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Lead Isotopic Study on the Dongnam Fe-Mo Skarn Deposit

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ABSTRACT: In Dongnam area, Cretaceous igneous rocks, such as diorite, porphyritic granite, and quartz porphyry intruded Paleozoic sedimentary rocks, such as Myobong slate and Pungchon limestone. The Dongnam Fe-Mo skarn deposits were imposed on the diorite(endoskarn) and the Myobong slate(exoskarn). The ore deposits consist mainly of magnetite and molybdenite with small amounts of sulfides, such as galena, sphalerite, pyrite, chalcopyrite, and pyrrhotite. The igneous rocks show nearly constant $^{206}Pb/^{204}Pb$(18.80~19.06) and $^{207}Pb/^{204}Pb$(15.71~15.72) ratios. Their $^{207}Pb/^{204}Pb$ ratios higher than the typical ratios of orogene suggest that the igeneous rocks were formed from lower crust(or mantle) - derived magma excessively contaminated by upper crustal materials such as high radiogenic Precambrian basement rocks. The lead isotopic compositions of the igneous rocks, the Pungchon limestone, and the ore minerals show a well defined linear array in $^{206}Pb/^{204}Pb$ - $^{207}Pb/^{204}Pb$ plot. The lead isotopic compositions of the igneous rocks are similar to those of magnetite and galena, which were formed at early skarn stage and significantly lower than those of altered quartz porphyry, molybdenites, and pyrite, which were formed at late epithermal alteration stage. Considering the systematic variation of the lead isotopic compositions in the ore minerals according to hydrothermal stages, the variation may be due to a relative variation in surrounding rock(Pungchon limestone) involvement in hydrothermal ore solution leaching the surrounding rock. Therefore, the variation of the lead isotopic compositions in ore minerals can be modeled in terms of the mixing of the leads derived from the igneous rocks as low radiogenic source and the surrounding rock(Pungchon limestone) as high radiogenic source.

INTRODUCTION

Major problems related to metallogeny are the origin of metal elements, the mineralization age, and the physico-chemical conditions of ore formation. The lead isotope method is considered as a powerful tool for solving the first two problems (Andrew et al., 1984; Fletcher and Farquhar, 1982; Oh et al., 1989). Galena can be considered to retain the initial isotopic composition of lead at the time of mineralization, because of the far greater abundance of lead over uranium and thorium. That is, the lead isotopic composition of galena is frozen and not changed by radioactive decay of uranium and thorium to lead since the ore formation. Because of its common nature, galena offers the informations on the lead isotopic systematics at the time of mineralization. Other sulfide minerals with high Pb/U and Pb/Th ratios also can be applied in similar way to galena.

In this study, the origin of the hydrothermal ore fluid of the Dongnam Fe-Mo skarn deposit has been deduced by using lead isotopic systematics. The Dongnam Mine is located around the Hambaeg basin in the north-eastern part of South Korea, where several skarn ore deposits are distributed (Fig. 1). This mine was exploited for magnetite from open-cuts in the early 1960's but it is currently closed. The ore minerals related to the skarnization are mainly magnetite and molybdenite with small amounts of galena and sphalerite. Manganese ores were also produced by supergene alteration.

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Previously, Chang(1983) studied on the mineralogy of the manganese ores and Seo et al.(1983) reported on the petrology and geochemistry of the skarn. More recently, petrological and geochemical features on the skarnization and the mineralization have been studied by Cheong(1988).

GEOLOGY

The geology of the study area comprises Cambro-Ordovician Myobong slate, Pungchon limestone, and Hwajeol Formation and Cretaceous intrusive rocks, such as diorite, porphyritic granite, quartz porphyry, and acidic dyke (Fig. 2). The main strike and dip of sedimentary rocks are N 10°~50° E and 10°~ 20° SE, respectively, and they vary irregularly at the contact zone with the igneous rocks.

The Myobong slate is the oldest formation in the study area. This rock is mainly distributed in the south-western part of the mine and also occurs as xenolith in the diorite at B pit. The slate is dark gray or dark green to green.

