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OCCURRENCE AND PALAEOHYDROLOGICAL SIGNIFICANCE OF AUTHIGENIC KAOLINITE IN THE ALDEBARAN SANDSTONE, DENISON TROUGH, QUEENSLAND, AUSTRALIA

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Abstract—Thin section, XRD, SEM, and isotopic techniques have been used to study authigenic kaolinite occurring in reservoir sandstones of the Lower Permian Aldebaran Sandstone. Where the unit is no longer an active aquifer, kaolinite is an intermediate-stage phase, and is highly depleted in deuterium (δD_{SMOW} = −115 to −99‰) and ^{18}O ($\delta^{18}O_{SMOW}$ = +7.8 to +8.9‰), indicating that precipitation must have been from meteoric water. Deep penetration of this water is linked to Late Triassic deformation and uplift of the Denison Trough sequence, an event which led to exposure of the Aldebaran Sandstone by the Early Jurassic prior to its re-burial beneath Jurassic and Cretaceous sedimentary rocks. The same water was probably involved in the creation of secondary porosity in the interval.

Where the Aldebaran Sandstone is presently undergoing meteoric flushing, kaolinite is relatively enriched in deuterium (δD_{SMOW} = −104 to −93‰) and ^{18}O ($\delta^{18}O_{SMOW}$ = +11.7 to +14.6‰), reflecting precipitation largely from post-Mesozoic meteoric water which was isotopically heavier than the Mesozoic water involved in intermediate-stage kaolinite precipitation. This temporal shift in meteoric water isotopic composition is related to the northward drift of the Australian continent to lower latitudes since the Mesozoic Era.

Key Words—Aquifer, Diagenesis, Kaolinite, Petrography, Secondary porosity, Stable isotope analysis.

INTRODUCTION

Authigenic kaolinite is an important component of many subsurface sandstones worldwide. Although its physical characteristics in sandstone reservoirs have been well documented, few studies have firmly established the palaeohydrological significance of the mineral within such settings. In particular, it is well known that the presence of authigenic kaolinite signifies the passage of acidic waters at some stage in the diagenetic history of the sandstone. However, the question of whether this water is produced by mudrock diagenesis at depth or is surface derived has been addressed by many workers merely on the basis of rather ambiguous circumstantial evidence. Accordingly, it is not surprising that conclusions concerning particular reservoirs have led to controversy, especially as they have an important bearing on suggested mechanisms of secondary porosity development.

Recently, stable isotope data on authigenic kaolinite have been used to constrain the conditions existing at the time of its formation (e.g., Longstaffe and Ayalon, 1987; Ayalon and Longstaffe, 1988; Glasmann *et al.,* 1989). Specifically, these data have provided direct evidence for the origin of fluids involved in kaolinite precipitation by allowing their isotopic composition to be calculated. Information gained using this technique has contributed to the construction of elaborate diagenetic models for the sandstone reservoirs of interest.

As part of a comprehensive diagenetic investigation of the Aldebaran Sandstone, authigenic kaolinite occurring in its sandstones at depth was studied using a variety of analytical techniques including stable isotope analysis. The purpose of this paper is to present the results of these analyses, and to discuss in particular the significant implications of the stable isotope data for the origin and evolution of fluids and mechanisms of secondary porosity formation in this important gas reservoir.

GEOLOGICAL SETTING

The Denison Trough is a deep continental sub-basin situated along the western margin of the Bowen Basin in east-central Queensland (Figure 1). It contains up to 6500 m of Permo-Triassic siliciclastic sedimentary rocks which, in the south, are overlain by Jurassic sedimentary rocks of the Surat Basin (Brown *et al.,* 1983). During the Late Triassic, the Denison Trough sequence was folded into a series of large, north-trending anticlines and synclines. Economic accumulations of hydrocarbon gas trapped within closures along some of these anticlines make the trough one of the more important onshore hydrocarbon provinces in Australia.

The Lower Permian Aldebaran Sandstone is the principal hydrocarbon reservoir in the Denison Trough. It consists of a thick sequence of sandstones, conglomerates, and minor mudrocks and coals which accu-

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Table 1. Kaolinite isotope data.

