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Mineralogy of Kaolin from Hadong-Sancheong Area, Korea

(하동-산청지역에서 산출되는 고령토의 광물학적 연구)

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ABSTRACT: The kaolin deposits in Hadong-Sancheong area, have been formed by supergene weathering of anorthositic rocks including anorthosite, leucogabbro, and gabbro. Kaolin consists chiefly of halloysite(10Å) and kaolinite with other minerals such as illite, vermiculite, plagioclase, hornblende, quartz, amorphous materials(allophane and silica), goethite, and hematite. Goethite and hematite are the major coloring agents of the reddish brown and other colored kaolins. Other common accessory minerals are magnetite, ilmenite, anatase, gibbsite, I/S, C/V, chlorite, lithiophorite, and birnessite. Paragonite, dravite, laumontite, clinozoisite, muscovite, scolecite, stellerite are locally found. Al substitution of Fe in goethite and hematite decreases from the surface zone toward the deeper zone. The kaolin deposits show three horizontal zoning; the upper reddish brown, middle pink, and lower white zones. All the zones are characterized by somewhat different mineralogy. The factors for the formation of kaolin deposits in Hadong-Sancheong area are 1) the presence of anorthositic rocks, 2) the low flat or gentle topography, 3) the favorable climate, and 4) the long-continued preservation of kaolins without erosion.

요약 : 하동-산청지역의 고령토는 회장암, 우백질 반려암 및 반려암으로 구성된 회장암질암의 풍화작용에 의하여 생성되었다. 고령토의 주구성 광물은 할로이사이트(10Å)와 캐올리나이트이며 일라이트, 버미큘라이트, 사장석, 각섬석, 석영, 비정질(알로팬, 실리카), 침철석, 적철석이 수반된다. 침철석과 적철석이 적갈색 및 기타 착색된 고령토의 주요 착색 요인이 된다. 그외 수반광물로써 깁사이트, 녹니석, I/S, C/V, 자철석, 일메나이트, 아나타제, 리치오포라이트, 버네사이트 등이 흔히 산출된다. 파라고나이트, 드래바이트, 로몬타이트, 클리노조이사이트, 백운모, 스콜레사이트, 스텔러라이트는 국지적으로 산출된다. 침철석과 적철석내 Al 치환량은 지면에 가까울수록 증가한다. 고령토 광산은 광물조성이 약간 다른 3개의 수평분대; 상부 적갈색대, 중부 분홍색대, 하부 백색대로 구분된다. 하동-산청지역의 고령토 광상 생성요인은 1) 회장암질암의 존재, 2) 평탄하고 완만한 지형, 3) 적당한 기후조건, 4) 오랜 동안의 광상의 보존등이다.

INTRODUCTION

The kaolin deposits in Hadong-Sancheong area are developed as a 50km long belt trending NS in the central southern part of the Korean peninsula. Its southern end is located 20km west of Jinju, Gyeongsangnam-do (province), Korea, and at 35°00'-35°30'N in latitude and 127°57'E in longitude (Fig. 1). The deposits have been the important source of kaolin for the ceramic industry in Korea and Japan. The total tonnage of reserves are about 65,000,000 tons.

The geology of the area was studied by Choi *et al.* (1964), Jeong (1980), Lee (1980), Song (1981), and Jeong and Lee (1986). Kaolin deposits of the area were studied by Kim and Kim (1964), Sang *et al.* (1972), Lee *et al.* (1977), and Sang (1980). Detailed mineralogical work has recently been made by Lee (1986), Kwon (1986), Jeong (1987), and Kim (1988).

GEOLOGICAL BACKGROUND

Geology of the area consists of Precambrian gneisses and anorthositic rocks, and Jurassic diorite, schistose granite, and syenite, and Cretaceous granite (Fig. 1). Precambrian gneisses consist of banded gneiss, migmatized gneiss, dioritic gneiss and granitic gneiss. They are intruded by anorthositic rocks.

