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K TRANSFER DURING BURIAL DIAGENESIS IN THE MAHAKAM DELTA BASIN (KALIMANTAN, INDONESIA)

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Abstract—Progressively buried sandstones and shales of the Mahakam Delta Basin in Indonesia were studied. Mineralogical, morphological and K-Ar isotopic data were obtained for clay, mica and feldspar minerals. The data indicate that K necessary for the illitization of illite/smectite mixed-layer minerals was supplied mainly from K-feldspar alteration within the sandstones and from mica within the shales. Most of the K-feldspar alteration for both the shale and sandstone samples were observed outside the main zone of illitization, which is restricted to the upper 2000 m of sediment. The feldspar grains were altered below this depth for both lithologies. Therefore, illitization requires an open sedimentary system. This is in contrast to the illitization model for deeply buried shales of the Gulf Coast. That system is commonly assumed to be a closed K system.

Key Words—Diagenesis, Illitization, K transfer.

INTRODUCTION

The quantity of illite layers generally increases for the illite-smectite (I/S) phase as the sediment containing I/S mixed-layer minerals are progressively buried in sedimentary basins (Powers 1959, 1967; Weaver 1959, 1960; Dunoyer de Segonzac 1970; Perry and Hower 1970; Hower et al. 1976; Boles and Franks 1979). The geochemical and mineralogic trends of illitization with depth for the Tertiary sedimentary sequence of the Gulf Coast area have served as a model to explain the diagenetic evolution of illite minerals for many deeply buried shales.

Perry and Hower (1970) made a detailed examination of mineralogical and chemical variations for deeply buried shales from the Gulf Coast. They found that the amount of K-fixation increases with depth for the mixed-layer clays during progressive conversion into illite-layers. However, the bulk samples K content remained invariant with depth. They concluded that the K derived from alteration of detrital minerals was largely redistributed among the authigenic clay minerals within each rock volume. These conclusions were supported by a study by Hower et al. (1976). Their study became the basis for the Gulf Coast diagenetic model that explains evolution of smectite minerals for buried sediments. Ahn and Peacor (1986) added to this theory. They reported a transmission electron microscopy study of illite-to-smectite transition for Gulf Coast sediments. The illite layers grew within smectite layers under a nearly closed system where small amounts of water facilitated the short distance ion transfers. On the contrary, Weaver and Beck (1971) proposed a chemical budget calculation for deeply buried Gulf Coast sediments. They concluded that K for the illitization of smectite had to be brought to the diagenetic site from outside by fluids moving upward through the sedimentary column. Based on Rb-Sr and Sm-Nd data of < 0.1 μm clay fractions of shales from the same region, Ohr et al. (1991) inferred that illitization occurred in an open system. But their conclusion was mainly based on assumptions about the behavior of Sr and REE, which did not properly address the issue of mobility of alkali elements to the clay minerals. Awwiller (1993) observed a significant K content increase for progressively buried mudrock samples from Tertiary strata of the Texas Gulf Coast. He claimed that part of this increase had to come from the import of K into the shale. This implies an open-system for these rocks.

Detrital feldspar grains of progressively buried sediments were reported to decompose as a result of reactions with saline subsurface fluids (Wollast 1967; Huang and Keller 1970; Petrovic 1976; Altaner 1986). Aronson and Hower (1976) observed, as Perry and Hower (1970) and Hower et al. (1976) reported, an increase of K in the clay interlayers simultaneously with the disappearance of K-feldspar. They concluded that there must be a direct transfer of K from the K-feldspars to the clays within the same volume of rock. Decrease of K-feldspar, but also an increase in albite with depth was observed by other investigators (Land and Milliken 1981). Diagenetic reactions have major effects on K-Ar isotopic dates of whole rocks and their K-bearing detrital and authigenic components (Perry and Hower 1970; Hower et al. 1976; Aronson and Hower 1976; Lee et al. 1985; Hamilton et al. 1987; Jourdan et al. 1987; Liewig et al. 1987 and Glasmann et al. 1989). This means that the method is well suited

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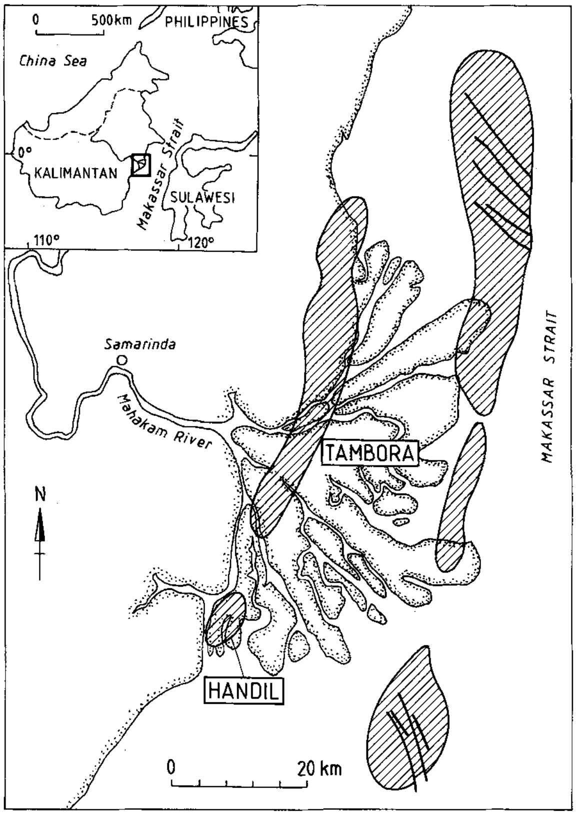

Figure 1. Location of the studied area and of the oil-fields from where the core samples were taken. The dashed areas contour the major oil fields in the near-shore and off-shore anticline structures.

to trace K transfers within sedimentary rocks. Perry (1974) examined the K-Ar systematics of clay size fractions from the Gulf Coast shales buried between 1580 and 5525 m. He observed that the proportion of illite layers in I/S of the <0.5 μm fraction increased from 20 to 80% as depth increased. This increase of illite layers was accompanied by decrease in the K-Ar dates from 164 ± 4 to 100 ± 2 Ma. Increase in temperature attendant with progressive burial of sediments was considered to cause diffusional loss of radiogenic ^{40}Ar from detrital sheet-minerals, as well as alteration of associated feldspars. This alteration induced pits and etches in the grains. It promoted K removal and probably preferential Ar diffusion along the structural defects of the feldspar minerals. Both alterations may have contributed to the age-depth trend.

