

Clay minerals of upper Cretaceous (Shiranish Formation) and lower Tertiary (Kolosh Formation) at selected sections from north Iraq

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Introduction

Due to their highly sensitivity to changes in the composition, temperature and pH values of their surroundings, clay minerals are commonly used as indicators of changing environmental conditions [1]. Smectite-rich clays in Cretaceous sediments were formed as residual clays due to erosion of thick pedogenic blankets developed under high temperature and seasonal variations in humidity [2]. In Cenozoic sedimentary successions, an increase in erosion and weathering promotes increase in rock-derived minerals, illite, kaolinite, chlorite, random mixed layers and feldspars, at the expense of pedogenic Al-Fe smectite. This general trend, roughly parallel to the cooling and warming periods indicated by climatic markers such as oxygen isotopes [3].

Clay minerals study of Cretaceous to Tertiary sediments from north Iraq in two sections at Duhok and Shaqlawa areas is conducted (Figure 1). The studied succession is characterized by a thick deposition of clastics and carbonates represented by Kolosh Formation (Late Paleocene-Early Eocene) and Shiranish Formation (Late Campanian-Maastrichtian). The Kolosh Formation comprises shale, sandstone, marl and thin limestone, which represents deposition in marginal marine depositional environment in a narrow rapidly subsiding trough. The Shiranish Formation is composed of thin bedded

Abstract

Clay minerals investigation is conducted using X-ray diffraction and scanning electron microscopy from the upper Cretaceous Shiranish Formation and lower Tertiary Kolosh Formation at two sections in Dohuk and Shaqlawa, northern Iraq. The study revealed the presence of smectite, palygorskite, illite, smectite-chlorite mixed layers, chlorite, and kaolinite. Smectite and palygorskite found to be formed authigenically in the marl of the Shiranish Formation, whereas, other minerals are detrital in origin especially in the Kolosh Formation. The mineral variation reflects the environmental changes from Cretaceous to Tertiary times. Variation in source rocks and change in drainage conditions may lead to increase in the effectiveness of leaching processes and hence transformation of smectite to palygorskite. These changes also effects on the prevalence of inherited or detrital types of clay minerals in the Kolosh sediments.

argillaceous limestone overlain by blue pelagic marl and represents deposition in outer shelf basinal setting [4].

The work aims to elucidate the paleoenvironmental interpretation of the Shiranish and Kolosh formations from north Iraq based on data deduced from clay mineral study.

Regional Geology

Location and tectono-sedimentary setting

North Iraq geologically is formed as a part of the extensive Alpine mountain belt of the Near East, which represented by Taurus-Zagros folding belt developed through collision of the Afro-Arabian and the Eurasian continents[5]. Iraq forms the northeastern part of the Arabian Plate, which lay in the southern hemisphere in high latitudes with dominant clastic sedimentation [6,7]. The Arabian Plate formed part of the long and wide northern passive margin of Gondwana bordering the Paleo-Tethys Ocean [8]. In the north margin of the Arabian Plate, a foreland basin was created in response to loading of the crust by thrust sheets generated because of compression. Basin formation in the region relates to crustal loading by the Zagros-Taurus mountain range. The evolution of the basin fill in a foreland basin system in terms of sedimentary

environment, succession thickness and vertical trends, is strongly dependent on the degree of

compressional tectonic activity [9].

