

# Fluid evolution in quartz vein-hosted tungsten mineralization in Chhendapathar, Bankura District, West Bengal: Evidence from fluid inclusion study

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Tungsten mineralization in Chhendapathar area is hosted by quartz veins that traverse mostly the metasediments in and around Jikhu Nala. Fluid inclusion microthermometric experiments reveal the presence of four distinct types of inclusions. These are: aqueous biphasic, monophasic carbonic, aqueous-carbonic and halite-bearing polyphasic inclusions. Salinity-temperature variation points towards the presence of two fluids of contrasting salinities and both independently followed simple cooling paths without any indication of fluid mixing. The P-T of mineralization was calculated from the intersection of coexisting and coeval aqueous biphasic, carbonic and halite-bearing inclusions. The deduced values range from 1.63 kb/361°C to 2.30 kb/385°C. However, the initial temperature must have been much higher as indicated from the high dissolution temperature (> 450°C) of halite. Transportation of tungsten in the high saline fluid was facilitated by cation-tungstate ion pairing, i.e., with the help of Na<sub>2</sub>WO<sub>4</sub> and/or NaHWO<sub>4</sub> complexes. A rapid fall in solubility in such fluid with falling temperature (in the range of 300–400°C), and by occasional fluid-rock interaction triggered precipitation of wolframite.

## 1. Introduction

Wolframite occurrence in the Chhendapathar (22°45'N, 86°45'E) area includes one of the few known tungsten deposits in India. For about half a century, it has attracted the attention of the mineral industry. The first incomplete attempt for prospecting the deposit was made by M/S Tata Sons & Co. Ltd. around 1920–22. Most of the previous work by private organizations were intended to facilitate ore prospecting, mining, ore reserve estimation etc. A comprehensive model to account for the evolution of ore fluid and precipitation of ore minerals is still lacking. Controversy exists regarding the source of ore elements. Chakravorty (1958, 1988) believes that the granites, exposed mostly along Jikhu Nala, is the source of ore elements, i.e., the granite is 'causal' and not casual to mineralization. He inferred mineralogical zoning around Chhendapathar granite, the high temperature tungsten and minor copper minerals occur close to the

granite, whereas the minor lead, zinc and silver minerals occur further away. On the contrary, Mondal (1993) advocates that the metasediments and metavolcanics supplied necessary ore elements whereas the granites only helped to mobilize the ore elements from metasediments (and metavolcanics) to structurally controlled suitable depositional locales. Chakravorty (1988) attempted dating of the mineralized veins on the basis of Rb-Sr isotope studies in the altered wall rock surrounding the veins. He obtained an age of 1048 ± 40 Ma for the veins and comparable ages of 1064 ± 25 Ma for the pegmatoids and leuco-granites respectively.

Most of the previous workers agreed upon the obvious 'hydrothermal' nature of the mineralization. However, fluid inclusion studies have not been taken up seriously. A survey of available literature (cf. Ghosh and Ghosh 1979; Sahu *et al* 1979) reveals limited effort in this direction with scanty data leading to inaccurate interpretation pertaining to the condition

