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Uranium and Thorium Anomalies in the ~2.5 Ga Vendodu Leucogranite, Nellore Schist Belt, SE India and its Potential to Generate Uranium Deposits

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ABSTRACT

The Vendodu intrusive, emplaced at 2483±3 Ma within the Nellore schist belt, SE India, is a K-rich per-aluminous A-type leucogranite composed of quartz and alkali feldspar (essential minerals), biotite, zircon, allanite, titanite, magnetite and apatite (magmatic accessory minerals) and muscovite, haematite, fluorite and uraninite (hydrothermal accessory minerals); zircon and uraninite could be both magmatic and hydrothermal. The Vendodu leucogranite is characterized by high Rb, Zr, Nb, Th, U and REE, low Ca, Al, Ba and Sr abundances, and large negative Eu anomalies. U content in the Vendodu leucogranite averages 17.63 ppm and is 6 to 11 times higher than average concentration of U in Upper Continental Crust (UCC) and Archaean granitoids. Similarly, Th content averages 61.38 ppm and is 3 to 20 times higher than average concentration of Th in Archaean granitoids and UCC. The U distribution in the Vendodu leucogranite is influenced by both magmatic and high-temperature hydrothermal processes. Processes that have enriched U have also increased LREE, Nb and Cr contents in the leucogranites. Geochemical proxies including Th/U, Zr/U and V/Yb suggest both magmatic and high-T hydrothermal (deuteric) enrichment of U over a wide range of temperatures and oxygen fugacities. High Th/U ratios in the Vendodu leucogranite (1.62–9.76) hint that the hydrothermal (deuteric) fluids were possibly magmatic. Petrographic and geochemical evidences suggest that the Vendodu leucogranite experienced magmatic and deuteric U enrichment that has potential to form mineralized zones either *in situ* or in pegmatitic veins.

INTRODUCTION

Archaean granitoids record evolution of the early continental crust and its ore deposits. Sites, rates and processes of crust formation significantly changed at the Archaean-Proterozoic boundary: the Na-rich granites of the early Archaean are largely succeeded by late Archaean K-rich granites (Kemp and Hawkesworth, 2003), which make up 20% of presently exposed Archaean cratons (Condie, 1993). The K-rich leucogranites contain high proportions of radioactive elements and are considered to have increased the upper crustal budget of K, Rb, Cs, U, Th, LREE, Zr, and Hf, and possibly form the sources for Palaeoproterozoic uraniferous conglomerates as well as unconformity-related U deposits in the Palaeoproterozoic intra-cratonic basins (Taylor, 1987; Cuney, 2014).

Uranium ores are characterized by a spectrum of compositions resulting from different geologic origins (Cuney, 2009). Spatio-temporally, the U-deposits range from Archaean-Proterozoic boundary to Recent and deep-seated magmatic cumulates to surface regoliths (Sarangi and Krishnamurthy, 2008; Cuney, 2009). The International Atomic Energy Agency (IAEA) proposed 15 distinct types of U deposits (Hore-Lacy, 2016). Granite-hosted uranium mineralization is one of the important U-deposits (magmatic and hydrothermal vein-type) (Cuney, 2009). Leucogranite hosted U deposits, in general, are low grade (<0.1 wt.% U) but large tonnage (>100 Mt) (Cuney, 2014). Four types of U deposits associated with leucogranites include: (1) primary disseminations and segregations, (2) primary mineralization in aplites and pegmatites (late differentiates; syngenetic), (3) high-T hydrothermal deposition in veins and fractures (epigenetic) and (4) low-T hydrothermal deposits due to meteoric water interaction (Cuney, 2009 and 2014).

In the present study, positive uranium anomalies in circa 2.5 Ga Vendodu leucogranite from the Nellore schist belt, SE India and highlight its potential to host U-deposits is reported.

