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Evolution of Glaucony in the Tertiary Marine Sediments in the Pohang Area, SE Korea

포항지역 제3기 해성퇴적층에서의 해록석 진화

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ABSTRACT : Glauconization was investigated by morphological and chemical analysis of glaucony from the Pohang Tertiary marine sediments. The glaucony, which is present sparsely in turbidite sandstones, shows two distinct stages in morphology and chemistry. Crystallization of glaucony starts preferentially from the interior of pellet-like grains, then to the outer region of the grains with increasing K and Fe, and decreasing Al. Furthermore, smectite-like glaucony is evolved into illite-like glaucony through interstratified glauconite-smectite with increases in K and Al, and decrease in Fe.

Key words : glauconization, glaucony, pohang, marine sediments, interstratified glauconite-smectite

요약 : 포항지역 제3기 해성퇴적층에서 산출되는 해록석의 생성과정을 해록석 형태와 화학분석에 의하여 연구하였다. 사암 내에서 산발적으로 산출되는 해록석은 뚜렷한 두 단계의 광물형태와 성분변화가 나타난다. 해록석의 결정화는 펠렛형태 입자의 내부에서 우선적으로 시작되어 외부쪽으로 K와 Fe 성분이 증가하고 Al 성분이 감소하는 경향을 보인다. 더욱이 스멕타이트와 비슷한 조성의 해록석은 K와 Al 성분이 증가하고 Fe 성분이 감소하면서 해록석/스멕타이트 혼합층광물 단계를 거쳐서 일라이트와 비슷한 조성의 해록석으로 진화하여 간다.

주요어 : 포항지역, 제3기 해성퇴적층, 해록석, 해록석-스멕타이트 혼합층 광물

Introduction

Glauconite is a hydrous Fe and Mg aluminous silicate clay mineral with a mica-like structure, which is characterized by significant amounts of Fe^{3+} in octahedral sheets and K in interlayers (Odom, 1984; Strickler and Ferrell, 1990). Glauconite can be easily differentiated from illitic minerals on the basis of structural, chemical and morphological characteristics, and geological occurrence. This mineral is commonly encountered in sandy sediments as an accessory mineral, and in most cases these sediments contain sedimentological evidence of marine origin (Burst, 1958a, 1958b; Hower, 1961; Odin and Matter, 1981; Odin, 1988). In other words, glauconite is developed commonly by contact with marine environment. Glauconite has been interchangeably used as a morphological or mineralogical term. The term of glauconite was used to describe not only a mineral species but also almost all types of greenish-colored grains or pellets in sediments. In addition, glauconite with expandable layers are frequently found in nature and therefore can not be described as a single phase glauconite but as interstratified glauconite-smectite. Odin and Matter (1981) proposed that the term "glauconite" be abandoned and introduced the general term "glaucony" for all morphological forms and the terms "glauconitic smectite" and "glauconitic mica" as end members of the glauconitic minerals. The similar case is that calcite is distinguished from limestones. The term glaucony is used in this paper.

Recently, Lee et al. (2002) reported the occurrence of glaucony in recent sediments of the Yellow Sea, whereas Lee and Paik (1997) reported it in the Ordovician formation. The present work also reports the occurrence of glaucony in the Tertiary marine sediments in the Pohang area. The Pohang basin is a marine sequence of clastic sediments which accumulated during Middle Miocene (Fig. 1). The formation of this basin is closely related with the origin of the East Sea (Son, 1996). In the basin, many drill holes have been made for hydrocarbon exploration. These drill holes