**Keywords.** Tungsten mineralization; Chhendapathar; inclusion microthermometry; P-T estimation; fluid evolution.

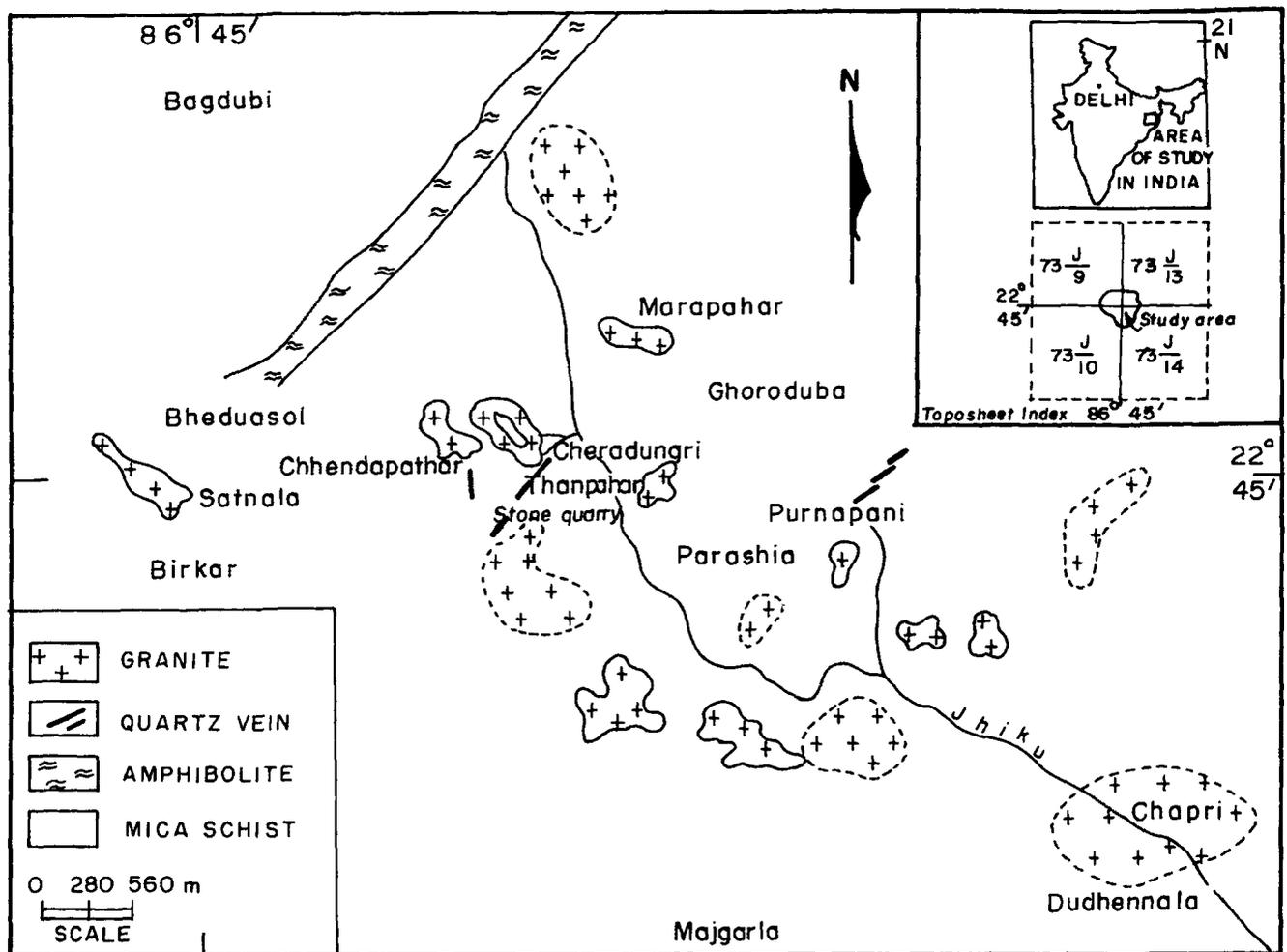


Figure 1. Lithological map in and around Chhendapathar, Bankura District, West Bengal.

of tungsten mineralization. In this communication, we have addressed the fluid evolution part as evidenced from fluid inclusion studies.

## 2. Geological background

The lithology of the area in and around Chhendapathar is dominated mainly by garnetiferous mica schists and leuco-granites which are regionally correlated with Iron Ore Stage of the Iron Ore Series of Dunn and Dey (1942). This is the eastern most extension of the Eastern Indian Shield and bounded by two mega lineaments, one running NNW-SSE and separating the Tertiary laterite from Precambrian rocks, and the other is the Dalma Thrust that runs nearly EW. The different rock units encountered in the field are metasediments, metabasics, granites and quartz veins (figure 1). **Metasediments** are represented by mica schists. Although these rocks, constitute the most dominant litho-unit, they are mostly altered and covered by thick aluvium and soil. The typical minerals in their order of abundance are biotite, muscovite, plagioclase, K-felspar and quartz. Garnet

and sphene are very rare and occur as accessory mineral phases. Mondal (1993) reported the presence of staurolite in mica schists. Quartz in these rocks commonly shows undulose extinction and occasionally are drawn out parallel to the schistosity. Garnet records retrograde alteration to biotite. They also sometimes contain inclusions of quartz and muscovite. Coarse-grained, melanocratic **amphibolite** occur in the field as long linear outcrop (figure 1). The rock is sometimes well lineated and the lineation is defined by the preferred orientation of hornblende. The different minerals present in amphibolite include hornblende, quartz, sphene, garnet, plagioclase and some opaques. Very coarse-grained **granites**, sometimes pegmatoid in nature, occur as small sporadic outcrop mostly along and close to the Jikhu Nala. The granites are composed of quartz, alkali felspar, plagioclase, biotite and hornblende. Muscovite and garnet are very rare. The accessory minerals are sphene, epidote and zircon. Commonly these rocks are gneissose and the gneissosity is defined by alternate layers of biotite and quartzo-felspathic minerals. **Quartz veins** are the host rock for ore minerals. They commonly occur within metasediments. There are three main vein

systems in and around Chhendapathar. The Thanpahar vein crops out as a single bodied lens with a length of 95 m and width 28 m. The vein dips 15°W and strikes 13° from north. Wolframite is the most dominant ore mineral in this vein. However, scheelite also occurs locally. Ore minerals occur as spotty and sporadic pockets, lenses, stringers and small disseminated grains. Besides the presence of these minerals, there are zones of oxidation in Thanpahar vein, characterized by the presence of cuprite, tenorite, malachite, goethite. The Cheradungri vein is a composite vein with an outcrop length of 80 m and width of 30 m, that trends NNW-SSE and dips due west. The Cheradungri vein either parallels or transgresses the schistosity. The contact is commonly sharp, with local development of zone of sericitization at the margin with the country rocks. The Purnapani group of veins comprises a number of en echelon mineralized veins trending WNW-ESE in the western part of the vein and nearly EW in the southern extension. The total strike length of the veins in the Purnapani group is 500 m and the average width is approximately 2 mt. The nature of mineralization and the mineral assemblage in the oxidized zone is similar to that in Thanpahar and Cheradungri.

