studies revealed U-enriched minerals from the basal Pur quartzite of the Samarkiya area (Shaji et al., 2007). Shaji et al. (2007) proposed episodic uranium mineralization, involving mobilization, during the protracted magmatic/metamorphic evolution (~1.9–1.4 Ga) of the host rocks. However, detailed geochemical characters and temporal status of individual uranium mineralization/mobilization events, linked to tectono-metamorphic evolution of the host rocks, have so far eluded us.

In the Samarkiya area, uraninite is volumetrically the most abundant U-bearing mineral (Shaji et al., 2007). Amongst all the primary U-bearing minerals, uraninite hosts the highest U-content in its structure (i.e., up to 88.2 wt%). Due to auto-oxidation caused by radioactive decay of uranium, pure uraninite ($U^{+4}O_2$) phase is very rare in nature (Dahlkamp, 1993), and thereby the non-stoichiometry and defects in the mineral favors cationic substitution (Janeczek and Ewing, 1992), which modifies and alters its formula to ($U^{4+}_{1-x-y-z}U^{6+}_{x}REE^{3+}_{y}M^{2+}_{z}$) $O_{2+x-(0.5y)-z}$ (Janeczek and Ewing, 1992; Finch and Murakami, 1999). Previous studies have demonstrated that uraninite is chemically active and readily exchanges elements or recrystallizes during subsequent fluid circulation events (Grandstaff, 1976; Kotzer and Kyser, 1993). Additionally, the content of trace (REEs and Y; hereafter ∑REY) and minor (Si, Ca, Fe, Al, K, and Na) elements within uraninite is a function of the physico-chemical conditions (temperature, redox state, and fluid composition) prevalent during uraninite formation (Alexandre and Kyser, 2005; Bonhoure, 2007; Mercadier et al., 2011; Eglinger et al., 2013; Pal and Rhede, 2013). Consequently, the concentrations of other elements in uraninite can act as geochemical tracers pertinent to its environment of formation. Further, geochemical characterization in conjunction with in situ dating of uraninite can provide constraints on the timing of mineralization and subsequent alteration, if any (Kempe, 2003; Deditius et al., 2007; Pal and Rhede, 2013). In this study, integrating geochemical characteristics and in situ chemical dating of uraninite, we aim at deciphering the mineralization types and geochemical vis-à-vis temporal evolution of uraninite.

2. Geological setting

2.1. Regional Geology

The ADFB comprises three major geological units, which in chronological order, from oldest to youngest, include the Banded Gneissic Complex (BGC), the Aravalli Supergroup and the Delhi Supergroup (Gupta, 1934; Heron, 1953; Raja Rao, 1976; Gupta et al., 1997; Fig. 1b). The BGC is divided into two disconnected terrains, BGC-I and BGC-II respectively constituting the southern and the central parts. The BGC-I is dominated by tonalite-trondhje mite-granodiorite suite and granitoids of the Archaean age (3.30–2.50 Ga; Wiedenbeck and Goswami, 1994; Roy and Kröner, 1996; Wiedenbeck et al., 1996). The BGC-II includes meta-igneous granulites (Sandmata Complex), and the meta-granitoids, amphibolites, and metasedimentary rocks, namely the Mangalwar Complex (MC; Guha and Bhattacharya, 1995). The rocks of the BGC-II mainly display Paleo-Neoproterozoic ages with vestiges of Archaean component (i.e., ~2.50–0.95 Ga) (Buick et al., 2006, 2010; Bhowmik et al., 2010; Roy et al., 2012; Ozha et al., 2016a). They are polychronous in nature, preserving Proterozoic crustal unit with an early metamorphic event (5.5 kbar and 520–550 °C) at ~1.82 Ga and a later high pressure event (8.0 kbar and 590–640 °C) at ~0.95–1.05 Ga (Buick et al., 2006, 2010; Bhowmik et al., 2010; Ozha et al., 2016a).

The MC constitutes vast stretch of schist and para- to ortho gneisses in the western part (i.e., at the contact of Sandmata Complex) of the BGC-II (Buick et al., 2006). On the contrary, in the central parts, the MC host garnetiferous biotite schist, which is unconformably overlain by the linear supracrustal belt of PB that evolved as pull-apart basin (Sinha-Roy et al., 1998) starting at ~1.74 Ga (Ozha et al., 2016a). The PB is a major syncline comprising rock units of quartzite/conglomerate (Pur Formation), metapelites, amphibolites, calc-silicate gneisses (Rewara Formation), banded iron formations (Tiranga Formation), and quartzite, mica schist with intercalations of marble and calc-silicate bands (Samodi Formation) (Fig. 1c). According to the recent petrological and geochronological studies of Ozha et al. (2016a), both the basement (MC) and the supracrustal (PB) rocks were subjected to two discrete amphibolite facies metamorphic events at ~1.37–1.35 Ga and ~1.05–0.99 Ga. In addition, a much older Paleoproterozoic