Fluid inclusion, geochemical, Rb–Sr and Sm–Nd isotope studies on tungsten mineralized Degana and Balda granites of the Aravalli craton, NW India

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Granitic plutons occurring within and to the west of the Delhi Fold Belt in the Aravalli craton, northwestern India are the result of widespread felsic magmatism during Neoproterozoic, some of which are associated with greisen and skarn tungsten deposits. In this paper, we present the result of our study on fluid inclusions, geochemistry and geochronology of two such tungsten mineralized granite plutons at Degana and Balda, and interpret the nature of ore fluid, and petrogenesis and age of these mineralized granites. Fluid inclusion study reveals coexistence of moderate and hyper-saline aqueous fluid inclusions along with aqueous-carbonic inclusions, suggesting their origin due to liquid immiscibility during fluid–rock interaction. Geochemically, the granites are peraluminous, Rb enriched, Sr and Ba depleted and highly differentiated. The Rb–Sr isotopic systematics yielded 795 ± 11 Ma for Balda granite and 827 ± 8 Ma for Degana granite. We show that major phase of widespread granitoid magmatism and mineralization during the Neoproterozoic (840–790 Ma) in NW India is coeval with breakup of the Rodinia supercontinent and infer a causal relationship between them.

Keywords. Neoproterozoic W deposit; Degana and Balda granites; fluid inclusion; geochronology; Aravalli craton; Rodinia.

1. Introduction

Tungsten mineralization is typically related to A- and S-type granites enriched in W, Sn are well above the Clarke values (Neiva 1984; Lehmann 1987; Huang and Jiang 2014). The mineralized granites are commonly emplaced at shallow crustal level and possess peraluminous characteristics, with enrichment in volatile elements (F, Li, and B; Neiva 1984; Zhao *et al.* 2001; Xie *et al.* 2009; Fogliata *et al.* 2012). Geochemical studies of granitic rocks

associated with various types of mineralization have been undertaken to establish criteria for mineral prospecting (Flinter *et al.* 1972; Tauson and Kozlov 1973; Tischendorf 1977; Drake 1981; Ohlander 1985; Lehmann 1987; Srivastava and Sinha 1997; Pirajno 2009).

In India, tungsten mineralization is mainly associated with Proterozoic granites in the western part of Aravalli craton (Degana, Balda and Sewariya; figure 1a) and in Sakoli schist belt of Bastar craton (Agargaon, Kuhi-Khobna;

Figure 1. (a) Regional geological map of NW India (inset shows the location of the study area; after Heron 1953; Roy 1988). (b) Geological map of Degana tungsten deposit (Pandian and Varma 2001). Inset shows the location of Degana in NW India. Abbreviation: MCG (medium to coarse grained granite porphyry); CGP (coarse grained granite porphyry); FGP (fine grained granite porphyry). (c) Geological map of Balda tungsten deposit (inset shows the location of the study area; after Singh and Singh 2001).

Banerjee *et al.* 1979; Gupta *et al.* 1980, 1997; Chattopadhyay *et al.* 1982; Bhoskar and Chande 1983; Jain and Mohan 1987; Bhattacharjee *et al.* 1993; Chattopadhyay *et al.* 1994; Banerji and Pandit 1995; Singh and Singh 1997, 2001; Srivastava and Sinha 1997; Pandian 1999; Pandian and Dutta 2000; Saha and Mohan 2000; Pandian and

Varma 2001; Sharma *et al.* 2003; Arif *et al.* 2017). Geochemical and geochronological data are available for several granites of Aravalli craton (table 1) including the Neoproterozoic Erinpura granite and Malani Igneous Suite (MIS), which occupy large areas in South Delhi Fold Belt and in the western part of Aravalli craton (Crawford and

Figure 1. (Continued.)

Table 1. Radiogenic isotopic ages of granitoids in NW India with present study.

Eon	Lithology	Method	Age (Ma)	Reference
Neoproterozoic	Sindreh felsic volcanics	U–Pb	765.9 ± 1.6	van Lente <i>et al.</i> (2009)
	Malani Igneous Suite	U–Pb	771 ± 2	Torsvik <i>et al.</i> (2001a)
	Abu granite	U–Pb	765 ± 5	Ashwal <i>et al.</i> (2013)
	Balda granite	Rb–Sr	795 ± 11	This study
	Degana granite	Rb–Sr	827 ± 8.2	This study
	Erinpura granite	Rb–Sr	900	Choudhry <i>et al.</i> (1984)
	Godhra granite	Rb–Sr	940	
Mesoproterozoic	Sendra granite	Pb–Pb	968 ± 8.2	Pandit <i>et al.</i> (2003)
	Saladipura granite	Rb–Sr	1400	
	Harsora granite	Rb–Sr	1600	
Paleoproterozoic	Chawand granite	Pb–Pb	1800	Sarkar <i>et al.</i> (1989), Deb <i>et al.</i> (1989)
Archean	Berach granite	Rb–Sr	2445 ± 100	
	Annasagar granite	Sm–Nd	2800	Tobisch <i>et al.</i> (1994)
	Gingala granite	Rb–Sr	2905 ± 109	Choudhry <i>et al.</i> (1984)
	Untala granite	Sm–Nd	3500	Macdougall <i>et al.</i> (1983)

Bold represents the present study.

Compston 1970; Choudhry *et al.* 1984; Tobisch *et al.* 1994; Torsvik *et al.* 2001a; Pandit *et al.* 2003; Ashwal *et al.* 2013), respectively. However, no geochronological data is available for the W-mineralized Balda and Degana granites. The Neoproterozoic period is significant time for large-scale magmatism and mineralization in NW India.

