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Rb-Sr isotopic study of the Hwacheon granite in northern Gyeonggi massif, Korea: A case of spurious Rb-Sr whole rock age

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ABSTRCT: The Yonghwasan pluton of the Hwacheon granite intruding the Precambrian metamorphic rocks of the northern Gyeonggi massif gives two apparent Rb-Sr whole rock isochrons depending on lithology: 177±11 Ma with an initial $^{87}Sr/^{86}Sr$ of 0.71538±0.00031 for mica granite and 134±10 Ma with an initial $^{87}Sr/^{86}Sr$ of 0.72183±0.00096 for garnet two-mica granite. However, the whole rock age for the garnet two-mica granite is much younger than mineral ages: Rb-Sr biotite (146.2±2.2 Ma) and muscovite (160.2±1.6 Ma), and reported CHIME monazite and K-Ar muscovite. Circumstantial evidences suggest that the young whole rock isochron may be the result from fortuitous alignment of data due to assimilation of country rocks, and thus meaningless, rather than resetting of whole rock Sr isotope ratios by later events such as post-emplacement heating and fluid infiltration. Considering the above, the best Rb-Sr whole rock age of the Yonghwasan pluton is estimated to be 172.0±6.8 Ma. The isotopic ages of this and previous studies for the garnet two-mica granite form a near linear cooling curve with a cooling rate of about 14.5 °C/Ma.

Key words: Rb-Sr isotope, whole rock age, mineral age, cooling history, false isochron

1. INTRODUCTION

A Rb-Sr whole rock isochron age is commonly used to determine the emplacement age of an igneous body, because it is believed that Rb-Sr whole rock system closes isotopic exchange near the solidus temperature of a magma. However, there are cases for distorted Rb-Sr whole rock age of an igneous body due to fluid infiltration (Kwan et al., 1982; Siebel, 1995), or to later thermal disturbance (Field and Raheim, 1979a and b; Compston et al., 1982). Also, it might be possible to have a distorted age in an isochron diagram by fortuitous alignment of data with heterogeneous initial isotopic ratios caused by assimilation of country rocks, as in the case of wrong Sm-Nd whole rock age (Chauvel et al., 1985). These observations caution that the Rb-Sr whole rock dating cannot be simply regarded as a reliable dating method of determining the emplacement age of an igneous body. Sagong et al. (2005) report several cases of false whole rock isochron age for the Phanerozoic granitic plutons in South Korea.

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In this study, we report a case for a distorted Rb-Sr whole rock age from the Hwacheon granite intruding the Precambrian Gyeonggi massif of Korea that might lead to a wrong interpretation about the emplacement age and the origin of the granite, and emphasize that multi-isotopic approach is necessary for better interpretation of the Rb-Sr whole rock data of a pluton.

2. GEOLOGIC BACKGROUND

An overall description of the Phanerozoic granitoids in South Korea is available in Chough et al. (2000) and Sagong et al. (2005). The general geology of the Hwacheon granite and its surrounding area has been described in detail by Sagong et al. (1997), and thus a brief summary is presented here. The Hwacheon granite intrudes the northeastern part of the Gyeonggi massif that is one of the Precambrian basement terranes in Korean Peninsula. The metamorphic rocks of the Gyeonggi massif around the Hwacheon granite consist mainly of banded biotite gneiss, leucocratic gneiss, quartzite and minor amphibolite (Fig. 1; Han, 1975; Park et al., 1997). The western part of the study area experienced greenschist to amphibolite facies metamorphism (Han, 1975), whereas granulite facies metamorphism has recently been reported by Lee and Cho (2003) just to the north of the study area. Cho et al. (1996) gave CHIME (chemical U-Th-Total Pb isochron method) monazite ages from a sillimanite garnet gneiss to about 15 km west of the Hwacheon granite and interpreted that the protolith of the gneiss formed about 1700 Ma, and suffered a peak metamorphism at 245±3 Ma with a later metamorphism at around 183-186 Ma.

