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EFFECT OF PERMO-CARBONIFEROUS CLIMATE ON ILLITE-SMECTITE, HAUSHI GROUP, SULTANATE OF OMAN

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Abstract—The Late Westphalian to Artinskian Haushi Group in the Sultanate of Oman consists of the glaciogenic Al Khlata Formation and the Gharif Formation which contains marginal marine, coastal plain, and fluvial sediments. The sequence was deposited during a global-warming event following the Permo-Carboniferous glaciation of Gondwana. Because of a varied subsidence history, these sediments range from the surface in the SE to almost 5000 m in the NW of the basin.

Mixed-layer illite-smectite (I-S) is an important constituent of the <2 μm size fraction of sandstone and shale samples in both formations at all depths. Different starting compositions lead to three distinct trends of illite layers in I-S versus temperature for different sedimentary environments and paleoclimatic conditions. The starting compositions of I-S at the surface range from an ordered I-S in the Al Khlata Formation to smectite-rich in the Upper+Middle Gharif members.

Physical, chemical and environmental factors were investigated as causes for the different starting compositions of I-S. Both formations share an identical burial history, paragenesis, thermal evolution, and source of detrital material. They differ only in environmental conditions during sedimentation. Thus, the variation in starting composition of I-S appears to be best explained by distinct weathering conditions during sedimentation of the three units. In particular, the expected low intensity of chemical weathering during glaciogenic conditions is marked by the presence of higher amounts of unstable volcanic and sedimentary rock fragments in the Al Khlata Formation.

Key Words—Haushi Group, Illite, I-S, Paleoclimate, Sultanate of Oman, Upper Paleozoic.

INTRODUCTION

The transformation of smectite into illite in interstratified I-S minerals with depth is a commonly observed phenomenon in siliciclastic sedimentary basins worldwide. The main factors controlling this transformation are potassium availability within the sediments and temperature (Pollastro, 1990; Pollastro, 1993). Other potentially important factors are time involved in the reaction (Pytte and Reynolds, 1989), pressure (Colton-Bradley, 1987), rock/water ratio of the sediments (Freed and Peacor, 1989), fluid and rock composition (Robertson and Lahann, 1981), the presence of organic acid anions (Small, 1994), and the starting composition of the I-S (Rettke, 1981; Pollastro, 1990; Vasseur and Velde, 1993; Wei *et al.*, 1996).

Starting with the groundbreaking works of Burst (1959, 1969), Powers (1959, 1967), Perry and Hower (1970), Hower *et al.* (1976) and others in the Gulf Coast area, many studies in sedimentary-basin modeling used various kinetic approaches of the smectite-to-illite transformation to calibrate the thermal history of a basin (Hillier *et al.*, 1995; Essene and Peacor, 1995; Elliott and Matisoff, 1996). The following criteria are critical when choosing a specific model: (1) single or multistage (Dutta, 1986; Velde and Vasseur, 1992), (2) first or higher order kinetics (Dutta, 1986; Pytte and Reynolds, 1989; Huang *et al.*, 1993), (3) the presence of dissolved species such as K^+ (Pytte and Reynolds, 1989; Huang *et al.*, 1993), or (4) whether it uses a fixed or a range of activation energies (Wei *et al.*, 1996). However, to apply the smectite-to-illite reaction as a calibration tool, the influence of the initial composition of I-S must also be taken into account.

The purpose of this study is to illustrate the influence of paleoclimate, specifically different weathering conditions, in generating different starting compositions of I-S mixed-layer clay minerals of continental sediments and to show how these primary differences affect the rate of later transformation of I-S minerals during burial.

GEOLOGIC SETTING

Interior Oman Sedimentary Basin

The Sultanate of Oman occupies the southeastern corner of the Arabian Peninsula. The Interior Oman Sedimentary Basin (IOSB) is a low-altitude desert area bounded by the Huqf-Haushi axis, a NNE- to SSW-running anticlinorium in the east and by the NE-SW oriented Jabal Qara arch in the south. In the north, the basin is bordered by the Oman Mountains. To the west, the basin grades into the Rub al Khali Basin of Saudi Arabia (Figure 1). Unlike the intensely studied geology of the Oman Mountains, relatively little is published on the geology of the interior (Tschopp, 1967; Al–Marjeby and Nash, 1986; Hughes Clarke, 1988; Levell *et al.*, 1988; Visser, 1991; Loosveld *et al.*, 1996; Millson *et al.*, 1996).

The sedimentary sequence within the IOSB reflects the basin's complex tectonic history which evolved in

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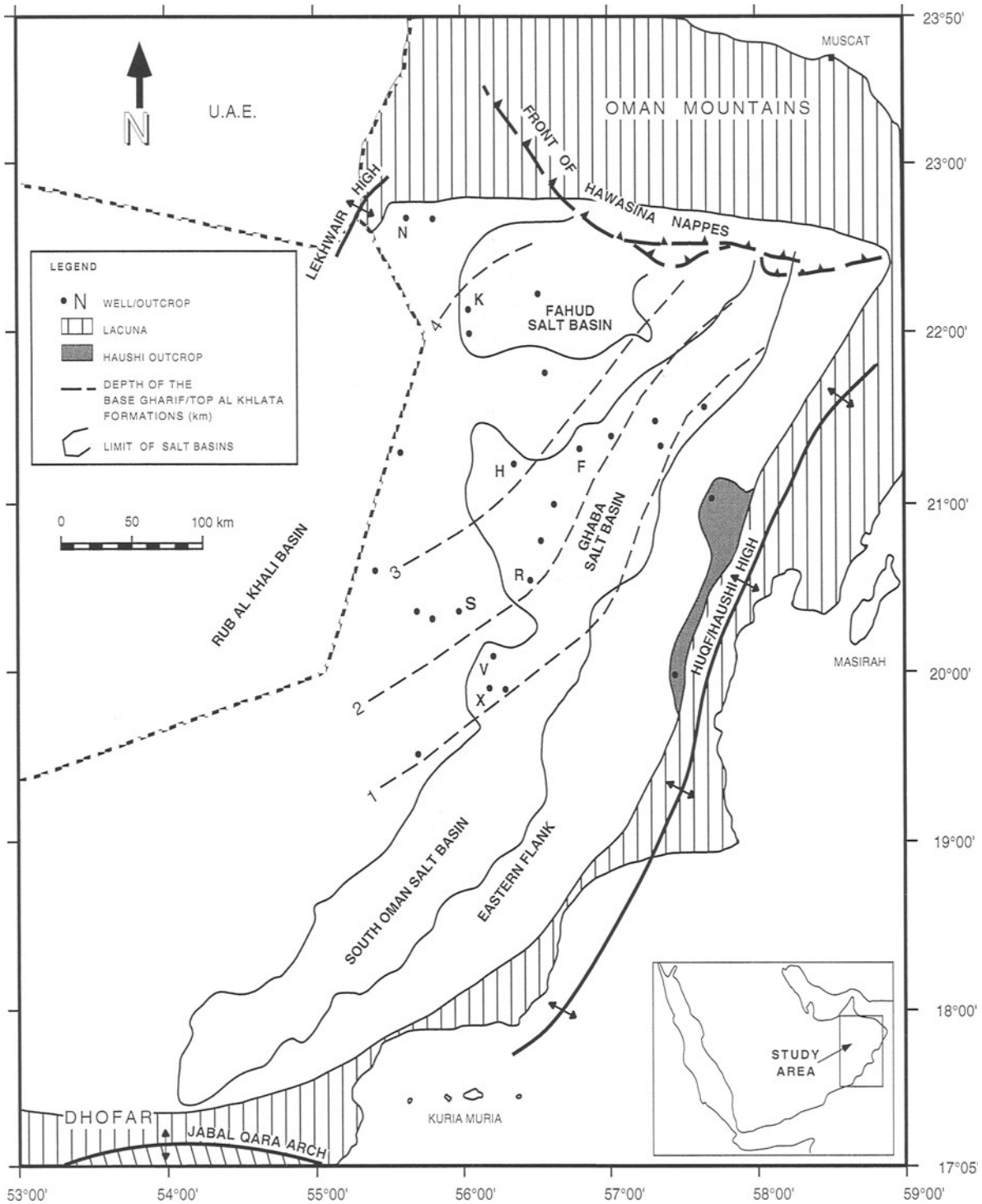


Figure 1. Location of studied wells and outcrops in the Interior Oman Sedimentary Basin. Dashed lines give the present day depth (in km) of the base of the Gharif/top of the Al Khlata Formation.