GEOLOGY AND PETROGRAPHY

The curvilinear Nellore Schist Belt (NSB) is the easternmost greenstone belt within the eastern Dharwar craton (Fig. 1A) and is sandwiched between the granulitic rocks of the Eastern Ghats belt (EGB) to the east and the intracratonic Cuddapah basin and TTG gneisses to the west (Fig. 1B). The NNE-trending NSB is up to ~600 km long and 30 to 130 km wide, and is composed of amphibolites, granite gneisses and metasediments, including banded iron formations (Vijaya Kumar et al. 2006). The present study area in the southernmost part of the NSB occurs between 2.7-2.6 Ga Archaean TTG to the west and the arc-related 1.8-1.7 Ga Eastern Ghats granite-migmatite complex to the east (Fig. 1C). Contacts between the different formations are either thrust or high-angle reverse faults (Fig. 1C). Hornblende- and biotite-granodiorite and leucogranite bodies are dispersed within the amphibolites and schistose rocks around the Kandra area (Fig. 1C). Leucogranites occur as domical stocks and elongate plutons within the schistose rocks. The strike directions of the host NSB terrane and the linear trails of leucogranite are conformable. A younger (1.85 Ga) ophiolite (Leelanandam, 1990; Sesha Sai, 2009; Vijaya Kumar et al., 2010; Saha, 2011), also occurs in the area, suggesting that the zone experienced repeated tectonothermal events. The leucogranites show intrusive relationships with the NSB, but thrusted and sheared contacts with the Kandra ophiolite complex (Fig. 1C).

The Vendodu leucogranite, member of the circa 2.5 Ga K-rich leucogranite magmatism (Vijaya Kumar et al., 2011), occurs as an isolated stock to the southwest of Kandra ophiolite complex within the Nellore schist belt (Fig. 1C). SHRIMP U-Pb dating of magmatic zircons indicate that the Vendodu leucogranite was formed at 2483±3

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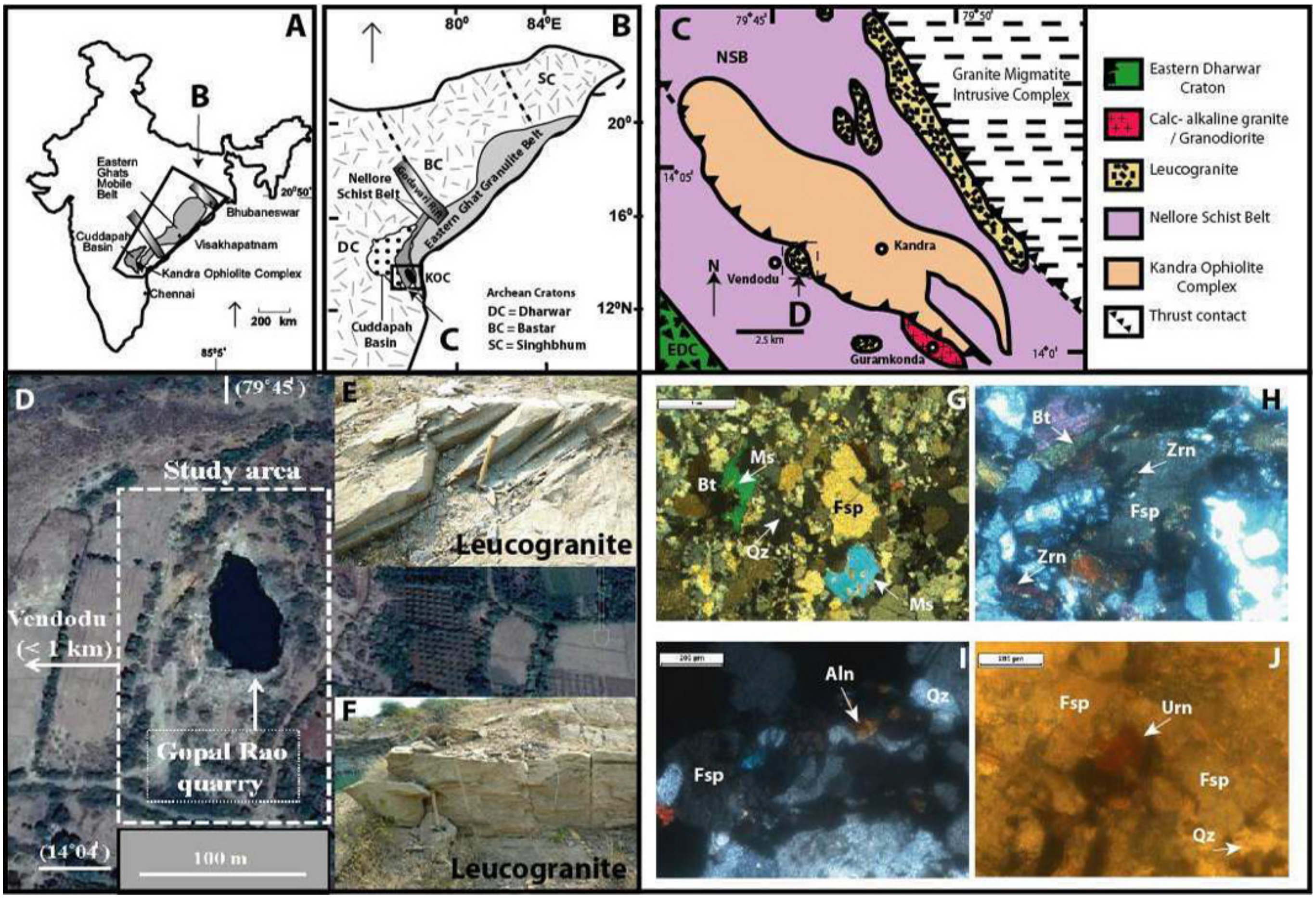