Anorthositic rocks consist of anorthosite, leucogabbro, and gabbro. They usually have banded structure due to primary layering and metamorphism. Cataclastic texture is well developed in anorthositic rocks. Plagioclase, hornblende, and olivine in the rocks are magmatic in origin, whereas tremolite, actinolite, and biotite are postmagmatic. They have undergone retrograde metamorphism which resulted in the formation of chlorite, muscovite, epidote, and albite (Jeong and Lee, 1986).

Many dikes are abundantly found in the anorthositic rocks which have undergone kaolinization. They are mostly of andesitic or lamprophyric, but rarely of acidic. Small quartz, clinozoisite,or zeolite veins are found in anorthositic rocks in places.

OCCURRENCE

Occurrence of kaolin deposits is confined to

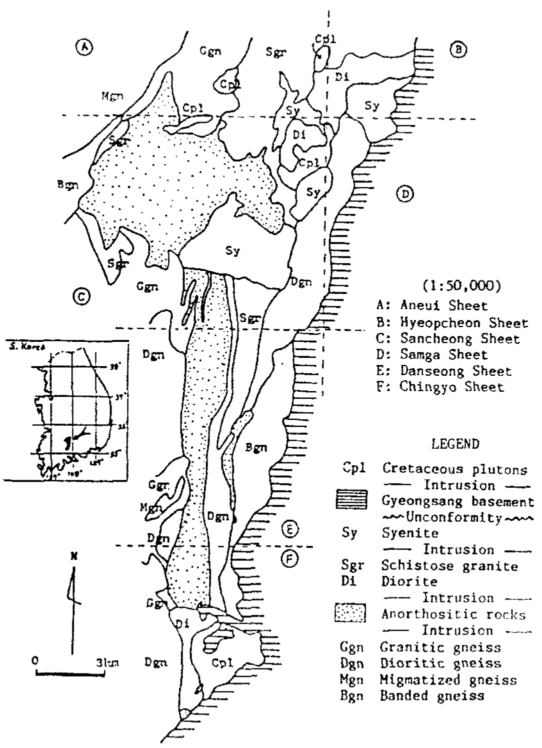

Fig. 1. Geologic map of Hadong-Sancheong area (Jeong, 1980).

the area of anorthositic rocks. They are generally localized on the low and gentle topography with a slope of less than 17° between 100 (the local base) and 350m in altitude.

The kaolin deposits are developed to the depth of 30 meters. The profile of kaolin deposits suggests the weathering origin. They usually have three horizontal zones; the upper reddish brown, middle pink, and lower white zones. The pink and white zones are usually the main objects of mining.

The reddish brown zone is rich in iron and shows no textures of parental rocks, whereas the pink and white zones preserve their original structures. The gneissose structures of various patterns are well preserved in the kaolin ores, although the original materials have been altered to clays and similar minerals. The present oriented structures of kaolin are the relict fabric of the parent anorthositic rocks. The color of the relict spots, streaks, and bands is yellowish, greenish, or brownish.

The white zone generally shows gradational transition to the underlying non-kaolinized parent rocks. In the transition zone from white zone to the underlying parent rocks, there is so-called "sandy kaolin" which consists of kaolin and sandy particles of plagioclase showing various stages of weathering. The kaolin deposits also contain large or small rounded masses of anorthositic rocks near the contact with underlying parent rocks. They are original anorthositic rocks which escaped from kaolinization and usually show sharp contact with kaolin. The contact of kaolin and the underlying parent rock is not uniform, but variable from place to place largely controlled by the topography of the area.

Kaolin is also found along the fissures in the anorthosite near the margins of the main kaolin deposits. Kaolins which are developed along the basic dykes are generally of high grade.

MATERIALS AND METHODS

According to the occurrence of kaolin, a systematic sampling was done for different color, texture, and depth. The collected samples were tightly packed in polyethylene bags and transported to preserve the original wet state and their original structure.