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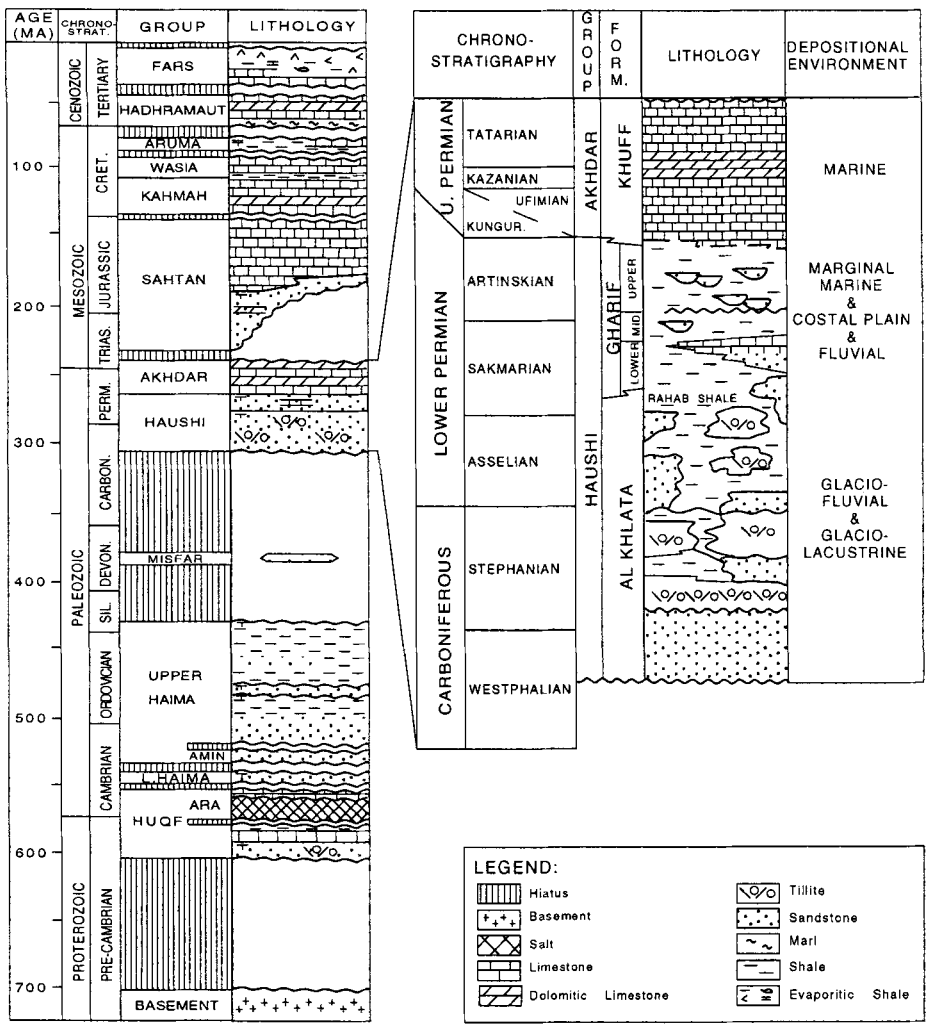


Figure 2. Generalized stratigraphy of the Interior Oman Sedimentary Basin modified after Loosveld *et al.* (1996).

six stages (Loosveld *et al.,* 1996). Stage I represents the Pan-African accretion of the Precambrian basement. Stages II and III, Early Proterozoic to Ordovician, cover two rifting cycles, followed by epeirogenesis and uplift from Late Silurian to Mid-Carboniferous with the general absence of sedimentation. The Late Carboniferous to Mid-Cretaceous stage IV is marked by the breakup of Gondwana and the development of the passive margin of the Arabian Plate. Stage V reflects the evolution of a foreland basin following the obduction of the Oman Mountains in the northern part of the basin during Late Cretaceous. The Tertiary stage VI is controlled by rifting in the Gulf of Aden in the south and the completion of the mountain-building process in the north, leading to relatively quiet conditions in the basin. From its deposition at the beginning of stage III onwards, episodic subsurface flow and dissolution of the Ara Salt modified the internal configuration of the IOSB (Loosveld *et al.,* 1996).

Haushi Group sediments

At the base of the Haushi Group, the most important hiatus (Figure 2) is located within the sedimentary sequence, reflecting the final phase of the Pan-African tectonic events, which affected Oman during most of the Upper Silurian to Lower Devonian (Al-Marjeby and Nash, 1986). Middle Devonian to Lower Carboniferous strata are also absent from the basin, possibly due to Hercynian uplift and erosion (Hughes Clarke, 1988; Husseini, 1992).

The predominantly clastic Haushi Group is divided into two units: the lower Al Khlata Formation and the upper Gharif Formation (Figure 2). The conglomerates, pebbly sandstones, sandstones, siltstones, shales, and diamictites of the Al Khlata comprise a great variety of lithofacies with extreme lateral and vertical variability due to its glaciogenic origin. This variability arises from the irregular substratum of the Upper

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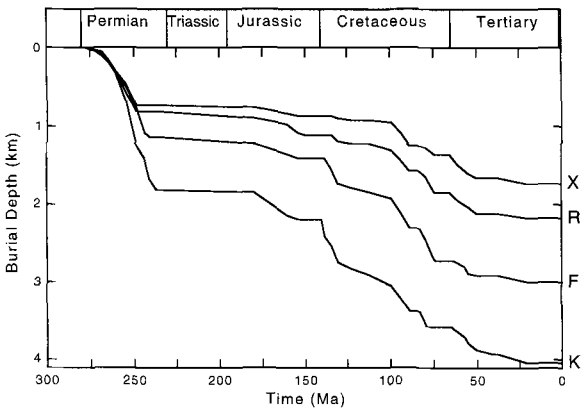


Figure 3. Burial history for the base of the Gharif/top of the Al Khlata Formation in wells X, R, F and K (for locations see Figure 1).

Haima Group, which was caused by salt tectonics of the Cambrian Ara salt (Braakman *et al.,* 1982; De la Grandville, 1982; Levell *et al.,* 1988). Only the uppermost unit of the Al Khlata Formation, the lacustrine Rahab Shale, provides a basin-wide correlatable horizon (Hughes Clarke, 1988). The Al Khlata is characterized by a variable sediment thickness ranging from 0 to >1000 m. These sediments have been dated as late Westphalian to early Sakmarian (Besems and Schuurman, 1987) and are mainly of continental origin.

The Gharif Formation, subdivided into a Lower, Middle and Upper member (Focke and van Popta, 1989; Mercadier and Livera, 1993), has been dated by palynomorphs to be of late Sakmarian to Artinskian age (Besems and Schuurman, 1987). Except along the northern and southeastern rim of the IOSB, the Gharif Formation is characterized by a sediment thickness varying from 200 to 350 m. The Lower Gharif consists of a transgressive coastal plain siliciclastic sequence which is replaced in the salt basins by a marine siliciclastic-bioclastic-oolitic limestone. The Middle Gharif contains a fining-upward sequence of cross-bedded sandstones interbedded with red-brown mottled siltstones and mudstones with strong calcrete development and root traces, followed by a basin-wide mudstone with less calcrete development and poorly preserved depositional textures. The Upper Gharif is composed of a multistory, coarse-grained sand sheet, grading into a fine-grained, fining-upward sand body embedded in red-bed paleosols towards the north and the Rub Al Khali Basin.

Following deposition of the siliciclastic Haushi Group, a widespread transgression across the northeastern margins of the Arabian Plate, caused by climatic warming, led to the deposition of the predominantly carbonate Akhdar Group (Kashfi, 1992; Alsharhan, 1993).

Paleoenvironmental reconstructions show that the Al Khlata has sediments of glacio-lacustrine, glacio-fluvial, and locally glacio-marine origin. The sediments were deposited during several advances and retreats of ice sheets during Late Paleozoic Gondwana glaciation. The Rahab Shale, a basin-wide lacustrine horizon, marks the onset of general warming. The shallow marine, coastal plain, and fluvial plain sediments of the overlying Gharif are the result of glacio-eustatic sea level changes superimposed on a general eustatic sea level rise during Gondwana deglaciation (Guit *et al.,* 1994). Appearance of red-bed diagenesis, typical of semiarid to arid conditions (Walker, 1976), within the Middle Gharif, is another clear indication for a general warming during Permian times in Oman.