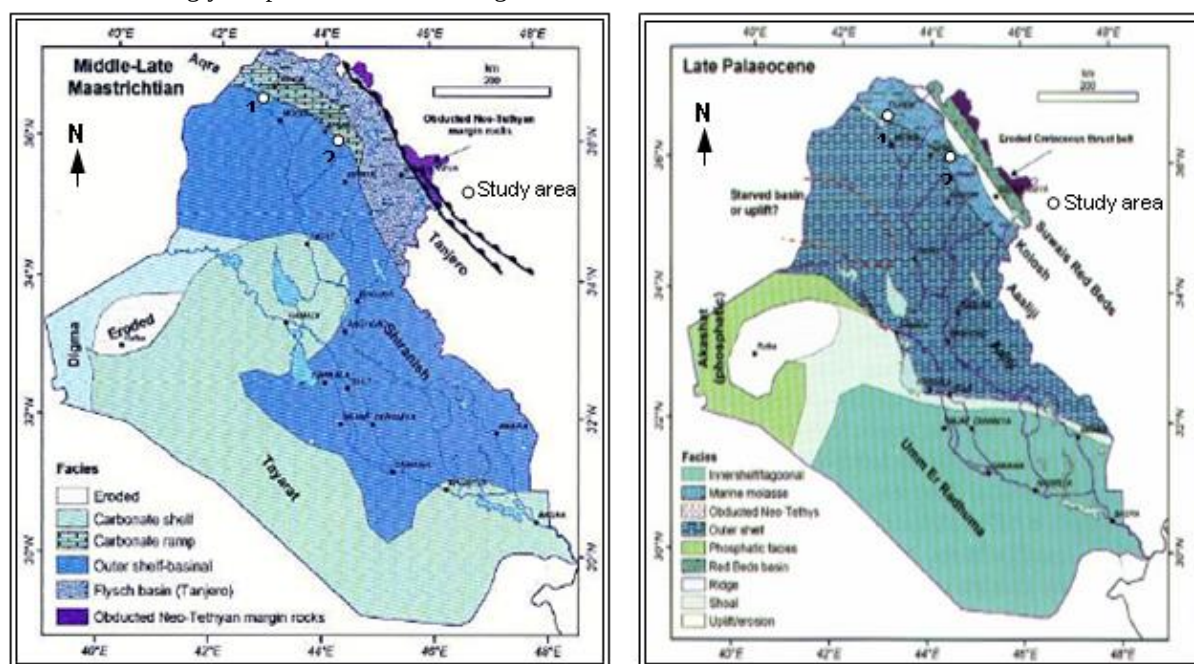


Figure (1): Facies and paleogeographic maps of Middle-Late Maastrichtian (left) and Late Paleocene (right) of Iraq, after (Jassim & Buday, 2006) [10]. The studied sections are : 1, (Dohuk) and 2, (Shaqlawah) of north Iraq

Stratigraphy and paleogeography

The Upper Cretaceous of north Iraq consists of the Shiranish marls and marly limestones, the Tanjero flysch-like clastics (upper Campanian-Maastrichtian) which are replaced laterally by neritic or littoral and reefal limestone, the Bekhme (upper Campanian-lower Maastrichtian) and Aqra (Maastrichtian) Formation and the Hadiena fragmental limestone and marl Formation (upper Campanian-lower Maastrichtian), [11], (Figure 1).

The Tanjero Formation was deposited as a flysch in a rapidly subsiding basin immediately in front of the thrust sheets of the obductional margin in the southern Neo-Tethys. The onset of flysch deposition occurred when these thrust sheets were elevated above sea level and rapidly eroded. Therefore, these sediments have local occurrence in northern Iraq and no continuous section of the formation is exposed at any locality of north Iraq [12 & 5].

The old Upper Cretaceous thrust belt was uplifted and shed clastics to the southwest into a new foreland basin in northeast Iraq and adjacent areas in southeast Turkey and southwest Iran (the Kolosh and Gercus formations of the Paleocene and Eocene age respectively). Sequences of this time span are equated with the Arabian Plate Megasequence AP10 [4].

The Kolosh Formation represents the sediment of the deepest and most mobile sedimentary basin of Paleocene-lower Eocene cycle of Iraq [13]. The Formation occurs in a broad belt, oriented approximately northwest-southeast, somewhat following the Zagros Mountains front (Figure 1). The formation lithologically is composed of black to grey

shale, sandstones and rare conglomerates alternating with thin sandy limestone beds.

The lithology of Shiranish Formation is similar throughout the areas of its exposure, which is composed of blue marl and thin bedded limestone. The deposition occurs in open marine with weak oxidation and alkaline conditions [13].

