Silver Mineralization of the Karuizawa Mine, Fukushima Prefecture, Japan

Asahiko Sugaki*, Arashi Kitakaze* and Kiyoshi Isobe**

Abstract: Silver deposits of the Karuizawa mine are network and vein types developed in fracture zone of Miocene rhyolite lava dome altered hydrothermally. The network ore body consists of veinlets along fracture and spots of ore filled up cavity after spherulite in rhyolite. Its ore is principally composed of barite, sphalerite and galena in association with small amounts of quartz, pyrite, marcasite, chalcopyrite, polybasite, pyrargyrite and argentian tetrahedrite (12 to 18 wt% Ag). On the other hand, the vein along fissure in silicified rhyolite consists mainly of galena, sphalerite, pyrite, marcasite and barite associated with quartz, chalcopyrite and small amounts of bournonite, argentian tetrahedrite, polybasite and pyrargyrite. These mineral assemblages in ores are similar to those of kuroko ore. In high silver ore, fine grained silver minerals such as polybasite, pyrargyrite and argentian tetrahedrite are usually found as inclusion in galena, and often show a pseudo-micrographic texture with galena. These silver minerals were crystallized at late stage of the mineralization as shown in Fig. 9. Homogenization temperature and salinity of fluid inclusion in barite are 230° to 320° C and 3.6 to 5.0 wt% NaCl equivalent, respectively. Hydrothermal alterations of silicification and adularization are conspicuously observed in or around the ore deposit. K-Ar age for adularia of the alteration product is 12.8±0.6 Ma which is approximately same as that of alteration rock of the kuroko deposits. The ore deposits of the Karuizawa mine are thought to have been produced by mineralization in the relation to formation of the kuroko deposits.

Introduction

The Karuizawa mine is situated at Yanaizu-town, Kawanuma-gun, Fukushima Prefecture in northeastern Japan (Fig. 1). It is located at about 15 km west of Aizuwakamatsu city and about 8 km south of the Aizuyanaizu railway station of Tadami line. It is an old mine discovered in the 16th century, and has been mined as famous silver mine in Japan from 1886 to 1895. Its production at that time was about 1,800 ton per month as crude ore and 300 kg per month in silver metal. After 1895, mining or exploration on the ore deposits of the mine has been carried out intermittently up to the present. In 1984, production of ore from the mine was about 700 tons per month as crude ore with about 200 g/t in silver. Very high grade silver ore contained 5 to 12 kg/t Ag sometimes occurs from the mine, but silver minerals in such ore have been not identified although argentite was described as only one of silver mineral by some authors (Harada, 1886; Watanabe, 1950: Nambu, 1969, Isobe et al., 1984). Thus, the present authors have made study to identify silver minerals in ore from the mine and make clear occurrence, mineral assemblage and paragenesis, and mineralization stage. From this study, ex-
istence of silver minerals such as polybasite, pyrargyrite and argentian tetrahedrite has been confirmed in the ore from the mine. However no argentite reported by former authors has been found. In this paper, their results are described below.

Geology

Geology around the Karuizawa mine consists of the Shiono and Ohyaji formations of Miocene age and the Izumi formation of Pleistocene age (KITAMURA et al., 1968). Lower part of the Shiono formation named as the Kamidaira tuff member is mainly composed of massive rhyolitic tuff, tuff breccia, sandy tuff, sandstone and mudstone. Upper part of the Shiono formation called as the Ginzan rhyolitic tuff member principally consists of the Ginzan rhyolite and its pyroclastic rocks. The Ginzan rhyolite occurs as lava dome in the member. It is usually a massive and compact rock, but sometimes shows distinctly flow structure. Also it occasionally has a lot of spherulite. Ore deposits of the Karuizawa mine occur in the Ginzan rhyolite. At the southern portion of the mine, andesite lava and its agglomerate belonging to the Ohyaji formation lie unconformably on the Ginzan rhyolitic tuff member of the Shiono formation. Meanwhile, at the northern portion of the mine, dacite lava, its welded tuff and tuff breccia of the Izumi formation covers unconformably on the Kamidaira tuff member of the Shiono formation.

