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GEOLOGY, MINERALOGY AND TEXTURAL INVESTIGATIONS ON TUNGSTEN DEPOSITS OF CHITRAL, KHYBER PAKHTUNKHWA (KPK), PAKISTAN

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Abstract

Investigations were carried out on tungsten ore deposits of district Chitral, Khyber Pakhtunkhwa (KPK) Province, Pakistan, in order to evaluate the ore deposits and subsequently to select a suitable process for beneficiation. Two important ore bodies of tungsten have been explored at Minike Gole and Besti Gole areas of district Chitral. The ore deposits were characterized by different techniques like microscopy, petrography, X-ray diffraction and chemical evaluation. The valuable mineral identified in these ores was found to be scheelite. The principal mineral constituents present in these ores were found to be quartz, clinozoisite, actinolite, magnetite, calcite, biotite and muscovite. The textural characteristics such as grain (crystal) size, shapes and mutual arrangement of component minerals in the ore bodies were also investigated. The ores were found to be fine-grained in nature. The degree of liberation of valuable scheelite mineral from associated valueless gangue minerals was studied by grain mounts technique. Mineralogical, textural and liberation studies revealed that both ores are amenable to beneficiation by froth flotation technique.

1. INTRODUCTION

Tungsten is a steel-gray metal in appearance. It has many unique properties and diversified industrial applications (Lassner and Schubert, 1999). Its abundance in the earth's crust is estimated to be 0.00015% (Foster, 1988). There are more than twenty well known minerals of tungsten but commercially important minerals are scheelite $CaWO_4$ and wolframite $(Fe,Mn)WO_4$ series (Blackburn, 1988). Tungsten mineralization takes place at high temperatures and pressures. Wolframite deposits are typically igneous in origin and wolframite occurs in quartz veins or segregated in pegmatites. Scheelite is usually found in skarn deposits formed through contact metamorphism by the replacement of limestone bed, in high-temperature hydrothermal veins and greisens; less commonly in granitic pegmatites (Zulfiqar and Hariri, 2006; Chowdhury, 2007; Tornos et al., 2008).

For extraction of metal from an ore deposit it is important to select the optimum economic process from the many available options. The selection of

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appropriate mineral processing technique largely depends on the nature of mineralization, texture of the ore body and size of liberation of the valuable minerals (Weiss, 1985). Mineralogical assemblage gives knowledge about the nature of valuable and gangue minerals and their proportion in the ore whereas the texture refers to the crystal habits, aggregation, dissemination, mode of occurrence and form of association within the ore.

Due to high specific gravity of tungsten minerals, the ores are generally beneficiated by gravity concentration methods. However, wolframite ores may be beneficiated by high intensity magnetic separation whereas scheelite ores can be processed by froth flotation depending on their texture. For complex ores, a combination of mineral processing techniques is applied to optimize metal recovery. Since different deposits vary greatly in their characteristics, therefore different flow sheets are adopted for their beneficiation (Wills, 1992).

