MINERALOGICAL AND GENETIC RELATIONSHIPS BETWEEN CARBONATE AND SEPIOLITE-PALYGORSKITE FORMATIONS IN THE NEOGENE LACUSTRINE KONYA BASIN, TURKEY

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ABSTRACT: Neogene (Upper Miocene-Pliocene) lacustrine sediments of northern Konya consist of conglomerate, sandstone, marl, mudstone, claystone, clayey limestone, and limestone. Limestones are white, beige-cream colored, fine-grained and contain remnants of plant roots. Brecciation, calcretion, mud cracks, and dissolution voids are common. Claystones are typical with white and green colors. Green claystone cropping out at the lower part of the sequence is alternated with mudstone and sandstone. White claystone alternating with carbonate units appears at the upper part of the sequence. Carbonate units are found as intercalated layers and lenses of conglomerate, sandstone, and mudstone.

Calcite, dolomite, feldspar, and quartz minerals are dominant in the study area. They are accompanied by sepiolite, palygorskite, smectite, chlorite, and illite minerals hosted by white colored clayey limestone and claystone. In addition, on the green colored claystones, minerals such as chlorite, smectite, and illite are formed.

SEM studies indicate that sepiolite-palygorskite fiber and fiber bundles cover calcite and dolomite. Calcite and dolomite are of hexagonal and rhombic crystal types. These minerals show a meniscus type cement which characterizes a vadose zone.

Paragenesis and textural features of the minerals determined in the study area indicate that precipitation should occur due to climate fluctuations ranging from arid, semi-arid, and wet conditions. Changes in climatic conditions does affect the lacustrine water chemistry and precipitation of carbonate and detrital units. Sepiolite and palygorskite form authigenically as a result of the calcretion of carbonate units in alkaline conditions, high Si and Mg activity, and low Al.

INTRODUCTION AND OBJECTIVES

Sepiolite and palygorskite type clay minerals are commonly found within the sulphates and carbonates of the Neogene lacustrine basins (Millot 1964; Parry and Reeves 1968; Starkey and Blackmon 1979; Galan and Ferrero 1982; Jones and Galans 1988; Suarez et al. 1989; Bellanca et al. 1993; Sanchez and Galan 1995; Galindo et al. 1996), in volcano-sedimentary lacustrine environments (Sheppard and Gude 1968; Starkey and Blackmon 1979, 1984), in upper Cretaceous-Tertiary marine areas (Hathaway and Sacks 1965; Fleischer 1972; Weaver and Beck 1977), in hydrothermally altered basic and ultrabasic rocks (Caillere and Henin 1949; Bonatti and Joensuu 1968), and in pedogenic neoformations of the paleosol (Singer and Norrish 1974; McLean et al. 1972; Watts 1980; Çavuşgil and Kapur 1985; Rodas et al. 1994).

The Neogene lacustrine basin north of Konya is a region where sepiolite and palygorskite are found associated with carbonates, although their distribution varies significantly in both vertical and horizontal directions. Therefore, the purpose of this study is to determine the textural and mineralogical characteristics of these Neogene carbonate and sepiolite-palygorskite units in respect to their genetical relationship.

METHODS

In this study, 95 clay-bearing samples were collected from six sites (Fig. 1). The mineralogical characteristics of the samples were determined through several techniques including XRD (Rigaku-Geigerflex) and SEM-EDX (Topcon Abt-60, Jeol. Jsm 84 A-Edx) methods. For petrographic studies, 50 thin sections were prepared from these samples. The mineralogical composition of the samples was examined with XRD under conditions of CuKα radiation and a scanning speed of 1°2θ/min. The samples were grounded in a porcelain mortar and dispersed in deionized water using a soil mixer. The fractions of the samples were washed three times with deionized water by centrifuging the slurry in order to remove excess cations which cause flocculation. The clay fraction (<2μm) was obtained based on the standard sedimentation method of Tucker (1988), and the excess water was removed by centrifuging. The smear glass samples were prepared from the remaining clay fraction. Air-dried, oriented, ethylene-glycol-solvated, demethyl sulphoxid, and 350°C/2h and 550°C/2h thermally treated samples were used for identification. Semi-quantitative estimates of both clay fraction and rock forming minerals were calculated by an external standard method (Brindley 1980).