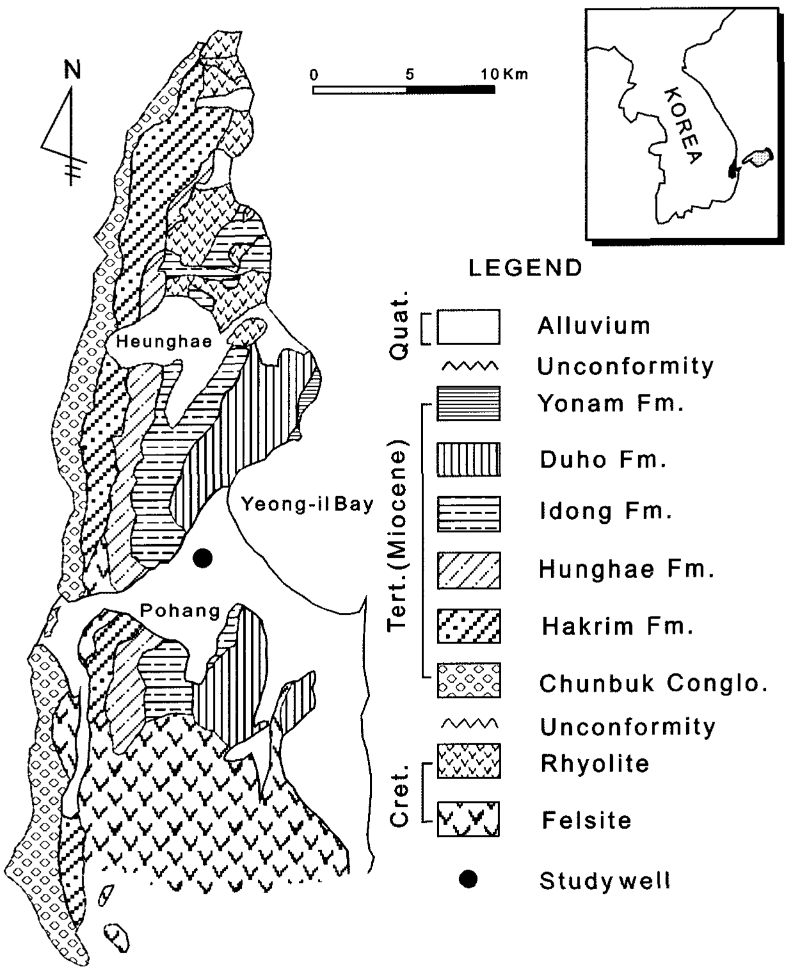

Fig. 1. Geologic map and the location of study well.

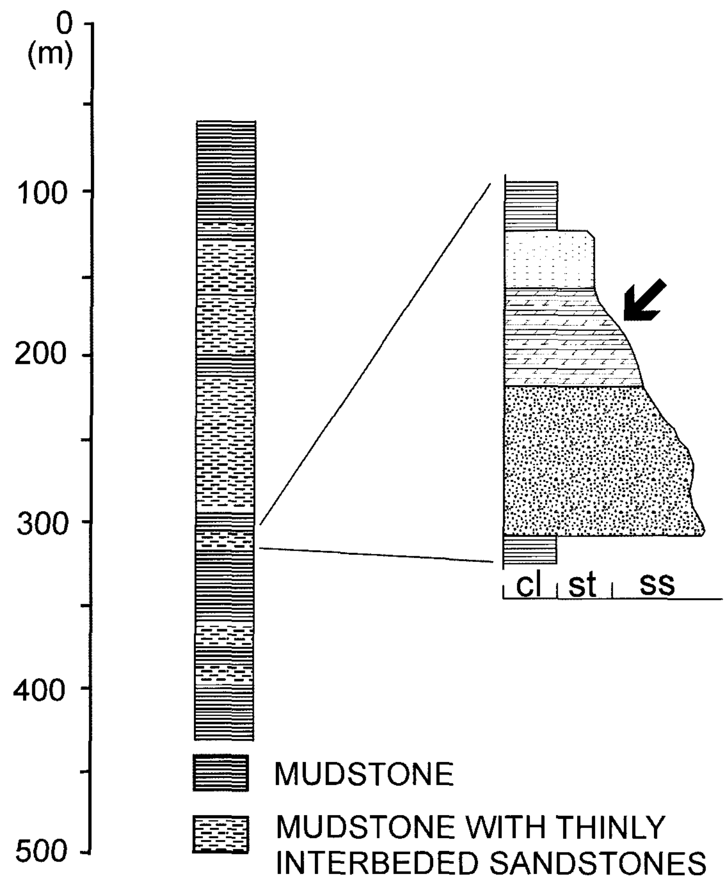

Fig. 2. Lithology of the study well. Arrow indicates the location of analyzed samples. cl: clay, st:silt, ss: sand.

revealed that the Tertiary marine sediments has a maximum depth of about 700 m and are dominated by fine-grained mudstones with thinly interbeded sandstones which show a fining upward sequence (Fig. 2). Glaucony are commonly found in the interbeded sandstones. The objective of this paper is to document the process of glauconization by investigating the chemistry of the glaucony from the Tertiary sediments.

Materials and Methods

Sandstones were collected at a depth interval of 300-305 m of a drill core that penetrated the Miocene marine sediments (Fig. 2). The sandstone samples were prepared into thin sections to observe the occurrence of glaucony under the polarizing microscope.

