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INTER-BASINAL COMPARISON OF THE DIAGENETIC EVOLUTION OF ILLITE/SMECTITE MINERALS IN BURIED SHALES ON THE BASIS OF K-Ar SYSTEMATICS

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Abstract—The K-Ar systematics of illite/smectite (I/S) mixed layers in deeply buried shales from the Gulf Coast, the North Sea and the Mahakam Delta basins have been compared to provide additional perspectives on the diagenetic evolution of these minerals. Comparison of the results suggests that illitization proceeds similarly in the 3 basins, at least for the increase in the illite-layer and K contents, despite differences in the provenance of the detrital components, the ages of deposition, the depths of burial and the tectonic history of the basins. Analysis of the trends with depth in the illite-layer and K contents of I/S-enriched size fractions of shales in the North Sea and the Mahakam Delta basins shows that these trends represent segments of the more complete trends from I/S minerals of the Gulf Coast area.

The trends with depth in the radiogenic ^{40}Ar contents and in the K-Ar ages of the I/S-rich fractions in the North Sea and Mahakam Delta basins suggest that, relative to the reference trends of the Gulf Coast area, the K-Ar system of the clay material is more dependent on the behavior of the radiogenic ^{40}Ar than on the occurrence or non-occurrence of detrital grains in the size fractions. Recasting of the available data suggests that retention of radiogenic ^{40}Ar by the illite-type minerals occurs in the intense illitization zone and release occurs in the deeper part of the basins. We therefore speculate that the illitization process of the I/S mixed layers of progressively buried shale-type sediments could be controlled by a transformation process integrating dissolution of detrital components in poral rock environments relatively impermeable to radiogenic ^{40}Ar. These excesses, which might be partly or completely erased in deeper parts of the sedimentary basins, question the application of the K-Ar dating method on clay minerals extracted from shales.

Key Words—Burial Diagenesis, Illite/Smectite Mixed Layers, Illitization, K-Ar Data, Sedimentary Basins, Shales.

INTRODUCTION

The most visible diagenetic change in many deeply buried argillaceous sediments is the increase in the amount of illite layers in the I/S mixed layers as a function of increase in burial depth. The trend of this depth-related mineralogic change is a rotated S-shaped curve depicting: 1) a segment at relatively shallow burial with almost no increase in the illite content as a function of depth; 2) a segment at intermediate burial with a rapid increase in the illite content of the mixed layers to a high amount; and 3) a deep-buried segment in which the high amount of illite layers in the I/S remains almost constant. The completed profile for this mineralogic change is well-illustrated by a number of studies on clays in Tertiary sedimentary rocks in the Gulf Coast region (Burst 1969; Perry and Hower 1970; Weaver and Wampler 1970; Weaver and Beck 1971; Perry 1974; Aronson and Hower 1976; Hower et al. 1976). This trend in the diagenetic transformation of smectite to illite has also been documented in other sedimentary basins (Jennings and Thompson 1986; Rinckenbach 1988; Burley and Flisch 1989; Glasmann, Larter et al. 1989; Glasmann, Lundegard et al. 1989). Time, temperature, chemical composition of the diagenetic fluids, availability of K, crystallography and chemistry of the parent minerals and fluid dynamics in the diagenetic zone were suggested as various important factors that can influence the diagenetic evolution. Opinions are still varied about the relative importance of these factors, leading to varied views about the mechanism of the change from smectite to illite in the Gulf Coast area, on the basis of either mineralogical and geochemical studies (Boles and Franks 1979; Freed 1981; Ahn and Peacor 1985, 1986, 1987; Pollastro 1985, 1993, 1994; Fisher and Land 1986; Land et al. 1987; Freed and Peacor 1989, 1992; Shaw and Primmer 1989; Primmer and Shaw 1991; Milliken 1992; Awwiller 1993), or isotopic studies (Yeh and Savin 1977; Yeh 1980; Land 1984; Morton 1985; Land and Fisher 1987; Ohr et al. 1991; Eberl 1993; Clauer et al. 1995). Consequently, the various studies of Gulf Coast sediments have significantly influenced thoughts about burial-induced illitization of smectite in sediments in other basins.

Isotopic data, especially K-Ar data, on clay minerals have proved extremely useful in the understanding of the historical aspects of the diagenetic change from smectite to illite in I/S (Clauer and Chaudhuri 1995). Studies of the Tertiary sedimentary sequence in the Gulf Coast basin have provided the most information about isotopic behaviors attending burial diagenesis.

