

## Oxygen and Hydrogen Isotopic Compositions of the Hwacheon Granite

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### 화천화강암의 산소와 수소 동위원소 조성

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**Abstract:** Oxygen and hydrogen isotopic compositions of the Jurassic peraluminous Hwacheon granite were measured, and compared with those of other Jurassic peraluminous Daebo granite in Korea.  $\delta^{18}\text{O}$  values for quartz and feldspar of the Hwacheon granite range from 8.2 to 10.6 and 5.8 to 9.0‰, respectively. Whole rock  $\delta^{18}\text{O}$  values for banded biotite gneiss country rocks surrounding the Hwacheon granites range from 8.1 to 9.4‰. Whole rock and biotite  $\delta\text{D}$  values for Hwacheon granite range from -84 to -113 and -107 to -113‰, respectively. Whole rock  $\delta\text{D}$  values for banded biotite gneiss country rocks range from -76 to -100‰. Both  $\delta^{18}\text{O}$  and  $\delta\text{D}$  values of the Hwacheon granite are characterized by low values compared to the 'normal' values for the fresh peraluminous granitic rocks. Low  $\delta^{18}\text{O}$  values of the Hwacheon granite resulted from fluid-rock interaction for a long period. Isotopic modelling result renders that a relatively low- $\delta^{18}\text{O}$  fluid below ~1‰ was involved in subsolidus isotopic exchange under a relatively high fluid/rock ratio (<-6). The fluid of meteoric origin has experienced a modification of oxygen isotopic composition as a result of fluid-rock interaction with the Hwacheon granite and surrounding metapelitic country rocks.

**Key words:** peraluminous granite, oxygen isotope, hydrogen isotope, Hwacheon granite, hydrothermal alteration, subsolidus isotopic exchange

**요약:** 중생대 쥐라기에 생성된 과알루미나질 화천화강암의 산소와 수소 안정 동위원소 조성을 알아보고, 이들 값이 다른 지역에 분포하는 쥐라기 화강암이 가지는 값과 어떠한 차이를 보이는지를 비교해보았다. 화천화강암을 구성하는 석영과 사장석은 각각 8.2에서 10.6‰과 5.8에서 9.0‰ 사이의  $\delta^{18}\text{O}$  값을 가지며, 화천화강암 주위에 분포하는 호상흑운모편마암의 전암  $\delta^{18}\text{O}$  값은 8.1에서 9.4 사이의 값을 갖는다. 화천화강암의 전암과 흑운모의  $\delta\text{D}$  값은 각각 -84에서 -113‰과 -107에서 -113‰ 사이의 값을 갖는다. 기반암의 전암  $\delta\text{D}$  값은 -76에서 -100‰ 사이의 값을 갖는다. 화천화강암의  $\delta^{18}\text{O}$ 과  $\delta\text{D}$  값은 과알루미나질 화강암이 전형적으로 가지는 값에 비해 낮은 값을 가지는 특징을 보여준다. 화천화강암이 가지는 낮은  $\delta^{18}\text{O}$  값은 오랜 기간 동안 이루어진 유체-암석 상호작용으로부터 기인한 것이다. 안정동위원소 모델링 결과는 약 1‰ 이하의 비교적 낮은  $\delta^{18}\text{O}$  값을 가지는 유체가 비교적 높은 유체/암석 비(<-6) 환경 하에서 동위원소 교환반응이 일어났음을 지시해준다. 이 유체는 천수가 화천화강암 및 주변에 분포하는 변성이질암과의 오랜 시간에 걸친 산소동위원소 반응 결과  $\delta^{18}\text{O}$  값이 무거워진 유체이다.

**핵심어:** 과알루미나질 화강암, 산소 동위원소, 수소 동위원소, 화천화강암, 열수변질, 고상선하 동위원소 교환반응

### Introduction

The peraluminous granitic rocks are known to origi-

nate from partial melting of the metapelitic rocks in the deep crustal levels (Chappell and White, 1974) and have  $\delta^{18}\text{O}$  values greater than 10‰ (O'Neil and

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Chappell, 1977; O'Neil *et al.*, 1977; Masi *et al.*, 1981; Lee *et al.*, 1982; Nabelek *et al.*, 1992). The Jurassic Daebo granites in Gyeonggi and Yeongnam massif, however, which also include peraluminous granitic rocks are characterized by low  $\delta^{18}\text{O}$  values mostly between 7 and 10‰ (Kim, 1991). The peraluminous granitic rocks are distributed along the northern margin of the Gyeonggi massif (Fig. 1a, Sagong *et al.*, 1997). We measured oxygen and hydrogen isotopic compositions of the Hwacheon granite which is peraluminous (Sagong *et al.*, 1997), in order to figure out if the Hwacheon granite has  $\delta^{18}\text{O}$  values greater than 10‰ which is 'normal' for the fresh peraluminous granitic rock (O'Neil and Chappell, 1977; O'Neil *et al.*, 1977; Masi *et al.*, 1981; Lee *et al.*, 1982; Nabelek *et al.*, 1992) and to compare with the Jurassic Daebo granitic rocks in Korea. The isotopic results for the Jurassic peraluminous Hwacheon granite from this study reveal that it also shows low  $\delta^{18}\text{O}$  values less than 10‰. In this paper, we tried to evaluate the possible causes for the lowering in  $\delta^{18}\text{O}$  values of the Hwacheon granite. Isotopic modelling has been conducted in order to elucidate the nature of fluid-rock interaction of the Hwacheon granite.

