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Oxygen Isotope Study of Mulgeum, Yangseong, Maeri and Kimhae Iron Ore Deposits in Gyeongnam Province, Korea

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Abstract: Mulgeum, Yangseong, Maeri and Kimhae iron ore deposits in Gyeongnam Province are hydrothermal skarn type magnetite ore deposits in propylitized andesitic rock near the contact with Cretaceous Masanite. Symmetrical zoned skarns are commonly developed around the magnetite veins. The skarn zones away from the vein are quartz-garnet skarn, epidote skarn and epidote-orthoclase skarn. Oxygen isotope analyses of coexisting minerals from andesitic rock, Masanite and major skarn zones, and of magnetite, hematite and quartz were conducted to provide the information on the formation temperature, the origin and the evolution of the hydrothermal solution forming the iron ore deposits. Becoming more distant from the ore vein, temperatures of skarn zones represent the decreasing tendency, but most $\delta^{18}O$ and $\delta^{18}O_{H2O}$ values of skarn minerals represent no variation trend, and also the values are relatively low. Judging from all the isotopic data from the ore deposits, the major source of hydrothermal solution altering the skarn zones and precipitating the ore bodies was magmatic water derived from the deep seated Masanite. This high temperature hydrothermal solution rising through the fissures of propylitized andesitic rock was mixed with some meteoric water, and occurred the extensive isotopic exchange with the propylitized andesitic rock, and formed the skarns. During these processes, the temperature and $\delta^{18}O_{H2O}$ value of hydrothermal solution were lowered gradually. At the main stage of iron ore precipitation, because all the alteration was already finished, the new rising hydrothermal solution formed only the magnetite ore without oxygen isotopic exchange with the wall rock.

Key words: iron ore deposits, oxygen isotope analyses, formation temperature, origin and evolution of hydrothermal solution

INTRODUCTION

Mulgeum, Yangseong, Maeri and Kimhae iron ore deposits in the south-eastern Gyeongnam Province are emplaced in the propylitized andesitic rocks which are in contact with or adjacent to Cretaceous Masanite.

There have been some previous studies about petrology, geologic structure and ore deposits in this area (Lee and Kim, 1964; Lee and Park, 1976; Hwang, 1957; Hwang and Kim, 1962; Kim and Kang, 1969; Kim, 1980; The Korean Mining Promotion Company, 1972, 1973, 1975, 1976, 1977, 1978, 1979, 1980, 1982; Lee, 1972, 1974; Lee and Lee, 1982; Miyazawa, 1978), but detailed studies about skarn and ore minerals, fluid inclusion, and ore genesis of major ore deposits were mainly carried by Woo *et al.* (1982) and Woo (1983).

In this paper, oxygen isotope analyses have been undertaken in an attempt to identify the formation temperatures of skarn and ore zones, and the origin and evolution of the hydrothermal solution related to the formation of iron ore deposits in the study area.

GENERAL GEOLOGY

The geology of Mulgeum, Yangseong, Maeri and Kimhae iron ore deposits area is mainly composed of granites, andesitic rocks, rhyolites and a small amount of gabbro and sedimentary rocks. Among the above rocks, those most importantly related to the origin of the iron ore deposits in the studied area are the andesitic rocks which form the wall rock of the ore deposits and the bordering granites

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(Fig. 1).

The andesitic rocks are widely scattered in the Gyeongnam Province. The colors of these rocks are greenish grey, dark grey and greyish black. The andesitic rocks bear phenocrysts of feldspar and hornblende whose diameter range from 0.2 cm to 0.5 cm. Under the microscope, the andesitic rocks show porphyritic texture. The phenocrysts consist mainly of plagioclase and hornblende with some orthoclase and biotite. Plagioclase phenocrysts show Albite and Albite-Carlsbad twin and zoned structure. The composition of the plagioclase ranges from An_{40} to An_{50}. The andesitic rocks usually contain some epidote pseudomorphs after feldspar and uralite pseudomorphs after clinopyroxene with small amount of calcite and pyrite. This suggests that this andesitic rock was already propylitized before the formation of iron ore deposits. Kwon (1979), Park (1980) and Kim (1980) reported that fresh andesitic rock is scarcely found in the Gyeongsang basin because of severe alteration. The andesitic rocks near the iron ore deposits in the study area commonly include much more orthoclase than those of the areas which are far from the ore deposits. In fact, the orthoclase content suddenly increases closer to the iron ore veins. These phenomena are considered to be resulted from plagioclase alteration to orthoclase by potassic metasomatism related to skarnization of the ore deposits. Here, the andesitic rocks near the iron ore veins are tentatively classified as the altered andesitic