Sagong et al. (1997) gave petrography and chemical data for the Hwacheon granite. The Hwacheon granite consists of two plutons: the southwestern medium-grained (0.5-4 mm) mica granite (Yonghwasan pluton) and the northeastern porphyritic medium-grained (0.3-2 mm) biotite granite (Chugongnyeong pluton), with the former intruding the latter. The Yonghwasan pluton can be further divided lithologically and geographically into the northern mica granite and the southern garnet two-mica granite (Fig. 1). Since it is not clear whether the muscovite is primary or secondary min-

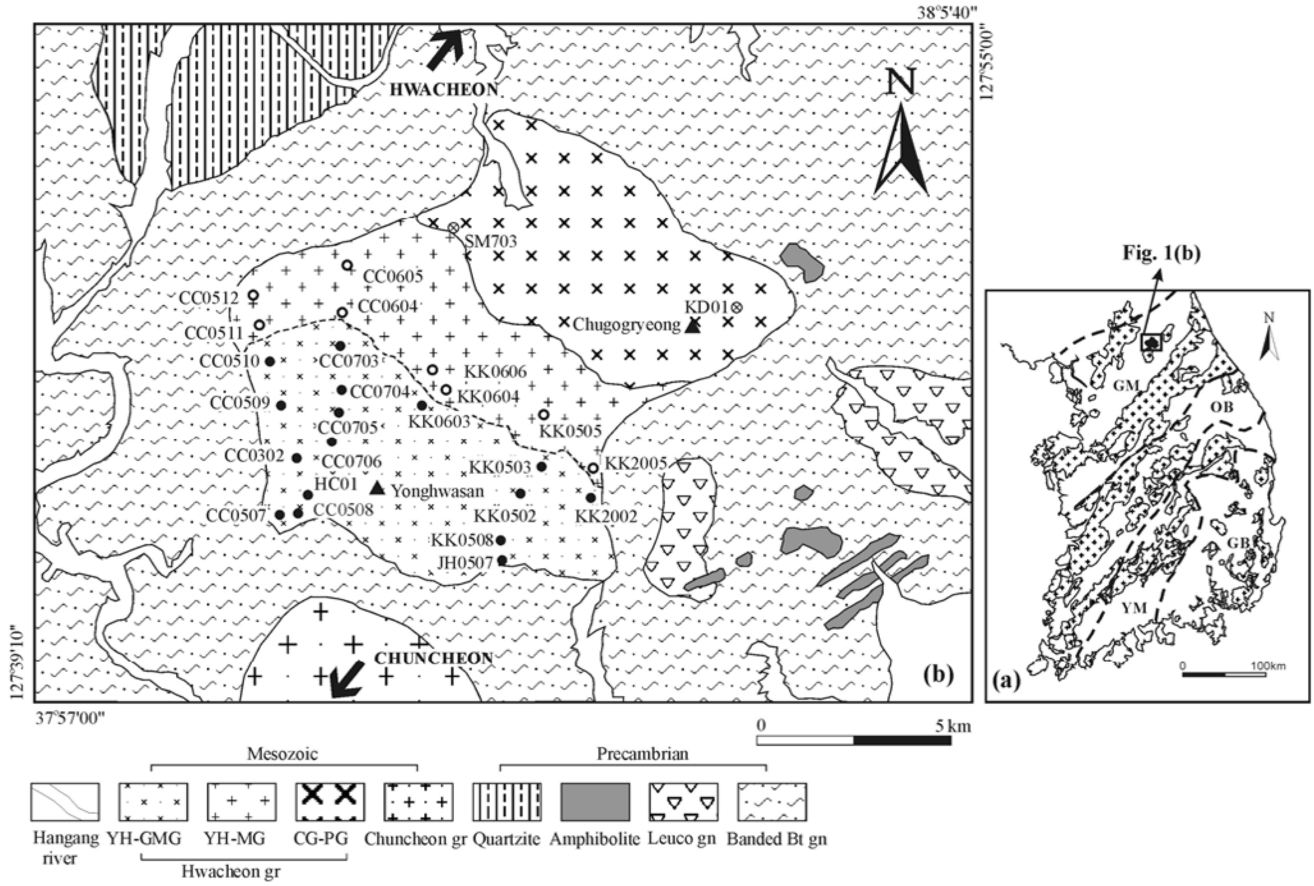

Fig. 1. (a) Distribution of Phanerozoic granites in South Korea. (b) Geologic map of the Hwacheon granite and sample locations, after Sagong et al. (1997).