The studied formations were deposited in area that was tectonically unstable. They have many equivalents towards southern Iraq and neighboring countries.

Materials and Methods

Clay mineral analysis was performed by x-ray diffraction of selected samples from the studied formations at both localities in northern Iraq (Figure 1). Forty-five samples (25 from Kolosh and 20 from Shiranish formations) were collected from the marl and claystone successions of the aforementioned localities. In general, distribution of clay and non-clay minerals in the studied marl and claystone samples from both Shiranish and Kolosh formations are nearly similar. Therefore, representative scans for x-ray diffractograms are included in Figure (2). These selected scans are chosen to represent the clay and non-clay fractions that revealed in the current study. This analysis is conducted at the School of Earth and Environmental Sciences of Wollongong University, Australia using Phillips Spellman DF3 diffractometer, with Cu- α radiation. The bulk compositions were determined based on method of Moore & Reynolds (1989), [14]. The clay fraction was then concentrated before they are analyzed by x-ray diffractometry techniques. Samples were dissociated in water, then

calcium carbonate was removed in 1/5 N hydrochloric acid. The fraction smaller than 2 μ m was decanted according to Stokes law and oriented pastes were made on glass slides [15]. Identification of the clay minerals was carried out using the data given by Carroll (1970), Thorez (1979), Brindly & Brown (1980), [16-18].

SEM analysis was performed using Cam Scan MV 2300 at the School of Material Engineering of Wollongong University, Australia.

Results

Clay mineral investigation of the studied upper Cretaceous-lower Tertiary clastics from both Dohuk and Shaqlawa sections proved the presence of, smectite, palygorskite, kaolinite, illite, smectite-chlorite mixed layers, and chlorite. The first three types, with traces of illite are proportionally higher in the Shiranish Formation, while S-Ch mixed layers

and chlorite, more available in the Kolosh Formation in addition to traces of kaolinite, smectite, palygorskite and illite (Figure 2). The non-clay components are composed quartz, feldspars and traces of calcite and dolomite form the main non-clay constituents of the claystones of the lower Tertiary Kolosh Formation, whereas, calcite forming the main mineral in the marl of the upper Cretaceous Shiranish Formation.

Scanning electron images of selected samples from the studied sections (Figures 3&4) revealed the presence of smectite, as framboidal shapes and palygorskite as long fibers in the form of delicate filamentous outgrowths from smectite. Kaolinite occurs in hexagonal forms with pitted surfaces, while illite found as fine flakes or as a crust and chlorite in small disc-shapes.