Electron microprobe analysis of glaucony was performed on polished thin sections of sandstones using a JEOL JCXA-733 electron microprobe. Prior to analysis, areas containing glauconites were located and mapped in polarized light. Many spots, traversing grains of glaucony, were analyzed using a 5 μm diameter beam and a 15 keV accelerating voltage. Structural formulae for glaucony were calculated assuming that all the Fe was ferric and using the total anion equivalency method based on the $O_{20}(OH)_4$.

X-ray powder diffraction data of glauconite could not be obtained due to difficulty in separating glaucony from rock specimens.

Results

Occurrence

Glaucony is sparsely distributed in the sandstones that are mainly cemented by carbonates. The glaucony is easily identified by their indigenous green color and pellet shape under the polarizing microscope. In the rock samples, however, there is difficulty in finding the glaucony minerals because of rarity in abundance. Most of the glaucony pellets have a smooth, rounded or lobate morphology and are internally composed of randomly arranged clay aggregates. The glaucony occurs as sand-sized grains with dark green color. Glaucony is very susceptible to compaction because the pellets of glaucony are quit soft. Therefore, deformation of glaucony is generally observed in the thin sections (Fig. 3). The glaucony often includes crystals of pyrites in the pellet-like grains. Shrinkage cracks is commonly found at the surface of glaucony. Rarely, well developed flakes are present as a part of the grain. Attention was paid to the occurrence and features of grains to analyze and interpret the process of glauconization.

Chemistry

Many grains of glaucony were analyzed with electron microprobe. However, a detailed analysis was concentrated on two grains that have different features between grains or within a

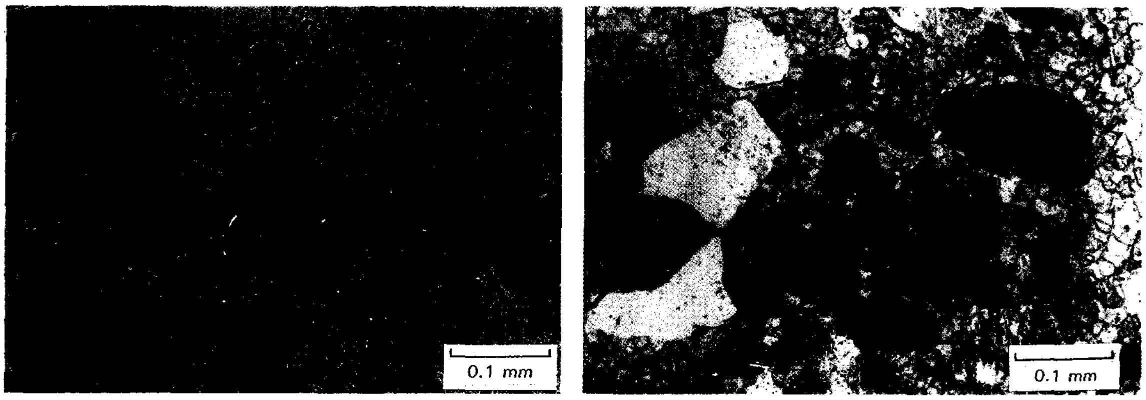

Fig. 3. An extended glaucony due to compaction (left), and broken and cracked glaucony (right).

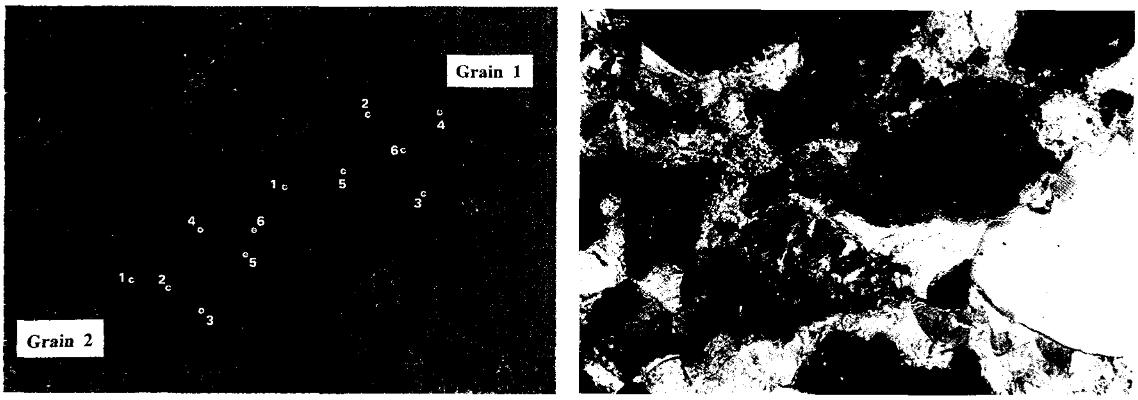

Fig. 4. Analyzed grains of glaucony (left: open nicol, right: crossed nicols). Numbers indicate points analyzed. Note well-developed flakes in the area of points 4, 5, and 6 of grain 2.