Burial and thermal history

Decompacted burial curves for four wells, based on stratigraphic data from Petroleum Development Oman L.L.C. (PDO) for the base of the Gharif Formation, are shown in Figure 3. The PetroMod software package was used for decompaction of the sedimentary sequence. This dynamic, deterministic forward two-dimensional simulator uses the finite element method (IES GmbH, 1994). In general, the subsidence history can be divided into four time intervals, two of them characterized by high sedimentation rates and the other two by non-deposition.

After deposition, the area was characterized by a high sedimentation rate, which was more pronounced along the Oman-Saudi Arabian border than along the SE coast of Oman. Sedimentation ceased during Middle Triassic and a 60 Ma period of relative stability with little or no sedimentation followed. A new phase of sedimentation started at the end of Lower Jurassic time (~180 Ma). Again, more sediment was deposited along the Oman-Saudi Arabian border than towards the SE. This second phase of sedimentation continued until the Eocene, except in the SE region where deposition ended in the Upper Cretaceous. The variation in the present day burial depth of the Haushi Group sediments, which show a generally deepening trend from the outcrop area in the Huqf region towards the NW (Figures 1 and 3), is the result of the regional variation in sedimentation rate and subsidence during the two phases of sedimentation.

Corrected present day borehole temperatures are available for many wells in the study area. In cases where no corrected temperatures exist, nearby wells were used to translate present-day burial depth into temperature values. Since no major uplift and erosion occurred in the northern part of the IOSB after deposition of the Haushi sediments (Figure 3), present temperatures are close to maximum burial temperatures. These findings are consistent with homogenization temperatures of late cements from different wells

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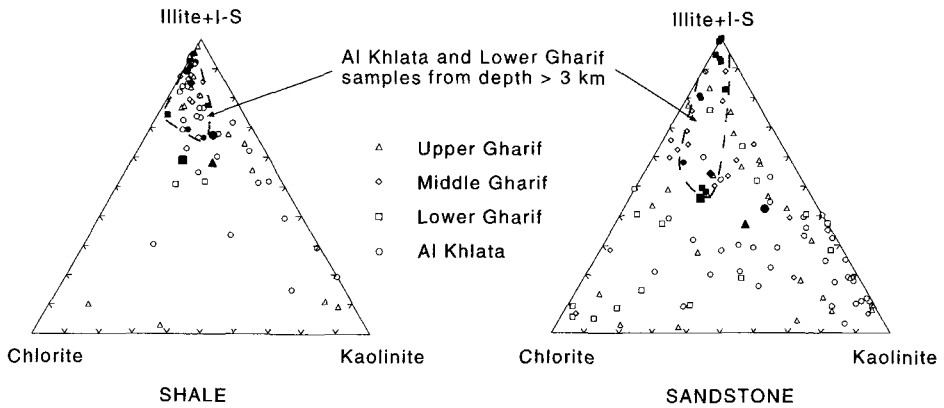


Figure 4. Clay mineral compositions of sandstones and shales from four different units of the Haushi Group. Filled symbols represent mean values.

which show values only up to 15°C higher than present day temperatures (Hartmann, 1996).

MATERIALS AND METHODS

Sampled materials

Material from 96 sandstone and 50 shale core samples of the Haushi Group were provided by PDO from 23 exploration wells situated in the northern part of the IOSB (Figure 1). Additionally, 15 sandstone and 14 shale samples were collected from shallow cores and from two locations in the outcrop area of the Haushi Group (Figure 1; Platel *et al.,* 1994). The sample depths for Haushi Group core material range from 14 to 4013 m.

Methods

For petrographic analyses, sandstone samples were impregnated with a high-temperature, blue-dyed epoxy resin. The polished thin sections were then stained for carbonate identification following the method of Dickson (1966). All thin sections were point-counted (400 points) using an optical microscope.

X-ray diffraction (XRD) determined the K-feldspar content of bulk-rock samples. Powdered samples were mixed with LiF as an internal standard at a weight-ratio of 10 : 1. Randomly-oriented samples were measured on a Philips PW-1710 diffractometer using CuKα-radiation. K-feldspar concentration was determined by comparing the ratio of the 27.5 °2θ (K-feldspar) and 38.7 °2θ (LiF) peaks in the sample with those of standards. The standard deviation was calculated from repeated (20) measurements of a standard mixture in which the amount of K-feldspar is <5%.

The analyses of clay minerals were performed on the <2 μm fraction. To avoid mixing of detrital clay minerals with clay minerals from shale rock fragments (to 2 vol%), or illitic material from altered feldspar grains, the sandstones were not powdered but only gently disaggregated into individual detrital grains. The crushed rock was dispersed by addition of sodium-metaphosphate in distilled water and separated in Atterberg settling tubes. The <2 μm fraction was then concentrated with a centrifuge and the slurry was sedimented on glass slides. Samples were run both air dried and after ethylene glycol saturation on a computer controlled Philips PW-1710 diffractometer in step scan mode using Ni-filtered CuKα-radiation, 0.05 °2θ/step and 3 s counting time per step. After data acquisition between 2–35 °2θ, the three regions, 2–11 °2θ, 14–20 °2θ and 23–26 °2θ were numerically decomposed into one to six symmetric Gaussian or Lorentzian peaks with help of "DECOMPXR", a computer program distributed by Lanson and Velde (1992). Percent illite in I-S mixed-layer clay minerals was determined after Moore and Reynolds (1989) from the difference in °2θ of the numerically decomposed I-S (001/002) and I-S (002/003) peak positions. Quantitative analysis of different clay minerals was performed with the method described by Moore and Reynolds (1989) using mineral intensity factors and peak intensities of the basal reflections from kaolinite (002), chlorite (004), illite (002), and I-S (002/003).

RESULTS

The clay mineral compositions of the Al Khlata and Gharif samples are shown in Figure 4. Generally, sandstones show a wider variety of clay mineral compositions than shales and the mean clay-mineral composition values for each unit are similar, as shown by the filled symbols in Figure 4. Compared to sandstones, the mean clay-mineral compositions in shales show a higher relative abundance of illite and I-S mixed-layer clay minerals. For both lithologies of the Al Khlata and Lower Gharif samples, a depth-related trend is also observable. Samples from >3 km are characterized by a higher abundance of illite and I-S mixed-layer clay minerals. This phenomenon is due to illitization (*e.g.,* kaolinite illitization), which was pos-

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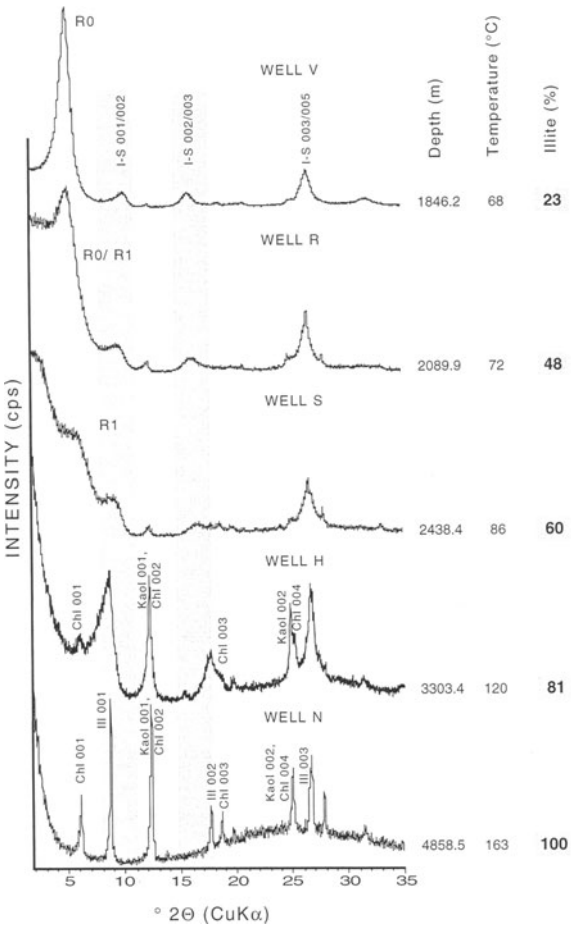


Figure 5. XRD traces of the <2 μm fraction after ethylene glycol saturation. The increase in illite layers in I-S mixed-layer clay minerals during burial is documented by the shift of the I-S (001/002) and I-S (002/003) peaks in the gray shaded areas.

