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CLAY MINERALIZATION OF THE NEOGENE AGED VOLCANICS OF THE NORTH-EASTERN SİVRİHİSAR (MÜLK-DEMİRCİ)

Zehra KARAKAŞ*, Baki VAROL* and Sonay BOYRAZ*

ABSTRACT.- The Neogene lacustrine units in the Mülk-Demirci region (NE Sivrihisar) are composed of detrital-evaporitic, volcanic and pyroclastic rocks. The Miocene volcanic rocks consist of lava flows of basalt and andesite composition and pyroclastic rocks are made up of agglomerate, tuff, altered tuff and tuffitic sandstone interbedded with altered claystone. These volcanic units change to dolomite, dolomitic limestone, marl and gypsiferous levels to the top. Some alteration zones in tuffs such as iron oxidation, limonitization, carbonatization and argillization are very noticeable with their yellow and red colors. Smectite is the main clay mineral which was formed as a result of alteration of volcanic and pyroclastic rocks in the study area. In some samples, smectite is accompanied mostly by feldspar, dolomite, calcite, opal-CT, quartz and partly by illite, gypsum and analcime. In the electron microscopy (SEM) studies, smectite morphology was distinguished with platy flakes that are developed as a honeycomb texture. In addition, it was also determined in SEM images that smectite mineralization mostly develops in dissolution voids and along fracture and fissures of volcanic glass or on the feldspar minerals. On the basis of field and laboratory studies, formation of smectite was found to be controlled by feldspar and volcanic glass which comprises the main component of tuffaceous units. Under hot and dry climate conditions, alteration of volcanic material in the lake water had an important role in the formation of smectite. Smectite was formed by hydrolysis of volcanic glass and alteration of feldspar.

Key words: Clay mineralization, Neogene, paleoclimate, pyroclastics, smectite, Sivrihisar, volcanic.

INTRODUCTION

The study area is located around the Mülk and Demirci villages (Ankara I27 c4, d3) at 25 km east of Sivrihisar in central Anatolia (Figure 1). Studies in the region were mostly conducted on sepiolite occurrences in the vicinity of study area (Bilgin, 1972; Kulaksız, 1981; Ece and Çoban, 1990; Karakaş, 1992; Gençoğlu et al., 1992; Yeniyol, 1992, 1993; Çoban, 1993; Bellance et al., 1993; Karakaş and Varol, 1993, 1994; Gençoğlu and İrkeç, 1994; Ünlü et al., 1995; Gençoğlu, 1996). The geological characteristics of the basement rocks and Neogene units in the region were studied by Weingart (1954), Erol (1955), Brelie (1956), Umut et al. (1991) and Gözler et al. (1996). Özbaş (2001) investigated mineralogical and geochemical properties of zeolites and associated minerals in the Mülk-Oğlakçı area. Temel (2001) studied geochemistry and petrology of the Miocene alkaline volcanism in the Oğlakçı area.

The aim of present work is to investigate clay mineralization in the Neogene volcanic and pyroclastic rocks around the Mülk-Demirci area and to study the formation conditions of argillization in regard to paleoclimate and bedrock characteristics.

MATERIAL AND METHOD

A total of 35 samples were collected from measured stratigraphic sections of the Karabayırlar Hill I, II, III and Mülk where volcanic, pyroclastic and detrital-evaporitic rocks are widely exposed (Figures 1 and 2). In addition to samples from the measured stratigraphic sections, 30 point samples were also collected from the Çakmak Hill, Çakmakçıkan Hill, Karabayırlar Hill, Hamdere Ridge and Hamam Ridge and around the Asarkale and Mülk villages. Thin sections were made from unaltered rock samples and they were studied with the Leitz brand polarizing microscope to determine their textural and minera-

* Ankara Üniversitesi Mühendislik Fakültesi Jeoloji Müh. Böl. 06100 Beşevler/Ankara
karakas@eng.ankara.edu.tr, varol@eng.ankara.edu.tr, boyraz@eng.ankara.edu.tr