## General Geology and Petrography

The Hwacheon granite, situated in the central northern part of the Gyeonggi massif (Fig. 1a), is a member of the peraluminous granitic rocks distributed in east-west direction in Gyeonggi massif. The Jurassic Hwacheon granite and late-Triassic Chuncheon granite located to the south of the Hwacheon granite intruded Precambrian metamorphic rocks of Yongduri gneissic Complex in Gyeonggi massif. The metamorphic rocks surrounding the Hwacheon granite consist of banded biotite gneiss, leucocratic gneiss, quartzite, and amphibolite. Metamorphic rocks to the north of the Hwacheon granite have experienced granulite-facies metamorphisms (Lee *et al.*, 1997; Lee and Cho, 2003)

The petrological and geochemical studies of the Hwacheon granite were documented by Sagong *et al.* (1997). The Hwacheon granite comprises two distinct bodies of Chugongnyeong porphyritic granite

and Yonghwasan mica granite (Sagong *et al.*, 1997). The Yonghwasan body is divided into mica granite in northern part and garnet-bearing mica granite in the southern part depending on the presence of garnet (Sagong *et al.*, 1997). Sagong *et al.* (1997) used the term 'mica granite' because it is not clear whether muscovite is magmatic or not. The Yonghwasan mica granite which has a gradational boundary with garnet-bearing mica granite in field intruded the porphyritic biotite granite containing K-feldspar phenocryst. The relatively small body of biotite monzodiorite which is not described in the previous studies occurs as a small body in Chugongnyeong porphyritic granite. The biotite monzodiorite contains the xenoliths of porphyritic biotite granite, but the relationship with mica granite and garnet-bearing mica granite is not known yet.

Modal compositions of the Hwacheon granite are presented in Table 1. The division of muscovite is accomplished on the basis of textural criteria only under microscope and electron probe analyses are necessary to make sure if all the primary muscovites in Table 1 are magmatic. The Hwacheon granite contains secondary minerals produced during hydrothermal alteration (e.g., muscovite, chlorite, sericite, and epidote and calcite). The volume percents of each secondary mineral are presented in Table 1. The porphyritic biotite granite consists of quartz, plagioclase, alkali feldspar, biotite, muscovite, sericite, chlorite, calcite, zircon, epidote, and opaque minerals. The phenocrysts are 2 cm in average, and up to 4 cm in size, and consist of microcline and perthite. The biotite monzodiorite is medium- to fine-grained, and consists of plagioclase, biotite, quartz, orthoclase, muscovite, zircon, apatite, and opaque minerals.

Mica granite is medium- to coarse-grained, and consists of quartz, plagioclase, alkali feldspar, biotite, muscovite, chlorite, sericite, and opaque mineral. Mica granite contacts garnet-bearing mica granite in the southern margin. Mica granite and garnet-bearing mica granite have similar textures and mineral assemblages excluding the presence of garnet as well as the more muscovite in the garnet-bearing mica granite. The abundance of garnet in the garnet-bearing mica granite tends to decrease gradually toward

**Table 1. Modal compositions of the Hwacheon granite.**

Rock type	Garnet-bearing mica granite					Mica granite			Porphyritic biotite granite		
Sample No.	A-6	B-10	D-1	D-7	I-8	B-3	B-6	E-9	A-15	A-17	A-22
Quartz	35.2	34.7	21.2	32.1	34.2	41.8	46.3	43.1	27.2	26.1	22.0
Plagioclase	28.4	23.2	27.2	23.9	20.4	31.0	16.5	14.5	10.1	13.5	16.4
Orthoclase	6.9	10.3	4.8	6.2	5.4	12.7	10.7	12.4	21.8	12.6	24.4
Microcline	6.8	7.2	23.7	17.2	16.4	6.8	9.4	12.7	21.2	18.6	13.8
Perthite	4.9	4.5	4.4	9.2	4.1	1.4	4.2	6.2	10.4	11.6	15.1
Biotite	6.0	8.1	4.4	3.3	4.7	3.7	3.8	4.4	2.2	1.9	2.8
Muscovite (primary)	2.4	2.9	3.8	1.4	5.1	-	1.1	0.1	1.0	3.0	1.5
Muscovite (secondary)	1.2	1.2	1.2	0.7	1.5	-	1.2	0.9	1.2	1.0	0.7
Garnet	0.3	0.8	0.6	-	1.8	-	-	-	-	-	-
Zircon	0.9	1.2	1.4	0.2	0.4	0.1	-	0.3	0.1	0.2	0.5
Chlorite	3.2	3.4	2.5	2.4	2.3	1.0	3.2	3.7	2.3	2.1	1.0
Sericite	3.8	2.5	4.8	3.4	3.7	0.8	3.1	1.6	2.3	5.2	1.5
Epidote	-	-	-	-	-	0.6	0.4	0.1	0.3	-	0.1
Opaque mineral	-	-	-	-	-	0.1	0.1	-	-	-	0.3
Calcite	-	-	-	-	-	-	-	-	-	4.2	-

the contact boundary with mica granite. Garnet-bearing mica granite tends to contain more and larger muscovite than that in the mica granite, whereas opaque minerals are abundant in mica granite rather than in the garnet-bearing mica granite. Two varieties of garnet occur in the garnet-bearing mica granite. One is anhedral but extremely fractured, coarse-grained (~2 mm) garnet, and contains inclusions such as quartz and feldspars. The other is euhedral, weakly fractured, fine-grained (~0.5 mm) garnet, and rarely contain inclusions.