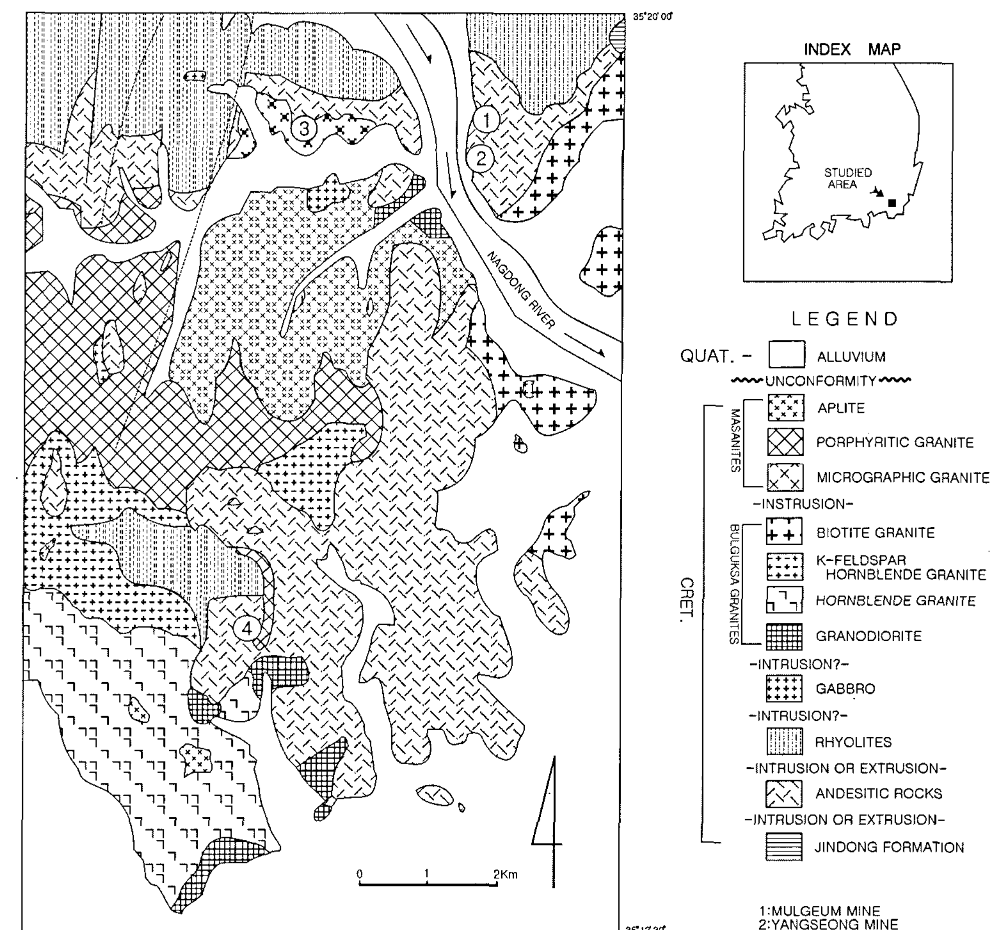

Fig. 1. Geologic map of Kimhae-Mulgeum area.

rock and the intensely altered andesitic rock.

The bordering granites are Masanite. Masanites have micrographic, porphyritic and aplitic textures, and consist of quartz, orthoclase, plagioclase, biotite, hornblende and magnetite. Many authors (Lee and Park, 1976; Park and Park, 1980; Lee, 1981; Jin, 1981; Tsusue *et al.*, 1981; Lee and Lee, 1982) suggested that Masanites are closely related to the ore genesis of many ore deposits in the Gyeongnam Province.

CHARACTERISTICS OF IRON ORE DEPOSITS

Mulgeum, Yangseong, Maeri and Kimhae ore deposits are skarn-type magnetite veins emplaced in propylitized andesitic rocks. The relevant igneous rock of these ore deposits as well as the wall rock, the skarn and ore minerals and the morphology of ore bodies are similar (Woo, 1983). The skarn minerals are mainly composed of garnet, quartz, epidote, orthoclase with a small amount of clinopyroxene and actinolite. Skarn zoning was developed on the boundary between the ore body and the wall rock. These skarns exhibit nearly symmetrical zoning on both sides of the ore body. In general, the sequence from magnetite vein to andesitic rock is quartz-garnet skarn, epidote skarn, epidote-orthoclase skarn and intensely altered andesitic rock. Small-scale skarn zoning occurs around the many veinlets in the adits of the ore deposits and also has the same sequence as the large scale zoning (Fig. 2).