Fig 1. Location of Eastern Ghats Belt and Kandra Ophiolite Complex within SE India (**A** and **B**) Vendodu leucogranites are present toward southern part of Kandra ophiolite suite (**C**); Gopal Rao quarry, the study area of present work is ~1 Km from Vendodu village (**D**); Representative outcrops of leucogranites are shown in **E** and **F**. Samples for the present study are collected from the Gopal Rao quarry. Representative photomicrographs of Vendodu leucogranites are shown in **G** to **J**. Leucogranite essentially contains subhedral feldspar (Fsp) and quartz (Qz) with minor amounts of muscovite (Ms), biotite (Bt) and allanite (Aln) as minor mineralogy; zircon (Zrn) is an important accessory in the leucogranites. Uraninite (light brown colour under reflected light; **J**) is present along grain margins. Mineral abbreviations are after Whitney and Evans (2010).

Ma (Vijaya Kumar et al., 2011). Samples for the present study come from the Gopal Rao quarry occurring ~1 km to the east of Vendodu village (Fig. 1D). The leucogranites are massive, fractured and traversed by few pegmatite veins (Figs. 1E and F). The leucogranite shows a medium- to coarse-grained hypidiomorphic texture (Fig. 1G). It contains quartz, alkali feldspar (+ subsolidus orthoclase perthite) as essential minerals (Fig. 1G). Biotite, zircon, allanite, titanite, magnetite and apatite are the important magmatic accessory minerals (Figs. H and I). Muscovite, haematite and fluorite, present in some of the studied samples, possibly represent products of late magmatic (deuteric) fluids. Uraninite occurs along grain boundaries in a few leucogranites (Fig. 1J). Reddish, ferruginous zones are recorded in some of the Vendodu pegmatites. The mineralogy of Vendodu leucogranite is similar to other U-mineralized granites elsewhere (see Mercadier et al., 2011; Simpson et al., 1979; Ballouard et al., 2017a).

WHOLE-ROCK GEOCHEMISTRY

Whole-rock major and trace element geochemistry of the Vendodu leucogranite was carried out on XRF and HR-ICP-MS respectively in the Geochemical Laboratories of National Geophysical Research Institute, Hyderabad. To determine accuracy and precision of the instrument, replicate analysis of the standards and a leucogranite sample were carried out. International granite (JG-1a) reference sample was used for calibration. The precisions for all major, trace and REE analyzed here were < 5% RSD (relative standard deviation) with comparable accuracy. A/CNK ratios (> 1 in all the samples) attest to per-aluminous nature of the Vendodu leucogranite. The Vendodu leucogranite is an Rb, Zr, Pb, Th, U and REE-rich biotite-bearing granite. High alkalis ($Na_2O + K_2O$), Rb, Nb, HREE, FeO^t/MgO and low Ca, Al, Ba, Sr and large negative Eu anomalies characterize the leucogranite as an A-type magma (Vijaya Kumar et al., 2011). High Ga x 10,000/Al ratios (1.94–3.14) also demonstrate the A-type nature of the Vendodu leucogranite. It was suggested that the leucogranite was formed by high-T (900–950°C), low pressure (4–6 kbar) partial melting of the calc-alkaline granite under vapor-absent, reducing conditions (Vijaya Kumar et al., 2011).