Ore Deposits

Ore deposits of the Karuizawa mine are two types of hydrothermal network and vein developing in the Ginzan rhyolite (HARADA, 1886; WATANABE, 1950; ISobe et al., 1984). Ores from them are principally composed of barite, galena, sphalerite, pyrite, marcasite and quartz, accompanied by small amounts of chalcopyrite, bournonite, silver minerals such as polybasite, argentian tetrahedrite and pyrargyrite, and kaoline. There are two ore deposits, Bonten and Ginsei, in the mine (Fig. 2).

Bonten deposit: Bonten deposit which is main

ore bodies in the mine has been mined at the Tsudo (0 m), Chugiri (+12 m), +30 m, +40 m, +60 m and +90 m levels. It is composed mainly of the stock work and vein developed in the fracture zone, 350 m in length, 120 m in width and 250 m in depth, at the southwestern portion of the Ginzan rhyolite altered hydrothermally. The ore body, 100 m long, 40 m wide and 60 m deep in scale, showing network develops in the northwestern part of mineralization zone as shown in Figs. 3 and 4. Its ore found as a lot of veinlets along fracture and many spot like forms filled up cavities after spherulite in the rhyolite consists mainly of fine grained aggregate of barite, sphalerite and galena in association with small amounts of quartz, pyrite, marcasite, chalcopyrite and silver minerals such as polybasite and argentian tetrahedrite (Fig. 5-A). It commonly contains 200 to 500 g/t in silver. Rhyolite of country rock is distinctly altered by silicification and adularization.

On the other hand, the veins, 5 to 40 cm in width, along fissures in silicified rhyolite run in general to N60°E to N60°W direction dipping 20° to 45°N as shown in Fig. 4. They contain
commonly 200 to 500 g/t, and sometimes 6,000 to 12,000 g/t in silver, and are principally composed of coarse grained aggregate of galena, sphalerite, pyrite, marcasite and barite in intimate association with quartz and chalcopyrite, and small amounts of bourronite, argentian tetrahedrite, polybasite and pyrargyrite. In general, the outer side of the vein consists almost of barite, 1 to 7 cm in width, while the inner side is composed mainly of sulfide minerals such as galena, sphalerite, pyrite and marcasite with small quantities of barite and quartz (Fig. 5-B). In high silver ore, fine grained silver minerals such as polybasite, pyrargyrite and argentian tetrahedrite are usually found as inclusions in galena microscopically. Colloform of marcasite, galena and sphalerite is commonly observed in the ore. The ore vein described above usually occurs in the silicification zone of rhyolite.

**Ginsei deposit**: It is situated at about 400 m east of the Bonten deposit and also occurs in the Ginzan rhyolite. It mainly consists of a ore body of network containing 200 g/t Ag, and has a scale of 50 m in length, 50 m in width and 50 m in depth. Ore minerals in veinlet of the network ore body are principally sphalerite, galena, pyrite and marcasite in association with small amounts of silver minerals such as polybasite, pyrargyrite and argentian tetrahedrite. Barite and quartz as principal gangue minerals are found in intimate association of ore minerals. Colloform banding of marcasite, galena and sphalerite is commonly observed in the veinlet of the network. Now this deposit is closed.

**Rock alteration**: Ginzan rhyolite usually is a
compact and massive rock, but partly shows flow structure having strike from N50°E to N10°W, and sometimes has a lot of spherulite. Around the Karuizawa mine, it is strongly affected by hydrothermal alteration such as silicification and adularization. Especially at the Bonten ore body, zoning of alteration is observed. That is, a central portion of alteration is characterized by silicification, and its margin becomes to the adularization zone. In weakly altered rhyolite, phenocrysts of quartz, plagioclase and biotite, 0.2 to 1.5 mm in size, are found in groundmass consisting of felsic minerals and glass. But biotite changes completely to chlorite.

Rhyolite silicified shows gray in color, and becomes to more compact and hard rock. Phenocrysts except quartz, and felsic minerals and glass in groundmass are replaced by fine grained aggregate of quartz, 0.01 to 0.05 mm in size. But, quartz phenocryst remains. Also spherulite, 1 to 5 mm in size, usually dissolves and then its cavity appears. It is sometimes filled up by fine grained aggregate of barite, galena, sphalerite and marcasite. The ore veins mainly develop in the silicification zone.

