Hydrothermal Alteration Related to Cretaceous Felsic Magmatism in the Gusi Mine, Southern Korea

Hi-Soo Moon*, Yul Roh*, In-Joon Kim**, Yungoo Song* and Hyun Koo Lee***

ABSTRACT: Gusi pyrophyllite deposit is located in the Haenam volcanic field in the southwestern part of the Korea Peninsula. This area is known for the occurrences of pyrophyllite, alunite and dickite. This volcanic field is composed of andesite, rhyolite and pyroclastic rocks of late Cretaceous age. The pyroclastic rocks are hydrothermally altered to phyrophyllite and kaolin minerals forming the Gusi deposits. The hydrothermally altered rock can be classified into the following zones on the basis of their mineral assemblages: quartz, pyrophyllite, dickite and illite-smectite zones, from the centre to the margins of the alteration mass. Such mineral assemblages indicate that the country rocks, most of which are the lower Jagguri Tuff, were altered by strongly acidic hydrothermal solutions with high aqueous silica and potassium activity and that the formation temperature of pyrophyllite is higher than 265°. The mechanism of the hydrothermal alteration is considered to be related to felsic magmatism.

INTRODUCTION

In general, some clay deposits are considered to be formed by acidic hydrothermal solutions at shallow levels(Iwao, 1972). but the alteration mechanisms are not simple. Some deposits are considered to have been formed by hydrothermal alteration associated with igneous intrusive activity(e. g., Kamitani, 1974), but in most other deposits, the hydrothermal alteration appears to be independent of intrusive activity. Furthermore, the mechanism of alteration zones around ores in felsic volcanic rocks has been extensively described by many workers(Matsumoto, 1968; Meyer et al., 1968; Corn, 1975; Utada, 1980). The alteration zones have been defined by the change of the mineral assemblages. In general, from the core of the ore or the vein to outer unaltered rocks, successive zones involve alteration of the felsic volcanic rocks to alunite, pyrophyllite, dickite, nacrite, kaolinite, sericite(illite)and montmorillonite.

Meyer and Hemley(1967) have classified alteration in most aluminosilicate rocks into prop-

A mumber of pyrophyllite, dickite and alunite deposits occur widely as a result of the hydrothermal alteration of the felsic volcanic rocks of Cretaceous age around the Haenam area, southwestern part of Korea(Cho and Moon, 1976). Gusi mine is one of the large pyrophyllite deposits. Though surface geology around the deposit is clear, mineral assemblages and alteration zones are developed vertically rather than horizontally. The mechanism of the hydrothermal alteration has not been clearly recognized.

The scope of the present study was limited to outline the vertical or horizontal distribution of the alteration minerals which could be identified

ylitic, intermediate argillic, advanced argillic, sericitic and potassic types. Intermediate argillic type is characterized by the presence of important amounts of kaolinite, montmorillonite or amorphous clay, principally replacing plagioclase ; this involves appreciable leaching of calcium, sodium and magnesium. Advanced argillic type is characterized by all feldspar being converted to dickite, kaolinite, pyrophyllite, diaspore, alunite or other aluminum-rich phases. These types are commonly observed as an epithermal alteration product. It indicates the extreme conditions of very low pH and relatively oxidizing environments(Hemley et al., 1969; Hemley et al., 1980 ; Rye et al., 1989) and may be associated with high-grade gold, silver and base-metal mineralization underlying and/or overlying these alteration zones(Wang, 1972; Ashley, 1974; Stoffregen, 1987; Hedenquist 1987; Berger et al., 1989; Hedenquist and Aoki, 1990).

Department of Geology, Yonsei University, 134 Shinchon-dong, Seodaemun-Ku, Seoul 120-749, Korea.

Geological Institute, Faculty of Science, University of Tokyo, 7-3-1 Hongo, Bunkyo-Ku, Tokyo 113, Japan.

^{***} Department of Geology, Chungnam Nat. Univ., Daejeon 305-764, Korea.

by a X-ray diffraction method using bulk samples and fine fractions(<2 micron size). It was also hoped to get some clues for the genesis of the deposit from the mineral assemblages. The observations in this paper are mainly the results of a study of two drill holes from the Gusi deposit.

GEOLOGICAL SETTING

Regional Geology

The volcanic rocks and related volcanogenic sediments are widely distributed in the Haenam area and erupted during the late Cretaceous. These rocks overly Precambrian metamorphic rocks and Jurassic granitic rocks unconformably, and were intruded by late Cretaceous granitic rocks.

The age relationships of the volcanic rocks distributed in the Haenam area have been reported by Moon et al.(1990). The volcanic field of the Heanam area began to form 94 Ma from a series of volcanic rocks consisting of an andesitic lava flow and pyroclastic rocks distributed in the northeastern part of the Hawon peninsula. These volcanic rocks are called the intermediate volcanics(Son et al., 1978). Subserquently, acidic ash-flow tuffs and rhyolites

were erupted(79-82 Ma) and distributed widely in the central part of the Hawon peninsula. Intermediate and acidic lava flow, and its tuffs were erupted in a final stage of volcanism in the eastern part of the Heanam are(68 Ma). The quartz porphyry which intrudes the volcanic rocks shows K-Ar ages of 63 Ma and 67 Ma respectively(Lee and Lee 1976).

General Geology

The geology(Fig. 1) and hydrothermal alteration of the Gusi mine have been studied in detail. The rocks, which can be correlated with the Haenam Group, are divided into four formations in ascending order: Mudeungsan Flow, Manamri Rhyolite, Jangguri Tuff and Songsanri Rhyolite.

The Mudeungsan Flow, distributed in the eastern part of the area studied, is composed of andesite, andesitic welded tuff and andesitic tuff breccia. They are dark gray to dark green in color and show flow and welding textures. The rocks consist of plagioclase, hornblende and small amounts of quartz and K-feldspar. Hornblende is altered to chlorite and epidote in some places. The size of breccia fragments in the rock is 5 cm to 40 cm. They strike N35 – 50 E and dip 30 – 40 NW.

