SUBMILLIMETER SCALE VARIATION OF THE OXYGEN ISOTOPE COMPOSITION OF GOLD-BEARING QUARTZ VEIN AT THE HISHIKARI DEPOSIT, JAPAN

Ken-ichiro Hayashi

Department of Mineralogy, Petrology, and Economic Geology,
Graduate School of Science, Tohoku University,
Aoba-ku, Sendai 980-8578, Japan

Low-sulfidation style ore deposits, the major source of Au, Ag, and Hg, are formed from neutral-pH, reduced hydrothermal solutions close to equilibrium with their host rocks. The waters are low-salinity (<1 wt % NaCl equiv.) but relatively gas rich (1-2 wt % CO_2), and are largely meteoric water. However, the contribution of magmatic components to the epithermal system, its temporal importance, and its relation to the source of ore metals are largely controversial. This paper describes a submillimeter scale $\delta^{18}O$(SMOW) variation of vein quartz from the Hosen-1 and Ryosen-6 veins at the Hishikari deposit, southern Japan, obtained by in situ laser ablation technique (Hayashi et al., 2000, 2001).

The Hishikari deposit in the southern Kyushu island of Japan is the most productive Au mine in Japanese mining history. Reserves including ore produced are estimated to be 5.2 Mt at an average grade of ~40 g/t Au; a total of at least 250 t of Au was precipitated in the 750 m wide vein system, which extends over 2 km along strike. It is a typical low-sulfidation style Au deposit, composed of several quartz-adularia veins hosted in basement metasedimentary rocks of the Cretaceous Shimanto Supergroup and overlying Quaternary andesitic volcanic rocks. K-Ar dating of adularia in ore indicates an early Pleistocene age of 1.25 to 0.6 Ma for Au mineralization.

The Hosen-1 vein consists of 8 different zones, which can be distinguished by the morphological features of quartz and adularia and are arranged symmetrically with respect to the center of vein. No cross-cutting relation is observed in the vein, and these 8 zones correspond to the temporal growth sequence, which began at the contact with wall rock and terminated at the center of the vein. Each zone can be divided into several minor bands.

In Figure 1, mineralogical and isotopic features of the Hosen-1 vein including morphological characteristics of quartz, the quartz/adularia ratio, abundance of ore minerals, grade of Au and Ag, and $\delta^{18}O$ of quartz are shown. The vein is composed of alternating crustified bands of clear quartz, milky quartz, quartz-adularia mixture, and lesser amounts of smectitic clay. The thickness of individual band varies from < 1 to ~10 mm. The quartz-adularia bands consist of fine-grained quartz and adularia. The quartz bands include relatively coarse euhedral quartz crystals which grew towards the center of the vein. So called 'bladed quartz', a radial aggregate of acicular quartz, which precipitated under a rapid nucleation of quartz during fluid boiling (Simmons and Christenson, 1994) is partly included in the period IV vein.

The overall range in $\delta^{18}O$ values obtained for the Hosen-1 vein quartz is +5.2 to +12.2 per mil, and $\delta^{18}O$ characteristics within an individual zone are variable. The largest variation is observed in periods IV, VI and VIII, while period VII which is

composed of fine grained milky quartz has a narrow $\delta^{18}O$ range (<1.5 ‰). Quartz in period IV changes its morphological feature frequently, which suggests rapid change of environment of mineralization. The highest grade parts of the vein also correspond to zones IV and V (131.1 ppm Au and 24.9 ppm Ag), where largest amount of electrum as well as other sulfide minerals have precipitated.
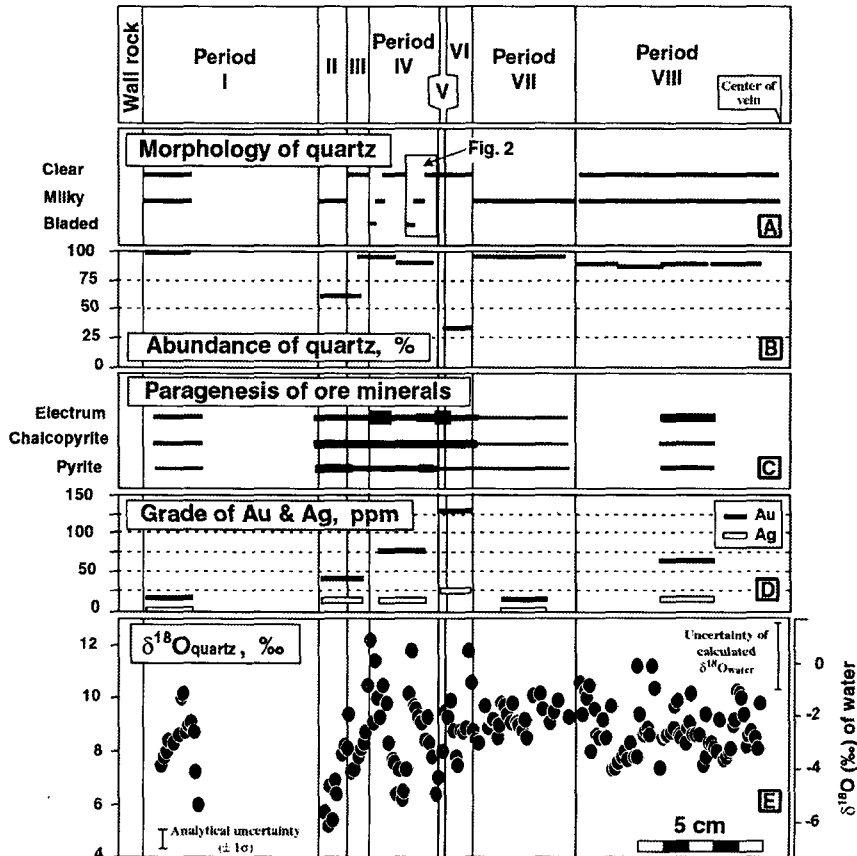