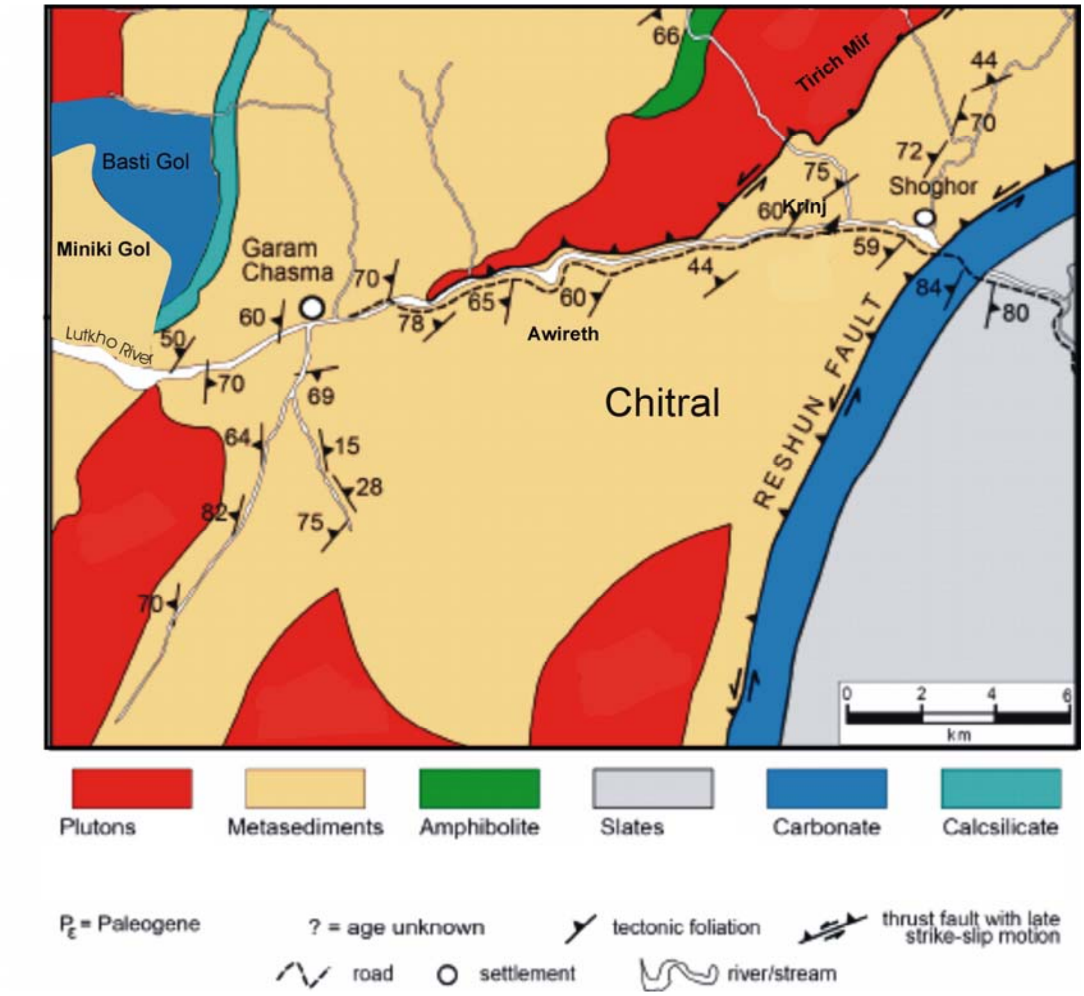

Fig. 1. Location map of Miniki Gol and Besti Gole tungsten ores, district Chitral, KPK Province, Pakistan

Tungsten deposits have been reported to occur in the Hindu Kush Range of northern Pakistan (Hildebrand et al., 2000). Zeschke (1961) first discovered tungsten as scheelite in this area during regional drainages survey using panning heavy mineral concentrate. Leak et al., (1989) of British Geological Survey in collaboration with team of Sarhad Development Authority, Peshawar, conducted a regional pan concentrate and stream sediments studies of Garam Chashma area, about 60 km to the north-west of Chitral City, in Khyber Pakhtunkhwa Province and discovered scheelite mineralization on the ridge to the north of Miniki Gole. The mineralization stretches from south of Lutkoh river to north-eastwards through Miniki Gole valley and extends as far as Besti Gole, at strike length of about 20 km (Figure 1). The deposit consists of a number of ore bodies. Indicated reserves of the ore have been calculated to be around 1.8 million tons however; the mineable reserves are estimated to be 0.5-0.75 million tons (Anwar, 2005). The present study is mainly focused on identification of minerals present in the ore, their relative amount, textural relationships between minerals within the ore and degree of liberation of valuable mineral from associated gangue minerals. These studies are particularly useful prior to the actual selection of the process and commercial plant designing.

2. GEOLOGICAL SETTING

Prospecting studies of samples collected from trenches and exposures show that scheelite mineralization mainly occurs in a calc-silicate quartzite rocks at Miniki Gol and adjoining areas. Two scheelite bearing calc-silicate quartzite units have been identified here. The most prominent horizon is fine-grained calc-silicate quartzite with maximum thickness of 4.5 m. The second horizon varies between 0.4 m to 1.0 m and is composed of medium-grained quartzite. Scheelites hosting quartzitic horizons are found within quartz mica schists. Quartzites are well developed in the upper reaches where as quartz mica schist is prominent at lower horizons. In addition to calc-silicate, some mineralization has also been found to occur in carbonate member of rock in Besti Gol samples. Sang Last is the only scheelite prospective area near by Dam-Dam Mountains in Besti Gol. The major ore body at Besti Gol is hosted by the granular limestone. According to Leake et al., (1961) scheelite mineralization occurs predominantly in clinozoisite-bearing calc-silicate quartzite beds within a sequence of mica schist and subordinate graphitic phyllite, mica quartzite, tourmalinite and feldspathic gneiss as strata bound deposits. But in our studies it has been found that in addition to quartzite, some scheelite is also hosted in limestone at Besti Gol area.

3. MATERIALS AND METHODS

Representative samples of the tungsten ore were collected from various localities of Miniki Gol and its adjoining areas such as Shish Gol, Jugh Kushu Gol, Aman Kushu Gol, and Baranazi Gol. Prospecting and evaluation work of scheelite mineralization were carried out using shortwave ultraviolet lamps during night time. The samples weighing about 20 kg each were obtained by excavation of trenches through drilling and blasting. The trenches were excavated in promising zones and the specimens were collected. The samples were brought to Mineral Processing Laboratory, PCSIR, Lahore, Pakistan, for chemical and mineralogical investigation. Portions of these samples were also mixed to prepare a composite representative sample. The ore samples were crushed using laboratory scale jaw crusher and roll crusher. Coning-quartering and riffling techniques of sampling were applied to prepare head sample for chemical analysis and for X-ray Diffraction studies. These were ground to minus 200 mesh size with the help of rod mill (Denver, USA).