In Degana area, phyllite of Delhi Supergroup is intruded by a granite pluton consisting of three successively emplaced porphyritic granites (medium to coarse grained granite porphyry, MCG; coarse grained granite porphyry, CGP; and fine grained granite porphyry, FGP), two generations of each aplites, greisen veinlets and wolframite mineralized quartz lodes (figure 1b; Pandian and Varma 2001). Zinnwaldite or lithian muscovite, topaz and fluorite are the common accessory minerals in the granitic rocks and major components of greisen and ore mineral assemblage.

In Balda area, mica schist of Sirohi Group is intruded by an older, highly deformed Erinpura granite gneiss, and a younger, massive, tourmaline leucogranite called Balda granite, which is responsible for W mineralization (figure 1c; Chatopadhyay *et al.* 1982). Wolframite bearing quartz veins are localized along sheared intrusive contact between the schist and Balda granite. Muscovite, tourmaline, apatite and fluorite are the accessory minerals in Balda granite and also occur in wolframite-bearing quartz veins.

In this paper, we report major, trace element and isotope geochemistry and age of intrusion of Degana and Balda granites and discuss their petrogenesis and significance of Neoproterozoic magmatism in the NW India. We also present the types and composition of fluid inclusions in greisenized granites, which are the footprints of magmatic-hydrothermal activity responsible for tungsten mineralization in these granites.

2. Samples and analytical methods

Five samples of greisenized MCG from Degana and three samples of greisenized tourmaline leucogranite from Balda were prepared as doubly polished wafers (300–500 μm thick) for fluid inclusion study.

Twelve samples of Degana granite, including four each of MCG (D-50, 51, 52, 56), CGP (D-57, 67, 68, 69) and FGP (D-70, 71, 72, 73), and four samples of Balda Granite (B-22, 24, 34, 37) were prepared for geochemical analysis. The powdered samples were homogenized by coning and quartering and 100 g of each sample was powdered ($< -120 \mu\text{m}$) to analyze for major oxides and trace elements by WD-X-Ray Fluorescence Spectrometer (Bruker, X4 Pioneer) at Central Instrumentation Facility, Pondicherry University. The major and trace element abundances are given in tables 3 and 4.

Nine samples of Degana granite (three each of MCG, CGP and FGP), three samples of Balda

Table 2. Fluid inclusion data of hydrothermal quartz from Degana and Balda granites.

Type	Phase	Tm ² _{CO} (°C)	Tm _{Clath} (°C)	Th ² _{CO} (°C)	T _{fm} (°C)	Tm _{Ice} (°C)	Low salinity (wt.%) NaCl equivalent	Th _{H₂O} (°C)	Tm solid (°C)	High salinity (wt.%) NaCl equivalent	Density (g/cc)
Degana granite											
I	L + V				−28 to −22.5 −50 to −47	−12 to −6	16–9.2	208–347 (L)			0.98–0.79
II	L ₁ + L ₂ + V	−64 to −56.6	3.6–14	15–27							0.90–0.86
III	L + V + S						11–4	226–350 (V) 205–318	195–337	30–41.5	1.12–1.23
Balda granite											
I	L + V				−26 to −22	−7.3 to −3.3	11–5.4	265–310			0.98–0.79
II	L ₁ + L ₂ + V	−60 to −56.7	8–15	10–25			<4	280–318			0.90–0.86
III	L + V + S							168–214	110–210	28–32	1.12–1.23

Tm²_{CO}: CO₂ melting temperature; Tm_{Clath}: Clathrate melting temperature; Th_{CO₂}: CO₂ homogenization temperature; T_{fm}: temperature of first melting; Tm_{ice}: temperature of ice (last) melting; Th_{H₂O}: H₂O homogenization temperature; Tm_{solid}: solid dissolution temperature. All temperature is °C, salinity expressed as wt.% NaCl equivalent and density expressed as g/cc.

granite and mineral fractions from these samples (muscovite, zinnwaldite, plagioclase) were prepared for Rb–Sr and Sm–Nd whole rock isotopic studies. The mineral fractions of plagioclase and zinnwaldite were separated from FGP (D-73) and plagioclase and muscovite were separated from Balda granite (B-34). For obtaining mineral fractions, the powdered samples reduced to about −120 μm size were subjected to heavy mineral separation with bromoform and magnetic separator (Model Frantz LB-1). Muscovite, zinnwaldite and plagioclase were hand-picked under binocular microscope to get pure mineral fractions. Micas were gently ground in agate mortar in acetone medium and inclusions were removed by repeated washings. The minerals and whole rock samples (about 30 and 100 mg, respectively) were precisely weighed and taken in a 3 ml Savillex vial and digested in HF, HNO₃ and HCl acids in steel lined pressure digestion vessels (Parr[®], USA). The digested samples were evaporated, dissolved in 1N HCl and split into two fractions. One of the fractions, isotopic tracer solution was added and equilibrated to determine Rb, Sr, Sm and Nd abundances by Isotope Dilution (ID) method. The solutions were passed through HCl columns to separate the Rb, Sr and REEs. The REE fractions were loaded on HDEHP column to separate Nd and Sm. The isotopic analysis was performed using thermal ionization mass spectrometer (model-Triton, Thermo-Finnigan) at National Facility for Geochronology in Department of Earth Sciences, Pondicherry University, India. Details of isotope analyses in this study are given in [Anand and Balakrishnan \(2010\)](#) and results of isotopic analysis are given in table 5.

3. Results

3.1 Fluid inclusion petrography

Fluid inclusions were observed in greisen quartz from Degana and Balda granites. The isolated and randomly distributed inclusions are classified as primary in origin. Inclusions showing secondary trails, planar arrays are designated as secondary in origin ([Roedder 1971](#)). The size of the inclusion vary from 4 to 20 μm. Three distinct types of primary fluid inclusions are observed in greisen quartz. Type I is aqueous bi-phase containing a liquid and vapour (L_{H₂O} + V_{H₂O}); type II is

Table 3. Chemical composition of Degana granites.