On the basis of XRD results obtained from 95 samples, 11 clay dominant samples were selected for SEM-EDX analysis of clay minerals. Hard broken, dried chips with fresh surfaces turned upward were adhered onto the sample holder for SEM and EDX analyses. SEM studies were performed on 350 Å thick Au film-coated sample chips, using a Giko IB.3 ion coater.

In addition, to identify the genetical relationship of sepiolite-palygorskite within carbonate units, 7 representitative samples were obtained from limestone, clayey limestone, claystone, sandstone, mudstone, and green colored claystone. They were analyzed for their major elemental compositions

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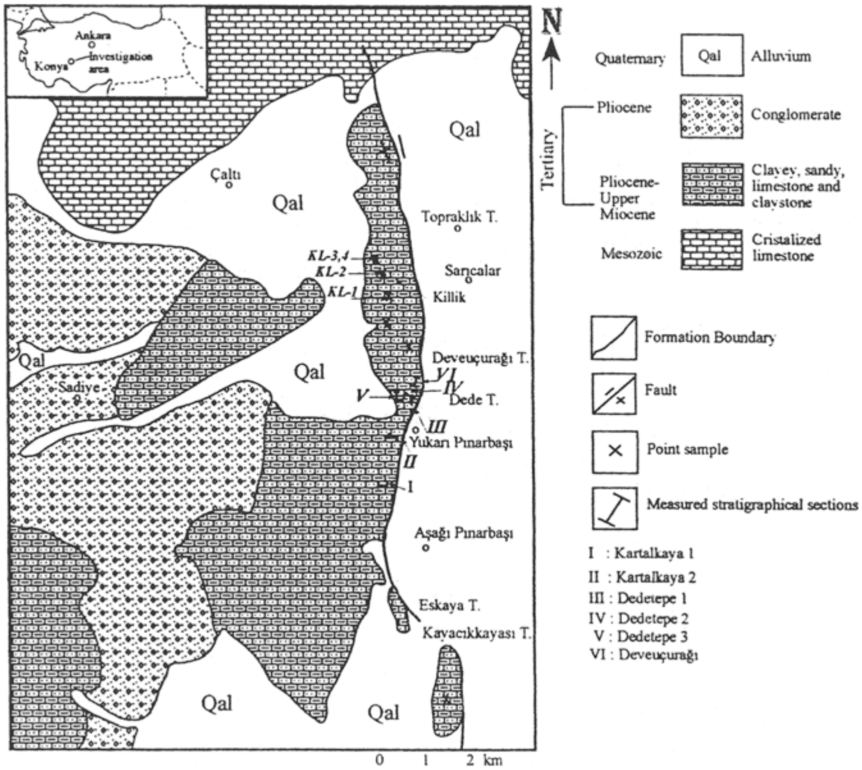

Figure 1. Generalized location and geological map of the northern Konya area (Modified from Özcan et al. 1990).

using atomic absorption, gravimetry, and optical spectrography methods.

RESULTS

Geology

The basement of the study area is represented by serpentinite, schist, and crystallized limestone units of Pre-Neogene age (Fig. 1; Özcan et al. 1990; Hakyemez et al. 1992). This is overlain by the alternation of limestone, sandstone, marl, mudstone, claystone, and conglomerate units (Fig. 2) of the upper Miocene-Pliocene. Here, *Cyprideis* sp., *Cyprideis torosa* (JONES), *Cyprideis seminulum* (REUSS), and *Darwinulla* sp. ostracode fossils of Neogene age are widely distributed in marl units.

The western part of the study area is dominated by an alluvial fan system. The units here are green in color and wedge laterally. The composition of these conglomerates varies from phyllite to micaschist, sericiteschist, and crystallized limestone pebbles of the basement rock. These pebbles are sorted poorly and their size varies between several mm and 6-10 cm. The pebbles are cemented with a fine-middle grained sand material. The sandstone unit is fine-grained and laminated. The laminates are 5-10 mm thick and, occasionally, they are deformated and appear as pocket-type wedging inside the claystones. Synsedimentary deformation structures are common in laminated levels (Fig. 3a). This unit is alternated with mudstone that contains biogenic footprints due to the dissolution voids (Fig. 3b). The upper units are intercalated with green claystones. The lateral distribution of this unit is very narrow, and it wedges from west to east in the area. These units are overlain by white, beige, cream colored limestones and claystones, which are widely found in the area. Limestone and claystone at the hanging wall of the stratigraphical sequence are alternated with each other several times. Brecciation, calcretion, mud cracks and dissolution voids are common (Fig. 3c). This unit contains white claystones. There is a gradational transition between limestone and claystone. Furthermore, limestone-lumps could be seen in the claystones. It is soft and friable outside the carbonate-lump, while inside it is massive. Therefore, there is a gradational alteration from the surface toward the center of the carbonate-lump (Fig. 3d).