eral, we prefer to use the terminology mica granite to describe the northern garnet-free rocks of the Yongwhasan pluton. The boundary between the two granites appears to be gradational, suggesting that the two rocks can be considered as differentiated products from a single magma. The mineral assemblage of the Yonghwasan pluton is quartz+K-feldspar +plagioclase+biotite (light brown to greenish brown)±garnet±muscovite with minor apatite, monazite, zircon and opaque oxides. The garnet in the garnet two-mica granite is euhedral to subhedral grain of 0.5-2 mm size and has no reaction relationship with biotite. These observations suggest that the garnet is of magmatic origin (Allan and Clark, 1981). The mineral assemblage of the Chugongnyeong pluton is quartz+plagioclase+perthitic K-feldspar+biotite (light brown to brown) with minor apatite, opaque oxides and zircon. The phenocryst of the porphyritic biotite granite is K-feldspar of 1-2.5 cm size. Secondary minerals include sericite and chlorite.

The garnet two-mica granite tends to have slightly lower SiO_2 content (73-76 wt%) than mica granite (74-79 wt%). All rocks are peraluminous, and have low MgO (<0.18 wt%), TiO_2 (<0.14 wt%), MnO (<0.09 wt%) and P_2O_5 (<0.04 wt%) contents. Al_2O_3, MnO and Na_2O contents tend to show smoothly decreasing trend with increasing SiO_2, while other major elements do not show coherent variation in Harker diagrams. Transition elements such as Ni (<1 ppm), V (<12 ppm), Cr (<6 ppm), Cu (<3 ppm) are uniformly low. Rb, Ga, Zr, Y, Th, U and Zn show a decreasing trend with increasing SiO_2. Sr increases with SiO_2, while Nb does not show coherent trend. The garnet two-mica granite tends to have higher MnO, Rb, Y and Ga contents than the mica granite. MnO, Y and Ga differences can be explained by presence of garnet in the garnet two-mica granite. In Rb-(Y+Nb) diagram of Pearce et al. (1984), all data belong to the volcanic arc granite field.

Cho et al. (1996) measured CHIME monazite ages from a garnet two-mica granite: 190-260 Ma apparent ages from some spots of grain cores, 74-154 Ma apparent ages from grain edges and rough-surface parts, and 172±5 Ma isochron age from the rest of data. They interpreted the 190-260 Ma as the age of xenocrysts derived from the surrounding gneisses and the 172±5 Ma as the emplacement age of the Yongwhasan pluton, although no interpretations were given for the younger apparent ages. Park et al. (1997) gave a K-Ar muscovite age of 151±4 Ma from the same sample as Cho et al. (1996) analyzed.

3. SAMPLES AND ANALYTICAL PROCEDURES

The samples analyzed for this study are five mica granites and six garnet two-mica granites from the Yonghwasan pluton (Fig. 1). The rocks of the Chugongnyeong pluton are rather severely weathered, and thus are not included in this study.

Isotopic analyses including chemical separation and mass spectrometry were performed at the Korea Basic Science Institute. All chemistries were done in a clean room with laminar flow benches. Acids and water used were double-distilled with quartz still or Teflon two-bottle method. About 100 mg of rock powders and mineral separates were mixed with highly enriched ^{84}Sr and ^{87}Rb spikes and then dissolved with a mixed acid (HF : $HClO_4$: HNO_3=3:1:1) in Teflon vessels. About 0.5 ml of sample solution was loaded onto 3.75 g of cation resin bed (Dowex AG50W-X8, H+ form, 200-400 #) charged into a quartz column (5 mm in diameter x 190 mm in length). Rb and Sr fractions were collected with 2.5N HCl.