U, Th abundances and U/Th ratios for the Vendodu leucogranites are given in Table 1. Average U and Th abundances and Th/U ratios for Archaean TTG, calc-alkaline granite and leucogranite (Kemp and Hawkesworth, 2003), and upper continental crust (Rudnick and Gao, 2003) are also shown for comparison. U in the Vendodu leucogranite ranges from 2.77 to 33.75 ppm and Th ranges from 27 to 77 ppm. Average U and Th abundances in Vendodu leucogranite (17.63 and 61.4 ppm) are high compared with values of the average concentration of U and Th in late Archaean granitoids and Upper Continental crust (UCC; Table 1). Th/U ratios in the Vendodu leucogranite are 1.62 to 9.76 with an average value of 4.53 (Table 1). Three weathered leucogranites (sample numbers ending with W) were analysed to

Table 1. U and Th abundances (ppm) in the Vendodu leucogranite, Nellore Schist Belt, Andhra Pradesh

	U	Th	Th/U
VD-02	33.75	54.58	1.62
VD-03	6.05	55.25	9.13
VD-4W	8.36	67.49	8.07
VD-5	18.95	70.60	3.73
VD-08	19.84	68.66	3.46
VD-09	15.71	48.60	3.09
VD-11	13.58	77.22	5.68
VD-12	17.86	53.90	3.02
VD-15	31.72	63.70	2.01
VD-17	16.97	67.16	3.96
VD-19	28.69	72.84	2.54
VD-21	2.77	27.03	9.76
VD-26	22.62	70.29	3.11
VD-32	18.29	62.98	3.44
VD-36W	6.69	62.22	9.30
VD-37	20.96	54.70	2.61
VD-39W	13.05	60.47	4.63
VD-41	6.77	53.39	7.89
VD-42	18.69	64.78	3.47
VD-43	19.98	69.51	3.48
VD-44	29.80	68.00	2.28
VD-45	16.72	56.92	3.40
Average U and Th abundances			
Vendodu Leucogranite	17.63	61.38	4.53
Archaean TTG*	1.6	6.9	4.31
Archaean Calc-alkaline granite*	2.4	21	8.75
Archaean Leucogranite*	2.09	3.4	1.63
Upper Continental Crust (UCC)#	2.7	10.5	3.89

U abundances >20 ppm in the Vendodu Leucogranite are shown in bold
* from Kemp and Hawkesworth (2003)
\# from Rudnick and Gao (2003)