Quartz in the mineralized quartz veins are transparent to bluish in color, whereas those in non-mineralized quartz veins are mostly transparent. No distinct mineral zoning is observed in these veins. Contrary to the observations of Chakravorty (1958), the present study does not reveal any preferred spatial concentration of ore minerals.

### 3. Fluid inclusion studies

Fluid inclusion studies were conducted to characterize the ore fluid in terms of its gross chemistry, pressure and temperature and also to trace a plausible evolutionary trend, that was responsible for W mineralization. The study was restricted to undeformed, well crystallized quartz from the mineralized quartz veins. Twelve doubly polished thin wafers with thickness between 30  $\mu$  to 50  $\mu$  were prepared. All the wafers were carefully studied under transmitted light microscope for a detailed petrographic classification of the inclusions. Most of the wafers were not suitable for microthermometric studies either because of their low visibility or because of the absence of workable inclusions and finally four were selected. Each wafer was broken into small chips of 3 to 5 mm in dimension. From each field of view, on an average 5–6 numbers of primary or pseudo-secondary inclusions were selected following the criteria of Roedder (1984) and Shepard *et al* (1985), for microthermometry. Only those inclusions, which gave reproducible values during the freezing-heating experiment, were used for data interpretation. The inclusion size varies from little less

than 10  $\mu$  to about 25  $\mu$ . Inclusions are circular, rectangular or negative crystal shaped. Petrographic study of the inclusions revealed the presence of four distinct types of inclusions on the basis of their contents at ambient laboratory temperature:

- 1) Type I inclusions: aqueous bi-phase inclusions containing a bubble of water vapor and aqueous liquid phase.
- 2) Type II inclusions: aqueous-carbonic inclusions containing a CO<sub>2</sub> rich liquid rimmed by H<sub>2</sub>O liquid. Attempts were made to study these inclusions but the inherent complexity and non-reproducibility of data inhibits further experiment with these inclusions. Moreover, the total homogenization temperature of H<sub>2</sub>O-CO<sub>2</sub> inclusions was difficult to obtain as they burst during heating runs because of building up of high internal pressure (cf., Shepard *et al* 1985).
- 3) Type III: monophasic carbonic inclusions containing a CO<sub>2</sub> liquid at ambient laboratory temperature. On slow cooling, a CO<sub>2</sub> vapor phase develops.
- 4) Type IV inclusions: containing water vapor, aqueous liquid and halite crystal. Halite was identified from its well developed cubic shape and colorless to greenish color.

### 4. Microthermometry

For the purpose of microthermometric experiments, a Fluid Inclusion adapted USGS gas flow heating-freezing system, also known as Reynolds stage (cf. Shepard *et al* 1985) has been used. The unit is placed on an X-Y stage of Leitz Laborlux D petrological microscope with a 100 watt illuminator. The chromel const. thermocouple is placed directly on the sample to tightly hold the sample in its place and also to reduce the uncertainty in temperature measurement. The unit operates in the temperature range of –195°C to 700°C. Freezing is achieved by flowing pre-chilled N<sub>2</sub> gas over the sample and pre-heated compressed air is used for heating. The unit is periodically calibrated by synthetic pure H<sub>2</sub>O (triple point 0°C) and pure CO<sub>2</sub> (triple point –56.6°C) inclusions supplied by the manufacturer. Additional calibrations were made by melting spec pure grade metal such as In (156.6°C) and Pb (327.5°C). In general, the accuracy of the system is 0.1°C during freezing and 1°C during heating.

The type I and type III inclusions were first frozen followed by gentle warming and then heating. The type IV inclusions were not frozen and they were only heated to record the liquid-vapor homogenization temperature ( $T_h$ ) and also the halite dissolution temperature ( $T_{s,NaCl}$ ). Aqueous bi-phase inclusions were completely frozen within –50°C and –125°C. However, some of the extremely metastable inclusions

