Geochemical Characteristics of Tungsten in Miyako Granitoid and Scheelite-bearing Aplitic Veins at the Yamaguchi Cu-W Skarn Deposit, Iwate, Japan

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ABSTRACT

Tungsten analyses were made on 16 samples collected from the North Miyako granitic body in Northeast Japan. Petrographic facies of the North granitic body vary from quartz diorite in the marginal zone (zone A), to tonalite and granodiorite (zone B), and to granite in the central zone (zone C). A large number of barren and scheelite-bearing aplitic veins are distributed around the Yamaguchi deposit which occurs in the contact aureole of zone C granite. The tungsten content of zone C granite is lower than that of the granitic rocks in zones A, B and the aplitic veins. It appears that tungsten in the differentiated granitic magma, which was associated with ore mineralization, was transported out of the magma chamber by magmatic fluids.

The tungsten content is generally low in the North Miyako granitic rocks but high in granitic rocks of Okinoshima zoned pluton, as well as in Otani granite and Busetsu granite from the Southwest Japan. In the case of magnetite-series, however, the behavior of tungsten in the Miyako granitic body from the tungsten metallogenic province is similar to that of the Okinoshima zoned pluton from molybdenum metallogenic province in Southwest Japan. Behavior of tungsten in granitic magma is affected by magmatic evolution during the process of saturation of granitic melt with magmatic fluid.

Keywords: Tungsten, Magmatic fluid, Devolatilization

INTRODUCTION

Ore elements such as tungsten, molybdenum and tin are important for understanding magmatic hydrothermal mineralization related to granitic magmatism. According to the computational models of magmatic devolatilization, behavior of ore metals such as tungsten, molybdenum and tin are affected by the generation of magmatic fluids separated from highly differentiated granitic magma [1].

Ishihara [2] divided the ore related granitoids into three provinces: molybdenum metallogenic province, tungsten metallogenic province, and tin-tungsten metallogenic province (Fig. 1). The molybdenum and tin-tungsten metallogenic provinces are related to the magnetite-series granitoids and the ilmenite-series granitoids, respectively [2]. The difference between those two provinces is the result of different oxidation states of granitic magma between the two series [2]. On the other hand, magnetite-series granitoids are related to both molybdenum and tungsten metallogenic province. It is possible that difference

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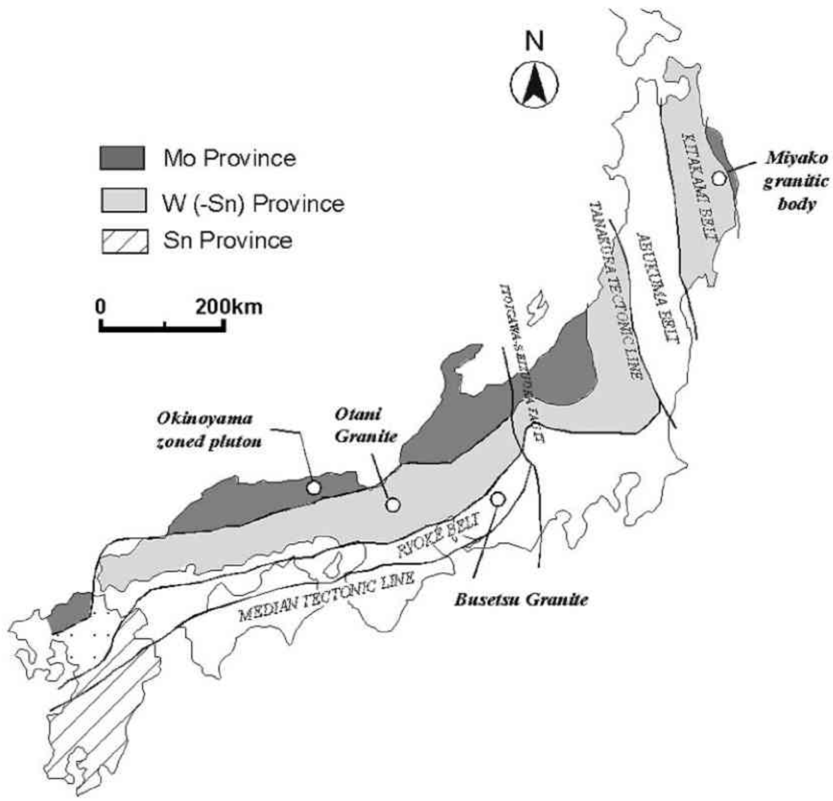

Fig. 1 Molybdenum and tungsten-tin metallogenic zoning of the Japanese islands (after Ishihara [6]).

between tungsten and molybdenum mineralization related to magnetite-series granitic magmatism is affected by mechanism of fractional crystallization and generation of magmatic fluids, separated from highly differential granitic magma, rather than the physicochemical state of granitic magma. In order to understand tungsten mineralization related to granitic magmatism, it is necessary to concentrate on tungsten occurring in granitic rocks.