Chemical analysis of the prepared sample was carried out in accordance with ASTM methods. Tungsten was determined colorimetry with stannous chloride using Spectrophotometer (Model: Spectronic-20 Bausch & Lomb, USA) in Analytical Laboratory of Mineral Processing Research Centre (MPRC). Silica was estimated by gravimetric method (Jeffery, et al.1997), Iron was found by oxidation reduction titration with standard solution of potassium dichromate (ASTM E 246-01). Aluminum was estimated by complexometric titration using standard solution of EDTA. Calcium and magnesium were determined by atomic absorption spectrophotometer (Model: Z-8000, Hitachi, Japan) (ASTM E-508). Alkali metals such as sodium and potassium were measured by flame photometer (PFP7, Jenway Limited, England) (ASTM E 395-02). Percentage of tungsten oxide in various samples of Miniki Gole and Besti Gole areas is reported in Table 1 and 2 respectively whereas complete chemical analysis of the representative samples is reported in Table 3.

The pulverized ore sample was run on X-ray Diffractometer (Model: D-5000, Siemens, Germany) in Mineralogy Laboratory, Institute of Geology, University of Punjab, Lahore. X-ray diffraction was done with a 0.02o step size per second. The scan angle was ranged from 0-80o and the spectrum obtained was matched with standard data. X-ray diffractrograms of both ores are shown in Figure 2 and 3 respectively. The mineral phases identified are given in Table 4 and percentage of minerals identified is shown graphically in Figure 4.

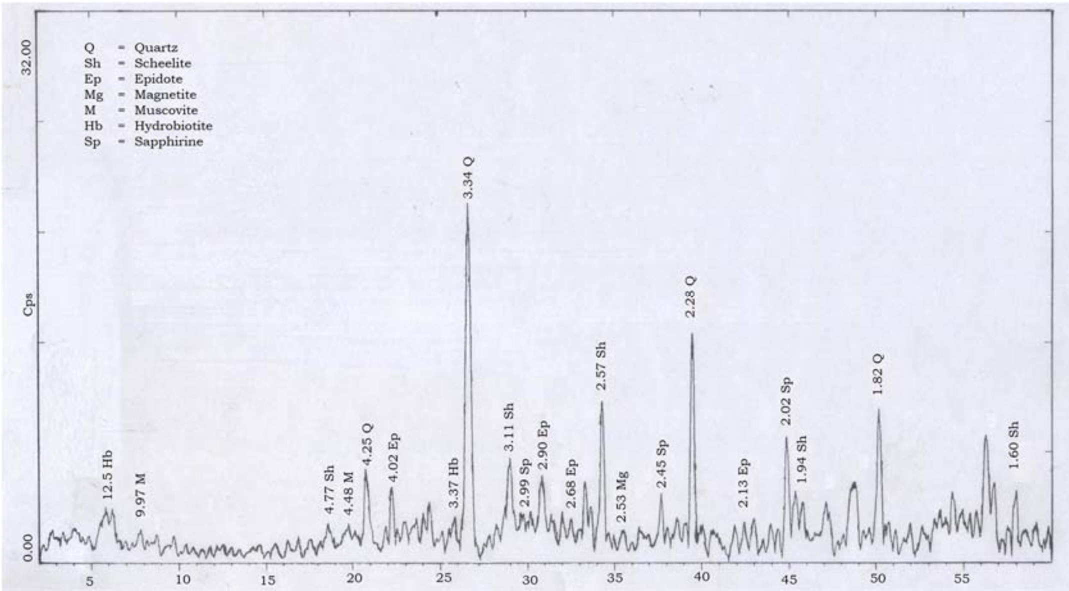

Fig. 2. X- Ray Diffractogram (XRD) of Miniki Gole tungsten ore, district Chitral, KPK Province, Pakistan

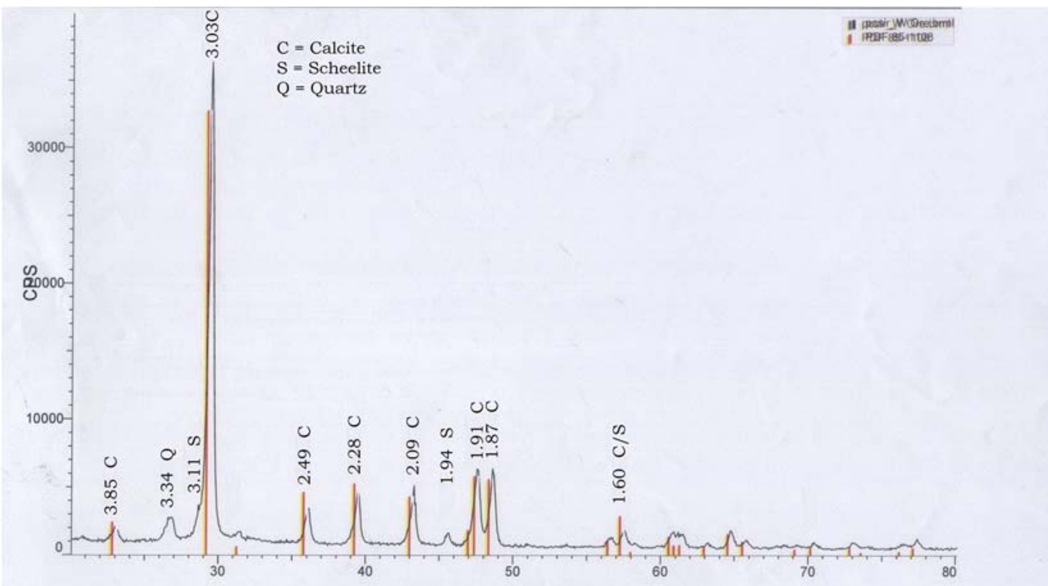

Fig. 3. X- Ray Diffractogram (XRD) of Besti Gole tungsten ore, district Chitral, KPK Province, Pakistan

