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Characterization of Mineralized and Barren quartz veins based on wall rock alteration and fluids inclusion studies: Case study from Degana tungsten deposits, Nagaur District, Rajasthan, India.

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Abstract: Tungsten Mineralization at Degana, North-Western India is associated with the Neoproterozoic Degana granite. The tungsten mineralization is mainly confined to the quartz veins with minor mineralization in disseminated forms and as stockworks in brecciated granite and phyllite. Mineralized quartz veins (MQVs) and post mineralized barren quartz veins (BQVs) are recognized based on theirfield relations, veins mineralogy and associated wall rock alteration. In the field, the BQVs show greater continuity and cut across the MQVs, suggest that they are younger than MQVs. Wolframite is the main ore mineral present in the MQVs, occurring as sparsely and sporadically distributed crystals or pockets. The MQVs are coarser in texture than BQVs and are composed of grey quartz, zinnwaldite, topaz, fluorite with wolframite and minor sulphides. The MQVs are bordered by greisen, silicification and potassic alteration zones which are characterized with altered mineral assemblages of secondary quartz, perthites, muscovite, biotite, zinnwaldite, topaz and fluorite. In contrast the BQVs are composed of light pink coloured quartz with plagioclase, muscovite, biotiteand locally secondary fluorite and topaz and lacks significant wall rock alteration. The MQVs are characterized by the presence of abundant multiphase and carbonic fluid inclusions, whereas BQVs contains aqueous inclusions with fewer multiphase and carbonic fluid inclusions. Micro-thermometric analysis indicates high homogenization temperature (T_h=335-487 °C) and high salinities (42-70 eq. wt% NaCl) in the MQVs and lower values in the BQVs. The abundance of CO_2-rich inclusions in the MQVs is explained by CO_2 effervescence, which causes the deposition of tungsten. Low salinities and temperature in the BQVs reflects mixing and dilution of the hydrothermal fluid by cooler meteoric waters, and the depletion of CO_2. The fluid inclusion data, wall rock alteration styles clearly discriminate the MQVs from BQVs.

Key words: barren quartz; Degana tungsten deposits; mineralized quartz; Nagaur; Rajasthan.

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1. Introduction

A 500 km long W-Sn belt extends from Balda in Rajasthan to Tosham in Haryana, North-Western India (Bhattacharjee *et al*., 1993). This belt hosts significant W-Sn deposits at Balda–Kalni–Kotaria, Degana and Tosham. At Degana, tungsten mineralization is associated with the A-type, per-aluminous, "Within Plate" granite (Chattopadhyay *et al*., 1994; Sukh Chain, 2004) which intrudes Pre-Cambrian meta-sedimentary rocks.

In this study we distinguish mineralized quartz veins (MQVs) from the barren quartz veins (BQVs) on the basis of Wall rock alteration and fluid inclusion study to constrain the nature of fluid responsible for the formation of mineralized and barren quartz veins.

Geological Setting

The present area forms a part of the Aravalli-Delhi metallogenic province in North Western India. The Precambrian rocks of this province have been subjected to two major orogenic events during the Proterozoic. They are represented by two major fold belts in the region namely Aravalli fold belt and Delhi fold belt, which comprise the Aravalli and Delhi Supergroups.

The Delhi fold belt contains a number of granite plutons ranging in age from 1600 m.ato 730 m.a. (Tobisch, 1994, Choudhary *et al.*, 1984). This late Proterozoic felsic magmatism commenced with the intrusion of the Erinpura

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granite (900 m.a., Choudhary *et al.*, 1984). Sinha Roy *et al.* (1998) suggested that emplacement of the Erinpura granite and its equivalent along and adjacent to the South Delhi fold belt was caused by intense compressive tectonism that followed the Delhi Supergroup sedimentation.

The rocks outcrops in the Degana area are confined to three small hillocks, locally named as the Rewat, Tikli and phyllite hills in an otherwise flat terrain covered by a thick blanket of sand and alluvium. The oldest rocks exposed in the area are represented by meta-sedimentary rocks of the upper Proterozoic Delhi Supergroup and includes phyllites with a few local quartzite band. The Degana pluton is represented by medium to coarsegrained granite, granite porphyry and rhyolite. Pandian and Verma (2010) have obtained 860±25 m.a. age for the Degana granite. The pluton is compositionally homogenous but shows various textural varieties. The present tungsten deposit is characterized by wide spread wall rock alteration which is found throughout the pluton. All the granite types in the area are characterized by the presence of very thin feldspathic veins (>1 cm wide) and dark coloured greisen veins (>5 mm wide) which indicate post magmatic alteration effects in the granite body. Aplite, numerous quartz veins and few pegmatite veins transverse both the granite body and country rocks. The pegmatite consists of quartz, plagioclase and muscovite. In the granite, mineralized quartz veins have intruded mostly along the joint plains with a NW-SE trend.

