Article

K-Ar ages of adularia at the Koryu epithermal Au-Ag deposit, Hokkaido in Japan

Toru Shimizu^{1, *}

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Abstract: Four new K-Ar ages of adularia at the Koryu Au-Ag epithermal vein deposit along with the mineralization stages were determined. One age is 1.4 Ma for the first mineralization stage and the other three ages are 1.19-1.10 Ma for the most economically important Au-Ag mineralization stage. Combined with published data of K-Ar ages, mineral description, and fluid inclusion study, the results suggest that the hydrothermal activities waxed and waned, forming ore veins with ten mineralization stages for 0.55 million years in the Koryu deposit.

Keywords: K-Ar age, epithermal Au-Ag deposit, Koryu, mineralization stages, adularia, duration of mineralization

1. Introduction

The K-Ar and ${}^{40}\text{Ar}/{}^{39}\text{Ar}$ ages of adularia (KAlSi₃O₈) are useful to constrain the duration of mineralization and the timing of gold and silver precipitation in the epithermal Au-Ag vein deposits because adularia repeatedly occurs during vein formation and is commonly associated with gold and silver minerals (e.g., Sekine et al., 2002; Leavitt et al., 2004; Sanematsu et al., 2006; Hames et al., 2009). Several K-Ar age studies have been conducted on adularia and sericite at the Koryu Au-Ag epithermal deposit (Sugaki and Isobe, 1985; Shimizu and Matsueda, 1993; Fujikawa et al., 1995). They showed that the ages varied between 1.2 and 0.85 Ma among different veins. This paper reports four new K-Ar ages of adularia: one from the first mineralizing band and three from the most prominent gold- and silver-mineralizing band. Combined with the previous age data and mineralogical data (i.e., mineral paragenesis and zoning, Shimizu et al., 1998), this paper clarifies the timing of the most important Au-Ag precipitation and duration of vein mineralization at the Koryu deposit.

2. Ore deposit and mineralogy

The Koryu Au-Ag deposit occurs within alternating mudstone and siltstone of Miocene (Hasegawa *et al.*, 1987) and consists of eight major ore veins (Fig. 1). The veins strike approximately east-west and dip steeply. The deposit was discovered in 1899 and recently produced about 3000 tons of ore per year with a grade of 40 g Au/t (Shimizu *et al.*, 1998). The mine was exploited over a lateral extent of up to 1 km and to depths of 180 m until the mine was closed in 2006.

Mineralogy at major ore veins Nos. 1, 2 and 3 was intensively investigated by Sugaki *et al.* (1984), Shimizu and Matsueda (1993), Ono and Sato (1994) and Shimizu *et al.* (1998). These studies showed that the ore minerals occurred in black-colored gold and silverrich bands ("ginguro"), as massive black ore with clay minerals, as disseminations in quartz, and as euhedral crystals in quartz vugs. The common ore minerals are pyrite, chalcopyrite, sphalerite, galena, electrum, acanthite-aguilarite, polybasite-pearceite and pyrargyriteproustite. Gangue minerals mainly consist of quartz with lesser amounts of adularia, manganocalcite, johannsenite, smectite, sericite, interstratified chloritesmectite, vermiculite-biotite, and a kaolin mineral.

3. Mineral paragenesis, zoning and distribution of adularia

The mineral paragenesis, zoning and distribution of adularia in Nos. 2 and 3 veins were investigated in detail by Shimizu *et al.* (1998) and summarized as follows. Figure 2 shows a mineral paragenesis compiled from mineralogical data at Locs. A, B and D in Figure 1. The mineralization is divided into two epochs: the earlier and later based on the cross-cutting relationships between the veins. The earlier and later epochs are divided into three stages (E-I~III) and seven stages (L-I-VII), respectively. Each stage is further divided into several substages according to the mineral assemblage

¹AIST, Geological Survey of Japan, Institute of Geology and Geoinformation

^{*} Corresponding author: T. Shimizu, Central 7, 1-1-1 Higashi, Tsukuba, Ibaraki, 305-8567, Japan. E-mail: t.shimizu@aist.go.jp

and growth textures of minerals.