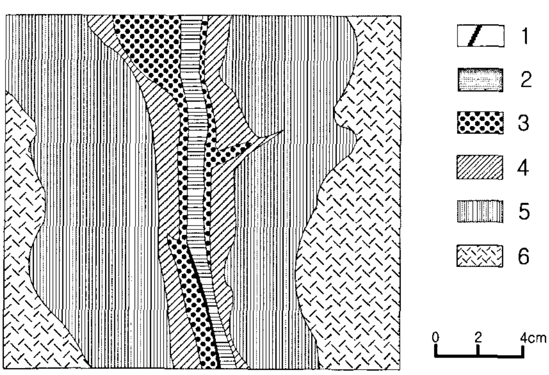

Fig. 2. The mode of occurrence of zoned skarn (-3L, E-cross, 324 m, Mulgeum iron ore deposits).
1. magnetite 2. quartz-garnet 3. epidote 4. epidote-orthoclase 5. intensely altered andesitic rock 6. altered andestic rock.

Quartz-garnet skarn is composed chiefly of aggregates of quartz and garnet crystals. Average grain size is about 0.4 mm, but may go up to about 2 mm in diameter.

Epidote skarn is composed chiefly of epidote and some quartz reaching about 0.5 mm in diameter. Some minerals may be associated; magnetite, sphene, garnet, chlorite, orthoclase, plagioclase, apatite and amphibole. Epidotes in this skarn range from Ps (content of pistacite) 26.9 to Ps 30.9.

Epidote-orthoclase skarn is composed chiefly of orthoclase and epidote. Average grain size is about 0.5mm but the phenocryst of orthoclase may go up to about 5mm in length. The epidotes in this skarn range from Ps 21.1 to Ps 29.7. Some minerals may be associated: magnetite, hematite, sphene, garnet, chlorite, plagioclase, actinolite and apatite.

Intensely altered andesitic rock chiefly occurs on the outer zone of epidote-ortoclase skarn, but in some cases is omitted, and intensely altered andesitic rock was in direct contact with the magnetite veins. This rock is mainly composed of orthoclase and plagioclase with small amount of epidote, magnetite, amphibole and calcite. Epidotes in this rock range from Ps 28.9 to Ps 32.6.

Altered andesitc rock chiefly occurs on the outer zone of intensely altered andesitic rock. This rock is mainly composed of plagioclase and amphibole with small amount of epidote, magnetite, clinopyroxene and uralite. Epidotes in this rock range from Ps 23.4 to Ps 28.6.

The ore minerals are composed chiefly of magnetite with small amounts of hematite, pyrite, chalcopyrite, pyrrhotite, sphalerite, galena and tetrahedrite.

OXYGEN ISOTOPE STUDIES

Analytical Procedures

As shown in Tables 1 and 2, samples used in this

Table 1. Oxygen isotope analyses of coexisting minerals in andesitic rock, Masanite and skarns.

Sample No.	Sample name	Sample location	Coexisting mineral	$\delta^{18}O$(‰)
PA-1	andesitic rock	far eastward from the ore deposits	plagioclase (An = 50%) magnetite	−0.19 −5.18
Gr-1	Masanite (micrographic granite)	near the Maeri ore deposits	quartz K-feldspar	+6.31 +5.32
AA-1	altered andesitic rock	Mulgeum ore deposits	plagioclase (An = 40%) epidote	+0.38 −3.77
La-1	intensely altered andesitic rock	Mulgeum ore deposits	K-feldspar epidote	+1.23 −0.74
Or-1	epidote-orthoclase skarn	Mulgeum ore deposits	K-feldspar epidote	+1.72 −0.83
ME-1	epidote skarn	Mulgeum ore deposits	quartz epidote magnetite	+2.37 −2.79 −8.79
YE-1	epidote skarn	Yangseong ore deposits	quartz epidote	+3.56 −2.52
RE-1	epidote skarn	Maeri ore deposits	quartz epidote	+6.82 +0.12
KE-1	epidote skarn	Kimhae ore deposits	quartz epidote	+2.81 −3.17
MG-1	quartz-garnet skarn	Mulgeum ore deposits 3rd level	quartz garnet	+0.27 −5.12
RG-1	quartz-garnet skarn	Maeri ore deposits	quartz garnet	+4.09 −0.23

study were mainly collected from the coexisting mineral pairs of andesitic rock, Masanite, and each skarn zone, and magnetite and hematite of ore zone and vuggy quartz of post-mineralizing zone from the ore deposits.