Isotopic ratios were measured using a VG 54-30 thermal ionization mass spectrometer equipped with nine Faraday cups. Rb fraction was dried with one drop of H_2SO_4 and loaded onto Ta single filament with H_2O. Sr fraction was treated with one drop of HNO_3 and loaded onto Ta single filament with 1M H_3PO_4. ^{87}Rb/^{85}Rb ratio was measured with a static mode, while ^{87}Sr/^{86}Sr ratio was measured with a dynamic mode. During the dynamic run, ^{84}Sr/^{86}Sr ratio was measured simultaneously with a static mode. Measured ^{87}Sr/^{86}Sr ratios were corrected for instrumental fractionation by normalizing to ^{86}Sr/^{88}Sr=0.1194. These ^{87}Sr/^{86}Sr ratios were further corrected for Sr contribution from spikes. Rb and Sr concentrations were determined by an isotope dilution technique. Replicate analyses of NBS 987 standard gave ^{87}Sr/^{86}Sr=0.71011±0.00003 (N=11, $2s_m$). Final ^{87}Sr/^{86}Sr ratios of samples are reported relative to ^{87}Sr/^{86}Sr=0.710250 for the NBS 987 standard. Total blank levels were below 200 pg for Rb and 400 pg for Sr during this study, and have negligible effect to the sample isotope data. Isochron parameters were calculated after the method of York (1969) using the ISOPLOT program of Ludwig (1990). Errors of calculated age and initial ratio are reported at the 95% confidence level. Errors used during the calculation of the isochron are 1% for Rb/Sr and 0.005% for ^{87}Sr/^{86}Sr ratios at the 95% confidence level. The analytical data are presented in Table 1.

4. RESULT AND INTERPRETATION

Rb and Sr contents measured by isotope dilution agree well with those by X-ray fluorescence method (Sagong et al., 1997). Garnet two-mica granite has higher Rb and lower Sr contents, and thus higher Rb/Sr ratio than mica granite.

The isotope data are plotted in a ^{87}Rb/^{86}Sr-^{87}Sr/^{86}Sr isochron diagram (Fig. 2a). Excluding one sample (JH0507) which is

Table 1. Rb-Sr whole rock and mineral isotope data for the Hwacheon granite

Sample	Rb (ppm)	Sr (ppm)	^{87}Rb/^{86}Sr*	^{87}Sr/^{86}Sr**	$\pm 2\sigma_m$	Mineral age (Ma)***
Garnet two-mica granite						
CC0507 WR	232.6	53.4	12.6	0.745775	0.000010	
CC0507-ms	892.4	3.7	836.	2.620824	0.000029	160.2±1.6
CC0507-bt	84.3	6.1	40.4	0.803554	0.000022	146.2±2.2
CC0509 WR	200.8	94.5	6.16	0.733547	0.000007	
CC0705 WR	187.5	131.1	4.15	0.729558	0.000010	
HC01 WR	179.7	42.7	12.2	0.744792	0.000014	
JH0508 WR	202.9	102.9	5.72	0.733004	0.000011	
JH0507 WR	171.3	61.9	8.05	0.755940	0.000011	
Mica granite						
CC0511 WR	131.0	146.6	2.59	0.721728	0.000011	
-duplicate	134.7	149.0	2.62	0.721907	0.000008	
CC0604 WR	133.7	103.8	3.73	0.725120	0.000009	
CC0605 WR	98.9	183.1	1.57	0.719328	0.000008	
KK0606 WR	74.2	301.9	0.711	0.717209	0.000009	
KK0606-bt	595.5	13.1	135.	1.026226	0.000022	161.3±1.6
KK2005 WR	137.9	126.5	3.16	0.723304	0.000007	

*Uncertainty is below 1%.
**Normalized to NBS987=0.710250
***Errors are at 95% confidence level
Abbreviations: WR, whole rock; ms, muscovite; bt, biotite