In the adularization zone, rhyolite becomes grayish white, white or yellowish white, and soft and slightly porous. Its phenocrysts of plagioclase and biotite, and felsic minerals in groundmass are almost replaced by fibrous or radial aggregate of adularia, 0.01 to 0.1 mm in length, but quartz phenocryst is found as relic. Spherulite also changes to fibrous aggregate of adularia, 0.1 to 1 mm long.

**Occurrence of Ore Minerals**

As ore minerals, galena, sphalerite, pyrite and marcasite associated with small amounts of chalcopyrite, bournonite, polybasite, argentian tetrahedrite and pyrargyrite are commonly found in ores from the network ore bodies and veins of the Bonten deposit. Also, barite, quartz and a small amount of kaoline as gangue minerals usually appear in close assemblage with ore minerals as above.

**Polybasite**: Polybasite is found microscopically as very fine grained crystal, less than 20 µm in size, enclosed in galena in the vein. It shows the pseudo-micrographic texture with galena, and sometimes appears as small grain, less than 10 µm in size, in a growth band of galena in closely association with argentian tetrahedrite, pyrargyrite, and bournonite (Fig. 6-A and B). Rarely, it occurs as aggregate of granular crystal, 0.1 to 0.3 mm in size, assembled with galena, sphalerite, marcasite, pyrite and chalcopyrite. In ore of the network ore body, polybasite is found as irregular forms, 0.2 to 0.8 mm in size, in association with galena, chalcopyrite, argentian tetrahedrite and sphalerite filled up to interspace or
Fig. 6 Microphotographs of ore minerals from the Karuizawa mine.

A: Pseudo-micrographic intergrowth of galena (gn) and polybasite (pl) in ore from the vein of the Bonten deposit. 
B: Pyrargyrite (pr) enclosed in growth band of galena (gn) crystal from the vein of the Bonten deposit. 
C: Granular crystals of bournonite (bn) in galena (gn) in association with argentian tetrahedrite (tt), polybasite (pl) and pyrargyrite (pr) in ore from the vein of the Bonten deposit. 
D: Colloform bands of sphalerite (sp), marcasite (mc) and galena (gn) in the network ore of the Gissei deposit.

Pyrargyrite: Occurrence of pyrargyrite in ore from the vein is similar to that of polybasite showing a pseudo-micrographic texture in galena in association with polybasite, pyrargyrite and bournonite (Fig. 6-B). In network ore, very small amounts of pyrargyrite appears as irregular forms assembled with chalcopyrite, polybasite, galena and marcasite.

Argentian tetrahedrite: Argentian tetrahedrite in ore from the vein is generally found under ore microscope as a very fine crystal, 2 to 20 μm in size, assembled with polybasite, pyrargyrite and sometimes bournonite as inclusion in galena. In this case, it shows a pseudo-micrographic texture with galena as same as that of polybasite. Occasionally, argentian tetrahedrite appears as relatively coarse grained crystal, 0.2 to 0.3 mm in size, associated with galena, polybasite, pyrargyrite, sphalerite and sometimes bournonite (Fig. 6-C). In ore from the network ore body, argentian tetrahedrite appears as irregular forms, 0.2 to 0.5 mm in size, filled up to interstice of crack of barite and marcasite in veinlet and spotted ores.
marcasite, quartz or barite in assemblage with galena, pyrite, chalcopyrite and polybasite.