Table 1. Electron microprobe analyses of glauconite grain 1

Analysis	1	2	3	4	5	6
SiO_2	54.15	54.14	54.40	51.65	52.12	50.39
Al_2O_3	12.71	12.77	11.30	10.34	11.30	10.21
Fe_2O_3	14.49	15.51	14.87	15.66	16.73	16.14
MgO	5.33	4.96	4.90	4.74	4.91	4.70
MnO	0.00	0.06	0.00	0.02	0.04	0.00
TiO_2	0.31	0.18	0.13	0.20	0.19	0.16
CaO	0.42	0.57	0.41	0.47	0.32	0.36
Na_2O	0.17	0.20	0.17	0.17	0.14	0.19
K_2O	6.95	7.29	7.12	7.00	7.78	7.65
Total	94.51	95.68	90.30	90.24	93.52	89.84
		Numbers of cations on the basis of 22 oxygens				
Si	7.42	7.37	7.43	7.49	7.34	7.40
Al(IV)	0.58	0.63	0.57	0.51	0.66	0.60
Σtetra	8.00	8.00	8.00	8.00	8.00	8.00
Al(VI)	1.47	1.42	1.35	1.25	1.22	1.17
Fe	1.49	1.59	1.62	1.71	1.77	1.78
Mg	1.09	1.01	1.06	1.02	1.03	1.03
Mn	0.00	0.01	0.00	0.00	0.00	0.00
Ti	0.03	0.02	0.01	0.02	0.02	0.02
Σocta	4.08	4.05	4.04	4.01	4.05	4.00
Ca	0.06	0.08	0.06	0.07	0.05	0.06
Na	0.04	0.05	0.05	0.05	0.04	0.05
K	1.21	1.27	1.31	1.29	1.40	1.43
Σinter	1.32	1.40	1.42	1.41	1.49	1.54
Chg(tetra)	0.58	0.63	0.57	0.51	0.66	0.60
Chg(octa)	0.80	0.86	0.92	0.97	0.88	1.00
Chg(inter)	1.38	1.49	1.49	1.49	1.53	1.60

grain (Fig. 4). These grains include all the features representative of the occurrence of glaucony. One of the grains has a form of rounded pellet with cracks at the surface (Fig. 4, grain 1), whereas the other has well developed flakes and an angular surface (Fig. 4, grain 2). The electron microprobe analyses were obtained from the spots traversing the grains. The results are presented in Table 1 and 2. Chemical fomular was calculated from the results of analysis. All Si^{4+} were assigned to tetrahedral sites, and preferentially Al^{3+} filled the remainder of the tetrahedral sites. The remaining Al^{3+} were allocated to the octahedral sites. In addition, layer charges were also calculated based on the chemical formula. Al content in the tetrahedral sheet varies in the wide range of 0.51 to 1.12 on the basis of $O_{20}(OH)_4$. The octahedral sheet has Fe atoms between 1.11 and 1.78, and Mg atoms between 0.62 and 1.17 on the basis of $O_{20}(OH)_4$. Fe and Mg contents are reversely proportional to the Al contents. Moreover, K in interlayer is reversely proportional to Si in tetrahedral sheet, probably indicating that the substitution of Al for Si in tetrahedral sheets is compensated for by the addition of K to interlayers. This is the same reaction as that in the well-known illitization reaction (Hower et al., 1976; Son, 1996; Son and Yoshimura, 1997; Son, et al., 2001). The analyses were plotted within the triangular composition diagram muscovite-celadonite-pyrophyllite proposed by Newman and Brown (1987) (Fig. 5). The muscovite end-member