There are very few previous studies related to the concentration of tungsten in granitic rocks and the genetic relationship among concentrations of ore metals in the granitic rocks [3–7]. In this paper, the tungsten concentration in the Miyako granitic body associated with Yamaguchi W-Cu skarn deposit is described. Tungsten concentration data are applied to deduce the genetic relationship between the ore forming fluid and crystallizing granitic melt, as well as to characterize the granitic magma involved in the mineral-melt- vapor reaction.

OUTLINE OF GEOLOGY

Large numbers of the magnetite-series granitic bodies are distributed in Kitakami belt of the Outer Zone in Northeast Japan. The Miyako granitic body is one of the Early Cretaceous magnetite-series zoned plutons in the North Kitakami belt. Ishihara [1] divided it into Mo and W metallogenic provinces of Kitakami belt, based on the location of the Yamaguchi W-Cu skarn deposit.

The Miyako granitic body is distributed in the magnetite-series granitic terrain and tungsten metallogenic province [1]. It has a size of about 50 km × 20 km in plan view with a balloon-like shape and a northwest-southeast extension (Fig. 2). The age of the emplacement of the Miyako granitoid was estimated by the K-Ar method to be 109~135 Ma [8] and by the Rb-Sr method to be 116~140 Ma [9]. The Miyako granitic body is divided into North and South bodies based on lithologic and compositional differences [10] (Fig. 1). Katada and Kanaya [11] reported that the South body intruded into the North body. The Yamaguchi Cu-W skarn deposit occurs in the contact aureole of the North Miyako granitic body.

On the basis of modal analysis, the North Miyako granitic body consists of quartz diorite (zone A), tonalite to hornblende-biotite grano-diorite (zone B) and hornblende-biotite granite (zone C) (Fig. 2). Ac-

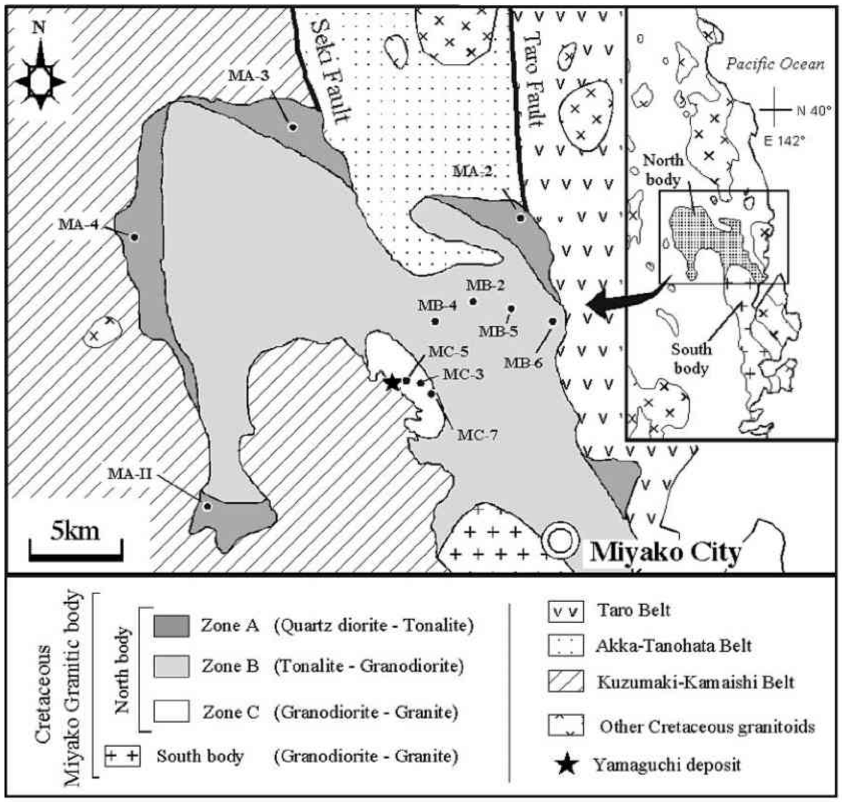

Fig. 2 Simplified geological map of the Miyako granitic body adapted from Nishioka et al. [10]. Numerals are sample localities presented in Table 2.

cording to the grain size of hornblende and biotite, the granitic rocks gradually change from fine-grained to coarse and non-equigranular from zone A through zone B to the boundary between zones B and C. The zone C granite frequently includes the country rocks as roof-pendant blocks. Aplite and pegmatite commonly occur in the zone C, close to the Yamaguchi deposit.