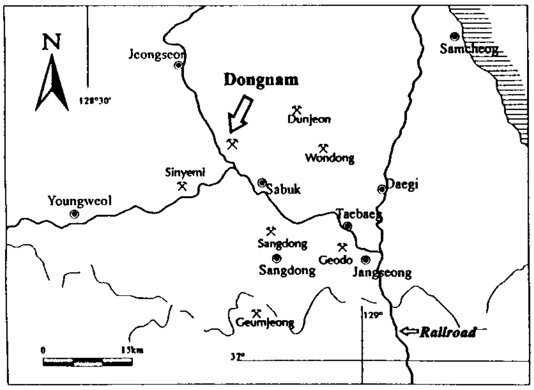

Fig. 1. Location of the Dongnam Mine.

The matrix of the slate consists of fine-grained quartz and feldspar, chlorite, and sometimes pyrite. The slate was partly replaced with the skarn at the contact with the quartz porphyry. The Pungchon limestone overlies conformably the Myobong slate. The limestone consists mainly of gray, dark gray, and milky white limestone units. The limestone was recrystallized near the contact with the diorite but not skarnized. The Hwajeol formation consists of shale, vermicular-shaped limestone, and gray limestone. The skarns were not developed in this formation.

The diorite intruded the Pungchon limestone in the central part of the study area. The lens-shaped diorite stock was divided into two parts by the fault of NNE direction. The diorite was replaced partly with the skarn. The diorite was also changed into pseudodiorite at the contact zone between the fault and the Pungchon limestone. According to the drilling exploration carried out by Korea Institute of Energy and Resources(Seo et al., 1983), the width and length of the diorite were about 120 m and 800 m, respectively. The diorite chiefly consists of plagioclase and amphibole with minor amounts of orthoclase, quartz, biotite, pyroxene, sphene, and apatite. Plagioclase corresponds to labradorite (An_{56}). Amphibole occurs as subhedral crystal and its composition is identified as tremolite to actinolite by electron microprobe analysis. Orthoclase is anhedral and occurs as interstitial grains. Pyroxene shows rather clear cleavages and coexists with plagioclase. Biotite is brown to dark brown in color and sometimes altered to chlorite along its cleavage. By modal analysis and normative classification, the diorite is monzo-diorite and classified as calc-alkaline and I-type granitoid by geochemical parameters, such as ACF diagram, $Fe^{3+}/(Fe^{2+} + Fe^{3+})$, $K/(Na+K)$, and $Al_2O_3/(Na_2O + K_2O + CaO)$ (Cheong, 1988).

The porphyritic granite intruded the Pungchon and Hwajeol Formations in the southeastern part of the study area. The rock is coarse-grained and shows porphyritic texture with the phenocrysts of K-feldspar from about 0.5 cm to 1.5 cm in size.

The quartz porphyry near the fault zone shows fractured quartz crystals resulted from mechanical stress. This indicates that the fault occurred after the intrusion of the diorite and the quartz porphyry. The quartz porphyry intruded the previous igneous rocks and skarn zones. It shows variable texture and mineralogy. The quartz porphyry distributed in the southern part of the area contains orthoclase phenocrysts. The apophysis of the quartz porphyry in the northern part of the area shows flowbanded texture and also porphyritic texture with large phenocrysts of quartz. At the extremity of A open pit (Fig. 2), disseminated molybdenites and molybdenite veinlets are observed in the quartz porphyry, which has been intensively sericitized and pyritized. Most of quartz porphyries were widely but weakly altered.