The purpose of the present study is to provide additional information about the diagenetic evolution of smectite-to-illite clay minerals in progressively buried sedimentary sequences. A combined mineralogical, morphological and K-Ar isotopic study of authigenic clay and detrital feldspar and mica fractions was conceived to address the issue of K behavior during burial diagenesis. The illitization of smectite in Gulf Coast sediments occured within shale-dominated lithology. Interpretation of isotopic data, especially radiogenic isotopic data, for clay minerals in shales is not well constrained because of the inherent difficulty of extricating the potential influence of any detrital component in the analyzed fractions (Clauer et al. 1992; Clauer and Chaudhuri 1995). We have included observations and analyses of both authigenic and detrital minerals from shale and associated sandstone samples. The choice for the sedimentary deposits of the Mahakam Delta Basin rests on three facts that we consider important for such a study: (1) the drainage basin is small and well defined; (2) the sedimentation is characterized by a fairly homogeneous supply; and (3) the depositional history is simple.

GEOLOGICAL SETTING

The Mahakam River Delta comprises an area of about 5000 km^2 in the Makassar Strait between Borneo and the Sulawesi Islands (Figure 1). The sedimentary deposits beneath the delta consist of sediments 8000 to 10,000 m thick where the first 4000 m are oil-bearing shale-sandstone. It is presently producing hydrocarbons and is being explored for additional reserves. The depositional history began in the early Miocene period (Van der Vlerk and Umgrove 1927). The Mahakam River drainage basin covers an area of approximately 75,000 km^2, which is about one-tenth the area of the island. It is surrounded by 2000 m high mountains. They were formed between 90 and 100 Ma ago or prior and represent the major source of clastic materials that have been transported by the Mahakam River since the beginning of the Miocene period (Priyomarsono 1985). The deltaic body comprises three sedimentary complexes separated by two transgressive systems (Samuel and Muchsin 1975; Rose and Hartono 1978).

Seismic studies indicate the occurrence of an anticline and syncline axes system oriented SW-NE, perpendicular to the progradation of the delta. Several of these SW-NE anticlinal axes have been recognized on- and off-shore (Magnier and Sansu 1975). Structuration induced sediment erosion at the top of the proximal anticline structure before renewed burial (Duval et al. 1992). Deformation began during the late Miocene period and could have resulted from collision between the islands of Borneo and Sulawesi (Katali 1978; Letouzey et al. 1990). The present-day temperatures measured for different wells made on the top of the Handil anticline structure (Figure 1), increase from about 60°C at 1100 m to about 80°C at 2000 m (Burrus and Bois 1989). These temperatures are higher for the overpressured zones. Assuming a surface temperature of 20°C, the present-day geothermal gradiant is about 30°C/km. It is slightly below that based on paleotemperatures determined from fluid inclusion microthermometry of quartz overgrowths, which are at 95–

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105°C at 2000 m and 160–165°C at 4000 m in the Handil field. In the Tambora field, the paleotemperatures range from 145 to 170°C at 4000 m based on the determinations of the same fluid inclusions (Rinckenbach 1988). In the Handil field, vitrinite reflectance increases from 0.4% at 1000 m to 0.5% at 1900 m and to 0.7% at 3000 m (Combaz and de Matherel 1978).

SAMPLE DESCRIPTION AND ANALYTICAL METHODS

Among the eight rock samples selected for this study, six belong to one well of the Handil field. The two deepest samples were collected in a well of the Tambora field, which is located in the same anticlinal structure as the Handil field (Figure 1). The analyses were carried out on both sandstone and shale samples from cores taken at four depth zones (185–350 m, 1980–2000 m, 2595–2610 m, and about 4230 m) within a Miocene-to-Quaternary stratigraphic interval. To avoid possible artificial grain reduction of framework detrital grains during sample preparation, the rocks were disaggregated by a thawing-freezing method. They were subsequently size fractionated following the procedure of Liewig et al. (1987). All fractions were investigated by X-ray diffraction (XRD) and electron microscopy.

Illite, I/S, kaolinite/dickite and chlorite amounts were determined for the different size fractions by comparing the heights of the major XRD [001] reflection peaks obtained for each mineral component on the untreated, ethylene-glycolated, hydrazine monohydrate vapored and heated aliquots of the same smear slides. Dickite was distinguished from kaolinite on the basis of SEM observations. Occurrence and semiquantitative contents of the framework minerals, such as quartz, K-feldspar, plagioclase and mica of the coarse size fractions were estimated by comparing the major [hkl] reflection peaks of each component on XRD's of powder preparations (Weber and Larqué 1969).

K-Ar isotopic analyses were carried out on the different size fractions following a technique similar to that described by Bonhomme et al. (1975). The samples were heated at 80°C for at least 24 h before Ar analyses. This heat treatment is to reduce atmospheric contamination due to the preparation. The K content were determined by flame spectrophotometry with a precision better than 2% for K contents greater than 0.1%. Each aliquot determination was framed by standard analyses. A periodic control of the $^{40}Ar/^{36}Ar$ ratio of the blank of the extraction line and the mass spectrometer provided a mean value of about 303.0, which is consistent with an atmospheric $^{40}Ar/^{36}Ar$ ratio at 295.5 (Nier 1950). During the course of this study, the international glauconite standard, GLO, was analyzed 15 times and its radiogenic ^{40}Ar content averaged 24.64 ± 0.30 (2σ) $\times 10^{-6}$ cm³/g STP. This value was compared to the reference value, which is $24.85 \pm 0.24 \times 10^{-6}$ cm³/g STP (Odin 1982). Analyses of all size fractions for each sample were not systematically possible because quantities of extracted material were limited.

RESULTS

Major changes were recorded for the mineralogy and K-Ar dates of the studied fractions from shale and sandstone samples. These changes are discussed in the following section.

Mineralogy

WHOLE-ROCK MINERALOGY. The framework grains of the sandstone samples for the shallow part of the sequence consist mainly of quartz, lithic fragments and alkali feldspar grains. The clay fractions consisted mainly of kaolinite, illite, I/S and chlorite (Table 1). The authigenic minerals were mostly pyrite and carbonate (siderite). Below 2000 m, the sandstones contained more detrital mica flakes (biotite and muscovite) in addition to quartz, lithic fragments and alkali feldspars. The authigenic minerals consisted of silica, dolomitic carbonate, kaolinite-dickite and illitic minerals occuring mainly as I/S with high amounts of illite layers. The kaolinite/dickite contents was approximately 50% of the <2 μm clay fraction. The remaining 50% was represented by illite, 20%; I/S, 20%; and chlorite, 10%. Kaolinte-dickite was most abundant in the 0.4–2 μm fractions, and I/S in the <0.4 μm fractions. The extent of silicification for the deeper section (>2000 m) of the sequence was positively correlated with the quantity of clay in the sandstones. Optical observation revealed that dolomitic cementation postdated silicification, but pre-dated dissolution of siderite, feldspar minerals and lithic fragments.

