Jour. Korean Inst. Mining Geol.
Vol. 26, No. 3, p. 259-265, 1993.

H/F Variation in Wolframites According to Depth and Temperature of Mineralization at Ssangjeon, Weolag, Cheongyang and Sannae Mines, Korea

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ABSTRACT: The variation of H/F coefficient (Hubnerite/Feberite coefficient) and the temperature of formation with depth have been studied for the hydrothermal tungsten-quartz veins from Ssangjeon, Weolag, Cheongyang, and Sannae mines. All samples were selected at the same paragenetic stage and mineral assemblege according to depth. The studied mines provide an opportunity to examine and regional variations in wolframite composition in different provinces.

The formation temperature is linearly related to the depth. At the Weolag, Cheongyang and Sannae mines, the H/F ratio of wolframite and filling temperatures of fluid inclusions in quartz show a general tendency to decrease at shallow levels, in spite of different geological settings. This implies the pH of fluid will decrease vertically with falling temperature and the high H/F ratio of wolframite in deep zone result in an increase of pH with depth. The Ssangjeon mine exhibits a tendency to increase slightly upwards at shallow level. This implies a different geochemical environment (high pH environment) for the Ssangjeon mine, as compared to the other three mines.

The H/F coefficient pattern suggests that it is a useful geothermometer for vein-type tungsten deposits in Korea. The H/F coefficient as an indication of depth should be examined for use in exploration.

INTRODUCTION

Compositional variations in wolframite are primarily controlled by pH, T and the aMn^{+2}/aFe^{+2} ratio of the fluid. The MnO/FeO ratio of wolframite is a geothermometer (Horner, 1979; Chernyshev and Ivanonva, 1969). It has been concluded empirically that there exists positive correlation between the MnO/FeO ratio and deposition temperature of wolframite (Taylor and Hosking, 1970; Leutwein, 1952). These investigators claimed that hubnerite is found mainly in the lower (high temperature) parts of wolframite veins.

Studies of vein type tungsten deposits have commonly indicated a general decrease in H/F ratio of wolframite away from the source area (Taylor and Hosking, 1970). However, many exceptions have been noted (Barabanov, 1971; Groves and Baker, 1972). Pegmatite type generally indicated high MnO contents for wolframites from pegmatite veins (Hosking and Polkinghorne, 1954). Limited studies of zoned crystals have generally indicated a possible inverse relationship between MnO/FeO ratio and temperature of deposition. However McIntire (1963) and Broecker and Oversby (1970) proposed that a temperature-dependent equilibrium existing between wolframite and other Fe- and Mn-bearing minerals play an important role.

The aims of present study are to determine the experimental variation for the H/F ratio with respect to the temperature of formation in different environment and to correlate it with depth and locality of wolfrmite-bearing vein deposits in Korea. For this study, the close association of wolframites and quartz and these ocurrences with depth were carefully considered to take samples from four mines Ssangjeon, Weolag (no. 2 vein), Cheongyang (Eastern no. 2 vein) and Sannae mine (Fig. 1). The formation temperature obtained from the homogenization temperature of the quartz, may imply that of the wolframite closely associated with the quartz.

GEOLOGY AND MINERALIZATION OF THE HYDROTHERMAL TUNGSTEN DEPOSITS

Ssangjeon mine: It is located within the Precambrian metamophic belt. The wolframite bearing quartz vein is developed within a pegmatite along the contact between the granitic gneiss on the footwall and the hornfels schist unit on the hanging wall. The ore vein, ranging from 3 to 40 m in width, is followed

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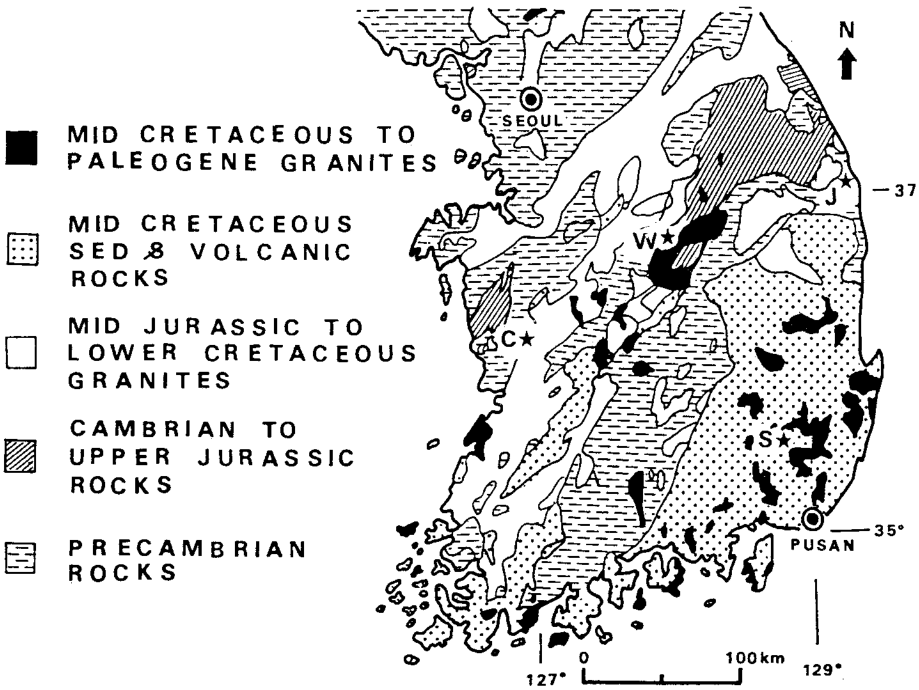