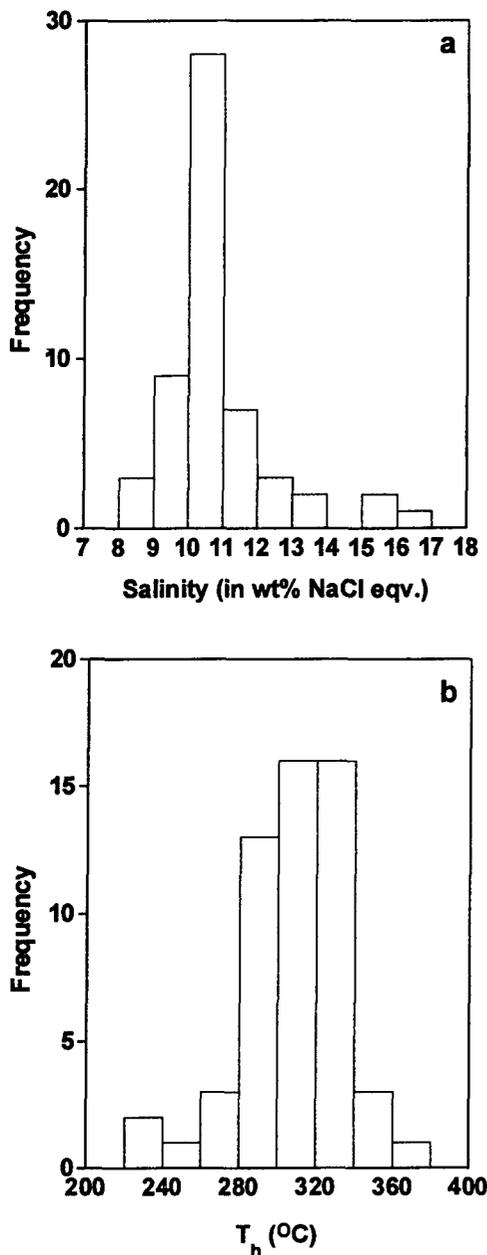


Figure 2. Frequency plots for aqueous bi-phase inclusions; (a) salinity (in wt% equivalent); (b) temperature of homogenization.

did not freeze even below  $-150^{\circ}\text{C}$ . The observed temperature of final ice melting ( $T_m$ ) values vary from  $-5.2^{\circ}\text{C}$  to  $-12.1^{\circ}\text{C}$  corresponding to salinity values of 8.10 and 16.05 wt% NaCl equivalent. The frequency of salinity is plotted in figure 2(a). There is a distinct peak in the salinity range of 9–12 wt% NaCl equivalent. All these aqueous bi-phase inclusions homogenized into the liquid phase. There was not a single instance of vapor phase homogenization. Although the  $T_h$  value ranges between  $220^{\circ}\text{C}$  and  $380^{\circ}\text{C}$  (figure 2b), a majority of the data lie within 280 and  $340^{\circ}\text{C}$ , thus defining a clear peak in this temperature range. Density of the type I inclusion varies from 0.749 to  $0.952\text{ gm/cm}^3$ . Most of the type III monophasic

carbonic inclusions are completely frozen at around  $-100$  to  $-105^{\circ}\text{C}$ . The final  $\text{CO}_2$  ice melting temperature ( $T_{m,\text{CO}_2}$ ) was recorded to lie between  $-56.6$  and  $-59^{\circ}\text{C}$ , with a majority of the values close to  $-56.6^{\circ}\text{C}$  (figure 3a). However, there are some inclusions where the last ice melting temperature is less than  $-56.6^{\circ}\text{C}$ . The lowering of  $T_{m,\text{CO}_2}$  beyond that of pure  $\text{CO}_2$  ice ( $-56.6^{\circ}\text{C}$ ) suggests the presence of some other incompressible gases, like  $\text{CH}_4$ , in the inclusions. The mole% of  $\text{CH}_4$  in  $\text{CO}_2$  was calculated using the data of Heyen *et al* (1982) and it varies between 1.07 and 11.69 mole%. The type III inclusions are therefore, effectively  $\text{CO}_2$ - $\text{CH}_4$  inclusions. All these inclusions homogenize by the disappearance of the vapor into the liquid ( $L_{\text{CO}_2} + V_{\text{CO}_2} \rightarrow L_{\text{CO}_2}$ ) and the temperature of homogenization ( $T_{h,\text{CO}_2}$ ) vary between  $4^{\circ}\text{C}$  and  $22^{\circ}\text{C}$  with a distinct peak in the range of 4 to  $12^{\circ}\text{C}$  (figure 3b). The density of pure  $\text{CO}_2$  and  $\text{CO}_2$ - $\text{CH}_4$  inclusions were obtained by using the equations of state by Brown and Lamb (1989) and the SOUP (Holloway 1981) subroutines in the FLINCOR software (cf, Brown 1989) respectively. The density of the  $\text{CO}_2$  inclusions varies between 0.65 and  $0.903\text{ gm/cm}^3$ . Barring the exception of one inclusion, all the polyphase inclusions homogenized by the disappearance of halite crystal. In some of the inclusions, the halite crystals did not dissolve even after being heated up to a temperature of  $450^{\circ}\text{C}$ . The halite dissolution temperature ( $T_{s,\text{NaCl}}$ ) is observed to lie between 256 and  $411^{\circ}\text{C}$  (figure 4b) and the liquid vapor homogenization occurred between 213 and  $346^{\circ}\text{C}$ . These are very high saline inclusions with salinity ranging from 34 to 54 wt% NaCl equivalent. Although there is no distinct peak, the salinity of these inclusion fluid centers around 38 to 42 wt% NaCl equivalent (figure 4a) and the density varies between 1.084 and  $1.235\text{ gm/cm}^3$ .