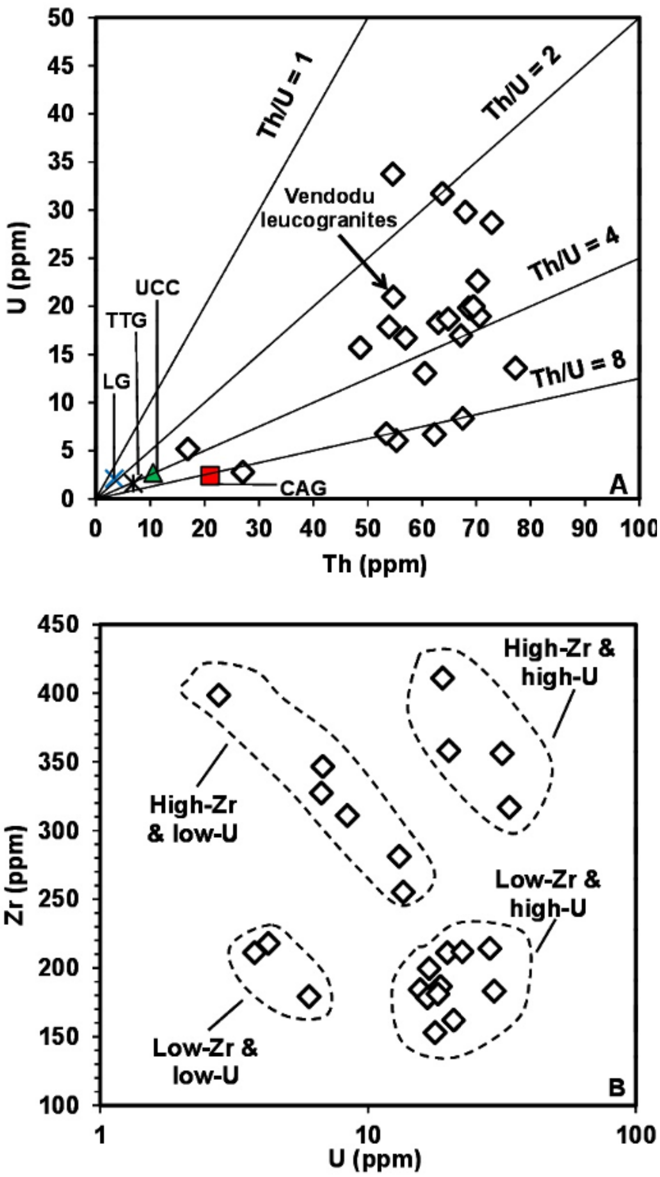

Fig. 2. U (ppm) *vs* Th (ppm) (**A**) and Zr (ppm) *vs* U (ppm) (**B**) variation plots for Vendodu leucogranites. Most of the Vendodu leucogranites have Th/U ratios between 2 and 8. U and Th abundances in the Archaean Tonalite-Trondhjemite-Granodiorite (TTG), calc-alkaline granite (CAG), leucogranite (LG) and Upper Continental Crust (UCC) are also shown for comparison. We infer that crystallization of U-bearing minerals from magmatic to deuteric stage over a wide range of temperatures and changing oxygen fugacities are responsible for variable Th/U and Zr/U ratios in the Vendodu leucogranites. For discussion, see the text.

decipher any effect of weathering on U abundances. In general, the weathered granites have lower U abundances (6–13 ppm) than the average abundance in the fresh samples indicating that some of the uranium is easily leachable.

Majority of the leucogranites have restricted Th contents (50–75 ppm) for a large variation in U (5–35 ppm; Fig. 2A), suggesting that Th-rich minerals like monazite are not responsible for the U enrichment. Th/U ratio is indicator of type of U-bearing minerals in granitoids. High Th/U ratios (>2) suggest that most of U is hosted in refractory minerals such as zircon and titanite in the Vendodu leucogranite (see Cuney, 2014). Uraninite may crystallize if U is > 10 ppm in peraluminous leucogranites (Forster et al., 2008; Cuney, 2014; Ballouard et al., 2017b) such as the Vendodu leucogranite. It is possible that high Th contents in the Vendodu leucogranite are due to Th-rich nature of the uraninite. Highly variable Th/U ratio in the Vendodu leucogranite can be attributed to magmatic and deuteric/hydrothermal formation of U-bearing minerals over a wide range of temperatures (see Forster et al., 2008; Ballouard et al., 2017a; Fig. 2A).

Zircon also may not be solely responsible for the U enrichment in the leucogranites as U and Zr do not correlate with each other (Fig. 2B) but share a complex relationship. The high-Zr leucogranites have both high and low U concentrations (Fig. 2B). Similarly, low-Zr leucogranites also have high and low U abundances (Fig. 2B). In the low-Zr and high-U leucogranites, U-carrying primary mineral phase could be uraninite and in the high-Zr and high-U leucogranites, U-carrying primary phase could be zircon. Average U content in the Vendodu zircons is ~ 1800 ppm (Vijaya Kumar et al., 2011); thus 1% of zircon is capable of providing 18 ppm to the whole rock chemistry, which is the average U content in the Vendodu leucogranite. However, some of the leucogranites in spite of having considerable zircon population have very low U contents (Fig. 2B) suggesting that the zircons them themselves may have highly variable U abundances. Indeed, some of the zircons considered to have grown in fluid-rich late magmatic (deuteric) environment in the Vendodu leucogranites have lower U abundances (Vijaya Kumar et al., 2011). U-Th and U-Zr relationships suggest that zircon and uraninite are primary U-bearing minerals, which have grown from magmatic to deuteric stages of crystallization.

Positive relationships between U and light REE, Nb and Cr and consequently inverse relationships between U and La/U, Nb/U and Cr/U (Fig. 3) suggest that processes that enriched U also increased La, Nb and Cr in the Vendodu leucogranites. It is possible that in addition to zircon and uraninite, U could have also accommodated in a refractory and complex U-Th-Nb-REE-Ti (?) mineral such as fergusonite or samarskite. However, these minerals in the Vendodu leucogranite are yet to be recorded. Alternatively, F-rich deuteric fluids may have precipitated LREE, Nb and Cr in addition to U along grain boundaries. U shows poor correlation with HREE and Y (Figure not shown) suggesting that U enrichment is accompanied by increase in LREE but not HREE and Y. Two different but parallel trends between U and plotted parameters (Fig. 3A, C and E) indicate at least two processes (magmatic and deuteric?) are responsible for U enrichment in the Vendodu leucogranites.