For the shallow section sequence (<2000 m), the shales typically consisted of quartz, micas and clays (Table 2). Minor amounts of pyrite, natrojarosite, K-feldspar and albite were also detected. The composition of the clay minerals for the <2 μm fractions consisted of 50% kaolinite, 20% illite, 30% I/S and trace amounts of chlorite. As compared to the shales of the upper section, those of the lower part (>2000 m) are slightly enriched with mica-type minerals and relatively depleted with quartz. Siderite is also a common secondary constituent and albite is relatively more abundant in the lower sequence section. The <2 μm clay minerals composition of the lower section shales (>2000 m) of the stratigraphic interval is approximately the same as that of the upper part (<2000 m).

CLAY MINERALOGY OF THE <0.4 μM FRACTION. The decrease in percent expandable layers of the <2 μm size I/S for both the sandstone and the shale lithologies (Figure 2) was studied in the well of Handil field by using a simulation code developed by Mossmann

Table 1. Mineralogic composition of the separated size fractions of Mahakam sandstones from different depths.

Samples	Clay fraction				Framework minerals				
	kaolinite	I/S	illite	chlorite	quartz	K-spars	plagio.	micas	others
Misedor (350 m)									
<0.4 μm	50	35	10	5	0	0	0	0	0
2–40 μm	nd	nd	nd	nd	49	1	0	49	1
40–63 μm	nd	nd	nd	nd	86	1	0	9	4
HD1 (1978 m)									
<0.4 μm	40	50	10	0	0	0	0	0	0
2–6 μm	nd	nd	nd	nd	33	2	0	57	8
10–20 μm	nd	nd	nd	nd	58	1	0	36	5
20–40 μm	nd	nd	nd	nd	70	0	0	25	5
40–63 μm	nd	nd	nd	nd	80	2	0	15	3
HD1 (2595 m)									
<0.4 μm	40	45	10	5	0	0	0	0	0
2–6 μm	nd	nd	nd	nd	31	1	1	67	0
6–10 μm	nd	nd	nd	nd	37	1	1	61	0
10–20 μm	nd	nd	nd	nd	58	1	1	40	0
20–40 μm	nd	nd	nd	nd	89	1	0	10	0
40–63 μm	nd	nd	nd	nd	91	1	0	7	1
TM29 (4228 m)									
<0.4 μm	50	30	20	nd	X	0	0	0	0
2–6 μm	nd	nd	nd	nd	47	0	14	38	1
6–10 μm	nd	nd	nd	nd	48	0	12	40	0
10–20 μm	nd	nd	nd	nd	56	0	7	37	0
20–40 μm	nd	nd	nd	nd	85	1	7	3	4
40–63 μm	nd	nd	nd	nd	90	0	6	2	2

The amounts are in percent for each category; I/S stands for illite/smectite mixed layers, K-spars for K-feldspars, plagio. for plagioclases, nd for not determined, X for abundant; among the other framework minerals are pyrite and siderite.

(1991). For the upper 900 m of sediment, the amount of expandable layers was constant at about 65 wt% with a random organization of the layering. Between 900 and 1200 m depth, the amount of expandable layers decreased to about 30%, while the layer organization became short-distance ordered. Between 1200 and 3000 m depth, the amount of expandable layers decreased progressively toward 20%. Between 3200 and 4000 m depth, the amount of expandable layers remained at approximately 10%, with long-distance ordering. The overall evolution of the amount of expandable layers in the <2 μm I/S seemed to have occurred in an abrupt step at about 1000 m. No significant difference in the amount of expandable layers in the I/S was detected depending on the lithofacies of the rocks (Figure 2). Characteristic XRD's for the <2 μm I/S are outlined for each major step together with the corresponding computed diagrams (Figure 3). The <0.4 μm clay fractions of the progressively buried sandstones consisted of similar contents of 40–50% kaolinite, 30–50% I/S, 10–30% illite, and 0–5% chlorite (Table 1). The equivalent <0.4 μm clay fraction of the associated shale samples contained similar amounts of the same components (Table 2).

Transmission electron microscopic observations of the I/S particles showed significant differences between the particles of the sandstones collected at the surficial (<1000 m) and the deepest (>3000 m) zones of the sequence. For the surficial zone, the particles were systematically of flake-type particles, which characterized a detrital origin (Figure 6A). For the deepest zone, they appear as lath-type particles, which implies a major reorganization including a dissolution-crystallization process (Figure 6I). Conversely, the I/S particles remain similar to the shale samples of the surficial (<1000 m) and the deepest (>3000 m) zones of the sequence. They were always flake-type, while presenting some discrete euhedral overgrowths around the flakes for the deeper zone (Rinckenbach 1988).

K-Ar Isotopic Data

SANDSTONES. Changes in K content with depth, among the different >0.4 μm size fractions, were only marked by a small decrease in K content between 2000 and 2600 m (Table 3, Figure 4). But the K content variation with depth for the <0.4 μm fraction appears different as K increased progressively to twice the value from a depth of 350 m to a depth of 2600 m. The K increase with increased depth was reversed below 2600 m. The K content of the <0.4 μm fraction at about 4230 m was similar to that near the top of the sequence.

Radiogenic ^{40}Ar contents for the 0.4–2 μm fraction remained approximately the same between 350 and 2000 m (5.93 to 6.20 × 10^{-6} cm^3/g). They sharply decreased to 3.5 × 10^{-6} cm^3/g between 2000 and 2600

Table 2. Mineralogic composition of the separated size fractions of Mahakam shales from different depths.

Samples	Clay fraction				Framework minerals				
	kaolinite	I/S	illite	chlorite	quartz	K-spars	plagio.	micas	others
Misedor (183 m)									
<0.4 μm	30	30	25	15	0	0	0	0	0
2–6 μm	nd	nd	nd	nd	81	0	1	14	4
6–10 μm	nd	nd	nd	nd	85	1	1	10	3
10–20 μm	nd	nd	nd	nd	93	0	0	5	2
20–40 μm	nd	nd	nd	nd	96	0	0	3	1
40–63 μm	nd	nd	nd	nd	97	0	0	3	0
HD1 (1973 m)									
<0.4 μm	35	45	20	0	0	0	0	0	0
2–6 μm	nd	nd	nd	nd	58	0	2	36	4
10–20 μm	nd	nd	nd	nd	65	1	2	28	4
20–40 μm	nd	nd	nd	nd	67	1	3	28	1
40–63 μm	nd	nd	nd	nd	56	1	2	35	6
HD1 (2605 m)									
<0.4 μm	30	50	20	0	0	0	0	0	0
2–6 μm	nd	nd	nd	nd	50	2	3	44	1
6–10 μm	nd	nd	nd	nd	66	0	2	32	0
10–20 μm	nd	nd	nd	nd	65	1	2	32	0
20–40 μm	nd	nd	nd	nd	66	0	2	32	0
40–63 μm	nd	nd	nd	nd	59	0	2	37	2
TM29 (4232 m)									
<0.4 μm	40	40	20	0	0	0	0	0	0
2–6 μm	nd	nd	nd	nd	25	0	5	60	0
6–10 μm	nd	nd	nd	nd	37	0	4	51	8
10–20 μm	nd	nd	nd	nd	44	1	5	42	8
20–40 μm	nd	nd	nd	nd	65	0	4	26	5
40–63 μm	nd	nd	nd	nd	63	0	6	27	4

The amounts are in percent for each category; I/S stands for illite/smectite mixed layers, K-spars for K-feldspars, plagio. for plagioclases, nd for not determined; among the other framework minerals are pyrite and siderite.

m. A small additional decrease occurred between 2600 and 4230 m (Table 3, Figure 4). The radiogenic ^{40}Ar contents for the fraction <0.4 μm between 2000 and 4230 m follow a depth trend similar to that of the coarse fraction, but the magnitude of decrease for the <0.4 μm fraction was about twice that of the >0.4 μm fraction.