Table 1. Percentage of WO_3 in samples collected from different blocks of Miniki Gole tungsten ore deposit, district Chitral, KPK, Pakistan

Block 1	B1-2	B1-5	B1-8	B1-11	B1-13	B1-16	B1-18	B1-19
% WO_3	0.179	0.135	0.585	0.165	0.157	0.225	0.280	0.341
Block 2	B1-54	B1-56	B1-58	B1-60	B1-86	B1-87		
% WO_3	0.197	0.516	0.350	0.198	0.596	0.250		
Block 3	B3-21	B3-24	B3-26	B3-29	B3-51	B3-53		
% WO_3	0.157	0.113	0.067	0.337	0.769	0.222		
Block 4	B4-32	B4-34	B4-37					
% WO_3	0.200	0.124	0.405					
Block 5	B5-92	B5-93	B5-94	B5-95	B5-96	B5-97	B5-98	B5-99
% WO_3	0.279	0.081	0.325	0.256	0.573	0.721	0.488	0.595
Block 5	B5-100	B5-101	B5-102	B5-103	B5-123	B5-126	B5-127	B5-128
% WO_3	0.297	0.314	0.435	0.139	0.550	0.279	0.581	0.418
Block 5	B5-129	B5-130	B5-132	B5-133	B5-134	B5-137	B5-138	B5-139
% WO_3	0.600	0.395	0.790	0.395	0.442	0.345	0.512	0.418
Block 6	B6-39	B6-42	B6-43					
% WO_3	0.168	0.517	0.149					
Block 7	B7-45	B7-49						
% WO_3	0.449	0.125						
Block 8	B8-47	B8-48						
% WO_3	0.091	0.114						
Block10	B10-89	B10-90	B10-91	B10-115	B10-140	B10-141	B10-142	
% WO_3	0.300	0.242	0.106	0.160	0.465	0.350	0.280	
Block11	B11-112							
% WO_3	0.419							
Block12	B12-116	B12-117	B12-121	B12-122	B12-124	B12-125		
% WO_3	0.495	0.117	0.060	0.218	0.215	0.135		

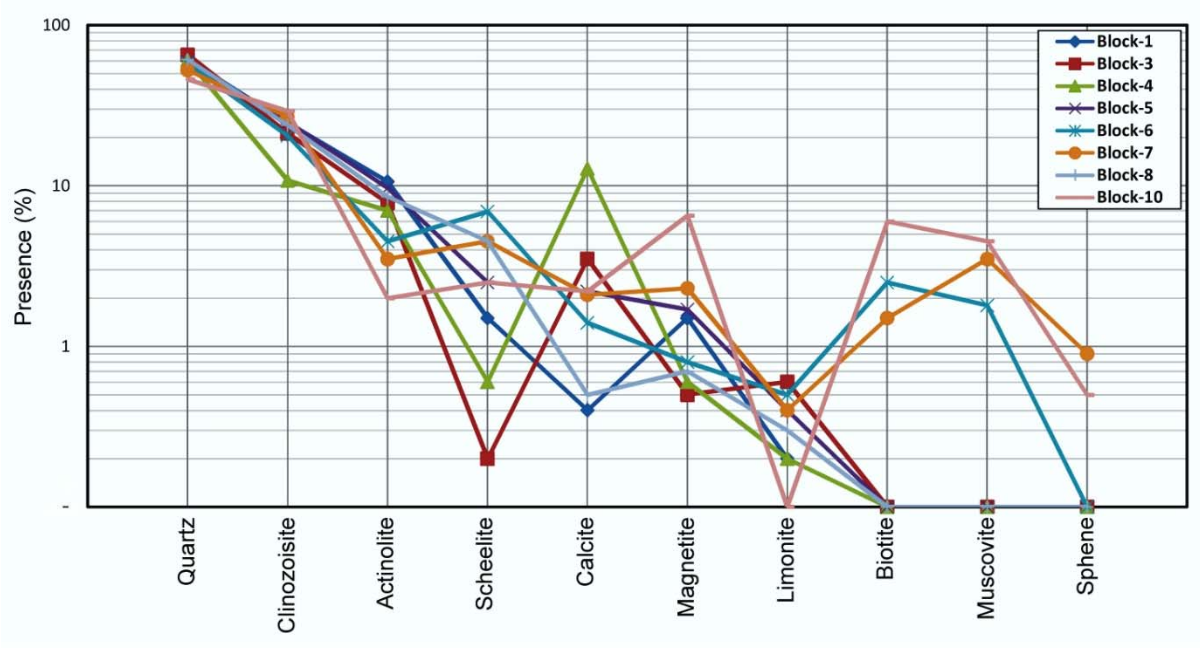

Fig. 4. Percentage of different minerals in various blocks of Miniki Gole tungsten ore deposit district Chitral, KPK Province Pakistan

Table 2. Percentage of WO_3 in samples collected from Besti Gole tungsten ore deposit, district Chitral, KPK Province, Pakistan

