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ESR DATING OF QUARTZ VEINS : COOLING HISTORY OF A HYDROTHERMAL SYSTEM

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ABSTRACT

The only method to date directly quartz crystals and veins is counting unpaired electrons in samples created by natural radiation and accumulated at some traps in proportion to their age, such as electron spin resonance (ESR) and thermoluminescence (TL) methods. In this study, the ESR dating method was employed on quartz veins which are exposed at the floor of Minase River, Akita Prefecture, Northeast Japan. 40 specimens of quartz vein were tested. ESR signals from Al inpurity center were detected in all specimens, and no other signal was observed. 5 specimens showed too much paramagnetic inpurity signals to calculate Al center ages, and 35 ESR ages were yielded; 6.4 - 24 ka for 34 specimens, and 0.47 ka for one. No systematic change in the ESR ages due to the sampling localities was observed. The ages 7.9 ka and 18 ka were calculated for 2 samples from the same vein. Slices from one vein of 20 cm thick yielded ages of 14 ka, 13 ka, 24 ka, 9.1 ka, 14 ka, 12 ka, and 8.5 ka, in which no systematic change was observed. Therefore, the difference between these ages has no effect, and thus the ages 6.4 - 24 ka for 34 specimens are roughly correlative. The field evidences suggest that the 0.47 ka quartz has been annealed by the younger hydrothermal activity. In conclusion, two phases of hydrothermal activity are found in this field; the main phase occured in 15±9 ka, and minor activity occured after it.

1. INTRODUCTION

Quartz is one of common constituent in hydrothermal deposits and have much information about past hydrothermal system. But no practical method has been developed to date quartz crystals and veins directly, except electron spin resonance (ESR) and thermoluminescence (TL) methods. In previous works, Takashima *et al.* (1987) discussed history of a hydrothermal system using 30 TL ages obtained from secondary quartz crystals in alteration halos, and Morifuji *et al.* (1989) reported ESR ages for three secondary quartz, one quartz sinter and one quartz vein specimens. The objective of this study is to clarify the cooling history of the hydrothermal system using sufficient number of ages obtained from quartz veins that have been precipitated from hydrothermal fluid.

2. STUDY AREA AND SAMPLES

The study area is located in the southeast of Akita Prefecture, Northeast Japan, 0.5 km south from recent hot springs (Oyasu-Daifuntou; over 90 °C), and 5 km east from Uenotai Geothermal Power Station (28.8MWe) (Fig. 1). Dated specimens are quartz veins which are exposed at the floor of Minase River for about 1 km (Fig. 2). Their country rocks are Miocene-Pliocene tuffs (Takeno, 1988), which have been hydrothermaly altered and silicified.

3. METHODS

3.1 ESR dating theory

ESR dating method uses unpaired electrons in samples created by natural radiation and accumulated at some defect and impurity traps in proportion to their age; thus the age is obtained by dividing the total dose (TD) determinged by ESR measurements by the annual dose (D) from natural radiation. The accumulation of unpaierd electrons starts at crystallization of the sample mineral, but trapped electrons are released and the age is reset to zero at a certain temperature. Therefore, if the sample has suffered such high temperature, the age counting is started at the reset temperature in its cooling history. Different traps in the same material have different reset temperatures, and it may provide important informations for the cooling rate. Unfortunately, the reset temperature depends on the cooling rate, but Toyoda and Ikeya (1991) gives some examples of reset temperature in cooling stage. For more detail, see Ikeya (1993).

3.2 Sample preparation

40 specimens of quartz vein were collected from the outcrop (Fig. 2). The country rock and weathered surface were cut off in order to eliminate external arpha and beta dose, and some of thick veins were sliced. Then quartz vein pieces were crushed in a mortar and were sieved to extract 0.125 - 0.25 mm grains. 0.1 g of extracted sample were enclosed in a test tube, and five tubes were prepared for each sample for artificial gamma irradiation.