SKARN AND ORE DEPOSIT

The skarns were imposed on the diorite (endoskarn) and the Myobong slate (exoskarn) along the contacts between both of the diorite and the slate. The textural characteristics of the host rocks were nearly obliterated. The structure of the skarns is very complex, because skarn zones were superimposed repeatedly on one another. The skarns occur as massive bodies and veins. The massive skarns preserve the relicts of original lithologies, such as the subhedral-granular texture of the diorite and the bedding texture of the slate. On the basis of mineral assemblages, the skarn zones from outer zones to inner zones are as follows; in endoskarn, altered diorite zone, pyroxene zone, epidote-pyroxene zone, epidote zone, and garnet zone and in exoskarn, altereds late zone, pyroxene zone, and epidote zone. The garnet zone and the epidote zone are the inner most zones in the endoskarn and the exoskarn, respectively. The typical structure of the endoskarn zones is schematically shown in Fig. 3.

The Dongnam skarn ore deposits consist mainly of magnetite and molybdenite with small amounts of galena and sphalerite. Other sulfides include pyrite, chalcopyrite, arsenopyrite, and pyrrhotite. Manganese oxides were formed near surface without any relation with the skarnization process. Magnetite mineralization was superimposed mainly on massive garnetite. Magnetites coexist with garnets and they occur as elongated massive ore bodies, which are isolated from each other. Moly-

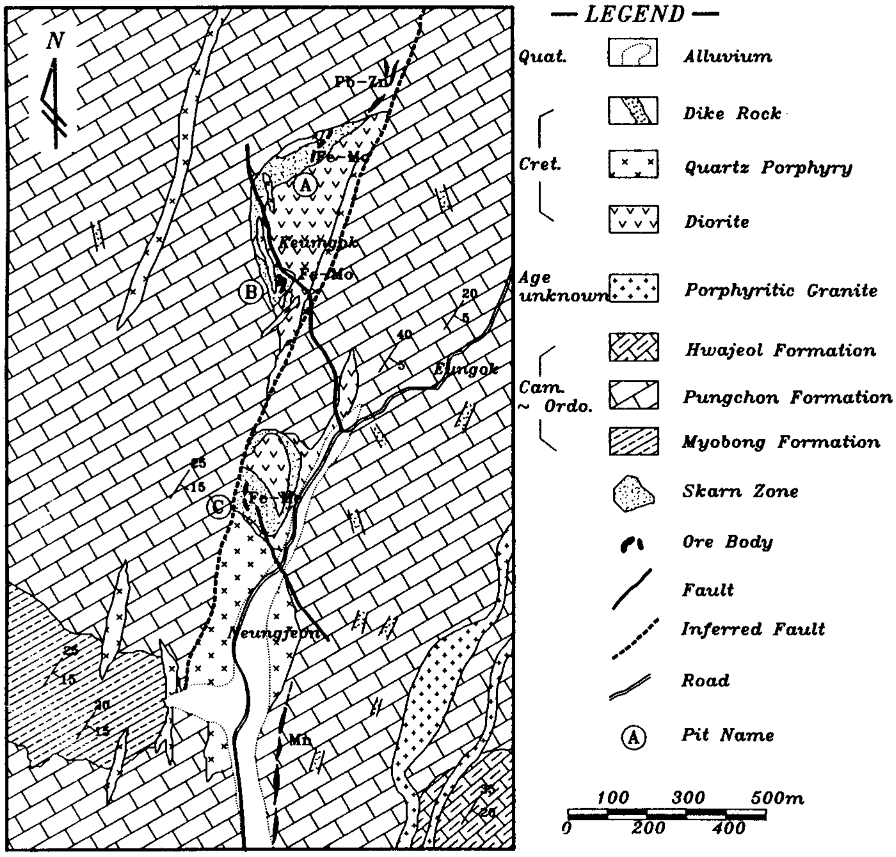

Fig. 2. The geologic map of the study area.

bdenites (MoS_2) occur as veins or disseminated aggregates, and they were precipitated with pyrite and other sulfides in calcite veins, which cut across the previous skarns and the diorite, and also in the secondary alteration zones formed along the fissures in the skarns. Molybdenites were closely related with secondary alteration zones formed after early skarn stage. They were enriched around calcite veins in the fissures of the diorite and they were nearly scarce or absent in the absence of calcite veins. The textures and distribution patterns of the skarns and the ore minerals suggest that the magnetites were precipitated simultaneously with or slightly later than early skarn minerals such as garnet or pyroxene, and that the molybdenites were probably precipitated from late epithermal fluid enriched in CaO, which infiltrated along the fissures in the previous skarns and the diorite.