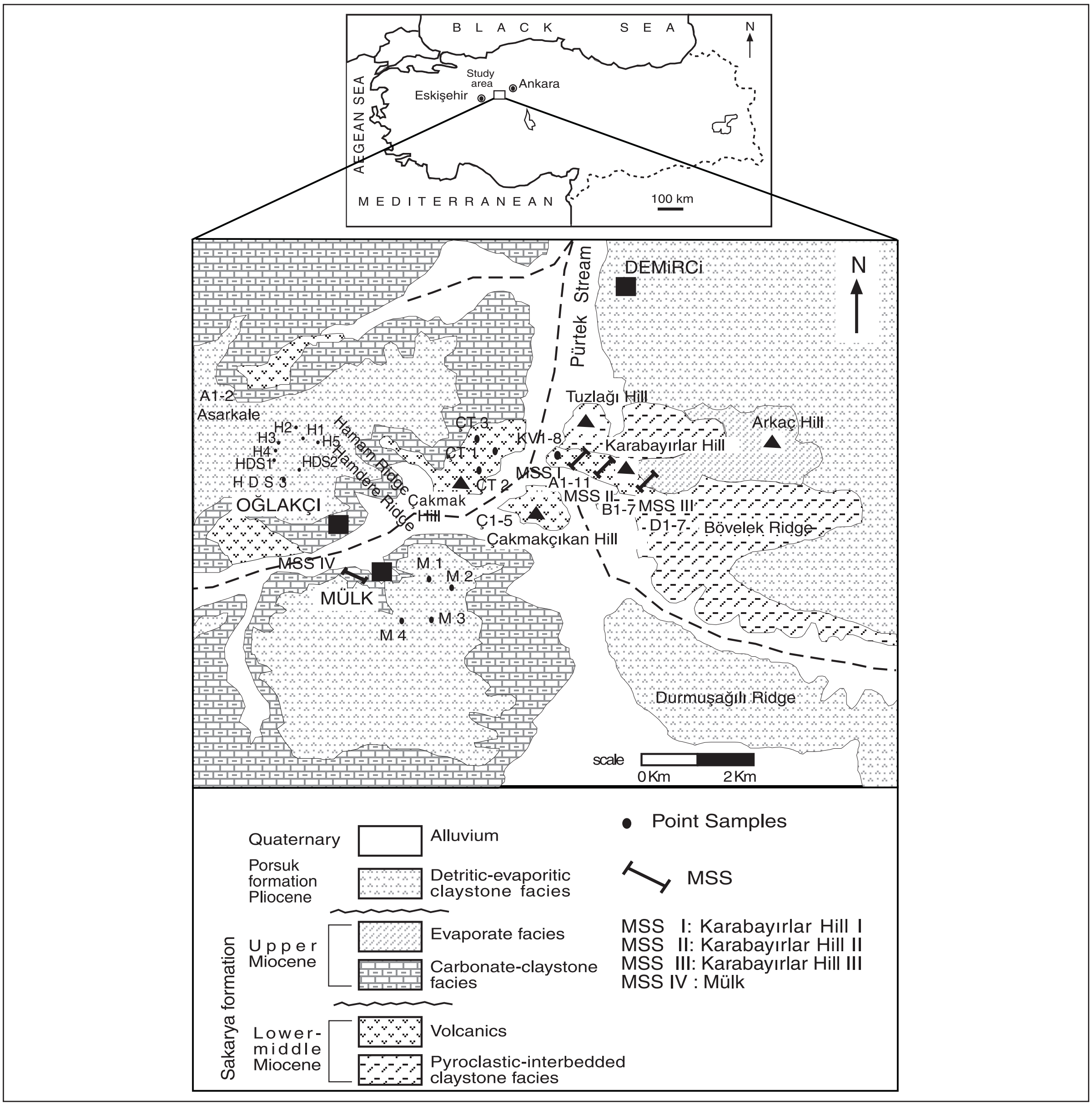

Figure 1- Location and geology map of the study area (Modified from Gözler et al., 1996).

logical properties. Volcanic and pyroclastic rocks were classified according to Streckeisen (1976, 1979) and Schimid (1981), respectively. Mineralogical compositions of clay minerals and partly and completely altered rock samples were determined with Rigaku-Geirgeflex X-Ray Diffractometer (XRD) device with whole rock (45 samples) and clay fraction (35 samples) analyses. Semi-quantitative mineralogical analysis of the samples was done (Gündoğdu, 1982) by the external standart method (Brindley, 1980). The morphologic properties and textural characteristics of samples which are dominated by argillization were studied with JEOL 840 A brand scan-

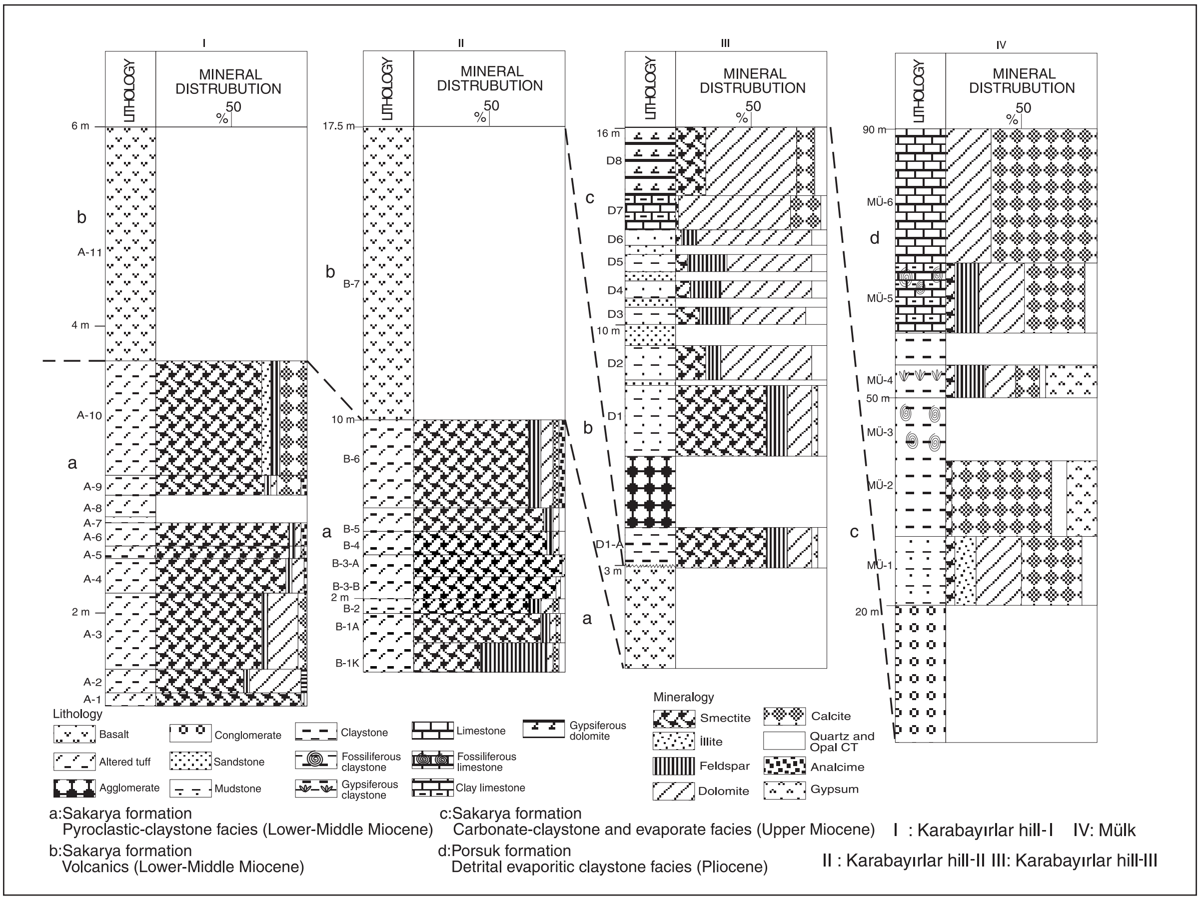

Figure 2- Lithological and mineralogical distribution in the Sakarya and Porsuk formations in the study area.

ning electron microscope (SEM). Moreover, thermal behaviors of clay minerals were determined with Rigaku Analyzer TAS 100 device at the General Directorate of Mineral Research and Exploration of Turkey (MTA).