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Much of this information is founded on the assumption of a simple geologic framework; hence, the derived information about clay diagenesis is not necessarily applicable to clay diagenesis in other sedimentary basins. The degree to which the Gulf Coast model of diagenetic clay formation applies to smectite illitization in other sedimentary basins where mineralogical profiles of diagenetic evolution of illite from smectite are different, such as the North Sea and the Mahakam basins, is critical to the understanding of the genesis of illite layers in I/S.

In this paper, we have compared the trends of the K-Ar data for I/S-rich size fractions in deeply buried shale sediments of the North Sea and the Mahakam Delta basins, for which only limited isotopic data are available, with those of similar size fractions in the Gulf Coast basin. The purpose is to point out the differences and similarities in the K-Ar isotopic trends of diagenetic illite-type clays from different sedimentary basins that belong to a similar rock lithology. This comparison was made even though: 1) the sediments in the different basins differ in present-day depths of burial, origin of detrital supply, paleo- and present-day temperatures and stratigraphic ages; and 2) the analyzed clay fractions may not be pure I/S, but may be variably "contaminated" by clastic components with their own isotopic signatures, which may lead to variations in the results due to heterogeneities in the analyzed material.

REVIEW OF THE PUBLISHED DATA

The mineralogical and K-Ar isotopic data for clay fractions of the early-to-middle Tertiary sediments in the Gulf Coast basin are from Hower et al. (1976) and Aronson and Hower (1976), whereas those of equivalent size fractions of the Jurassic sediments in the North Sea basin and of the late Tertiary sediments in the Mahakam Delta basin are from Glasmann, Larter et al. (1989) and Rinckenbach (1988), respectively. The illitization of I/S in shale-type sediments of the 3 basins occurred at different times and also apparently at different depths. As the analyzed Gulf Coast clay fractions were recovered from drill cuttings that integrated 100-m stratigraphical intervals and as the clay fractions from the other 2 basins were extracted from individual drill-core samples, we have recast the K-Ar and mineralogic data for the clay minerals from North Sea and Mahakam Delta basins in terms of 100-m stratigraphic intervals beginning at a 1 km depth to portray all the trends similarly, as functions of depth. We have also decided to keep the original units used by the authors for the K or K_2O and radiogenic ^{40}Ar contents.

The Gulf Coast Area

Figures 1a and 2a illustrate the trends of change in the amounts of illite layers, K_2O and radiogenic ^{40}Ar contents and K-Ar dates of the <0.1 μm I/S size fractions across a stratigraphic interval extending from 1.1 to 5.5 km depth in the Oligocene sedimentary sequence. The changes in the contents of both the illite layers and K_2O, which fairly follow each other with some variations in the upper 1-km interval, are characterized by rapid increases between 2.5 and 4 km. Below 4 km depth, both the illite-layer and the K_2O contents of the I/S remain at high values, again with some random fluctuations in narrow limits. Like the K_2O contents, the radiogenic ^{40}Ar contents remain constant for the top 1-km interval and then increase rapidly between 3 and 4 km depths. Below 4 km, the ^{40}Ar contents decrease progressively, unlike the K_2O contents which randomly fluctuate.

The K-Ar dates increase from 54 Ma for the <0.1 μm fraction at a depth of 1.2 km to 58 Ma for that at 2.5 km and then sharply decrease from 58 Ma to 46 Ma below 2.5 km, across a 0.6 km burial increase. The 46 Ma date remains essentially unchanged for an additional burial of about 0.6 km. But below 3.5 km, another sharp decrease occurs from 46 Ma to 34 Ma by an increase in depth of only about 0.1 km. The further trend in the K-Ar dates, as a function of depth from 3.6 km to 5.5 km, corresponds to a profile characterized by a slight decrease with "zig-zag" variations between 35 and 30 Ma. The zig-zag trend in the K-Ar dates cannot be strictly related to variations in radiogenic ^{40}Ar, which decrease progressively, but the effect may be attributable to the variations in the K_2O contents. The zig-zag pattern in the K-Ar dates and the K_2O variations across the bottom 1.5-km interval can be explained in terms of combined effects of loss of radiogenic ^{40}Ar, presence of varied but minute amounts of some detrital minerals in the <0.1 μm fractions that cause the scatter, and dilution of the clay minerals by non-K-bearing minerals such as kaolinite and quartz, even if not detected by X-ray diffraction.