Fig. 1. Simplified geologic map of the Republic of Korea showing the location for the studied mines (J; Ssangjeon mine, W; Weolag mine, C; Cheongyang mine, S; Sannae mine).

along strike for 1000 m horizontally and 400 m vertically. The tungsten grade and tonnage increase where the ore vein thickens. The ore vein also displays chemical zonation with depth; becoming enriched in W, Cu and Sn and depleted in As, Ni, Co and Bi with depth (Kim et al., 1979). The paragenesis is divided into three stages: 1) early pegmatitic stage, 2) main tungsten stage and 3) late fluorite stage (So, et al., 1983). The major minerals are wolframite, scheelite, and arsenopyrite with accessory pyrite, chalcopyrite, stannite and rare bornite. K-feldspar occurs as main alteration product. It is characteristic that wolframite crystals are partially replaced by scheelite; with the replacement progressing along fractures and cleavages to develope a shredded texture.

Weolag mine: It is located within the Hwanggangri mineralized region of the Ogcheon metamophic belt. The Weolag mine consists of a series of the main parallel sets of fissure-filling quartz veins (width; 2-60cm) which extened from Late Cretaceous granite (K-Ar dates from 87 to 90 m.y., Choo, 1971) into hornfels and calc-silicate rocks. The veins can be followed along strike for about 1 km horizontally and 400 m vertically. The paragenesis is divided into three stages: 1) early molybdenite-wolframite stage, 2) iron sulfide-scheelite stage, and 3) late carbonate stage (So, et al., 1983). The major minerals are wolframite, schellite, molybdenite, bismuthinite, and native bismuth, with accessory sphalerite, galena, chalcopyrite, pyrite, and pyrrhotite. Strong sericitic alteration is developed in wall rock.

Sannae mine: It is located within the Cretaceous Gyeongsang basin. The hydrothermal vein type deposits which comprise the mine occur within an Upper Cretaceous quartz monzonite (Rb-Sr dates to 75.5 m.y., Choo and Kim, 1981) which intrudes the middle Cretaceous sedimentary and volcanic rocks of the basin. The age of mineralization is 65± 3 m.a. based on K-Ar data of sericite from quartz veins within the mine (Fletcher and Rundle, 1977). The ore vein varies from 0.1 to 2 m in width. A distinct metallic and mineral zonation is apparent; scheelite/molybdenite ratio and scheelite/wolframite ratio decrease with depth, respectively. The fissure-filling quartz veins can be followed along strike for about 3 Km horizontally and 400 m vertically. The mineral paragenesis is divided into three stages: 1) early molybdenite, 2) main tungsten, and 3) late carbonate (Shelton et al., 1986). The major ore minerals are molybdenite, wolframite and scheelite together with pyrite and minor amounts of rutile, hematite, and chalcopyrite are contained within the quartz vein.

Cheongyang mine: It is located in Precambrian biotite gneiss adjacent to a contact with a small granitic stock of Mesozoic age. The granite has been regionally intruded by quartz porphyry dikes of uncertain age. The Cheongyang W-Mo deposits are composed of numerous fissure-filling quartz veins contained within the Precambrian biotite gneiss and granite porphyry. Some ore veins are closely associated with quartz porphyry. The deposits are comprised of 6 to 7 quartz veins varying from 5 to 100 cm in width and extending up to 2 Km in length and 500 m in depth. The paragenesis can be devided into three distinct stages: 1) wall rock alteration stage (sericitic alteration) 2) early vein stage, 3) late vug stage. The major minerals are wolframite and molybdenite with small amount of pyrite, sphalerite, galena, chalcopyrite, scheelite, fluorite, rhodochrosite, calcite, beryl, muscovite and rare arsenopyrite, chalcocite, marcasite, bismuthinite, native bismuth, cosalite and stannite.