**Galena:** Galena is a most principal ore mineral from the vein and network ore body. It occurs as band as aggregate of granular crystals, 1 to 2 mm in size, in association with sphalerite, pyrite, marcasite and barite in the inner side of the vein, and sometimes fills up interstice or crack of barite. Silver rich ore from the vein commonly consists of massive aggregate of fine grained galena, 0.1 to 0.5 mm in size, associated with sphalerite, marcasite, pyrite and quartz, and galena encloses fine grained crystal of sulfosalt minerals such as polybasite, argentian tetrahedrite, pyrargyrite and bournonite microscopically as shown in Fig. 6-A, B and C. Meanwhile in ore from the network ore body, galena usually appears as colloform band with marcasite and sphalerite as seen in Fig. 6-D. It fills up many cavities after dissolution of spherulite in altered rhyolite in association with barite, sphalerite and pyrite. **Sphalerite:** Sphalerite also is a principal ore mineral same as galena, and occurs as aggregate of granular form, 1 to 2 mm in size, in close association with galena, pyrite, marcasite, barite and quartz in the vein. Under microscope, it is sometimes accompanied with fine grained aggregate of bournonite, chalcopyrite, polybasite and pyrargyrite. While, sphalerite in ore from the network ore body commonly shows a colloform banding texture with marcasite and galena. Under euhedral crystal, 0.5 to 1.0 mm in size, in the druse of quartz or barite together with galena in central portion of the vein and sometimes in small cavity after spherulite. Sphalerite contains no chalcopyrite dots.

**Marcasite:** Marcasite occurs in the vein and network. In the vein, it is aggregate of irregular forms, 0.5 to 1.0 mm in size, associated with galena, sphalerite, pyrite and barite, and small amounts of bournonite, pyrargyrite, polybasite and argentian tetrahedrite. Marcasite is also found as aggregate of granular form, 0.5 to 0.6 mm in size, in the network ore. In the case, interspace and crack of marcasite aggregate are filled up by galena, sphalerite, pyrite, and sometimes argentian tetrahedrite, pyrargyrite or polybasite. It commonly shows colloform banding, up to 2 mm in width, with galena and sphalerite (Fig. 6-D).

**Pyrite:** Pyrite appears as euhedral or subhedral forms, 0.1 to 0.5 mm in size, accompanied by galena, sphalerite, marcasite, barite and quartz in the vein. It is also enclosed in galena and sphalerite as euhedral crystals, 0.01 to 0.05 mm in size. It rarely shows a colloform texture with marcasite and sphalerite. In ore from the network, pyrite is found as fine grained crystal, 0.01 to 0.03 mm in size, in association with marcasite, galena and sphalerite, and sometimes chalcopyrite, polybasite and bournonite in the veinlet.

**Chalcopyrite:** Small amounts of chalcopyrite is recognized microscopically as irregular forms, 0.02 to 0.05 mm in size, accompanied by argentian tetrahedrite, polybasite, pyrite, galena, marcasite and sphalerite in the vein and veinlet of the network ore body.

**Bournonite:** Bournonite appears only microscopically in very small amounts as inclusion, 0.03 to 0.1 mm in size, in galena in assemblage with pyrargyrite, polybasite and argentian tetrahedrite (Fig. 6-C) and sometimes marcasite, sphalerite, pyrite and chalcopyrite.

**Gangue minerals:** As gangue minerals, barite, quartz and small amounts of kaoline occur in veinlet of the network and vein in association with ore minerals as described above.

Barite which is a most principal mineral from the mine is usually found in the outer side of the vein as aggregate of euhedral crystals, 2 to 5 mm in size, in assemblage with galena, sphalerite and marcasite. In the network ore body, it also occurs as platy crystal of euhedral form, 0.5 to 2 mm in size, in close association with quartz, galena, sphalerite and pyrite in veinlet.

Quartz commonly occurs as aggregate of granular crystals, 0.2 to 1 mm in size, together with galena, sphalerite, marcasite, and pyrite in central portion of the vein. It is also found as aggregate of fine grained form, 0.2 to 0.5 mm in size, in association with galena, sphalerite, marcasite and chalcopyrite in veinlet of the network. The amounts of quartz are fairly less than those of barite.
A small amount of kaoline appears as veinlet, 1 to 2 cm wide, cutting barite and galena in the vein. There is frequently found druse structure of barite and quartz in central portion of the vein. In the druse, euhedral crystals of galena and sphalerite are found. Galena from the druse has no silver minerals.

Silver Minerals

Optical properties and chemical compositions of the silver sulfosalt minerals from the Karuizawa mine are described below. No their X-ray powder diffraction data have been obtained because grain size of silver minerals is very fine and their quantity is very slight. 

**Polybasite:** Polybasite shows distinct pleochroism from grayish white with bluish green tint to grayish white with purplish tint in color, and strong anisotropism changing its interference color from dark brown to yellowish gray.