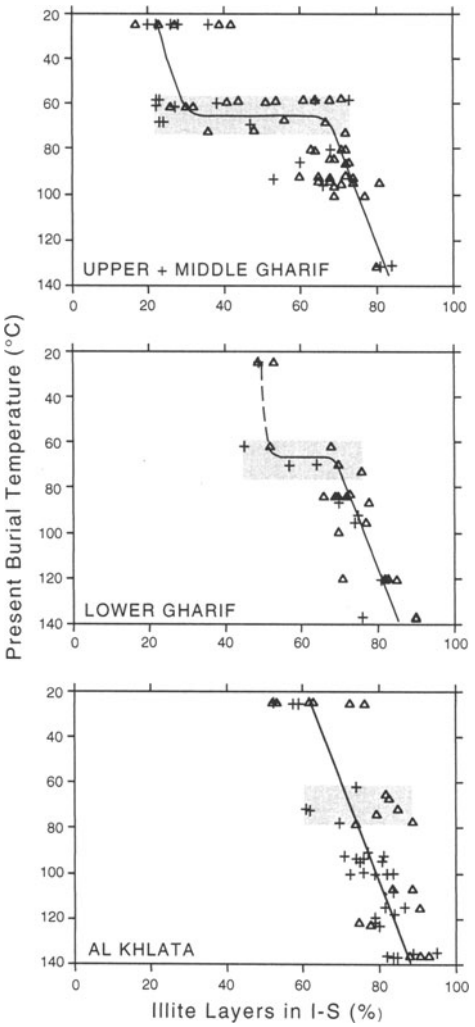


Figure 6. Composition-temperature relations for sandstones (triangles) and shales (crosses) for the Al Khlata, Lower Gharif and Upper+Middle Gharif units. Note that the starting composition and the depth/temperature trend of illite layers in the I-S mixed-layer clay minerals depends on the stratigraphic unit. The gray area highlights the isothermal reaction zone.

itively identified only in samples from the depth >3 km (Hartmann *et al.*, 1999).

Figure 5 shows XRD traces typical of the <2 μm fraction of Haushi Group sediments. The increase in illite layers in I-S is documented by the shift of both I-S peaks in the range of 8–11 °2θ and 15–17.7 °2θ. Separating the samples into the three stratigraphic units, Al Khlata, Lower Gharif, and Upper+Middle Gharif (Figure 6), reveals that the starting composition of the I-S contains a higher percentage of illite layers for Al Khlata samples compared to samples from the Upper+Middle Gharif, with the Lower Gharif I-S composition between. In addition, the rate of increase of percent illite in I-S with depth is clearly different for the three units. In the case of the Upper+Middle Gharif unit (Figure 6), a considerable degree of heterogeneity in the lowest temperature range exists, probably reflecting a range of initial compositions of detrital I-S during and after deposition of the sediments (Pollastro, 1990). A drastic increase of percent illite in I-S occurs in the temperature interval between 60–80°C. The observed trend can be interpreted as an S-shaped composition-temperature relation with a starting composition of about 20% illite in the I-S minerals. Such a transformation trend is typical for Tertiary Gulf Coast sandstones (Boles and Franks, 1979; Bruce, 1984), but was also recognized in older sediments, such as the Mississippian in the Anadarko Basin, Oklahoma (Weaver and Beck, 1971). The composition-temperature trend for the Lower Gharif can be interpreted similar to the Upper+Middle Gharif, with the exception of a starting composition of ~50% illite in the I-S mineral. In contrast, the Al Khlata unit

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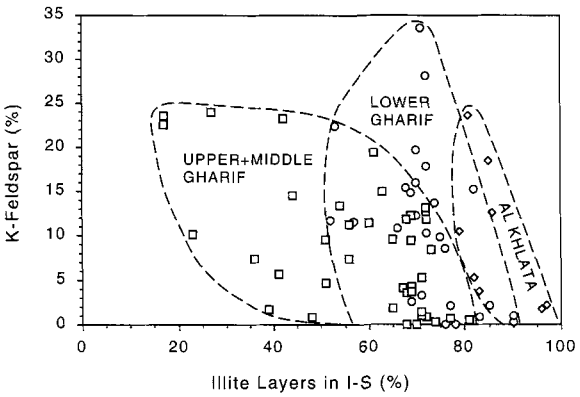


Figure 7. K-Feldspar content versus illite layers in I-S clay minerals of sandstones from the Al Khlata, Lower Gharif and Upper+Middle Gharif units. Note that the majority of sandstones contains K-feldspar even in samples where the illitization of I-S mixed-layer clay mineral has taken place.

exhibits the smallest increase of percent illite in I-S with increasing temperature, shown by the linear correlation with a starting composition of ~60% illite in I-S. A comparable trend was described by Gharabi and Velde (1995) for the Paleozoic Illinois Basin, where the authors explain the high illite content in I-S minerals at the surface by a significant basin inversion, with erosion of overlying sediments exposing previously deeper buried samples. Both the burial history of the study area (Figure 3), and the close vertical spacing between the three units with no erosional boundaries, excludes basin inversion as the cause for the higher percent illite in I-S in the Al Khlata and the Lower Gharif Formations.

Although the data points show a large scattering even for a specific unit, bulk-rock XRD analysis of the sandstones from all three units suggests a decrease in K-feldspar content with increasing illite layers in the mixed-layer clay minerals (Figure 7). This decrease is similar for all three units, albeit there is a large difference in starting composition of the I-S mixed-layer clay minerals.

INTERPRETATION

To understand the three different trends in the transformation of smectite to illite in I-S mixed-layer clay minerals with increasing depth or temperature, the controlling factors of the transformation need to be considered. For clarity these factors have been grouped as (1) physical: temperature, time, and pressure; (2) chemical: potassium availability, fluid and rock composition, rock/water ratio of the sediments, and presence of organic matter and, (3) depositional environment: starting composition of the I-S mixed-layer clay mineral and weathering conditions.

Physical factors

The physical factors time, temperature, and pressure are controlled by the age and subsequent burial history of the sediments studied. Final deposition of the uppermost Gharif sediments is dated at the end of Artinskian time, ~20 Ma after final deposition of Al Khlata sediments during Lower Sakmarian time (Figure 2; Besems and Schuurman, 1987). This difference in age of deposition is negligible compared to the ~280 Ma duration of burial diagenesis of the Haushi Group (Figure 3).

Temperature and pressure during burial diagenesis are a function of the burial history of the sediments (Figure 3). Since there is no hiatus or tectonic event between deposition of the Gharif and Al Khlata Formations, they share an identical burial history resulting in comparable thermal and pressure histories. These similarities do not indicate any significant differences in the physical factors during burial diagenesis of the two formations. Thus, physical factors are not thought to be the causes for the observed differences in the starting composition and the depth/temperature trends of I-S mixed-layer clay minerals.

Chemical factors

The chemical factors (potassium availability, fluid and rock composition, rock/water ratio, and presence of organic matter) are reflected in the detrital composition and the paragenetic sequence. The detrital composition was determined for 255 sandstone samples with grain sizes ranging from coarse to fine.