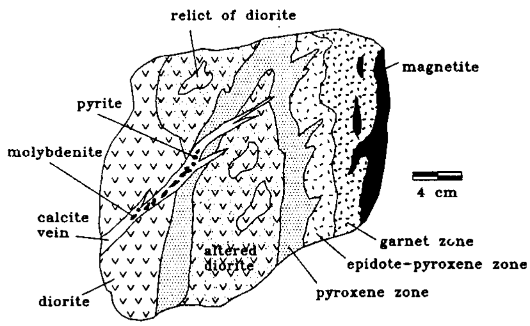

Fig. 3. Schematic diagram showing endoskarn zones from outer to inner zones: altered diorite zone, pyroxene zone, pyroxene-epidote zone, and garnet zone.

LEAD ISOTOPES

Samples

To investigate the source of metals in the hydrothermal system, lead isotopic compositions and U, Th and Pb concentrations were determined in the ore minerals, such as magnetite, molybdenite, pyrite, and galena. The Pungchon limestone and the intrusive rocks, such as diorite, porphyritic granite, and quartz porphyry were also analyzed. All analyzed samples were collected from

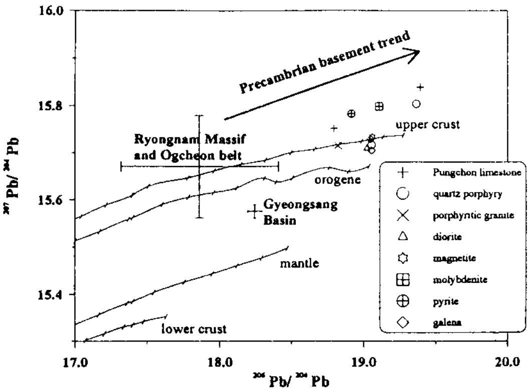

Fig. 4. $^{207}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ plot. The present lead isotopic compositions of mantle, upper crust, lower crust and orogene are from Zartman and Haines (1988). Lead isotopic compositions of the granitic rocks in the Gyeongsang Basin and the Ryongnam Massif - Ogcheon Belt are taken from Kwon (1992). Tick marks along each curve indicate progressively older time in 0.1-Ga increments.

the open pits(A, B, and C in Fig. 2).

ANALYTICAL PROCEDURE

About 100~150 milligrams of each sample except galena were dissolved in mixed acid(HF, HNO_3, and $HClO_4$=3 : 1 : 1), using hot plate set to 150℃. The dissolved samples were dried 3 times with 6N HCl, and dissolved in about 0.5 ml of HBr. After centrifuging, the samples were loaded on anion exchange resin (AG 1X 8, Cl^- form, 100~200#), and lead was extracted using HBr and HCl. Galena samples were dissolved in aqua regia (HCl : HNO_3=3 : 1), using hot plate, and they were loaded directly on the filament without the purification procedure. Lead was loaded on a Re filament, using the standard silicagel-phosphoric acid technique. Isotopic ratios were measured using VG 54-30 mass spectrometer at K.B.S.C.(Korea Basic Science Center). The data were normalized using the NBS SRM 981 standard. Used acid and water were purified by quartz sub-boiling system and two bottle sub-boiling system. Total blank level was about 1 ng for lead. U, Th, and Pb concentrations were determined by ICP-MS(Inductively Coupled Plasma Mass Spectrometer of VG PQ2+ model) at K.B.S.C.. Samples for ICP-MS measurements were dissolved by the reflux procedure after Park et al.(1992).