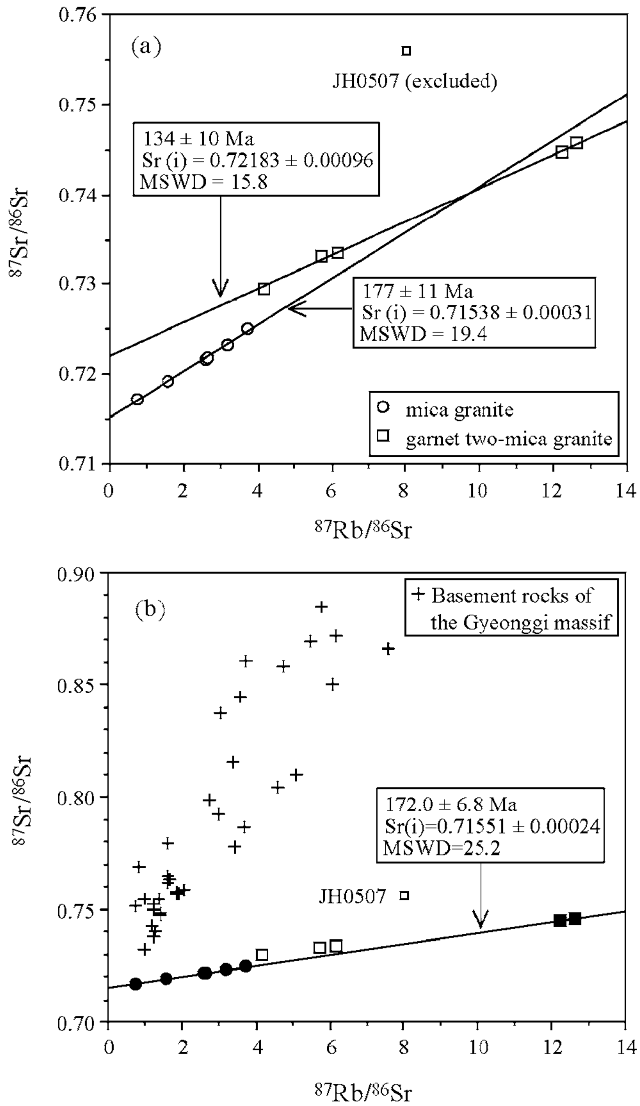

Fig. 2. (a) $^{87}Rb/^{86}Sr$-$^{87}Sr/^{86}Sr$ isochron diagram for the Yonghwasan pluton of the Hwacheon granite. Note that mica granite and garnet two-mica granite define distinctly different isochrons. (b) $^{87}Rb/^{86}Sr$-$^{87}Sr/^{86}Sr$ isochron diagram for the Yonghwasan pluton and basement rocks of the Gyeonggi massif (Choo, 1983; Kwon et al., 1999). The 172 Ma isochron for data with closed symbols is a preferred interpretation for the emplacement age of the Yongwhasan pluton. Compare with Fig. (a) above. See text for discussion.

a significant outlier relative to the rest of samples, the data for the two types of granites form two different linear arrays. The data for the mica granite define 177±11 Ma (MSWD=19.4) with an initial $^{87}Sr/^{86}Sr$ of 0.71538±0.00031, while those for the garnet two-mica granite define 134±10 Ma age (MSWD=15.8) with an initial $^{87}Sr/^{86}Sr$ of 0.72183±0.00096. The data for mica-whole rock pairs give the following ages: a muscovite age of 160.2±1.6 Ma and a biotite age of 146.2 ±2.2 Ma from a garnet two-mica granite, and a biotite age of 161.3±1.6 Ma from a mica granite (Table 1). The K-Ar muscovite age of 151±4 Ma (Park et al., 1997) from a garnet two-mica granite is compatible with the Rb-Sr mineral ages from the same lithology.

Apparently the whole rock Rb-Sr isotope data indicate that the Yongwhasan pluton is not a single pluton, and that the mica granite emplaced in middle Jurassic was intruded by the garnet two-mica granite in early Cretaceous, with a time span of ~40 Ma. However, this interpretation is not compatible with gradational boundary between the two granites observed in the field. The 134±10 Ma whole rock age of the garnet two-mica granite is much younger than the reported CHIME monazite of 172±5 Ma (Cho et al., 1996) and K-Ar and Rb-Sr mica ages described above. Since the CHIME monazite age has no requirement such as homogeneous initial isotope ratio, as does Rb-Sr whole rock age, and has a blocking temperature of 650-700 °C (Suzuki et al., 1994) approaching solidus temperature of a hydrous granite magma, it can be considered that the CHIME monazite age represents an age very close to the intrusion of the garnet two-mica granite, which agrees well with the Rb-Sr whole rock age of mica granite. Thus, the two lithologically different granites of the Yonghwasan pluton are considered as differentiates from a single magma. The conclusion is also supported by a K-Ar muscovite age of 151±4 Ma (Park et al., 1997) from the same sample for which the CHIME monazite age was measured and by Rb-Sr mica ages of this study, since K-Ar or Rb-Sr mica ages cannot be older than a Rb-Sr whole age from the same pluton, considering higher blocking temperature of Rb-Sr whole rock than that of K-Ar or Rb-Sr mica systems.