Sagong *et al.* (1997) have documented that ages of mica granite and garnet-bearing mica granite are  $177.3 \pm 3.8$  Ma and  $133.5 \pm 3.2$  Ma, respectively based on whole-rock Rb-Sr method, but also suggested the possibility that they might have been crystallized 168 Ma ago as a one body. Cho *et al.* (1996) reported  $172 \pm 5$  Ma for the garnet-bearing mica granite on the basis of CHIME (chemical U-Th-total Pb isochron method) age of monazite. Park *et al.* (1997) reported  $151 \pm 4$  Ma for the garnet-bearing mica granite based on K-Ar dating of muscovite from the same sample as Cho *et al.* (1997) used for CHIME.

### Sampling and Analytical methods

Samples were collected from the Hwacheon granite

and surrounding metamorphic rocks in the Hwacheon area (Fig. 1). Whole rock samples were trimmed with a diamond saw to prepare fresh surfaces for analysis. Mineral separates of feldspar, quartz, biotite, hornblende, and garnet for isotopic analysis were extracted from flat slabs by diamond drilling. All sample powders were stored in a vacuum oven about 12 hours at 250°C prior to loading for oxygen and hydrogen isotopic analyses. Samples for oxygen isotopic analyses were prepared using BrF<sub>5</sub> method of Clayton and Mayeda (1963), and oxygen isotopic ratios were determined using Finnigan Mat 252 mass spectrometer at Indiana University. Water for hydrogen isotopic analyses of whole rocks and mineral separates was liberated by melting in vycor tubes using a method modified slightly from that of Vennemann and O'Neil (1993). Hydrogen was produced by heating the liberated water with zinc in sealed pyrex tubes. Water yields were measured by using the ion gauge of the mass spectrometer inlet system, calibrated with a series of water standards. Hydrogen isotopic ratios and water content were measured on VG Prism II stable isotope ratio mass spectrometer at the Korea Basic Science Institute (KBSI). Results are reported in delta notation relative to V-SMOW for O and H. NBS-28 quartz (NIST Reference Material 8546) and NCSU quartz

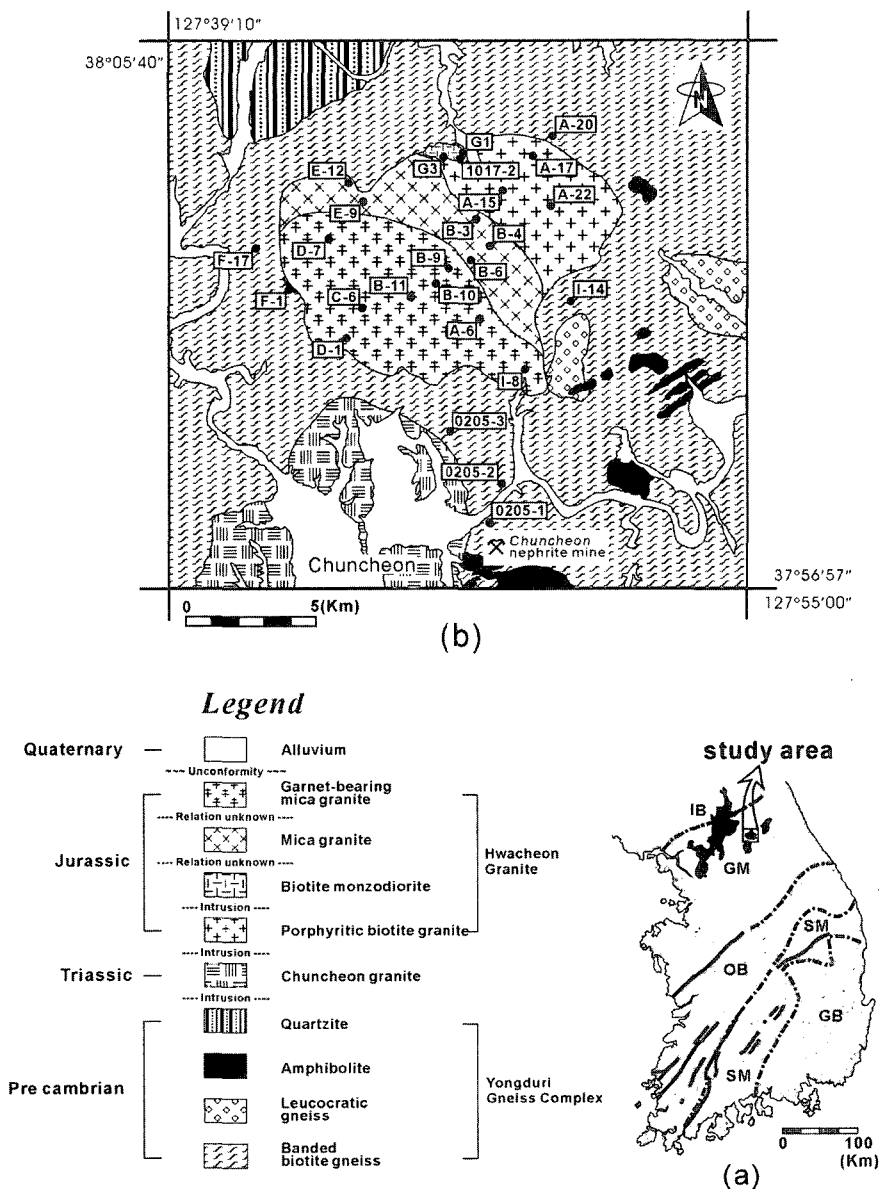


Fig. 1. (a) A simplified geologic map of the Korean peninsula. Location of the study area is shown as a box. Dark colored area represents the distribution of the peraluminous granitic rocks in northern Gyeonggi massif after Sagong *et al.* (1997). Abbreviations: IB, Imjingang Belt; GM, Gyeonggi massif; OB, Okcheon Belt; SM, Sobaeksan massif; GB, Gyeongsang Basin. (b) Geologic map showing the distribution of the Hwacheon granite with sample locations after Sagong *et al.* (1997).