	D-50	D-51	D-52	D-56	D-57	D-67	D-68	D-69	D-70	D-71	D-72	D-73
Major oxides												
SiO ₂	67.39	71.32	69.79	68.33	71.86	71.72	71.89	68.18	68.87	68.94	73.73	71.66
TiO ₂	0.02	0.03	>0.02	>0.02	0.02	0.03	0.02	0.14	0.02	>0.02	>0.02	0.02
Al ₂ O ₃	15.62	14.42	16.63	16.24	16.36	14.01	14.37	15.87	15.81	15.19	15.90	14.91
Fe ₂ O ₃ (Tot)	5.70	2.54	5.48	8.08	5.72	2.67	2.43	2.72	3.86	5.59	3.49	2.07
MnO	0.48	0.15	0.40	0.24	0.22	0.15	0.14	0.07	0.12	0.48	0.08	0.06
MgO	>0.02	0.04	0.06	0.30	0.04	>0.02	0.19	>0.02	>0.02	>0.02	0.32	>0.02
CaO	0.49	0.74	0.60	0.17	0.53	0.66	0.59	1.33	0.65	0.08	0.77	0.62
Na ₂ O	3.24	2.78	2.08	2.42	2.06	2.36	3.43	3.05	2.39	2.25	2.20	3.54
K ₂ O	3.83	5.21	3.47	3.39	3.16	4.95	4.54	5.65	4.35	4.67	4.16	4.55
P ₂ O ₅	>0.02	>0.02	>0.02	>0.02	>0.02	>0.02	>0.02	0.04	>0.02	>0.02	>0.02	>0.02
Total (wt.%)	96.77	97.23	98.50	99.17	99.96	96.55	97.59	97.04	96.07	97.21	100.6	97.43
Trace elements (ppm)												
Sc	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10
Cr	54	36	56	47	51	58	14	54	10	57	50	54
Ni	36	47	41	60	30	38	44	58	42	49	54	48
Rb	1850	1171	1722	1029	1342	1485	1111	1194	1060	1213	1421	1907
Sr	98	117	97	122	88	99	112	113	125	107	114	104
Y	74	66	68	63	62	70	64	69	58	62	68	80
Zr	64	114	55	118	57	73	103	104	186	79	99	76
Nb	16	18	16	21	15	17	18	19	17	18	19	17
Ba	<10	<10	<10	<10	<10	<10	<10	<10	113.8	<10	<10	<10
A/CNK	2.0656	1.6518	2.7067	2.7148	2.8447	1.7592	1.6793	1.5824	2.1388	2.1685	2.2334	1.7112
Y + Nb	89.9	83.6	83.6	83.8	76.8	86.3	81.6	87.8	74.9	80.5	86.7	96.5
CIPW normative mineralogy (wt.%)												
Quartz	32.54	33.76	43.41	39.96	46.66	38.31	33.12	25.73	37.62	37.81	42.52	32.59
Plagioclase	32.56	28.51	22.23	23.34	21.39	24.63	33.44	33.59	25.39	21.2	23.16	34.54
Orthoclase	25.35	33.24	22.81	22.46	20.48	31.92	28.85	36.04	28.65	30.81	26.17	28.89
Corundum	3.78	1.98	5.97	5.96	6.05	2.5	1.89	1.67	4.29	4.51	4.36	2.09
Hypersthene	4.37	1.87	4.26	6.33	4.03	1.97	2.11	2.07	3.09	4.31	2.97	1.36
Ilmenite	0.02	0.03	0	0	0.02	0.03	0.02	0.15	0.02	0	0	0.02
Magnetite	1.37	0.59	1.3	1.94	1.35	0.62	0.56	0.63	0.92	1.34	0.8	0.48
Zircon	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.03	0.01	0.01	0.01	0.02
DI	90.45	97.4	88.45	85.76	88.53	94.86	95.41	95.36	91.66	89.82	91.85	96.02

Table 4. Chemical composition of Balda granites.

	B-22	B-24	B-34	B-37
Major oxides				
SiO ₂	72.02	74.11	70.85	69.73
TiO ₂	0.03	0.14	0.04	0.03
Al ₂ O ₃	15.34	14.34	15.99	16.80
Fe ₂ O ₃ (Tot)	0.93	1.17	1.30	0.59
MnO	0.02	0.03	0.02	0.01
MgO	0.33	>0.02	0.08	0.04
CaO	0.60	1.26	0.69	1.82
Na ₂ O	4.06	3.50	4.13	2.19
K ₂ O	5.54	5.03	5.23	6.32
P ₂ O ₅	0.43	0.05	0.43	0.60
Total (wt.%)	99.31	99.63	98.77	98.14
Trace elements (ppm)				
Cr	16	40	20	27
Ni	50	51	49	58
Rb	1064	278	958	544
Sr	147	154	124	139
Y	50	35	49	40
Zr	108	180	105	127
Nb	17	16	17	18
Ba	<10	203	<10	<10
A/CNK	1.5048	1.464	1.591	1.6271
Y+Nb	66.9	50.3	65.9	58.6
CIPW normative mineralogy (wt.%)				
Quartz	26.45	31.58	26.36	31.08
Plagioclase	35.27	35.8	36.64	24.34
Orthoclase	34.19	30.86	32.6	39.71
Corundum	1.74	0.63	2.24	3.05
Hypersthene	1.27	0.61	0.97	0.44
Ilmenite	0.03	0.15	0.04	0.03
Magnetite	0.21	0.26	0.29	0.14
Apatite	0.83	0.1	0.84	1.18
Zircon	0.01	0.02	0.01	0.02
DI	95.91	98.24	95.6	95.13

aqueous-carbonic containing liquid CO₂ and liquid H₂O (L_{H₂O} + L_{CO₂}), some of type II inclusions have V_{CO₂} > 90% that contain a small amount of or no water. Type III is polyphase inclusions (L_{H₂O} + V_{H₂O} + solid; figure 2).