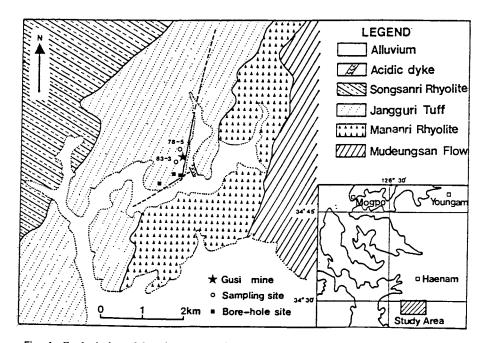


Fig. 1. Geological and location maps of the Gusi mine area showing drilling sites.

The Manamri Rhyolite, distributed in the eastern part of the Gusi mine, is white to pale brown in color and shows flow texture. The rock is deposited conformably above the Mudeungsan Flow, and consistes mainly of plagioclase and small amounts of quartz, sericite and hematite. It strikes N10°-45° and dips 15°-35° NW.

The Jangguri Tuff distributed in the central part of the area studied, including the Gusi mine, is composed mainly of welded tuff and lithic tuff, and overlies conformably the Manamri Rhyolite. It is light gray to pale green in color, and has a welding texture. Black shales are sometimes intercalated within the tuffs. The upper part of this formation contains andesite and rhyolite pebbles derived from the lower part of the Mudeungsan Flow and Manamri Rhyolite. Both formations strike N10°-40° E and dip 7°-25° NW.

The Songsanri Rhyolite, distributed in the Western part of the area studied, is light brown, light gray and pale green in color, and has a flow texture. The rock consists of quartz, plasioclase and K-feldspar and overlies the Jangguri Tuff striking N30° – 50° E and dipping 20° – 50° NW.

The Gusi area is cut by a north to northeast trending high-angle fault with vertical displacement up to several tens of meters. The volcanic rocks and volcanogenic sediments of the area have suffered extensive prorylitic alteration. The Jangguri Tuff in the Gusi mine area has undergone intens hydrothermal alteration producing pyrophyllite deposits. Lenticular-shaped or somewhat irregularly-shaped pyrophyllite deposits were mainly embedded in the lithic uiff having a high silica content, and forming part of the lower horizon of the Jangguri Tuff.

SAMPLE AND METHOD OF INVESTIGATION

Nineteen samples from the outcrop and twenty-one samples from the two drill holes were examined(Fig. 2). The core samples studied were collected from the two drill holes in the central part of the Gusi pyrophyllite deposit. The bulk samples and less than 2 micron size fractions were investigated by petrography, XRD, thermal and chemical analysis.

The bulk samples were first analyzed by XRD to identify mineral assemblages. The bulk rock chemistry of some selected samples were obtained by Inductively Coupled Plasma Spectrometer (ICP) on the dehydrated basis.

For the petrographic studies and electron mic-

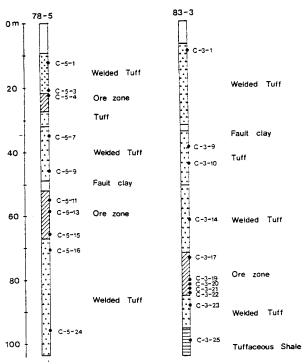


Fig. 2. Lithological coloumnar sections of the drilled holes for 78-5 and 83-3 showing sampling horizon.

roprobe analysis(EPMA) ordinary and impregnated thin sections were prepared. In order to obtain clay mineral assemblages in each alteration halo, less than 2 micron size fractions were separated from the powdered bulk samples by sedimentation after ultrasonic treatment for a few minutes. XRD patterns were obtained on powder and oriented mounts. All oriented mounts were analyzed by following treatments: heated at 250° and 500°C for two hours, and ethylene glycolated. Additional acid treatment was carried out to identify interstratified minerals. After obtaining mineralogical data, representative minerals such as pyrophyllite, illite, chlorite and kaolin minerals were analyzed with Jeol JCX 733 electron microscobe in Yonsei University, using the wavelength dispersive system. Alteration zones were defined on the basis of their mineral assemblages and occurrences.

MINERAL ASSEMBLAGES AND ALTERATION ZONING

The mineralogical compositions of the bulk samples were determined by XRD method and are listed in Table 1. The first column of Table 1 shows the predominant clay minerals in the

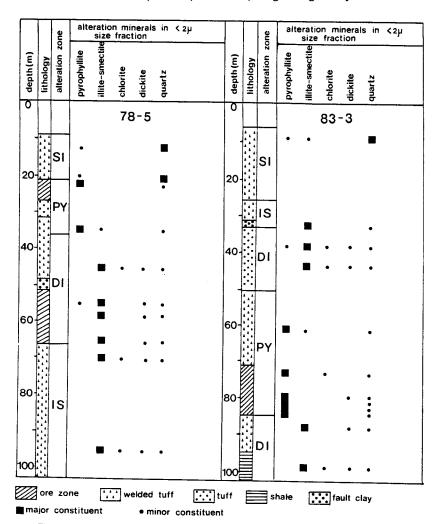


Fig. 3. Logs of two wells showing lithology, alteration zone and clay mineral assemblages of the <2 micron size fractions.

bulk samples excluding non-clay minerals. The second column shows the associated clay minerals and the last column shows the non-clay minerals. Fig. 3 shows the mineral assemblages of each altered zone. The hydrothermal altered rocks can be divided into four alteration zones (Fig. 4) on the basis of mineral composition in the bulk samples using optical and XRD methods(Niskanen, 1964). The alteration zones, going from the center to the margin of the alteration process. The amount of the non-clay minerals are usually less than that of the clay minerals in all alteratin zones except for quartz zone. In order to avoid confusion, the definition of alteration used here needs to be clarified. "Intense alteration" means that parent materials

except for quartz were altered to authigenic phyllosilicates with the original volcanic rock textures being almost completely destroyed. "Morederate alteration" means that the textures of volcanic rocks partly preserved and the parent materials can be identified.