Besti Gole	BG-1	BG-2	BG-3	BG-4	BG-5	BG-6	BG-8	BG-9	BG-10	BG-11
% WO_3	0.480	0.460	0.158	0.135	0.159	0.182	0.204	0.114	0.113	0.078

Table 3. Chemical composition of Miniki Gole and Besti Gole tungsten ores district Chitral, KPK Province, Pakistan

Constituents	LOI	WO_3	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O
Miniki Gole	3.14	0.34	65.53	11.67	5.65	10.43	0.54	0.96	0.38
Besti Gole	37.04	0.32	6.60	0.80	1.02	52.11	2.29		

Table 4. Mineralogical composition of Miniki Gole (composite sample) and Besti Gole tungsten ores, district Chitral, KPK Province, Pakistan

Mineral Name	**Formula**	Miniki Gole (%)	Besti Gole (%)	**Specific**
Scheelite	$(CaWO_4)$	2.4	2.0	6.0
Calcite	$(CaCO_3)$	1.5	90.5	2.71
Quartz	(SiO_2)	60.5	6.5	2.65
Epidote (Clinozoisite)	$Ca_2Al_3(SiO_4)(Si_2O_7)(O,OH)$	24.3	--	3.3
Amphibole	$Ca_2Mg_5(Si_8O_{22})(OH)$	5.2	--	3.2
Magnetite	(Fe_3O_4)	1.8	--	5.2
Limonite	(FeO.(OH)	0.5	--	4.0
Biotite	$K(Mg,Fe)_3(AlSi_3O_{10})(OH,F)_2$	2.2	--	4.3
Muscovite	$K,Al_2(AlSi_3O_{10})(OH,F)$	1.2	--	2.8
Sphene	$(CaTiSiO_5)$	0.4	--	3.53

Petrography of the selected samples was carried out with polarizing microscope (Meiji, Japan) in Petrography laboratory, Institute of Geology, University of Punjab, Lahore. The photomicrographs of the representative samples are presented in Figures 5-14.

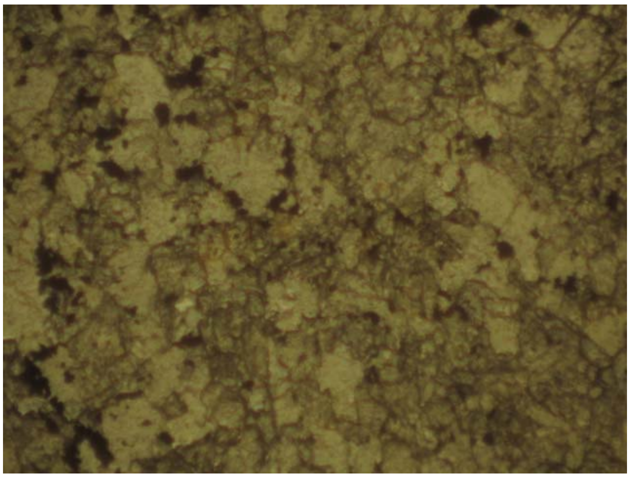

Fig. 5. Off-white to yellowish brown (angular to sub-rounded) grains of scheelite embedded in quartz and clinozoisite grains in plane polarized light (PPL) (40×)

The liberation of scheelite mineral was measured by grain mounts. The crushed ore was ground in a rod mill for 15 minutes and separated into a number of size fractions using different sieves (ASTM E 11-70). The grain mounts of various size fractions were prepared by placing a small amount of sample in one drop of glycerin on a glass slide and covering with a cover slip. These were analyzed under the microscope at a magnification of 40X. The number of liberated and locked mineral grains in each sieve fraction were noted and the percentage liberation was calculated. About 100 particles were counted for each slide. The data expressing the liberation of scheelite in various size fractions of Miniki Gole and Besti Gole is shown in the Table 5 and 6 respectively.

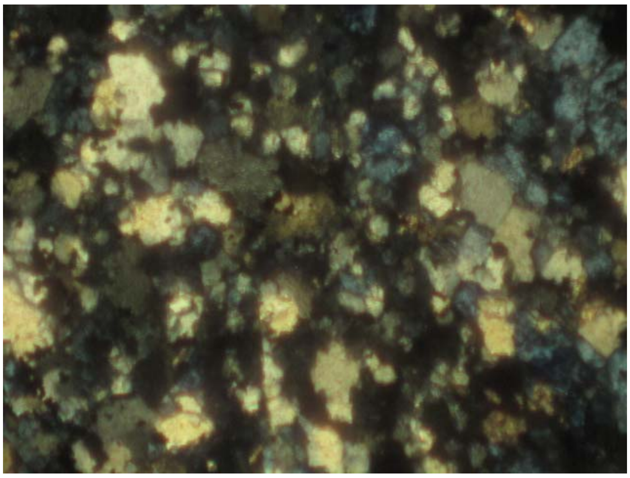

Fig. 6. Same photomicrograph under cross polarized light (XPL) (40×) showing light bluish clinozoisite matrix and yellowish to light brown crystals of scheelite in gray quartz