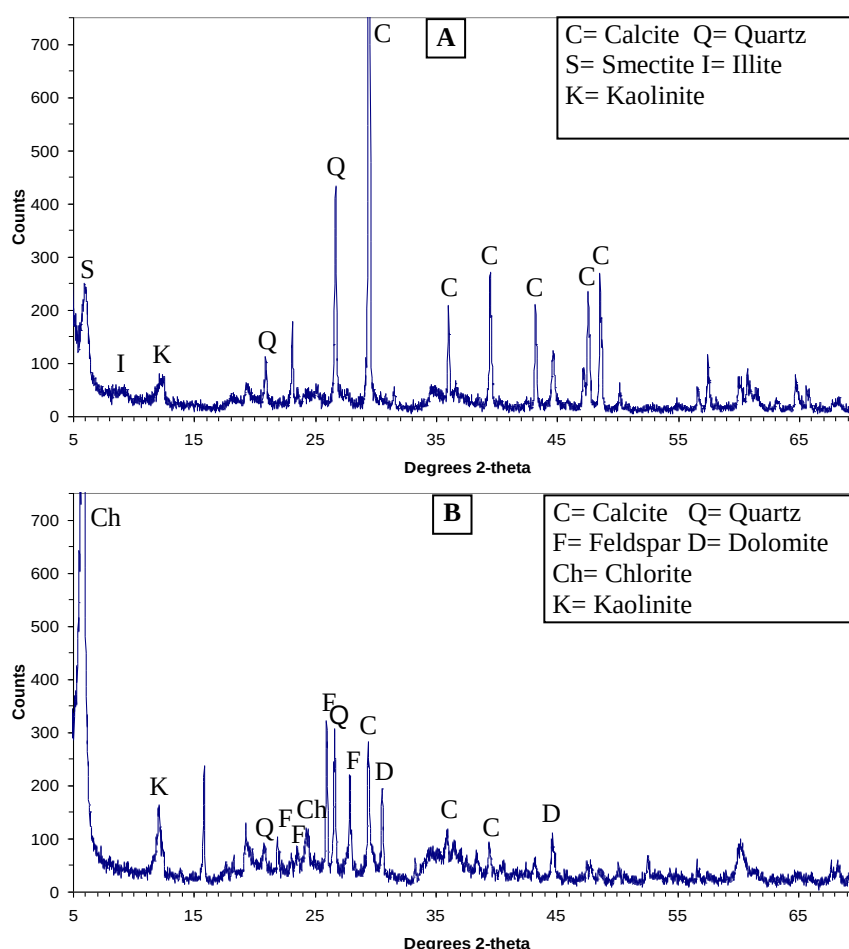


Figure 2: Representative x-ray diffraction scans for the bulk samples from marl of Shiranish Formation (A), and from claystones of Kolosh Formation (B), Shaqlawa section.

Discussion

Clay minerals in the clastic rocks of the Cretaceous-Tertiary succession as represented by Shiranish and Kolosh formations from north Iraq are either of terrigenous (detrital) and/or authigenic and diagenetic in origin. Clay minerals, in general, are chemically unreactive in deep oceans and undergone little change in the zone of weathering [19, 20 and 2]. This

supports the view that most of clay minerals in sediments are of detrital origin. However, clay minerals could also be formed because of diagenesis of sediments. Berner (1971), [21] concluded based on the distribution of clay minerals in oceans that minor proportion of clay minerals may undergo complete structural transformation during diagenesis. He also concluded that kaolinite and smectite could

be formed by crystal growth in the basin of deposition at the expense of muscovite, k-feldspar and plagioclase. Kaolinite, however, is more likely to be inherited from kaolinitic source since kaolinite is very unlikely to form in seawater [22].

The presence of various clay minerals in the Cretaceous-Tertiary succession may reflect the change in the source rocks and change in drainage conditions (in the zone of weathering) which may lead to increase in the effectiveness of leaching processes and hence transformation of specific minerals to others (e.g. smectite to palygorskite in the present study), [2].

Smectite, the abundant clay mineral in the upper Cretaceous Shiranish Formation in both sections and is derived from many types of rocks, these include; acidic, and basic igneous, metamorphic and sedimentary rocks [22]. He also suggested that smectite can be neoformed (as authigenic mineral) in alkaline environment in which dissolved silica, alumina, and magnesium are abundantly available. These conditions were reported as prevailing conditions during deposition of the Shiranish Formation in northern Iraq [23 and 12]. Scanning electron images show smectite as framboidal and platy forms, which may reflect its authigenic formation of this mineral (Figures 3A and 4A).

The marine transgressive depositional nature of Shiranish Formation is resulted from the early Maastrichtian transgression over the foreland basins on the passive margins of the Arabian Plate forming what is called "Piggy-back basins" [24]. These basins were fragmented into shallow sub-basins due to listric faults [25], in these fragmented basins. Marine conditions of deposition of the Shiranish Formation may enhance the authigenic deposition of various clay minerals like smectite and palygorskite.

palygorskite may be formed or transformed from the precursor smectites (Figures 3A and 4B-C) These delicate filamentous outgrowths from a platy nucleolus are similar to those portrayed by Chamley (1989), [2] for the transformation of precursor clays. This mineral also formed authigenically as long flexuous fibers in Shiranish marl (Figure 4A) or detrital in origin as short and broken flakes in the claystones of Kolosh Formation (Figure 3D-E).