The variation of K-Ar dates for the >0.4 μm fraction follows two trends depending upon depth. For the upper 2000 m interval, the average K-Ar date increased from 90 Ma at 350 m to 107 Ma at 2000 m. It progressively decreased from 107 Ma at 2000 m to 58 Ma at 4230 m for the lower interval. The K-Ar dates for the <0.4 μm fraction decreased progressively from 80 Ma at 350 m to 27 Ma at 2600, and from 27 Ma at 2600 m to 22 Ma at 4230 m (Table 3, Figure 4). The youngest date (21 Ma) recorded for the <0.4 μm fraction of the sandstone samples at the deepest part of the stratigraphic interval remained higher than the Miocene sedimentation age (which was approximately 13–14 Ma at 4230 m depth). A conspicuous feature for the K-Ar date-depth trend for the <0.4 μm fraction between 350 and 2000 m was that the rapid decrease in the values was accompanied by a rapid increase for the K content and the nearly invariant radiogenic ^{40}Ar content.

SHALES. The K contents of the >0.4 μm fraction increased progressively with depth. The K values at a depth of 4230 m was almost twice those at 180 m. By contrast, the profile of the variation of K content with depth for the <0.4 μm fraction can be divided into an increase between 180 and 2000 m and then a nearly constant content between 2000 and 4230 m (Table 4, Figure 5). The rate of increase of K content with depth for the <0.4 μm fraction for the upper 2000 m was about the same as that for the >0.4 μm fraction in the same stratigraphic interval. Thus, the profiles of K content variation with depth for the coarse and fine fractions of the shales was distinctly unlike those for corresponding size fractions of the sandstones.

The trend in the variation of radiogenic ^{40}Ar content with depth for the >0.4 μm fraction of the shales was marked by increases in the top 2000 m interval and near constant K contents at depths between 2000 and 4230 m. The <0.4 μm fractions have a profile that was characterized by a decrease in the radiogenic ^{40}Ar content for the top 2000 m and near constant radio-

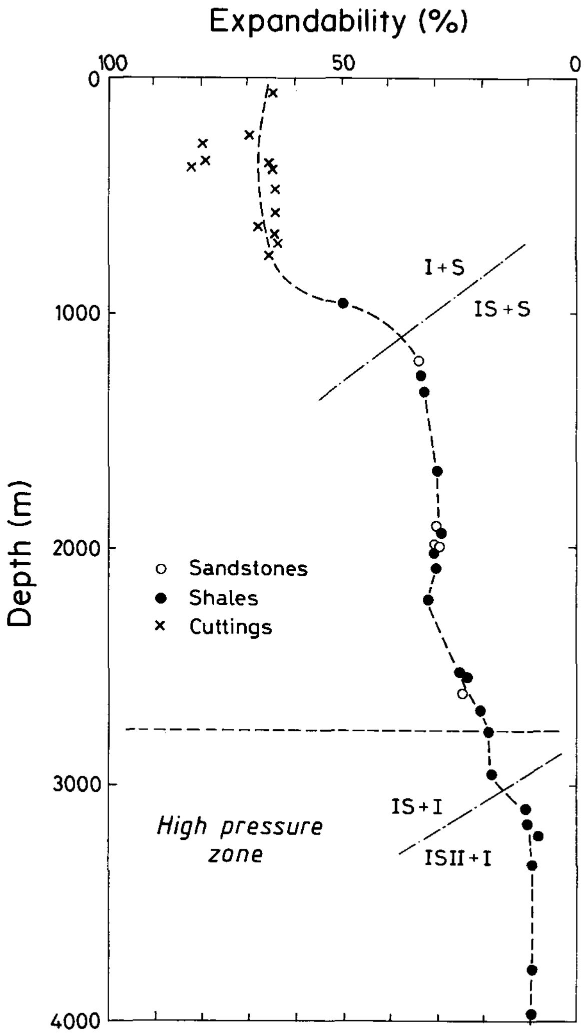

Figure 2. Percent of expandable layers in the illite-smectite mixed-layer minerals relative to the depth of both sandstone and shale samples in the Handil oil-field.

genic ^{40}Ar content for the 2000-to-4230 m interval (Table 4, Figure 5). The variation of the K-Ar dates with depth for the >0.4 μm fraction decreased with increasing depth. The rate of decrease for the upper 2600 m of the interval appears to be higher than that for the lower 1600 m interval (Table 4, Figure 5). The dates for the >0.4 μm fraction varied widely between 180 and 2600 m, ranging from 94 to 127 Ma at 180 m, and from 61 to 98 Ma at 2600 m. The decreasing trend of K-Ar dates with increasing depth continues beyond the 2600 m depth. The spread of the dates for the >0.4 μm fractions was diminished to between 61 and 69 Ma at 4230 m.

The trend of the K-Ar date-depth relationship for the <0.4 μm fraction of the shales was similar to that of the >0.4 μm fraction. The absolute value of the date was smaller for the <0.4 μm fraction (Figure 5). The K-Ar date decreased from 80 Ma at 180 m to 54 Ma at 2000 m. Any further depth increase seems to have a negligible effect on the values. The K-Ar date of the <0.4 μm fraction was 50 Ma at 4230 m. The K-Ar dates decreased within the 180-to-2000 m interval and was accompanied by an increased K content and a decreased radiogenic ^{40}Ar content. Near constant dates between 2000 and 4230 m were attendant with nearly invariant K and Ar contents (Figure 5).