Sample	Borehole	Depth (m)	δD (‰)$_{SMOW}$	$\delta^{18}O$ (‰)$_{SMOW}$
ED1-6	GSQ Eddystone 1	893.84	—	7.8
ED4-17	GSQ Eddystone 4	514.34	—	11.8
MV1-7[1]	AAR Merivale 1	1404.95	−105	9.2 8.6
MV2-4[1]	AAR Merivale 2	1407.50	−115	8.7 9.1
SP1-5	AAR Springvale 1	730.47	−99	8.0
SS4-5	GSQ Springsure 4	152.07	−104	11.7
SS10-3	GSQ Springsure 10	469.04	−93	14.6

[1] Replicate samples analysed for $\delta^{18}O$.

mulated in a wide range of fluvio-deltaic and nearshore marine environments. Over most of the trough, the underlying unit is the Cattle Creek Formation, a thick sequence of marine mudrocks and sandstones. Directly overlying lithologies are marine mudrocks and sandstones of the Ingelara and Freitag Formations. The Aldebaran Sandstone occurs virtually throughout the Denison Trough, with exposure restricted to within the Springsure, Serocold, and Consuelo Anticlines along the western side of the trough. Elsewhere it is covered by up to 1.4 km of Permo-Triassic strata of the upper Bowen Basin sequence, Jurassic sedimentary rocks of the Surat Basin, and Cenozoic basalts and alluvium.

BURIAL HISTORY

The Aldebaran Sandstone has experienced two major cycles of burial and uplift (Baker, 1991; Baker and

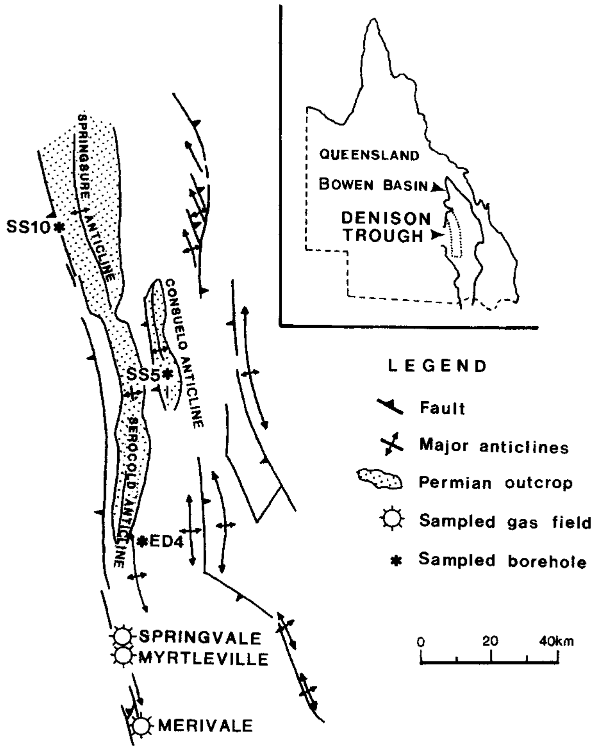

Figure 1. Denison Trough location and structure (from Brown *et al.*, 1983). ED = Eddystone, SS = Springsure.

Caritat, 1992). The first burial episode occurred during the Permian and Triassic Periods, and was terminated by major Late Triassic deformation and uplift. Stratigraphic and vitrinite reflectance data indicate that maximum burial depths and temperatures ranging from 1500 to 3000 m and 90° to 150°C, respectively, were attained at the end of this first burial phase (Baker, 1991). Following the Late Triassic deformation and uplift episode, which led to exposure of the unit in the area where Permian sedimentary rocks now outcrop, a second burial phase occurred during the Jurassic and Early Cretaceous, when over 1000 m of Surat Basin sediments accumulated over the Denison Trough. Late Triassic maximum temperatures were not exceeded during this second burial phase. This was in turn followed by a final phase of uplift and erosion during the Late Cretaceous. With post-uplift denudation of the land surface continuing to the present day, most of the overlying Surat Basin sequence has since been removed, resulting in the exhumation of deformed and eroded Permo-Triassic sedimentary rocks, including the Aldebaran Sandstone, in the north.

METHODOLOGY

The kaolinite isotopic study focussed on seven fluvio-deltaic and nearshore marine sandstone samples collected from depths of between 152 and 1408 m in boreholes located within the Springvale-Myrtleville (ED1-6, SP1-5) and Merivale (MV1-7, MV2-4) Fields, and within the Permian outcrop area along the Springsure, Serocold and Consuelo Anticlines (ED4-17, SS4-5, SS10-3) (Table 1 and Figure 1). Although a large number of other kaolinite-bearing samples from the Aldebaran Sandstone were collected, all were unsuitable for this study because they contained significant amounts of other clay minerals from which the kaolinite could not be separated. Samples were examined in thin section, and the <2.0-μm fraction of each was analysed by XRD. SEM was used to provide additional information on paragenetic relationships. Kaolinite for XRD and isotopic analysis was separated from sandstones by gentle crushing of the rock, ultrasonic disaggregation, settling (a step repeated three times for each sample) and centrifugation. Hydrogen and oxygen isotopic compositions were determined using the stan-