The Bergen High Area in the North Sea Basin

The Bergen High area is located on the eastern flank of the North Viking graben in the Norwegian part of the North Sea basin. Major oil-producing fields, such as the Huldra and Veslefrikk fields with productions of hydrocarbons from Jurassic rocks, occupy part of a major fault-block structure in this area. Progressive changes from randomly ordered smectite-rich to ordered illite-rich clays for the I/S have been found in these fields (Glasmann, Larter et al. 1989). The orderings of the I/S in the Huldra field appear to have occurred over a deeper zone than in the Veslefrikk field. Fluid inclusion data for quartz overgrowths have shown that temperatures during the growth of these secondary minerals were about 180 °C in the Huldra field, as compared to the present geothermal temperature of 150 °C at 4 km depth, and about 270 °C in the Veslefrikk field, as compared to the present geo-

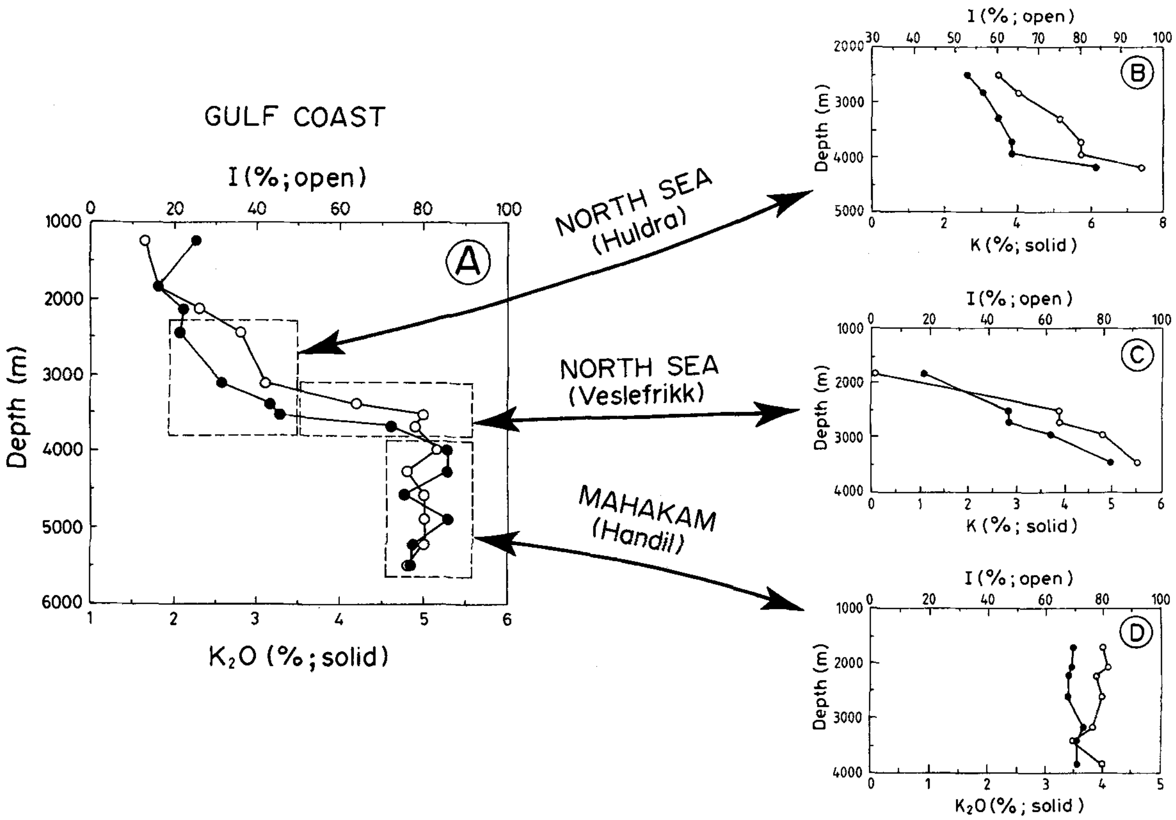

Figure 1. Relationships between depth and the amounts of illite layers and K of the <0.1 μm illite/smectite mixed layers from progressively buried shale-type sediments in the Gulf Coast Basin (A, after Aronson and Hower 1976), and the Huldra and Veslefrikk fields of the North Sea Basin (B and C, after Glasmann, Larter et al. 1989), and of the <0.4 μm illite/smectite mixed layers from equivalent sediments in the Mahakam Delta Basin (D, after Rinckenbach 1988).

thermal temperature of 170 °C at the same depth of 4 km.