GEOLOGY

The basement of the Sivrihisar Neogene basin is represented by Paleozoic metamorphic rocks consisting of schist, gneiss and marble and Mesozoic ophiolitic complex, granite and granodiorites (Kibar et al., 1992; Gözler et al., 1996). Exposures of the basement rocks are observed around the Sazak and Baharözü villages at NE of Sivrihisar town, outside of the study area. The Neogene units which have a wide distribution in the region unconformably overlie the basement rocks. In previous geologic and industrial raw material studies conducted in and around the study area, Neogene units were investigated under different formation names. Gençoğlu and İrkeç (1994) described the lacustrine units as the İlyaspaşa (Miocene) and Sakarya (Pliocene) formations while the same units were named as the Porsuk (Miocene) and Hüyüklü (Pliocene) formations by Gözler et al. (1996). In the present work, lacustrine units in the area were described as Miocene Sakarya and Pliocene Porsuk formations (Boyraz, 2004) (Figures 1 and 2). The mappable lithofacies of the Sakarya formation, from bottom to the top, are pyroclastic - interbedded claystone, volcanics, carbonate - claystone and evaporate facies. The pyroclastic - interbedded claystone facies is represented by agglomerate, tuff, altered tuff and tuffitic sandstone. Altered claystone levels and tuff levels which are mostly observed in multi-colorings are repeatedly alternated (Figure 3a, b). In addition to argillization, iron oxidation, limonitization and carbonatization are also detected in tuffs (Figure 3c). These alteration zones are noticeable with yellow, red, greenish yellow and green colors. Volcanics overlying the pyroclastic-interbedded claystone facies are observed as basaltic and andesitic lava flows (Figures 1, 2 and 3a). Basalts show porphyry-afanitic texture and gas vesicles while andesites are characteristic with light pink-beige colors, compact structure and afanitic texture. Basalts and andesites that are observed as partly or completely altered are typical with yellow, red and reddish brown colors. The volcanics in the region are exposed along E-W extending fault systems which were developed as a result of N-S extending compressional regime which was followed by an extensional tectonism (Temel, 2001; Özen and Sarıfakıoğlu, 2003). Basalts and andesites in the study area were dated as Early-Middle Miocene (14-18 Ma) (Temel, 2001). These volcanics are overlain with a low-angle angular unconformity by carbonate-claystone and evaporate facies which is represented from bottom to the top by dolomite, dolomitic limestone, mudstone and gypsiferous units (Figures 1, 2 and 4). Some Gastropod species such as *Planorbarius* sp., *Lymnaea* sp., *Helix mrazeci* Sévastos, 1922, *Abida* sp. and *Mostus* sp. (Upper Miocene) were determined in clay, marl and organic material-rich units in this facies (Taner, 2004). On the basis of data obtained from radiometric and paleontological dating studies, the age of the Sakarya formation is accepted as Miocene.

The Sakarya formation is overlain with a low-angle angular unconformity by detritic-evaporitic claystone facies of the Pliocene Porsuk formation (Figures 1). This facies is composed of a repetition of deposition package consisting of red-gray colored conglomerate-sandstone, gray-green colored claystone, gypsum, gypsiferous mudstone, dolomite, dolomitic claystone and limestone (Figures 1 and 2). Some Gastropod species such as *Valvata crusitensis* (Fontannés, 1886), *Gyraulus* (G.) *ignoratus* Schickum-Puissegur, 1977, *Emmericia rumana* Tournouér, 1880 were determined in green claystone and marl units and the age of the Porsuk formation is accepted as Late Pliocene (Romanian) (Taner, 2004). In addition, in these units, some ostracoda fossils such as *Ilyocypris* sp, *Potamocypris similes* (Müller, 1894), *Pseudocandona* cf. *compressa* (Koch, 1837), *Hemicyprideis dacica grekoffi* Carbonnel, 1971, *Cyprideis* cf. *torosa* (Jones, 1850), *Candona neglecta* Sars, 1888, *Candona* sp. were also found which yielded Pliocene-Early Pleistocene age. Moreover, spore-pollen studies conducted on brown clayey, clay, marl and organic material-rich units yielded the presence of biozone assemblages which indicate Late Pliocene age. These assemblages are Tubuliflorae and Liguliflorae types (Batı, 2004). In addition, this dating was also verified by micro/macro mammalian assemblages such as *Testudo* sp., *Micromys* sp., *Occitamomys* sp., *Prolagus* sp. and *Promimomys* sp. (Saraç, 2004).

Red colored Quaternary units which unconformably set above the Neogene lacustrine deposits are composed of red-brown conglomerate, mudstone, sandstone and alluvium.

PETROGRAPHY

Regarding mineralogical compositions and textural characteristics, volcanic and pyroclastic rocks in the study area were described as tuff, basalt and andesite.

On the basis of their rock fragment, volcanic glass and crystal content abundance, tuffs comprising the dominant lithological assemblage of pyroclastic rocks are investigated as lithic, vitric and crystal tuff. The vitric tuffs are completely composed of volcanic glass. In volcanic glass which is generally observed as fragmented and rounded grains, some alteration products such