Sedimentary Petrography

Generally, the texture of Neogene lacustrine carbonates is micritic. XRD analyses show that most of the carbonate rocks are dolomite (Fig. 2). Ostracods and Gastropods are biogenic grain components of the micritic limestones (Fig. 3e). The rest are ooids and intraclasts (Fig. 3f). The shell of the Ostracods and Gastropods is somewhat protected; however, in some places, it is crystallized or dolomitized and partly broken into pieces. The gaps of protected shells contain different

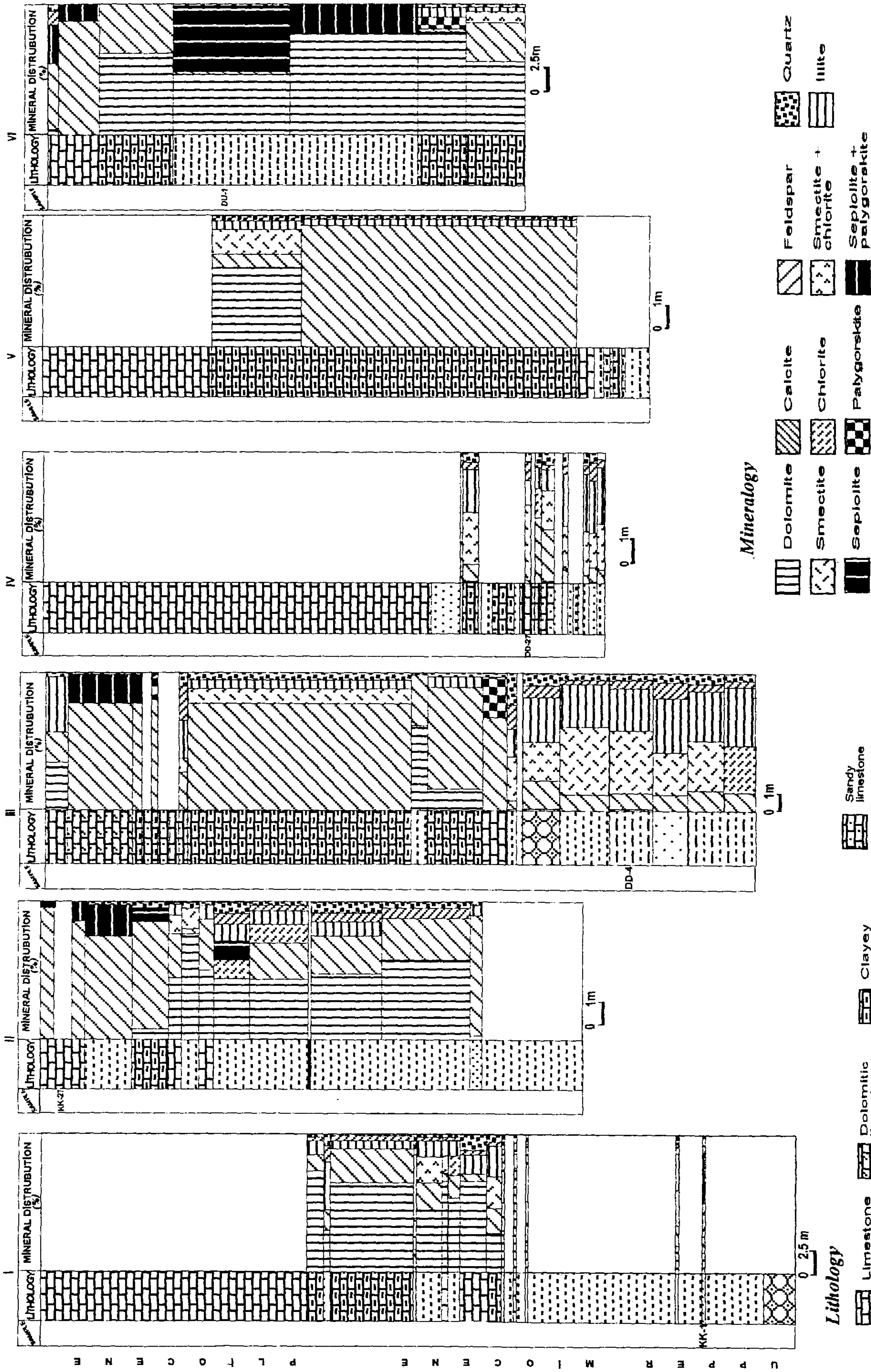

Figure 2. Distribution of the principal lithology and bulk mineralogy in the area of northern Konya.

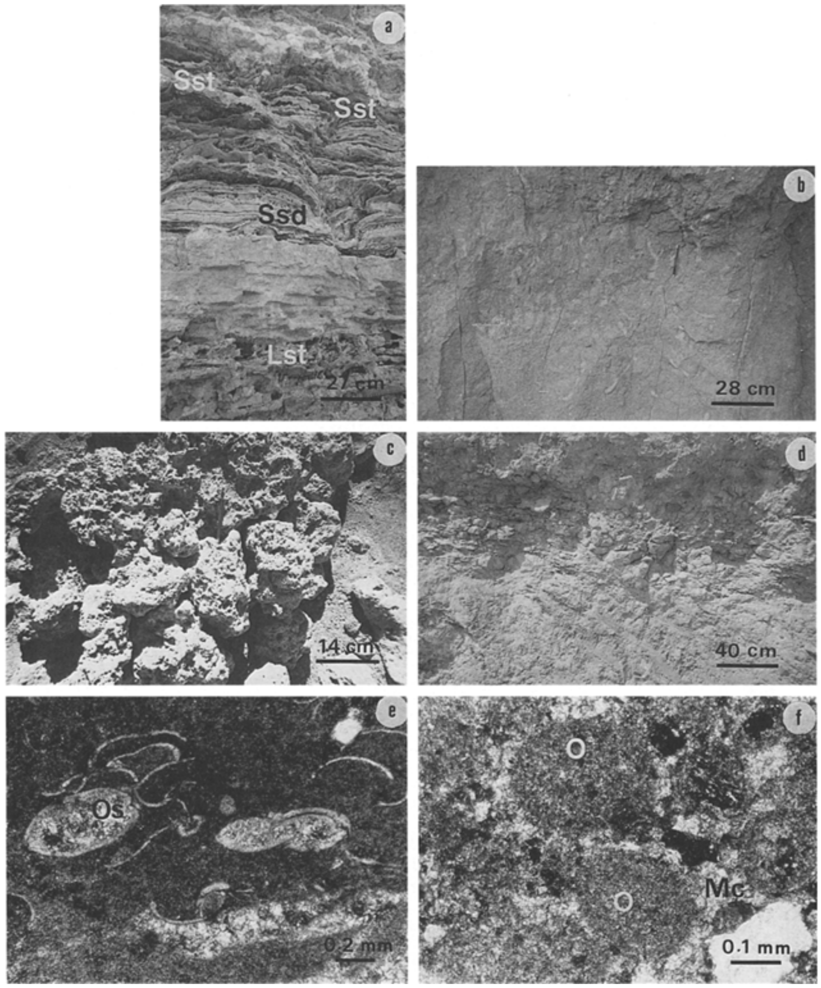

Figure 3. (a) Dedetepe (II) Section: Pocket type sandstone wedged inside the claystone (Sst) and synsedimentary deformation structures (Ssd) are common in dissolution voidy limestones (Lst). (b) Biogenic footprints filling dissolution voids in mudstone unit. (c) The view of sepiolitic material neoformed in the dissolution cracks of completely altered limestone. (d) The view of limestone-lumps in claystone. (e) Shell of the Ostracod (Os) inside the micritic matrix. (f) The Development of sparitic meniscus type cement (Mc) and dissolution voids around ooid (O).

fillings types. These are generally quartz, micrite, sparite, and coarse calcite crystals.