When the data for the mica granite are regressed with those for two garnet two-mica granite having highest Rb/Sr ratios (samples CC0507 and HC01), it gives 172.0±6.8 Ma (MSWD=25.2) with an initial $^{87}Sr/^{86}Sr$ of 0.71551±0.00024 (Fig. 2b), agreeing very well with the CHIME monazite age. Thus, we think that this might be a better interpretation of the Rb-Sr whole rock data for the Yonghwasan pluton. In this case, the four data of garnet two-mica granite plotting above the 172 Ma isochron can be interpreted as the result of assimilation of country rocks, although the sample JH0507, the outlier, may have suffered post-emplacement alteration. Such a relationship is shown in Fig. 2b. Also plotted in Fig. 2b along with the data for the Hwacheon granite are Rb-Sr whole rock data for the Precambrian basement rocks of the central Gyeonggi massif reported by Choo (1983) and Kwon et al. (1999). All the data for the basement rocks plot well above the isochron defined by the Hwacheon granite data and form a much steeper slope. When calculated back at 172 Ma, the $^{87}Sr/^{86}Sr$ ratios of basement rocks range from 0.725 to 0.875, which is much higher than those of the Hwacheon granite. This observation indicates that the Hwacheon granite did not form by direct partial melting of exposed Precambrian basement rocks. Thus, the initial Sr isotope ratios of the Hwacheon granite suggest that the

Hwacheon granite formed either by partial melting of the lower crust with lower Sr isotopic ratios than the exposed basement rocks as commonly supposed (Hamilton et al., 1979), or by mixing crust-derived magma with some mantle-derived component.

5. DISCUSSION

5.1. Cooling History

Figure 3 shows a cooling history of garnet two-mica granite in age-blocking temperature diagram, using the data from Cho et al. (1996), Park et al. (1997) and this study. Blocking temperatures used are: 300±50 °C for Rb-Sr biotite (Jager et al., 1967), 350±50 °C for K-Ar muscovite (Purdy and Jager, 1976), 500±50 °C for Rb-Sr muscovite (Purdy and Jager, 1976), and 675±25 °C for CHIME monazite (Suzuki et al., 1994). Relative ages from each isotopic system fit very well with its known blocking temperature, indicating a rather simple cooling history for the garnet two-mica granite. This means that no later thermal events, if any, happened above the blocking temperature of Rb-Sr biotite for the garnet two-mica granite. The near linear cooling curve corresponds to a cooling rate of about 14.5 °C/Ma.

There are just a few studies on the cooling rate of the Phanerozoic granitoids in South Korea. The cooling rate of the Hwacheon granite is similar to that (~10 °C/Ma) of a Triassic granodiorite body in Inje-Hongcheon area, about 40 km southeast of the Hwacheon granite (Jwa et al., 1990), but is much lower than those (26 to 133 °C/Ma) of the Cretaceous plutons in the Gyeongsang basin (Lee et al., 1995).

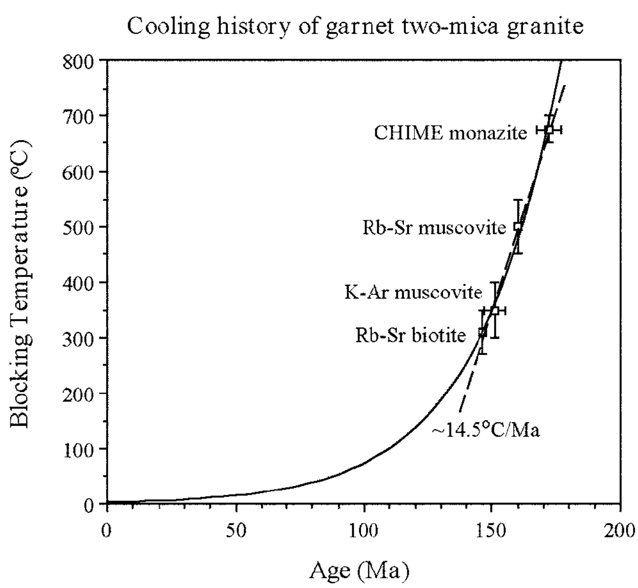

Fig. 3. Age-temperature diagram showing a simple cooling history of garnet two-mica granite. A linear fitting of the data gives a cooling rate of about 14.5 °C/Ma.

The tendency that the Cretaceous plutons have higher cooling rate than the Jurassic and Triassic ones (this study; Jin et al., 1984) agrees well with the observation that the former is shallow intrusive and the latter is deep one (e.g., Cho and Kwon, 1994).