Fig. 1. Sequential mineralogical and oxygen isotopic change of the gold-bearing quartz-adularia vein, the Hosen-1 vein at the Hishikari mine, plotted on a relative time scale recorded by the crustified bands. (A) Color and textural appearance of quartz. (B) Molal abundance of quartz in quartz-adularia multiple bands. (C) Semi-quantitative estimation of relative abundance of ore minerals. (D) Concentrations of Au and Ag. (E) Oxygen isotopic compositions of quartz determined by *in situ* laser ablation technique. The $\delta^{18}O_{water}$ is calculated using a fractionation factor between water and quartz at T= 206.1°C. Since samples smashed into small pieces, any systematic data was not obtained from the later part of period I because of the paucity of temporal information.

The $\delta^{18}O$ of vein quartz is principally controlled by the $\delta^{18}O$ value of the hydrothermal solution which was responsible for the precipitation of quartz and the temperature of crystallization. The best estimation of temperature during the precipitation of vein minerals is from homogenization temperature (Th) of fluid inclusions. All fluid inclusions observed are simple two-phase type, small bubble containing liquid inclusion. Fluid inclusions large enough to measure Th were found in

quartz and adularia of period I, IV, VI, and VIII. However, because of the restriction of size of host minerals, detailed temperature excursion during the growth of vein is difficult to reconstruct. The whole range of Th varies from 170.4° to 250.8°C. Any pressure correction on Th was not applied, because depth of vein formation at the Hishikari deposit was shallow (Izawa et al., 1990). The $\delta^{18}O$ values of hydrothermal solution were calculated from the fractionation factor of oxygen isotopes between quartz and water (Matsuhisa et al., 1979), and $\delta^{18}O$ values of quartz at 206.1°C, the average of all the Th data (n = 50). The calculated range of $\delta^{18}O_{water}$ is –6.0 to 1.0 per mil (Fig. 1E).

Close association of fertile part with isotopically heterogeneous part suggests that the mechanism to precipitate significant amount of gold also produced the large variation of oxygen isotopic value of water. In the period IV vein, two cycles of characteristic $\delta^{18}O$ profile are included. The cycle begins with a rapid increase of $\delta^{18}O_{quartz}$ to ~12 per mil and followed by the gradual decrease to ~6 per mil. Period IV vein also consists of two sequential changes of quartz morphology, which begins with precipitation of bladed quartz followed by milky quartz and later clear quartz. This sequence is shown in Fig. 2; the early bladed quartz has high $\delta^{18}O$, and it gradually decreases and final coarse euhedral quartz has the lowest $\delta^{18}O$.
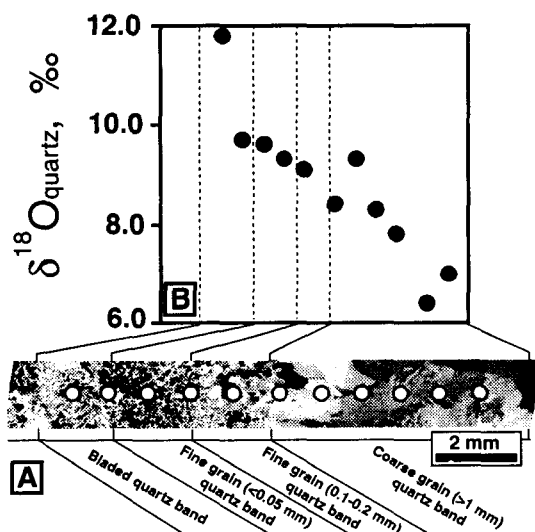
Fig. 2. Photomicrograph (A) and $\delta^{18}O_{quartz}$ (B) of period IV bands in Fig. 1. In accordance with the morphological change from early bladed quartz, middle fine grain quartz to late coarse euhedral quartz, the $\delta^{18}O$ values rapidly increase to ~12 ‰ then gradually decrease to ~6 ‰. Open circles in (A) show position of laser spots where oxygen was extracted for isotopic analysis.