Standard mineral separating procedures were used, including hand picking, magnetic and heavy liquid techniques. Additional procedures were applied to improve the purity of some of these mineral separates. From quartz-feldspar-calcite mixture, quartz separates were prepared using a H_2SiF_6 and HCl solution.

After identification of purity of minerals by X-ray diffraction, the separated minerals were isotopically analyzed. Isotopic analyses were made using standard techniques (Clayton and Mayeda, 1963).

All oxygen isotopic data are reported in δ-notation as per mil deviations from the SMOW standard (Craig, 1961). Results are reproducible to +0.1 per mil or less for all samples. Isotopic measurements were made using a Nuclide RMS 6-60, 6 inch double collecting mass spectrometer at Case Western Reserve University.

Isotopic Results

All oxygen isotopic results of silicate and oxide minerals are listed in Tables 1 and 2.

Andesitic rock: This rock is relatively fresh andesitic rock collected from the Kijang area which is located in the far eastern part of this study area. The $\delta^{18}O$ values of coexisting plagioclase (An = 50%) and magnetite in this rock are −0.19‰ and −5.18‰ respectively (Table 1). The $\delta^{18}O$ value of plagioclase is lower than feldspar in typical andesite (about +6‰, Taylor, 1974). It is considered because this andesitic rock was propylitized.

Oxygen isotope fractionation between plagioclase and magnetite yield the temperature of 631°C, using the equations of Bottinga and Javoy (1973). It can be considered the temperature at the time of propylitization of this andesitic rock.

We can calculate the oxygen isotope composition of the water, $\delta^{18}O_{H2O}$, at the time of propylitization, using the temperature 631°C and the equations of Bottinga and Javoy (1973), for α plagioclase-water (T), and α magnetite-water (T), and we obtained $\delta^{18}O_{H2O}$ = +0.32‰.

Masanite (micrographic granite): Because of its timely and spacely possibility of relevant igneous rock to the ore deposits and its possible role in the genesis of the ore-forming fluids, we analyzed the coexisting quartz and K-feldspar in Masanite near the Marie ore deposits.

The $\delta^{18}O$ values of quartz and K-feldspar are +6.31‰ and +5.32‰ respectively. The $\delta^{18}O$ values of quartz and K-feldspar are lower than quartz (+8 ~ +14‰) and feldspar (+7 ~ +13‰) in typical plutonic granites (Taylor, 1974). Oxygen isotopic fractionation between quartz and K-feldspar in Masanite yield the temperature of 717°C using the equations of Bottinga and Javoy (1973) for α quartz-K-feldspar (T). Also, we calculated the oxygen isotope composition of the water, $d^{18}O_{H2O}$, at the time of granite crystallization, using the temperature 717°C and the equations of Bottinga and Javoy (1973) for α quartz-water (T) and α K-feldsapr-water (T), and we obtained $\delta^{18}O_{H2O}$ = +5.83‰. The oxygen isotopic composition of this water is in the range of typical magmatic water (+5.5 ~ +10.0‰, Taylor, 1979).

Altered and intensely altered andesitic rock: The $\delta^{18}O$ values of the coexisting plagioclase (An = 40%) and epidote in the altered in the altered anedsitic rock are +0.38‰ and −3.77‰, and the coexisting K-feldspar and epidote in the intensely altered andesitic rocks are +1.23‰ and −0.74‰ respectively. Oxygen isotopic fractionation between feldspar and epidote using the equations of Mattews *et al.* (1983), yield the temperatures of 115°C in altered andesitic rock and of 461°C in intensely altered andesitic rock respectively.

Epidote-orthoclase skarn: The $\delta^{18}O$ values of the coexisting K-feldsapr and epidote in the epidote-orthoclase skarn of the Mulgeum ore deposits are +1.72‰ and −0.83‰ respectively.