Wall rock alteration

Mineralized quartz veins (MQVs) are bordered by thick mica rich greisen selvages consisting grey quartz, zinnwaldite, topaz, fluorite with wolframite and minor sulphides (pyrite, pyrrohotite, arsenophyrite and chalcopyrite). The barren quartz veins (BQVs) are pinkish in colour and are composed of light pink quartz with plagioclase, muscovite, biotite and locally secondary fluorite and topaz.

Based on field relationship, petrographic features and geochemical characteristics, Sukh Chain and Srivastava (2005) identified three alteration zonesiegreisen, silicification and potassic out wards from the MQVs. These alteration zones do not show sharp boundaries but lateral variations in mineralogy and texture have been used as criteria for distinguishing these alteration zones in the field. A systematic mineralogical, geochemical and textural variation is observed in each alteration zones. The altered minerals assemblage of the greisen zones are quartz, muscovite, zinnwaldite, topaz, fluorite, wolframite + altered feldspars. Topaz is more abundant than fluorite in this zone. In contrast potassic zone is characterized by the development of fresh microcline, the breakdown of feldspars and biotite and the dominance of fluorite than topaz.

2. Material and methods

Fluid inclusion study

Samples of quartz from both MQVs and BQVs were collected and sections for fluid inclusions micro-thermometric analysis were prepared. The identification of primary, psedo-secondary and secondary inclusions was based on the criteria of Roeder (1981) and only primary and psedo-secondary inclusions were chosen for analysis. Micro-thermometric measurements were carried out at Jammu University on a Linkam THMSG-600 heating and freezing stage mounted on a Nikon microscope with long working distance objective lenses. Inclusions that showed sign of leakage or necking were not analysed. The fluid inclusions data were checked by repeating the experiment. In general, temperature recorded during heating experiment was repeatable to ±2 °C. The heating and freezing data of different types of the inclusions is summarized in **Table-1**.

3. Results

Fluid inclusion petrography and Heating and freezing Data

Different inclusions assemblages are present in the quartz from MQVs and BQVs. Based on the phases present in the inclusions at room temperature, 4 types of inclusions are recognized and are discussed below.

Multiphase Inclusions (Type-I): These inclusions are characterized by the presence of 1 or more daughter crystals; they occur mostly as primary and psedo-secondary types and are mostly present in the MQVs. Their size varies from 10 -25 μm and have rounded shapes **(Fig., 1a).** Three different daughter crystals (halite, sylvite and rarely anhydrite) were identified in multiphase inclusions in the MQVs. In the BQVs halite is the only solid phase present.

Table 1. Heating and Freezing data of the different inclusion types in the Mineralized (M) and Barren (B) quartz veins of Degana area.

Temperature (°C) of various attributes		Multiphase (Type-I)	CO_2 Monophase (Type-II	Bi-phase (H_2O-CO_2) Type-III		Aqueous bi-phase (Type-IV)
				CO_2-rich	H_2O-rich	
Homogenization (T_h)	M	300-465	254-375	260-408	255-375	155-245
	B	300-325	250-350	275-300	265-395	145-185
Clathrate (Tm. Clath)	M	-	-	+ 1.0 to +5.0	-	-
	B	-	-	+ 1.5 to + 4.0	-	-
CO_2 Homogenization ($T_{h.CO2}$)	M	-	- 4.0 to +11.0	+16.0 to +23.5	-	-
	B	-	- 6.5 to +13.0	+12.0 to +26.0	-	-
Final Ice melting (Tm.ice)	M	-	-	-	-	- 9.5 to -22.5
	B	-	-	-	-	-7.5 to -20.5