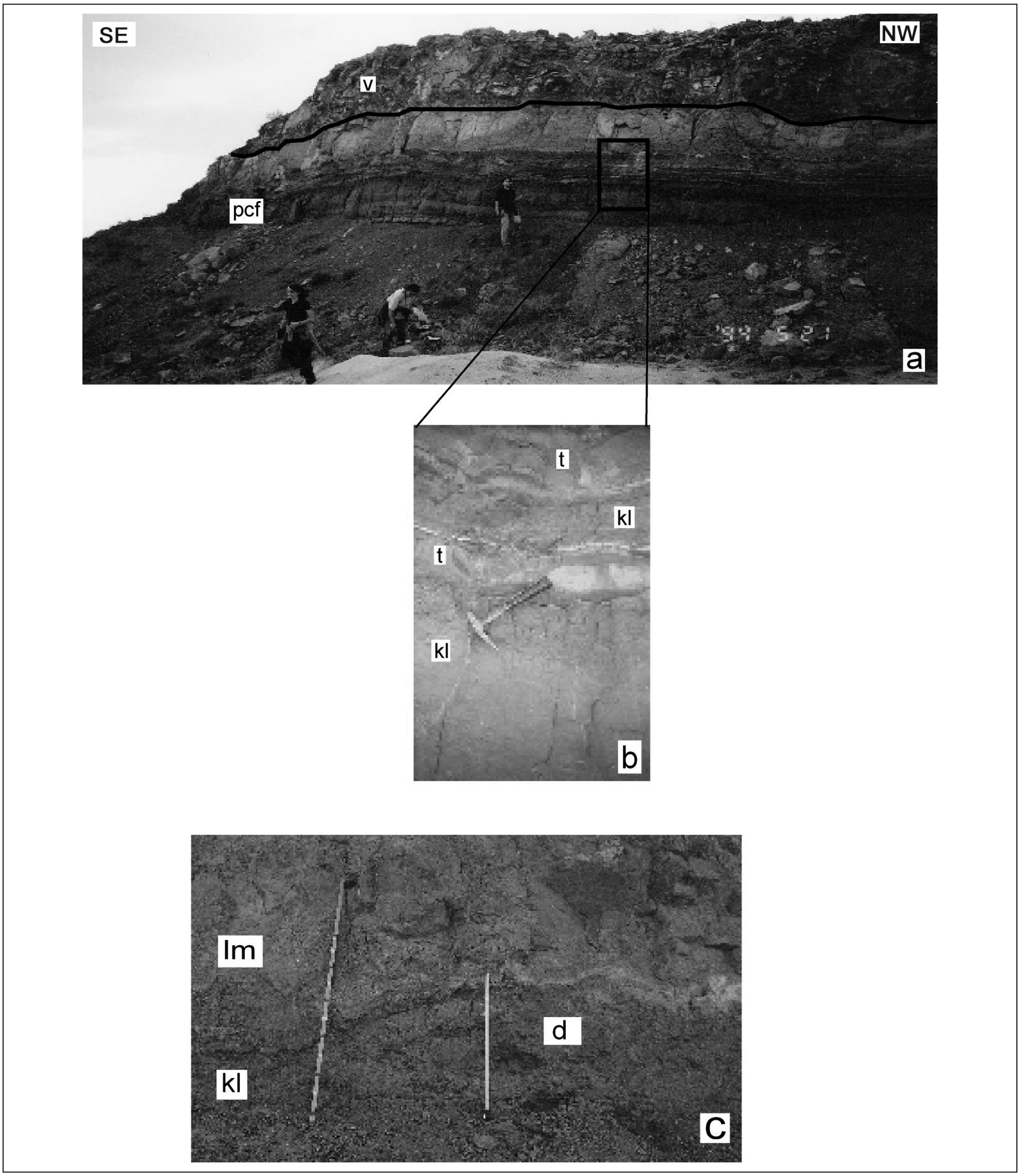

Figure 3- a. Vertical and horizontal relations between pyroclastic-interbedded claystone facies (pcf) and volcanics (v) at the Karabayırlar Hill;
b. Close view of multicolored, altered tuffs (t) and alternated claystone levels (kl);
c. Argillization (kl), limonitization (lm) and iron oxidations (d) in the pyroclastic-interbedded claystone facies.

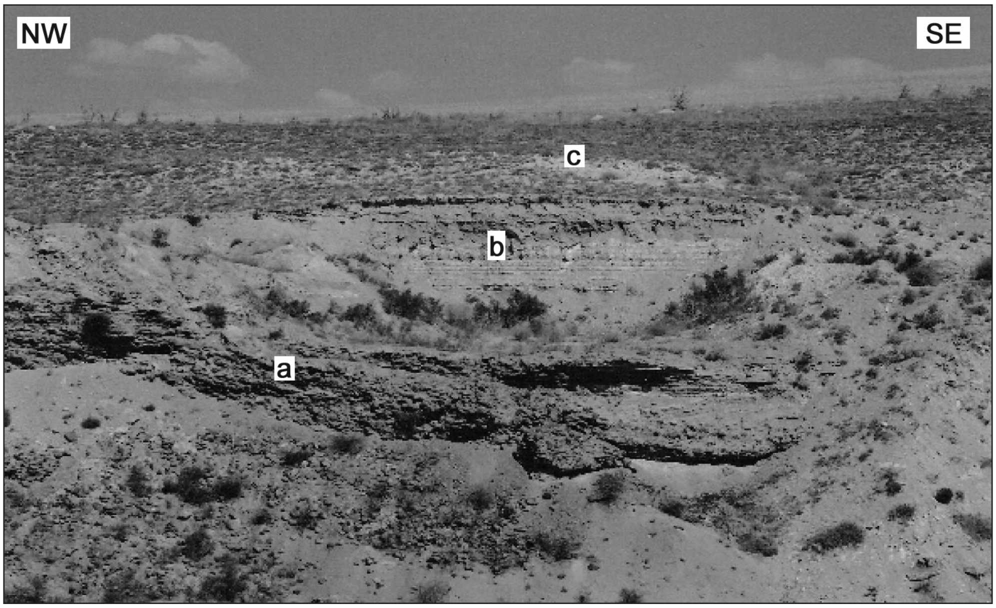

Figure 4- The transition between pyroclastic-interbedded claystone facies (a), carbonate-clay stone (b) and evaporate (c) facies.

as argillization and carbonatization are partly noticeable (Figure 5a). Lesser amount of opaque mineral was also determined in the volcanic glass. In the study area, vitric tuff was intensely found at the Karabayırlar Hill (MSS I and II) (Figures 1, 2). The lithic tuffs are made up dominantly of volcanic rock fragments (andesite) and partly of quartz and biotite phenocrystals (Figure 5b). Litic tuffs were also observed at the Karabayırlar Hill (MSS I and II) (Figures 1, 2). The crystal tuffs are composed of plagioclase, biotite, hornblende and quartz phenocrystals and volcanic glass. The flow texture observed in crystal tuffs is noticeable with oriented plagioclase microcrystals and biotite and hornblende are partly or completely transformed to opaque minerals (Figure 5c). Like vitric and lithic tuffs, the crystal tuffs are also observed at the Karabayırlar Hill (MSS I and II) (Figures 1, 2).

The basalts in the area have porphyric hypocrystalline texture. In addition, amigdaloidal texture is also observed which is formed by filling of gas vesicles with the secondary quartz crystals. The rock is composed mainly of plagioclase and pyroxene phenocrystals together with microlite and crystallites (Figure 5d). Labrador plagioclase displays alteration signs such as iron oxidation, carbonatization and argillization. Most part of pyroxene phenocrystals were iddingsitized and form glomeroporphyric texture. In the study area, basalts are exposed at the Karabayırlar (MSS I and II), Çakmak and Çakmakçıkan Hills (Figures 1, 2).

Andesites are composed of plagioclase (andesine), hornblende and biotite and show trachytic texture. Plagioclases are subhedral and display a zoning texture (Figure 5e). In addition, hornblende and biotite are transformed to opaque form (Figure 5f). Their typical exposures are found around the Asarkale (Figure 1).

XRD DETERMINATIONS

In order to determine the relation between bedrock and clay minerals which were formed in association with alteration of volcanic and pyro-