RESULT AND DISCUSSION

The lead isotopic compositions and concentrations of U, Th, and Pb of the samples are presented in Table 1. Fig. 4 illustrates $^{206}Pb/^{204}Pb$ versus $^{207}Pb/^{204}Pb$ relation for the samples. The characteristic values of the lead isotopes in specific tectonic environments (Zartman and Haines, 1988) and in the granitic rocks taken from Gyeongsang Basin, Ryongnam Massif, and Ogcheon Belt (Kwon, 1992) were also plotted in Fig. 4 for comparison.

Table 1. U, Th, and Pb abundances and lead isotopic compositions of samples collected from the Dongnam skarn deposit.

Sample No.	$^{207}Pb/^{204}Pb$	$^{206}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	U(ppm)	Th(ppm)	Pb(ppm)
Pungchon limestone						
PC 01	19.393 ± 1	15.839 ± 1	38.121 ± 3	0.8	1.4	18.1
PC 02	18.796 ± 3	15.751 ± 2	38.890 ± 6	0.1	1.0	1.9
Quartz porphyry						
B 16	19.057 ± 1	15.716 ± 2	38.429 ± 8	1.8	14.3	12.0
B 36	19.366 ± 1	15.804 ± 1	37.987 ± 2	3.6	12.2	45.4
Porphyritic granite						
F 8	18.824 ± 1	15.714 ± 1	38.699 ± 3	4.2	24.3	35.7
Diorite						
DO 06	19.024 ± 2	15.708 ± 2	38.360 ± 5	1.6	9.5	18.5
Magnetite	19.061 ± 5	15.734 ± 4	38.470 ± 10	0.6	2.0	3.5
Molybdenite	19.110 ± 1	15.798 ± 1	38.288 ± 4	0.6	14.7	398.8
Pyrite	18.917 ± 1	15.783 ± 1	38.595 ± 2	0.4	4.7	88.5
Galena 1	19.057 ± 1	15.708 ± 1	37.896 ± 3			
Galena 2	19.050 ± 5	15.729 ± 4	38.018 ± 11			

Notes : 1. Pb isotope ratios are corrected by NBS 981 standard, which are $^{208}Pb/^{204}Pb$ = 36.617 ± 29(N = 16.2σ S.E.), 2. Errors in isotope ratios are reported as 2σ S.E., 3. U, Th, and Pb concentrations are determined by ICP-MS.

In Fig. 4, two Pungchon limestones show rather different lead isotopic compositions each other and they have higher $^{207}Pb/^{204}Pb$ ratio the galena and magnetite.

The igneous rocks, except the quartz porphyry sample B36, show nearly constant $^{206}Pb/^{204}Pb$(18.82~19.06) and $^{207}Pb/^{204}Pb$(15.71~15.72) ratios. The porphyritic granite is the least radiogenic among the analyzed samples. Two quartz porphyries show different lead isotopic compositions each other. In case of the quartz porphyry sample B36, it is more plausible to explain its higher lead isotopic composition relative to the other quartz porphyry as the contamination due to the alteration by epithermal fluid. As stated earlier, most of quartz porphyry was widely but weakly altered.

The igneous rocks in the study area have higher $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratios, compared with the granitic rocks from Ryongnam Massif and Ogcheon Belt. The igneous rocks have also higher $^{207}Pb/^{204}Pb$ ratio than orogene rocks(Fig. 4). Generally, $^{207}Pb/^{204}Pb$ ratio is a sensitive indicator of the amount of upper crustal materials incorporated in primary magma. Accordingly,

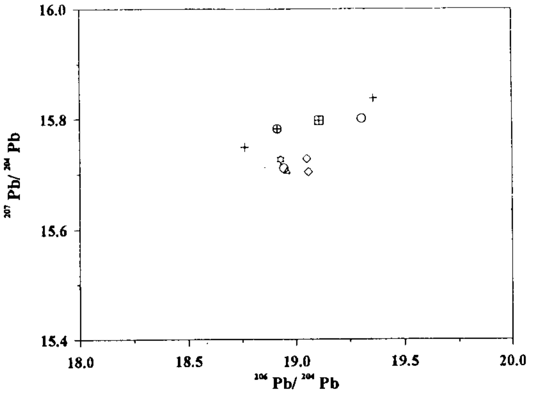

Fig. 5. The lead isotopic compositions of ore minerals and those of rocks recalculated from the mineralization age of 76 Ma. Calculation is carried out using the present lead isotopic compositions, U, Th, and Pb concentrations. Symbols are the same as in Fig. 4.