## 5. Fluid evolution and discussion

Salinity vs. temperature of the final homogenization plot of the aqueous bi-phase and poly-phase inclusions (figure 5) show two independent paths of fluid evolution; each characterized by a simple cooling history. The high saline, high temperature fluid is presumably derived from granitic melt (cf. Roedder 1971; Burnham and Ohmoto 1980) which, in this case, is represented by the Chhendapathar granite. The relatively less saline fluid is of some non-magmatic origin; may be heated meteoric or metamorphic water. Polyphase and aqueous biphasic inclusions are not found to coexist within the same sample. Coexisting and cogenetic  $\text{CO}_2$ - $\text{CH}_4$  inclusions are found in association with the halite-bearing and also with aqueous biphasic inclusions. The coexistence of halite-bearing inclusions and  $\text{CO}_2$ - $\text{CH}_4$  inclusions is indicative of a parent  $\text{H}_2\text{O}$ - $\text{CO}_2$ - $\text{CH}_4$ -NaCl fluid. The independent fluid evolutionary paths for high

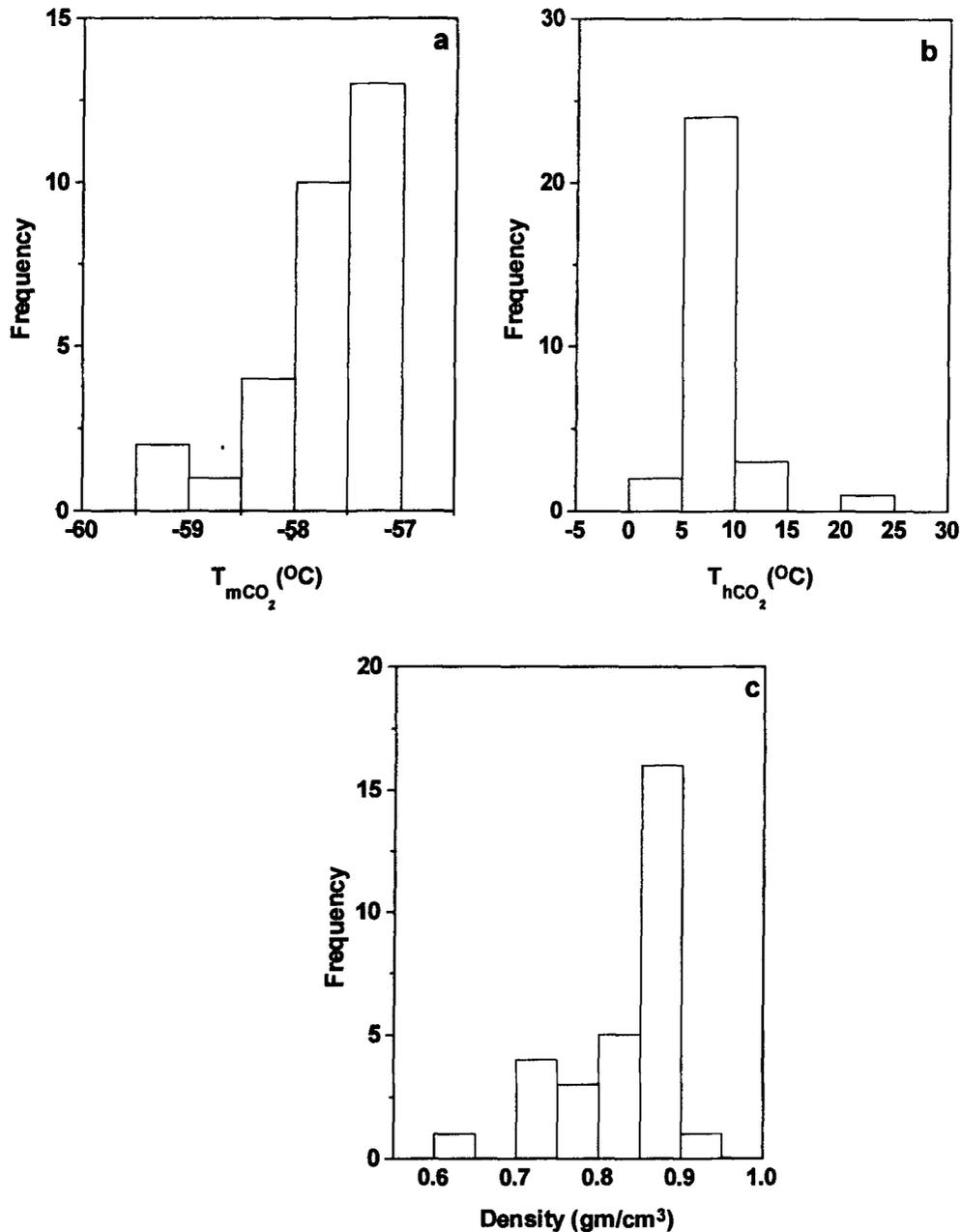


Figure 3. Frequency plot for CO<sub>2</sub>-CH<sub>4</sub> inclusions; (a) temperature of CO<sub>2</sub> ice melting ( $T_{mCO_2}$ ); (b) temperature of homogenization ( $T_{hCO_2}$ ) and (c) density (in gm/cm<sup>3</sup>).

saline and low saline inclusions, without any intermediate salinity, suggests that there was no extensive mixing between these two fluids. This is possible because of a large difference in salinity that inhibited any chemical equilibrium between these two fluids. By the time the low saline fluid has reached the ore depositional site, the major bulk precipitation of wolframite along with other ore minerals was possibly nearly complete.