3.2 Microthermometry

3.2.1 Greisen quartz from Degana granite

In the type I inclusion, first ice melting temperature is observed at −28 to −22.5°C (H₂O–NaCl–KCl) and −50 to −47°C in some of the inclusions (H₂O–CaCl₂–MgCl₂). The final ice melting temperature of these inclusions varies from −12 to −6°C, corresponding to the salinity values of 16

to 9.2 wt.% NaCl equivalent. The type I inclusions are homogenized into liquid between 208 and 347°C (L_{H₂O} + V_{H₂O} → L_{H₂O}). In the type II inclusions, Tm_{CO₂} was recorded to lie between −64 and −56.6°C and Tm-clathrate is recorded between 3.6 and 14°C. The partial Th_{CO₂} in these inclusions varies between 15 and 27°C and temperature of homogenization of these inclusion observed at 226 to 350°C (vanishing of liquid phase). The type III inclusions are homogenized by the disappearance of solid crystal first (195 and 337°C, corresponds to salinity of 30 to 41.5 wt.% NaCl equivalent), followed by the total homogenization (L_{H₂O} + V_{H₂O} + S → L_{H₂O}) recorded between 205 and 318°C (table 2).

3.2.2 Greisen quartz from Balda granite

The final ice melting temperature for type I inclusions varies from −7.3 to −3.3°C, corresponding to salinity values of 11 to 5.4 wt.%. Homogenized temperatures (to liquid) for type I inclusions are between 265 to 310°C. The CO₂ ice melting temperatures (Tm) for type II inclusions range from −60 to −56.7°C. The type II inclusions show lowering CO₂ melting (<−56.6°C) and higher CO₂ clathrate melting temperature (>10°C) indicates that they are CO₂ dominated with possible trace amount of dilutant of CH₄ or N₂ rich fluids. This is further confirmed by the laser Raman microscopy. Corresponding partial homogenized temperatures (Th) for type II inclusions recorded lie between 10 and 25°C. The total Th for these inclusions is between 280 and 318°C (vanishing of liquid phase). The type III is polyphase inclusions, which is homogenized by the disappearance of solid crystal between 110 and 210°C (corresponds to salinity of 28–32 wt.% NaCl equivalent). The temperature of homogenization varies from 168 to 214°C (L_{H₂O} + V_{H₂O} → L_{H₂O}; table 2).

3.3 Laser Raman microprobe

Vapour phases in the fluid inclusions were analysed by Thermo Almega XR dispersive Raman Spectrometer with 532 nm laser. The presence of H₂O, CO₂ and CH₄ in vapour phases is identified with typical Raman shift values. Hump in Raman spectra between 3000 and 3900 cm^{−1} suggests aqueous nature of fluid. Raman spectra at 1278/1382 and 1280/1384 cm^{−1} signify the presence of CO₂ and spectra at 2911 and 2909 cm^{−1} indicate presence of CH₄ (figure 3).

Figure 2. Photomicrographs of fluid inclusion in grey quartz from Degana and Balda W deposit. (a–d) Fluid inclusions in grey quartz from Degana granite and (e–h) fluid inclusions in grey quartz from Balda granite.

4. Geochemical characteristics of Degana and Balda granites

The geochemical results for Degana and Balda granites are having wide range of SiO_2 (68–74 wt.%) and a narrow range of alkali-oxide abundances. The Al_2O_3 content for these granites are 14 to 17 wt.%, whereas Fe_2O_3 content in Balda granite is relatively lower (0.6–1.3 wt.%) than Degana granite (2–8 wt.%). The samples of Balda and Degana granites fall in granite field (figure 5) when their normative mineral composition is plotted in the Quartz–Alkali feldspar–Plagioclase feldspar (QAP) classification diagram of Streckisen (1974). Among the trace elements of Ba and Sc, concentrations are very low and below the detection limit of WD-XRF (<10 ppm) except in two samples (D-70 and B-24). The Sr concentration is also low (55–186 ppm) in granites except in sample D-70 and B-24. The Rb content of Balda and Degana granites is quite high and ranges from 958 to 1907 ppm except for B-24, Zr content varies from 55 to 186 ppm, Nb is 15–21 ppm, and Y is 34–79 ppm. Different bivariate plots were prepared for SiO_2 vs. other major oxides to examine the composition of the mineralized granites. It is also observed that there is a small variations in $\text{Fe}_2\text{O}_{3(\text{tot})}$, K_2O and MnO , whose concentration decreases with increasing SiO_2 abundances (figure 6). Moniar and Piccoli (1989) showed that the variation of Al_2O_3 against SiO_2 indicates different tectonic setting distinguished from orogeny (higher values of Al_2O_3) to anorogenic setting (figure 7). The molecular proportion of $\text{Al}_2\text{O}_3/(\text{CaO}+\text{Na}_2\text{O}+\text{K}_2\text{O})$ in Balda

and Degana granites is >1 and peraluminous, which is the characteristic feature of S-type granite. The relative range of A/CNK ratio is a wider range in Degana granite as compared to Balda granite, which is attributed to greater proportion of aluminosilicate minerals in Degana than the Balda granite.

Another characteristic feature of these granites is the enriched Rb, Zr, Nb, Y and depletion of MgO , CaO , Sr and Ba and as a result Rb/Sr ratio is high (figure 8). The CaO content decreases with increasing silica which could be related to *in situ* crystal fractionation of plagioclase feldspar (Chappell and White 1974). Pearce *et al.* (1984) discriminated granites formed in different tectonic settings, based on the concentration of Rb, Y, Nb. The Ba content is very low in Degana and Balda granites as in tungsten mineralized granites reported from other regions, because of alkali feldspar fractionation and volatile enrichment in the parent magma (Singh and Singh 1997, 2001). It is also observed that all the samples of Balda granite and Degana granite fall in the syn-collision granite field (figure 9).