3.3 ESR measurement and irradiation

The intensity of ESR signals were measured by a ESR spectrometer (JEOL JES-FE1XG) at room temperature and liquid nitrogen temperature (77 K) with a 100-kHz field modulation and a microwave power of 1 mW. 5 gauss magnetic field modulation width was used for measurement at 77K to simplify the hyperfine structure of ESR signals (Imai and Shimokawa, 1988), and signal intensities were measured by peak-to-peak height. A ^{60}Co source was used to irradiate samples for 30, 60, 90 and 120 minutes for each sample.

3.4 Calculation of dose rate

The natural radiation dose rate was calculated based on chemical analyses for quartz veins and country rocks. The concentrations of K_2O, U, and Th were determined by instrumental analysis for country rocks and the residue of quartz vein samples for ESR mesurement (Table 1). Alpha, beta and gamma doses were calculated with following coefficients:

$$D\alpha=2.78*C_U+0.74*C_{Th}$$
$$D\beta=0.15*C_U+0.03*C_{Th}+0.69*C_{K_2O}$$
$$D\gamma=0.11*C_U+0.05*C_{Th}+0.21*C_{K_2O}$$

where $D\alpha$, $D\beta$ and $D\gamma$ are Alpha, beta and gamma doses respectively, and C_U, C_{Th} and C_{K_2O} are contents of U, Th and K_2O respectively.

Annual dose (D) was calculated for each sample summing up internal alpha and beta doses from quartz vein, external gamma dose from the country rock which decreases exponentially with the distance from dated part of quartz vein, and cosmic dose which was estimated to be 0.19 mGy after Aitken (1985). 0.15 was used as alpha dose attenuation factor. Then Annual dose from the country rock $D\gamma r$ is

$$D\gamma r=\{\exp(-0.15*D1)+\exp(-0.15*D2)\}*D\gamma c$$

where D1 and D2 are distances from dated quartz on both side, and $D\gamma c$ is gamma dose of the country rock. External dose rate De is

$$De= D\gamma r+D_{cos}$$

where D_{cos} is cosmic dose. Internal dose rate Di is

$$Di=0.15*D\alpha q+D\beta q$$

where $D\alpha q$ and $D\beta q$ are alpha dose and beta dose of quartz respectively. Therefore, annual dose D for dated quartz is

$$D = De + Di$$

Radioactive disequilibrium and water effect were neglected.

4. RESULTS

4.1 Detected ESR signals

An example of observed ESR spectra is shown in Fig.3a. ESR signals from Al inpurity center were detected in all specimens at 77 K, and no other datable signal was observed. Broad signals similar to sine curves are judged to be caused by paramagnetic impurities such as Fe and Mn. Numerical compensation procedure for paramagnetic inpurity signals is shown in Fig. 3. An artificial sine curve is generated as an approximation of the paramagnetic signal, and then is subtracted from observed ESR spectrum; thus an almost flat baseline is obtained and Al signal intensity can be measured. This procedure was applied to 25 specimens. 5 specimens showed too much paramagnetic inpurity signals to cancel numerically and to calculate Al center ages.

The Al center signal increased in propotion to gamma irradiation, and no saturation was observed. The signal growth was plotted as a function of artificial dose for each specimen, and fitted to a straight line obtained by the least squares method. Total doses (TD) were determined by extrapolating the growth line to the zero ordinate.

4.2 Calculated Age

35 ESR ages were yielded as shown in Table 1; 6.4 - 24 ka for 34 specimens, and 0.47 ka for one. No relation has been found between ESR ages and sampling localities (Fig. 2).

4.3 Error

It is difficult to evaluate errors theoretically because of too many uncertain factors, such as erosion rate and loss of Rn gas in U- and Th-series disintegration. Therefore, errors for ESR age is estimated practically and directly from ages themselves. The ages 7.9 ka and 18 ka were obtained for 2 samples from the same vein, and the distance between them is about 1 m. Slices from one vein of 20 cm thick yielded ages of 14 ka, 13 ka, 24 ka, 9.1 ka, 14 ka, 12 ka, and 8.5 ka, in which no systematic change was observed (Fig. 4). Hence, the difference between these ages has no effect, and thus the ages 6.4 - 24 ka for 34 specimens are judged to be consistent with each other. Additional efforts are still needed to evaluate age errors.