High saline fluids are rather common in many tungsten deposits in other parts of the world (Wood and Vlassopoulos 1988). Ivanova (1988) and Ivanova *et al* (1987) recognized a number of Russian deposits, where salinity of tungsten ore forming fluid goes even up to 70 wt% NaCl equivalent. This is of prime impor-

tance as the solubility of W in saline solution is quite high to account for the transport and subsequent precipitation of wolframite and other associated ore minerals to give rise to economic ore deposits. Wood and Vlassopoulos (1988) are of the opinion that solubility of W increases with increasing salinity. They have argued, on the basis of their experimental studies, that chloride complexing is not a viable mechanism for the transport of tungsten in natural hydrothermal condition. However, they propose that chloride complexing may be extremely important to transport other elements such as Fe, Ca, Mn which are common constituents of W minerals. The experimental studies of these workers show that cation-tungstate ion pairing is the most effective mechanism especially

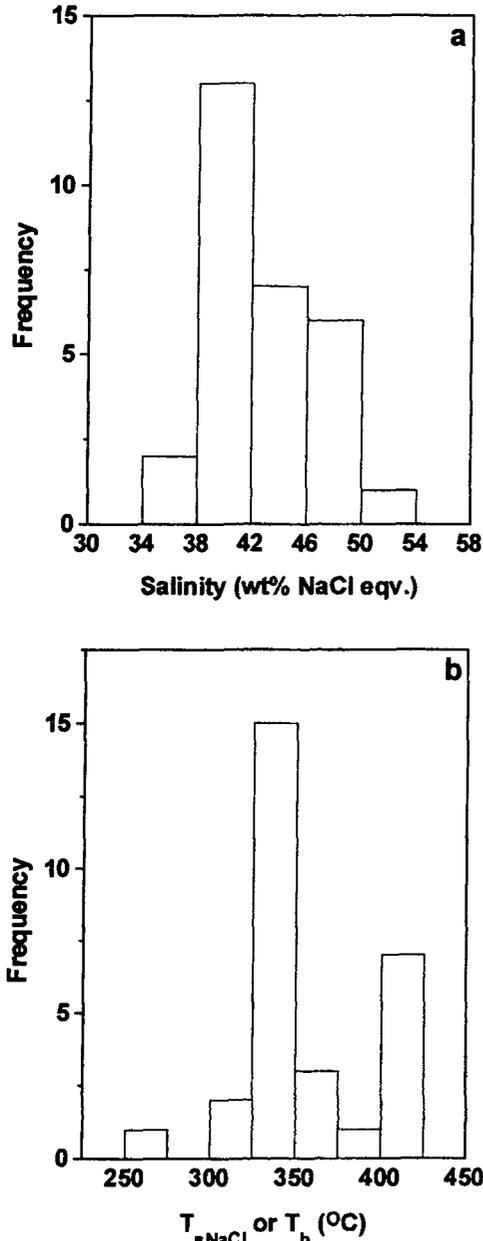


Figure 4. Frequency plot for halite bearing inclusions; (a) salinity (wt% NaCl equivalent) and (b) temperature of final homogenization ( $T_{sNaCl}$  or  $T_h$ ).

when the ore fluid is high saline. The common soluble complexes,  $NaHWO_4$  and  $Na_2WO_4$ , are formed by the following reactions



Reaction 1 will be important in pure water and reactions 2 and 3 will be more effective in saline fluids. The polyphase inclusions from Chhendapathar indeed contain halite (NaCl) as the most common daughter mineral. Therefore,  $Na_2WO_4/NaHWO_4$  are the most feasible complexes that have transported tungsten to the depositional locales. The common precipitation

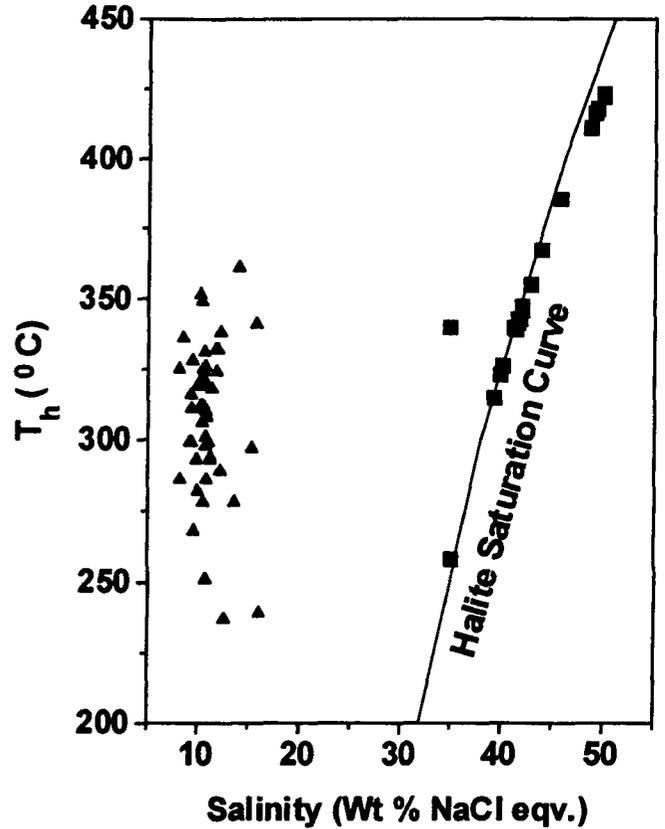
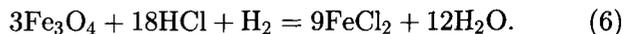