(B. Showers, North Carolina State University) have  $\delta^{18}\text{O}$  values of  $9.6 \pm 0.1$  and  $11.5 \pm 0.1\%$ , respectively, in stable isotope laboratory at Indiana University. The analytical reproducibility is 1 for  $\delta\text{D}$ . The

chemical compositions of chlorite of the Hwacheon granite were measured by an electron microprobe analyzer (CAMECA SX-50) at the KBSI.

## Results and Discussion

Results of stable isotopic measurements are presented in Table 2. Ranges of  $\delta^{18}\text{O}$  values for various minerals from the Hwacheon granite are as

follows: quartz, 8.2-10.6‰; feldspar, 5.8-9.0‰; biotite, 4.3-5.1‰. Whole rock  $\delta^{18}\text{O}$  values for the surrounding banded biotite gneiss range from 8.1 to 9.4‰, and  $\delta^{18}\text{O}$  values of garnet and hornblende from the amphibolite country rock are 5.1 and 5.9‰, respec-

**Table 2. Oxygen and hydrogen isotopic compositions and water content of the Hwacheon granite and surrounding metamorphic rocks.**

Sample No.	$\delta^{18}\text{O}_{\text{Qtz}}$	$\delta^{18}\text{O}_{\text{Fds}}$	$\delta^{18}\text{O}_{\text{Bt}}$	$\delta^{18}\text{O}_{\text{WR}}$	$\delta\text{D}_{\text{WR}}$	$\text{H}_2\text{O}$ (wt.%)
	(‰ vs. V-SMOW)					
<b>Igneous rocks</b>						
<b>Garnet-bearing mica granite</b>						
A-6	10.2	7.9			-113	0.5
B-10	8.4	5.8			-107	0.5
D-1	8.2	7.9			-93	0.59
D-7	10.6	9.0			-104	0.5
I-8	9.8	8.5			-84	
B-9						
B-11						
C-6						
<b>Mica granite</b>						
B-3	8.6	7.7			-112	0.57
B-4					-116	0.5
B-6	9.2	7.6			-110	0.5
E-9	9.2	6.9			-100	0.44
E-12					-95	1.5
<b>Biotite monzodiorite</b>						
1017-2		7.4	4.3		-107 (Bt)	
G1		7.3	5.1		-113 (Bt)	
G3						
<b>Porphyritic biotite granite</b>						
A-15	8.9	7.1			-101	0.5
A-17	9.1	6.0			-95	1.3
A-22	8.2	7.0			-99	0.4
<b>Country rocks</b>						
<b>Banded biotite gneiss</b>						
A-20				8.4	-76	0.9
F-17				8.1	-96	0.8
I-14				8.6	-89	0.9
0205-1				8.5	-93	1.4
0205-2				9.3	-100	0.6
0205-3				9.4	-91	1.1
<b>Amphibolite</b>						
F-1		5.1 (Grt)	5.9 (Hbl)			

Qtz, Fds, Bt, WR, Grt, and Hbl represent quartz, feldspar, biotite, whole rock, garnet, and hornblende, respectively.

tively. Whole rock and biotite  $\delta D$  values for granitic rocks range from -84 to -113‰ and -107 to -113‰, respectively. Whole rock  $\delta D$  values for banded biotite gneiss country rocks range from -76 to -100‰.

#### Potential causes of lowered $\delta^{18}O$ values

$\delta^{18}O$  values of quartz and feldspars of the Hwacheon granite ranging from 8.2 to 10.6‰ and 5.8 to 9.0‰, respectively, are low compared to the 'normal'  $\delta^{18}O$  values ( $> \sim 10$ ‰) for the fresh peraluminous granitic rocks originated from metapelitic protoliths (O'Neil and Chappell, 1977; O'Neil *et al.*, 1977; Masi *et al.*, 1981; Lee *et al.*, 1982; Nabelek *et al.*, 1992). Considering the fact that other constituent minerals such as biotite, muscovite, garnet, chlorite, sericite and epidote (Table 1) tend to prefer  $^{16}O$  to  $^{18}O$  compared to quartz and feldspar (Garlick and Epstein, 1967; Criss, 1999), whole rock  $\delta^{18}O$  values of the Hwacheon granite are not expected to be higher than 10‰.

The low  $\delta^{18}O$  values of the granitic rocks may indicate either crystallization from a low- $^{18}O$  magma or subsolidus isotopic exchange with a low- $^{18}O$  fluid. A low- $^{18}O$  peraluminous magma can be originated from either partial melting of a low- $^{18}O$  peraluminous source rock at depth or assimilation of low- $^{18}O$  rocks attributed to oxygen isotopic exchange with a low- $^{18}O$  fluid, during ascent of a magma.