5. DISCUSSION

5.1 Thermal stability of Al center

Thermal stability of Al center in quartz have been determined in some previous works. Imai and Shimokawa (1989) reported that the intensity of Al center signal begins to decrease at 200 °C and is annealed completely at 300 °C. Falgueres *et al.* (1991) reported that the ESR intensity of Al center was reduced to 20 % of initial intensity by annealing at 180 °C for 16 hours. Toyoda and Ikeya (1991) calculated closure temperature of Al center age to 149 °C in cooling rate of 1 °C/ka. As a rough estimation, the reset temperature of Al center age is inferred to be 150 - 200°C; further discussion is no use here because of too large errors for obtained ages.

5.2 Cooling history of the hydrothermal system

ESR ages of 34 in 35 specimens are considered to be consistent, and no local heterogeneity is detected. Therefore, it is concluded that the temperature of this hydrothermal system have dropped to 150 - 200 °C at 15±9 ka and ceased after it, in relatively rapid cooling rate.

The age 0.47 ka is considerably younger than all other ages in this study. 0.47 ka quartz was taken from an older vein in a fragment of country rock included in a 1-m-wide fracture zone filled by younger quartz which yielded no age because of much paramagnetic inpurity signals. This fracture zone extends for about 100 m near the northern end of this outcrop. The age 0.47 ka is too young to believe, but it is concordant with the field evidence, i.e., it is judged that the 0.47 ka quartz has been annealed by the younger hydrothermal activity which has been formed the fracture zone.

Three problems remain unsolved to discuss cooling history of hydrothermal systems using vein ESR age; firstly too large error and many error factors, secondly uncertainty of reset temperature, and thirdly single electron trap of specimens from this field. The next step study should include detailed error estimation and experimental comfirmation of reset temperature, and other specimens including multiple defects should be used.

6. CONCLUSION

The ESR signal of the Al centers yields ages ranging 6.4 - 24 ka with one exception 0.47 ka for a system of quartz veins. Based on these ages, two phases of hydrothermal activity are identified in this hydrothermal system; in the main phase, the temperature of the hydrothermal sustem have dropped to 150 - 200 °C at 15±9 ka and ceased in relatively rapid cooling rate; in later phase, minor activity formed a fracture zone and annealed surrounding rocks.

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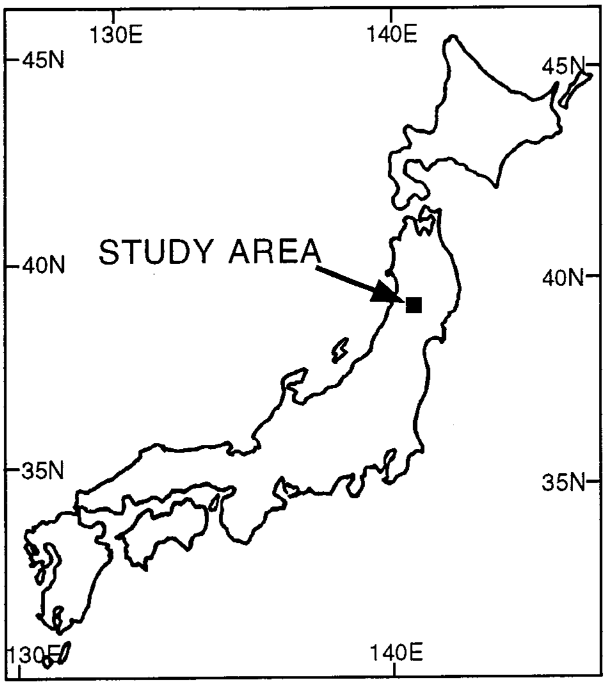

Figure 1. Locality of study area.