The temperatures 372°C by oxygen isotopic fractionation between K-feldspar and epidote using the equation of Mattews *et al.* (1983) is geologically reasonable. Using this temperature and equation of O'Neil-Taylor (1967), we calculated the oxygen isotopic composition of the water. The results are −1.86‰.

Epidote skarn: In the Mulgeum ore deposits, the coexisting quartz, epidote and magnetite of the epidote skarn were analyzed. In the Yangseong, Maeri and Kimhae ore deposits. only the coexisting quartz and epidote were analyzed. As shown in Table 1, the $\delta^{18}O$ value of magnetite in Mulgeum ore deposits is −8.79‰ and the $\delta^{18}O$ values of quartz and epidote range from +2.37‰ to +6.82‰ and from +0.12‰ to −3.17‰ respectively, but except the values of Maeri ore deposits (Qt: +6.82‰ Epi: +0.12‰), all have nearly similar values.

Oxygen isotope fractionations between quartz-magnetite and quartz-epidote yield the temperatures of 362 ~ 433°C (Mulgeum), 312 ~ 326°C (Yangseong), 284 ~ 295°C (Maeri), and 317 ~ 328°C (Kimhae), using the equations of Bottinga and Javoy (1973), and Matthews and Schliestedt (1984), for α quartz-magnetite (T) and α quartz-epidote (T) respectively.

The temperatures of three other ore deposits are relatively somewhat lower than those of the Mulgeum ore deposits. Judging from other geological data, it is considered that the formation temperature of this skarn is 433°C.

Here, using the temperature 433°C and the equations of Matsuhisa *et al.* (1979) for α quartz-water (T), we calculated the oxygen isotopic composition of the water at the time of this skarn formation. The value of $\delta^{18}O_{H2O}$ is −0.97‰.

Quartz-garnet skarn: In the Mulgeum and Maeri ore deposits, the coexisting quartz and garnet mineral pairs were analyzed (Tabel 1). Using the diagram of Kiffer (1982), oxygen isotope fractionations between quartz and garnet yield the temperatures 527°C (Mulgeum) and 587°C (Maeri) respectively.

Table 2. Oxygen isotope analyses and $\delta^{18}O_{H2O}$ values of magnetite, hematite and vuggy quartz.

Sample No.	Sample name	Sample location	Mineral	$\delta^{18}O$ (‰)	$\delta^{18}O_{H2O}$ (‰)
		Mulgeum ore deposits			
MM-1	ore vein	1st level main vein	magnetite	+0.53	+6.16
MM-2		3rd level west vein	magnetite	+0.06	+5.69
MM-3		main vein	magnetite	+0.27	+5.89
MM-4		east 2nd vein	magnetite	−1.68	+3.95
MM-5		east 5th vein	magnetite	−10.04	−4.41
MM-6		6th level main vein	magnetite	−2.52	+3.11
MM-7		8th level main vein	magnetite	−2.19	+3.44
MH-1		E4E5 vein	hematite	−12.10	−6.23
YM-1		Yangseong ore deposits	magnetite	−8.93	−3.30
RM-1		Maeri ore deposits	magnetite	−2.52	+3.11
KM-1		Kimhae ore deposits	magnetite	−0.19	+5.44
MQ-1	vuggy quartz	Mulgeum ore deposits	quartz	+2.57	−1.36
RQ-1		Maeri ore deposits	quartz	+4.66	−1.19
KQ-1		Kimhae ore deposits	quartz	+3.13	−1.90

Using the above temperatures and the equations of Matsuhisa *et al.* (1979) and Bottinga and Javoy (1973) for α quartz-water (T), we calculated the oxygen isotopic composition of the water. The results are −1.79 ~ −2.44‰ (Mulgeum), and +2.25 ~ +2.46‰ (Maeri) respectively.

Magnetite: The $\delta^{18}O$ values of 10 magnetite samples from the ore veins of four ore deposits range from −10.04‰ to +0.53‰, but except two values of Mulgeum east fifth vein (−10.04‰), and Yangseong main vein (−8.93‰), all have relatively narrow range values (+0.53 ~ 2.52‰). The trend of the values is roughly decreased from main vein to east vein, and from upper level to lower level (Table 2). Referring to the temperatures of quartz-garnet skarn, we can assume that the magnetites were precipitated at about 600°C.

Using this temperature and the equation of Bottinga and Javoy (1973) for α magnetite-water (T), we calculated the oxygen isotopic composition of the ore fluid. The results range from −4.41‰ to +6.16‰, but except those of Mulgeum east fifth vein (−4.41‰) and Yangseong vein (−3.30‰), the others range from +3.11‰ to +6.16‰.