5. Geochronology

The Rb–Sr data obtained on Balda and Degana granites and minerals were plotted in separate isotope evolution diagrams using Isoplot 2.49 version for evaluation of their respective ages (Ludwig 2008). The Rb–Sr isotopic data for the whole rock samples of Balda and Degana granites and

Figure 3. Representative Raman spectra of fluid inclusions in grey quartz from Degana and Balda W deposit. (a) H₂O, CO₂ doublets and CH₄ spectrum of type II inclusion in Degana granite; (b) H₂O, CO₂ doublets spectrum of type II inclusion in Balda granite.

mineral separates are given in table 5. The Sm–Nd isotopic data for whole rock samples of Balda and Degana granites are given in table 5. Balda and Degana granite samples have much higher Rb abundances (544–1907 ppm) compared to Sr (98–147 ppm) and as a result ⁸⁷Rb/⁸⁶Sr ratios and ⁸⁷Sr/⁸⁶Sr ratios are also quite high. Muscovite and zinnwaldite have very high ⁸⁷Rb/⁸⁶Sr ratios (1150 and 1551), whereas feldspar has ⁸⁷Rb/⁸⁶Sr ratio of 10.75. The Sm concentrations in many samples are relatively high and therefore the ¹⁴⁷Sm/¹⁴⁴Nd ratio also higher than what is expected for typical

LREE enriched granites. The ε_{Nd} values calculated for these samples for an age of 800 Ma range from –11 to +6.90, where ε_{Nd} is defined by DePaolo (1981) as:

$$\epsilon_{Nd} = [10000 \times \{(\frac{^{143}\text{Nd}}{^{144}\text{Nd}})_{\text{sample}} - (\frac{^{143}\text{Nd}}{^{144}\text{Nd}})_{\text{CHUR}} / (\frac{^{143}\text{Nd}}{^{144}\text{Nd}})_{\text{CHUR}}\}] \quad (1)$$

Whole rock samples of Balda and Degana granites do not define isochron in Rb–Sr isotope evolution diagram. Magmas represented by Degana

Figure 4. Salinity *vs.* total homogenization temperature diagram illustrating the distribution pattern for the data points from the Degana and Balda granites.

Figure 5. QAP classification of plutonic igneous rocks (after Streckeisen 1974). The Balda and Degana granites fall in the granite field (medium-coarse grained granite, MCG), (coarse grained granite porphyry, CGP), (fine grained granite porphyry, FGP), (Balda granite, BG).

and Balda granites must have been highly heterogeneous in terms of $^{87}\text{Sr}/^{86}\text{Sr}$ and could have assimilated country rocks resulting in inhomogeneous $^{87}\text{Sr}/^{86}\text{Sr}$ initial ratios.

One of the samples of Degana granite (D-73) along with its pure mineral fractions of zinnwaldite and plagioclase defined an imprecise isochron corresponding to 843 ± 11 Ma with initial ratio of 0 ± 16 . It may be noted that the initial ratio is lower than the Basaltic Achondrite Best Initial (BABI) value of 0.69897 ± 0.000003 , which

represents $^{87}\text{Sr}/^{86}\text{Sr}$ initial ratio of the earth. The whole rock sample, D-73 may not truly represent Sr-isotope composition and $^{87}\text{Rb}/^{86}\text{Sr}$ ratio of the aliquot which was used for mineral separation. Such heterogeneities are expected from granite samples that have uneven distribution of micaceous minerals. Due to this $^{87}\text{Rb}/^{86}\text{Sr}$ initial ratio was brought down and error on the isochron, age is also high. When we consider only zinnwaldite and plagioclase, age obtained is 827 ± 8.2 Ma with initial ratio of $^{87}\text{Sr}/^{86}\text{Sr}$ is 0.7603 ± 0.0034

Figure 6. Harker's variation diagrams of SiO_2 vs. other major oxides in Degana and Balda granites (values are plotted in wt.%).

Figure 7. Variations diagram of SiO_2 vs. Al_2O_3 in Degana and Balda granites (after Moniar and Piccoli 1989). IAG: Island Arc Granite, CAG: Continental Arc Granite, CCG: Continental Collision Granite.

Figure 8. Ternary plot of Degana and Balda granites in Rb–Sr–Ba ternary diagram (after El-Bousely and Sokkary 1975).

Figure 9. Y+Nb vs. Rb tectonic discrimination diagram for Degana and Balda granites (after [Pearce et al. 1984](#)). Oceanic ridge granites (ORG), within-plate granite (WPG), volcanic arc granite (VAG) and syn-collisional granites (syn-COLG).

(figure 10b). Although this represents a two-point isochron the age obtained is valid because zinnwaldite has very high $^{87}\text{Rb}/^{86}\text{Sr}$ ratio of 1550.83 and $^{87}\text{Sr}/^{86}\text{Sr}$ is 19.081. In view of this if any reasonable initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio is assumed for the crustally derived magma, the age will remain same within its analytical uncertainty. Hence, the above two-point isochron, age of 827 ± 8.2 Ma represents the time of crystallization and closure of zinnwaldite and plagioclase for Rb–Sr isotopic system in the Degana granite (D-73; figure 10b). The Balda granite (B-34) was also subjected to mineral separation and isotopic data of pure fractions of muscovite, plagioclase along with Balda granite is given in figure 10(a). The three points are collinear; the slope corresponds to 795 ± 11 Ma with initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 1.193 ± 0.077 . It is also noted that MSWD is 1.6 and thus isochron age represents the time of crystallization of Balda granite.