Table 2. Electron microprobe analyses of glauconite grain 2

Analysis	1	2	3	4	5	6
SiO_2	51.51	53.44	53.44	50.57	49.75	49.43
Al_2O_3	13.45	10.90	10.70	18.64	20.87	17.35
Fe_2O_3	14.40	16.46	16.46	11.80	10.70	12.13
MgO	4.97	5.68	5.23	3.65	3.01	3.71
MnO	0.00	0.01	0.03	0.03	0.00	0.02
TiO_2	0.29	0.26	0.24	0.14	0.13	0.19
CaO	0.49	0.38	0.47	0.30	0.29	0.30
Na_2O	0.13	0.13	0.17	0.13	0.11	0.15
K_2O	7.22	7.26	7.38	8.36	8.49	7.89
Total	92.77	94.51	95.01	93.62	93.35	91.16
	Numbers of cations on the basis of 22 oxygens					
Si	7.27	7.41	7.48	7.00	6.88	7.04
Al(IV)	0.73	0.59	0.52	1.00	1.12	0.96
Σtetra	8.00	8.00	8.00	8.00	8.00	8.00
Al(VI)	1.50	1.19	1.22	2.05	2.29	1.95
Fe	1.52	1.72	1.71	1.23	1.11	1.30
Mg	1.03	1.17	1.07	0.75	0.62	0.79
Mn	0.00	0.00	0.00	0.00	0.00	0.00
Ti	0.03	0.03	0.03	0.01	0.01	0.02
Σocta	4.08	4.10	4.03	4.05	4.04	4.06
Ca	0.07	0.06	0.07	0.04	0.04	0.05
Na	0.04	0.04	0.05	0.04	0.03	0.04
K	1.29	1.28	1.30	1.48	1.50	1.43
Σinter	1.40	1.38	1.41	1.56	1.57	1.52
Chg(tetra)	0.73	0.59	0.52	1.00	1.12	0.96
Chg(octa)	0.75	0.84	0.96	0.60	0.50	0.60
Chg(inter)	1.48	1.43	1.48	1.60	1.61	1.57

represents charge arising from substitution in the tetrahedral sheet, the celadonite end-member represents charge arising from octahedral substitution, and the pyrophyllite end-member is uncharged. As shown in the diagram, Most of the analyses fall within the region of glauconite-smectite as well as glauconite. This suggests that composition of the Pohang glaucony is constrained by variation of component layers in interstratified glauconite-smectite. In addition, it should be noted that the interstratified glauconite-smectite is evolved to glauconitic illite with a tendency toward the tetrahedral charge end.

Discussion

Two grains of glaucony were chemically analyzed by electron microprobe to understand the process of the formation of glaucony in the Yeonil sediment (Fig. 4; Table 1 and 2). The analyses were obtained from the points traversing the grain. One of the two grains contains well-developed flakes (Fig. 4, grain 2). The analysis shows that composition varies even within the same grain. This variation can probably indicates the process of glauconization and the transformation of glauconitic smectite into glauconitic illite.

In the number 1 grain of Figure 4, there is a tendency for the contents of K and Fe to increase, and for the content of Al to decrease from the outer part to the inner (Fig. 6). In the number 2 grain, the analyses of the flakes (point 4, 5, and 6) are reversely related with