Figure 5. Temperature vs. salinity plot for aqueous bi-phase (solid triangles) and poly-phase (solid squares) inclusions. See text for discussion.

mechanism for ferberite is



Experimental work by Chou and Eugster (1977) and that of Boctor *et al* (1980) shows that in chloride-bearing hydrothermal solutions,  $Fe_2O_3$  and  $Fe_3O_4$  occur mostly in  $FeCl_2$  state. But, as has been pointed out by Eugster (1985), the co-transportation of  $FeCl_2$  and  $NaHWO_4/Na_2WO_4$  in hydrothermal fluids is unlikely. The possible sources, therefore, are the ferromagnesian minerals and magnetite in the country rocks. The ore fluid on its way to the depositional locale might have scavenged Fe from the country rocks. One of the possible reactions of Fe leaching from magnetite is



In contrast to reaction 4, reaction 5 does not evolve any acid. On the other hand, HCl generated from the reaction must be consumed to facilitate precipitation. Felspar, in the wall rock, is the most suitable candidate for acid consumption. In such cases, it is expected that the ore minerals will concentrate at the contact between the vein and the wall rock. In Chhendapathar, wolframite is disseminated in the quartz vein and there is no preferential concentration of ore minerals in the contact zone. Therefore, reaction 5

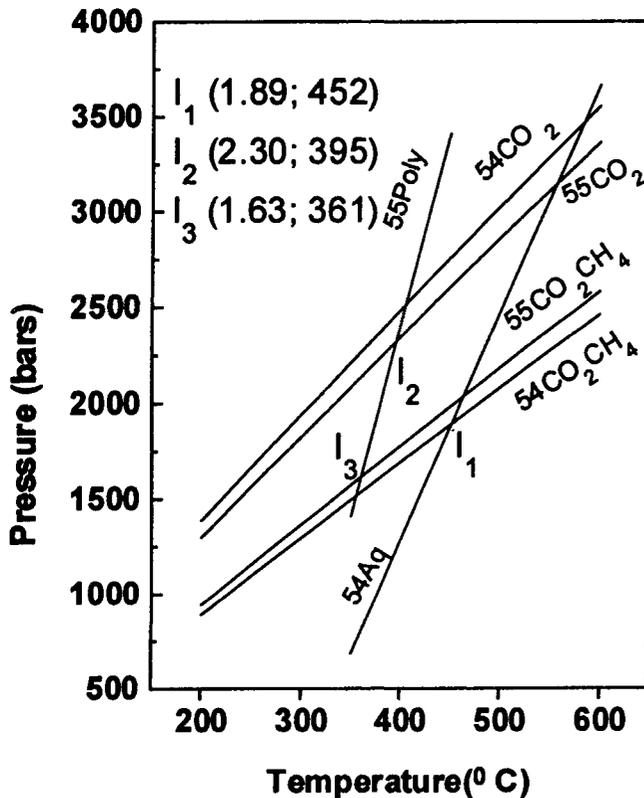


Figure 6. P-T calculation from isochore intersection method. Aq., poly., CO<sub>2</sub> and CO<sub>2</sub> CH<sub>4</sub> stands for aqueous bi-phase, polyphase, carbonic and CO<sub>2</sub>-CH<sub>4</sub> inclusions. See text for discussion.

seems to be more effective for the precipitation of wolframite. However, Chakravorty (1958) noted that mica, wolframite and rarely tourmaline are “commonly attached” to the walls of the veins with a central filling of massive quartz. Therefore, reaction 4 was possibly of local importance, where the acid released due to wolframite precipitation was consumed by the wall rock minerals, thereby facilitating concentration of ore minerals in the walls.

Theoretical calculations of Heinrich (1990) suggest that the amount of soluble tungsten in hydrothermal ore fluids, in equilibrium with a granitic rock or quartzo-feldspathic minerals, falls rapidly with decreasing temperature and this is more pronounced at a temperature around 300°C. Further this is true both in rock-buffered and fluid-buffered situations. Consequently, most of the tungsten deposits are formed within the temperature range of 300–400°C (Jaireth *et al* 1990). The pressure-temperature of ore precipitation, in Chhendapathar, has been calculated from the intersection of isochores of coexisting (and coeval) polyphase and carbonic inclusions following the method of Roedder and Bodnar (1980) as shown in figure 6. The practical guidelines for the choice of coeval inclusions for isochore intersection are discussed by Mishra and Panigrahi (1999). The equations of Brown and Lamb (1989) and that of Zhang and Franz (1987) were used to construct the isochores