The Yamaguchi Cu-W skarn deposit occurs in the contact aureole of zone C granite. The distance between the Yamaguchi deposit and the granitic rocks in zone C is less than 100 m. Tungsten mineralization associated with scheelite ($CaWO_4$) occurs in aplitic veins. The scheelite-bearing aplitic veins cut massive skarn, which commonly contains the aggregates of fine-grained scheelite crystals mainly near the veins. The scheelite-bearing aplitic veins are abundant of albite, plagioclase, and titanite with subordinate amount of K-feldspar, quartz and scheelite.

ANALYTICAL PROCEDURE AND RESULTS

Major elements and tungsten analyses were made on about 16 samples taken from granitic rocks and aplitic veins from the North Miyako granitic body. Locations of the analyzed samples are illustrated in Fig. 2. The concentration of SiO_2 was analyzed using XRF at Akita University (RIGAKU 3270) using fusion glasses made from a mixture of powdered sample and alkali fluxes in the proportion of 1 : 5. Tungsten was analyzed by ICP-MS (PQ-3, VG Elemental Co.) at Akita University. The methods of sample digestion and preparation of solution for ICP-MS analyses were the same as described by Sato et al. [12].

Results of analyses on SiO_2 and W are given in Table 1. The results from the duplicate analyses for tungsten concentration in the Japanese geological standards (JB-1 and JG-2) and the Miyako granitic rocks (MA-2, MB-2 and MC-5), used to calculate experimental error are shown in Table 2. Coefficients of variations (CV) of tungsten concentration were less than 12% for concentrations > 0.15 ppm in samples JB-1a, JG-2, MA-2 and MB-2. A larger CV value (34.1%) for MC-5 probably results from the smallest concentration of tungsten. The detection limit of tungsten concentration by ICP-MS, calculated after Yoshida et al. [14], is approximately 0.02

Table 1 Analytical results (contents of SiO_2 and W) for samples of the granitic rocks and aplitic veins

Sample No.	Rock type	SiO2 (wt.%)	W (ppm)
North Miyako granitic rocks			
Zone A			
MA-2	Biotite-hornblende quartz diorite	61.64	0.40
MA-3	Biotite-hornblende quartz diorite	54.55	0.15
MA-4	Biotite-hornblende quartz diorite	55.92	0.18
MA-II	Biotite-hornblende tonalite	64.67	0.46
Zone B			
MB-2	Biotite-hornblende granodiorite	64.20	0.56
MB-4	Biotite-hornblende granodiorite	65.69	0.34
MB-5	Biotite-hornblende tonalite	62.88	0.63
MB-6	Biotite-hornblende tonalite	62.68	0.39
Zone C			
MC-3	Biotite-hornblende granite	72.16	0.19
MC-5	Biotite-hornblende granite	72.34	0.15
MC-7	Biotite-hornblende granite	71.46	0.28
Aplitic veins			
BA-1	Barren aplitic vein	76.42	0.63
BA-2	Barren aplitic vein	75.14	0.93
BA-4	Barren aplitic vein	76.59	1.10
MM-0	Scheelite-bearing aplitic vein	58.38	8000

ppm (2σ).

DISCUSSION

Figure 3 shows the variation diagram of tungsten against SiO_2 concentrations for the North Miyako granitic body in Northeast Japan. For comparison, data for similar plutons in Southwest Japan, such as Okinoyama zoned pluton (magnetite-series: molybdenum metallogenic provinces), the Busetsu granite (ilmenite-series: barren zone) and the Otani granite (ilmenite-series: tungsten metallogenic province) after Ishihara [6, 7] are also plotted in the same diagram.

The tungsten content in the North Miyako granitic body slightly increases from zone A (0.15–0.46 ppm) to zone B (0.34–0.63 ppm) and then decreases from zone B to zone C (0.15–0.28 ppm). However, the tungsten content drastically increases from zone C to the barren aplitic veins (0.63–47.67 ppm). Tungsten content in scheelite-bearing aplitic veins is ca. 0.8 wt% [15] that is mush higher than in the granitic rocks from all zones.

The trends of concentration of tungsten in the North Miyako granitic body and aplitic veins are similar to those of copper [15]. Ogata et al. [15] suggested that behavior of Cu in the granitic rocks in the North Miyako granitic body was affected by a mechanism that differs from simple magmatic fractional differentiation and also transportation out of the granitic melt by fluid coexisting with the melt. Therefore, tungsten in granitic magma might have been transported out of the magma chamber by magmatic fluids.

The Tungsten concentration in the North Miyako granitic body is lower than that in the other plutons of Southwest Japan (Fig. 3). The tungsten concentration in granitoids is somewhat similar to tin contents, which are generally high in Southwest Japan and low in Northeast Japan [16]. Ishihara and Terashima [16] suggested that the low tin granitoids of Northeast Japan and high tin granitoids of Southwest Japan were formed by granitic magmatism which occurred in intra-oceanic island arc and continental margin, respectively.