EVIDENCES FOR POSSIBLE URANIUM MINERALIZATION IN THE VENDODU LEUCOGRANITE

Partial melting or fractional crystallization rarely produces uranium deposits (Cuney, 2014); late magmatic fluid (deuteric) enrichment is an important process of U enrichment especially in the U-rich A-type granites. Hydrothermal process seems to be necessary for mobilization of U from the primary carrier minerals and subsequent precipitation in favorable environments. Mobilization of U is relatively easier if it resides in the easily dissolvable uranium oxides rather than refractory minerals such as zircons and titanite (Cuney, 2014). In peraluminous leucogranites U is also hosted by uranium oxides in addition to zircon, allanite and titanite (Cuney, 2014). Additionally, magma with high F contents tends to contain higher U abundances. Fluorite is an important indication for U mineralization and major path finder elements for magmatic U deposits are Th, Nb and F (Rogers et al., 1978).

Igneous uranium deposits generally contain high concentration of Th. Th/U ratios increase from 2 to 3 in mafic rocks to 5 to 6 in the felsic differentiates. U partitions into late-stage fluids due to oxidation of U whereas Th is less efficiently separated into hydrothermal fluids; therefore, primary igneous deposits tend to have Th/U ratios much higher than uranium deposits associated with pegmatites, veins and supergene enriched zones (Rogers et al., 1978; Bowden et al., 1995). Therefore, it is suggested that decoupling of U and Th concentrations in the Vendodu leucogranite is due to magmatic and high-T hydrothermal (deuteric) enrichment of U over a wide range of temperatures and oxygen fugacities (see Figs. 2A and 4).

High oxygen fugacity favours transport of U (Dubessy et al., 1987) as hexavalent uranyle ion (UO_2^{2+}) (Cuney, 2009) and gets precipitated as UO^{4+} under reducing conditions. Potential reductants in a granitic magma include magnetite, ilmenite and sulfides. $D^{V/melt}$ decreases with increasing f_{O_2}, therefore, at high f_{O_2} conditions melts tend to have much higher V than the melt crystallized at low f_{O_2} conditions (see Huang et al., 2015). Although V absolute concentration is controlled by fractionating mineral assemblages, V (redox sensitive element) ratio with Yb, Y, Sc and Ga (single valent elements with similar bulk distribution coefficients) are less sensitive to magmatic differentiation

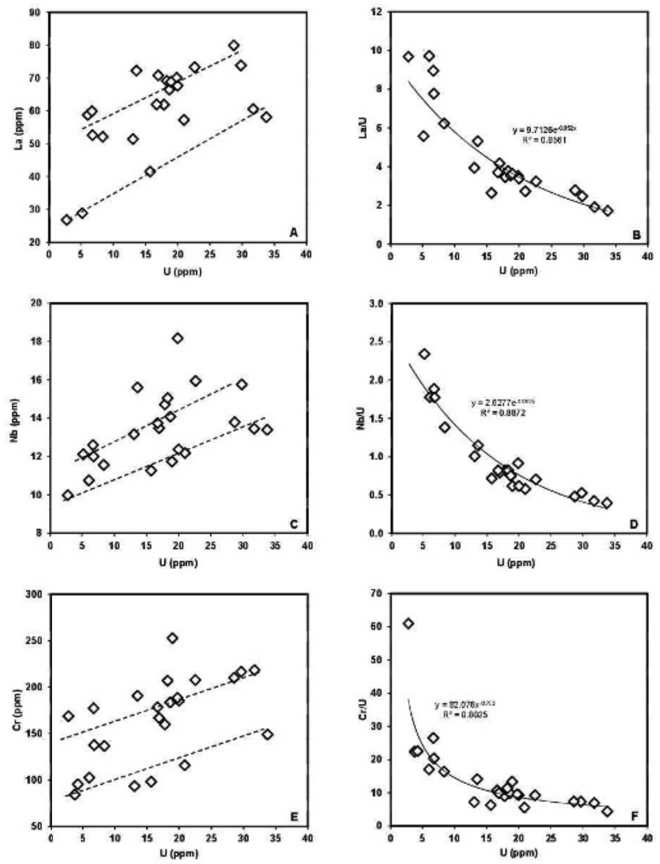

Fig. 3. La (ppm) and La/U (**A** and **B**), Nb (ppm) and Nb/U (**C** and **D**) and Cr (ppm) and Cr/U (**E** and **F**) variation against U (ppm) in the Vendodu leucogranites. Note the two different, albeit, parallel trends between U and La, U and Nb and U and Cr plots. For discussion, see the text.