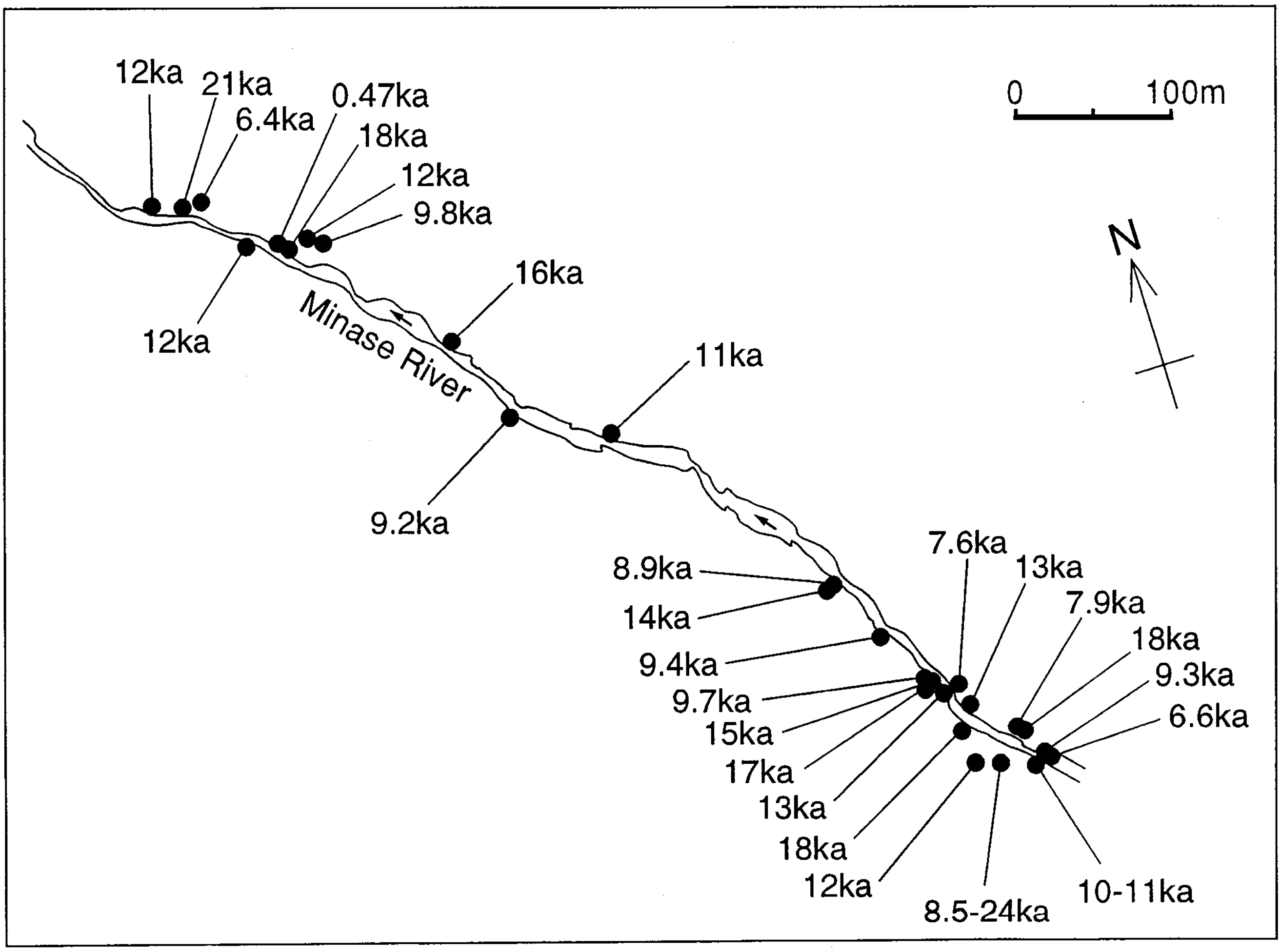

Figure 2. Sampling points and ages.