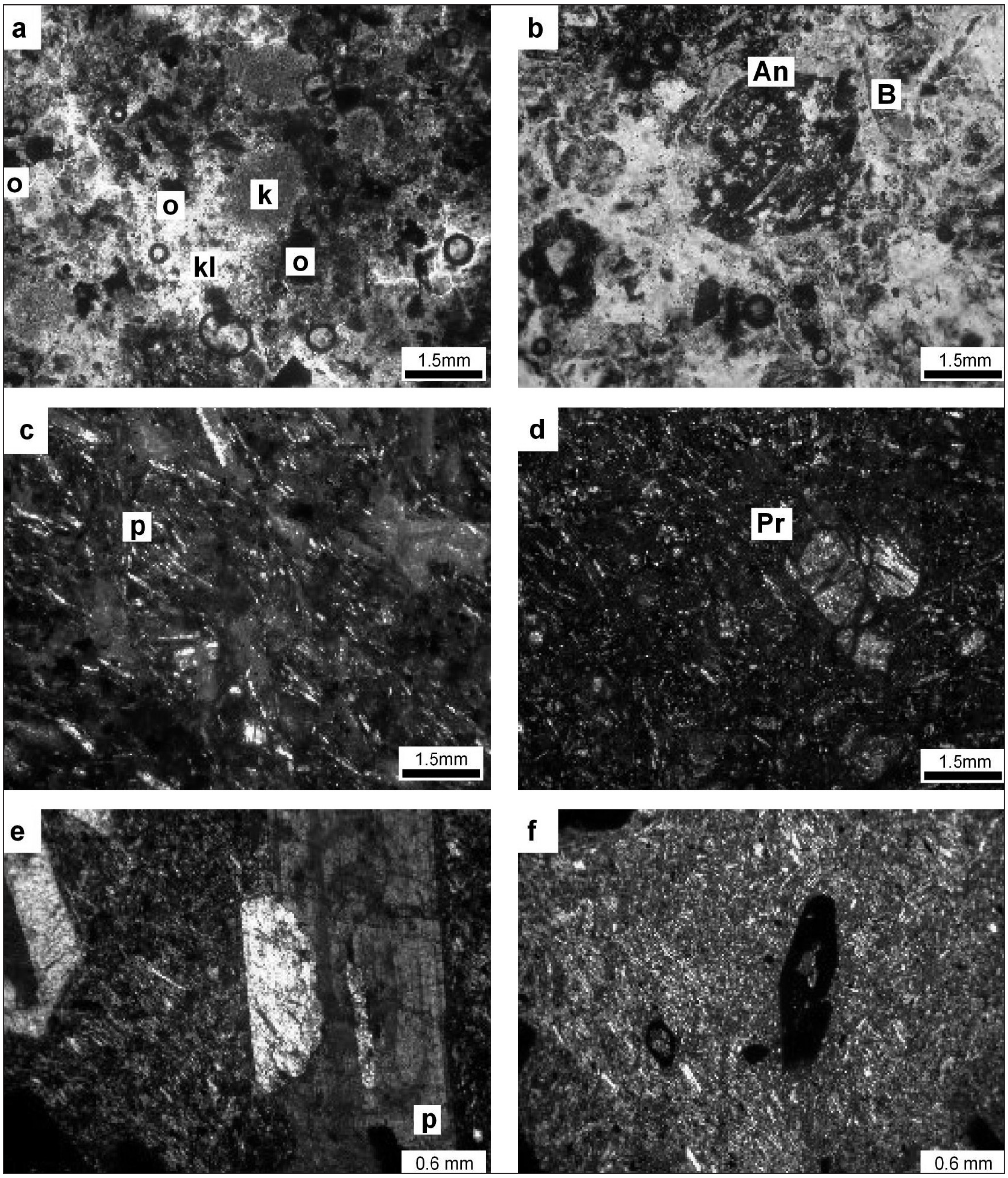

Figure 5- Microscopic views of pyroclastic and volcanic rocks:

a- Argillization (kl) and carbonatization (k) developing in volcanic glass of vitric tuffs and opaque minerals (o) (under crossed polars - sample no. A-4);

b- Andesite (An) rock fragment and biotite (B) phenocrystal in lithic tuffs (under plane polarized light, sample no. A-5);

c- Plagioclase (p) microlites oriented with flow texture observed in crystal tuffs (under crossed polars - sample no. A-9);

d- Pyroxene (pr) phenocrystals and plagioclase microlites which display iddingsitiza tion along edges and fissures of basalts (under crossed polars - sample no. Ç-2);

e- Sub-hedral and zoning plagioclase (p) phenocrystals in andesites (under crossed polars - sample no. A-1);

f- Opaqued hornblende in andesites (under crossed polars - sample no. A-2).

clastic rocks, fresh and altered samples from these units were analyzed with XRD method.

Clay fraction studies of volcanic and pyroclastic units indicate that the dominant clay is smectite although illite exists in a few samples (Figures 2 and 6a). The abundance of smectite varies with the alteration degree of volcanic and pyroclastic rocks. In intensely altered samples, smectite is the main clay mineral (90%-100%) (Figures 2 and 6a). In moderately less altered samples, the abundance of smectite in whole rock analysis was found to be ranging from 40 to 70% (Figures 2 and 6b, c). In whole rock analysis, smectite is accompanied by feldspar, dolomite, calcite, opal-CT, quartz and lesser amount of illite, analcime and gypsum (Figures 2 and 6a, b, c). In the analysis, smectite was described with the maximum peak intensity at 15.12 A° - 15.93 A° which belongs to (001) reflection surface (Figure 6a, b, c). In addition, peaks at 5.06 A°, 4.49 A° and 2.56 A° with higher 2 values were also helpful for identification of smectite. The intensity of (001) peak which is the first basal reflection is narrow and symmetrical thus indicating that smectite is well crystallized. According to 14.76 A° -15.93 A° (001) reflection values, smectite was determined as Ca type smectite (Moore and Reynolds, 1989) (Figures 6a, b, c).

In whole rock analysis, feldspar was identified at 3.18 A° - 3.20 A° and 3.22 A°, dolomite at 2.89 A°, calcite at 3.04 A°, quartz at 3.34 A° and analcime at 3.43 A° peaks (Figures 6a, b, c). In whole rock analysis of samples which were altered at varying degrees and contain smectite as the only clay-size component, there exists a proportional inverse relation between smectite and feldspar (Figure 2). In samples which are particularly intensely altered and enriched in smectite, the feldspar content decreases while in samples which are less altered and represented by low smectite content, the feldspar content shows a proportional increase (Figures 2 and 6a, b, c).

(001) reflection of smectite which was treated with ethylene glycol shifted to 16.67 A° (Figure 7). The reflections at 350 and 550ºC were observed at 9.66 A° and 9.76 A°, respectively.

In X-ray diffractograms of whole rock samples, rising of background by 2θ = 15 A° indicates the presence of volcanic glass of amorphous character (Jones and Segnit, 1971).

DTA DETERMINATIONS

Thermal characteristics (phase transformations) of sample B3A which was determined as smectite by XRD studies were studied with DTA-TG analysis. In DTA analysis of smectite, the first intense endothermic peak was observed around 148.2 °C (Figure 8). The second small endothermic peak at 220 °C is typical for Ca-smectite (Özkan and Erkalfa, 1977). In addition, there are also two endothermic peaks at 653.4 °C and 873.1 °C. Temperatures of these endothermic peaks are suitable for dioctaedric smectites (Paterson and Swaffield, 1987). The first two endothermic peaks reflect the humidity loss and the third one stands for the loss of interlayer water. The weight loss in the first two reactions is 15.3% and 7.6% in the third reaction (Figure 8). The peak observed at 810.1 °C at the DTA curve is related to impurities rather than smectite.