For mineral identification, both the field moist and dried bulk samples were analyzed by X-ray diffraction using nickel-filtered Cu $K\alpha$

radiation of JEOL JDX-5P X-ray diffractometer operated at 30kV 20mA. Clay mineral sample was separated by handpicking under stereomicroscope and size-fractionation, and iron oxide minerals were concentrated by 5M NaOH selective dissolution method (Kämpf and Schwertmann, 1982). Size-fractionation was made by sedimentation after disaggregation and ultrasonic agitating. These treated samples were prepared for X-ray work as random specimens and preferred oriented one by smearing on slide glass. The prepared specimens were saturated with K and Mg by their 1N chloride solution, and treated with ethylene glycol and glycerol vapor at 60°C.

Different forms of iron (crystalline and amorphous) in kaolin were selectively extracted by dithionite-citrate-bicarbonate (DCB) (Mehra and Jackson, 1960) and acid oxalate (Blackmore *et al.*, 1981). The same specimen was completely decomposed with HF-H_2SO_4-HNO_3 to estimate total Fe content. The extracted and total iron were analysed by atomic absorption spectrophotometer model IL251 using air-acetylen flame. Chemical analyses of kaolin samples were made by wet method.

TEM analyses was made by JEOL JEM 200CX operated at 160 kV. The specimen for TEM observation was prepared by laying the clay particles in distilled water on the copper grit coated with carbon.

RESULTS AND DISCUSSION

Mineral Composition

Clay minerals identified from kaolin samples are halloysite, kaolinite, nacrite, illite, vermiculite, mixed layer illite-smectite (I/S), mixed layer chlorite-vermiculite (C/V), chlorite, and momtmorillonite. Other associated minerals are gibbsite, muscovite, plagioclase, hornblende, quartz, goethite, hematite, ferrihydrite, magnetite, ilmenite, anatase, lithiophorite, and birnessite. Nacrite, kaolinite, illite, smectite, and I/S are found in between quartz vein and enclosing kaolins. Among these minerals, plagioclase, quartz, hornblende, muscovite, magnetite, and ilmenite are the residual materials which escaped weathering during the formation of kaolin deposits.

Most kaolins consist chiefly of halloysite (10 Å type) and kaolinite (together 60-95%) with variable content of other minerals such as illite (up to 15%), vermiculite (up to 10%), plagioclase (up to 30%), gibbsite (up to 30%), hornblende (up to 3%), quartz (up to 4%), and amorphous material (allophane and silica) (up to 20%). Goethite aand hematite are also important constituents of the reddish brown and colored kaolin. Quantitative modal analyses by Chung's matrix-flushing method (Chung, 1974a) of kaolin samples are given in Table 1.

Table 1. Quantitative mineralogical analyses of kaolins from Hadong-Sancheong, Korea

Mineral	1	2	3	4	5	6	7	8	9
Halloysite	18	15	20	20	40	38	53	57	57
Kaolinite	50	45	40	55	45	43	24	38	19
Illite	15	11					11		12
Vermiculite	4	6	5	5					
Gibbsite			1					1	
Hornblende				3			1	1	
Plagioclase				6		1		1	
Quartz		1	4		1	2			1
Goethite	2	3	10	5	3				2
Hematite	2								2
Amorphous	9	19	20	6	11	16	9	2	7
Total	100	100	100	100	100	100	100	100	100

1: Reddish brown,
2, 3, 4: Yellowih brown,
5, 6: Pink,
7, 8, 9: White

Chemistry of Kaolin

Chemical analyses of some kaolin samples are given in Table 2. The average SiO_2, Al_2O_3, and Fe_2O_3 content of 300 samples is given in Table 3. The white kaolin is considerably pure but the pinkish or brownish kaolin is high in Fe_2O_3 suggesting the high content of iron oxides. Presence of hematite and goethite was proved by X-ray and TEM studies.