5.2. Spurious Rb-Sr Whole Rock Isochron

We have seen that the younger apparent Rb-Sr whole rock age of 134 Ma defined by five garnet two-mica granite data (Fig. 2a) is much younger than the K-Ar or Rb-Sr mica ages, and thus cannot represent the emplacement age. However, the question "Is there a chance for this age to have any geologic meaning?" still remains. We will discuss two possibilities for those data to define the apparent isochron.

First, the data for the five garnet two-mica granites can be considered as two clustered groups, somewhat similar to a two-point isochron. In this case, lower Rb/Sr samples could have been disturbed for some reasons, while the higher Rb/Sr samples remained as closed system. The reasons for disturbance could be due to initial Sr isotopic heterogeneity caused by assimilation, or post-emplacement alteration. The monazite xenocryst in garnet two-mica granite reported by Cho et al. (1996) indicates assimilation of country rock. Whatever the reason, the age would not have any geologic meaning in this case.

Second, complete isotopic resetting after the emplacement of the granite by fluid infiltration or thermal event can result in geologically meaningful age. There are not many age data suggesting later thermal event reported at 134 Ma around the study area. The Chuncheon granite occurring to the south of the Hwacheon granite has a K-Ar hornblende age of 210.5±5.0 Ma (Jin et al., 1993). Since the Chuncheon granite is older than the Hwacheon granite, it cannot be a heat source for later thermal disturbance. Cho et al. (1996) reported from garnet two-mica granite CHIME monazite apparent ages of 74-154 Ma from grain edges and rough-surface parts. These apparent ages might suggest some later thermal disturbances. However, at present it is not clear about what caused the thermal disturbances or if they were strong enough to cause complete resetting of Rb-Sr whole system of garnet two-mica granite. Siebel (1995) reported a similar case as in the Yonghwasan pluton in his geochronological study of the Leuchtenberg granite in Germany. The Leuchtenberg granite shows a continuous differentiation trend from coarse-grained porphyritic biotite granite to medium- to fine-grained garnet-bearing muscovite granite, but the two granites have different ages. The Rb-Sr whole rock age of biotite granite is 326±2 Ma, while that of garnet muscovite granite is 317±2 Ma, with a time span of about 10 Ma. Siebel (1995) attributed the age difference to later hydrothermal event, noting that K-Ar mica ages of 326-323 Ma from garnet muscovite granite are similar to the Rb-Sr whole rock age of biotite granite and are older than the Rb-Sr whole rock age of garnet muscovite granite. He proposed that the small grain size, low Sr content (2-13 ppm) and high Rb/Sr ratio of garnet muscovite granite make it susceptible to resetting of initial whole rock Sr ratios, while Rb-Sr whole rock system of the coarse-grained biotite granite with higher Sr content was essentially unaffected, when hydrothermal fluids infiltrated the granite. However, this mechanism is difficult to apply in the case of the Yonghwasan pluton of this study. That is, it is difficult to explain how hydrothermal fluids can affect only the garnet two-mica granite of the Yonghwasan pluton while leaving mica granite intact, since the grain size of garnet two-mica granite is very similar to that of the mica granite and Sr contents (43-131 ppm) of garnet two-mica granite are not particularly low as in the case of the Leuchtenberg granite. Also, it is hard to imagine that the fluid infiltration occurred only in particular lithology.

Therefore, we tentatively conclude that the 134 Ma age defined by garnet two-mica granite is geologically meaningless.

6. CONCLUSION

We documented the Rb-Sr whole rock age defined by the garnet two-mica granite phase of the Hwacheon granite does not represent the emplacement age by field relationship and by comparing CHIME monazite age (Cho et al., 1996), K-Ar muscovite age (Park et al., 1997), and Rb-Sr mica ages of this study. Especially notable is that the Rb-Sr whole rock age is much younger than the K-Ar or Rb-Sr mica ages. Among the possible reasons for the apparent isochron of the garnet two-mica granite, a fortuitous alignment of data to form an isochron without geologic meaning is favored. This study shows that Rb-Sr whole rock method is not always reliable one of dating emplacement of granites, and that it is important to check Rb-Sr whole rock age with other isotopic systems for correct interpretation of Rb-Sr whole rock data of a granite pluton.

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