In the low-sulfidation epithermal environment, precipitation of gold from ore-forming fluid is caused by decomposition of gold-bisulfide complex during the reduction of hydrogen sulfide concentration. This process may simultaneously occur during fluid boiling, dilution, oxidation, and precipitation of sulfide minerals (Hayashi and Ohmoto, 1991). Appearance of bladed quartz suggests the fluid boiling, and rapid increase of $\delta^{18}O_{quartz}$ from ~6 per mil to ~12 per mil is consistent with intense fluid boiling. However, the calculation of Rayleigh distillation using a fractionation factor of oxygen isotope between water and vapor (Horita and Wesolowski, 1994) indicates that

more than 92 percent of fluid is necessary to boil in order to increase the $\delta^{18}O_{quartz}$ up to 6 per mil. This large proportion of boiling is unlikely to occur during vein formation. Therefore, the rapid increase of $\delta^{18}O$ was probably caused by the combination of the introduction of fluid with $\delta^{18}O = \sim1.0$ per mil and the fluid boiling. The gradual decrease of $\delta^{18}O_{quartz}$ shown in Fig. 2 is reasonably explained by mixing with water of $\delta^{18}O = \sim-6$ per mil.

Although detailed temperature excursion during vein formation could not be clarified in this study, Th of fluid inclusions suggests wide range of temperature between 170° and 250°C. Higher temperature of deeply circulated fluid is also suggested by Izawa et al. (1990) and Matsuhisa and Aoki (1994). Izawa et al. (1990) discussed that temperature of ore-forming fluid was near 250°C in a moment. If temperature decreased systematically from 250°C to 170°C during the formation of typical growth zone shown in Fig. 2, calculated $\delta^{18}O$ values of water were ~3.1 per mil at the earlier stage and ~7.7 per mil at the later stage, respectively.

The most likely sources of above two fluids are water of magmatic origin and meteoric water. The $\delta^{18}O$ of local meteoric water around the Hishikari mine is -6.6 to -6.3 per mil, approximately agrees with the lowest $\delta^{18}O_{water}$ of this study (-6.0 ‰), and this value probably has not changed significantly during last ~1 Ma of the Hishikari vein formation. Since typical magmatic water has $\delta^{18}O$ range of 7±2 per mil, the fluid with $\delta^{18}O = \sim1$ per mil is not a typical one. Therefore, ore-forming fluid is either magmatic fluid that has been diluted to some extent by mixing with meteoric water at shallow depth, or circulating meteoric water that has undergone isotopic exchange with country rocks at elevated temperatures.

The submillimeter scale oxygen isotopic variation of vein quartz clearly demonstrates that the formation of epithermal vein is not a quiescent process. Intermittent opening of vein triggered the introduction of deep fluid to the epithermal system, and associated boiling and subsequent mixing with meteoric water caused the precipitation of precious metals.

References

Hayashi, K., Maruyama, T. and Satoh, H. (2000), Resource Geol., 50, 141-150.
Hayashi, K., Maruyama, T. and Satoh, H. (2001), Econ. Geol., 96, 211-216.
Hayashi, K. and Ohmoto, H. (1991), Geochim. Cosmochim. Acta, 55, 2111-2126.
Horita, J. and Wesolowski, D. J. (1994), Geochim. Cosmochim. Acta, 58, 3425-3437.
Izawa, E. et al. (1990), J. Geochem. Explor., 36, 1-56.
Matsuhisa, Y. and Aoki, M. (1994), Econ. Geol., 89, 1608-1613.
Matsuhisa, Y. et al. (1979), Geochim. Cosmochim. Acta, 43, 1131-1140.
Simmons, S. F. and Christenson, B. W. (1994), Amer. J. Sci., 294, 361-400.