DISCUSSION

At the deepest level, the <0.4 μm fractions from the sandstone sample yields K-Ar dates of 22 Ma and the shale sample yields a K-Ar date of 50 Ma. These values are older than the depositional age of 14 Ma. This indicates that the fractions contain detrital K-bearing minerals with varied detrital isotopic memories. The variation trends for the K-Ar dates and not the specific time nor the number of times of sediment diagenesis are pertinent for this study. The aim was to address the behavior of K among the fine (<0.4 μm) clay fractions and the coarse (>0.4 μm) mica- and feldspar-enriched fractions. It was important to relate the burial evolution of both lithofacies relative to surfacial references. Also, it was assumed that sediment deposited during the last 14 Ma was fairly homogeneous among each facies. Hence, the presence of the detrital memory for the analyzed clay samples does not hinder the understanding of the physical and chemical controls for the diagenesis of these sediments.

Sandstones

Between 350 m to 2000 m, changes in the K-Ar dates of the <0.4 μm fraction was marked by a decrease that was attendant with increased illitization inducing an increased K content while the radiogenic ^{40}Ar content remains invariant. The K-Ar date across this stratigraphic interval decreased by about 40% from 80.3 at 350 m to 47.7 Ma at 2000 m, whereas the I/S expandanbility decreased by about 35% (70 to 35%). As the content of radiogenic ^{40}Ar slightly increased during the interval, the I/S XRD data indicated an increase for the illite layers in an amount correlating to measurable K change. Therefore, the decrease of 40% for the K-Ar date is similar to the decrease of 35% for the expandability of the I/S. Unlike the <0.4 μm fraction, the >0.4 μm fraction does not show distinct changes in either the K content or the K-Ar date

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Figure 3. XRD diagrams for the different types of illite-smectite mixed-layer minerals (A) relative to the depth and (B) to the corresponding computed diagrams.

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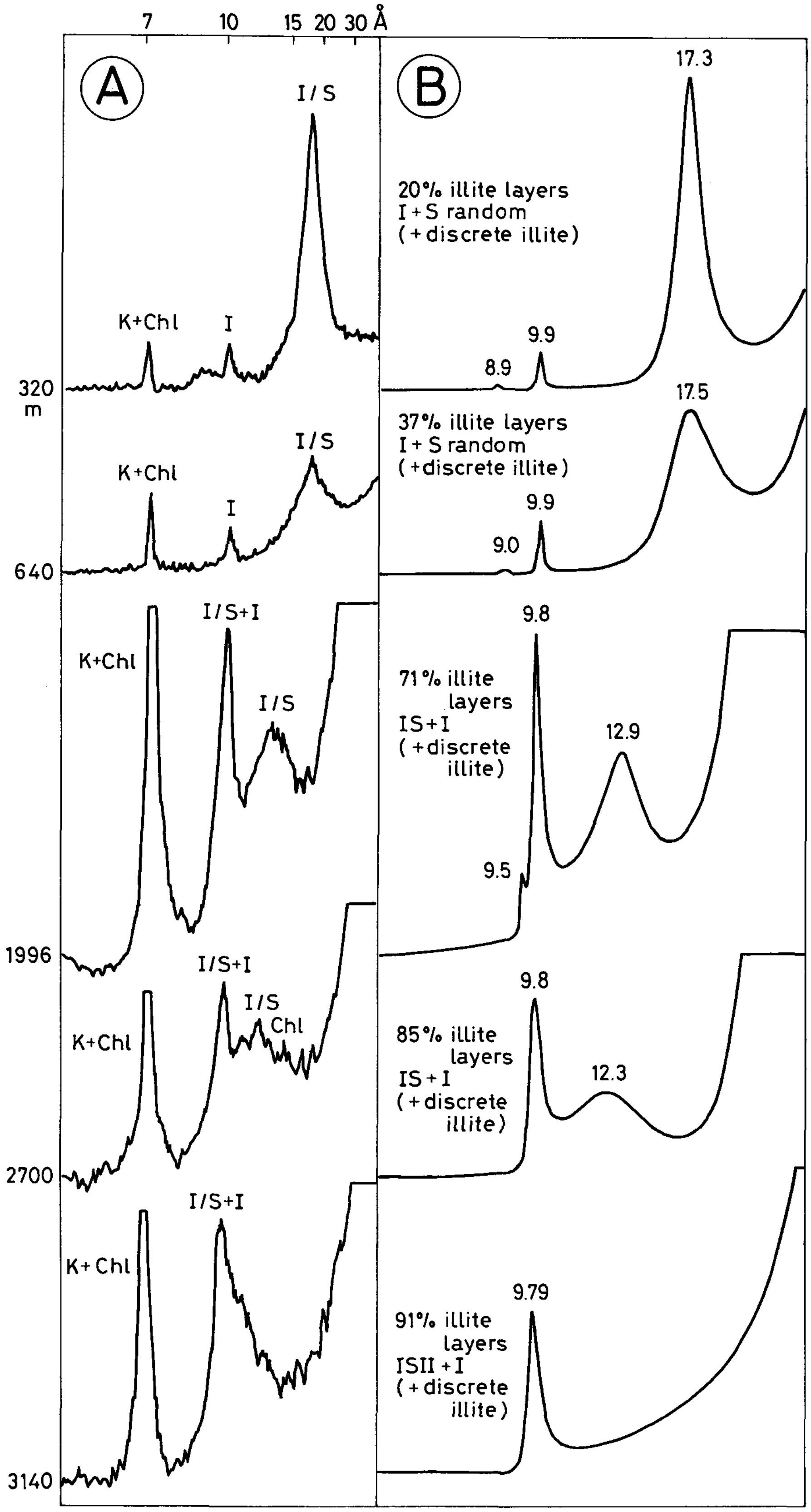

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Table 3. K-Ar isotopic data of different size fractions of Mahakam sandstones from different depths.

Samples	K_2O_m (%)	K_2O_{adj} (%)	rad. ^{40}Ar (%)	rad. ^{40}Ar (10^{-6} cm^3/g)	Age (Ma ± 2σ)
Misedor (350 m)					
<0.4 μm	2.10	4.66	80.10	5.93	80.3 ± 5.0
2–40 μm	0.91	nd	40.99	2.76	91.6 ± 4.7
40–63 μm	0.88	nd	47.64	2.65	91.0 ± 4.1
HD1 (1978 m)					
<0.4 μm	3.98	6.63	13.44	6.20	47.7 ± 1.8
2–6 μm	0.98	nd	70.44	3.33	102.4 ± 3.3
10–20 μm	0.87	nd	67.55	2.88	100.1 ± 3.4
20–40 μm	0.75	nd	85.66	2.81	112.7 ± 3.2
40–63 μm	0.70	nd	84.78	2.65	113.7 ± 3.3
HD1 (2595 m)					
<0.4 μm	4.13	7.50	23.90	3.55	26.5 ± 2.2
2–6 μm	0.83	nd	64.50	2.33	85.2 ± 3.0
6–10 μm	0.59	nd	78.02	1.75	89.7 ± 2.8
10–20 μm	0.46	nd	63.82	1.37	90.4 ± 3.3
20–40 μm	0.35	nd	67.90	1.03	89.4 ± 3.1
40–63 μm	0.25	nd	62.73	0.66	80.4 ± 2.9
TM29 (4228 m)					
<0.4 μm	1.92	7.68	28.91	1.35	21.6 ± 2.0
2–6 μm	0.78	nd	88.86	1.12	44.1 ± 1.2
6–10 μm	0.23	nd	82.12	0.51	67.3 ± 2.0
10–20 μm	0.26	nd	89.64	0.53	60.7 ± 1.7
20–40 μm	0.18	nd	47.88	0.29	48.9 ± 2.2
40–63 μm	0.20	nd	38.35	0.46	68.8 ± 3.8