Palygorskite formed as authigenic mineral in lagoons and evaporitic basins by chemical sedimentation [22 and 26] or by transformation from precursor clays during early diagenesis by direct crystallization in calcareous soils, or as results of hydrothermal alteration of basaltic glass in the open oceans in association with fore-arc basins [27].

Illite is formed diagenetically or in weathering zones due to alteration of muscovite, biotite, and k-feldspar [28]. Transported smectite also changes to illite as it enters seawater where the chemical environment is completely different from that of weathering and transporting environments [21]. In the present study, illite occurs as flakes or as crusts (Figures 3 C&F; 4

B&E) which may indicate the altered form of illite from older feldspars or other silicate minerals.

Kaolinite commonly are of detrital origin derived mainly from igneous rocks rich in potsh feldspars or from the reworking of older sedimentary rocks [26]. Presence of detrital kaolinite also is an indication of relatively little leaching effect and chemical weathering in the source area [2]. The presence of hexagonal plates with pitted surfaces (Figure 4F), eroded plates (Figures 3E & 4D), and of plates which are relatively oriented face-to-face (Figure 3B) could be interpreted as retransported and redeposited (detrital) kaolinite [29 and 30].

Chlorite is derived from the weathering of rocks rich in ferromagnesian minerals that contains high Mg, Fe, and Ca and that is excellent in the basic igneous and metamorphic rocks [26]. These rocks are common in the ophiolitic complexes of Iraq and Turkey, which may form the source of the sediments in the Kolosh Formation [13 and 31]. Scanning electron micrographs show small disc-shape chlorite (Figure 3C-D) reflecting inheriting nature from older chlorite- rich or derivation as detrital clays from older igneous rocks.

In the claystones of the Kolosh Formation, the smectite and palygorskite mostly are of detrital origin. The subsiding shallow marine nature of deposition with common turbidity currents does not allow the chemical precipitation of these clay minerals and mostly favor the detrital nature of clay minerals. Smectite and palygorskite mostly were derived from older silicate minerals such as pyroxene, amphibole, and serpentine associated with basic rocks [32 and 33].

The overall variation reflects differences in behavior as a function of grain size, cation exchange capacity and thus differential flocculation. The interaction of these parameters is inferred to be responsible for distribution of smectite and kaolinite and invariably related to depth and distance of the ultimate site of deposition.

shallow conditions is reflected by the presence of larger size and less ionic absorption in kaolinite while deposition in more deeper conditions is commonly indicated by presence of smectite and mixed layers which is of small size fraction and higher absorption of ions [34].

During period of increased rate of terrigenous sedimentation during uplift/or subsidence or transgression, the calcareous content of the sediments was progressively reduced by dilution of terrigenous detritus and this culminated towards the contact between the Cretaceous to Tertiary. The succeeding, characteristically coarse-grained conglomerate is likely to reflect a higher episodic uplift and the consequent deposition of these sediments by higher turbidity currents, probably as part of submarine fans. Later on, the basin was relatively subdued and shallower during the Tertiary accompanying the deposition of the Kolosh Formation.

This configuration, more or less, coincides with the event of Laramide Orogeny [13], which starts at the end of Mesozoic (upper Cretaceous) and the beginning of Cenozoic (lower Tertiary), and affected particularly the northern part of Iraq.

Furthermore, the deposition of the upper part of the Shiranish Formation marks a sudden deepening in water because of increasing of subsidence during which thick sequence of argillaceous pelagic facies is deposited [35]. This phase of transgression, which is developed during middle-upper Maastrichtian, seems to be pursued under less stable tectonic conditions [36 and 37]. The abundance of fine terrigenous materials in the upper part of the formation is appearing to be associated with the water depth increase. This increase may relate to the culmination of upper Cretaceous tectonic unrest, which leads to the increase of clastic influx into the basin, especially in northeast Iraq (Tanjero Formation). The transgression continued until the end of Maastrichtian time when a regional regression took place. The area was subjected to other transgression whereby the lower Tertiary (Paleocene) Kolosh flysch sediments was brought into the area.