If low  $\delta^{18}O$  values of the Hwacheon granite result from crystallization from low- $\delta^{18}O$  magma, a question of how source rock with a low- $\delta^{18}O$  value could have been located at depth is raised. The downward infiltration of surface derived fluids to depths of ca. 12 km has been documented (Wickham and Taylor, 1985). Large-scale convection of fluids within the crust has been also suggested (e.g., Etheridge *et al.*, 1983). However, it is generally not accepted that the pervasive circulation of surface derived fluids readily occur below the brittle-ductile transition, because permeability decreases at deep crustal levels (i.e.  $> 10$  km) due to increased ductility prohibits fluid flow during regional metamorphism (Valley and O'Neil, 1982). Using the hornblende geobarometry, Cho and Kwon (1994) documented that the Jurassic Daebo granite in Korea formed at

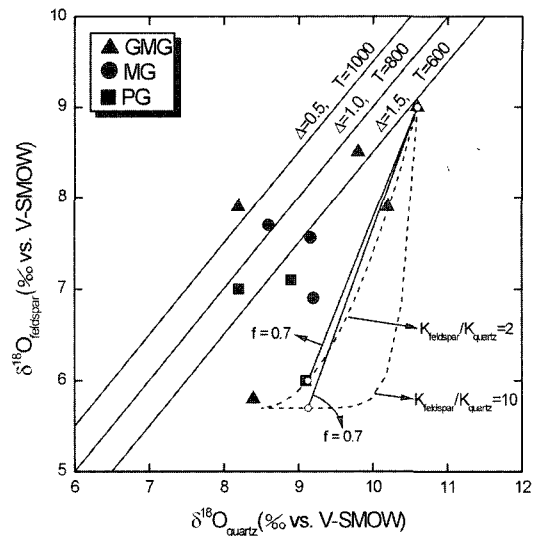


Fig. 2.  $\delta^{18}O_{\text{quartz}}$  vs.  $\delta^{18}O_{\text{feldspar}}$  plots for the Hwacheon granite. Broken lines represent exchange trajectory calculated for 300°C for  $k_{\text{feldspar}}/k_{\text{quartz}}$  values of 2 and 10, respectively.  $f$  represents a constant value for the fractional degree of exchange in quartz toward the final, equilibrium with fluid. Solid lines representing  $f=0.7$  are shown for  $k_{\text{feldspar}}/k_{\text{quartz}}$  values of 2 and 10, respectively. Parallel lines represent mineral pairs in equilibrium at different temperatures. The isotherms are based on Zheng (1993). GMG, garnet-bearing mica granite; MG, mica granite; PG, porphyritic biotite granite.

depth of ca. 12 to 28 km. It is unlikely, therefore, that low- $^{18}O$  characteristics of the Hwacheon granite originated from partial melting of low- $^{18}O$  metapelitic rocks lowered in  $\delta^{18}O$  values due to downward infiltration of surface derived low- $^{18}O$  fluid into deep crustal levels. In addition, the relatively low  $\delta^{18}O$  values of the surrounding banded biotite gneiss (8.1 to 9.4‰) indicate that the possibility of lowering in oxygen isotopic composition during fluid-rock interaction with low- $\delta^{18}O$  fluid. It is not unreasonable to assume, therefore, that subsolidus isotopic exchange with external fluids during fluid-rock interaction was a major process to produce low and variable oxygen isotopic compositions of the Hwacheon granite.

In order to evaluate if the variable and low  $\delta^{18}O$  values of the Hwacheon granite reflect magmatic record or subsolidus isotopic exchange with external fluids during water-rock interaction, we plotted  $\delta^{18}O$  values of feldspar against the  $\delta^{18}O$  values of quartz

in Fig. 2. The isotherms in Fig. 2 are based on Zheng (1993). The quartz-feldspar pairs with  $\Delta$  values  $> 1.5$  indicate that temperatures are too low to be magmatic suggesting subsolidus isotopic exchange with externally derived fluid at low temperature environment.

However, the samples plotted parallel to the isotherms with the relatively low  $\Delta_{\text{quartz-feldspar}}$  are probably explained by variable amounts of exchange with low- $^{18}\text{O}$  external fluid during long period of hydrothermal activities. Re-equilibration of the feldspar and quartz could have occurred during prolonged heating of pre-existing disequilibrium arrays, which results in decrease of the magnitude of  $\Delta_{\text{quartz-feldspar}}$  and moves samples to the left in Fig. 2 towards the high-temperature equilibrium fractionation lines.

#### Fluid-rock Interaction

Gregory and Criss (1986) extensively described the

isotopic effects of fluid-rock interactions in closed and open systems. In order to estimate possible sources of fluid involved in hydrothermal alteration of the Hwacheon granite, isotopic modelling has been conducted and the result is plotted in Fig. 2. In the modelling, we assumed that initial oxygen isotopic compositions of quartz and feldspar are 10.6 and 9.0‰, respectively which are the highest values measured from the Hwacheon granite. Hydrothermal alteration of the Hwacheon granite at low temperature seems to cause formation of secondary hydrous minerals (e.g., chlorite, epidote, muscovite, and sericite) and the accompanied isotopic exchange is likely to result in  $\Delta_{\text{quartz-feldspar}}$  values  $> 1.5$ ‰. We estimated chlorite formation temperature ranging from 170 to 330°C from Cathelineau and Nieva (1985) thermometer (Table 3) and used 300°C for isotopic simulation. The ratios of oxygen isotopic exchange rate of feldspar to quartz ( $k_{\text{feldspar}}/k_{\text{quartz}}$ ) of 2 and 10 were used