Mineralogy

Halloysite: According to previous works (Kim *et al.*, 1964; Sang *et al.*, 1972; Lee *et al.*, 1977; Sang, 1981), halloysite in Hadong-Sancheong

Table 2. Chemical analyses of kaolin samples from Hadong-Sancheong, Korea

	1	2	3	4	5
SiO_2	63.50	40.10	48.40	47.30	50.40
TiO_2	0.25	0.10	0.79	tr	tr
Al_2O_3	10.10	41.40	23.50	19.30	19.40
Fe_2O_3	0.49	0.42	6.72	0.80	0.30
MnO	tr	tr	0.10	tr	tr
MgO	2.01	0.45	3.16	0.98	0.02
CaO	1.55	6.25	0.80	11.40	12.50
Na_2O	4.80	2.49	0.50	3.28	3.25
K_2O	2.10	0.96	1.39	0.16	0.19
H_2O	14.70	7.35	14.10	16.30	13.30
Total	99.50	99.52	99.46	99.52	99.36

1,2: white kaolin, 3: brown kaolin,
4: pink kaolin, 5: yellowish white kaolin.

Table 3. Range of chemical composition of 300 kaolin analyses from Hadong-Sancheong, Korea

	Reddish brown kaolin	Pink kaolin	White kaolin
SiO_2	35.2 – 50.1	32.3 – 50.2	43.6 – 49.7
Al_2O_3	31.0 – 43.2	34.1 – 41.7	33.7 – 40.2
Fe_2O_3	3.0 – 19.8	0.9 – 3.0	0.5 – 1.5

area occurs as 10Å type as well as 7Å type. The X-ray study on the size-fractionated specimens and dried at 100°C shows that halloysite occurs as 10Å type rather than 7Å type in the natural state, and the abundant 7Å phase is due to the disordered kaolinite(Jeong and Kim, 1989). Frequent observation of book morphology under SEM supports such conclusion.

Halloysite in kaolins occurs as tubes of various length and thickness (Fig. 2). Halloysite growing directly on the surface of plagioclase is usually of long tube. Sizes of hallosite tubes are less than 0.1 μm in diameter and 2 μm in length. However, halloysite which was formed by the alteration of scolecite in veins has spherical or crooked tubular morphology (Fig. 3). Halloysite which has been deironated by DCB method shows 0.18-0.71% Fe_2O_3 for 5 different samples with average of 0.4% Fe_2O_3. It suggests that the haloysite itself is considerably pure and that the

Fig. 2. Transmission electron micrograph of halloysite in common kaolin.

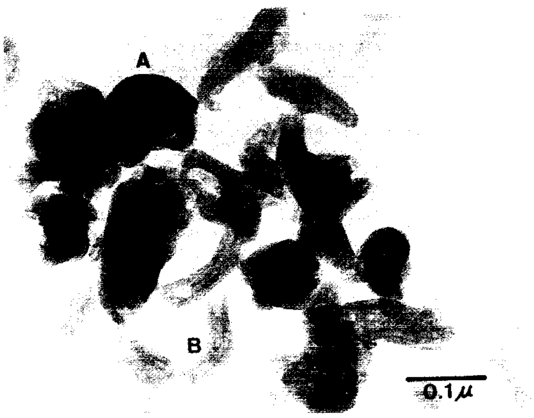

Fig. 3. Transmission electron micrograph of halloysite formed by alteration of scolecite in veins. Halloysite occurs as spherules (A) and crooked tube (B).

iron content of most kaolin samples is due to the impurity iron oxide and hydroxide which grew independently on the halloysite tubes.

Kaolinite: Kaolinite shows book-like morphology in kaolin samples. It is relatively large (25-60 μm) compared with halloysite, but disordered (Jeong and Kim, 1989). Kaolinite which occurs as narrow parallel zones along the quartz veins is well crystallized.

Illite: Illite is widely dispersed in kaolin samples and its content fluctuates from 0 to 20%. Samples with yellow bands usually contain more illite than other ones. Illite occurs usually as relatively large irregular plates ranging from 0.4

to 16 μm. The pure illite sample which concentrated by DCB and 5M NaOH methods shows 1.4% Fe_2O_3.