Conclusions

Clay mineral distribution in the Cretaceous-Tertiary succession from Dohuk and Shaqlawa sections from

north Iraq revealed the dominance of authigenic smectite and palygorskite in the marine upper Cretaceous Shiranish Formation with lesser amounts of illite. The authigenic minerals were formed and preserved in alkaline environment in which dissolved silica, alumina, and magnesium are abundantly available. These conditions were prevailed during deposition of the Shiranish Formation. Sometimes, post-depositional diagenetic reactions affect in the transformation of palygorskite from precursors smectite. In the lower Tertiary Kolosh claystones, the dominance of chlorite, mixed-layers of smectite-chlorite and illite at the expense of smectite and kaolinite may refer to dominance of warm conditions that serve the preservation of these minerals in addition to influx of terrigenous materials eroded from older Cretaceous successions in the area. The distribution of clay minerals in the studied succession coincides with the environmental change from Cretaceous to Tertiary and reflects the change in the source rocks and change in drainage conditions which may lead to increase in the effectiveness of leaching processes and hence transformation of smectite to palygorskite and in the prevalence of inherited or detrital types of clay minerals in the Tertiary sediments.

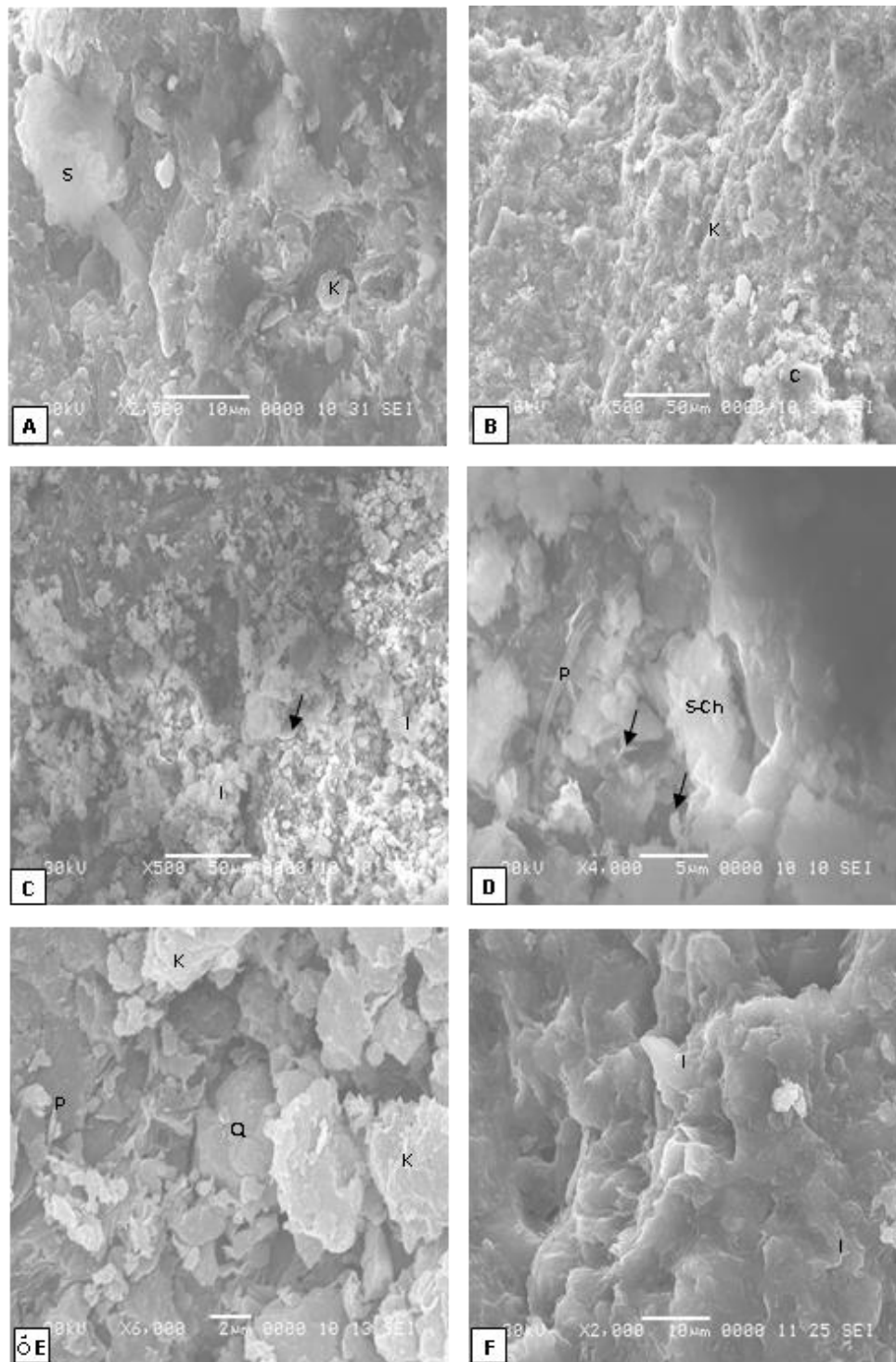


Figure 3: Scanning electron microphotographs (SEM) showing various morphologies of the clay minerals in the studies succession: A- framboidal smectite (S) and kaolinite plates (K). B- common kaolinite hexagonal plates (K) embedded in carbonates (C), both photos from the Cretaceous Shiranish marls. C- illite white flakes (I) and small disc-shape chlorites (arrow). D- broken fiber of palygorsite (P) and chlorite (arrows) and smectite-chlorite mixed layer (S-Ch). E- detrital quartz (Q), eroded kaolinite plates (K), and short palygorskite fibers. F- common illite as flakes and white crusts (I). photos C-F from the claystones of Tertiary Kolosh Formation, Shaqlawa section.