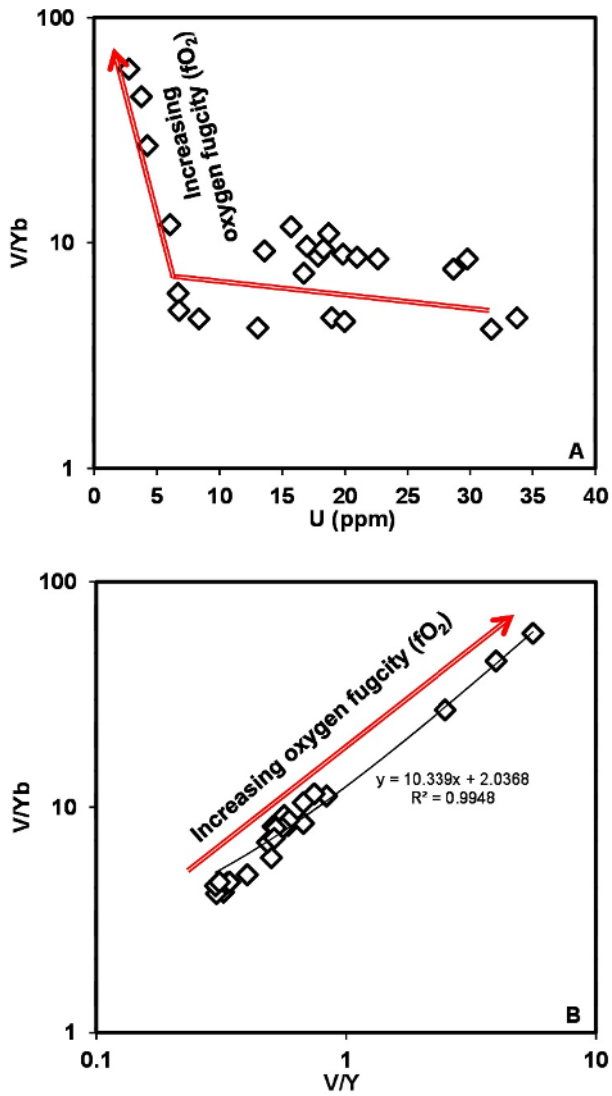

Fig. 4. V/Yb *vs* U (ppm) (**A**) and V/Yb *vs* V/Y (**B**) variation plots for Vendodu leucogranites. Note the inverse relationship between U content and V/Yb ratios. For discussion, see the text.