Quartz Zone

This zone is mainly composed of fine-grained quartz with small amounts of kaolin, pyrophyllite, and some other minerals. Vuggy quartz is rarely found in this zone. This zone is distributed in the top of the alteration envelope of the deposit.

Table	1.	Mineralogical	compositions	of	bulk	samples.
1 4010		Millioralogical	compositions		Cuin	builipies.

Sample	Predominant	Other Clay	Non-Clay
No	Clay Mineral	Minerals	Minerals
J-1	IS	Py	Qz, Fd
J-2	IS	Py	Qz
C-5-1	Py	-	Qz
C-5-3	Py	_	Qz
C-5-4	_	_	Qz
C-5-7	Ka	IS, Py	Qz
C-5-9	IS	Ch, Ka	Qz
C-5-11	Py	IS, Ka	Qz
C-5-13	IŚ	_	Qz
C-5-15	IS	Ka	Qz
C-5-16	IS	Ch, Ka	Qz
C-5-24	IS	Ch, Ka	Qz
C-3-1	_	IS, Py	Qz
C-3-9	Ka, IS	Ch	Qz
C-3-10	Ka, IS	Ch, Ka	Qz
C-3-14	Py	IS	Qz
C-3-17	$\mathbf{P}_{\mathbf{y}}$	Ch	Qz
C-3-19	Py	Ka	Qz
C-3-20	Py	_	Qz
C-3-21	Py	_	Qz
C-3-22	Рy	_	Qz
C-3-23	Ka, IS	_	Qz
C-3-25	Ka, IS		Qz

Keys: IS; Illite-smectite interstratified mineral, Py; Pyrophyl lite, Fd; Feldspar, Ka; Kaoline mineral, Ch; Chlorite, and Qz; Quartz.

Pyrophyllite Zone

The rock in this zone is pale green to green in color. Predominant mineral is pyrophyllite with an appreaciable amount of dickite and quartz as an alteration product. The aletration of the wall rock adjacent to the channel of hydrothermal solution is the pyrophillite zone. Diaspore is rarely found in the central part of the zone. Original minerals and volcanic glasses are completely replaced, and form phyllosilicates. The primary texture of volcanics can not be recognized and indicates the intensity of the alteration.

Dickite Zone

Dickite zone consists mainly of kaolinite, dickite and quartz with small amount of alunite and chlorite. This zone distributes between pyrophyllite and Illite-smectite zones, and is intensely altered.

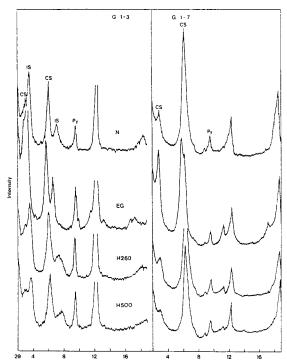


Fig. 4. X-ray diffraction patterns of oriented, glycolated, and heated interstratified minerals. Left; Gl-7 and right; Gl-9. IS; Illite-smectite interstratified mineral, CS; Chlorite-smectite interstratified mineral, and Py; Pyrophyllite.

Illite-Smectite Zone

This zone is situated in the outer part of the deposits, and is moderately aktered. The degree of alteration is easily distinguished because of the absence of pyrophyllite and abundance of illite minerals and of unaltered parent minerals. This zone is characterized by the abundance of illite. Illite-smectite interstratified mineral occur in this zone.

CHARACTERISTICS OF THE REPRESENTATIVE CLAY MINERALS

The fine fraction(<2 micron size) was examined by XRD and EPMA to identify the clay minerals. The results are listed in the colum in Fig. 3. Illites are abundant in all alteration zones except for the Quartz zone. Although kaolin minerals are the predominant mineral of the bulk samples from the Dickite zone, it is not the major constituent of the fine fraction because it usually occurs as a massive rock which is not easy to disperse.

Pyrophyllite

XRD results show that the pyrophyllites are 2M polymorphs. Electron microprobe analyses of pyrophyllite show no significant compositional variation in samples collected from different parts of the Pyrophyllite zone(Table 2). Al ion in the tetrahedral site ranging from 0.01 to 0.03 indicates that the very limited substitution of Al for Si occurred. Al ion in the octahedral site ranges from 98.7 to 99.7% of the total octahedral cations. The number of total octahedral cations ranging from 3.93 to 3.98 is close to the ideal formula, (Al₄)(Si₈xAlx) O₂₀ (OH)₄. Pyrophyllite occurs as a lenticular body in the central part of the alteration zone and as irregular shaped-body in the pyrophyllite zone. Within the kaolin zone, it occurs as a thin vein along the fractures. As shown in Table 2, the chemical compositions of pyrophyllites in each zone, are reasonably uniform.

Table 2. Electron microprobe analyses and structural formulae for pyrophyllites.

Sample						
No.	_ 1	2	3	4*	5*	6*
SiO ₂	64.73	65.75	65.52	66.42	65.79	65.39
TiO_2	0.00	0.00	0.00	0.00	0.00	0.00
Al_2O_3	27.32	27.96	27.51	28.06	27.62	27.54
FeO**	0.07	0.07	0.10	0.03	0.36	0.28
MnO	0.00	0.02	0.00	0.00	0.00	0.02
MgO	0.01	0.01	0.01	0.00	0.01	0.02
CaO	0.00	0.14	0.01	0.05	0.10	0.06
Na ₂ O	0.10	0.17	0.14	0.12	0.14	0.17
K ₂ O	0.01	0.02	0.36	0.10	0.40	0.20
Total	92.39	94.14	93.65	94.78	94.42	93.68
!	Number	of ion	s on the	basis (0(22)	
Si	7.99	7.98	8.00	8.00	7.99	7.99
Al(IV)	0.01	0.02	0.00	0.00	0.01	0.01
T.tet***	8.00	8.00	8.00	8.00	8.00	8.00
Al(VI)	3.97	3.97	3.96	3.98	3.94	3.95
Ti	0.00	0.00	0.00	0.00	0.00	0.00
Fe	0.01	0.01	0.01	0.00	0.04	0.03
Mn	0.00	0.00	0.00	0.00	0.00	0.00
Mg	0.00	0.00	0.00	0.00	0.00	0.00

^{*} Sample number from 4 to 6 occur as thin vein dickite zone, the rest of them are from ore zone.