the lead isotopic characteristics of the igneous rocks in the study area can be explained as the involvement of upper crustal materials. This means that the igneous rocks might be formed from the magma derived from partial melting of upper crustal materials or lower crust(or mantle)-derived magma contaminated by upper crustal materials. According to Park et al.(1993), the lead isotopic compositions of the Precambrian granitic gneisses in the Taebaegsan area were very variable and radiogenic ($^{206}Pb/^{204}Pb$ = 18.418~57.352, $^{207}Pb/^{204}Pb$ = 15.741 ~20.496). Accordingly, the magma derived from the deep-seated the Precambrian gneissic basement rocks might be more radiogenic than the igneous rocks in the study area, under the condition that the isotopic equilibrium was maintained during the partial melting. Consequently, the partial melting of upper crust materials is not appropriate to the origin of the igneous rocks in the area. Considering that the igneous rocks of the study area are more radiogenic than those from Ryongnam Massif and Ogcheon Belt, the former might be more contaminated by upper crustal materials than the latter. Therefore, the upper crustal materials such as the Precambrian gneissic basement rocks high radiogenic in the Taebaegsan area may be regarded as one of possible contaminants for the formation of the igneous rocks in the area.

The lead isotopic compositions of the igneous rocks and the limestone at the time of the mineralization can be obtained from the magnetite mineralization age of 76 Ma(Park et al., 1986). Park et al. dated the phlogophite coexisting with magnetite to 76 Ma by K-Ar method. The correction for radiogenic decay is a function of Pb, U, and Th contents of the samples and of the time elapsed since the closure of the system. Of course, the U/Pb and Th/Pb ratios of the samples could have been changed by the migration of these elements. But, the mineralization age is so young that the correction for radiogenic decay can be executed without large error as long as the samples do not have abnormally high parent/daughter element ratio. In Fig. 5, the lead isotopic compositions of the rocks were recalculated from the time of magnetite mineralization. In Fig. 4 and Fig. 5, magnetite and galena show relatively constant lead isotopic compositions. Their $^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ ratios are similar to those of the porphyritic granite and the diorite. Molybdenite and pyrite, which were formed at late epithermal alteration stage, have higher $^{207}Pb/^{204}Pb$ ratios than galena and magnetite coexisting with garnet which is early skarn mineral. These lead isotopic characteristics shown in the ore minerals can be interpreted in terms of the mixing of leads derived from different sources.

Two Pungchon limestone samples, which are one of the surrounding rocks, show different isotopic compositions each other. The lead concentration of the limestone sample PC01 is strongly higher than that of the limestone sample PC02. The sample PC01 is the most radiogenic in both $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratios among the analysed samples. Sometimes, a limestone shows the variation with a wide range in lead isotopic compositions, and it can be used for U-Pb or Pb-Pb dating (Kwon and Park, 1993; Moorbath et al., 1987; Park and Cheong, 1993).

Possible sources of the leads could be the surrounding rocks of the igneous rocks and the hydrothermal ore solution differentiated from the igneous rocks. The porphyritic granite and the diorite are the least radiogenic and they show the lowest $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratios. These igneous rocks can be regarded as low end member in the mixing relation. The high end member can be considered as the Pungchon limestone.