Table 2 Duplicate analytical results (n = 5) of geochemical reference samples and the North Miyako granitic rocks

	R.V. (ppm)	mean (ppm)	σ (ppm)	CV (%)	R.E. (%)
JB-1	17.1	11.87	0.55	4.6	−31
JG-2	23	20.60	0.26	1.3	−10
MA-2	—	0.40	0.03	8.2	—
MB-2	—	0.56	0.06	11.4	—
MC-5	—	0.15	0.05	34.1	—

R.V.: recommended value[13]
mean: arithmetic means of duplicate analyses
σ: standard deviations of duplicate analyses
CV: coefficients of variation
R.E.: relative error between R.V. and mean value.

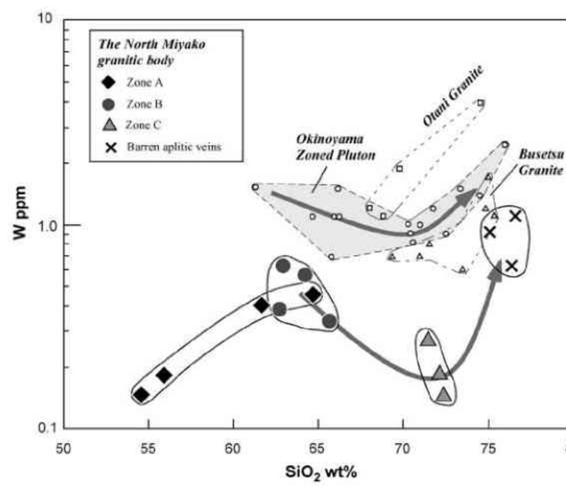

Fig. 3 Tungsten content (ppm) plotted against SiO_2 wt% in granitic rocks.

Behavior of tungsten in the granitic melt is different between magnetite-series and ilmenite-series granitoids. In the magnetite-series granitoids, variation of tungsten content in the North Miyako granitic body is similar to that in the Okinoyama zoned pluton. On the other hand, tungsten contents in the Otani and Busetsu granites of ilmenite-series increase with increasing SiO_2 concentration. Therefore, it is possible that the Okinoyama granitic magma coexisted with magmatic fluid that transported tungsten out of granitic melt.

Tungsten and molybdenum species are contained mainly in OH complexes and in some cases in Na- and K- hydroxide complexes in the magmatic fluid [17, 18]. The solubility of tungsten in magmatic fluid coexisting with the granitic melt increases with increasing pressure and irrespective of the chloride concentration in magmatic fluid [19]. On the other hand, the solubility of molybdenum in magmatic fluid increases with increasing pressure and/or the chloride concentration in magmatic fluid [18, 19]. Shinohara et al. [20] suggested that the partition ratio of chloride (the ratio of Cl concentration in fluid to Cl concentration in melt) exhibits a strong positive pressure dependence. Therefore, W/Mo ratio of ore-forming magmatic fluid was probably determined by the depth of devolatilization of the granitic melt.

In the Okinoyama zoned pluton, high tungsten concentration among granitic rocks is observed in the granitic porphyry [7], which is the hypabyssal rock emplaced at shallow environment, rather than the North Miyako granitic body. Shimazaki [21] indicated that the depth of formation of the deposit in the inner zone of Southwest Japan is relatively shallower compared to the Kitakami belt of Northeast Japan. It is possible that the depth of emplacement and devolatilization of the North Miyako granitic body is relatively deeper than that of the Okinoyama zoned pluton. Chlorine concentration in apatite from magnetite-series granitoids distributed in the outer Southwest Japan is higher than in other similar granitoids in the Kitakami belt [22]. Therefore, chloride concentration of magmatic fluid coexisting with granitic magma of the North Miyako granitic body is relatively lower than those of the Okinoyama zoned pluton. Additionally, W/Mo ratio of the mineralization in the North Miyako granitic body is relatively higher than that in the Okinoyama zoned pluton, which is related to the high chloride bearing fluid. Ishihara [23] suggested that magnetite-series granitoids in W metallogenic province belonged to a different original magma and tectonic setting which are characteristic of granitoids in Mo metallogenic province. However, it is emphasized that the W/Mo ratio of the magnetite-series granitoids may be controlled by factors such as the devolatilization depth related to the formation of ore metal complex.

Tungsten and molybdenum mineralization associated with magnetite-series granitic magmatism is affected mainly by magmatic evolution in the process of saturation of a granitic melt with magmatic fluid. Study of behavior of tungsten in granitic melt is important to understand its behavior as an ore metal during volatile exsolution and magmatic crystallization.

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