K_2O_m stands for measured content, K_2O_{adj} for content adjusted to the amounts of K-bearing phases in the <0.4 μm fractions, rad. ^{40}Ar for radiogenic ^{40}Ar.

within the 350 to 2000 m interval. The radiogenic ^{40}Ar content of the >0.4 μm fraction also appears to remain almost invariant. Evidence from scanning electron microscope (SEM) observation indicated K-feldspar grain alteration (maximum 5 to 10%). This is supported by the comparison of SEM photos showing K-feldspars buried at 350 m (Figure 6B) and 1980 m (Figure 6C). The amount of K-feldspar in the rocks of the upper section (<2000 m) were approximately 1% as estimated from thin-section petrographic observation.

The expandability for the I/S of the <0.4 μm fraction at 2600 m depth decreased by about 10%, while the K-Ar date decreased by about 45% from 48 Ma at 2000 m to 27 Ma at 2600 m. This K-Ar date significant decrease, in comparison to the small decrease in expandability as observed from XRD data, supports the significant decrease of radiogenic ^{40}Ar loss relative to constant K content. Unlike the <0.4 μm fraction, the >0.4 μm fractions within this interval exhibited a decrease of the K content, which was not induced by significant changes for the mineral compositions. But, like the <0.4 μm fraction, the >0.4 μm fractions also yielded a lower radiogenic ^{40}Ar content and lower K-

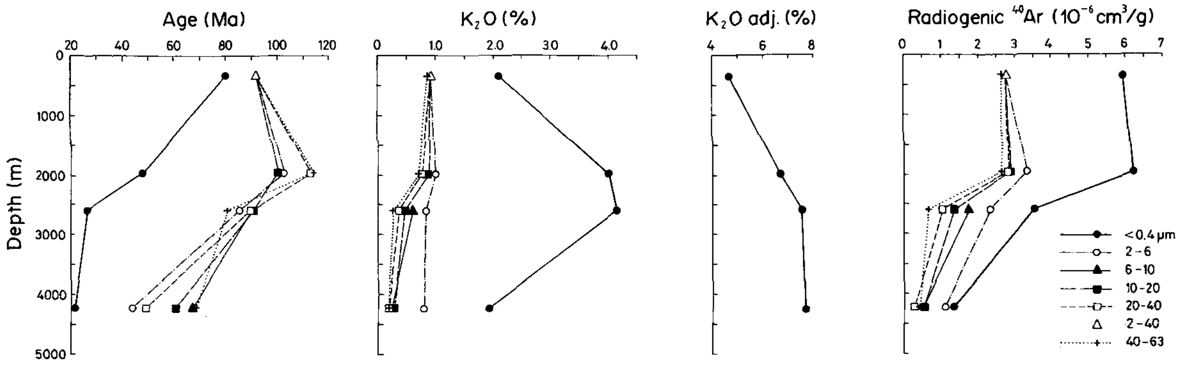

Figure 4. Variations in the K-Ar dates, K_2O contents and radiogenic ^{40}Ar contents of different size fractions of the sandstones collected at different depths in the Mahakam sediments.

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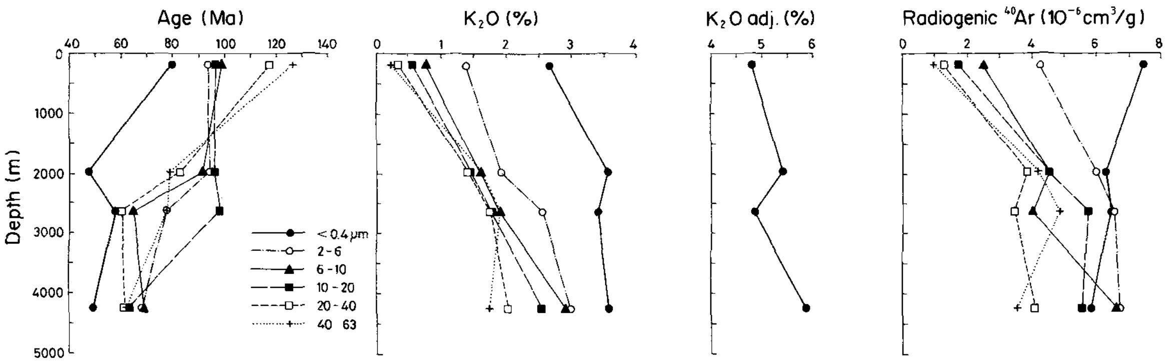

Figure 5. Variations in the K-Ar dates, K_2O contents and radiogenic ^{40}Ar contents of different size fractions of the shales collected at different depths in the Mahakam sediments.

Ar dates while depth increased from 2000 to 2600 m. The evidence from electron- and petrography-microscope observations indicated that the K-feldspars were highly altered at the 2000 to 2600 m interval relative to those for the 350 and 2000 m interval. The relative K-feldspar abundances for the two intervals were about equal. The decreased K-Ar dates with increased depth for the coarse (>0.4 μm) fraction appeared to be related to a significant loss of radiogenic ^{40}Ar with some loss of K as a result of K-feldspar alteration. This K-feldspar grains alteration for the sandstones can be seen by comparing SEM photos showing K-feldspar grains at 1980 m (Figure 6C) and 2600 m (Figure 6D).

When the 2600 m data was compared to the data from the 4230 m depth, there was an additional 10% decrease in the expandability of the I/S for the <0.4 μm fraction at 4230 m, which means an addition of

Table 4. K-Ar isotopic data of different size fractions of Mahakam shales from different depths.