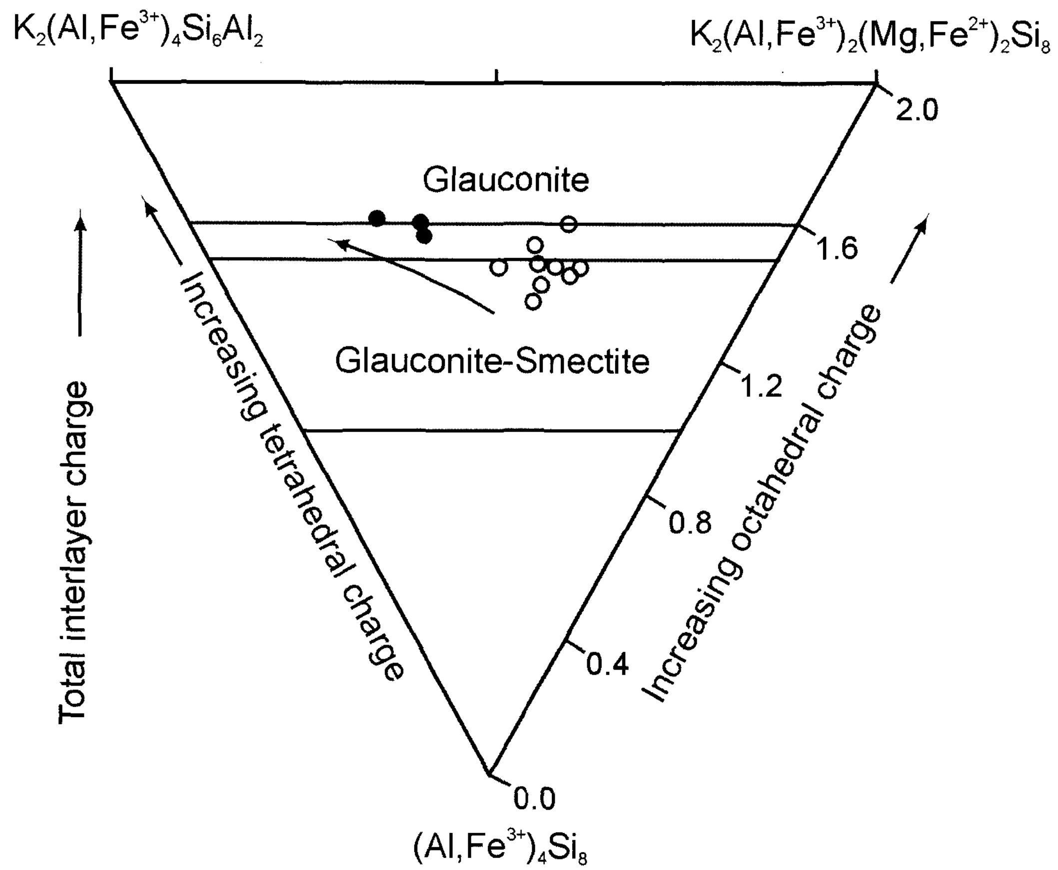

Fig. 5. Position of analyses of glaucony in the triangular composition diagram. Closed circles indicate the analyses of 4, 5, and 6 in grain 2 of Figure 4.

those of the other part (points 1, 2, 3) for the contents of K and Fe, whereas directly related for the content of K and Al (Fig. 7).

The relationships between components are considered further from the analyses. Fe and Al(VI) contents are reversely proportional to each other. On the other hand, Fe and K contents are reversely proportional to each other in the analyses of flakes (Fig. 7, point 4, 5, and 6), whereas they are nearly proportional to each other in the analyses of the other part of the grain. Furthermore, K and Al(VI) contents are also directly proportional to each other in the analyses of the flakes, while in the other part of the grain they are reversely proportional to each other. Fe and Al(IV) components are reversely proportional to each other in the analyses of the flakes. K and Al(IV) contents are proportional to each other in all analyses (Fig. 6 and 7).

Glaucony has been known to be formed in the marine sediment (Odin and Matter, 1981; Odin, 1988). In particular, Odin (1988) investigated the process of glauconization in the Quaternary marine sediment. According to him, glaucony is changed from ochreous green through light green to dark green in color while the burial depth is increasing. The evolution leads to the increase of the volume of grain, and thus provokes cracks in the grain surface (Fig. 8). The green pellets with a number of poorly organized globules due to the initial biogenic material dissolved are changed into homogeneously dense grains during the evolution. In the sense of mineralogy, Odin (1988)