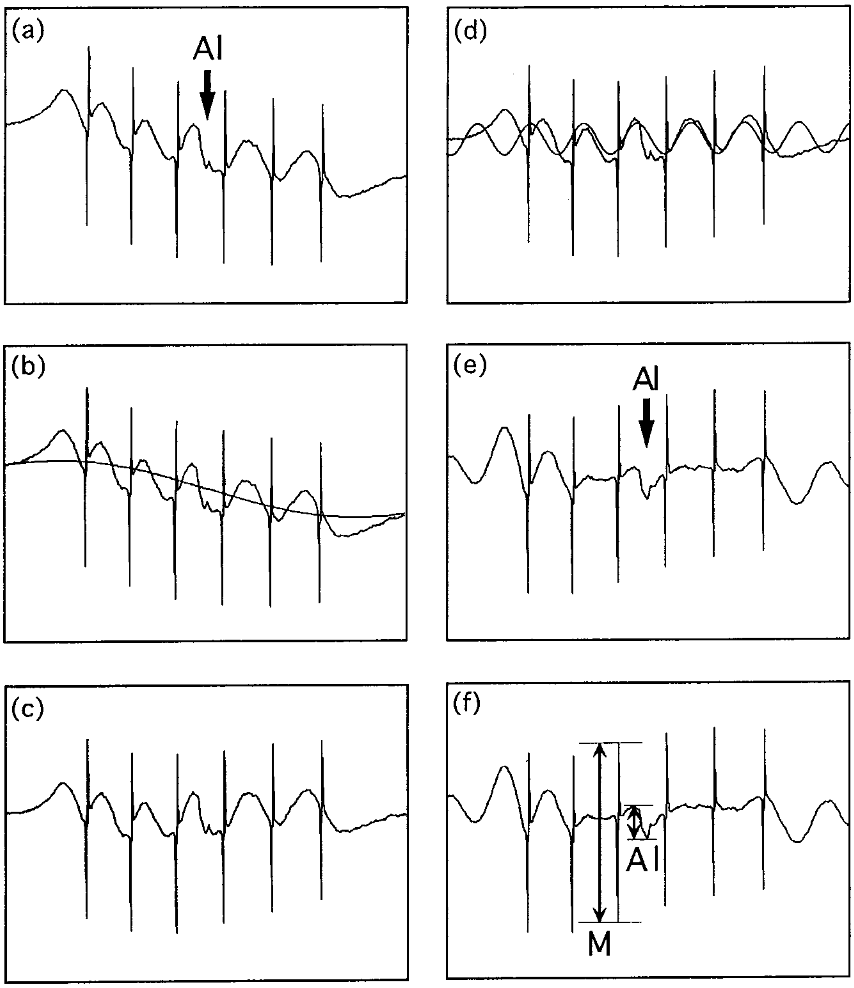

Figure 3. Compensation procedure of paramagnetic inpurity signals. Al:Al center signal. (a)obtained spctrum. (b)curve fitting to Fe signal. (c)result of subtraction. (d)curve fitting to Mn signal. (e)result of subtraction. (f)measured Al signal intensity (Al) is normalized by marker signal intensity (M).