Samples	K_2O_m (%)	K_2O_{adj} (%)	rad. ^{40}Ar (%)	rad. ^{40}Ar (10^{-6} cm^3/g)	Age (Ma ± 2σ)
Misedor (183 m)					
<0.4 μm	2.64	4.80	79.90	7.44	79.9 ± 3.8
2–6 μm	1.37	nd	53.98	4.25	93.8 ± 3.8
6–10 μm	0.77	nd	57.53	2.52	98.6 ± 3.8
10–20 μm	0.54	nd	54.81	1.73	96.7 ± 3.9
20–40 μm	0.33	nd	61.80	1.29	117.6 ± 4.3
40–63 μm	0.23	nd	57.00	0.97	126.5 ± 5.0
HD1 (1973 m)					
<0.4 μm	3.53	5.43	47.37	6.24	54.0 ± 2.6
2–6 μm	1.91	nd	82.37	5.96	94.3 ± 2.6
6–10 μm	1.58	nd	77.75	4.48	92.1 ± 2.7
10–20 μm	1.43	nd	81.85	4.54	96.0 ± 2.8
20–40 μm	1.39	nd	75.64	3.81	83.1 ± 2.5
40–63 μm	1.60	nd	76.84	4.16	78.9 ± 2.5
HD1 (2605 m)					
<0.4 μm	3.39	4.84	36.80	6.41	57.8 ± 3.2
2–6 μm	2.53	nd	78.60	6.49	77.9 ± 2.3
6–10 μm	1.87	nd	90.90	3.99	65.0 ± 2.0
10–20 μm	1.75	nd	85.30	5.69	98.1 ± 2.7
20–40 μm	1.73	nd	84.00	3.43	60.5 ± 2.0
40–63 μm	1.88	nd	68.90	4.83	78.0 ± 2.5
TM29 (4232 m)					
<0.4 μm	3.57	5.91	48.40	5.79	49.7 ± 2.1
2–6 μm	2.98	nd	76.53	6.68	68.2 ± 2.0
6–10 μm	2.93	nd	77.05	6.64	69.0 ± 2.0
10–20 μm	2.54	nd	69.41	5.53	62.8 ± 2.0
20–40 μm	2.02	nd	66.28	4.07	61.5 ± 2.0
40–63 μm	1.74	nd	64.59	3.55	62.2 ± 2.1

K_2O_m stands for measured content, K_2O_{adj} for content adjusted to the amounts of K-bearing phases in the <0.4 μm fractions, rad. ^{40}Ar for radiogenic ^{40}Ar.

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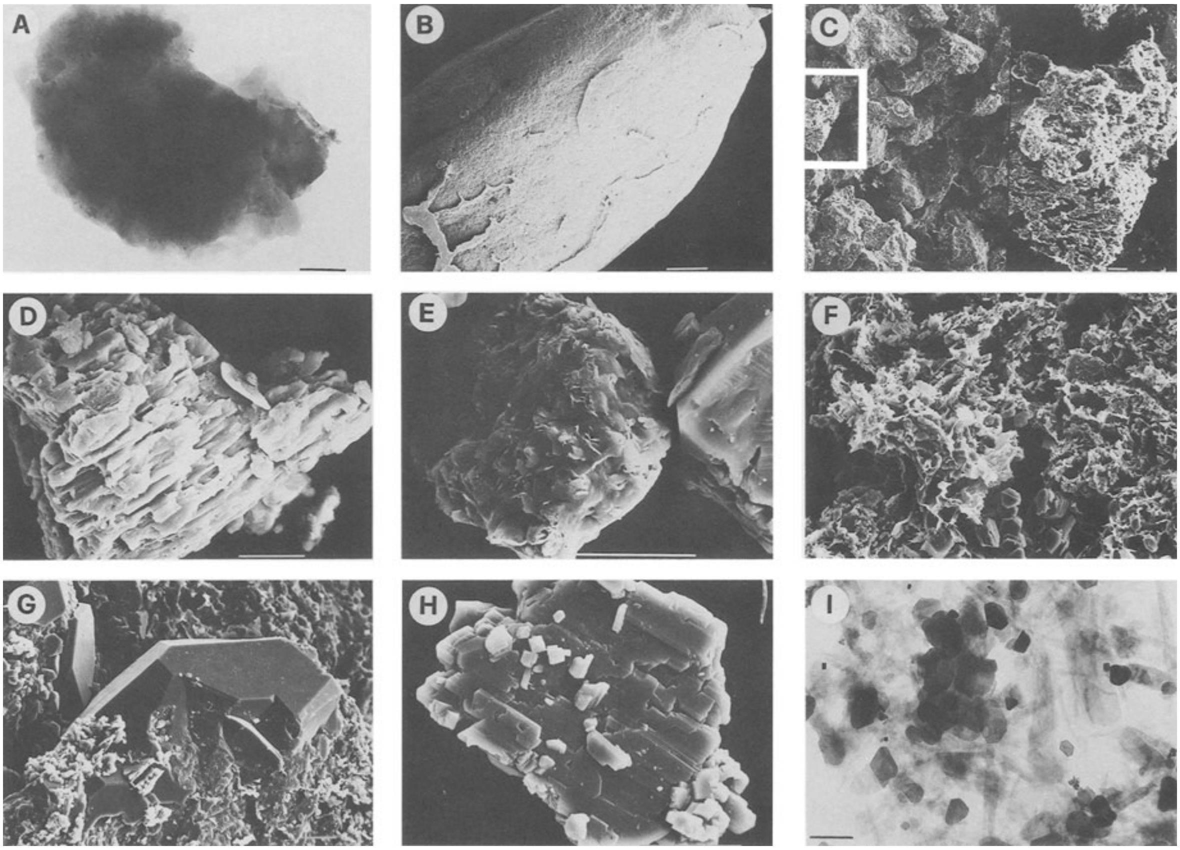

Figure 6. SEM images of detrital K-feldspars and illite-smectite mixed-layer minerals and of authigenic albite and illite of sandstone samples at different depths. The bars represent 10 μm in all cases except for image A (0.4 μm), C (100 μm) and I (1 μm). The letters represent: (A) = detrital illite-smectite particle (350 m); (B) = detrital K-feldspar grain (350 m); (C) = altered K-feldspar grain (1980 m); (D) = altered plagioclase grain; (E) = altered K-feldspar grain with a piece of quartz grain (2600 m); (F) = K-feldspar relics with pristine dickite particles (4230 m); (G) = quartz overgrowths (4230 m); (H) = authigenic albite (4230 m); and (I) = authigenic lath-shaped illite particles with euhedral dickite particles (4230 m).

K. The increase in depth has produced decreased K-Ar dates by about 19% from 27 Ma at 2600 m to 22 Ma at 4230 m. The chemical data obtained from clay analyses shows decreased K (53%) and radiogenic ^{40}Ar (62%) contents. These decreases result not only from dilution of the clay minerals by a slightly increased amount of kaolinite/dickite, but also by abundant quartz, probably authigenic in origin and small in size. These occurences explain the apparently anomalous relationship between the observed illitization of I/S increase and the decreased K content for the <0.4 μm fraction at this depth. The correlation is obvious when the K contents were calculated on the basis of the amount of the K-bearing clays (Figure 4). The decrease of K-Ar dates is dependent on both the K and the ^{40}Ar contents. The K change for this interval was not sufficient to explain the decreased K-Ar dates, so that a small amount of radiogenic ^{40}Ar loss from the particles has to be assumed between 2600 m and 4230 m as a result of burial. The K-Ar dates for the >0.4 μm fractions decreased between 2600 and 4230 m while the radiogenic ^{40}Ar content decreased but the K content remained fairly constant. The microscopic observations show that the K-feldspars were so deeply altered within this interval that only grain relics can be identified. They are systematically associated with euhedral booklets of dickite and quartz overgrowths (Figures 6E and 6F), and euhedral albite crystals were also frequent within the deeply buried sandstones (Figure 6H).