Spherical grains showing similarity to ooids are another grain component determined in the limestones. The size of these grains ranges from 0.25-0.40 mm (Fig. 3f). Generally, the nucleus of the grains are uncertain and semi-opaque, which indicates the presence of organic materials (Williamson and Picard 1974; Friedman et al. 1973). Also, there are some gaps which appeared by washing the ooid nucleus. These voids, from place to place, are filled with quartz, calcite, and sparicalcite. The nucleus of some ooids contain epidote and muscovite type terrigenic materials, which originated from the surrounding metamorphic rocks. The ooid grains are cemented by another sparicalcitic cement. Although there are some voids developed between ooids grains, the presence of voids with the sparicalcitic cement indicates that there are water and air between the ooid grains. Sparicalcite cement forms due to the crystallization of water that circulated between the ooids. This type of cement is called meniscus cement (Strong et al. 1992).

XRD Determinations

All the facies studied including: sepiolite-palygorskite, associated clays, and non-clay minerals were examined by X-ray diffraction.

It has been found that calcite, dolomite, feldspar, and quartz minerals are generally common in the study area (Fig. 2). These minerals are often accompanied by sepiolite, palygorskite, chlorite, smectite, and illite minerals. The mineral paragenesis changes due to the variations in lithologies observed in the area. Also, these mineral paragenesis are not uniform in the vertical and horizontal directions, which are shown in different proportion alternations. Lithologically, sepiolite and palygorskite appear in white claystone and clayey dolomitic limestone. These units also contain chlorite, smectite, illite, feldspar, and quartz minerals. Green claystone, mudstone, and sandstone do not contain sepiolite, palygorskite, and dolomite. Spatial mineral distributions of dolomite, smectite, and chlorite minerals are dominant in Kartalkaya. Towards the north (Dedetepe 1 and 3), dolomite decreases and calcite increases (Fig. 2). Besides the smectite and chlorite, the proportion of sepiolite and palygorskite increases in Dedetepe. In the north (Deveuçuragi and Killik), smectite and chlorite minerals completely disappear and sepiolite-palygorskite and dolomite increase.

The (110) 100 peak of oriented sepiolites moved from 12.166 Å to 12.267 Å after glycolation. The palygorskite mineral changed from 10.517 Å to 10.593 Å (Fig. 4).

SEM Determinations

The carbonate samples containing mostly ooid nucleus and cement were analyzed by SEM after thin section determination.

The nucleus of ooids is mainly spherical shaped and has different diameter micro-lumps (Fig. 5a). The sizes of these micro-lumps is between 2-5 microns. The nucleus of the modern ooids that were reported by Siesser (1973) is similar to what we found. Also, there are tubular and spherical structures in the cement matrix around the ooids. Furthermore, the cement around ooids is composed of coarse grained calcite crystals. The sizes of these calcite crystals are measured to be 10-80 microns. Spherical shaped micro-lumps were identified in these idiomorphic calcite crystals of our samples. These idiomorphic hexagonal calcite crystals surround the ooids of micritic character in the form of cement