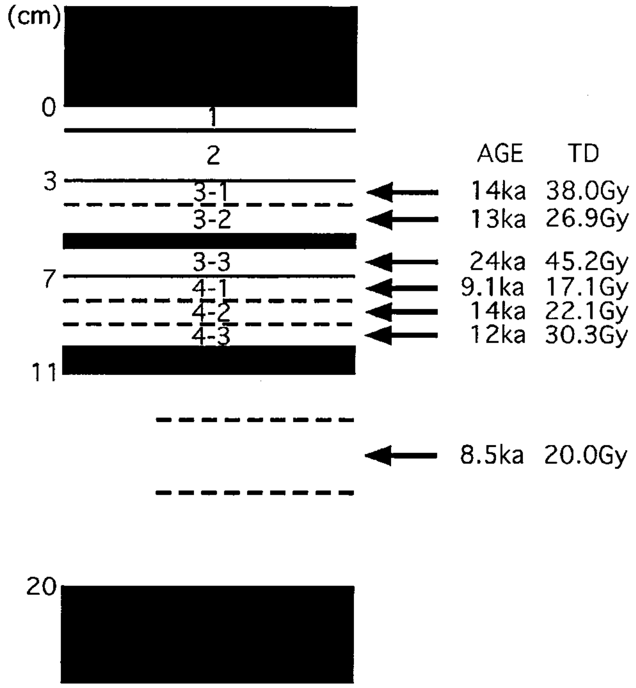

Figure 4. TDs and ages of a 20 cm thick quartz vein.

Table 1. Age calculation. *:slices from a 20cm thick vein (see Figure 4). Chemical analyses by Activation Laboratories Ltd. (Pack 4E).

sample No.	TD (Gy)	vein thickness	country rock dose (mGy/y)	internal dose (mGy/y)	annual dose (mGy/y)	age (year)
91073001-1	26.90	11cm	0.91019	0.95120	2.05139	13113
91073006	17.00	4cm	1.75263	0.61710	2.55973	6641
91073007	25.00	3cm	1.88913	0.61710	2.69623	9272
91073101-3-1	38.00	20cm*	1.00168	1.44060	2.63228	14436
91073101-3-2	26.94	20cm*	0.89973	1.02020	2.10993	12768
91073101-3-3	45.22	20cm*	0.75482	0.92170	1.86652	24227
91073101-4-1	17.11	20cm*	0.70306	0.97880	1.87186	9141
91073101-4-2	22.13	20cm*	0.68068	0.70310	1.57378	14062
91073101-4-3	30.33	20cm*	0.66692	1.72330	2.58022	11755
91073101-5	20.00	20cm*	0.83668	1.31670	2.34338	8535
91073103	38.37	> 1m	0.33071	2.32980	2.85051	13461
91073106	58.75	4cm	1.20798	1.97970	3.37768	17394
91073107	27.60	4cm	1.20798	0.38660	1.78458	15466
91073108	16.40	5cm	1.12069	0.38660	1.69729	9662
91073113	29.00	2cm	1.08294	1.80160	3.07454	9432
91080102	26.40	2.5cm	1.60749	0.46210	2.25959	11684
91080103	33.00	6cm	1.23636	0.40700	1.83336	18000
91080104-1	25.32	5cm	1.61564	0.51700	2.32264	10901
91080104-2	21.17	5cm	1.59268	0.23440	2.01708	10495
91080105	17.00	8cm	1.09120	0.95120	2.23240	7615
92080520	19.00	4cm	0.09497	0.75130	1.03627	18335
92080522	15.20	2cm	0.17817	0.34470	0.71287	21322
92080524	25.67	3cm	1.41603	0.54450	2.15053	11937
93080904	24.80	2cm	1.80387	0.20710	2.20097	11268
93080907	16.20	3cm	1.28602	0.28290	1.75892	9210
93080909	27.20	5cm	1.43555	0.07580	1.70135	15987
94080501	33.80	2cm	1.58941	0.96520	2.74461	12315
94080503	31.40	4cm	1.47456	0.92040	2.58496	12147
95080901	0.50	1cm	0.11894	0.75130	1.06024	472
95080903	12.00	2cm	1.08294	0.07630	1.34924	8894
95080904	23.40	2cm	1.08294	0.39990	1.67284	13988
95080906	14.67	2cm	1.58941	0.51730	2.29671	6387
96080605	22.33	5cm	1.16331	0.92040	2.27371	9822
96080902	13.33	10cm	1.11752	0.37220	1.67972	7938