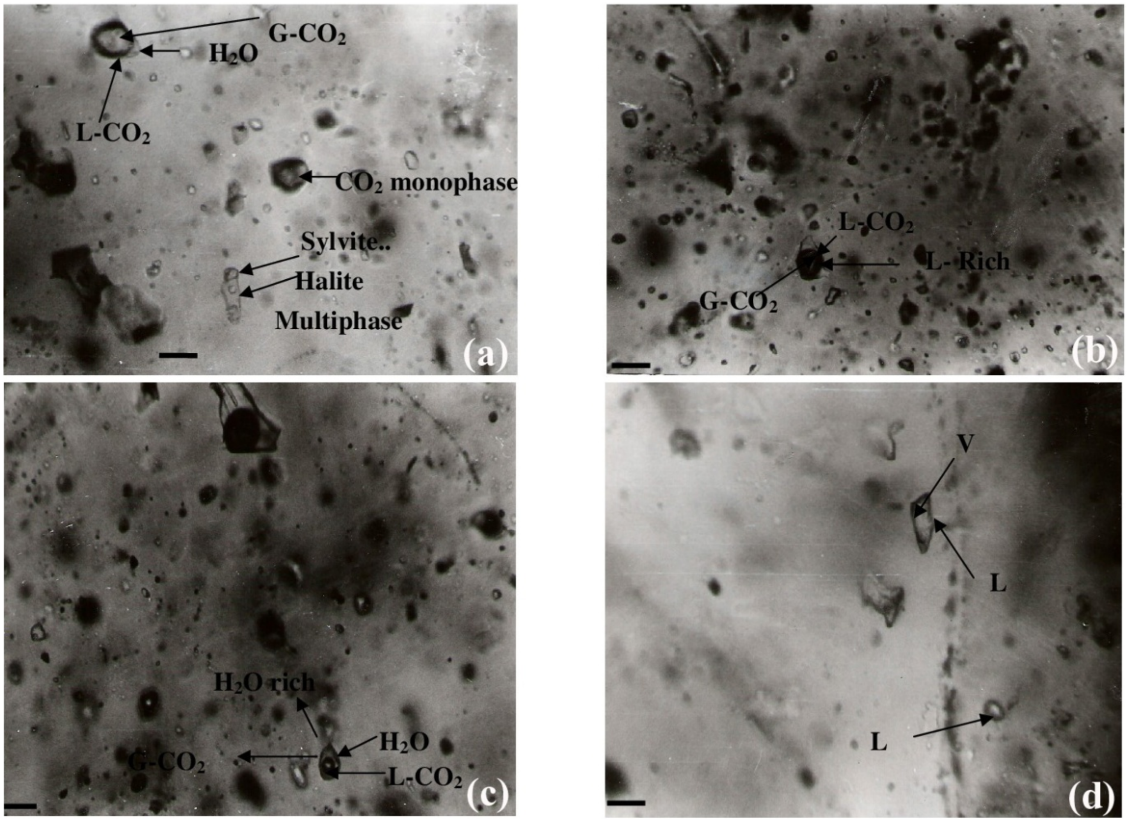

Fig.1(a-d): (a)-Micro photograph of Multiphase inclusion, (b) Microphotograph of CO_2 Mono phase inclusion, (c) Microphotograph of H_2O-CO_2 Bi-phase inclusions, (d) Aqueous bi-phase inclusions. The length of bar is 1 μm

The dissolutions temperature of halite ($T_{d.halite}$) ranged from 307–415 °C in the MQVs and between 310 and 390 °C in the BQVs, whereas sylvite dissolved (Td.sylvite) between 160 and 265 °C in MQVs. The total homogenization temperature (T_h) of these inclusions ranged from 300–465 °C in MQVs mostly falling in the range of 350 to 440 °C. In BQVs these inclusions homogenized between 325 and 376 °C, with a prominent peak between 300 and 325 °C. **(Fig., 2a**). All the multiphase inclusions homogenized into the liquid phase.