Monomineralic and polycrystalline quartz (6%–98%), plagioclase and K-feldspar (2%–49%), and various rock fragments (to 90%) are the main detrital components of the sandstones. Detrital mica is only a minor component (<5%) and present over the whole depth range. The samples plot in a Q-F-RF diagram (McBride, 1963) mainly in the quartzarenite, subarkose and arkose fields (Figure 8). Samples with >10% rock fragments are the exception; these are mainly sandstones containing reworked caliche clasts (Upper+Middle Gharif) or sandstones from the Al Khlata Formation in the outcrop area. The relatively high mineralogical maturity of the Lower Gharif and Al Khlata sandstones is partly due to the inferred source, *e.g.,* continental block, and partly caused by dissolution of geochemically unstable feldspars and rock fragments during burial diagenesis (Hartmann, 1996). The higher average feldspar content of the Upper+Middle Gharif points to a multi-origin of the Gharif sandstones, including reworked older units (*e.g.,* Haima, Huqf) and Al Khlata sediments (Silva *et al.,* 1996). Both the intra-plate position of the Sultanate of Oman during Upper Paleozoic times (Loosveld *et al.,* 1996) and Dickinson's (1985) classification of sandstone provenance agree with a continental block

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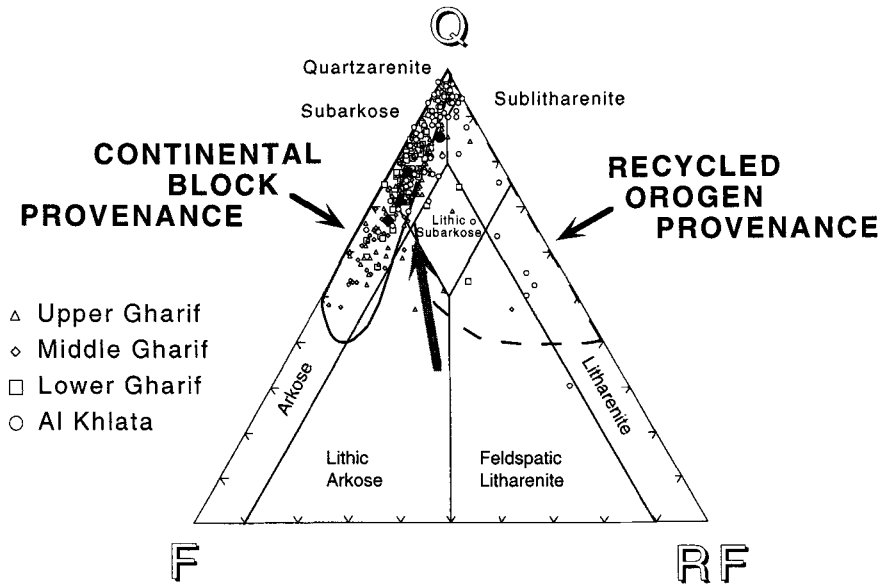


Figure 8. Composition of sandstones from the Haushi Group shown in the Q-F-RF diagram of McBride (1963). Filled symbols represent mean values. The gray arrow marks the most likely path of geochemical alteration during burial diagenesis. The two source areas marked are from Dickinson (1985).

origin for the majority of the sandstone material (Figure 8).

Diagenetic alterations of the sandstones within the Gharif Formation (Hartmann, 1996) and Al Khlata Formation (Ramseyer, 1983; Hartmann *et al.,* 1999) were studied. Because of the similar burial history, the burial diagenetic alterations are comparable within the generalized paragenetic sequence for the two formations (Figure 9). Eogenetic alterations, reflecting the near-surface conditions during and shortly after deposition of the sandstones, vary significantly. Gharif sandstones of the Upper and Middle units contain *in situ* caliche and detrital grains with tangential clay rims and hematite. These early features are indicative of semiarid to arid conditions during sedimentation (Walker, 1976; Wright, 1992). They are not present in the glaciogenic, continental Al Khlata Formation (Ramseyer, 1983; Hartmann *et al.,* 1999).

The bulk-rock XRD analysis of the sandstones revealed that the vast majority have detectable amounts of K-feldspar even at depths >4 km (or >130°C present day burial temperature) where I-S mixed-layer clay minerals contain >80% illite layers (Figures 6 and 7). Thus, K-feldspar is present at depths where the transformation of I-S mixed-layer clay minerals is practically complete. This observation implies that potassium availability was similar in all three stratigraphic units during burial and therefore can be ruled out as a source for the different trends at shallow burial (Figure 6).

In each depositional environment, glaciogenic for the Al Khlata Formation and semiarid to arid for the Gharif Formation, little organic matter was present after deposition because of either an initially low level caused by unfavorable climatic conditions (glaciogenic) or oxidation as revealed by the red color of the sediments typical for semiarid to arid conditions (Walker, 1976). Input of organic material during hydrocarbon migration in the subsurface, a second source for organic acid anions, is similar for both formations (Grantham *et al.,* 1987).

The data presented suggest that the above chemical factors were comparable for the three stratigraphic units. Thus, the observed differences in the starting composition and depth/temperature trends of illite layers in I-S mixed-layer clay minerals are not caused by marked differences in chemical factors.

Environmental factors

Other possible causes for the variable starting composition and the resulting difference in the three trends of the I-S mixed-layer clay minerals may be variable source material and the climatic conditions during weathering, transport, and deposition.

Petrographic information from thin sections together with evidence from paleoenvironmental reconstructions show that the Gharif sandstones contain reworked Al Khlata material (Silva *et al.,* 1996) with few additional clasts derived from older Paleozoic and Proterozoic rocks of south Oman. The source of the Al Khlata material is more difficult to explain. The general S-N and SE-NW orientation of meltwater channels in south Oman (Heward, 1990; Silva *et al.,* 1996) indicates that the main source for the Al Khlata material was Proterozoic basement and Paleozoic siliciclastic sediments from south Oman and neighboring

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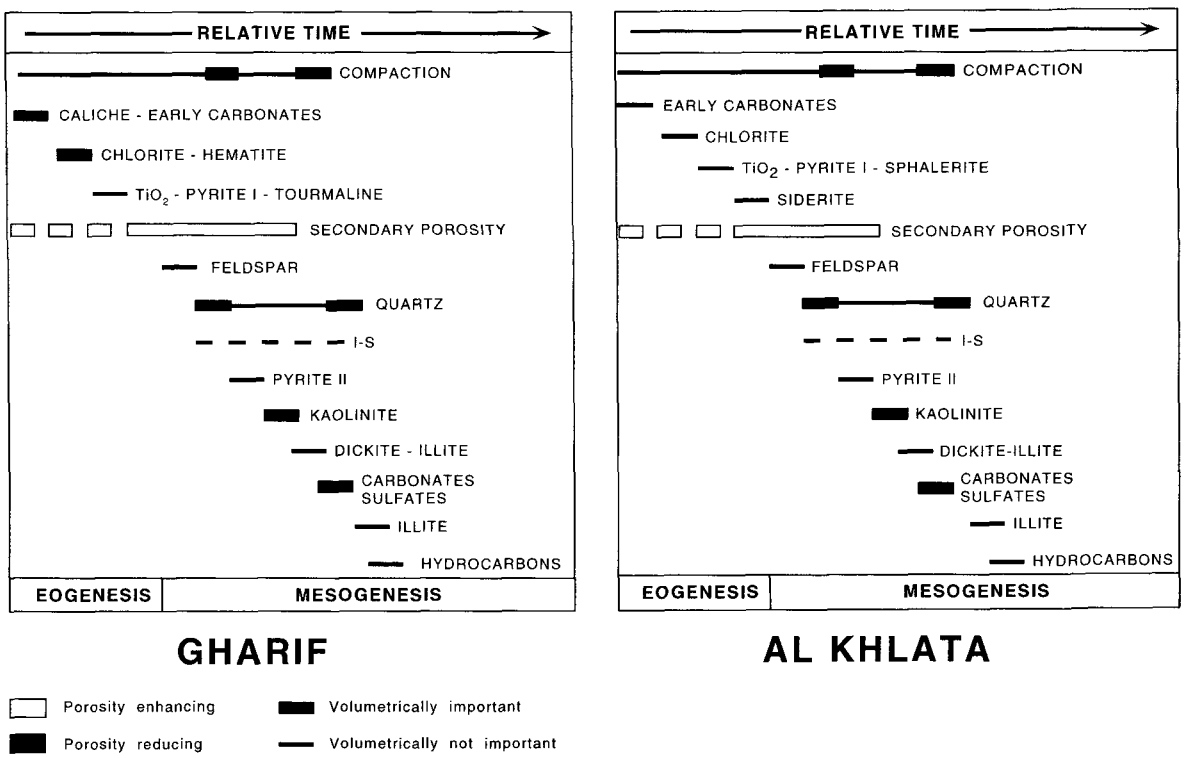


Figure 9. Generalized paragenetic sequence for the Gharif Formation and the Al Khlata Formation sediments (Hartmann *et al.*, 1999).

Yemen. Additional reworked Paleozoic sandstones and shales may be derived locally from the Huqf-Haushi High and the proposed SE-prolongation of Arabia, the paleo-highland at the western end of the Lut block (Husseini, 1992).