EXPERIMENTAL PROCEDURE

Fluid Inclusion Study

First of all, it was necessary to measure the temperature accompanying the mineralization process along the depth sampled for each mine. To do this, used fluid inclusions in quartz sampled from same vein, which was considered to have grown nearly simultaneously or appeared to be coprecipitated with wolframite in same stage and paragenesis was used. The fluid inclusions were examined in thin (1 mm or less) doubly polished plates. Most heating measurements were made on a Leitz Microscope 350 and a 1350 Heating Stage. The heating stage was calibrated relative to the melting point of Merck standards. Three types (type I, type II, type III) of primary fluid inclusions recognized in this study are classified according to the terminology of Nash (1976).

Liquid-rich type I and vapor-rich type II fluid inclusions are intimately associated in some samples from all studied mines and homogenize at similar temperatures. These observations indicate periodic boiling of the fluids during mineralization. Therefore, no pressure correction is necessary for inclusions trapped along the two phase boundary in the system H_2O-NaCl.

Geochemical Study

Quarts and wolframites were sampled in the plan

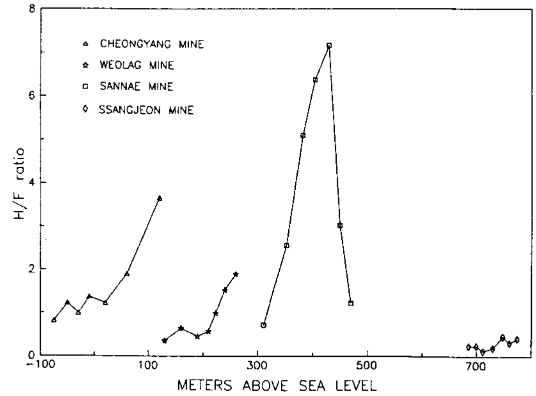

Fig. 2. Relation of H/F ratios in wolframite to depth. Symbols mean average H/F ratio.

of a vein along a well defined profile which began at the thermal source of the system. All samples were selected at the same stage in paragenesis. After sampling, each wolframite specimens was pulverized and purified for X-ray analysis.

X-ray diffraction analysis (chart speed; 1/8 min, internal standard; SiO_2, unfillted Cu radiation) were performed to determine compositions in solid solution FeWO4-MnWO4. The dependence of the unit cell parameter on the composition of $FeWO_4$-$MnWO_4$ solid solutions obey Vegard's rule and is described by equation: a (Å)$=4.732+0.00098\times MnWO_4$ (mol.%) ± 0.002 (Chernyshev et al., 1976).

The Sannae samples are estimated for comparision the X-ray fluorescence technique and X-ray powder diffraction method. There is a fair agreement between the XRF and the XRD composition estimates.

RESULT

The analytical results and relationships from deposits will be presented and followed by a general discussion.

Local and Regional Variation in H/F Coefficient

All studied mines provide an opportunity to study local and regional variation in wolframite composition in different provinces. There is a relatively restricted range of composition. The compositions of wolframite from the Ssangjeon mine are significantly different from those of other mines (Fig. 2). Table 1 present the values and local variation of H/F ratio in wolframites. The variation range of H/F ratio in wolframites from the Sannae mine is the greatest (0.72

Table 1. Temperature for fluid inclusion in quartz associated with the wolframite.

Sample No.	Location(level)	Elevation(m)*	H/F ratio	T_h(℃) Range (average)
		SSANGJEON MINE		
S1	Seungbu	774	0.41	244~273 (257)
S4	12 m/Sangjeon	760	0.31	252~325 (291)
SC2	Ssangjeon	748	0.46	271~336 (299)
M3	Main	730	0.19	312~326 (317)
C3	Central	712	0.12	284~359 (334)
L5	14 m/lower one	700	0.24	292~372 (317)
L6	Lower one	686	0.23	341~387 (362)
		WEOLAG MINE (No. 2 vein)		
MM2	Main	260	1.90	221~331 (272)
ML10	10 m/Lower one	240	1.52	232~357 (286)
ML4-1	Lower one	223	0.99	284~317 (303)
ML1-2	−10 m/Lower one	210	0.58	289~346 (308)
ML2-5	Lower two	190	0.46	281~367 (323)
ML3-11	Lower three	160	0.65	293~372 (317)
ML4-4	Lower four	130	0.36	284~384 (355)
		CHEONGYANG MINE (Eastern No. 2 vein)		
U3-1	Upper two	120	3.68	223~289 (249)
M3	Main	60	1.90	235~322 (268)
M14	Lower one	20	1.22	221~301 (267)
L2	Lower two	-10	1.36	243~325 (273)
L3	Lower three	-30	0.99	272~366 (343)
L4	Lower four	-50	1.22	275~370 (345)
G1	Eastern	-75	0.81	327~390 (361)
		SANNAE MINE		
S1	One	470	1.22	290~352 (325)
S2	Two	450	3.03	301~375 (340)
S3	Three	430	7.17	340~414 (376)
S5	Five	405	6.37	361~424 (392)
S6	Six	382	5.10	406~453 (425)
S7	Seven	352	2.57	411~451 (427)
S9	Nine	311	0.72	425~455 (440)