Chemical compositions for polybasite from the Karuizawa mine obtained by an electron probe microanalyser (EPMA) are shown in Table 1. It contains some amounts of copper from 5.2 to 8.9 wt% (6.3 to 10.8 at%) and arsenic from 0.1 to 5.6 wt% (0.1 to 5.4 at%) in addition to silver, antimony and sulfur. Accordingly its composition shows to a wide range from 30 to 95% Sb/(As+Sb) in atomic%. Relation between Cu/(Ag+Cu) and Sb/(As+Sb) in atomic% is shown in Fig. 7. As seen in this figure most of the analytical data are plotted in compositional area of the polybasite field (HALL, 1967), but some of them are in that of the pearceite area. The wide variation of polybasite composition is thought to be caused by difference of mineral assemblage and mineralization, but it is not sure. No compositional zoning of polybasite is observed in the crystal.

**Pyrargyrite:** Composition of pyrargyrite is given in Table 2. It contains some amounts of arsenic and copper besides silver, antimony and sulfur, but amounts of arsenic and copper are only less than 1.1 wt% (1.1 at%) and 0.5 wt% (0.6 at%), respectively. It has from 3 to 7% as As/(As+Sb) in atomic %, corresponding to proustite molecule (Ag₃AsS₃).

**Argentian tetrahedrite:** Analytical data of argentian tetrahedrite are shown in Table 3. Its tennantite composition as As/(As+Sb) are from 7 to 26%. Tetrahedrite contains from 12 to 18 wt% (7.0 to 10.3 at%) Ag, from 4.0 to 5.7 wt% (3.7 to 5.2 at%) Zn and from 0.4 to 1.2 wt% (0.4 to 1.3 at%) Fe. Its Fe/(Fe+Zn) is 4 to 16% in atomic %, thus argentian tetrahedrite from the mine corresponds to zinc rich one.

**Fluid Inclusion**

Homogenization temperature and salinity of fluid inclusions in barite from the vein composed mainly of barite, galena and sphalerite at the Tsudo and +30 m levels have been measured by using Linkam TH-600 heating and cooling stage. Inclusions measured are two phases, vapor and liquid, and 20 to 30 μm in size. Histogram for homogenization
Table 2 Analytical data of pyrargyrite from the vein in the Bonten deposit of the Karuizawa mine.

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Table 3 Chemical composition of argentian tetrahedrite from the network ore and vein in the Bonten deposit of the Karuizawa mine.

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Discussion and Conclusion

The Karuizawa deposits are network and vein types developed in the Ginzan rhyolite. The network ore bodies and veins of the deposit, are frequently high in silver, but very low in gold, usually less than 0.1 g/t. The ores mainly consist of galena, sphalerite, marcasite, barite and quartz with small amounts of silver minerals such as polybasite, pyrargyrite and argentian tetrahedrite, and bournonite. The silver minerals usually occurs as minute inclusions in galena and sometimes as fine grained aggregate in association with galena, sphalerite, marcasite, pyrite, chalcopyrite and bournonite in veinlet of the network and the vein. They usually show a pseudo-micrographic texture with galena microscopically.

Watanabe (1950) and Isobe et al. (1984) reported occurrence of argentite as only one of silver mineral from the mine, but in the present study there has been found no argentite in the ores from the network and vein of the mine. Existence of polybasite, pyrargyrite and argentian tetrahedrite as silver minerals is recognized for the first time by this study. However, no electrum occurs.

Polybasite is a principal silver mineral from the mine and appears usually as inclusion in galena in association with pyrargyrite, argentian tetrahedrite and bournonite. The chemical composition of polybasite has a wide range from 30 to 95 Sb/(Sb + As) in atomic%, and is principally plotted in the area of polybasite-arsenopolybasite series (Hall, 1967) as shown in Fig. 7.

Although argentian tetrahedrite from the epithermal gold-silver veins is in general iron rich (Sugaki et al., 1982, 1984), tetrahedrite from the mine is zinc rich (4.0 to 5.7 wt%) in comparison with its iron content (less than 1.2 wt%). It is similar to that from the kuroko deposits.