Shales

The profile and magnitude of change for the expandability of I/S with depth of the <0.4 μm fraction for the shale samples were similar to those of the sandstones. Unlike the expandability of the I/S, the K-Ar dates of the <0.4 μm fraction between shale and sandstone rocks, although changing in the same sense with depth, were not equal at equivalent depths. The shale <0.4 μm fraction systematically has an older apparent

K-Ar age than those for the sandstones. The relatively higher K-Ar dates for the <0.4 μm fraction of the shales relative to equally buried sandstones was related to contamination of the analyzed fractions by detrital micaceous or K-feldspar components. But a different alteration process of the I/S might also be considered on the basis of the study by Rinckenbach (1988). He claimed that the downward evolution of I/S for sandstones was a dissolution-recrystallization process, while that of the I/S for shales was a transformation process. During transformation, the reorganization occurred in an almost closed system and the transformed clay particles might have incorporated "detrital" Ar. The $^{40}Ar/^{36}Ar$ ratio was enriched with ^{40}Ar coming from the detrital lattices and unable to escape.

The K-Ar dates for the shales <0.4 μm fraction, changed from 80 Ma at 185 m to 54 Ma at 1975 m. This 32% decrease in K-Ar date was accompanied by about a 35% decrease in I/S expandability. The K content increased markedly due in part to an increase of the K-bearing phases content, while the radiogenic ^{40}Ar content decreased slightly. The overall result suggests that the decreased K-Ar dates were mainly induced by illitization of the I/S. The shales >0.4 μm fraction had K-Ar dates that generally decreased within the 185–1975 m interval, while the K and radiogenic ^{40}Ar contents increased in relation to increased mica content. The direct cause of the decreased K-Ar dates for the >0.4 μm fractions cannot be positively identified, but the K increase was more pronounced than that of the radiogenic ^{40}Ar.

Between the 2000 and 2600 m depth, the K-Ar dates of the <0.4 μm fraction increased by about 7%, due to a small K content decrease and a small radiogenic ^{40}Ar content increase. The increased K-Ar date from 54 Ma at 2000 m to 58 Ma at 2600 m is believed to have been induced by slight changes in the relative amount of the detrital mica components. The amount of mica was especially high for the 2–6 μm size fraction. This occurrence has probably obscured the effects of the transformation process. The K-Ar dates of the <0.4 μm fraction from 2000 to 4230 m interval decreased by about 7% from 54 Ma to 50 Ma. The I/S expandability decreased by about 20%. The decrease was induced by a very small increase in K content and a small decrease in radiogenic ^{40}Ar content. The K-Ar dates for the >0.4 μm fraction of the same interval decreased like the <0.4 μm fraction. The cause cannot be clearly identified because the SEM observations were not very discriminating as they are from the sandy material. Probably, they resulted from the combined effects of radiogenic ^{40}Ar loss and K addition.

Another Perspective for Diagenetic Illitization

The K-Ar dates obtained provide new information about the behavior of K during the burial-induced illitization process of smectite. Those for the clay fraction of the sandstone samples indicate that the rate of K change with depth for the upper stratigraphic interval, between 350 and 2000 m, is higher than that for the lower stratigraphic interval between 2600 and 4230 m. These two intervals also differ with respect to the degree of change for the K-Ar dates. The cause or causes for the two trends should give an account of how K supplied the site where most conversion of smectite into illite took place. In order to add further information, a mass balance calculation have been developed to account for the K behavior (Furlan 1994).

The microscopic observations of the sandstones show that the lower interval of the sequence (>2600 m) has a high amount of illite layers in the I/S (70 to 90% illite layers). This interval corresponds to the zone in which the K-feldspar grains are highly altered and where a large amount of kaolinite/dickite minerals crystallized. In the upper interval (<2000 m), the first 1000 m have a low percentage of illite layers (35%). The main illitization zone is located at approximately 1000 m where the illite layers percentage increase from 35 to 70%. This upper interval (<2000 m) corresponds to the zone in which the K-feldspar crystals were the least altered. These correspondances do not indicate that the amount of K was derived locally from the alteration of K-feldspar for the conversion of smectite or I/S to illite. Petrographic estimates have set a value of about 1% for the amount of K-feldspar present in the sandstone whole rocks within the upper 350 to 2000 m zone of illitization. Five to 10% of the feldspar volume has been altered. For the same zone, the amount of I/S was estimated to be about 5% and the amount of change in the I/S expandablity was shown to be as much as 35%. The calculated amount of K available from the K-feldspar alteration is at least six times less than the amount of K needed for the illitization of smectite within the upper illitization zone (<2000 m). This suggests that an additional supply of K had to come from outside this illitization interval. A potential source of this additional amount of K needed for the illitization of smectite for the upper 300–2000 m intervals came from the lower section (>4000 m). This stratigraphic zone was marked by dickite and silica precipitation as quartz overgrowths. The same calculation for the released K for the lower section indicated that about 10 times more K was available than needed to complete illitization. Even if less reliable, because of difficulties in evaluating the relative amounts of K released by altered K-feldspar grains and altered mica flakes, a similar calculation on K availability for shales has been made. It showed the same problem of the lack of K in the illitization zone. Therefore, such a model of illitization requires that fluids carrying the needed K for the illitization necessarily moved upward across the stratigraphic interval (faults) or along the anticlinal slopes. Such movements were suggested by Weaver and Beck (1971), and Awwiller (1993) for the illitization process of the sediments for the Gulf coast area.

CONCLUSION

Mineralogical, morphological and K-Ar isotopic data on authigenic clay and primary detrital K-feldspar minerals gave further insights into clay diagenesis for progressively buried Tertiary sediments of the Mahakam Delta Basin. The data suggested that part of the K needed for illitization of the illite/smectite clays in sandstones was supplied by upward migrating fluids. They altered K-feldspar minerals at greater depths and left behind most of the Si and Al to form dickite and quartz overgrowth. This model differs from the Gulf Coast model that portrays illitization as a result of the local supply in K from alteration of K-feldspars associated with the clay minerals. The current model suggests that illitization occured within a chemically open system and was consolidated by a mass-balance calculation. However, the clay minerals of shales tend to give consistently older K-Ar dates relative to those of the sandstones. The reasons are still unclear, but can be related to the closed-system of these rocks that can induce illitization by "transformation."

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