Kaolin minerals

Kaolin minerals are predominant in all alteration zones of the deposit and are kaolinite and dickite. Kaolinites is more abundant than dickite, but its content is somewhat variable. Kaolinites are concentrated in the weathered surficial part of the illite-smectite zone as a supergene alteration products. Kaolinite has friable characteristics. It shows a characteristic 7 A peak and typical pseudohexagonal morphology on SEM image. The chemical composition of kaolin minerals occurring in each alteration zone shows an almost identical composition as that of the ideal formula, Al₂Si₂O₅(OH)₄. Results of some representative samples are listed in Table 3. In some samples, broad 7 Å peak appears after heat treatment at 550°C for two hours. It indicates that dickite may exist in these samples. The hkl reflections of some randomly mounted samples are also assigned to dickite. Dehydroxylation temperature of dickite appeared at about 600°C which is less than the expected value of a well ordered one at about 700°C. This low temperature indicates some degree of disorder.

Chlorite

EPMA results of authigenic chlorite occurring

Table 3. Electron microprobe analyses and structural formuale for kaolin minerals.

Sample						
No.	1	2	3	4	5	6
SiO ₂	46.93	46.72	46.43	47.16	47.55	47.42
TiO_2	0.00	0.00	0.00	0.00	0.00	0.04
Al_2O_3	39.45	39.28	38.70	39.82	38.94	39.15
FeO	0.01	0.02	0.07	0.03	0.04	0.00
MnO	0.00	0.00	0.00	0.00	0.00	0.00
MgO	0.00	0.07	0.00	0.00	0.00	0.01
CaO	0.07	0.03	0.00	0.01	0.02	0.00
Na ₂ O	0.00	0.01	0.00	0.00	0.10	0.00
K_2O	0.01		0.04	0.03	0.05	0.01
Total	86.47	86.13	85.24	87.05	86.70	86.63
	Number	of ion	s on the	e basis	0(14)	
Si	4.014	4.013	4.028	4.007	4.056	4.045
Ti	0.000	0.000			0.000	
	0.000	0.000	0.000	0.000	0.000	0.003
Al	3.977	3.976	0.000 3.957	0.000 3.988		0.003 3.936
Al Fe					0.000 3.915 0.003	3.936
	3.977	3.976	3.957	3.988	3.915	3.936 0.000
Fe	3.977 0.001	3.976 0.001	3.957 0.005	3.988 0.002	3.915 0.003	3.936 0.000 0.000
Fe Mn	3.977 0.001 0.000	3.976 0.001 0.000	3.957 0.005 0.000	3.988 0.002 0.000	3.915 0.003 0.000	3.936 0.000 0.000 0.001
Fe Mn Mg	3.977 0.001 0.000 0.000	3.976 0.001 0.000 0.000	3.957 0.005 0.000 0.000	3.988 0.002 0.000 0.000	3.915 0.003 0.000 0.000	3.936 0.000 0.000

^{**} All iron reported as FeO

*** Total number of cations in tetrahedral and octahedral site.

Sample							
No.	1	2	3	4	5	6	7
SiO ₂	23.64	23.97	24.54	25.33	24.93	25.00	25.35
TiO ₂	0.02	0.01	0.03	0.03	0.01	0.02	0.01
Al_2O_3	22.19	22.42	23.31	23.84	23.69	23.37	23.56
FeO*	33.80	32.69	23.59	22.27	22.34	24.18	21.02
Mno	0.85	0.92	2.28	2.33	2.53	2.03	2.27
MgO	5.13	5.67	11.55	12.39	12.35	11.51	12.92
CaO	0.00	0.00	0.01	0.01	0.02	0.00	0.02
Na ₂ O	0.00	0.00	0.00	0.00	0.00	0.00	0.00
K ₂ O	0.00	0.00	0.01	0.00	0.00	0.01	0.02
Total	85.63	64.68	85.32	86.20	85.88	86.12	85.17
		Numb	er of ions on	the basis 0(2	8)		
Si	5.37	5.41	5.32	5.38	5.33	5.37	5.41
Al(IV)	2.63	2.59	2.68	2.62	2.67	2.63	2.59
T. tet**	8.00	8.00	8.00	8.00	8.00	8.00	8.00
Al(VI)	3.32	3.36	3.27	3.34	3.29	3.28	3.34
Ti	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Fe	6.43	6.17	4.27	3.95	3.99	4.34	3.75
Mn	0.16	0.18	0.42	0.42	0.46	0.37	0.41
Mg	1.74	1.91	3.73	3.92	3.93	3.68	3.73
Ca	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Na	0.00	0.00	0.00	0.00	0.00	0.00	0.00
K	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T.oct**	11.65	11.61	11.70	11.64	11.69	11.68	11.63

Table 4. Electron microprobe analyses and structural formulae for some representative chlorites.

along the fracture in the Dickite zone are reported in Table 4. According to the recommendation of AIPEA Nomenclature Subcommittee(Bailey, 1980), most chlorites are ferroaluminian clinochlore and two of them are magnesian chamosite(Ripidolite by Hey's classification). Authigenic chlorites also occur in the illite-smectite zone, but those are too finegrained to obtain microprobe data. Cathelineau (1988) considered that the Al content in the tetrahedral site of chlorites and the K content of illite increase with temperature. Al ions in the tetrahedral sites of the present samples range between 2.59 and 2.63, indicating that they were formed under almost identical temperatures.