The mineralization sequences of ore and gangue minerals from the mine is inferred
from the data on occurrence of minerals, mineral assemblage and ore texture in ores from the network and vein as shown in Fig. 9. As seen in the figure, barite was continuously crystallized from earliest to middle or late stages of the mineralization. Sphalerite, pyrite, galena and quartz were formed at relatively early stage of the mineralization together with barite. Meanwhile galena was also continuously produced from early to late stages, but its crystallization was conspicuously performed at middle stage. Marcasite and chalcopyrite were formed at middle stage of the mineralization after pyrite. The silver minerals such as polybasite, pyrargyrite and argentian tetrahedrite, and bournonite were crystallized during late stage mineralization together with galena of fine grained aggregate. Small amounts of sphalerite, marcasite, barite and quartz were also accompanied by silver minerals at late stage.

Homogenization temperature and salinity of fluid inclusions in barite range from 200° to 380°C (mostly from 230° to 320°C) and from 2.6 to 5.0 wt% NaCl equivalent, respectively. Both the values are slightly higher than those (200° to 260°C and 0 to 2 wt%) from epithermal gold-silver quartz veins reported by Enjoji and Takenouchi (1976), Takenouchi (1981) and Sugaki et al., (1984) etc.

K–Ar age for adularia as hydrothermal alteration product of rhyolite at the Chugiri level is 12.8 ± 0.6 Ma (Sugaki et al., in preparation). This age is approximately same as the K–Ar age, 10.7 to 15.6 Ma (MITI, 1980, 1981) for hydrothermally altered rock of the kuroko deposits. Mineral assemblages of sphalerite, galena and barite in the ore from the mine are also similar to those of kuroko ore. Thus, the ore deposits of the Karuizawa mine are thought to have been produced by mineralization in the relation to formation of the kuroko deposits, but at shallow depth near sea floor of middle Miocene.

Acknowledgment: The authors are deeply indebted to staffs of the Karuizawa mine for their kind cooperation during our field survey. A part of expense for this study was defrayed by the Grant-in-Aid for Scientific Research from the Ministry of Education of Japan. The authors offer their sincere thanks to the Ministry of Education.

References


福島県軽井沢山の銀鉱化作用

冨木浅彦・北風 蠼・磯部 清

要旨：軽井沢山は福島県河沼郡柳津町軽井沢にあり、会津若松市の南西方約17 kmに位置する。本鉱床は新第三紀中新世の銀山流紋岩中に半晶する網状および脈状鉱体で梵天および銀盛柄鉱床があり、金に乏しい銀鉱、亜鉛鉱を産する。いずれも流紋岩の破砕帯あるいは裂か帯に発達した脈状鉱脈および鉱脈で、重晶石、黒鉛鉱、方鉛鉱の脈状鉱脈を主とし、これに少量の黄鉛鉱、白鉛鉱、黄銅鉱、亜鉛鉱および銀鉱物を伴う。銀鉱物として含銀基固面銅鉱(12～18 wt% Ag)、青銅鉱および淡红色銀鉱がみられる。これらはいずれも方鉛鉱中の包有物として見られ、方鉛鉱と擬文象状組織を呈するが、とき方鉛鉱、黒亜鉛鉱、白亜鉛鉱、黄鉛鉱、黄銅鉱、重晶石、石英と組み合う細粒集合として産する。

上記の鉱物組合せは黒鉱とそれによく類似し、銀鉱物もまた黑鉱中のものと同一種である。母岩は結晶および氷長化石作用を受けて、亜鉱の大部分は結晶帯、一部氷長石帯にみられる。熱水変質帯の氷長石について得たK-Ar年代値は12.8±0.6 Maで黒鉱鉱床変質母岩のそれとは近い。以上本研究によって得た資料は本鉱床が黒鉱生成に関係ある鉱化作用によって生じた可能性を示唆している。

文中的地名・固有名詞の漢字による表記
Bonten 梵天, Chugiri 中切, Ginzai 銀盛, Ginzan 銀山, Izumi 和泉, Kamidaira 上平, Karuizawa 軽井沢, Kawanuma 河沼, Nijusshaku 二十尺, Ohyaji 大谷地, Shiono 塩野, Tsudo 通洞, Yanaizu 柳津