**Table 3. Representative electron microprobe analysis (in wt.%) of chlorites of the Hwacheon granite.**

Rock type	Garnet-bearing mica granite					Mica granite				Porphyritic biotite granite		
Sample No.	D-1	I-8				B-3	B-6			E-9	A-15	
SiO <sub>2</sub>	31.16	22.66	21.75	21.19	21.61	21.47	21.39	21.39	21.87	26.19	25.67	26.93
TiO <sub>2</sub>	0.95	0.03	0.05	0.10	0.04	0.07	0.13	0.04	0.11	0.77	0.04	0.34
Al <sub>2</sub> O <sub>3</sub>	21.67	21.58	21.37	21.81	21.13	21.26	21.08	21.04	20.57	18.96	20.70	19.34
FeO	30.66	43.86	44.72	42.44	42.93	41.42	44.07	43.59	44.35	38.39	26.75	24.24
MnO	0.79	0.96	1.07	1.37	1.64	0.76	0.95	0.80	1.37	0.77	1.23	1.40
MgO	1.25	0.32	0.28	0.29	0.32	2.11	0.80	0.84	0.78	0.91	13.76	14.91
CaO	0.20	0.03	0.04	0.01	0.04	0.05	0.07	0.07	0.04	0.35	0.01	0.34
Na <sub>2</sub> O	0.12	0.00	0.02	0.05	0.10	0.00	0.12	0.02	0.01	0.98	0.00	0.02
K <sub>2</sub> O	3.69	0.03	0.05	0.03	0.08	0.04	0.04	0.01	0.02	1.15	0.00	0.23
Total	90.49	89.46	89.34	87.29	87.90	87.18	88.65	87.80	89.13	88.47	88.15	87.76
Number of cations on the basis of 28 oxygens												
Si	6.564	5.221	5.069	5.018	5.102	5.047	5.020	5.054	5.114	5.950	5.448	5.679
Al(IV)	1.436	2.779	2.931	2.982	2.898	2.953	2.980	2.946	2.886	2.050	2.552	2.321
Al(VI)	3.944	3.079	2.937	3.104	2.982	2.935	2.850	2.912	2.782	3.027	2.626	2.485
Ti	0.150	0.006	0.009	0.017	0.007	0.012	0.023	0.006	0.019	0.132	0.006	0.055
Fe <sup>2+</sup>	5.401	8.451	8.714	8.405	8.475	8.140	8.649	8.612	8.672	7.295	4.748	4.274
Mn	0.141	0.187	0.212	0.274	0.328	0.151	0.189	0.159	0.271	0.148	0.221	0.250
Mg	0.392	0.110	0.096	0.103	0.114	0.741	0.281	0.297	0.272	0.309	4.355	4.688
Ca	0.044	0.008	0.009	0.003	0.011	0.013	0.017	0.018	0.011	0.085	0.001	0.078
cations	18.074	19.841	19.976	19.906	19.916	19.993	20.009	20.005	20.028	18.997	19.957	19.829
Fe/(Fe+Mg)	0.932	0.987	0.989	0.988	0.987	0.917	0.969	0.967	0.970	0.959	0.522	0.477
T(°C)*	169.96	312.57	328.66	334.12	325.20	331.06	333.92	330.30	323.93	235.11	288.41	263.90

\*Chlorite-formation temperature calculated from Cathelineau and Nieva (1985)

because quartz is relatively resistant to oxygen isotopic exchange (Cole and Ohmoto, 1986). Isotopic modelling result indicates that fluid with relatively low  $\delta^{18}\text{O}$  value below  $\sim 1\text{‰}$  might have been involved in the subsolidus isotopic exchange of the Hwacheon granite. Oxygen isotopic composition of fluid ( $< \sim 1\text{‰}$ ) is estimated from the best fit of the horizontal part of the exchange trajectories to the lowest  $\delta^{18}\text{O}$  value of feldspars in the Hwacheon granite. However, due to the fact that the fluid may have been isotopically modified during the fluid-rock interaction by oxygen isotopic exchange with the granite or with metapelitic country rocks, this value can not be considered to represent the primary isotopic signature of the fluid. In order to figure out source of fluid lowered  $\delta^{18}\text{O}$  values of the Hwacheon granite, we used hydrogen isotopic composition of the rocks.  $\delta\text{D}$  values of whole rocks and biotite of the Hwacheon granite range from  $-84$  to  $-113\text{‰}$  and  $-107$  to  $-113\text{‰}$ , respectively.

The relatively low  $\delta\text{D}$  values can result from crystallization from a D-depleted magma formed due to magma exsolution (Nabelek *et al.*, 1983; Taylor *et al.*, 1983; Brigham and O'Neil, 1985), or subsolidus isotopic exchange with a low-D fluid. Taylor *et al.* (1983), Nabelek *et al.* (1983), and Brigham and O'Neil (1985) have documented that low  $\delta\text{D}$  values in igneous rocks can result from degassing of a D-enriched vapor phase rather than isotopic exchange with a low- $^{18}\text{O}$  fluid. Low  $\delta\text{D}$  rocks which record D-enriched vapor-phase exsolution generally must exhibit a trend of decreasing  $\delta\text{D}$  with decreasing  $\text{H}_2\text{O}$  content. For the Hwacheon granite,  $\delta\text{D}$  values show no well-defined trend with water content (Fig. 3), which argues against crystallization from a low- $\delta\text{D}$  magma resulted from magma degassing as a mechanism for the low-D granitic rocks.