HULDRA FIELD. The available K-Ar isotopic data for <0.1 μm I/S-enriched fractions in the Huldra field cover a limited stratigraphic interval of about 1.5 km between 2.5 and 4 km depths. Across this stratigraphic interval, the I/S changes progressively from 60% illite layers in the fractions at 2.5 km depth to 80% illite layers at 3.8 km, following which the amount increases very rapidly to 95% illite layers at a depth of 4 km (Figure 1b). The increase of K contents as a function of depth closely follows that of the illite content in the I/S, suggesting that the clays were not significantly diluted by any other mineral. The trend in the change of the radiogenic ^{40}Ar contents follows closely that of K contents across much of the stratigraphic interval, which supports the lack of K-bearing detrital components. Like the K contents, the radiogenic ^{40}Ar contents also increase very sharply in the bottom 0.2 km of the interval (Figure 2b).

While both the K and the radiogenic ^{40}Ar contents essentially increase with depth at a nearly uniform rate, at least for the top 1.3-km interval of the section, the K-Ar dates remain more or less constant between 75 and 80 Ma (Figure 2b). The whole trend suggests a slight increase in the K-Ar dates, due mainly to the deepest sample, the K-Ar date of which increasing noticeably to 84 Ma. In this size fraction, both the K and radiogenic ^{40}Ar contents increase downwards, but differently as the K-Ar date increases.

VESLEFRIKK FIELD. In the <0.1 μm size fractions, the amount of illite-layer contents in the I/S increases rapidly from 20% at a 1.8 km depth to slightly more than 90% at 3.5 km, although the illite-layer contents remain constant at 65% in 2 adjacent samples at 2.5 to 2.7 km (Figure 1c). As in Huldra field, the trend of increase in the K contents of the clay fractions in the Veslefrikk field closely follows that of increase in the illite-layer contents of the I/S, suggesting no major dilution effect on the clay samples by the presence of non-K-bearing minerals. The radiogenic ^{40}Ar contents also increase as a function of depth increase, essentially between 2.7 and 3.5 km. Thus, both the K and radiogenic ^{40}Ar contents increase across the entire stratigraphic interval between 1.7 and 3.5 km.