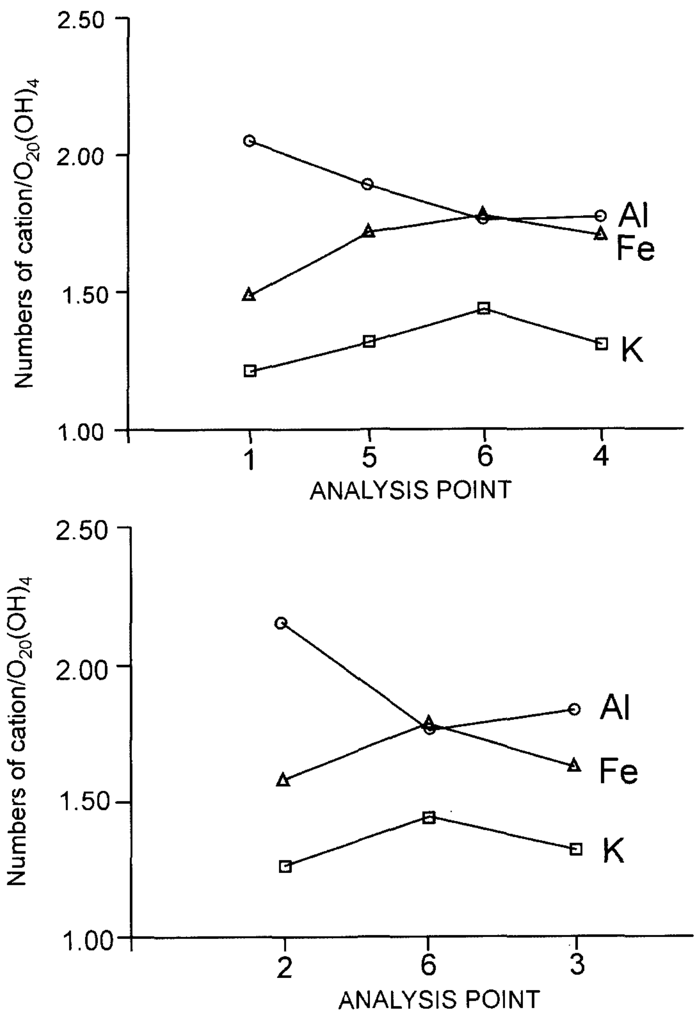

Fig. 6. Compositional variation within grain 1 in Figure 4.

shows that the initial glaucony at the shallow depth is dominated by the composition of kaolinite. The process of glauconization leads the initial glauconite to a 14Å smectite-type mineral, and then to the illite-type mineral. The model suggested by Odin (1988) can be constructed by a three-stage process of glauconization in the sense of morphology and crystal growth (Fig. 8). In stage 1, the fecal pellet is deposited in a shallow environment that is made up of the mud sediment, probably enrich in biogenic particles since the mud-eaters most probably select a mud rich in organic matter. The initial fecal pellets are dominated by kaolinite with quartz and carbonate mineral. In the next stage, a large proportion of the initial biogenic material is dissolved. Thus, this leads to a high porosity in the pellets. In the last stage, well-shaped microcrystals (rosette or flakes) are present in most of the grains but for the periphery. There are also a volume increase and cracks in the grain surface as described above. As a result, the great portion of the initial detrital material has been removed and the authigenic material initially crystallized has been rearranged, i.e., recrystallized. Moreover, in the initial stage of glauconization, glauconitic smectite is formed, that is rich in Fe content

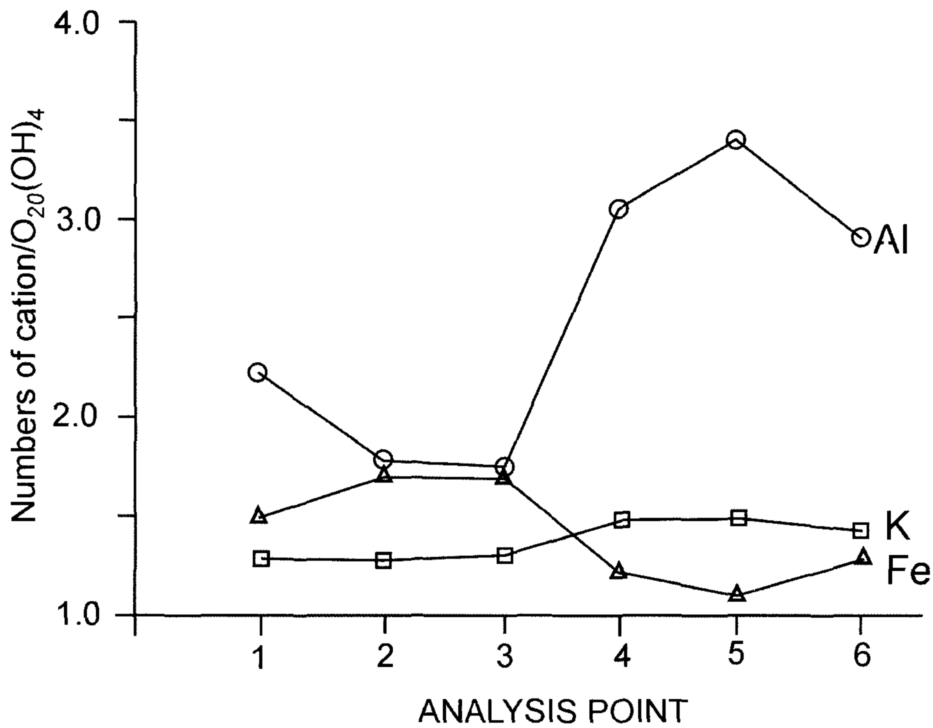

Fig. 7. Compositional variation within grain 2 in Figure 4.

but poor in K content. In the next stage, potassium is introduced into the glauconitic smectite and layer distance is reduced, and thus the glauconitic smectite is actively changed into glauconitic illite. At that time of the transformation, The glauconitic smectite is evolved by way of interstratified glauconite-smectite into glauconitic illite. It is a quite similar process to the transformation of smectite into illite. Most of the Pohang glauconites show the chemical composition of interstratified glauconite-smectite. Only a few analyses are plotted in the region of glauconite (Fig. 5).