The mineral assemblage for the earlier mineralization stages is quartz, adularia, and manganocalcite with small amounts of ore minerals whereas that for the later mineralization stages is quartz, adularia, interstratified chlorite/smectite and large amounts of ore minerals. The earlier mineralization occurs only at No. 2 vein whereas the later occurs both at Nos. 2 and 3 veins: stages L-I to VII at No. 3 vein and stages L-III, VI and VII at No. 2 vein. The Au-Ag ore forming episodes are stages E-II and L-I, III, IV, and V. The stage L-III band is the largest and richest zone of Au-Ag mineralization; abundant gold and silver minerals (e.g., electrum) precipitated in the stage III band at bonanzas 1 and 2 (Fig. 1) where the vein is thickest (~2 m). The stage L-III band is seen in many outcrops between Nos. 2 and 3 veins.

Adularia is identified in a number of stages and substages: E-I, E-II-a, b, L-I-a, b, c, L-II, L-III-c, e, g, i, L-VI-a, and L-VII (Fig. 2). The mineral generally occurs as rhombic crystals in these stages except for columnar crystals in substage L-III-i. On an ore deposit scale, large amounts of gold and silver minerals are present in stage L-III band that contained abundant adularia. However, on a handspecimen to microscopic scale, gold and silver minerals coexist with clay minerals and quartz in the bands that alternately occur with adulariaquartz bands.

4. Sample descriptions

Four samples for K-Ar dating were collected from veins in different locations. One of the stage E-I (Fig. 3a) was from an outer band of the No. 2 vein that was approximately 5 mm in thickness adjacent to host rocks at Loc. A. The band was clearly older than the inner bands in the outcrop, hand specimen, and thin section. Ore minerals were not seen in the hand specimen but small amounts of pyrite, sphalerite and galena coexist with adularia and manganocalcite in thin sections (Shi-



Fig. 1 Exploration adits of Koryu mine (above) and generalized longitudinal projection onto a vertical plane of the Nos. 1, 2 and 3 veins (below). Modified after Shimizu *et al.* (1998). The characteristics (strike, dip, length, width) of each vein are as follows; No. 1 vein: N80-90°W, 65-85°S, 550 m, 0.3-1.5 m, No. 2 vein: N75-80°E, 70-80°N, 400 m, 0.1-0.4 m and No. 3 vein: N80°E-N80°W, 70-88°N, 450 m, 0.2-2.0 m. Locations (Loc.) A to F correspond to those for mineralogical, fluid inclusion and stable isotopic studies by Shimizu *et al.* (1998). Samples for K-Ar age determination in the present study were obtained from underlined locations A, H, I and J that correspond to those in Table 1. Dotted lines enclose the high Au-Ag grade zone. Oval shaded areas in No. 3 vein are bonanzas that were mineralogically studied in detail by Shimizu *et al.* (1998).

mizu *et al.*, 1998). Two of the stage L-III (Figs. 3b and c) were collected from inner portions of the No. 3 vein at Locs. H and I. One (Fig. 3d) was a float at Loc. J, No. 2 vein. In the latter three samples, an adularia band (\sim 1 cm in maximum thickness) commonly alternated with a ginguro band.

5. Procedure for K-Ar dating

Adularia was separated from the samples by handpicking after slight crushing and carefully checked for contamination by binocular microscope examination. In order to concentrate adularia and remove coexisting manganocalcite, the sample of the stage E-I was treated with hydrochloric acid (0.5N). All the samples were washed by distilled water and were dried in the oven at 110 °C. We prepared a few grams of each sample for K-Ar age determination. The measurements were carried out by Geological & Nuclear Sciences Ltd. and Mitsubishi Materials Co., Ltd. for stage E-I and L-III samples, respectively.

6. Results

Combined with previous data, the results of four K-Ar ages are listed in Table 1. The age of the stage E-I (1.4 ± 0.1 Ma) is the oldest and the three ages of the stage L-III (1.10 ± 0.12 , 1.19 ± 0.07 and 1.19 ± 0.08 Ma)