The geological history of the southern part of Oman, an important primary source area for the Al Khlata Formation and the Gharif Formation, is known to some extent (Sykes and Abu-Risheh, 1989; Visser, 1991). Burial history curves show that Late Proterozoic, Cambrian, and Ordovician sediments were rapidly buried to almost 4000 m, then uplifted by the final phase of the Pan-African tectonic events which affected Oman during most of the Upper Silurian to Lower Devonian times (Al-Marjeby and Nash, 1986) (Figure 10). Furthermore, Middle Devonian to Lower Carboniferous strata are missing in the basin, possibly related to Hercynian uplift and erosion (Hughes Clarke, 1988; Husseini, 1992). These phases of tectonic uplift brought sediments of the Huqf and Haima Groups to the surface, where they were eroded and reworked into the Al Khlata sediments (Figure 10). The deep burial of the proposed detrital source for the Al Khlata Formation is most likely the cause of the high content of illite layers in the I-S mixed-layer clay minerals still present in the surface samples of the Al Khlata Formation. This interpretation is in accordance with implications from paleolatitude reconstructions (Hughes Clarke, 1990; Beydoun, 1991), which show a rapid northward drift for Oman during Haushi deposition, bringing it from 50° south at the end of the Carboniferous to close to the equator at the end of Permian time (Figure 11). The colder climatic conditions during Al Khlata deposition, resulting from the southern position and the Gondwana glaciation (Beydoun, 1991; Husseini, 1992), and thus the less pronounced intensity of chemical weathering, support an inherited illite-rich composition of the I-S mixed-layer clay minerals. The rapid warming and change from a cold to a semiarid/arid climate during deposition of Haushi sediments, as confirmed by palynology (Love, 1994; Broutin *et al.*, 1995), are again in accordance with the observed decrease of illite layers in the starting composition of I-S mixed-layer clay minerals from the base to the top of the Haushi Group (Figure 6). Illite weathering might be the key to understand the observed pattern of the widespread starting composition of I-S mixed-layer clay minerals in the Haushi Group sediments.

Illite weathering

Weathering of illite to smectite is a multistage chemical process (Figure 12), whereby the smectitization occurs through intermediate I-S mixed-layer clay mineral phases (Thorez, 1989). In a Si-rich environment with moderate or low leaching combined

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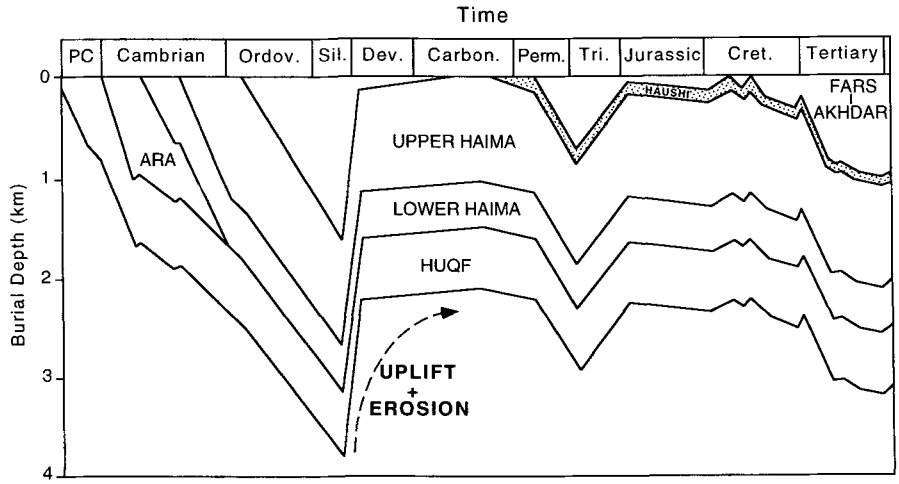


Figure 10. Burial history for south Oman, one of the proposed source areas for the detritus of the Haushi Group sediments (after Sykes and Abu-Risheh, 1989; Visser, 1991). Note that the proposed sources, the Huqf and the Haima Groups, were rapidly uplifted from deep burial and continuously eroded.

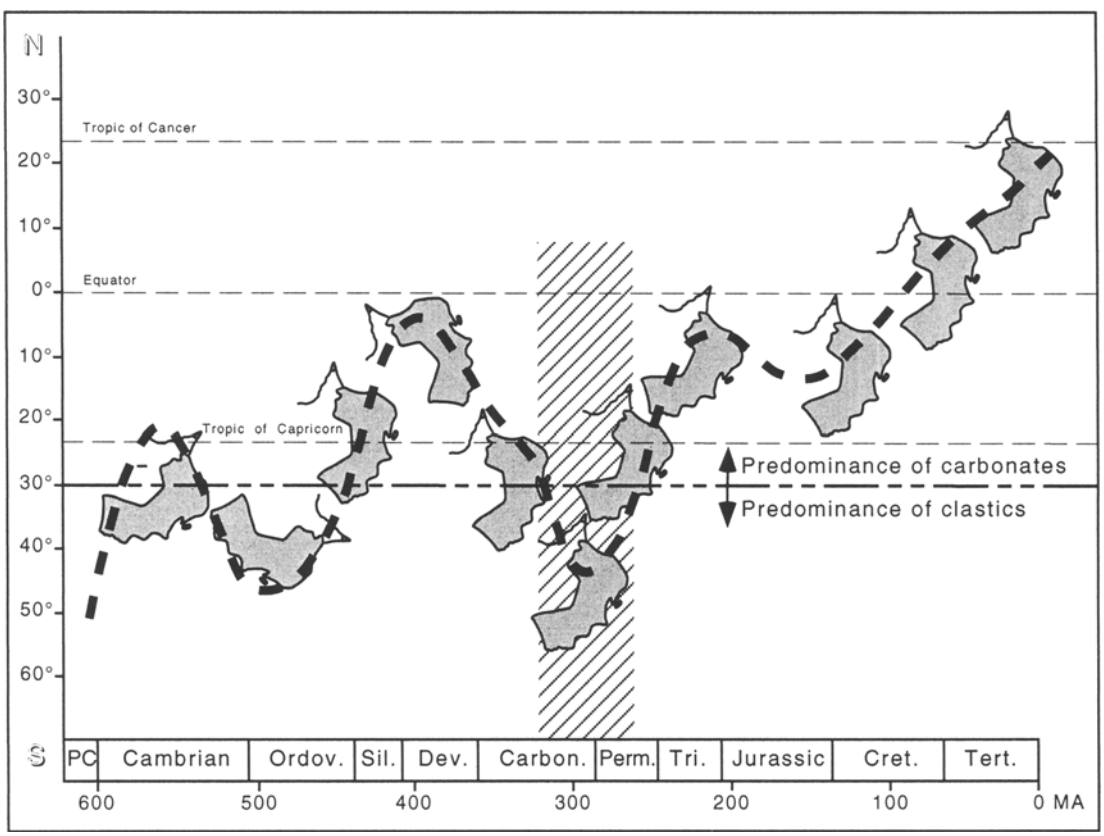


Figure 11. Paleolatitude reconstruction for the Sultanate of Oman. The shaded area represents the studied time interval. Location north of 30° south latitude is predominantly documented in carbonate sediments, whereas siliciclastics were deposited when Oman was located further to the south (after Hughes Clarke, 1990 and Beydoun, 1991).

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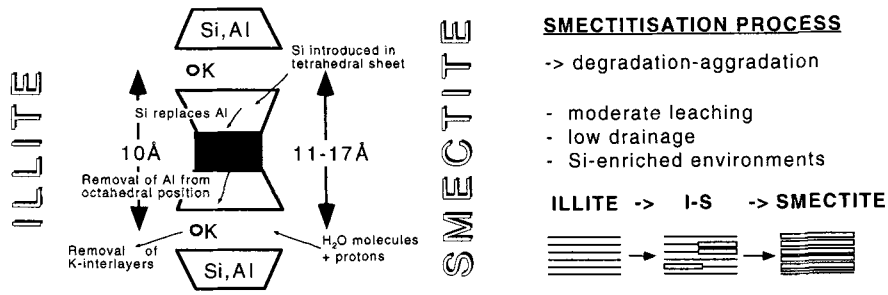


Figure 12. Schematic presentation of ion movement during illite weathering, showing the possible geochemical pathway for the smectitization process through an intermediate I-S mixed-layer clay mineral stage (modified after Thorenz, 1989).

with low drainage, smectite will be the final phase. The intensity of weathering largely determines if illite decomposes to smectite or to an intermediate I-S mixed-layer phase (Thorez, 1989). In soil environments, this often leads to a very complex assemblage of I-S mixed-layer clay minerals (Velde, 1992).