*Meters above sea level

to 7.17). Followed by the Weolag mine (0.36 to 1.90), the Cheongyang mine (0.81 to 3.68), and the Ssangjeon mine have the smallest range (0.12 to 0.46). The median values of $MnWO4$ composition (mol.%) in wolframite from the Sannae mine have the highest (71.5), followed by the Cheongyang mine (57.8) and the Weolag mine (44.1), and the Ssangjeon mine have the lowest value (21.2). Generally the variation ranges increase with H/F coefficient proportionally. These phenomena are related to the formation temperature of wolframite (refer to next section).

The changes in chemical behavior during the crystallization of wolframite under various geological conditions are shown by their concentration and varying H/F ratios.

H/F Coefficient and Filling Temperature with Mineralization Depth

Filling Temperature with Depth: Thirty doubly polished sections of quartz closely associated with wolframite were prepared from different levels in all studied mines. Fluid inclusion filling temperatures show a consistent relationship with respect to depth (Fig. 3). A general tendency for filling temperature to rise with decreasing depth can be noted. The results of heating studies on inclusions from various depth are presented in Table 1 and Fig. 3.

In the Ssangjeon mine, the temperature decreases from an average of 362℃ on Lower 1 level to an average of 257℃ on Seungbu level. In the Weolag mine, filling temperature decreases from an average of 355℃ on Lower 4 level to an average of 272℃ on Main level. In the Cheongyang mine, filling temperature decreases from an average of 361℃ on Eastern level to an average of 249℃ on Upper 3 level. In the Sannae mine, the temperature decreases from

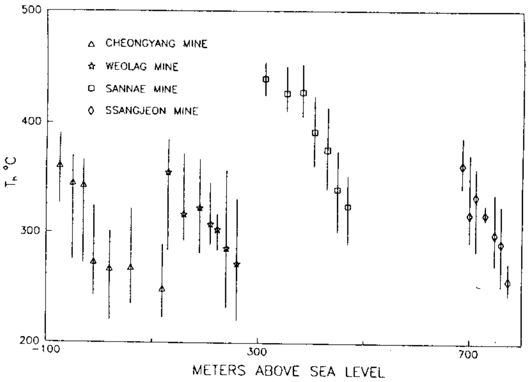

Fig. 3. Patterns of homogenization temperature and depth vs. each mines. Average temperatures are shown by symbols.

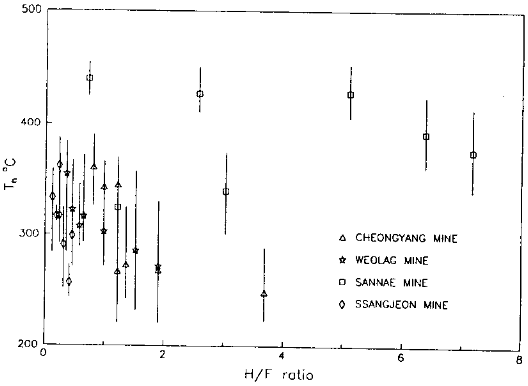

Fig. 4. Relation between H/F ratio and homogenization temperature. Average temperature are shown by symbols.

an average of 440℃ on No. 9 level to an average of 325℃ on No. 1 level. Although there is a significant overlap of homogenization temperature ranges, there is a consistant decline of average temperature according to depth. The H/F ratios for individual samples also show an interesting relationship with respect to depth (elevation). Data from these mines indicate a positive linear relationship in all stuided mines. When filling temperatures are plotted against elevation for individual mine (Fig. 4). The temperatures, however, show no systematic decreasing curve with depth. The average gradient is predominantly in the range of 5 to 10℃ per 10 m, occasionally exceeding 20℃.