Stages		Earlier			Later																					
Ban Sube		E-I	E-I E-II E-III		i L-I			L-II	L-III							L-IV				L-V L		L-VI	L-VI			
	in may width (m		a¦b¦c	a¦b	a	b	c	d		a	b	c	d	e ¦ f	g	h	i	a	b	c¦ d	1	a¦b	а	b	с	
Minerals		5	7	25			59		30					100					4	1		32		79		21
	Quartz			H	_	1	1	1			1		*	*2	-			1				-				
Gangue minerals	Adularia	**1			*1	-	-	1 1 1		ſ		*2			_								-			*3
	Manganocalcite		╞┊╞																							
	Johannsenite		-																							
	Chlorite/smectite †										-			-		-										
	Smectite					_					1															
	Pyrite			-		_		 			-			-		-						-				
	Sphalerite		-			_		1 1 1			_							_				-				
	Chalcopyrite		-			-					_		_	-	-	_		_		_		-				
	Galena					_		1			_			-		_		_				-				
als	Electrum					_					_		_	_	-	_		_				-				
Ore minera	Acanthite -aguilarite					_		 			_					-		_								
	Polybasite -pearceite					-		 			-		-			_		_		_						
	Pyrargyrite -proustite										_					-						-				
	Mckinstryite					1	1	1 1 1																		
	Jalpaite							1 1 1 1			1															
	Hessite					_		1							1											
	Ag-Te-Se-S mineral					-		- - - - -																		
	Tetrahedrite					-					į.															

Fig. 2 Mineral paragenesis of the Koryu Au-Ag deposit after Shimizu *et al.* (1998). Mineralogical data for the stages E-I, II and III were from Loc. A in Figure 1. The data for the stages L-I to VII were from Locs. B and D in Figure 1. For the detailed difference of mineralogy among these locations, see Shimizu *et al.* (1998). The width of vertical column for stages reflects the relative ratio of maximum band width for stages in the veins. The width of column for the stages with the band width less than 50mm is equally displayed (stages E-I, II and III, L-II, V and VII). The dotted boundary in the column denotes the substage boundary. The stage (or substage) for K-Ar age dating: *1 = 1.2±0.7 Ma, *2 = 1.19±0.09 Ma, *3 = 0.85±0.13 Ma (Shimizu and Matsueda, 1993), **1 = 1.4±0.1 Ma(This study), **2 = 1.10±0.12, 1.19±0.07 and 1.19±0.08 Ma that were obtained from unidentified substages in stage III (This study). † Interstratified minerals.

among different locations are concordant within error of analysis.

7. Discussion

Newly determined four ages and previously reported three ages along with the sequence of mineralization stage are between 1.4 and 0.85 Ma (Fig. 4). This range contains other ages from unidentified stages: 1.0 ± 0.3 Ma for adularia at 60 mL+20 mL sublevel, No. 1 vein (Sugaki and Isobe, 1985) and 1.09 ± 0.20 Ma from sericite at No. 8 vein (Fujikawa *et al.*, 1995). The data (Fig. 4) indicate that earlier mineralization started around 1.4 Ma, forming stage E-I to III bands of No. 2 vein. This was followed by later mineralization (1.2-0.85 Ma), forming stage L-I to VII bands at No. 3 veins and stage L-III, VI and VII bands at No. 2 veins. The main gold and silver mineralization (L-III) occurred from 1.19 to 1.10 Ma.

Combined with the fluid inclusion study for different mineralization stages (Shimizu et al., 1998), the K-Ar ages along with the mineralization stages suggest that episodic hydrothermal activities waxed and waned, forming ten mineralization stages (E-I~III and L-I~VII) for 0.55 million years, during which time the temperature of the hydrothermal fluids was as high as 283 °C in stage E-III, but mainly around 250 °C in most stages except for stage L-VII (as low as 206 °C). Because the formation of adularia in the last stage L-VII was followed by that of quartz within the same stage (Fig. 2), the obtained age of stage L-VII adularia did not represent the ending of the mineralizing events. However, judging from the restricted distribution of the stage VII band in the center of the vein at two bonanzas in No. 3 vein (Fig. 3 in Shimizu et al., 1998) and lowest formation temperature (206 °C) among the ten mineralization stages above, it is suggested that hydrothermal activi-



Fig. 3 Modes of occurrence of adularia for K-Ar dating in hand specimen scale at the Koryu Au-Ag deposit. A white arrow in each picture indicates the growth direction of minerals. Locs. A, H, I and J correspond to those in Figure 1. a. Sample No. Sm-940126-3 (a polished slab) from Loc. A. Adularia occurs as fine-grained crystals (<0.5 mm) within a narrow band (stage E-I) containing many fragments of mudstone. b. Sample No. Sm-900711-10 from Loc. H. Adularia with subhedral rhombic shapes occurs after ginguro and clay. The K-Ar dating was conducted for adularia on top. c. Sample No. Sm-910729-3 from Loc. I. Adularia with subhedral rhombic shape intermittently occurs with ginguro and clay. d. Sample No. Sm-9501207-1 (a polished slab) from Loc. J. Adularia band consisting of euhedral-subhedral rhombic shapes is followed by a gingruo band.</p>

ties responsible for major mineralizations at the Koryu deposit ceased by 0.85 Ma.