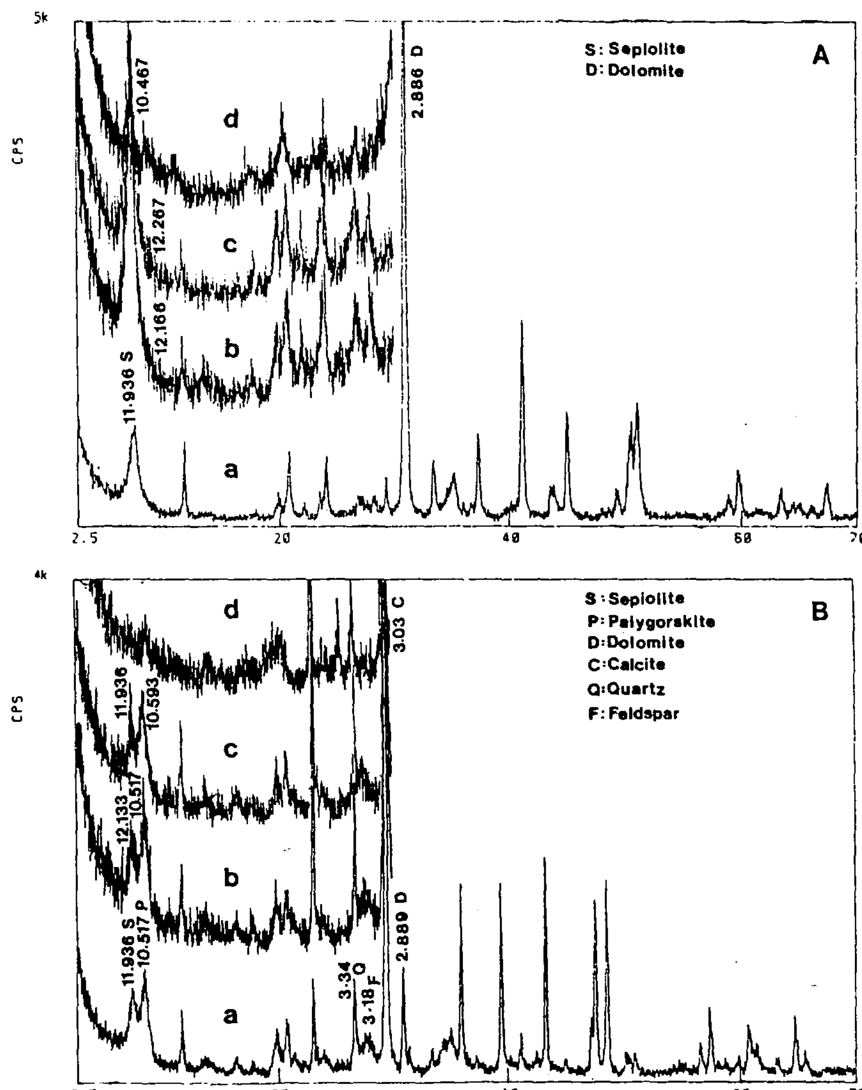

Figure 4. X-ray diffraction of sepiolite (DU-1). (A) and palygorskite (KL-4) (B) minerals. [a. Air dried, b. Oriented, c. Ethylene Glycol, d. Heated].