Variation of H/F ratio with Depth. Amosse (1978) has suggested that the H/F coefficient in wolframite is related to the distance of vein mineralization from granitic intrusives. In Fig. 2 the variation of H/F ratios with different depths of mineralization are presented in detail. There is a consistent trend of increasing H/F ratio with decreasing elevation except for the Ssangjeon mine. The Ssangjeon mine show a tendency to decrease (Fig. 4). These results indicate that changes in the temperatures and/or compositions of the wolframite took place with mineralization depth.

DISCUSSION

Compositional variations in wolframite are primarily controlled by pH, T, and aMn^{+2}/aFe^{+2} ratio of the fluid. The thermodynamic data compiled by Lvanova and Khodakovskiy (1968), Horner (1979) suggest that at low [W] and $[Me^{+2}]$ concentrations the temperature dependence of the solubility product of $MeWO_4$ is the major factor governing the precipitation of $MeWO_4$. That is, the higher the solubility product of $MeWO_4$ and the lower the $[Me^{+2}]$ and [W] concentrations, the higher the precipitation temperature of $MeWO_4$. Thus with falling temperature the wolframite composition coexisting with a fluid of constant cation concentration ratio becomes more ferruginous. The thermodynamic parameters for $FeWO_4$-$MnWO_4$ solid solution could allow the determination of the equation representing the variation of the molar Mn value with the physicochemical conditions such as temperature and Fe, Mn activity in the mineralizing solution.

This phenomenon is a consequence of a decrease in the H/F ratio of the mineralizing solution during the mineralization process. The lack of Fe- and Mn-bearing minerals in equilibrium with wolframite suggests that the decreasing in ratio of Mn to Fe is related to this process, the deposited wolframite having a Mn/Fe ratio greater than that of the fluid. $FeWO_4$ and $MnWO_4$ show quite different behavior in solubility. $FeWO_4$ can from over a wide temperature range, but $MnWO_4$ from only at temperature above 200℃ ($FeWO_4$ requires more acidic solutions whereas $MnWO_4$ needs neutral to weakly alkaline conditions). At the Weolag, Cheongyang and Sannae mine, the H/F ratio of wolframite and filling temperature of fluid inclusion in quartz show a general tendency to decrease upwards at shallow levels in spite of different geologic settings. Decreasing temperature pH with depth result in an increase of high H/F ratio of wolframite with depth. The Ssangjeon mine present a tendency to slightly decrease at shallow level. It means that different geological environments are present for the other mines.

The H/F coefficient pattern suggests the use as geothermometer in vein type tungsten deposits of Korea. The depth control of H/F coefficient should be closely examined for use in exploration.

ACKNOWLEDGEMENT

This paper was supported by CMR (Center for Mineral Resources Research) which is greatly acknowledged.

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Manuscript received 18 March 1993

쌍전, 월악, 청양 및 산내 철·망간중석 광산의 유체포유물 온도와 심도에 따른 H/F 값의 변화

박 맹 언

요 약: 본 논문은 심도별 시료 채취가 가능한 국내 주요 함중석 열수석영맥 광상 (쌍전, 월악, 청양, 산내광산)에서 산출되는 철·망간중석과 동일맥 중 공생단계에 있어서 밀접하게 수반되는 같은 광화시기의 석영을 대상으로 유체포유물 온도, 철·망간중석에서의 H/F값 및 광화작용의 심도와의 상관관계를 밝히기 위해서 시도 되었다.

연구대상 함중석 열수석영맥광상에서 철·망간중석의 H/F값은 생성온도와 광화작용의 심도에 따라 특징적인 상관관계를 보여준다. 월악, 청양 및 산내광산에서 산출되는 철·망간중석의 H/F값은 광화온도가 높을수록, 광화심도가 깊을수록 증가된다. 이는 광화용액의 pH값이 냉각과 모암과의 반응에 따라 감소되었음을 시사한다. 쌍전광산의 경우 생성온도가 높아짐에 따라서 상대적으로 제한된 범위내에서 H/F값이 증가되나, 심도에 따라서는 상기 광산과는 상반된 결과를 나타낸다. 이러한 결과는 알카리장석과 견운모등의 침전에 따른 pH값의 증가를 유도한 물리·화학적 환경이 조성되었음을 의미한다.

본 연구에서 밝혀진 함중석-휘수연 열수 석영맥에서 산출되는 철·망간중석의 심도에 따른 생성온도와 H/F값의 특성은 유사한 지질환경의 물리화학적 조건(동일한 공생단계)이 형성되면 지질온도계로서의 활용이 가능하며, H/F값은 열수석영맥에서 산출되는 철·망간중석광의 심도 탐광시 고려되어야 할 요건으로 심도추정 및 탐사지침으로 활용할 수 있을 것으로 사료된다.