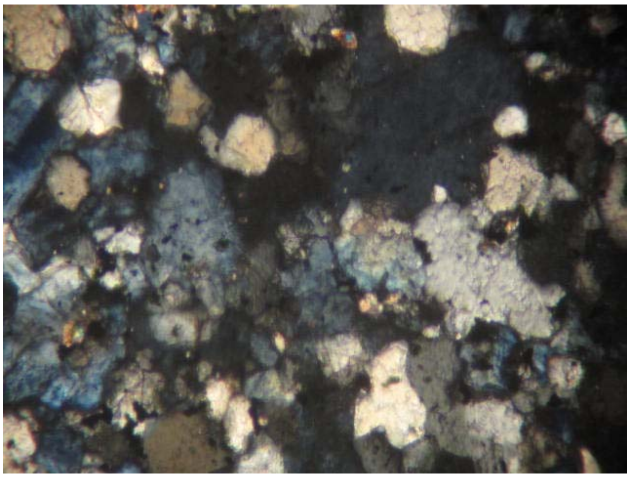

Fig. 7. Another view under high magnification (80×) shows grayish white to yellowish brown scheelite crystals in XPL

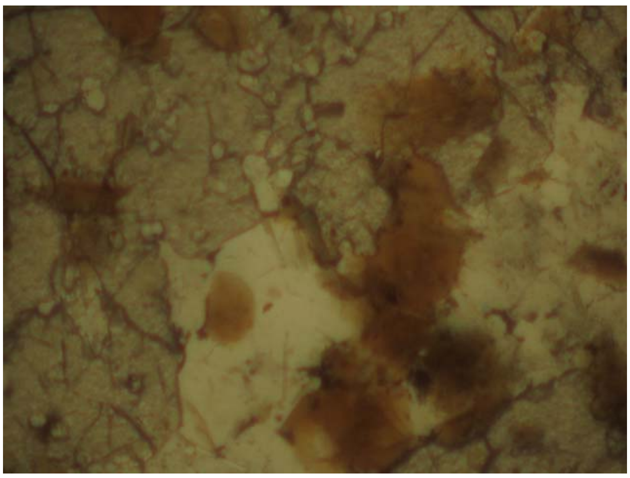

Fig. 8. Magnetite grains looks gray with brownish tint in PPL (80×)

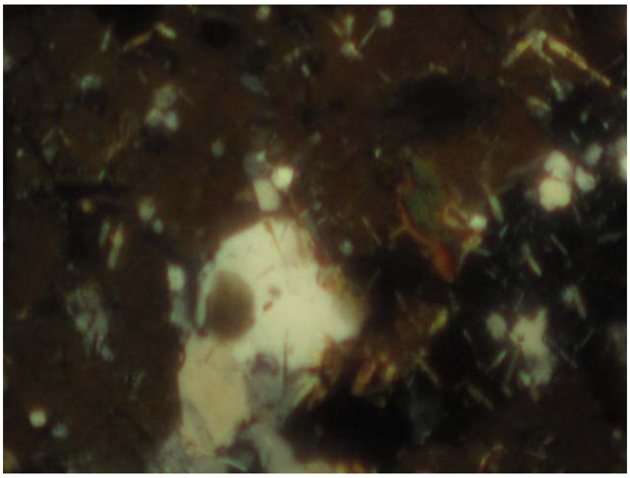

Fig. 9. Magnetite grains observed dark black under XPL in same photomicrograph (80×)

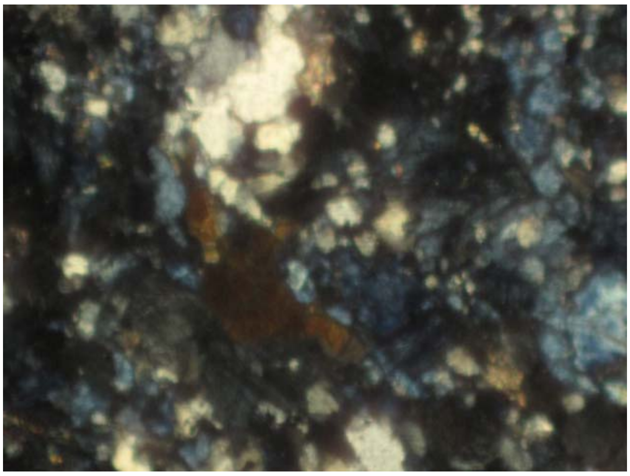

Fig. 10. An intermixing of off-white (medium to fine grained) scheelite, gray quartz, bluish clinozoisite, brownish red biotite and black magnetite in XPL (80×)

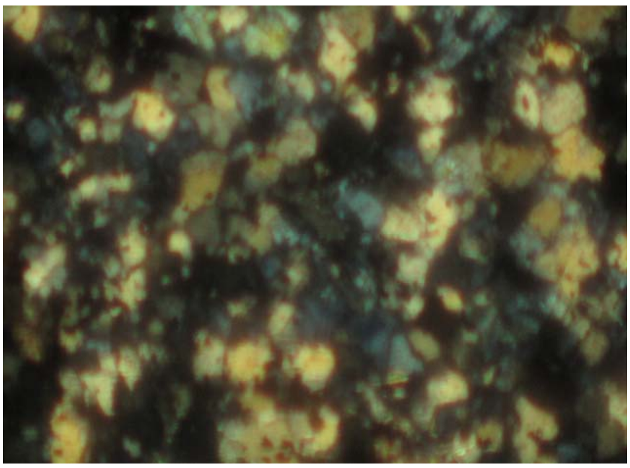

Fig. 11. Dark brown grains of biotite and bluish gray clinozoisite having different orientations in XPL (40×)