The Rb–Sr isotope system has Rb–Sr closure temperature of around 400°C for biotite and 50°C higher for phlogopite ([Jenkin et al. 2001](#); [Willigers et al. 2004](#); [Nebel and Mezger 2008](#)). Zinnwaldite, also trioctahedral mica, is expected to have same closure temperature as that of phlogopite. Thus the Rb–Sr isochron age based on zinnwaldite and plagioclase for the Degana granite sample (D-73) represents time of cooling of the granite body to $<400^\circ\text{C}$. Several studies have shown that igneous rocks that were neither subjected to metamorphism nor heated to temperature $>400^\circ\text{C}$ give Rb–Sr biotite or phlogopite ages that match with U–Pb zircon ages within analytical uncertainties ([Nebel](#)

Table 5. Rb–Sr and Sm–Nd isotopic composition of Balda and Degana granites.

Sample no.	Rb (ppm)	Sr (ppm)	$^{87}\text{Rb}/^{86}\text{Sr}$	$^{87}\text{Sr}/^{86}\text{Sr}$	2σ (error) $\times 10^{-6}$	Sm (ppm)	Nd (ppm)	$^{147}\text{Sm}/^{144}\text{Nd}$	$^{143}\text{Nd}/^{144}\text{Nd}$	2σ (error) $\times 10^{-6}$	ϵ_{Nd} ($t = 800$ Ma)	$f_{\text{Sm}/\text{Nd}}$	T_{DM} (Ma)
B-24	1064	147	71.18	2.27747	1	0.4	2	0.33	0.512778	6	-11.1	0.6838	-485
B-34	958	124	179.33	1.236444	7	1	3	0.18	0.512516	6	-0.2	-0.1099	2491
B-37	544	139	23.85	0.822536	1	1	3	0.15	0.512627	5	4.8	-0.2465	1216
D-50	1850	98	190.06	4.66187	4	8	29	0.17	0.512148	6	-7.2	-0.1256	3659
D-51	1110	112	77.94	7.97979	3	10	40	0.18	0.512362	4	-3.9	-0.0755	3737
D-52	1171	117	94.44	3.12488	3	13	42	0.20	0.512426	3	-4.9	0.0363	10840
D-57	172	97	165.99	19.1422	7	6	32	0.10	0.512348	6	3.7	-0.4668	1123
D-68	1194	113	155.30	12.4332	6	7	40	0.11	0.512544	4	6.9	-0.4338	903
D-69	1060	125	102.08	1.78254	2	14	69	0.14	0.512563	1	4.8	-0.3107	1145
D-70	1213	107	112.42	14.0149	9	6	28	0.14	0.512011	6	-6.0	-0.3107	2478
D-72	1907	104	130.49	6.65737	3	11	30	0.18	0.512463	2	-2.1	-0.0654	3479
D-73	1029	122	552.01	5.52213	8	9	45	0.12	0.512476	3	4.3	-0.3715	1140
B-34 Muscovite	1410	208	1150.02	14.226	1								
B-34 Plagioclase	229	42	10.75	1.275142	1								
D-73 Zinnwaldite	1521	74	1550.83	19.0806	7								
D-73 Plagioclase	225	2.0	20.44	1.0018	1

Figure 10. (a) Rb–Sr isochron plot of plagioclase+ muscovite+Balda granite; (b) Rb–Sr mineral isochron plot of zinnwaldite+plagioclase mineral isochron in Degana granite.

et al. 2011). As the Degana and Balda granites were not metamorphosed, the Rb–Sr zinnwaldite-feldspar or muscovite-whole-rock-feldspar isochron ages represent time of igneous cooling of these bodies to $<400^\circ\text{C}$. Because the size of these granitoid bodies are small ($<4\text{ km}^2$) they must have cooled to $<400^\circ\text{C}$ within a million year from the time of their intrusion. Hence, we consider that Rb–Sr isochron ages reported above are robust and date the time of igneous intrusion of the Degana and Balda granites.

Sm–Nd isotopic analysis was carried out on three whole rock samples of Balda granite and nine whole rock samples of Degana granite (table 5). Both Degana and Balda samples do not define any meaningful correlation in the isochron diagram, indicating that their magmas were characterized

by heterogeneous initial $^{143}\text{Nd}/^{144}\text{Nd}$ ratio inherited either by partial melting of diverse sources or due to variable extents of crustal assimilation.

6. Discussion

6.1 Hydrothermal process of mineralization

In the greisen quartz of Degana granite, T_h of type I inclusions ranges between 208 and 347°C and the salinity ranges from 16 to $9.2\text{ wt.}\%$ NaCl equivalent, and in the type II inclusions, T_h ranges from 228 to 350°C and the salinity from 11 to $4\text{ wt.}\%$ NaCl equivalent. Similarly, in the Balda granite, the type I inclusions are homogenized between 265 and 310°C , and the salinity ranges from 11 to $7.2\text{ wt.}\%$ NaCl equivalent, and the type II inclusions are homogenized between 280 and 318°C and the salinity is $<4\text{ wt.}\%$ NaCl equivalent. In the type III inclusions, daughter crystal is dissolved first, followed by the disappearance of vapour bubble. The co-existence of type I to type III inclusions and the comparable range of T_h with different mode of homogenization to liquid in type I inclusion, to vapour in type II inclusion are evidences of fluid immiscibility during hydrothermal process (figure 4).