It is more likely, therefore, that the relatively low  $\delta\text{D}$  values of the Hwacheon granite were attributed to hydrogen isotopic exchange with low-D fluid of meteoric origin. From the stable isotopic studies of the Chuncheon nephrite deposits located to the south of the Hwacheon granite, Yui and Kwon (2002) suggested that meteoric water with  $\delta^{18}\text{O}$  value of  $\sim -13\text{‰}$  and  $\delta\text{D}$  value between  $-96$  and  $-83\text{‰}$  has

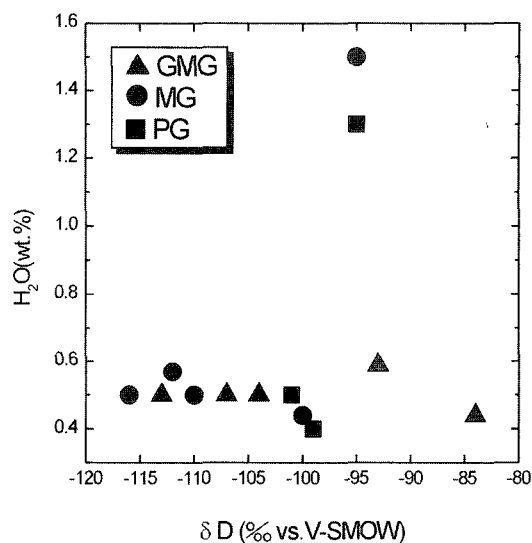


Fig. 3.  $\text{H}_2\text{O}$  (wt.%) content vs.  $\delta\text{D}_{\text{whole rock}}$  (‰ vs. V-SMOW) for the Hwacheon granite. Symbols are the same as in Fig. 2.

been involved in the formation of the nephrite deposits, which is considered to be generated at 230 to 260 Ma (Park and Noh, 2000). Likewise, low- $^{18}\text{O}$  and low-D fluid of meteoric origin might have been involved in the hydrothermal alteration of the Hwacheon granite and surrounding country rocks. However, oxygen isotopic compositions of meteoric fluid must have been modified as a result of fluid-rock interaction during hydrothermal circulation for a relatively long period of time in the study area. On the contrary, however, hydrogen isotopic composition of the circulating meteoric water has been much less affected and not experienced modification in  $\delta\text{D}$  value during fluid-rock interaction because water contains much more hydrogen in it relative to rocks.

Assuming that the fluid flux is large compared to the mineral-fluid isotopic exchange rates, it is possible to estimate fluid/rock ratios from the steep slope of the disequilibrium array of open system. We applied open system behavior to the estimation of the nature of the isotopic exchange of the Hwacheon granite because isotopic exchange trajectories must show the negative slope in  $\delta$ - $\delta$  plot for a closed system (Gregory and Criss, 1986; Criss, 1999), which is not the case of the Hwacheon granite. The slope of the disequilibrium array of the



Hwacheon granite is about 1.45. Therefore, a constant value for the fractional degree of exchange in quartz toward the final, equilibrium with fluid ( $f_{\text{quartz}}$ ) is over about 0.7. From the open system isotopic fluid/rock equation of Taylor (1977):

$$\text{water/rock} = -\ln(1 - f)$$

the effective water/mineral ratio for quartz is calculated to be at least 1.2. From the  $f_{\text{quartz}}$  value, the  $f_{\text{feldspar}}$  value can be calculated from the equation (Gregory and Criss, 1986):

$$f_{\text{feldspar}} = 1 - (1 - f_{\text{quartz}})^{k_{\text{feldspar}}/k_{\text{quartz}}}$$

For the  $k_{\text{feldspar}}/k_{\text{quartz}}$  value of 2 and 10,  $f_{\text{feldspar}}$  can be computed to be 0.910 and 0.99994, respectively, and water/rock for feldspars is 2.4 and 11.7, respectively. Assuming that the Hwacheon granite simply consists of quartz and feldspar, it is estimated that the overall bulk effective fluid/rock ratio was below ~6.

## Conclusions

Oxygen and hydrogen isotopic studies reveal that both  $\delta^{18}\text{O}$  and  $\delta\text{D}$  values of the Jurassic Hwacheon granite are characterized by low values compared to 'normal' value for the peraluminous granitic rocks. These characteristics are similar to other Jurassic Daebo granites (Kim, 1991). Low and variable  $\delta^{18}\text{O}$  values of the Hwacheon granite primarily resulted from subsolidus isotopic exchange with a low- $\delta^{18}\text{O}$  and low-D fluid of meteoric origin. Isotopic exchange at solid state occurred for a long period from high temperature to relatively low temperature. Isotopic modelling result renders that a relatively low- $^{18}\text{O}$  fluid below  $\sim 1\%$  has been involved in the hydrothermal alteration and isotopic exchange under high fluid/rock ratio ( $< \sim 6$ ). The fluid responsible for alteration of the Hwacheon granite and surrounding country rocks has experienced a modification of oxygen isotopic composition as a result of fluid-rock interaction by passing through a long pathway.