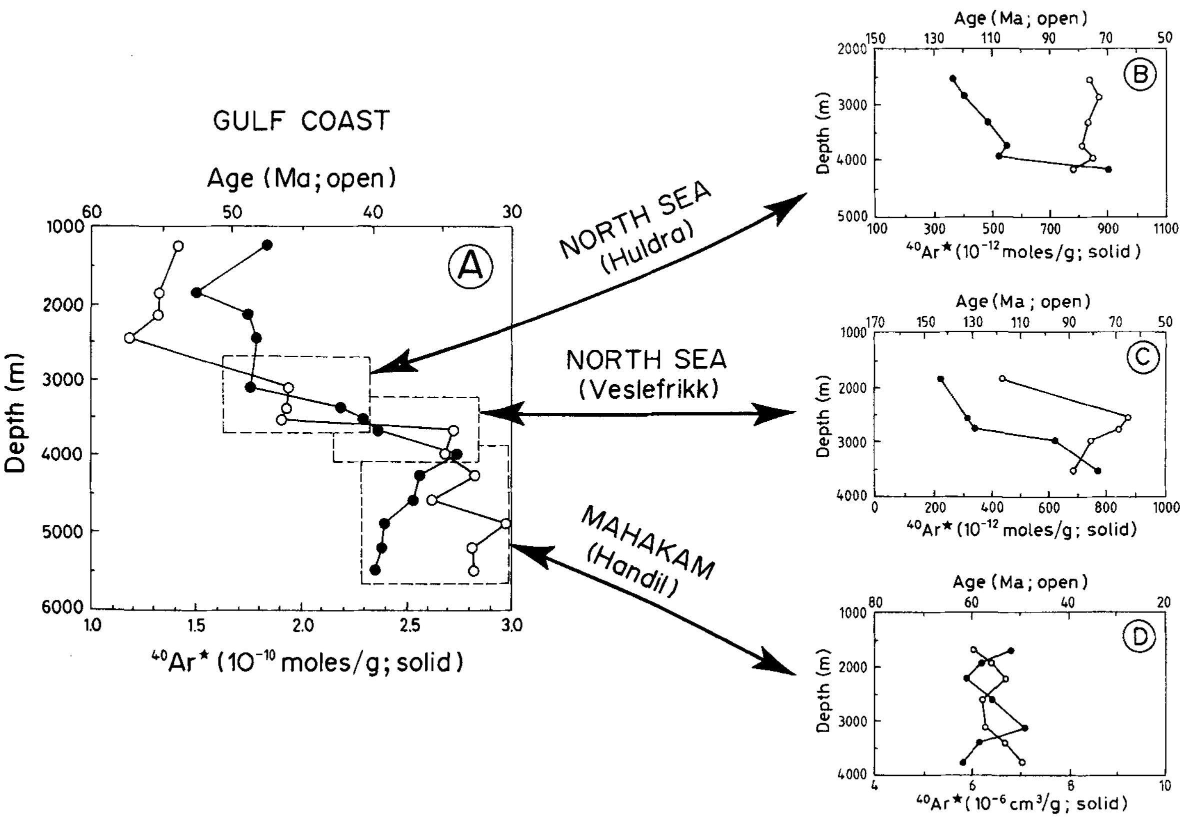

Figure 2. Relationships between depth, the amounts of radiogenic ^{40}Ar and the K-Ar apparent ages of the <0.1 μm illite/smectite mixed layers from progressively buried shale-type sediments in the Gulf Coast Basin (A, after Aronson and Hower 1976), and the Huldra and Veslefrikk fields of the North Sea Basin (B and C, after Glasmann, Larter et al. 1989), and of the <0.4 μm illite/smectite mixed layers from equivalent sediments in the Mahakam Delta Basin (D, after Rinckenbach 1988).

By contrast, the K-Ar dates decrease sharply from 118 Ma to 65 Ma within the interval between 1.7 and 2.5 km, but this trend in the age turns to a strong increase from 65 Ma at 2.5 km to 90 Ma at 3.5 km (Figure 2c). This increasing trend in the K-Ar dates could be related to increasing minute amounts of detrital K-bearing minerals with depth in the size fractions dated.

The Mahakam Delta Basin

The clay fractions examined for the Mahakam Delta basin belong to the late-Tertiary sequence in the Handil field, located on the eastern side of Borneo Island (Indonesia). Unlike the Gulf Coast and the North Sea basins, the illitization of I/S in the Mahakam Delta basin appears to have occurred at a shallow depth with a burial of possibly less than 2 km, allowing for the erosional removal of 0.6 to 0.8 km of sediments, as suggested by Letouzey et al. (1990). Fluid inclusion data from analyses of quartz overgrowths indicate temperatures of about 100 °C at 1 km depth and 160–170 °C at 4 km, in contrast with a relatively low present-day temperature of about 120 °C at 4 km (Rinckenbach 1988). Relatively higher paleotemperatures occurring during most of the illitization process were also found in the North Sea basin.

The <0.4 μm clay fraction of the entire stratigraphic interval between 1.5 and 4 km depths consists of a constant mixing of kaolinite-dickite and I/S. The illite content of the <0.4 μm I/S generally remains unchanged between 80 and 85%, except for a zone at about 3.5 km where the illite content is only 70%. The K_2O contents remain nearly constant across the stratigraphic interval between 1.5 km and 4 km depths, although a small increase appears to have occurred at about 3 km depth (Figure 1d). While the K_2O contents are nearly invariant, the radiogenic ^{40}Ar contents change in a zigzag manner, characterized by a decreasing trend between 1.7 and 2.2 km, followed by a progressive increase in the value across the interval between 2.2 and 3.2 km, and finally by a decreasing trend at the bottom interval between 3.2 and 4 km (Figure 2d).