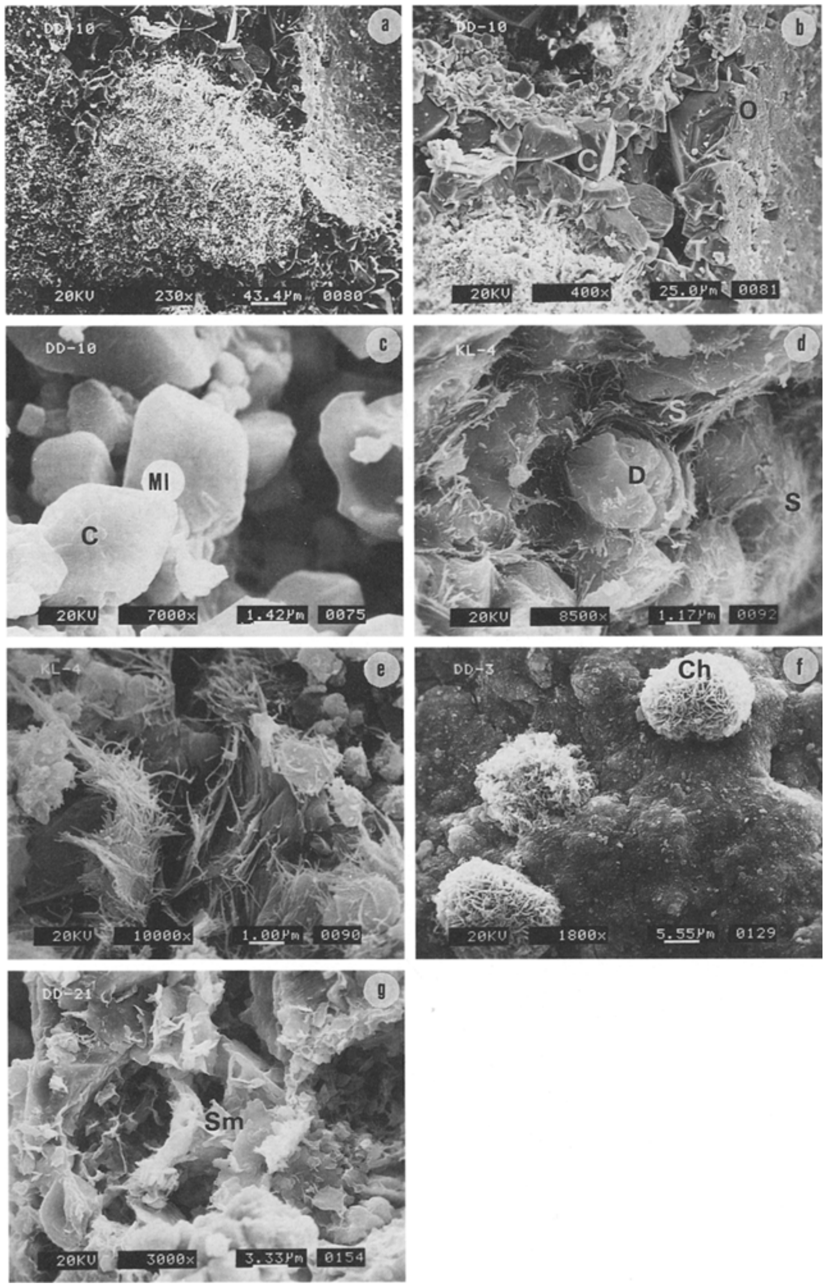

Figure 5. (a) Idiomorphic calcite crystals formed meniscus type cement around ooid grain. (b). A close view of the idiomorphic calcite crystals (C) formed meniscus cement around and between the ooid grains (O). (c) Micro-lumps (Ml) grown on idiomorphic calcite crystals in spari-calcite (C) around the ooids. (d) Concentric sepiolite fibers (S) grown around half authomorph dolomite crystals (D). (e) View of fiber and fiber bundle of sepiolite and palygorskite matrix. (f) Authigenic chlorite (Ch) in green colored sandstone (g) Authigenic smectite (Sm) minerals grown in green colored sandstone.

(Fig. 5b). Idiomorphic calcite crystals are known from the meniscus cement of the vadose zone, as stated by Strong et al. (1992). Beside this, the presence of micro-lumps in the nucleus of ooids and the cement show that the ooid was formed by algae and micro-organism activities (Bathurst 1968; Fig. 5c).

SEM analysis of the sepiolitic dolomite shows that sepiolite fibers generally surround dolomite (Fig. 5d). These fibers are either rare or fiber bundle types formed around dolomite crystals which are idiomorphic rhomboedric crystals. SEM determinations of the KL-4 sample show that the edge of the dolomite crystals is corroded and even semi-rounded. There are voids between these semi-rounded and rounded dolomite crystals. Sepiolite and palygorskite fiber bundles concentrically surround dolomite as a matrix inside the voids (Fig. 5e). In the same sample, the dolomite crystal is corroded and voids are increased. Therefore, the corroded dolomite results in increases of voids which are infilled by sepiolite-palygorskite fibers as a cement matrix.

Smectite and chlorite minerals were identified through SEM-EDX and XRD analyses on green claystone and sandstone units. Chlorites are generally scattered as rounded lumps of chlorite leafs (Fig. 5f). The close view of these rounded chlorite lumps indicates an authigenic origin of honeycomb type. Smectite was identified as an authigenic mineral in the voids of sandstone and claystones (Fig. 5g).

Chemical Analysis

Representative samples of green and white claystone, clayey limestone, limestone, sandstone, and mudstone were chosen for chemical analysis. Generally, the CaO value of the samples is high and Na_2O and K_2O are very low (Table 1). MgO, Al_2O_3, Fe_2O_3, and SiO_2 values are variable. The changes in the oxide value are related to the variations of mineral composition of the different lithologies. Al_2O_3, Fe_2O_3, and SiO_2 values in mudstone, and SiO_2 and CaO values in sandstones are high. These high values were probably originating from detritus materials transported by flowing water draining the western part of the area. The high Al_2O_3 values in these units originated from smectite and chlorite that were determined by the XRD. MgO, SiO_2, and CaO values are high in the Sepiolite dominant DU-1 sample. The high CaO value shows the presence of carbonate minerals as well as sepiolite-palygorskite. This indicates that the formation of sepiolite and palygorskite is in a close relationship with the carbonate units. Furthermore, low values of Na_2O and K_2O support the opinion mentioned above.