Fluid immiscibility refers to unmixing of originally homogenous fluid into two phases of contrasting physical and chemical parameters. Hence, the ore fluid is interpreted to be essentially aqueous carbonic fluid, and may be defined as a H_2O – NaCl – $\text{CO}_2 \pm \text{CH}_4$ system. The close association of CO_2 -rich inclusions and H_2O -rich inclusions in groups and along the same trail also suggests fluid immiscibility. It is interpreted that fluids of type I and type II inclusions co-existed during mineralization and type III inclusions indicate increasing salinity of hydrothermal fluid as a result of fluid–rock interaction leading to greisenisation (Vijay Anand *et al.* 2010; Krylova *et al.* 2012), which also contributed to ore mineralization. According to Pearson’s hard soft acid base (HSAB) principle, tungsten which occurs as WO_4^{2-} in hydrothermal fluid is a hard base and readily combines with hard acid H^+ to form H_2WO_4 (tungstic acid) in hydrothermal solution. Therefore, tungsten is transported in acidic hydrothermal solution (Giggenbach 1997). Greisenisation is a neutralization reaction in which H^+ is consumed in the conversion of alkali feldspars to zinnwaldite or

Figure 11. The Ab–Or–Qz system with the ternary cotectic curves and eutectic minima from 500 to 5000 bar, showing plot of Balda and Degana granites (MCG, CGP, FGP). Note the effects of increasing pressure and the water-saturated liquids in equilibrium with quartz and alkali feldspar at different confining pressure (Winter 2001).

muscovite. Such reaction destabilizes H_2WO_4 resulting in precipitation of wolframite.

6.2 Petrogenesis

The Balda and Degana granites are associated with tungsten mineralization and therefore it is important to understand their petrogenesis. The Balda granite samples fall close to the ternary minima corresponding to $\text{P}_{\text{H}_2\text{O}}$ (water saturated conditions) values of 0.5, 1, 3, 5 kb, whereas few samples of Degana granites fall close to the minima in normative albite-orthoclase-quartz ternary diagram (Winter 2001). Thus magmas representing Balda granites and some of the Degana granites could have been generated by partial melting of continental crustal sources under water saturated conditions. Whereas, other Degana samples plot in the primary phase field of K-feldspar indicating that their magmas could have been generated either at higher temperature than the ternary minima or by partial melting of sources under relatively dry conditions (Winter 2001; figure 11).

Trace elements of Rb, Sr and Zr abundances in magmas generated by partial melting of continental crust (average composition given in Rollinson 1993) and fractional crystallization of plagioclase

were models to infer nature of sources and dominant magmatic processes. Mineral-melt distribution co-efficient values from Arth (1976), Pearce and Norry (1979) were used and batch melting of continental crust source leaving residue consisting of 50% plagioclase, 40% clinopyroxene and 10% orthopyroxene was assumed. The results of trace element modeling in granite system are shown in figure 12(a, b).

In Degana and Balda granites, Sr abundances are much lower and Rb abundances are higher that has been calculated in melts formed by partial melting of continental crust. The high Rb content indicate that it behaves as an incompatible element during partial melting and neither mica group of minerals nor alkali feldspar was present in the residue in significant quantities. Although fractional crystallization of plagioclase could explain lower Sr abundances in the Balda and Degana granites, Rb concentration is much higher and cannot be explained by this process. Therefore, basaltic rocks could not be the source for magma representing these granites. Rb and Sr abundances calculated for different extents of partial melting of the continental crust are much closer to that of granites (figure 12a).

The range in Rb content is nearly same except that few samples of Degana granite have higher

Figure 12. (a) Trace element concentrations (Sr vs. Rb) of Degana and Balda granite comparison to partial melting and fractional crystallization of continental crust by leaving a basaltic residue of Plg:Cpx:Opx = 50%:40%:10%. (b) Trace element concentrations (Sr vs. Zr) of Degana and Balda granite comparison to partial melting of continental crust by leaving a basaltic residue of Plg:Cpx:Opx = 50%:40%:10%.

values. The Sr abundances are somewhat lower by about 50–100 ppm compared to the model melt composition. This difference can be explained if magma representing these granites had undergone fractional crystallization of plagioclase feldspar or source had lower Sr concentration than assumed. Compared to the Degana granite, the Balda granite represents magma formed by higher extents of melting (figure 12a). Thus magmas representing Degana and Balda granites could have been generated by partial melting of continental crustal rocks and have undergone fractional crystallization of plagioclase.

Zr and Sr abundances for the Degana and Balda granite samples show a positive correlation, whereas, melts calculated for different extents of partial melting of average crust shows a negative trend (figure 12b). This is because Sr was assumed as a compatible element, while Zr as an incompatible element. However, zircon is usually present as an accessory mineral within the continental crustal rocks and partial melting of these rocks under hydrous conditions (700–800°C) would have resulted zircon in the residue. The granite magmas formed would have been saturated with zircon and fractional crystallization of zircon

Figure 13. (a) $Nd_t - f_{Sm/Nd}$ plot for Degana granite (MCG, CGP, FGP) and Balda granite; (b) $Sr_t - Nd_t$ plot for Degana and Balda granites; CHF: Contaminated Hydrothermal Fluids, ACC: Archean Continental Crust, CHUR: CHondrite Uniform Reservoir. Abbreviations. $f_{Sm/Nd}$: fractionation factor Sm/Nd; T_{DM} : T-depleted mantle; Sr_t : Initial value of Sr; Nd_t : Initial value of Nd.

could not be ruled out. Hence, Zr would have also behaved as compatible element and magma representing Degana and Balda granites had equilibrated with zircon and plagioclase confirming their continental origin (Ito *et al.* 1987).