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## References

- Brigham, R.H. and O'Neil, J.R., 1985, Genesis and evolution of water in a two-mica pluton: a hydrogen isotope study. *Chem Geol*, 49, 159-177.
- Cathelineau, M. and Nieva, D., 1985, A chlorite solid solution geothermometer. The Los Azufres (Mexico) geothermal system. *Contrib. Mineral. Petrol.* 91, 235-244.
- Chappell, B. W. and White, A. J. R., 1974, Two contrasting granitic types. *Pacific Geol.*, 8, 173-174.
- Cho, D.-L. and Kwon S.-T., 1994, Hornblende geobarometry of the Mesozoic gneissoids in South Korea and the evolution of crustal thickness. *Jour. Geol. Soc. Korea*. 30, 41-61.
- Cho, D.-L., Suzuki, K., Adachi, M. and Chwae, U., 1996, A preliminary CHIME age determination of monazites from metamorphic and granitic rocks in the Gyeonggi massif, Korea. *J. Earth and Planet. Sci. Nagoya Univ.*, 43, 49-65.
- Clayton, R. N. and Mayeda, T. K., 1963, The use of bromine pentafluoride in the extraction of oxygen from oxides and silicates for isotopic analysis. *Geochim. Cosmochim. Acta*, 72, 43-52.
- Cole, D.R. and Ohmoto, H., 1986, Kinetics of isotopic exchange at elevated temperatures and pressures. In: Valley, J.W., Taylor, H.P., O'Neil (eds.). *Stable isotopes in high temperature geological processes*, *Rev. in Mineralogy*, 16, Min. Soc. Am., 91, 41-90.
- Criss, R.E., 1999, *Principles of stable isotope distribution*, Oxford University Press, 254 p.
- Etheridge, M. A., Wall, V. J. and Vernon, R. H., 1983, The role of the fluid phase during regional metamorphism and deformation. *Journal of Metamorphic Geology*, 1, 205-226.
- Garlick, G.D. and Epstein, S., 1967, Oxygen isotope ratios in coexisting minerals of regionally metamorphosed rocks. *Geochim. Cosmochim. Acta* 31, 181-214.
- Gregory, R.T., and Criss, R.E., 1986, Isotopic exchange in open and closed system. In: Valley, J.W., Taylor, H.P., O'Neil (eds.). *Stable isotopes in high temperature geological processes*, *Rev. in Mineralogy*, 16, Min. Soc. Am., 91, 91-127.
- Kim, K.H., 1991, *Isotope Geology*, Mineumsa, 552 p.
- Lee, D.E., Friedman, I., and Gleason, J.D., 1982, The oxygen isotope composition of granitoid and sedimentary rocks of the southern Snake Range, Nevada. *Contributions to Mineralogy and Petrology*, 79, 150-158

- Lee, S. R. and Cho, M., 2003, Metamorphic and tectonic evolution of the Hwacheon granulite Complex, central Korea: Composite P-T path resulting from two distinct crustal-thickening events. *Journal of petrology*, 44, 197-225.
- Lee, S. R., Cho, M., Cheong, C.-S. and Park, K.-H., 1997, An early Proterozoic Sm-Nd age of mafic granulite from the Hwacheon area, South Korea. *Geoscience Journal*, 1, 136-142.
- Masi, U., O'Neil, J.R., and Kistler, R.S., 1981, Stable isotope systematics in Mesozoic granites of central and northern California and southwestern Oregon. *Contributions to Mineralogy and Petrology*, 76, 116-126.
- Nabelek, P. I., O'Neil, J. R. and Papike, J.J., 1983, Vapor Phase exsolution as a controlling factor in hydrogen isotope variation in granitic rocks: the North Peak granitic stock, Utha. *Earth Planet. Sci. Lett.*, 66, 137-150.
- Nabelek, P.I., Russ-Nabelek, C., and Haeussler, G.T., 1992, Stable isotope evidence for the petrogenesis and fluid evolution in the Proterozoic Harney Peak leucogranite, Black Hills, South Dakota, *Geochimica et Cosmochimica Acta*, 56, 403-417.
- O'Neil, J.R. and Chappell, B.W., 1977, Oxygen and hydrogen isotope relations in the Berridale Batholith, southeastern Australia. *Journal of the Geological Society of London*, 133, 554-571.
- O'Neil, J. R., Shaw, S.E., Flood, R.H., 1977, Oxygen and hydrogen isotope compositions as indicators of granite gneiss in the New England Batholith, Australia. *Contrib. Mineral. Petrol.*, 62, 313-328.
- Park, K. H., Lee, B. J., Cho, D. L. and Kim, J. B., 1997, Explanatory text of the geological map of Hwacheon sheet (scale 1:50,000). Korea Inst. Geol. Mining and Materials.
- Park, K.-H. and Noh, J. H., 2000, Geochemical study on the genesis of Chuncheon nephrite deposits. *Jour. Petrol. Soc. Korea*, 9, 53-69.
- Sagong H., Kwon, S.-T., Jeon, E.-Y. and Mertzman, S. A., 1997, Petrology and geochemistry of the Hwacheon granite. *Journal of the Geological Society of Korea*, 33, 99-110. (in Korean with English abstract)
- Taylor, H. P. Jr., 1977, Water/rock interactions and the origin of H<sub>2</sub>O in granitic batholiths. *J. Geol. Soc. Lond.*, 133, 509-558.
- Taylor, B.E., Eichelberger, J.C. and Westrich, H. R., 1983, Hydrogen isotopic evidence of rhyolitic magma degassing during shallow intrusion and eruption. *Nature*, 306, 541-545.
- Valley, J. W. and O'Neil, J. R., 1982, Oxygen isotope evidence for shallow emplacement of Adirondack anorthosite. *Nature*, 301, 226-228.
- Vennemann, T. W. and O'Neil, J. R., 1993, A simple and inexpensive method of hydrogen isotope and water analyses of minerals and rocks based on zinc reagent. *Chem. Geol. (Isotope Geosci. Sec.)*, 103, 227-234.
- Wickham, S. M. and Taylor H. P. Jr, 1985, Stable isotopic evidence for large-scale seawater infiltration in a regional metamorphic terrane; the Trois Seigneurs Massif, Pyrenees, France. *Contrib. Mineral. Petrol*, 91, 122-137.
- Yui, T.-F. and Kwon, S.-T., 2002, Origin of a Dolomite-related jade deposit at Chuncheon, Korea. *Econ. Geol.*, 97, 593-601.
- Zheng, Y.-F., 1993, Calculation of oxygen isotope fractionation in anhydrous silicate minerals. *Geochim. Cosmochim. Acta*, 57, 1079-1091.

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