The interpretation of the chemical data can be based on Odin's premise described above. Judging from the evolution model by Odin (1988) and the results of the analysis, glaucony crystals are developed preferentially in the interior of the grain compared with the outer part. Furthermore, glauconitic smectite is evolved into glauconitic illite through interstratified glauconite-smectite (Fig. 5). The glauconite from this study area has two distinct stages of morphology and variation in chemical composition. The first stage is dominated by interstratified glauconite-smectite with no flakes. In the stage, there are the increases of Fe and K content and the decrease of Al content through the evolution. The second stage is characterized by the development of flakes of glauconite. There are tendencies for Fe content to decrease and for K and Al contents to increase.

Glaucony has been regarded as one of the most reliable indicator of low sedimentation rate in marine settings (Amorosi, 1995). In the Pohang sediment, however, glaucony is present in a sandstone sequence of turbidites with a low concentration (Fig. 2). This indicates that the Pohang glaucony was reworked by turbidity current, then evolved with increasing depth of burial. Glauconization starts from a K-poor glauconite-smectite, which progressively evolves toward an end member constituted by a K-rich glauconitic mica. K-feldspar and detrital muscovite can be a likely source for the potassium.

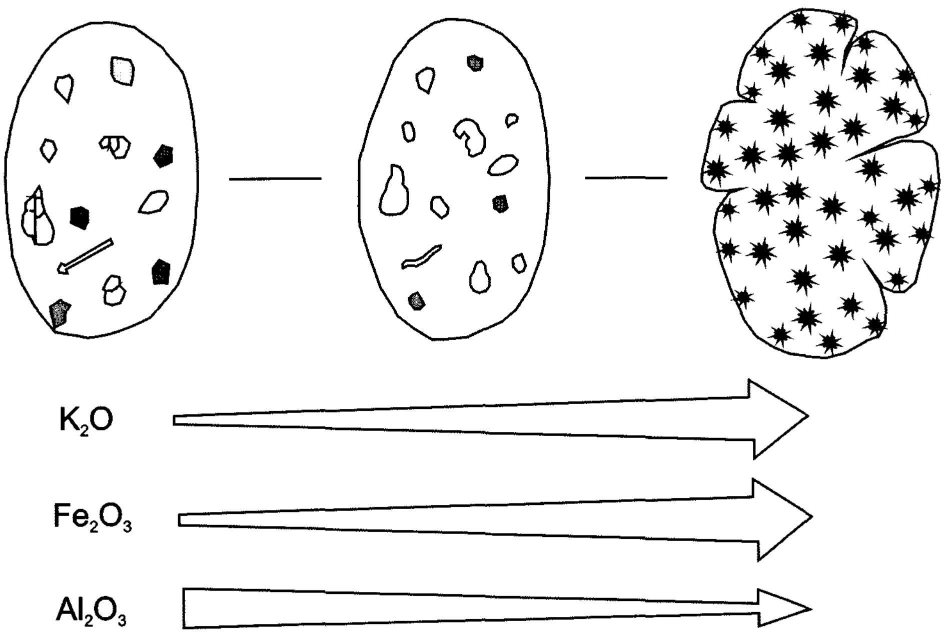

Fig. 8. Glauconization model suggested by Odin (1988). Thickness of arrows implies the quantity of components.

Conclusions

This study shows that the Pohang glaucony supports an evolution model of glaucony suggested by Odin (1988). Development of glaucony crystals occurs preferentially in the interior of the grain compared with the outer part. This glauconization proceeds with increasing K and Fe contents, and decreasing Al content.

In addition, glauconitic smectite is evolved into glauconitic illite through interstratified glauconite-smectite, accompanying increases in K and Al contents, and deceasing Fe content. This reaction is quite similar to the well-known illite-smectite reaction.

As a whole, the evolution of glauconite in the Pohang Tertiary marine sediments can be characterized chemically by increase in K content.

Acknowledgments

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