Vermiculite: Vermiculite is abundantly included in the kaolins which have yellowish brown or greenish yellow bands, streaks, or spots. It is the alteration product of hornblende. Hornblende relics are remained in some vermiculite-rich kaolins. Its size ranges from macroscopic to microscopic. The basal spacing collapses to 10Å at 500°C or by K-saturation. Vermiculite is closely associated with goethite. DCB treatment removed 5.12% Fe_2O_3 from the vermiculite specimen separated from raw sample. The pure vermiculite prepared by DCB method has 8.98% Fe_2O_3. It implies that vermiculite is one of the major impurities affecting the grade of kaolins.

Mixed layer chlorite/vermiculite: Mixed layer chlorite/vermiculite (C/V) occurs in the lower zone than vermiculite. It is usually green, greenish yellow in color and micaceous in morphology. X-ray diffraction pattern of oriented specimen shows that reflection at 29.5Å collapses to 24.5Å at heating to 550°C and expands to 30.1Å at ethylene glycol treatment.

Chlorite: Chlorite is associated with C/V in the green spots or bands in kaolins. Its presence is readily overlooked by the reflections of C/V or vermiculite, but the intensity of 14Å reflection enhances at heating to 550°C (Brindley and Ali, 1950).

Montmorillonite: Montmorillonite is rarely found in kaolin, but it is usually found as a zone or mixed zone with illite, kaolinite and/or I/S along the quartz vein in kaolin deposits. The (001) basal reflection expands to 17Å and 18Å after treatment with ethylene glycol and glycerol, respectively, and collapses to 10Å after heating to 110°C.

Gibbsite: Gibbsite occurs mainly in the upper reddish brown zone and surface soil. It is contained in the kaolins up to 40%.

Goethite: Goethite is the main impurity mineral which causes the kaolins colored to yellow, yellowish brown, or reddish brown. It is usually associated with vermiculite and illite in the pink zone, and with hematite in the reddish brown zone. It shows lath or star-like forms under TEM. The goethite laths are about 0.05μm in width and below 0.3μm in length.

Fe in the goethite structure is partially replaced by Al. The Al content of goethite ranges from 6 to 25% AlOOH. Goethite in the reddish brown zone contains higher Al than that in the pink zone.

Hematite: Hematite is the red coloring agent in kaolins. It occurs mainly in the reddish brown zone. It has the subrounded or hexagonal plates of 0.2-1.0μm in diameter.

The Al content of hematite ranges from 5 to 8% Al_2O_3. The substitution of Fe by Al in hematite decreases with depth as in goethite.

Ferrihydrite: Ferrihydrite is an iron oxide having the composition of $5Fe_2O_3 \cdot 9H_2O$ (Chukhrov *et al.*, 1973). Its crystallinity is very poor so that it is difficult to detect with X-ray diffraction, but can be selectively extracted with acid oxalate which dissolves amorphous Fe (Schwertmann and Taylor, 1977). The ferrihydrite occurs mainly in the reddish brown zone. Its content ranges from 0.06 to 3.65%.

Manganese oxides: Small manganese oxide nodules are frequently found in kaolins. They consists of lithiophorite and/or birnessite.

Genesis

The origin of Hadong-Sancheong kaolin deposits has been regarded as due to the weathering of the anorthositic rocks by Kim and Kim(1964), Sang *et al.* (1972), Lee *et al.* (1977), Lee (1986), Kwon (1986), and Jeong (1987). However, Sang (1980) suggested the hydrothermal origin of halloysite in relation to quartz or zeolite veins. Recent study shows that the Hadong-Sancheong kaolins are evidently of weathering origin. The weathering origin of kaolin deposits has been observed in relation to the parent rocks, topography, climate, and mineral distribution.

Relation to parent rocks: Distribution of kaolin deposits shows that the chemistry and fabric of the rocks are important factors for their formation. It is important to note that the kaolin deposits are confined to the area of anorthositic rocks. Halloysite is nearly always detected as thin or thick crust on the outcrops of the anorthositic rocks. It implies that the calcic plagioclase-rich rocks were the most pertinent material for the formation of kaolin minerals.