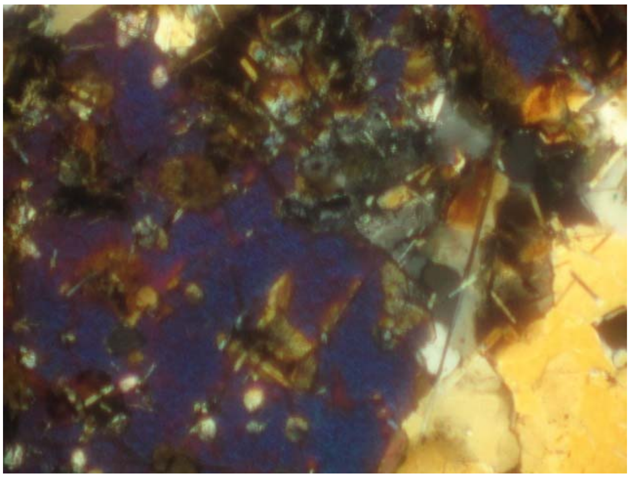

Fig. 12. The clinozoisite (light-bluish gray) associated with gray quartz in XPL (×40)

4. RESULTS AND DISCUSSION

Chemical analysis of the different samples collected from various sites of the deposits (Table 1 and 2) shows quite variation in WO_3 content of the ore. On the basis of this information, it is inferred that distribution of scheelite would vary from place to place. However, the chemical analysis of representative composite sample presented in Table-3 shows that the average WO_3 content in the bulk sample is 0.34% in Miniki Gole and 0.32% in Besti Gole. The obtained grade of the ore is quite sufficient to exploit it on commercial scale to produce tungsten concentrate (Srivastava & Pathak, 2000). Pandey et al., (2001) processed a low-grade scheelite ore (0.3 % WO_3) into a pre-concentrate assaying 13% WO_3 The pre-concentrate either can be directly utilized for extraction of tungsten (W) metal by hydrometallurgical treatment or it can be further upgraded into a higher grade concentrate (Guar, 2006). It is also clear from Table 3 that the main impurity is SiO_2 in Miniki Gole ore. It is present due to quartz and to some extent other silicate minerals as is obvious from Table 3. After SiO_2 the other major impurity is iron which is present in the form of magnetite. CaO is a part of calcite and scheelite where as Al_2O_3, Na_2O and K_2O are mainly associated with silicate minerals. It is also obvious that major impurity is CaO in Besti Gole ore being 90% along with minor amount of quartz scheelite. It is due to calcite mineral which hosts scheelite.

Table 5. Degree of liberation of scheelite in different size fractions of Miniki Gole tungsten ores (composite sample)

Size Fraction	Free Scheelite	Locked Scheelite		Free Gangue	Liberation of
-50+70	**3.63**	**8.39**	**12.02**	**87.98**	**30.20**
-70+100	**4.86**	**5.26**	**10.12**	**89.88**	**48.02**
-100+140	**4.93**	**3.20**	**8.13**	**91.87**	**60.64**
-140+200	**5.20**	**2.18**	**7.38**	**92.62**	**70.46**
-200+270	**5.25**	**0.86**	**6.11**	**93.89**	**85.92**
-270	**5.28**	**0.22**	**5.50**	**94.50**	**96.00**

Table 6. Degree of liberation of scheelite in different size fractions of Besti Gole tungsten ores (composite sample)

Size Fraction	Free Scheelite	Locked Scheelite	Free Gangue	Liberation of
-50+70	**4.10**	**7.24**	**88.66**	**36.15**
-70+100	**4.76**	**4.84**	**90.04**	**49.58**
-100+140	**5.82**	**3.70**	**90.48**	**61.13**
-140+200	**6.46**	**2.51**	**91.03**	**72.02**
-200+270	**6.55**	**1.23**	**91.32**	**84.19**
-270	**6.76**	**0.27**	**92.97**	**96.15**

The X-ray diffraction analysis (Figure 2 and 3) confirms the presence of considerable amount of scheelite mineral. It was identified by JCP.CAT search/match programme of the X-ray diffractometer that the other peaks corresponds to gangue minerals which mostly comprised of predominant quantities of quartz, calcite, epidote (clinozoisite), amphibole (actinolite), magnetite, biotite, muscovite, sphene in Miniki Gole areas and calcite and quartz in Besti Gole areas.

The microscopic examination of thin sections of the ore samples shows that scheelite looks off-white to yellow in colour with greasy luster in plane polarized light (PPL) and grayish white to yellowish brown in cross polarized light (XPL). It has massive and granular shape crystals which occur as medium to fine grains embedded in quartz grains (Figures 5-7). Under the microscope, the grain size of scheelite shows wide variations. It ranges from coarse crystals of 0.2 mm to fine disseminated particles of 0.03 mm. However, the majority of grains have a size in the range of 0.08 to 0.12 mm. The grain habits range from anhedral to euhedral tetragonal crystals with highly lustrous gray faces and a moderately high birefringence of 0.016 under crossed Nichols.