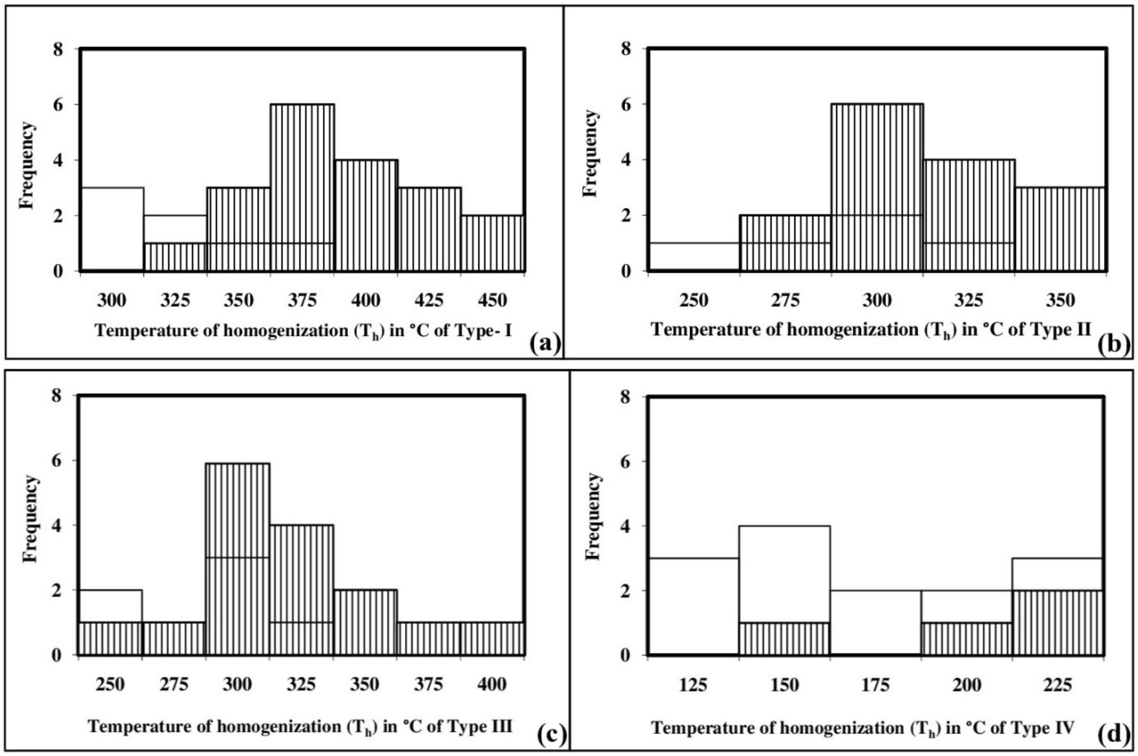

Fig.2(a-d). (a) Type- I; (b) Type- II; (c) Type-III, and (d) Type- IV. Figures showing homogenization temperature pattern of different inclusion types in MQVs and BQVs. Empty bars represent the Mineralized and line bars represents barren quartz veins.

Mono-phase CO_2 Inclusions (Type-II): These inclusions contain 90 - 95 % gas (**Fig., 1b**) such inclusions are more abundant in the MQVs than in the BQVs. The shape of the cavity is mostly elliptical and sometime irregular with size varying from 5 to 15 µm. These types of inclusions are rare in the BQVs.

$T_{m.CO2}$ in the MQVs ranged between -60.1 to -63 °C, where as in the BQVs it ranged between -60.1 to – 61.5 °C. Depressions of the expected temperature of CO_2 melting ($T_{m.CO2}$) in MQVs is slightly more than in the BQVs and is likely due to the presence of some other gas phase such as CH_4 or N_2. The CO_2 component homogenized ($T_{h.CO2)}$ into the vapour phase between -4 to +11 °C in the MQVs and -6.5 to +13 °C in the BQVs **(Fig., 3a)**. On further heating homogenization of these inclusions in the MQVs (mostly to the liquid phase) ranged from 254 to 375 °C with the majority of inclusions homogenized in between 300 and 350 °C. In the BQVs these inclusions homogenized into the vapour phase in between 250-350 °C with the majority homogenizing between 250-300 °C **(Fig., 2b)**.

H_2O-CO_2 (Bi-phase) inclusions (Type-III): these inclusions are characterized by the presence of two immiscible phases of CO_2 and H_2O at room temperature. These inclusions mostly occur as isolated primary inclusions in the mineralized veins with sub-rounded to elliptical shapes **(Fig., 1c)**. The size of such inclusions varies from 10 - 25 µm. In these inclusions, the H_2O –CO_2 ratio varies from inclusions to inclusions. Based on the proportion of CO_2 and H_2O, these inclusions are of two sub types, CO_2 rich bi-phase and H_2O rich bi-phase.