CONCLUSION AND DISCUSSION

The Neogene lacustrine sequence of northern Konya is composed mainly of siliciclastic detritus and carbonate units. The lower part of the sequence consists of green clay, mudstone, sandstone, and conglomerate of the detritus units, which is deposited in an alluvial fan system. The hanging wall of the sequence is characterized with limestone-type carbonate units. The carbonate units are generally alternated with clayey limestone and white claystones. In dry periods, following the precipitation of carbonate units under wet conditions, mud cracks and dissolution voids formed. Meteoric water moving downward through these cracks and dissolution voids caused washing and dissolution of the dolomite and limestone units (Siesser 1973; Velde 1985; Semeniuk and Meagher 1981). Siesser (1973) stated that the formation of meniscus cement around ooids and intraclasts of the diagenetic caliche in Southern Africa was related to the dry period which follows the half-dry period of the lacustrine environment. We identified the meniscus cement around the ooids and intraclasts like Siesser (1973). This indicates that, from time to time the climate is rainy and sometime it is dry.

The presence of limestone-lumps in claystones indicates that alteration occurred in wet conditions during dry seasons (Figs. 3c and 3d). The climatic conditions are generally arid, semi-arid, and wet. The annual rainfall is high. During the wet periods, CO_2-rich rainwater easily penetrates into and dissolves the carbonate units. The porous carbonate units ensure that the infiltration rate is high. Relatively high air temperatures and low humidity in summer causes extreme evaporation of surface and near surface waters. In addition, brecciation and alteration of the units show seasonal changes. These micromorphological textures were presented in the caliche environment (Gouide 1972, 1983). Sepiolite and palygorskite are the dominant clay minerals in the caliche

Table 1. Chemical result of the different lithologies in the study area.

Oxides	KK-27 ✶	KL-2 ✶	KL-1 ✶✶	DU-1 ✶✶✶	KK-1 ✧	DD-4 ✧✧	DD-27 ✧✧✧
SiO_2	1.7	0.5	6.97	24.20	38.49	45.57	6.48
MgO	1.1	0.8	7.77	17.25	5.34	2.8	3.13
CaO	53.3	54.22	42.61	21.68	19.45	10.02	2.42
Al_2O_3	0.2	0.1	0.6	1.41	7.25	15.42	15.83
Na_2O	0.1	0.1	0.1	0.1	0.5	0.8	0.31
K_2O	0.1	0.1	0.1	0.3	2.22	3.71	3.02
Fe_2O_3	0.1	0.1	0.3	0.81	2.82	6.31	6.75
MnO	0.1	0.1	0.1	0.1	0.1	0.1	0.1
TiO_2	0.1	0.1	0.1	0.1	0.5	0.7	0.81
P_2O_5	0.1	0.1	0.1	0.1	0.1	0.1	0.2
H_2O^-	0.05	0.1	0.5	1.61	1.26	2.10	2.12
L.I.	43.05	43.57	40.77	32.32	21.96	12.37	4.84
Total	99.90	99.89	100.02	99.98	99.99	100.00	100.01

✶: Limestone ✶✶: Clayey limestone ✶✶✶: Claystone
✧ : Sandstone ✧✧ : Mudstone ✧✧✧ : Green colored claystone

environments (Watts 1980; Hay and Winggins 1980; Hutton and Dixon 1981). The claystone of the area is dominated by sepiolite and palygorskite. Therefore, the neoformation of these minerals is attributed to the calcretion of the carbonate units. SEM analyses show that sepiolite and palygorskite fiber and fiber bundles are grown on and around the dolomite and calcite crystals. Meniscus type cement composed of idiomorphic calcite is grown between and around the ooids. This has a direct relation with the calcretion in the vadose zone (Siesser 1973; Strong et al. 1992).

The limestone in the area must have precipitated during the rainy season, and changes to the arid climate conditions caused the development of cracks. During the wet period, water moved upward inside the vadose zone by capillarity pressure dissolving the carbonate and detritus minerals. An increase in the evaporation caused an increase in the pH, Mg, Ca, Si, and Al of the environment. Sepiolite and palygorskite minerals precipitated because of changes in the physico-chemical condition of the water that circulated within the cracks (Hay and Wiggins 1980; Watts 1980; Hutton and Dixon 1981; Singer and Norrish 1984; Lang et al. 1990). Due to the circulated water activity in the carbonate cracks, brecciation texture is common in sepiolite-palygorskite bearing claystone (Arakel 1979, 1982).

Results of chemical analysis show that CaO values are high in sepiolite-palygorskite-dominant claystone and carbonate units. These values are similar to those from caliche units (Gouide 1972). Our field observation and the results of mineralogical and chemical analyses yield that the Konya sepiolite-palygorskites form authigenically as a result of the calcretion of carbonate units in alkaline conditions, high Si and Mg, and low Al.

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