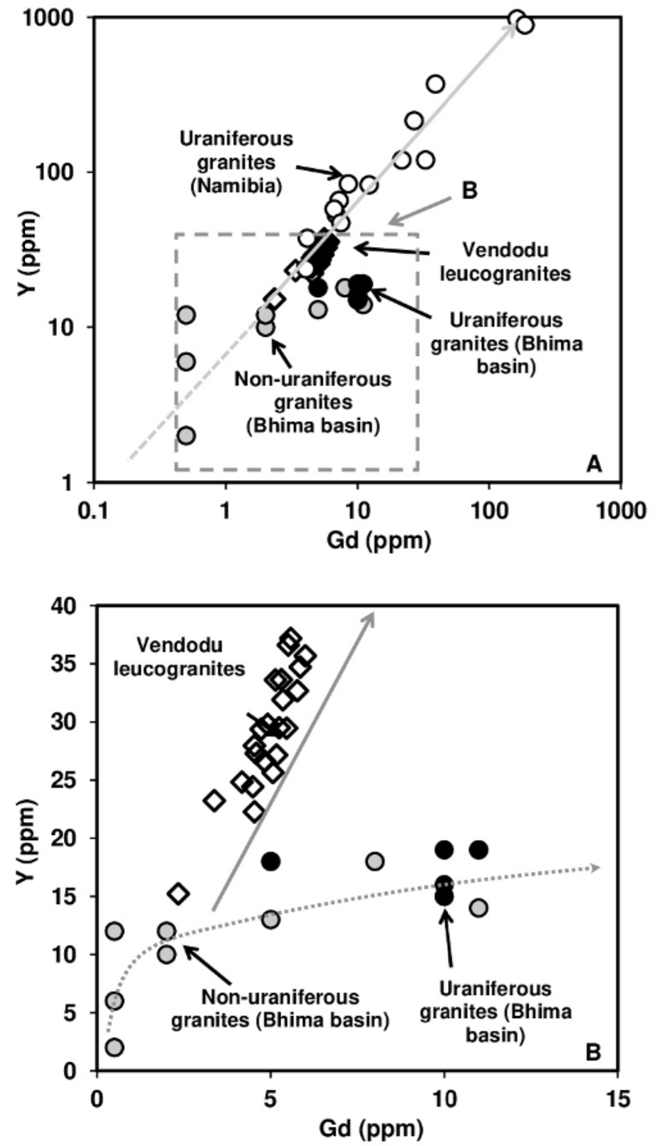

Fig. 5. Y (ppm) *vs* Gd (ppm) variation plot for Vendodu leucogranites. Uraniferous and Non-uraniferous granites from Damara orogen, Namibia (Chen et al., 2017) and Bhima basin, Southern India (Patnaik et al., 2016) are also shown for comparison (**A**). The Vendodu leucogranites show trend similar to that of Namibia leucogranites (**A**) but different from that of Bhima basin leucogranites (**B**). For discussion, see the text.

and are good geochemical proxies for f_{O_2} conditions (Laubier et al., 2014; Huang et al., 2015 and references therein). V and Yb have some similar D (distribution coefficients) during fractional crystallization of silicate magma under constant f_{O_2}conditions (Farley, 1994). However, under high oxidation conditions f_{O_2} (>FMQ) V/Yb ratio increases, whereas under low f_{O_2} (<FMQ) conditions V/Yb ratio decreases. Under oxidizing conditions U occurs as U^{+6} which is mobile; therefore, low U samples should show high V/Yb ratios. The Vendodu leucogranites clearly show such a pattern (Fig. 4A). When tested with other geochemical proxies for oxygen fugacity such as V/Ga, V/Y and V/Sc, they have also given similar results (for example see Fig. 4B). In summary, the leucogranites with high V/Yb, V/Ga, V/Y and V/Sc ratios, formed at high f_{O_2} (oxidizing conditions), have low U contents and also modal haematite. The low U contents in some of the Vendodu leucogranites thus do not seem to be representative of original magmatic U values.

U enrichment associated with leucogranites may take place by two distinct mechanisms: (1) magmatic + high-T syngenetic hydrothermal and (2) low-T epigenetic hydrothermal. Since Y/Gd ratio is independent of high-T fluid activity and fractional crystallization, it gives important clues regarding the process of U mineralization and possible primary carries of U. Two genetically distinct types of U mineralization associated with leucogranites: Damara orogen, Namibia (U mineralization was largely due to magmatic and high-T hydrothermal; Chen et al., 2017) and Bhima basin, southern India (U mineralization was largely due to low-T hydrothermal vein type; Dhana Raju and Babu, 2003; Patnaik et al., 2016) were selected. The Y/Gd ratios in the Vendodu leucogranite are constant and fall along the trend for Namibia U-bearing leucogranites (Fig. 5A). The Y/Gd ratios in the Vendodu leucogranites (4.9-7.5) are within the range shown by constituent zircons (5-16; zircon data from Vijaya Kumar et al., 2011). Whereas the uraniferous leucogranites of Bhima basin have relatively more Gd contents than the Vendodu leucogranites and show distinctly different trend (Fig. 5B). It is speculated that U enrichment in the Vendodu leucogranite is due to magmatic and high-T hydrothermal processes.

CONCLUSIONS

The present study on the positive U anomalies in the circa 2.5 Ga Vendodu leucogranite indicates that the (1) the mineralogy of Vendodu leucogranite (quartz and alkali feldspar + biotite, zircon, allanite, titanite, magnetite and apatite + muscovite, haematite, fluorite and uraninite) is similar to that of U-mineralized granites elsewhere, (2) the average U and Th contents in the Vendodu leucogranite are respectively 6 to 11 and 3 to 20 times higher than the average U and Th abundances in the late Archaean granitoids and upper Continental Crust, (3) uraninite and zircon could be primary U- and Th-carrying minerals, (4) leucogranites with high La, Nb, Cr, Y, Gd contents and $(La/Yb)_N$ ratios, and low V/Yb and Eu/Eu* ratios could be primary targets of U mineralization in the Vendodu leucogranite and associated pegmatites. Detailed studies in this direction are in progress.

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