The CO_2-rich bi-phase inclusions are abundant among all types of inclusions present. They are mainly primary inclusions, and are present mostly in the MQVs and less commonly in BQVs. Their size varies from 10 - 20 µm. The CO_2 and H_2O in these inclusions are immiscible at room temperature (25 °C). These inclusions contain variable gas contents, which are normally much less, in comparison to liquid (CO_2+ H_2O)**.** The liquid CO_2 inclusions are more common in the MQVs than BQVs.

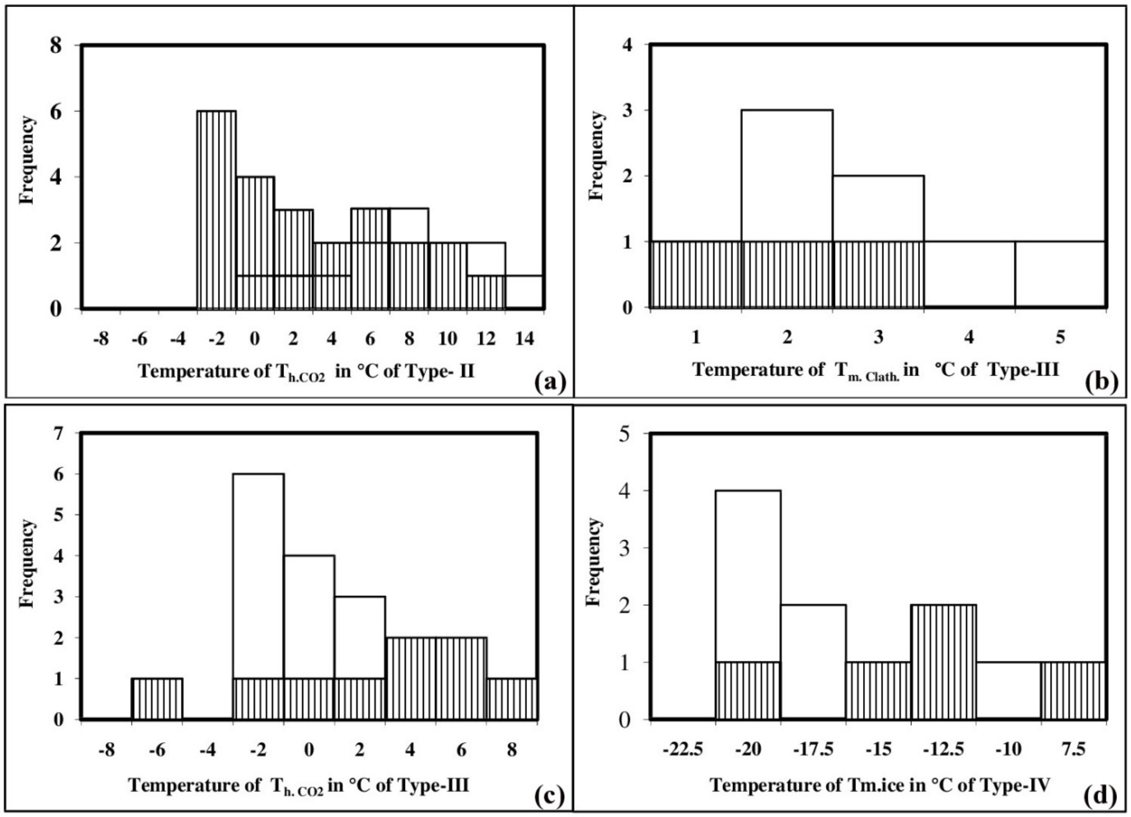

Fig. 3(a-d). Figures showing freezing data pattern of different inclusion types in MQVs and BQVs. Empty bars represent the Mineralized and line bars represents barren quartz veins.