SEM DETERMINATIONS

The clay-dominant (smectitization) samples which were by determined to be variously altered by field observations, microscopic investigations and XRD analyses were also studied with scanning electron microscope (SEM).

In scanning electron microscope studies, it was observed that smectite has a well developed platy structure and morphology of honeycomb texture (Figure 9a). In general, smectite develops in fissures, fractures and dissolution voids of the volcanic glass (Figure 9b). It was noticed that the smectite with honeycomb texture develops on spherical opal-CT (Figure 9c). In some samples, smectite develops on and along the edges of feldspar as well as volcanic glass (Figure 9d).

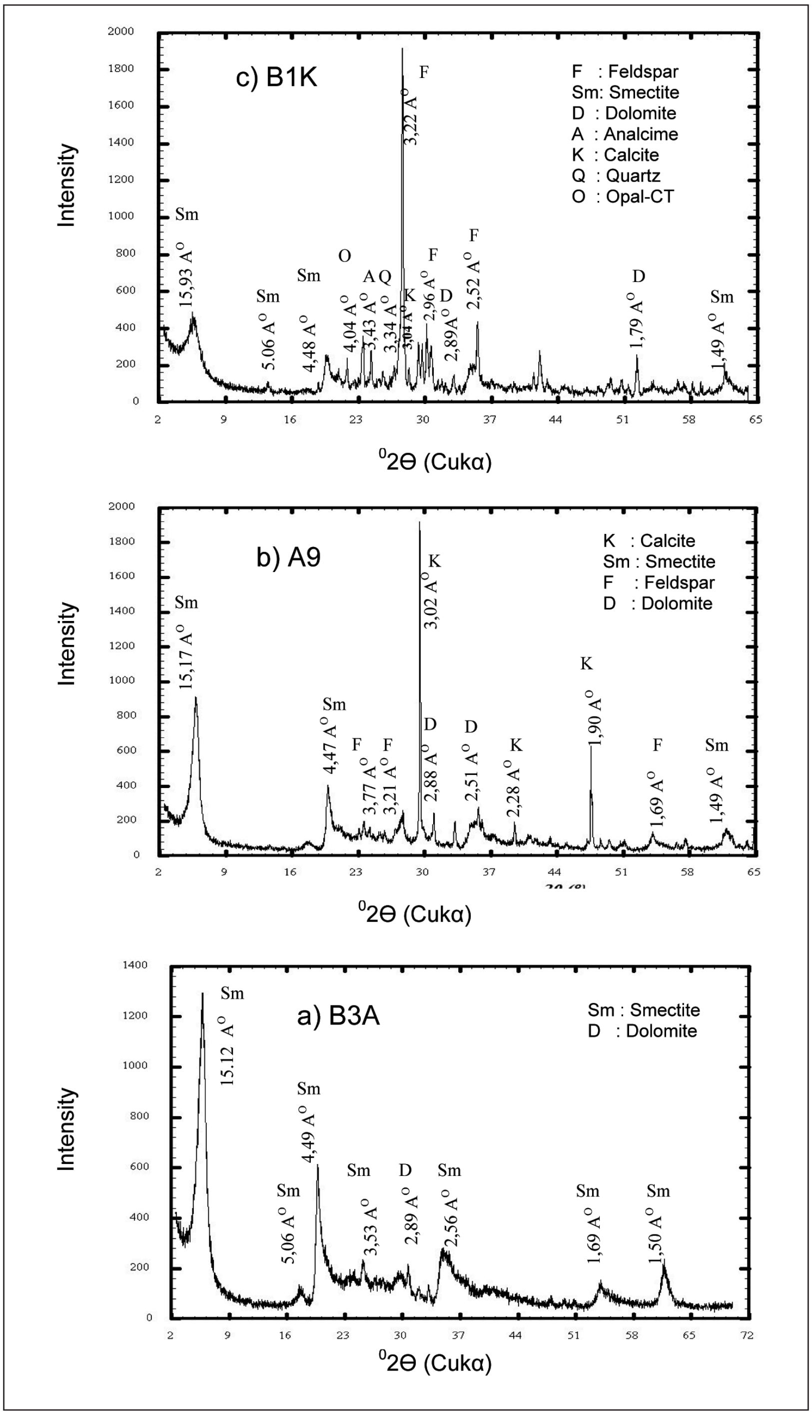

Figure 6- Whole rock XRD diffractogram of tuff samples which were altered in varying degrees (a. altered tuff, sample no. B3A; b. less altered tuff sample no. A9; c. partly altered tuff, sample no. B1k).