The main kaolin deposits are developed along the rocks which were subjected to severe cataclastic deformation. The plagioclase grains are considerably crushed and sheared in the anorthositic rocks underlying the large kaolin deposits. Many dykes are also found in these cataclastic zone. It implies that the rock fabric also was an important factor for the deep kaolinization of the parent rocks. It is evident that the small and large fissures in rocks facilitated the penetration of groundwater and its reaction with parent rocks, resulting in the rapid formation of thick kaolin deposits. The occurrence of high-grade kaolins near the abundant dykes suggests that dykes served for the better movement of groundwater and accelerated the kaolinzation.

Relation to topography: Distribution of kaolin deposits also implies that the topography may be important factor for the formation of kaolin deposits. The kaolin deposits are generally developed on the low and gentle topography with a slope of less than 17° between 100m (the local base) and 350m in altitude. They are also frequently found in the flat plains. However, kaolin deposits are also found on the higher land than 350m, if there is a large flat or gentle topography. Such phenomena suggest that the gentle topography served for the conservation of groundwater for reaction with parent rocks and long-continued preservation of already formed kaolins for the large kaolin deposits.

Relation to climate: The annual rainfall in the Hadong-Sancheong area is about 1,500mm. it may be enough groundwater to kaolinize the anorthositic rocks. The rainfall coming mainly in the hot humid summer could facilitated the kaolinization of parent rocks.

Relation to mineral distribution: Mineral zoning is recognized megascopically and microscopically in the profiles of the kaolin deposits. Three clay zones, that is, the upper reddish brown clay, middle pink clay, and lower white clay zones, are generally developed in the profile. The mineral compositions of three zones are summarized in Table 4. The distribution of somewhat different mineral species in different zones in the profile suggests the variable physicochemical environment with depth.

Table 4. Mineral distribution in the profile of the Hadong-Sancheong kaolin deposits.

Zones in profiles	Minerals
Reddish brown zone	halloysite, kaolinite, gibbsite vermiculite, hematite, goethite ferrihydrite
Pinkish zone	halloysite, kaolinite, vermiculite, C/V, illite, goethite
White zone	halloysite, chlorite, C/V, illite
Parent rocks (Anorthositic rocks)	plagioclase, amphiboles, sericite

The upper zone which is characterized by gibbsite, kaolinite, ferrihydrite, and hematite is usually above the watertable and in the oxidation state. The leached iron is oxidized and accumulated as iron oxides resulting in the reddish brown zone. Presence of gibbsite in this zone indicates an intensive leaching of silica leading to enrichment of aluminum.

The middle zone which is characterized by goethite is in the weak oxidation state. The iron released at the time of dissolution of amphibole has been precipitated as goethite on newly formed vermiculite and C/V.

The lower zone is characterized by the absence of iron oxides. It suggests that this zone is in the less oxidation state. The iron released at the time of formation of chlorite, and C/V from amphibole, is not precipitated as stable oxides.

Various clay minerals are also found in zones along the quartz veins of a few centimeters thick. The entire zone is usually less than 1 m, usually several tens of centimeters in thickness. The zoning is of three types: 1) Quartz-illite-I/S-kaolinite-nacrite. 2) Quartz-I/S,smectite-kaolinite, nacrite 3) Quartz-I/S, smectite, laumontite, 10Å halloysite. It is important to note that no clay zoning is found in the fresh parent rocks along quartz vein. This implies that the clay zoning along quartz vein is the weathering product.

Based on the above discussion, it can be concluded that the kaolin in the Hadong-Sancheong area has been formed by supergene weathering from the anorthositic rocks. The factors for the formation of kaolin deposits of the area are 1) the presence of the cataclastically deformed anorthositic rocks, 2) the low flat or gentle topography, 3) the favorable climate, and 4) the long preservation of kaolins without a considerable erosion.

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