Illite and Interstratified Minerals

Illite occurs as the predominant mineral in the illite-smectite zone. It is uaually associated with an appreciable amounts of interstratified minerals such as illite/montmorillonite interstratified minerals(I/M) and chlorite/montmorillonite interstratified minerals(C/M). I/M and C/M

minerals can be identified more easily in the fine fractions. Even though illite shows basal spacing at about 10 Å region, it exhibits some degree of interstratification with montmorillonite. The mineral with 29 Å basal spacing consists of chlorite interstratified with montmorillonite. As shown in Fig. 3. 29 Å basal spacing is expanded to 31 Å after glycol treatment and collapsed with heat treatment 500°C. This is a low-charge corrensite as defind by AIPEA Nomenclature Committee(Bailey, 1982). It is characterized by a relatively sharp basal peak indicating regularity in the montmorillonite and chlorite layering.

I/M mineral was identified by expanding basal spacing from 25Å to 27 Å after glycolation. EPMA results of I M minerals are reported in Table 5. K₂O content of I/M minerals renging from 7.19 to 8.20% is less than that of mica. Interlayer charge, liying between 1.4 and 1.6, indicates that proportion of expanding layer is not greater than 5%. It shows good agreement by XRD method, suggested by Tomita et al.(1988). Illite and montmorillonite also can be identified by scanning electron microscopy (SEM).

^{*} All iron reported as FeO.

^{**} Total number of cations in tetrahedral and octahedral site.

Table 5. Structural formulae for illite-smectite interstratified minerals.

Sample			-				
No.	_1	2	3	, 4	5	6	7
Si	6.35	6.46	6.36	6.52	6.51	6.54	6.57
Al(IV)	1.65	1.54	1.64	1.48	1.49	1.46	1.43
Al(VI)	3.96	3.90	3.91	3.91	3.91	3.61	3.66
Ti	0.00	0.00	0.00	0.00	0.00	0.04	0.02
Fe**	0.09	1.13	0.11	0.14	0.10	0.38	0.36
Mg	0.02	0.05	0.01	0.01	0.03	0.06	0.06
Mn	0.00	0.00	0.00	0.00	0.00	0.01	0.01
Ca	0.01	0.04	0.01	0.01	0.02	0.03	0.03
Na	0.17	0.13	0.16	0.16	0.16	0.08	0.08
K	1.35	1.27	1.36	1.29	1.25	1.42	0.39
T.oct***	0.53	1.34	1.53	1.46	1.43	1.53	0.50
Si/Al	1.13	1.19	1.15	1.21	1.21	1.29	1.29

- * Number of ions on the basis 0(22).
- ** All iron reported as FeO.
- *** Total number of cations in terrahedral and octahedral site.

BULK CHEMICAL AND TEXTURAL CHANGE DUE TO ALTERATION

Twenty-four samples from the drill core and unaltered tuff collected from the surface were analyzed by ICP(Table 6). As shown in Fig. 2, C-series samples were collected from the drill core and J-series samples were collected from the unaltered tuff horizon in the south-western extent of the main ore zone.

The data shows very pronounced chemical variation among each altered zone, including the unaltered tuff. In considering the chemical data, it is assumed that alumina is an immobile element and hence contents of altered rock are the same as those of unaltered ones. On the basis of this assumption, chemical analyses were normalized. The result is shown in Fig. 5. Some components show a regular variation from zone to zone reflecting their mineralogical assemblages. FeO is closely related to pyrite contents of the rock. CaO, Na₂O and K₂O probably reflect alteration intensity. For example, those elements in the intensely altered zone(Pyrophyllite zone) are depleted compared with those of other alteration zones. It indicates that parent materials, mainly volcanic glasses, are intensely

Table 6. Partial chemical analyses of bulk samples.

Sample										
No.	Al_2O_3	FeO	Fe_2O_3	MgO	CaO	Na ₂ O	K_2O	TiO ₂	P_2O_5	MnO
C-3-9	13.33	1.14	0.61	0.85	0.26	0.10	2.63	0.15	0.10	
C-3-10	17.12	1.24	2.11	1.05	0.28	0.12	3.45	0.13		0.09
C-3-14	18.09	0.27	0.03	0.03	0.09	0.08	0.83	0.32	0.07	0.17
C-3-17	28.01	0.20	0.05	0.05	0.21	0.08	0.33	0.30	0.05	0.00
C-3-19	10.23	0.19	0.39	0.02	0.06	0.03	0.10		0.20	0.00
C-3-20	18.50	0.21	0.01	0.02	0.06	0.03	0.42	0.19	0.04	0.00
C-3-21	25.07	0.16	0.18	0.02	0.08	0.03	1.93	0.19	0.06	0.00
C-3-22	14.06	0.14	0.03	0.01	0.05	0.06	0.48	0.22	0.01	0.00
C-3-23	25.50	0.14	1.21	0.21	0.05	0.00		0.12	0.04	0.00
C-3-25	14.33	2.26	0.85	3.33	5.42	0.10	6.16	0.15	0.04	0.00
C-5-1	7.16	0.00	0.03	0.00	0.03		3.67	0.51	0.13	0.15
C-5-3	5.17	0.00	0.03	0.01	0.03	0.02	0.00	0.21	0.03	0.00
C-5-4	23.55	0.18	0.14	0.01	0.03	0.01	0.00	0.21	0.02	0.00
C-5-7	18.51	0.12	0.52	0.01	0.00	0.06	0.00	0.33	0.08	0.00
C-5-9	15.07	0.21	1.45	0.12		0.19	2.73	0.24	0.06	0.00
C-5-11	17.84	0.13	0.16	0.77	0.25	0.12	2.85	0.21	0.06	0.16
C-5-13	19.50	0.31	0.10	0.02	0.12	0.13	1.49	0.23	0.08	0.00
C-5-15	14.63	0.37	0.51	0.03	0.26	0.20	3.62	0.35	0.08	0.00
C-5-16	13.31	0.62	1.03	0.11	0.19	0.10	3.03	0.20	0.04	0.00
C-5-24	17.74	4.67	0.18		0.27	0.09	2.47	0.21	0.08	0.01
J-1	14.22	1.07	1.07	1.20	0.26	0.21	2.66	0.28	0.04	0.09
J-2	12.28	0.14	0.30	0.35	0.13	0.64	5.19	0.23	0.04	0.01
J-3	11.49	0.14	0.30	0.16	0.09	1.89	2.54	0.11	0.02	0.04
	11.77	0.41	0.79	0.56	0.05	0.07	4.06	0.21	0.05	0.02

Core samples; C-3-9~C-5-24 and less altered samples; J-1~J-3.