Considering that the lead isotopic compositions of the altered quartz porphyry and molybdenite formed at late epithermal alteration stage approach to those of the limestone, the systematic increase of the lead compositions from the igneous rocks to the limestone through the altered quartz porphyry and the ore minerals may be due to a relative increase in surrounding rock(limestone) involvement in the magma and hydrothermal ore solution or due to the difference in the lead isotopic compositions according to the alteration stages of hydrothermal fluids leaching various surrounding rocks which had different compositions in place. The ore minerals have higher $^{207}Pb/^{204}Pb$ ratios for a given $^{206}Pb/^{204}Pb$ ratios. This suggest that they received an isotopically different lead compositions during the mixing.

CONCLUSIONS

1. In the Dongnam mine, Cretaceous igneous rocks, such as diorite, porphyritic granite, and quartz porphyry intruded Paleozoic sedimentary rocks, such as the Myobong slate and the Pungchon limestone. The Fe-Mo skarn deposits were developed on the diorite(endoskarn) and the Myobong slate(exoskarn). They show distinctly zonal structures.

2. The igneous rocks have nearly constant $^{206}Pb/^{204}Pb$ (18.80~19.06) and $^{207}Pb/^{204}Pb$(15.71~15.72) ratios. They show higher $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratios than the granitic rocks from Ryongnam Massif and Ogcheon Belt. This suggests that the igneous rocks of the study area might be derived from lower crust(or mantle)-derived magma excessively contaminated by upper crustal materials such as the Precambrian gneissic basement rocks high radiogenic in the Taebaegsan area. The lead isotopic compositions of the igneous rocks are similar to those of magnetite and galena, which were formed at early skarn stage, whereas significantly lower than those of molybdenite and pyrite, which were formed at late epithermal alteration stage.

3. The lead isotopic compositions of the igneous rocks, the Pungchon limestone, and the ore minerals can be modeled in terms of the mixing of the leads derived from the igneous rocks as low radiogenic source and the surrounding rock(Pungchon limestone) as high radiogenic source. That is, the systematic increase in the lead isotopic compositions may be due to a relative increase in surrounding rock(Pungchon limestone) involvement in the magma and the hydrothermal fluids.

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동남 스카른 광상에 대한 납 동위원소 연구

장호완 · 정창식 · 박희인 · 장병욱

요 약 : 동남 스카른 광상은 백악기에 고생대 퇴적암류를 관입한 섬록암에 의해 형성되었다. 스카른의 모암은 섬록암과 묘봉 슬레이트인데 스카른 누대구조에서 모암의 종류에 따라 각각 특징적인 광물공생군의 체계적 변화를 관찰할 수 있다. 섬록암, 화강반암, 석영반암으로 구성된 화성암류들은 플룸보텍토닉 모델의 조산대에 비해 높은 $^{207}Pb/^{204}Pb$ 비를 보이는데 이러한 현상은 하부지각 (또는 맨틀)에서 유래된 마그마가 상부지각물질에 의해 심히 혼화되어진 것으로 해석되며 이는 영남육괴와 옥천계에 분포하는 화강암류가 보여주는 특징과 유사하다.

현재의 납 동위원소 비와 우라늄, 토륨, 납 농도로부터 구한 광화작용 당시(76 Ma)의 화성암 및 풍촌석회암의 납 동위원소 비는 비교적 일정하지만 광화작용 후기로 갈수록 광석광물들의 납 동위원소 비가 높아진다. 초생 스카른 시기에 형성된 자철석과 방연석의 납 동위원소 비는 화성암류의 것과 거의 유사하지만 후기 열수변질시기에 형성된 휘수연석, 황철석, 그리고 열수변질된 석영반암의 납 동위원소 비는 풍촌석회암의 비에 접근하고 있다. 이는 서로 다른 기원을 가진 납들이 열수 변질시기에 따라 그 혼화의 정도가 달라지게되어 광석광물들의 납 동위원소 비에 차이가 있게 된 것으로 여겨진다.

광석광물의 납 동위원소 비의 변화특징에 의하면 섬록암과 화강반암을 형성한 마그마로부터 분화된 낮은 납 동위원소 비를 갖는 열수용액과 높은 납 동위원소 비를 갖는 주변모암(풍촌석회암)이 혼화의 기원물질로 판단된다.