The K-Ar dates obtained for the clay fractions of the shale-type sediments are in excess of the late Tertiary age of deposition, clearly indicating that detrital phases are present in the size fractions analyzed. The overall trend for the K-Ar dates increases with depth with some zig-zagging variations probably induced by the occurrence of minute amounts of detritals (Figure 2d). However, the K-Ar trend is also related to the trend of change in the radiogenic ^{40}Ar contents across the entire interval: the K-Ar dates are very dependent on the behavior of the radiogenic ^{40}Ar, which could mean in turn that diffusion of radiogenic ^{40}Ar may have occurred in this case.

DISCUSSION

The mineralogic changes in the I/S are characterized by increases in the illite-layer content versus depth, which follows the increases in the K content. Figure 1 broadly illustrates similar changes in these contents in the Gulf Coast and North Sea basins; they can be described by rotated S-shaped curves. This shape also exists for the I/S in the Mahakam Delta basin, but at shallow depths of less than 1 km (Rinckenbach 1988). The curves differ in changes in the slope with respect to both the depth and the amount of illite layers in the I/S. The comparison between the trends for change in both the amounts of illite layers and K contents between the I/S highlights that, for equivalent depth intervals, the I/S from Huldra field in the North Sea basin are less illitized than the I/S from Veslefrikk field and that they are much less illitized than those of Handil field in the Mahakam Delta basin. Also, in contrast, the I/S of Huldra and Handil fields are more evolved at similar depths than those of the Gulf Coast basin, while those of Veslefrikk field are less evolved (Figure 1).

In fact, the depth-related trends of the illite-layer, the K and the radiogenic ^{40}Ar contents and the K-Ar dates in the I/S size fractions of shales from North Sea and Mahakam Delta basins correspond well to different segments of the more complete trends of the I/S size fractions from shales of the Gulf Coast basin (Figures 1 and 2). This overall agreement suggests that the illitization process proceeds similarly in the shale-type rocks of the 3 basins, despite significant differences among them. That of the North Sea is characterized by basement adjustments with faulted blocks and by old dominant Caledonian detrital supply, that of the Mahakam Delta is subsiding in a tectonically active compressive region and is supplied by early Tertiary detrital silicic sediments and that of the Gulf Coast is broadly subsiding and filled by sediments containing mostly pedogenic-type smectite material. The fact that the K-Ar characteristics of the I/S of shale-type rocks from different basins are very similar implies that the K-Ar systematics of these minerals behave consistently during the illitization process, although the results for an individual basin may not reflect the entire evolutionary trend seen in the Gulf Coast basin.

In failing to recognize that the records of individual basins are incomplete, many have interpreted the data differently. For instance, the interval of intense illitization where the illite-layer and K contents increase rapidly should theoretically be characterized by decreases of the K-Ar dates, because of a combined increase of K and an expected decrease of radiogenic ^{40}Ar due to structural reorganization. In fact, the K-Ar dates never decrease at this interval: they either remain constant, as in the Gulf Coast basin and in Huldra field, or they increase, as in Veslefrikk field. These unexpected trends obviously occur without apparent significant changes in the amounts of detrital components in the size fractions. What should be considered is that the trends in the K-Ar dates depend to a large degree on the behavior of the radiogenic ^{40}Ar in the evolution of the minerals. As the contents of the radiogenic ^{40}Ar systematically increase in the interval of intense illitization, we are inclined to believe that an increasing amount of Ar might be either retained or even incorporated in the mineral structures during illitization. It should be considered that the poral environment of the rocks in which the clay particles were illitized was partly or completely impermeable to Ar release. The K-Ar dates are seen to be randomly scattered in the deeper parts of the Gulf Coast and Mahakam Delta basins, but they show a tendency to decrease with depth. In these zones, the K contents remain narrowly constant, but the scatter suggests that the amounts of minute quantities of detrital grains may change. Such occurrences explain the scatter, but they do not explain the overall decrease in the K-Ar dates. It might be that either a delayed release when retained, or diffusion when incorporated, of radiogenic ^{40}Ar occurs at greater depths. This does not automatically mean that mineralogic changes had to occur to the clay particles in these deeper parts to allow Ar diffusion, as Ar release may have been facilitated by physical modifications of the rocks. However, it cannot be excluded that the illite-type clays were affected further in the deeper parts of the basins with related Ar diffusion.