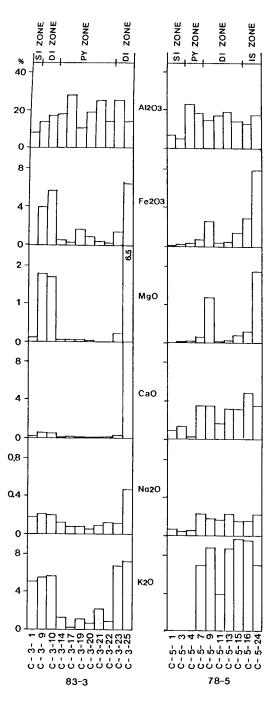


Fig. 5. Contents of major elements four different alteration zones. Al₂O₃ is real value and the rest of them are normalized to the Al₂O₃ content assuming that chemistry of altered and unaltered rock is uniform.

altered. Analyses of samples from the drill hole 78-5 do not show a regular variation with depth. It probably due to superimposed alteration. Therefore, chemical variation cannot be interpretated as a single change of chemical composition in the solution with a drop of temperature. Another possible interpretation for such an irregularity is that each type of alteration zone, or envelope, was probably formed from the solutions having originally different chemical composition, but it is not likely to occur in this deposit. Because mineralogical assemblage of each alteration zone in the Gusi deposit is almost identical, which indicates that superimposed alteration proceeding with the solution having almost identical chemical character.

Potassium depletion of the drill hole 78-5 is not observed in the illite-smectite zone where the illite phase is predominant. Parent material is not expected to be totally replaced as the alteration is less intense. The apparent enrichment of potassium in illite-smectite zone from the drill hole 83-3 is probably due to the fact that the parent material remains unaltered.

Alteration zones in this deposit are gradational. However, textural change due to alteration can be monitered from the drill core and surface outcrops. Consequently, alteration intensity decreases from the central part of the deposit towards the outermost part. Weakly altered rocks(outer illite-smectite zone) preserves characteristic textures of the original rock and contains occasionally unaltered volcanic materials whereas intensely altered rocks do not. In general, intensely altered rocks show simpler or more monominerallic assemblage than those of a less - intensely alteration. For intance, pyrophyllite ore developed in intensely rocks consists of wholly pyrophyllite or predominantly pyrophyllite with a small amount of quartz and kaolinite. Unaltered parent materials can hardly be detected in this ore. The quartz zone, distributed in top of the alteration envelope, shows a characteristic occurrence with both vuggy and friable quartz. Massive silicified rocks are more widely distributed than the friable rock and consist mainly of cryptocrystalline intergrown quartz which is resistant to erosion and hence froms a capping to the depposit.

DISCUSSION AND CONCLUSION

The parent rocks of the hydrothermal altered altered the Gusi deposit are the lithic and welded tuff of the Jangguri Tuff. These volcanic rocks have the relatively higher permeability than those of outer unaltered rhyolite. These rocks

were easily altered by a process of metasomatism with large amounts of fluid passing through channel especially along fault planes. Pyrophyllite and Dickite zones were developed close to the fault plane, which indicates that high angle fault plane served as a channel of thermal solution. The fairly large size of the alteration zone and microscopic texture of the altered rock suggest that alteration be proceded by infiltration process.

From the available data for diagenetic alteration of sediment and hydrothermal alteration of tuff under active geothermal system, there is such a tendency with increasing the temperature(Steiner, 1968; Hower et al., 1976; Bole and Frank, 1979; Utada, 1983). Mineralogical transformation from montmorillonite to illite starts to occur at about 80 to 140 °C. When the temperature increases to about 130 to 180 °C, all water is lost and the mineral structure is completely changed to illite structure. The presence of I/M mineral in illite-smectite zone suggests that the alteration temperature of this zone had not reached 200°C. Inoue and Utada(1983) proposed that the degree of ordering of interstratification of hydrothermal mica/ semctites changes discontinuously from g=0 to g=1 and from g=1 to g=2, whereas the conversion is continuous in diagenetic clay. Such a discontinuous trend from g=0 to g=1 could not be monitered because of lacking of data of the studied samples. However, the occurrence of illite clearly shows that it was formed by hydrothermal alteration.

The percentage of expandable layer and the tetrahedral Al content of I/M occurred in the Gusi pyrophyllite deposit is less than 5% for most samples ranging between 1.43 and 1.77 per O₂₀ OH₄. It indicates that smectite is almost converted into a mica with a sericitic composition. The I/M formed by hydrothermal alteration and those of diagenetic origin have dominantly non-exchangrable interlayer K(Hower and Mowatt, 1966; Eslinger et al., 1979; Inoue and Utada, 1983; Nadeau and Bain, 1986). Therefore, the number of K ions as interlayer cation in Table 4 cannot be interpretated for the genetic environment, but high K in the interlayer suggests high degree of illitization occurred at a considerably high temperature, definitely more than 100℃