The quartz is present as medium to coarse grained matrix mineral (0.5 to 2.0 mm). Quartz appears as colorless when viewed under plane polarized light and has some shade of gray (typical grayish) when viewed under crossed polarizer (Figures 5-7). Magnetite grains are also coarse to medium, subhedral to anhedral and observed as gray with brownish tint in plane polarized light and dark black when viewed under crossed polarizer. It has no birefringence. Magnetite is partially altered to limonite (white blue) on exposed surface (Figures 8-9). Calcite is present as anhedral colorless grains. It is almost indistinguishable from other minerals under plane polarized light because of the distorted surface of the crystal, combined with its high birefringence and interference order.

Biotite looks as dark brown in plane polarized light and brown to dark red in cross polarizer (Figures 10-11). Muscovite is colorless with good cleavage. Epidote, mostly clinozoisite, is identified by its light-gray coloration in plane polarized light. Under cross polarizer, it is the light-bluish gray mineral. Photomicrographs of epidote (clinozoisite) in plain polarized light and in crossed polar are shown in Figures 11-12.

For efficient ore beneficiation, the knowledge of predominant range of grain (crystal) size is an important factor. It helps to take decisions concerning the grind size to which the ore should be ground in order to liberate the valuable mineral. Although, the grain size affects the recovery of any valuable element, but it is particularly important when dealing with precious and rare metals, whose full recoveries are normally required (Ferrara et al., 1989). Grinding of the ore is the most energy consuming unit operation in ore processing. Grinding at the coarse size, not only save the energy but also helps to reduce the production of slimes. The slimes generated not only decreases the recovery but also hinder in achieving the desired grade. Therefore, ore is ground to a size at which maximum minerals grains are liberated at the coarsest possible size, avoiding over-grinding (King, 1994).

The degree of liberation of economic minerals in new ores is often determined to estimate the required grind size (Ferrara et al., 1989). Table 5 and 6 represents the degree of liberation of scheelite mineral in various size fractions. It is clear from these Tables that liberation increases with a reduction in particle size and about 84 % scheelite grains are liberated when the ore is ground to minus 200 mesh (74 µm). Practically, the ore is grind to a size where the economic factor of the process is maximized and particle size is never reduced to achieve 100 % liberation (King, 1994). Therefore, in order to have a sufficient proportion of liberated scheelite grains with minimum level of locked particles, these ores should be ground from 80 to 90 % minus 200 mesh size. As mentioned earlier, the average grain size of scheelite also falls in this range.

Tungsten minerals are very amenable to gravity concentration because of their high specific gravity (Rao, 1996). In present ore, scheelite has specific gravity from 5.9 to 6.1 and associated gangue minerals have specific gravity ranging from 2.6 to 4.0 except magnetite. Due to this significant difference in specific gravity, it is possible to upgrade this ore by gravity concentration technique which can reduce most of the undesirable minerals. Magnetite having specific gravity (5.0 to 5.5) close to scheelite may be recovered along with scheelite concentrate. But, it is possible to separate it from concentrate by magnetic separation because of its high susceptibility.

However, the amenability of ores to beneficiation depends not only on nature of minerals but also on their texture (Adams, 1986). The ore under investigation is fine-grained in texture and some of scheelite crystals are finely disseminated within the host quartzite. Liberation of such fine scheelite grains (30-50µm) from the associated gangue minerals requires sufficient grinding. Since scheelite tends to slime badly on excessive grinding due to its brittle and friable nature. Therefore, gravity concentration techniques may not succeed to produce desired grade concentrate with high recovery.

However, to produce economic grade tungsten concentrate with acceptable recovery, such ore can be treated either by a combination of physical beneficiation techniques such as gravity concentration at coarser size followed by re-grinding of the pre-concentrate and subsequently its flotation or by an all over froth flotation process. Flotation is an important process in mineral processing which utilize the physical and chemical properties of different minerals for their separation. The scheelite mineral ($CaWO_4$) generally responds well to flotation process using carboxyl acid collector (Wills, 1992). The mineralogical composition of the ore indicates that only calcite ($CaCO_3$) may interfere in its flotation because of similar surface behaviour. Therefore, this ore can be beneficiated by single froth flotation process. The associated gangue minerals such as quartz and various silicate minerals can be depressed using sodium silicate or sodium pyrophosphate as depressant while the carbonate gangue mineral (calcite) may be depressed either with quebracho or tannins.

5. CONCLUSIONS

The only valuable tungsten bearing mineral identified in the samples collected from various localities of Miniki Gole deposit and Best Gole is scheelite while the rest of the matter is associated gangue minerals. The ore bodies comprise of mainly calcite in Besti Gole and quartz and clinozoisite as major minerals while actinolite, magnetite, calcite, biotite, muscovite and sphene as minor minerals in Miniki Gole deposit. Thin sections of the ores reveal that both deposits are fine-grained in texture and minerals phases are finely disseminated in host quartz. The distribution of free and locked scheelite grains of various size fractions of the ores show that these should be ground to more than 80% minus 200 mesh size to have an economic degree of liberation. It is concluded that mineralogical and textural observations of the ore indicate that the froth flotation is more suitable technique for up-gradation of these ores as compared to gravity concentration methods.

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