of CO<sub>2</sub>-CH<sub>4</sub> and H<sub>2</sub>O-NaCl inclusions respectively. The isochores were plotted with the help of the FLINCOR program (Brown 1989). The calculated P-T, for sample No. 55, range from 2.30 kb/395°C (I<sub>2</sub>) to 1.63 kb/361°C (I<sub>1</sub>). For the less saline fluid (sample No. 54), the estimated P-T is 1.89 kb and 452°C (I<sub>3</sub>) (figure 6). Further, the high P-T intersection (at 3.46 kb/585°C) has not been taken into account, because isochores cannot be constructed in the H<sub>2</sub>O-NaCl system beyond 3.00 kb by the available P-V-T-X systematics in the above system (*c.f.*, Brown 1989). On the other hand, the deduced values (I<sub>1</sub> and I<sub>3</sub>) are well within the critical P-T range of Heinrich (1990) and Jaireth *et al* (1990). However, there is evidence in favor of higher initial temperature (and pressure) of the ore fluid. Halite crystals in some of the polyphase inclusions dissolved at a temperature of 420°C and there are some, which did not dissolve even after being heated up to 450°C. The absence of coexisting and cogenetic CO<sub>2</sub>/CO<sub>2</sub>-CH<sub>4</sub> inclusions and polyphase inclusions with higher  $T_{s,NaCl} > 420^\circ\text{C}$  values exclude the possibility of calculating the P-T of entrapment by isochore intersection method. Nevertheless, the P-T estimation has been attempted by the intersection of inclusion isochore and the corresponding halite liquidus drawn along  $T_{s,NaCl}$ , where the halite dissolution temperature is higher than the liquid-vapor homogenization (*i.e.*  $T_{s,NaCl} (422^\circ\text{C}) > T_h (290^\circ\text{C})$ ), according to the method outlined by Shepard *et al* (1985) and Bodnar (1994). The halite liquidus at the  $T_{s,NaCl}$  have been assumed to be pressure independent. This is a reasonable approximation since at low temperature dP/dT slope of the halite liquidus is negligible (*cf.* Gunter *et al* 1983). The pressure obtained by this method is 3.12 kb (figure 7). It must be noted that the P-T (3.12 kb, 422°C) estimated from this method gives the minimum conditions of entrapment for the corresponding inclusion.

Distribution of the majority of ore veins within schistose rocks around Chhendapathar granite (Chakravorty 1958) is strong evidence in favor of the genetic linkage of the granite and the productive quartz veins. For proper understanding of the evolution of the ore-bearing veins vis-à-vis granitic activity, the geochemical characterization of the granites is necessary. However, from the presence of muscovite, biotite, garnet and its two-felspar nature, possibly indicate that the granite belongs to the ‘peraluminous’ clan. H<sub>2</sub>O-saturated granitic melt exsolved a high temperature, high saline and CO<sub>2</sub>-CH<sub>4</sub> bearing fluid after its emplacement at a shallower level and sufficient crystallization. The ubiquitous presence of CO<sub>2</sub>-CH<sub>4</sub> bearing inclusions suggests a reduced nature of the ore fluid. Burnham and Ohmoto (1980) and Jackson *et al* (1982) have explained the Sn-W potential of such magmatic aqueous fluid. The reduced nature of such fluid has been stressed by Eugster (1985) and Heinrich (1990). After separation from the granitic melt, the

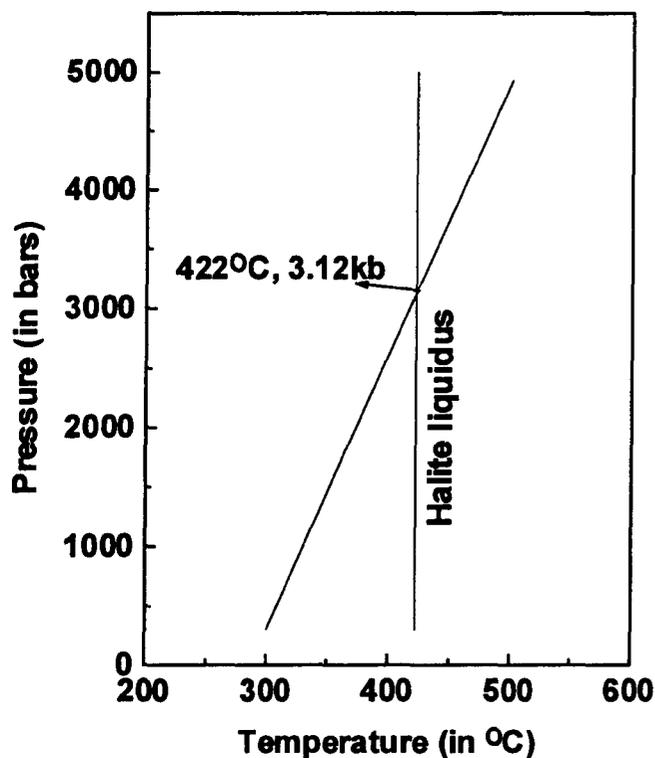


Figure 7. P-T diagram showing intersection between isochore of a polyphase, halite-bearing inclusion and the corresponding halite liquidus through the temperature of halite dissolution. See text for discussion.

ore fluid was focused to favorable locales. During its journey to the depositional site it has followed a simple cooling path and the P-T values fell from more than 3.12 kb/422°C to 1.63 kb/361°C. A genetically different and less saline fluid that has evolved independently following a simple cooling path, was also focused to the depositional site. This fluid (metamorphic or heated meteoric water), on its way to the depositional locale, might have scavenged pertinent ore elements from the metasediments and added to mineralization at certain places. Therefore, the two fluids although show seemingly independent evolutionary paths, were both responsible for the tungsten mineralization to variable extents at Chhendapathar.

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