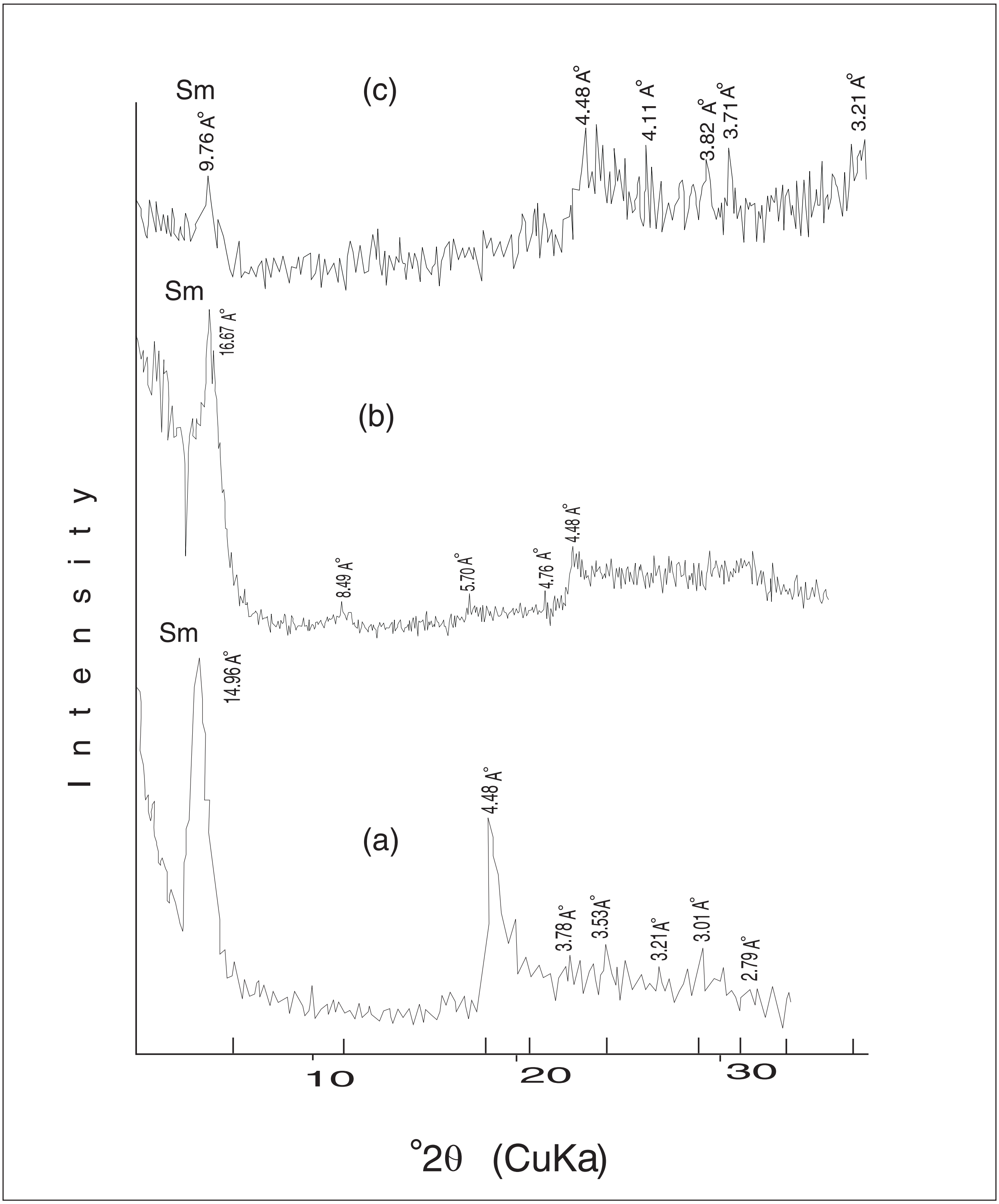

Figure 7- XRD diffractogram of clay fraction of nearly pure smectite (sample no B3A), a: normal; b: glycolated; c: heated (550ºC).

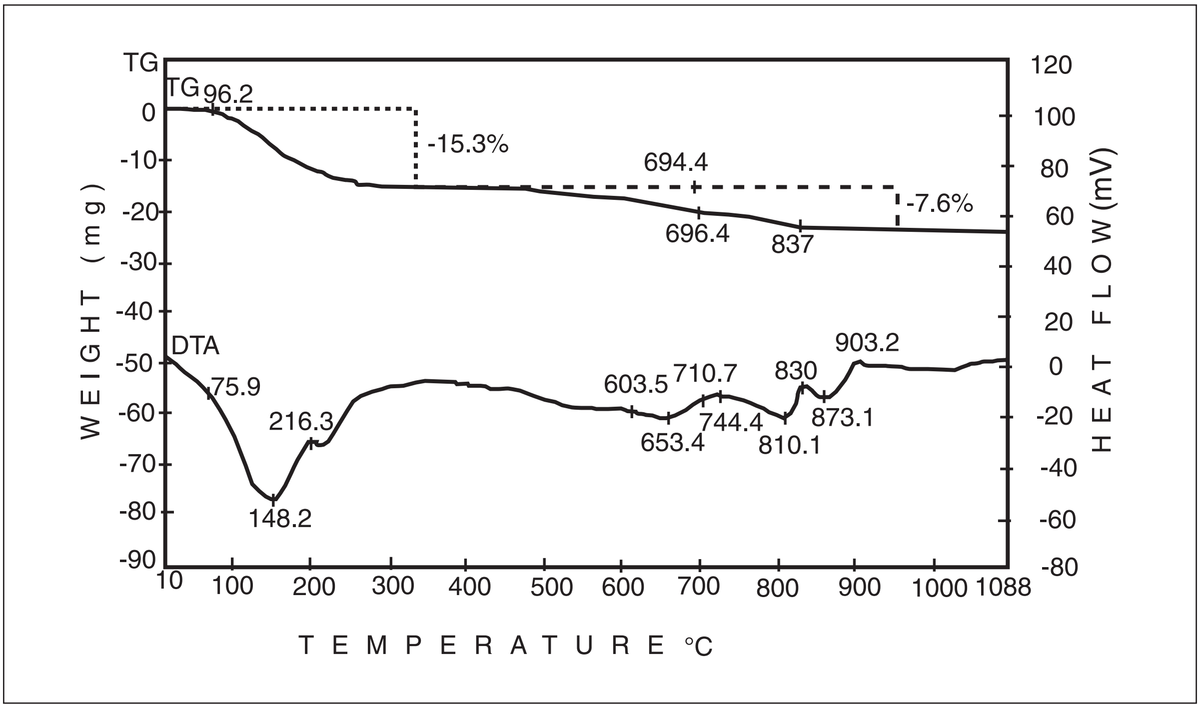

Figure 8- DTA and TG thermogram of nearly pure smectite (sample no B3A).

This may indicate that in addition to volcanic glass and opal-CT, feldspar also had a role in formation of smectite. Christidis et al. (1995), Kadir and Karakaş (2002), Besbelli and Varol (2002) also indicated that smectite was derived from alteration of volcanic glass and feldspar.

DISCUSSION AND RESULTS

The Neogene lacustrine units which are widely exposed in the study area are described as the Miocene Sakarya formation and Pliocene Porsuk formation (Figures 1 and 2). The Sakarya formation starts with pyroclastic-interbedded claystone facies that is characterized by agglomerate, tuff, altered tuff and tuffitic sandstone and continues with a volcanic sequence (Lower-Middle Miocene) represented by lava flows of basalt and trachyandesite composition. These units are overlain with a low-angle angular unconformity by carbonate-claystone and evaporate facies (Upper Miocene) which are composed of claystone, dolomite, limestone, mudstone and gypsum. The Pliocene units which cover this lithologic assemblage with a low-angle angular unconformity are comprised by a repetition of sequence consisting of conglomerate, sandstone, claystone, mudstone, gypsum, gypsiferous mudstone, dolomitic claystone and limestone.

According to field and laboratory observations, Neogene (Miocene and Pliocene) lacustrine units in the region were deposited under varying volcanism, tectonism and paleoclimate conditions. In this respect, there are different facies in the study area showing horizontal and vertical changes. The volcanic activity in the region was started in the Early-Middle Miocene (Temel, 2001; Özen and Sarıfakıoğlu, 2003). In the first period of deposition when the volcanism was active, lacustrine deposits which were originally depleted in evaporitic minerals were changed to dolomite-evaporite facies with increasing evaporation at arid and sub-arid climate conditions.