In soils from cold and temperate climates, such as those found during the deposition of Al Khlata sandstones, rates of weathering reactions tend to be reduced. This is generally the result of both the temperature dependence of the rate constants, and the effect of organic acids derived from plant material (Ugolini and Sletten, 1991). Weathering in cold areas is quite distinct from other climates, and a common result is the formation of interstratified clays (Righi and Meunier, 1995). Climatic changes leading to semiarid to arid conditions during Middle and Upper Gharif time, with only moderate leaching due to the scarcity of water, still favor smectitization. Weathering conditions during the Lower Gharif were somewhere between the conditions described for the Al Khlata and Upper+Middle Gharif.

The influence of illite weathering on the I-S mixed-layer clay mineral composition is clearly evident in the starting composition found in the surface samples of the three Haushi Group units: Al Khlata, Lower Gharif, and Middle+Upper Gharif, with 60, 50 and 20% illite layers in I-S mixed-layer clay minerals, respectively (Figure 6). In addition, the pronounced difference in the starting composition is visible in samples down to 80°C present day burial temperatures, as indicated by the three distinct composition-temperature trends of the I-S mixed-layer clay minerals. Above this temperature, the composition-temperature trends of the I-S mixed-layer clay minerals are identical for the three units. The effect of changing primary composition of illite through changes in the weathering conditions is thus clearly recognizable in shallow buried sediments where the temperature did not exceed 80°C (Figure 6).

CONCLUSIONS

Evidence from the sample composition, paragenetic sequence, and the burial and thermal history excludes potassium availability, temperature, time, pressure, rock/water ratio of the sediments, fluid and rock composition, or the presence of organic acid anions as likely causes for the differences in the starting composition and the composition-temperature trends of I-S mixed-layer clay minerals in the Haushi Group sediments. As a consequence, the most plausible cause for the observed compositional trends is that these sediments had a wide range of starting compositions in the I-S mixed-layer clay minerals. Furthermore, since the Al Khlata and the Gharif Formations share a comparable primary mineralogical composition, differential weathering, caused by a pronounced change of climate during deposition, is the most likely reason for the wide range in starting compositions.

This climatically induced compositional variability in the lower-temperature range must be considered when using I-S minerals for calibration in thermal modeling. In addition, the initial compositional variability of I-S mixed-layer clay minerals also affects the calculated amounts of water and ions released during illitization of I-S and their effect on burial diagenesis.

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REFERENCES

Al-Marjeby, A. and Nash, D. (1986) A summary of the geology and oil habitat of the Eastern Flank Hydrocarbon Province of South Oman. *Marine and Petroleum Geology*, **3**, 306–314.

https://doi.org/10.1346/CCMN.1999.0470203 Published online by Cambridge University Press

Alsharhan, A.S. (1993) Facies and sedimentary environment of the Permian carbonates (Khuff Formation) in the United Arab Emirates. *Sedimentary Geology,* **84,** 89–99.

Besems, R.E. and Schuurman, W.M.L. (1987) Palynostratigraphy of late Palaeozoic glacial deposits of the Arabian Peninsula with special reference to Oman. *Palynology,* **11,** 37–53.

Beydoun, Z.R. (1991) Arabian Plate hydrocarbon geology and potential—A plate tectonic approach. *American Association of Petroleum Geologists Studies in Geology,* Tulsa, Oklahoma, **33,** 1–78.

Boles, J.R. and Franks, S.G. (1979) Clay diagenesis in Wilcox Sandstones of Southwest Texas: implication of smectite diagenesis on sandstone cementation. *Journal of Sedimentary Petrology,* **49,** 55–70.

Braakman, J.H., Levell, B.K., Martin, J.H., Potter, T.L., and van Vliet, A. (1982) Late Palaeozoic Gondwana glaciation in Oman. *Nature,* **299,** 48–50.

Broutin, J., Roger, J., Platel, J.P., Angiolini, L., Baud, A., Bucher, H., Marcoux, J., and Al-Hasmi, H. (1995) The Permian Pangea. Phytographic implications of new palaeontological discoveries in Oman (Arabian Peninsula). *Comptes Rendus de l'Académie des Sciences de Paris, Série IIa,* **321,** 1069–1086.

Bruce, C.H. (1984) Smectite dehydration-its relation to structural development and hydrocarbon accumulation in Northern Gulf of Mexico Basin. *American Association of Petroleum Geologists Bulletin,* **68,** 673–683.

Burst, J.F. (1959) Postdiagenetic clay mineral environmental relationships in the Gulf Coast Eocene. In *Clays and Clay Minerals: Sixth National Clays and Clay Minerals Conference Proceedings,* A. Swineford, ed., Pergamon Press, London, 411 pp.

Burst, J.F. (1969) Diagenesis of Gulf Coast Clayey Sediments and Its Possible Relation to Petroleum Migration. *American Association of Petroleum Geologists Bulletin,* **53,** 73–93.

Colton-Bradley, V.A. (1987) Role of pressure in smectite dehydration-effects on geopressure and smectite-to-illite transformation. *American Association of Petroleum Geologists Bulletin,* **71,** 1414–1427.

De la Grandville, B.F. (1982) Appraisal and development of a structural and stratigraphic trap oil field with reservoirs in glacial to periglacial clastics. *American Association of Petroleum Geologists Memoir,* **32,** 267–286.

Dickinson, W.R. (1985) Interpreting provenance relations from detrital modes of sandstones. In *Provenance of arenites,* G.G. Zuffa, ed., D. Reidel, Dordrecht, 333–361.

Dickson, J.A.D. (1966) Carbonate identification and genesis as revealed by staining. *Journal of Sedimentary Petrology,* **36,** 491–505.

Dutta, N.C. (1986) Shale compaction, burial diagenesis, and geopressures: a dynamic model, solutions and some results. In *Thermal modelling in sedimentary basins,* J. Burrus, ed., Editions Technip., Paris, 149–172.

Elliott, W.C. and Matisoff, G. (1996) Evaluation of kinetic models for the smectite to illite transformation. *Clays and Clay Minerals,* **44,** 77–87.

Essene, E.J. and Peacor, D.R. (1995) Clay mineral thermometry-a critical perspective. *Clays and Clay Minerals,* **43,** 540–553.

Focke, J.W. and van Popta, J. (1989) Reservoir Evaluation of the Permian Gharif Formation, Sultanate of Oman. *Society of Petroleum Engineers,* **17978,** 517–528.

Freed, R.L. and Peacor, D.R. (1989) Variability in temperature of the smectite/illite reaction in Gulf Coast sediments. *Clay Minerals,* **24,** 171–180.

Gharabi, M. and Velde, B. (1995) Clay mineral evolution in the Illinois basin and its causes. *Clay Minerals,* **30,** 353–364.

Grantham, P.J., Lijmbach, G.W.M., Posthuma, J., Hughes Clarke, M.W., and Willink, R.J. (1987) Origin of crude oils in Oman. *Journal of Petroleum Geology,* **11,** 61–80.

Guit, F.A., Al-Lawati, M.H., and Nederlof, P.J.R. (1994) Seeking new potential in the Early-Late Permian Gharif play, west Central Oman. *Middle East Petroleum Geoscience,* 447–462.

Hartmann, B.H. (1996) Diagenesis and pore-water evolution of the Lower Permian Gharif Formation (Sultanate of Oman). Ph.D. thesis, Geologisches Institut der Universität Bern, Switzerland, 342 pp.

Hartmann, B.H., Juhász-Bodnár, K., Ramseyer, K., and Matter, A. (1999) Polyphased quartz cementation and its sources: a case study from the Upper Palaeozoic Haushi Group sandstones, Sultanate of Oman. In *Quartz cementation in oil field sandstones,* R. Worden and S. Morad, eds., International Association of Sedimentologists Special Publication 29, in press.

Heward, A.P. (1990) Salt removal and sedimentation in Southern Oman. In *The Geology and Tectonics of the Oman Region,* A.H.F. Robertson, M.P. Searle, and A.C. Ries, eds., Oxford Geological Society, 637–652.

Hillier, S., Mátyás, J., Matter, A., and Vasseur, G. (1995) Illite/smectite diagenesis and its variable correlation with vitrinite reflectance in the Pannonian Basin. *Clays and Clay Minerals,* **43,** 174–183.

Hower, J., Eslinger, E.V., Hower, M.E., and Perry, E.A. (1976) Mechanism of burial metamorphism of argillaceous sediment: 1. Mineralogical and chemical evidence. *Geological Society of America Bulletin,* **87,** 725–737.