These inclusions typically show two immiscible liquids, aqueous liquid and liquid CO_2 and a CO_2 rich gas phase. The Tm.$_{CO2}$ of these inclusions ranged between -60 to -60.5 °C in the MQVs and between -60 to -61 °C in BQVs. On further heating the ice completely melted, but the vapour bubble remained distorted in shape by the presence of solids Clathrate. The Clathrate dissociates between +1 to +5 °C ($T_{m.clath.}$) in MQVs and between +1.5 to +4 °C in BQVs (**Fig., 3b**). The CO_2 homogenized into the liquid phase between + 16 to + 23.5 °C (T_{hCO2}) in the MQVs mostly between + 4 to 16^0 C and between +12 to +26 °C in the BQVs (**Fig., 3c**). In both types of veins majority of these inclusions homogenized finally into the liquid phase, CO_2 rich inclusions (Type-IIIA) in the MQVs homogenized between 260 – 408 °C and between 275 - 300^0 C in BQVs **(Fig., 2c**). The H_2O-rich CO_2 –H_2O inclusions (Type-IIIB) homogenized in to the vapour phase between 255 – 375 °C in the MQVs and 265 - 395 °C in BQVs.

Aqueous Inclusions (Type-IV): Aqueous inclusions are abundant in BQVsand are uncommon in MQVs. These inclusions are characterized by the presence of H_2O in liquid and vapour phase. In these inclusions, the liquid –gas ratio is variable but liquid is always more than vapour. The shape of the cavity is mostly irregular (**Fig., 1d)** and their size is highly variable ranging from less than 5 - 30 µm. In the MQVs, the degree of fill ranges between 0.7 - 0.9, where as in the BQVs it ranges between 0.25 - 0.45. At places they are seen cutting across the trails of the other types of inclusions. This indicates that the aqueous inclusions trappedat a late stage after the entrapment of other types of inclusions.

During freezing studies of these inclusions liquid first appeared between -38 .5 to -40.0 °C (T_{FM}) in the MQVs and -38 to -39 °C in BQVs. Further heating final ice melting ($T_{m.ice}$) was noted between -9.5 to -22.5 °C in MQVs, and between -7.5 to +20.5 °C in BQVs (**Fig., 3d**). In these inclusions the vapour bubble gradually disappeared during heating and the inclusions homogenized finally into the liquid phase. The homogenizing temperature of these inclusions ranged between 155 and 245 °C in the MQVS, whereas in the BQVs these inclusions homogenized at lower temperature from 145-185 °C **(Fig., 2d).**

Composition and Density of Fluid

The salinity of the multiphase inclusions was determined using the solid dissolution method of Sourirajan and Kennedy (1962). The total salinity of the inclusions containing the halite and sylvite solid phases was determined using the H_2O-NaCl-KCl Model system by using the dissolution temperature of NaCl and KCl. The density of multiphase inclusions was determined using the equation of state of Klevtsov and Lemmlein (DATE). The salinities of H_2O-CO_2biphase inclusions were calculated from the clathrate melting temperature using the equation of Collins (1979). $T_{h.CO2}$ was used to calculate salinity of CO_2-rich inclusions and densities of Type-II and Type-III were calculated from $T_{h.CO2}$ using the equations of Valakovicbh and Altumin (1968). The salinity of aqueous inclusions was determined using the final ice melting ($T_{m.ice}$) using the equation of Crawford (1981). The density of inclusions was calculated from degree of fill using the equation of Shepherd *et al.* (1985).

The salinity of multiphase inclusions in MQVs ranged between 52 - 70 eq. wt.% NaCl, whereas, it was slightly lower in the BQVs (39-42 eq. wt.% NaCl). The density of the multiphase inclusions in the MQVs ranged between 1.1 and 1.35 g/cm^3, whereas in the BQVs the density was 1.02 -1.08 g/cm^3. The salinities of H_2O-CO_2 bi-phase inclusions ranged between 10-15 eq. wt.% NaCl in MQVs and 9-12 eq.wt.% NaCl in BQVs. The density of Type-II inclusions varies from 0.88-0.94 g/cm^3 in MQVs and in BQVs the density 0.65-0.80 g/cm3.The density of the H_2O-CO_2 bi-phase inclusions in MQVs and BQVs ranged between 0.84-0.77 g/cm^{3}and 0.54-0.75g/cm^3 respectively. The density of the aqueous inclusions in the MQVs and BQVs ranged between 1-1.05 g/cm^3 and 0.98-1.02 g/cm^3 respectively.