The similar durations of mineralization from two other epithermal Au-Ag deposits were reported: 0.64 million years at the Hishikari deposit (Sekine *et al.*, 2002; Sanematsu *et al.*, 2005; Sanematsu *et al.*, 2006; Tohma *et al.*, 2010) and 0.53 million years at the Cibaliung deposit, Indonesia (Harijoko *et al.*, 2004).

Stage	Locations	Sample No.	Material (XRD analyzed)	K wt%	Rad ⁴⁰ Ar (scc/gm x 10 ⁻⁵)	Rad ⁴⁰ Ar %	Age (Ma) (Uncertainty: 1σ)	References
E-I	No.2 vein, Loc. A	Sm-940126-3	Adularia, quartz	5.98 5.98	0.030 0.032	22 33	1.4±0.1	b
L-I-a	No.3 vein, Loc. D	Sm-900709-6	Quartz>> adularia	0.3 0.3	0.0013 0.0014	6.1 4.7	1.2±0.7	а
L-III-c	No.3 vein, Loc. G	Sm-900712-19-6	Adularia	12.3 12.3	0.058 0.056	34.4 30.4	1.19±0.09	а
L-III	No.3 vein, Loc. H	Sm-900711-10	Adularia	10.7 10.7	0.047 0.044	16.5 16.0	1.10±0.12	b
L-III	No.3 vein, Loc. I	Sm-910729-3	Adularia	12.5 12.6	0.057 0.061	30.6 24.4	1.19±0.07	b
L-III	No.2 vein, Loc. J	Sm-9501207-1 †	Adularia	9.64 9.61	0.045 0.044	27.6 23.4	1.19±0.08	b
L-VII	No.3 vein, Loc. D	Sm-900404-A-1	Adularia, quartz	1.07±0.03 1.07±0.03	0.004 0.003	49.4 48.2	0.85±0.13*	а

Table. 1 K-Ar ages of adularia from Koryu Au-Ag deposit.

Locations correspond to those in Figure 1. References: a. Shimizu and Matsueda (1993), b. This study. †: float. *: An average value of 0.91 ± 0.19 Ma and 0.81 ± 0.17 Ma was obtained by duplicate measurements (Shimizu and Matsueda, 1993). Constants used for the age calculation are; $\lambda\beta = 4.962 \times 10^{-10}$ /yr, $\lambda\epsilon = 0.581 \times 10^{-10}$ /yr and 40K/K = 1.167 x 10⁻² (atom. %).



Fig. 4 The K-Ar ages of the Koryu Au-Ag deposit. The error bars extend one standard deviation. Shaded area denotes the timing of the main Au-Ag mineralization stage (L-III). References: (a) Shimizu and Matsueda (1993), (b) This study.

8. Conclusions

1. The K-Ar ages of adularia along with the mineral paragenesis showed that the earlier mineralization initiated at 1.4 Ma, forming stage E-I to III bands mainly composed of quartz, adularia, manganocalcite and small amounts of ore minerals. The mineralization was followed by a later mineralization, forming stages L-I to VII bands with quartz, adularia, interstratified clay minerals and large amounts of ore minerals between 1.2 and 0.85 Ma. The stage III band, the most economically important gold and silver mineralization zone, occurred in an interval of 1.19-1.10 Ma.

2. The duration of major mineralizing events at Koryu hydrothermal system is estimated to be 0.55 million years.

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北海道光竜浅熱水性金銀鉱床の氷長石の K-Ar 年代

清水 徹

要 旨

本研究では、北海道光竜浅熱水性金銀鉱床で産出した氷長石に関し、新たに四つの K-Ar 年代値を得た. それらの値 を鉱脈の鉱化ステージ区分に沿って報告する. 四つの年代値のうち一つは 1.4 Ma であり、最も初期の鉱化ステージから 得られた値である. 一方、他の三つは 1.19 から 1.10 Ma の範囲であり、最も主要な金銀鉱化ステージから得られた値で ある. 光竜鉱床では、熱水活動の盛衰とともに、十個の鉱化ステージからなる鉱脈が 55 万年間にわたって形成された.