Hematite: The $\delta^{18}O$ value of the only one hematite from Mulgeum ore deposits is −12.10‰.

Assuming this hematite was precipitated at about 550°C, we calculated the oxygen isotopic composition of the ore fluid, using the equation of Bottinga and Javoy (1973) for α magnetite-water (T). The result is −6.23‰.

Vuggy quartz: In the Mulgeum, Maeri and Kimhae ore deposits, the $\delta^{18}O$ values of vuggy quartz of post mineralization stage were analyzed. The results are +2.57‰, +4.66‰ and +3.13‰ respectively (Table 2).

The formation temperatures of vuggy quartz by fluid inclusion study were reported as 402°C (Mulgeum), 331°C (Maeri) and 360°C (Kimhae) respectivety (Woo, 1983).

So, in this vuggy quartz zone, we calculated the oxygen isotopic composition of the water using the fluid inclusion temperature data and the equation of Matsuhisa *et al.* (1979) for α quartz-water (T). The $\delta^{18}O_{H2O}$ values related to the formation of vuggy quartz in Malgeum, Maeri and Kimhae ore deposits are −1.36‰, −1.19‰ and −1.90‰ respectively.

DISCUSSION

The analyzed $\delta^{18}O$ values of the minerals from the ore deposits in this area, except the minerals of Masanite, are relatively low. The calculated temperatures from the oxygen isotopic fractionation of the coexisting mineral pairs in the each alteration zone, represent the decreasing tendency from the ore vein to the propylitized andesitic rock. It can be considered because the early stage alteration products were completely changed to the later stage alteration products on the same place, by successively altering action of the hydrothermal solution, and the temperature of the early stage hydrothermal solution was high at first, but during the diffusion through the andesitic rock from the center of the fissure to the margin, its temperature decreased.

On the other hand, the calculated $\delta^{18}O_{H2O}$ values represent nearly no variation trend through the skarn zones, except those of magnetites. Because the $\delta^{18}O_{H2O}$ values of magnetites of ore zone are more similar to those of Masanite than those of propylitized andesitic rock, it can be considered that the major source of the hydrothermal solution formed this ore deposits, was Masanite, and some local meteoric water was mixed.

The caculated $\delta^{18}O_{H2O}$ values of the skarn minerals of alteration zone represent the relatively low values. It can be considered because there have been extensive isotopic exchange of the magmatic origin hydrothermal fluid with the propylitized andesitic rock, causing a lowering of $\delta^{18}O_{H2O}$ in the alteration zone.

Among the $\delta^{18}O_{H2O}$ values of magnetites in the ore zone, those of Mulgeum east fifth magnetite vein (−4.41‰), and Yangseong magnetite (−3.30‰) is especially low, and referring to these values are close to those of hematite (−6.23‰), it is considered because these magnetites were somewhat changed to hematites by martitization. And, the $\delta^{18}O_{H2O}$ values of first level main vein magnetite (+6.16‰) and third level main vein magnetite (+5.89 ~ +5.90‰) of Mulgeum ore deposits, are somewhat higher than those of Masanite (+5.83‰). It can be considered that the major origin of the hydrothermal solution was the higher $\delta^{18}O_{H2O}$ value-magmatic water possibly derived from the more deeper seated Masanite. And, at the main stage of iron ore precipitation, because all the alteration was already finished, the oxygen isotopic exchange with the wall rock was nearly not occurred, so most $\delta^{18}O_{H2O}$ values of magnetite are similar to those of magmatic water.

CONCLUSIONS

The oxygen isotopic analyses were utilized to acquire a general picture of the genesis of the iron ore deposits of Mulgeum area as followings;

1. The major source of the hydrothermal solution altering the skarn zones and precipitating the ore bodies was magmatic water derived from deep seated Masanite.
2. This high temperature hydrothermal solution rising through the fissures of propylitized andesitic rock was mixed with some meteoric water, and occurred the extensive isotopic exchange with the propylitized andesitic rock, and formed the skarns. During these processes, the temperature and $\delta^{18}O_{H2O}$ value of hydrothermal solution were lowered gradually.
3. At the main stage of iron ore precipitation, because all the alteration was already finished, the new rising hydrothermal solutions formed only the magnetite ore without oxygen isotopic exchange with wall rock.

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