Henley and Ellis(1983) summarized that each alteration zone characterized by specific mineral has a certain range of formation temperature. According to them, C/M zone corresponds to temperature range between 130 and 170°C and Chlorite zone between 170 and 200°C. Mineral

stability relation in the system Al₂O₃ - SiO₂-H₂O were investigated by Hemley et al.(1980). They concluded that dehydration temperature at 1 Kb H_2O is 273 ± 10 °C and 300°C ±10 °C for kaolinite - pyrophyllite - quartz and kaolinite-pyrophyllite - diaspore, respectively. Dickite- diaspore-pyrophyllite assemblages typically occur in sillicic and intermediate silicate rocks that have undergone extreme acid leaching, producing advanced argillic assemblages. Such environment are most commonly seen in recent volcanic terranes and have been widely described(Utada, 1980; Kamitani, 1974; Stofferegen, 1987; Hedenquist, 1987). Intense acid alteration is, of course, a common expression of relatively shallow hydrothermal alteration processes operating in volcanic system because under these condition the ionization and oxidation of magmatically derived acid constituents havd reached maximum. And although water was assumed to be pure phase in this system, it is reasonable to consider that the fluid phase was rich in Na+ K+ , H+, Ca+, Cl-, aqueous sillica and others. Therefore, the factor that controls the alteration processes is not only temperature and pressure but also the chemical compositions of the solution such as pH, aNa⁺, aK⁺, and so on("a" representing the activity). On the basis of the chemical data and mineral assemblages a rough estimation of the alteration temperature of each zone in the Gusi pyrophyllite deposit can be made. It is likely that Illite-smectite zone was formed at about under 200°C, dickite corresponding a temperature over 200°C but less than 265℃ and pyrophyllite higher than 265℃.

Alteration process can be summarized as follows. In the center of hydrothermal activity, pyrophyllite was formed by interaction between hydrothermal solution and parent materials at relatively high temperatures (greater than 265°C). Disseminated diaspore in the pyrophyllite zone indicates that the formation temperature reached up to 300°C and the thermal solution was strongly acidic. Hemley et al.(1980) suggested that higher temperature and high aqueous silica activity favour pyrophyllite relative to kaolinite. Strong acidic alteration is commonly found in epithermal alteration processes occurring in volcanic system because under these condition the ionization and oxidation of magmatically derived acidic constituents have been greatly increased. Under this circumstance, the reactions between volcanic material and hydrothermal solution in the Gusi deposit can be assumed as follows:

The hydrothermal solutions gradually alter to tuff by infiltration processes toward the outer

part of the alteration envelope, and were mixed. or diluted with groundwater or interstitial pore solution and hydroxyl water, mainly from the volcanic glass. This solution at a relatively lower temperature in an early stage of the alteration process is likely to form smectite. Subsequently, smectite was assumed to react with convecting relatively high temperature hydrothermal solutions, resulting in the transformation of smectite to illite or chlorite. Absence of pure end member of montmorillonite and presence of small amount of I/M mineral in outer alteration zone agree with above assumption. Eberl and Hower(1977) stated that Na-semectite is stable to at about 400°C in the solution with high Naactivity. This fact suggests that conversion from smectite to I/M mineral or illite were controlled by not only temperature but also composition of hydrothermal solution. The empirical relationship between Al in tetrahedral position in chlorite and temperature was made by Cathelineau (1988). The chlorite samples used for geothermometry were collected from the pyrophyllite zone. The formation temperature of chlorite calculated from a empirical formula suggested by them ranges from 250 to 265°C. Pyrophyllites which were formed at a higher temperature in an relatively later stage of alteration are distributed in the innermost part of the deposit. Thin pyrophyllite vein indicates superimposed alteration cuts the Dickite and Pyrophyllite zones. It is concluded that the Gusi deposit was formed by shallow hydrothermal alteration processes operating in a volcanic system.

ACKNOWLEDGEMENT

HSM thanks the Korea Science and Engineering Foundation for financial support of this work. This work carried out as a part of the project No. 88610. We also thank Dr. I. Wilson(ECC Internatinal, St. Austell) for a helpful review and improving English of the manuscript.

REFERENCES

- Ashley, R. P.(1974) Goldfield mining district. Nevada Bur. Mines and Geology, Rept., v. 19, p. 49-66.
- Bailay, S. W.(1980) Summary of recommendation of AIPEA nomenclature committee. Clays Clay Miner., v. 28, p. 73-38.
- Bailey, S. W.(1982) Nomenclature for regular interstratifications. Am. Miner., v. 67, p. 394-398.
- Berger, B. R. and Henley, R. W. (1989) Advances in the understanding of epithermal gold-silver deposits, with special reference to the western United States. Econ. Geol. Monograph, v. 6, p. 405-423.

- Boles, J. R. and Frank, S. G.(1979) Clay diagenesis in Wilcox sandstones of southwest Texas: implications of smectite diagenesis on sandstone comentation. J. Sed. Pet., v. 23, p. 55-77.
- Cathelineau, M.(1988) Cation site occupancy in chlorites and illites as a function of temperature. Clay miner., v. 23, p. 471-485.
- Cho, H. I. and Moon, H. S.(1978) Alunite deposits in Korea. Korea Research Inst. of Geoscience & Mineral Resources, KIGAM Bull. -02, p. 1-100(in Korea).
- Corn, R. M.(1975) Alteration-mineralization zonning, Red Mountain, Arizona. Econ. Geol., v. 70, p. 1437-1477.
- Eberl, D. D. and Hower, J.(1977) the hydrothermal transformation of sodium and potassium smectite into mixed layer clay. Clays Clay Miner., v. 25, p. 215-227.
- Eslinger, E., Highsmith, P., Albers, D. and De Mayo, B.(1979) Role of iron reduction in the conversion of smectite to illite in bentonites in the Disturbed Belt, Montana. Clays Clay Miner., v. 27, p. 327-338.
- Hedenquist, J. W.(1987) Mineralization associated with volcanic-related hydrothermal systems in the Circum-Pacific Basin: in Horn, M. K., ed., Transactions of the Fourth Circum-Pacific Energy and Mineral Resources Conference: Singapore, p. 513-524.
- Hedenquist, J. W. and Aoki, M.(1990) Meteoric interaction with magmatic discharges in Japan, and its significance regarding the potential for mineralization. In "High-temperature acid fluids and associated alteration and mineralization" (Extend Abstracts), Geol. Survey Japan, Tsukuba, Japan, p. 63-73.
- Hemley, J. J., Hostetler, P. B., Gude, A. J. and Mountjoy, W. T.(1969) Some stability relations of alunite. Econ. Geol., v. 64, p. 599-612.
- alunite. Econ. Geol., v. 64, p. 599-612.