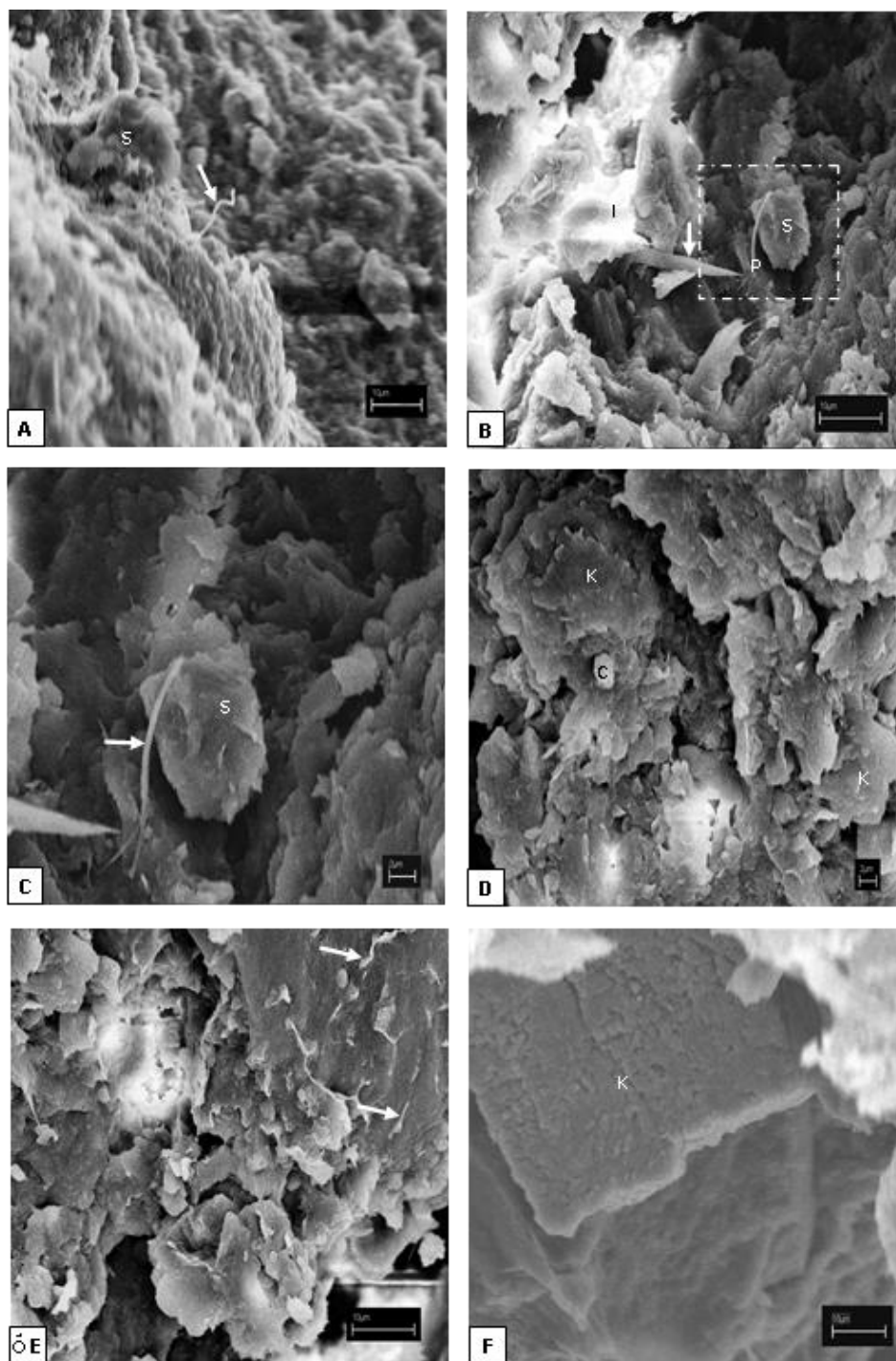


Figure 4: SEM photomicrographs illustrating A- authigenic palygorskite fibers (arrow) and smectite (S) in framboidal shape. B- palygorskite outgrowth (P) from platy smectite (S), illite flakes (I) and laths of mica (arrow). C- enlarged view of B, showing the outgrowing of palygorskite (arrow) reflecting its diagenetic origin from precursor smectite (S), Cretaceous Shiranish Formation. D- Kaolinite (K) in eroded or erose plates and fine calcite (C) crystals. E- illite flakes (arrows) or crusts. F- pitted-surface hexagonal plates of kaolinite (K) reflecting its detrital origin, Kolosh Formation, Shaqlawa section

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المعادن الطينية في تكاوين الشرائش (الطباشيري الأعلى) وكولوش (الترشري المبكر) في مقاطع مختارة من شمال العراق

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الملخص

تمت دراسة المعادن الطينية باستخدام حيود الاشعة السينية والمجهر الماسح الالكتروني لنتابعات تكويني الشرائش (الطباشيري الأعلى) والكولوش (الترشري المبكر) في مقطعي دهوك وشقلاوة شمالي العراق. أظهرت الدراسة تواجد معادن السمكتايت، الباليغورسكايت، الالاييت، الصفائح المزدوجة من سمكتايت-كلووايت، الكلورايت، والكاؤولينايت. السمكتايت والباليغورسكايت قد تكونت موضعيا في صخور المارل لتكوين الشرائش بينما تبين الأصل الفتاتي لبقية المعادن الطينية في تكوين الكولوش. تغيرات المعادن الطينية تعكس التغيرات البيئي من الكريتاسي-الترشري. التغيرات في صخور المصدر وتغيرات ظروف التصريف قد تؤدي الى زيادة قابلية عملية الخلب بما يعزز من تحول السمكتايت الى الباليغورسكايت وتكونهم موضعيا. هذه التغيرات تؤثر أيضا على شيوع المعادن الموروثة فتاتي الأصل في ترسبات الترشري.