The samples of Balda and Degana granites were plotted in ϵNd_t and ϵSr_t diagram (figure 13b). One of the Balda granite and four Degana granite samples have positive ϵNd values of +3.7 to +6.9 and ϵSr close to zero. Magma representing these samples must have been derived from sources that had long term depletion of LREEs such as depleted mantle. The other samples have variably negative ϵNd values and positive ϵSr values and show considerable spread, which could be explained by mixing of long term LREE depleted sources and LREE enriched sources, such as depleted mantle and upper continental crust. The possibility of such mixing is also indicated by curve I and curve II, wherein magmas representing samples falling along curve I could have been derived by partial melting of mixed sources having long term LREE depleted and enriched rocks with high Rb/Sr ratios (figure 13b). Samples falling along the curve II and also represents magmas derived by partial melting of mixed sources as mentioned above however, having low Rb/Sr ratios. If granite magmas formed from long term LREE depleted sources had undergone assimilation of old continental crustal rocks then they will resemble samples falling along the curve I. However, extent of contamination required will be quite high. Therefore, magmas representing Degana and Balda granites could have

been derived by partial melting of LREE depleted basaltic rocks and LREE enriched lower and upper crustal rocks with low and high Rb/Sr ratios, respectively.

The granite samples with positive ϵNd values are light REE enriched and therefore have negative $f_{Sm/Nd}$ values, where

$$f_{Sm/Nd} = \frac{[(^{147}Sm/^{144}Nd)_{sample} - (^{147}Sm/^{144}Nd)_{CHUR}]}{(^{147}Sm/^{144}Nd)_{CHUR}}. \quad (2)$$

Although their magmas could have been derived by partial melting of long term LREE depleted sources, at the time of melting they were enriched in LREE and additionally as a result of low extents of partial melting of such sources, granitic magmas were further enriched in LREE. The samples that have variable negative ϵNd values were derived by partial melting of long term LREE enriched sources such as Archean or Paleoproterozoic continental crust (Chemale *et al.* 2011). However, one of the samples of Balda granite (B-24) has highly negative ϵNd value of -11.1 and also has high Sm/Nd ratio and falls in the forbidden quadrant in ϵNd vs $f_{Sm/Nd}$ diagram (figure 13a and b). This unusual character of this sample could be a result of extensive alteration by hydrothermal fluids, which had changed its Sm/Nd ratio as suggested by Zacharia (1996) for metavolcanic rocks of the Ramagiri greenstone belt of the Dharwar craton, India.

7. Significance of Neoproterozoic magmatism (900–700 Ma)

The present study has yielded ages of 795 ± 11 Ma for Balda granite and 827 ± 8 Ma for Degana granite. The acid magmatism during 900–700 Ma in Aravalli craton is significant time for east Gondwana amalgamation, continental dynamics and ore genesis.

The MIS is one of the largest felsic provinces in NW India and characterized by three different phases of magmatic activity—basaltic and felsic volcanic flows (Phase I), granitic intrusions (Phase II) and felsic-mafic dyke complexes (Phase III, Bhushan 2000; Pandit *et al.* 2003; Just *et al.* 2011). It is related with crust melt during extension and/or an active hotspot (Meert and Lieberman 2008). Many authors have studied about MIS magmatic activity and most of the researchers suggested as it can be Andean type active continental margin (Meert 2001; Torsvik *et al.* 2001a,b; Ashwal *et al.* 2002; Gregory *et al.* 2009). Similarly, the reported ages of 800 Ma for tonalitic basement rocks, Sindreh felsic volcanics and Harsani granodiorites in NW India have comparable ages with Balda and Degana granite (Maheshwari *et al.* 2001; van Lente *et al.* 2009; Pradhan *et al.* 2010).

The Neoproterozoic magmatism in NW India can be compared with equivalent Neoproterozoic igneous activities in Australia, Seychelles, Madagascar based on geochemical and geochronological similarities (Torsvik *et al.* 2001b; Ashwal *et al.* 2002; Pandit *et al.* 2003; Collins 2006; Pradhan *et al.* 2010). In Australia, Neoproterozoic magmatism is reported of 850–780 Ma (Black *et al.* 1997; Turner *et al.* 1998), similar age of volcanic rocks in the mainland and Rb–Sr dating of mafic dykes is 850 Ma reported in Western Australia are the products of crustal melting of above a mantle plume (Robey *et al.* 1989).

The Neoproterozoic age ranges for granitoids and mafic dyke complex in Seychelles and NE Madagascar is attributed to subduction of Mozambique Ocean (Santosh *et al.* 1989; Radhakrishna and Mathew 1996; Handke *et al.* 1999; Bhushan 2000; Miyazaki *et al.* 2000; Torsvik *et al.* 2001b; Tucker *et al.* 2001; Ashwal *et al.* 2002; Pandit *et al.* 2003) and it is suggested as Andean type active continental margin related with coeval arc activity in Seychelles and Madagascar (Windley *et al.* 1994; Collins and Windley 2002; Collins 2006).

Ashwal *et al.* (2013) have also reported that Malani Igneous Suite (MIS) and Praslin Group granitoids of the Seychelles, along with North Madagascar formed now fragmented components of an Andean-type magmatic arc on the margin of the Rodinia supercontinent. The tectonic settings for Malani magmatic activity is suggested to have generated by a hotspot source or lithospheric melting and thinning at the base of the crust (Sharma 2004). The magmatic rocks of the MIS and Mt. Abu granites are part of this arc, and suggested to have been contiguous with 700–800 Ma arc-related magmatic rocks of the Seychelles and Madagascar. The earlier episode was well developed in India, Australia, South China, Seychelles and Madagascar and the latter phase spread to South Africa (Li *et al.* 2003). The possible explanation for these magmatic episodes is mantle plume under the Rodinia supercontinent that eventually led to its breakup during Neoproterozoic (Preiss *et al.* 1978; Choudhry *et al.* 1984; Buhn and Stanistreet 1997; Walter and Veevers 1997; Preiss 2000; Wingate and Giddings 2000; Li *et al.* 2003; Meert 2003; Pandit *et al.* 2003; Wang and Li 2003; Li *et al.* 2008; Meert and Lieberman 2008; van Lente *et al.* 2009; Ashwal *et al.* 2013; Jin *et al.* 2016).

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