 Hemley, J. J., Montoya, J. W., Marinenko, J. W. and Luce, R. W.(1980) Equilibria in the system Al₂O₃-SiO₂-H₂O and some general implications for alteration/mineralization processes. Econ. Geol., v. 75, p. 210-228.
- Henley, R. W. and Ellis, A. J. (1983) Geothermal systems ancient and modern; a geothermal review. Earth-Sci. Rev., v. 19, p. 1-50.
- Hower, J., Eslinger, E. V., Hower, M. E. and Perry, E. A. (1976) Mechanisms of burial metamorphism of argillaceous sediments: 1. Mineralogical and chemical evidence. Bull. Geol. Soc. Am., v. 87, p. 725-737.
- Inoue, A. and Utada, M.(1983) Further investigations of a conversion series of dioctahedral mica/smectites in the Shinzan hydrothermal alteration area, Northeast Japan. Clays Clay Miner., v. 31, p. 401-412.
- Iwao, S.(1972) Genesis of clay(II). Jour. Clay Sci. Japan, v. 12, p. 81-95(in Japanese).
- Kamitani, M.(1974) hydrothermal alteration at the Uku "Roseki" mine, Yamaguchi Prefecture. Min. Geol., v. 24, p. 31-43(in Japanese).
- Lee, D. S. and Lee, H. Y. (1976) Geological and

geochemical study on the rock sequences containing oily materials in Southern coast area of Korea. Jour. Korea Ins. Min. Geol., v. 9, p. 183-218.

Meyer, C and Hemley, J. J.(1967) Wall rock alteration. In Barnes, H.L., ed., Geochemistry of Hydrothermal ore deposits,. New York, Holt Rinehart and Winston, Inc., v. 204, p. 183-218.

Meyer, C.(1968) Ore deposits at Butte, Montana. In Ore Deposits of the U. S., 1933-1967. J. D. Ridge, ed., New York, Am. Inst. Min. Eng., p. 1373-

1416.

- Moon, H. S., Kim, Y. H., Kim, J. H. and You, J. H. (1990) K-Ar ages of alunite and sericite in altered rocks and volcanic rocks around the Haenam area. J. Korea Inst. Min. Geol., v. 23, p. 135-141.
- Nadeau, P. H. and Basin, D. C.(1986) Composition of some smectites and diagenetic illite clays and implications for their origin. Clay Clay Minerals, v. 34, p. 455-464.

Niskanen, E.(1964) Reduction of orientation effects in the quantitative XRD analysis Kaolin minerals. Am. Mineral., v. 49, p. 705-714.

Rye, R. O., Bethke, P. M. and Wasserman, M. D.(1989) Diverse origins of alunite and acid-sulfate alteration: Stable isotope systematics. U. S. G. S., Open File Rept., p. 1-14.

Son, J. D., Yoon, H.S., Kim, H. Y., Ko, M. J., Cho, D. H. Koo, J. H. and Kim, S. G.(1980) Geology, geochemical and geochemical exploration for the Cretaceous strata containing oily shale in Haenam Mogpo area. KIGAM(KIER) Bull., no. 21, p. 51(in Korean).

Srodon, J., Morgan, D. J., Eslinger, E. V., Erberl, D. D. and Karlinger, M. R.(1986) Chemistry of illite/smectite and end-member illite. Clays Clay Miner., v. 16, p. 368-378.

Steiner, A. (1968) Hydrothermal rock alteration at Wairakei, New Zealand. Clays Caly Miner., v. 16,

p. 193-213.

Stofferegen, R.(1987) Genesis of acid-sulfate alteration and Au-Cu-Ag mineralization at Summitille, Colorado. Econ. Geol., v. 82, p. 1575-1591.

Tomita, K., Takahashi, H. and Watanabe, T.(1988) Quantification curves for mica/smectite interstratifications by X-ray powder diffraction. Clays Clay Miner., v. 36, p. 258-262.

Utada, M.(1990) Hydrothermal alteration related to igneous activity in Cretaceous and Neogene formationd of Japan. Min. Geol., Spce. Issue, no. 8, p.

67-83.

Velde, V. and Brunsewitz, A. M.(1986) Compositional variation in component layer in illite/smectite.

Clays Clay Miner., v. 34, p. 651-657.

Wang, Y.(1972) Wall rock alteration of late Cenozoic mineral deposit of Taiwan: Mineralogical and Physicochemical aspects. Acta Geol. Taiwanica, no. 16, p. 29.

Manuscript received 31 December 1990.

전남 해남지역 구시광상의 화산활동에 수반된 열수변질작용 및 생성환경

문희수·노열·김인준·송윤구·이현구

요약 : 구시납석광상은 전남 해남지역의 중생대 백악기 화산암류를 모암으로 산출된다. 모암을 이루는 화산암류는 대부분 응회암류이나 일부 유문암의 변질산물도 있으며, 이들의 열수변질 산물로서 구시광상의 납석 및 도석을 형성 시켰다. 이 변질작용의 결과로 형성된 광물조합에 의하여 이들은 변질대의 중심부로부터 외각으로 가면서 납석, 디카이트, 석영 및 일라이트─스멕타이트대로 구분된다. 이러한 변질광물의 분포양상은 모암인 장구리웅회암이 상대적으로 높은 Si와 K이온 활동도를 갖는 강한 산성 열수용액에 의한 변질작용에 의하여 형성되었음을 지시한다. 이들 광물의 안정영역을 고려할 때 일라이트─스맥타이트대는 200℃ 이하에서, 디카이트대는 200~250℃ 범위에서, 납석대는 260℃ 이상의 온도 영역에서 형성 되었음을 지시한다. 납석대의 일부에서 다이수로모어를 함유하는 것으로 보아 이때의 최고 온도는 300℃ 정도에 이르렀을 것이다. 이들 변질작용은 본역에 분포된 화산암류를 형성시킨 화산활동에 기인된 열수변질작용으로 해석된다.