A common explanation for the behavior of the K-Ar systematics of clay size fractions in progressively buried shale rocks is the variable dilution effect by non-K-bearing minerals where K content decreases while the illite-layer content appears to remain constant or increases. This dilution effect should not basically affect the K-Ar dates, as illustrated by some samples of the Gulf Coast basin, but it does influence those of the Gulf Coast fractions buried at 2.5, 4.5 and 5.2 km. Where an increase in K content occurs while the illite-layer content remains constant, it may be related to the presence of some detrital K-bearing minerals. The presence of these minerals in the I/S fractions may induce a more significant increase in radiogenic ^{40}Ar contents than in K contents and, consequently, an increase in the K-Ar dates, because the increase is expected to depend on the age and the K content of the detrital K-bearing minerals. But this is not the case for some samples for which the K-Ar dates decrease instead: at 3.8, 4.3 and 5.0 km depth for the Gulf Coast samples, and at 3.1 km depth for the Mahakam samples. Part of the trend with constant K content accompanied by decreases in both the radiogenic ^{40}Ar content and the K-Ar date should be ascribed to a loss of radiogenic ^{40}Ar due to diffusion while the K amount remained unchanged. This is illustrated by few samples from the Gulf Coast basin at 4 and 4.2 km depth and from the Mahakam Delta basin at 1.8, 2.1, 2.3, 3.4 and 3.8 km depth. This is the only interpretation that we are able to provide, as it is difficult to consider that changes in the amount of K-bearing detrital components do not affect the K and radiogenic ^{40}Ar contents in a way that increases the K-Ar dates of I/S.

An additional interesting feature in the evolutionary trends involving rapid increase in illite-layer content is a trend of unchanged K-Ar dates, while both K and radiogenic ^{40}Ar contents increase. This is evident from data in the Gulf Coast basin and Huldra field in the North Sea basin. Eberl (1993) considered this as an example of Ostwald ripening. Alternatively, a possible explanation of these constant K-Ar dates with increases in both K and radiogenic ^{40}Ar is that the trend may be related to a binary mixing of nearly coeval detrital I/S clays that differ in their K and ^{40}Ar contents. The essentially unmodified detrital signatures of the clay components, as recorded in the 0.5-km thick stratigraphic interval within the 1.5-km thick zone of conversion of smectite to illite in the Gulf Coast basin and in the 1.5-km thick stratigraphic interval in Huldra field, could then be viewed as limited illitization-related fluid movements through these stratigraphic intervals because of less fluid movements or less K activity.

CONCLUSIONS

Interesting similarities exist among the K-Ar systematics of clay fractions extracted from progressively buried shale-type sediments in the sedimentary basins of the Gulf Coast area, the North Sea and the Mahakam Delta. An important feature in the foregoing discussions about the various trends in the K-Ar dates of illite/smectite size fractions from shales is that diagenetic illite tends to accommodate significant excesses of radiogenic ^{40}Ar, even though the effect of such excesses may be overwhelmed by the uptake of K. If so, it calls to question the meaning of K-Ar dates of clays extracted from buried shales. For instance, arguments about using the increases in K and ^{40}Ar contents with depth to date diagenesis would have to be modified if such incorporation of excess ^{40}Ar occurs.

On the basis of these observations, we are tempted to believe that the illitization process for illite/smectite clays from shales within a deeply buried diagenetic zone might proceed along a transformation mode. The structure of the clay particles is essentially maintained with the retention of part or even all of the inherited radiogenic ^{40}Ar, and with the incorporation, concurrently in the new layers, of additional ^{40}Ar provided by the alteration of detrital components in the immediate environment. Dissolution of detrital components might create insolated microenvironments with high partial pressure of radiogenic ^{40}Ar. The whole supply of K for the illitization is the sum of K coming from neighboring detrital grains that are essentially in contact with the clay particles plus that imported by fluids from locations well outside the zone of illitization. We agree with Awwiller (1993) that at least part of the K for the illitization in the Gulf Coast basin had to come from outside the illitization site, and with Furlan et al. (1996) that part of the K needed for the illitization of illite/smectite clays in the Mahakam Delta basin had to come from deeper parts of the basin. The slow decrease in the K-Ar dates of the size fractions enriched in illite/smectite mixed layers from deeper parts of the sequences might be ascribed to a partial loss or release of radiogenic ^{40}Ar with little or no change in the K contents. This could have been induced by a cannibalistic dissolution-crystallization in poral environments from which ^{40}Ar was able to escape.

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