4. Discussion

Two types of quartz veins present in the Degana tungsten deposits shown contrasting mineral assemblages. The mineralized quartz veins (MQVs) are bordered by zinnwaldite-rich greisen, whereas the BQVs are accompanied by muscovite alteration with little or no zinnwaldite. Three different types of wall rock alteration are associated with MQVs in the Degana tungsten deposits. Greisenization directly related to the action of volatile rich hydrothermal fluids on the host rock. The common presence of miarolitic cavities in the Degana granite indicates that hydrothermal fluids were ex-solved from the magma during the late stages of cooling. The presence of stockworks, quartz veins and post-magmatic changes in the host rock indicate the evolution of an aqueous phase during crystallization of the Degana granite.

It is suggested that the separation of an aqueous phase from the crystallizing magma took place simultaneously with the crystallization of coarse grained granite, leaving behind a dry melt to form aplite dykes. Type –I inclusions shows high homogenization temperatures (335-487 °C) and high salinities (45-70 eq.wt.% NaCl), which supports a magmatic origin of ore fluid (Roedder, 1971; Pollard *et al.*, 1991; Sharma *et al.*, 1994) in the Degana tungsten deposits. This alteration pattern is absent around BQVs, which is interpreted to be due to differences in composition, pH and volatile concentration in the hydrothermal fluid that formed the barren quartz veins.

The fluid inclusion petrography, inclusion types, abundance composition and densities suggested significant differences in homogenization temperature and fluid composition for the formation of MQVs and BQVs. Micro-thermometric analysis of the different fluid inclusions shows initial high T_h (335-487 °C) and high salinity (52-70 eq. wt.% NaCl) in the MQVs and lower temperature of homogenization (325-376 °C) and salinity (40-42 eq. wt.% NaCl) in BQVs.

The similar homogenization temperature of the multiphase inclusions and carbonic inclusions in MQVs with contrasting salinities can be explain by the un-mixing (boiling) of fluid as suggested for other Sn-W deposits elsewhere (Shepherd *et al.*, 1985; Lourenco *et al.*, 2001). In the Degana tungsten deposits the CO_2 bearing inclusions (Type-III) have variable CO_2:H_2O rations. These inclusions variably homogenized into either the liquid or vapour phase over nearly the same temperature range (270-350 °C). Touret (1977) and Bodnar *et al.* (1986) suggested that boiling of an H_2O-rich fluid could result in separation of a vapour phase rich in CO_2. This explains the entrapment of

Type-III inclusions with a wide range of CO_2: H_2O ratios.

The brecciation of Degana granite decreases the confining pressure, which caused ex-solution of a CO_2-bearing vapour from the initial Type-I fluid. Due to CO_2 un-mixing, the pH of the fluid increased and the deposition of tungsten could have effectively taken place (Somani and Srivastava, 2000). However, a small change in salinity with decreasing temperature is observed in the barren quartz veins. The lower salinity (9-42 wt.% NaCl) and densities (0.22-0.42) of the barren quartz veins suggested the possibility of mixing of comparatively cooler meteoric water in the primary fluid at the later stages of fluid evolution, and as a result the salinities of the fluid inclusions decreases from MQVs to BQVs.

As the fluids at Degana are dominantly NaCl solutions with CO_2, the transport of W in the form of chloride complexes are is favoured for the Degana tungsten deposits. However, the role of CO_2 in W transportation cannot be completely ruled out, as suggested by the common presence of carbonic inclusions in major tungsten bearing quartz veins.

Somani and Srivastava (2000) observed a substantial increase in the chloride contents of the fluid inclusions from the granitic quartz to vein quartz. The present study also supports the existence of high salinity and high temperature fluids during the first stage of the fluid ex-solution from magma. This supports the view that W at Degana might have been transported in the form of chloride complexes such as (WO_3) CL- and /or WO_2Cl_2, perhaps in addition to carbonic complexes.

Srivastava and Sinha (1995) have synthesized the available information from the known tungsten mineralization of western India to evaluate the relevance of wall rock alteration mineralogy to tungsten exploration and suggested that intense greisenization of the host rock with quartz-zinnwaldite-fluorite mineral assemblages may be used for targeting vein-type tungsten mineralization, such as that found around mineralized quartz veins at the Degana tungsten deposits.

Based on field characteristics, differences in the mineral assemblages of these veins, post-magmatic changes in the host rock surrounding the quartz veins and fluid inclusion data, barren quartz veins and mineralized quartz veins may be distinguished. Further, the prominent difference in characteristics of the fluid inclusion in the two different generations of quartz veins has the potential to be used as an exploration guide for tungsten –bearing quartz veins in the area.

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