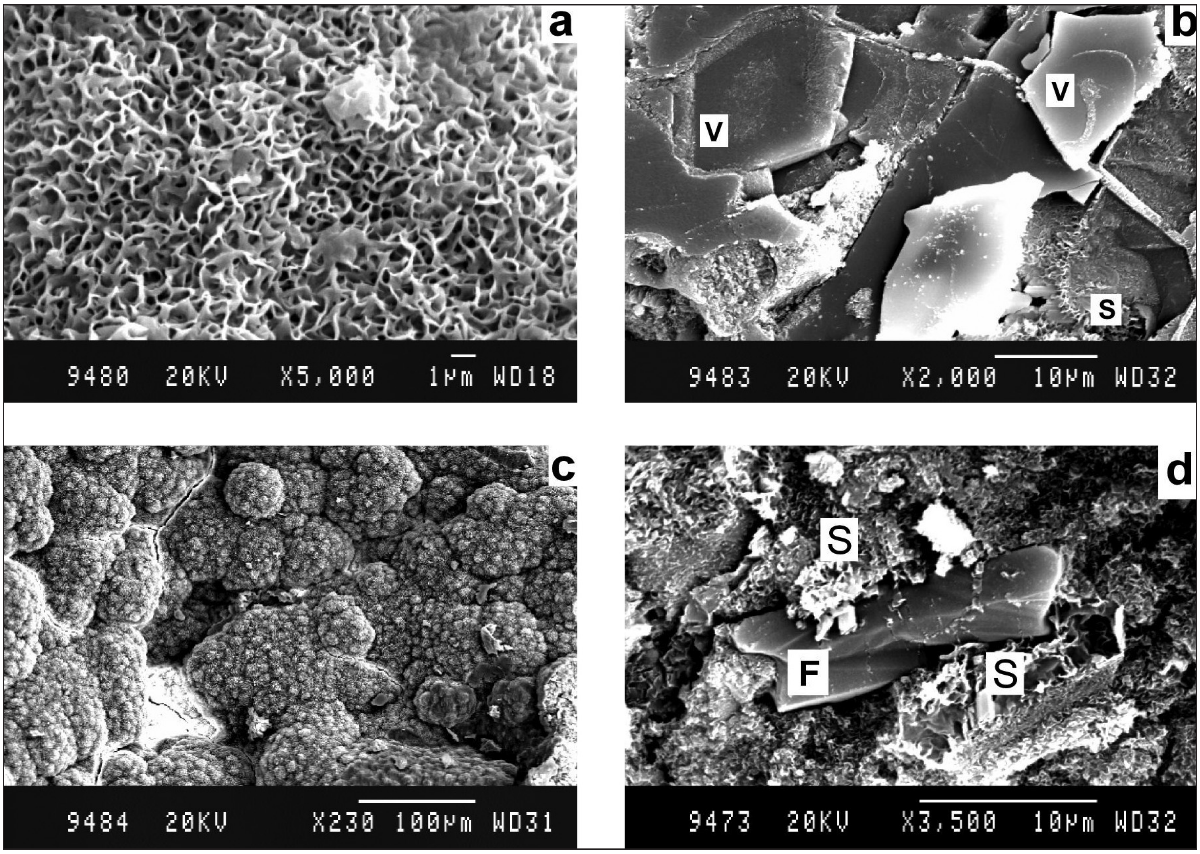

Figure 9- SEM image of smectites.
a: Platy and honeycomb textured smectite (sample no B-1);
b: Smectite mineral developing along edges and surfaces of fractured, platy volcanic glass (V) (sample no KV-8);
c: Smectite forming on spherical opal-CT (sample no KV-8);
d: Smectite (S) developing around and on the feldspar mineral (F) (sample no B-5).

It was determined that smectite is the main clay mineral in alteration zones of tuff and basalts which comprise the volcanic and pyroclastic units of the Sakarya formation. Considering the role of whole rock assemblage and textural characteristics in formation of smectite, alteration of volcanic ash and tuff is the main agent for the deposition of smectite (Grim and Güven, 1978). Particularly, smectite formation was controlled by the presence of volcanic glass and feldspar which are the main components of tuffaceous units. In XRD studies, in addition to the association of volcanic glass, opal-CT, smectite and feldspar which comprise the main components of volcanic units, a proportional inverse relation between smectite and feldspar was also determined which indicates that these minerals are derived from the same source (Figures 2, 6 and 9). Alteration of feldspars results in formation of smectites (Millot, 1970; Furnes, 1975; Chamley, 1989; Tucker, 1992; Kadir and Karakaş, 2002). Alteration of feldspar facilitated great amount of Ca input to the environment which is supported by carbonates occurrences partly shown in fractures. In addition, gypsum was precipitated with the increase of Ca and SO_4 activities due to arid conditions of the environment (Yeniyol, 1987).

Under hot and arid conditions, alteration of volcanic material in the lake water had an important role in formation of smectite. This is in support of the presence of dolomite above the volcanic units which reflects hot and arid climate regime in the area. Smectite is related to hot-arid climate regime and it is known to occur under low rainfall and weak drainage conditions (Chamley, 1989). In addition, it is stated that analcime which is accompanied with smectite in some samples, is occured in saline and alkaline lacustrine environment, indicating that arid and semi-arid climate conditions (Mariner and Surdam, 1970; Gall and Hyde, 1989; Hartley et al., 1991; Renaut, 1993; Türkmenoğlu et al., 1995; Karakaş and Kadir, 2006). While arid conditions were prevailing in the region, units were deposited in association with depth changes in the lake which rapidly deepened due to increasing tectonic activity. The change in lithology in the environment depending on change of depth in the lake area was studied by Hardie et al. (1978). Waters which affected the porous and permeable volcanic units in the study area washed and dissolved the tuffaceous units. Smectite was formed as a result of hydrolysis of volcanic glass and alteration of feldspar. As also evidenced in SEM studies, smectite occurs in weak zones developing in dissolution voids and along the fissure and fractures of volcanic glass indicating that ions freed by water circulation were effective in formation of smectite. Stages which follow the forming of smectite, contributes to rich in Na, Al, and K ions of lacustrine water and increasing pH, resulting in analcime occurrences (Gall and Hyde, 1989; Stamatakis, 1989; Hartley et al., 1991, Karakaş and Kadir, 2006). Illite minerals accompanied with smectite in altered tuff samples were derived from clay-sized biotite (Bayhan and Yalçın, 1990; Gümüşer and Yalçın, 1998). On the contrary, illite minerals in mudstone samples were originated from detritic form and was transported to lacustrine environment.

As a result, formation of smectite was controlled by the presence of volcanic glass and feldspar that comprise the main components of volcanic and pyroclastic rocks. Alteration of volcanic material in the lake water under hot and arid conditions had an important role in the smectite formation. Formation of smectite is closely related to hydrolysis of volcanic glass and alteration of feldspar.

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