Huang, W.L., Longo, J.M., and Pevear, D.R. (1993) An experimentally derived kinetic model for smectite-to-illite conversion and its use as a geothermometer. *Clays and Clay Minerals,* **41,** 162–177.

Hughes Clarke, M.W. (1988) Stratigraphy and rock unit nomenclature in the oil-producing area of interior Oman. *Journal of Petroleum Geology,* **11,** 5–60.

Hughes Clarke, M.W. (1990) *Oman's Geological Heritage.* Department of Public Affairs and Information, Muscat: Petroleum Development Oman, 247 pp.

Husseini, M.I. (1992) Upper Palaeozoic tectono-sedimentary evolution of the Arabian and adjoining plates. *Journal of the Geological Society of London,* **149,** 419–429.

IES GmbH (1994). PetroMod®. Vers 2.4. Jülich, 603 pp.

Kashfi, M.S. (1992) Geology of the Permian super-giant gas reservoirs in the greater Persian Gulf area. *Journal of Petroleum Geology,* **15,** 465–480.

Lanson, B. and Velde, B. (1992) Decomposition of X-ray diffraction patterns: A convenient way to describe complex I-S diagenetic evolution. *Clays and Clay Minerals,* **40,** 629–643.

Levell, B.K., Braakman, J.H., and Rutten, K.W. (1988) Oil-bearing sediments of Gondwana glaciation in Oman. *American Association of Petroleum Geologists Bulletin,* **72,** 775–796.

Loosveld, R.J.H., Bell, A., and Terken, J.J.M. (1996) The tectonic evolution of Interior Oman. *GeoArabia,* **1,** 28–51.

Love, C.F. (1994) The palynostratigraphy of the Haushi Group (Westphalian-Artinskian) in Oman. In *Micropalaeontology and hydrocarbon exploration in the Middle East,* M.D. Simmons, ed., Chapman and Hall, London, 23–39.

McBride, E.F. (1963) A classification of common sandstones. *Journal of Sedimentary Petrology,* **33,** 664–669.

Mercadier, C.G.L. and Livera, S.E. (1993) Applications of the formation micro-scanner to modelling of Palaeozoic reservoirs in Oman. *Special Publication of International Association of Sedimentologists,* **15,** 125–142.

https://doi.org/10.1346/CCMN.1999.0470203 Published online by Cambridge University Press

Millson, J.A., Mercadier, C.L., Livera, S.A., and Peters, J.M. (1996) The Lower Palaeozoic of Oman and its context in the evolution of a Gondwana continental margin. *Journal of the Geological Society,* **153,** 213–230.

Moore, D.M. and Reynolds, R.C. (1989) *X-ray Diffraction and the Identification and Analysis of Clay Minerals.* Oxford University Press, New York, 332 pp.

Perry, E.A. and Hower, J. (1970) Burial diagenesis in Gulf Coast pelitic sediments. *Clays and Clay Minerals,* **18,** 165–177.

Platel, J.P., Philip, J., Bourdillonde Grissac, C., Babinot, J.F., Roger, J., and Mercadier, C. (1994) Characteristics of the Campanian transgression onto the Haushi-Huqf basement (East-Oman). Stratigraphy, geodynamic setting and palaeoenvironments. *Bulletin de la Société Geologique de France,* **165,** 147–161.

Pollastro, R.M. (1990) The illite/smectite geothermometer—Concepts, methodology and application to basin history and hydrocarbon generation. In *Applications of thermal maturity studies to energy exploration,* V.F. Nuccio and C.E. Barker, eds., Society of Economic Paleontoligists and Mineralogists, Tulsa, Rocky Mountain Section, 1–18.

Pollastro, R.M. (1993) Considerations and applications of the illite/smectite geothermometer in hydrocarbon-bearing rocks of Miocene to Mississipian age. *Clays and Clay Minerals,* **41,** 119–133.

Powers, M.C. (1959) Adjustment of clays to chemical change and the concept of the equivalence level. *Clays and Clay Minerals,* **6,** 309–326.

Powers, M.C. (1967) Fluid-release mechanisms in compacting marine mudrocks and their importance in oil exploration. *American Association of Petroleum Geologists Bulletin,* **51,** 1240–1254.

Pytte, A.M. and Reynolds, R.C. (1989) The thermal transformation of smectite to illite. In *Thermal history of sedimentary basins: Methods and case histories,* N.D. Naeser and T.H. McCulloh, eds., Springer-Verlag, New York, 133–140.

Ramseyer, K. (1983) Bau eines Kathodenlumineszens-Mikroskopes und Diagenese-Untersuchungen an permischen Sedimenten aus Oman. Ph.D. thesis, Geologisches Institut der Universität Bern, Switzerland, 153 pp.

Rettke, R.C. (1981) Probable burial diagenetic and provenance effects on the Dakota Group clay mineralogy. *Journal of Sedimentary Petrology,* **51,** 541–551.

Righi, D. and Meunier, A. (1995) Origin of clays by rock weathering and soil formation. Origin and mineralogy of clays. In *Clays in the Environment,* B. Velde, ed., Springer-Verlag, Berlin, 44–161.

Robertson, H.E. and Lahann, R.W. (1981). Smectite to illite conversion rates: Effect of solution chemistry. *Clays and Clay Minerals,* **29,** 129–135.

Silva, F.P., Silva, A.C., Martinius, A.W., and Weber, K.J. (1996) The Tórtola Fluvial System: An Analogue for the Upper Gharif of the Sultanate of Oman. *GeoArabia,* **1,** 325–342.

Small, J.S. (1994) Fluid composition, mineralogy and morphological changes associated with the smectite-to-illite reaction: An experimental investigation of the effect of organic acid anions. *Clay Minerals,* **29,** 539–554.

Sykes, R.M. and Abu-Risheh, A.K. (1989) Exploration of deep Palaeozoic and Pre-Cambrian plays in the Sultanate of Oman. In *OAPEC/ADNOC seminar on deep formations in the Arab Countries: hydrocarbon potential and exploration techniques, October 1989,* Proceedings of Country Reports and Case Studies, Abu Dhabi, E71–E133.

Thorez, J. (1989) Between the crystal and the solutions. A graphical overview of the passage to, from and of the clay minerals in the lithosphere during weathering. In *Weathering; its Products and Deposits, I. Processes.,* K.S. Balasubramaniam, V.P. Evangelou, G. Faure, J. Goni, P.L.C. Grubb, P.A. Hill, O. Lahodney-Sarc, A.J. Melfi, E. Mendelovici, A.P. Nikitina, W.F. Pickering, and S.S. Augustithis, eds., Theophrastus Publishers S.A., Greece, 49–120.

Tschopp, R.H. (1967) The general geology of Oman. In *7th World Petroleum Congress,* Mexico, 231–249.

Ugolini, F. and Sletten, R.S. (1991) The role of proton donors in pedogenesis as revealed by soil solution studies. *Journal of Soil Science,* **151,** 1–21.

Vasseur, G. and Velde, B. (1993) A kinetic interpretation of the smectite-to-illite transformation. In *Basin modelling: advances and applications,* A.G. Doré, ed., Norwegian Petroleum Society Special Publication 3, Amsterdam, 173–184.

Velde, B. (1992) *Introduction to clay minerals.* Chapman & Hall, London, 199 pp.

Velde, B. and Vasseur, G. (1992) Estimation of the diagenetic smectite to illite transformation in time-temperature space. *American Mineralogist,* **77,** 967–976.

Visser, W. (1991) Burial and thermal history of Proterozoic source rocks in Oman. *Precambian Research,* **54,** 15–36.

Walker, T.R. (1976) Diagenetic origin of continental red beds. In *The continental Permian in Central, West and South Europe,* H. Falke, ed., D. Reidel, Dordrecht, 240–282.

Weaver, C.E. and Beck, K.C. (1971) Clay water diagenesis during burial: how mud becomes gneiss. *Geological Society of America Special Paper 134,* 96 pp.

Wei, H., Roaldset, E., and Bjorøy, M. (1996) Parallel reaction kinetics of smectite to illite conversion. *Clay Minerals,* **31,** 365–376.

Wright, V.P. (1992) Paleosol recognition: a guide to early diagenesis in terrestrial settings. In *Diagenesis III. Developments in Sedimentology,* **47,** K.N. Wolf and G.V. Chilingarian, eds., Elsevier, Amsterdam, 591–619.

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