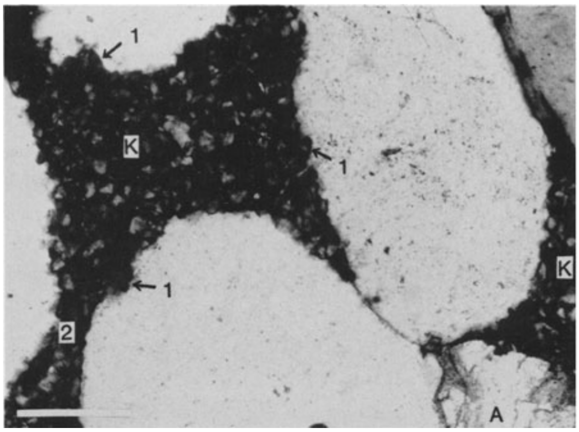

Figure 2. Photomicrograph showing kaolinite (K) infilling areas made available by ankerite dissolution. Evidence for ankerite decementation having occurred is severely corroded and embayed margins of quartz grains (1), elongated areas between adjacent quartz grains (2), and the presence of small ragged patches of oxidized ankerite (A). Yellowbank #3, 25°26′E 148°23′S, 1336.05 m, crossed polars, scale bar = 200 μm.

dard analytical techniques of Clayton and Mayeda (1963) and Coleman *et al.* (1982). All isotopic data are presented in per mil with respect to Standard Mean Ocean Water (SMOW). The reproducibility of standard δD and $\delta^{18}O$ values was 3‰ and 0.2‰ or better, respectively. XRD traces of the kaolinites prior to isotopic analysis confirmed their relative mineralogical purity, showing tectosilicates and other phyllosilicates to constitute less than 5 to 15% of each sample. Isotope results were not corrected for impurities.

RESULTS

Sandstone petrology

Representing typical reservoir sandstones of the Aldebaran Sandstone, the seven samples are medium-grained, well-sorted sublitharenites (Folk *et al.*, 1970), with an average QFR ratio of 79:7:14. Quartz is mainly monocrystalline with non-undulatory extinction, and subrounded. Feldspar varieties are orthoclase, microperthite, sodium-rich plagioclase and microcline. K-feldspar dominates over plagioclase. Rock fragments consist of acid to intermediate volcanics, fine-grained siliciclastics, granite, and low-grade metamorphics. Accessories are mica, tourmaline, zircon, and rutile. Detrital matrix is absent. Visual porosity varies between 2.6 and 9.6%.

Authigenic minerals present in the seven samples are quartz, calcite, dawsonite [$NaAlCO_3(OH)_2$], ankerite, siderite, kaolinite, illite, and allevardite ordered illite-smectite. Authigenic quartz contents are less than 3% of the bulk rock. Carbonate and clay abundances range from 0 to 6.8% and 9.4 to 14.4%, respectively. Kaolinite accounts for around 90% of the clay mineral fraction.

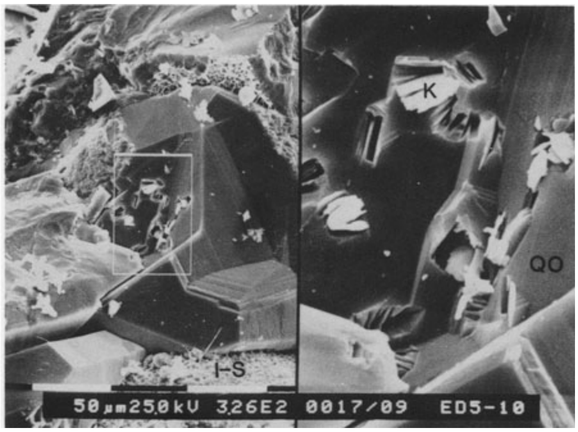

Figure 3. SEM photomicrograph showing kaolinite (K) booklets engulfed by quartz overgrowth (QO). Relation interpreted to indicate that the two are coeval or the overgrowth is the later phase. Illite-smectite (I-S) grain rim also shown. Eddystone #5, 25°00′E 148°29′S, 1084.84 m.

Kaolinite description

Authigenic kaolinite in the Aldebaran Sandstone forms delicate booklet and accordian-like (sub-vermicular), loosely to densely packed, randomly orientated aggregates of euhedral pseudohexagonal plates which line and fill primary and secondary pores and replace labile grains. Significantly, in samples from the Springvale-Myrtleville and Merivale Fields (as well as from other southern Denison Trough gas fields), it tightly infills enlarged intergranular voids within small areas characterised by open packing where margins of detrital grains are highly etched (Figure 2). Small corroded patches of ankerite occur within these areas, suggesting that the original voids were created as a result of ankerite dissolution. Commonly, kaolinite is engulfed by quartz overgrowths (Figure 3) and calcite (Figure 4).

Oxygen and hydrogen isotope data are given in Table 1 and are plotted in Figure 5. Kaolinite in the four

Figure 4. SEM photomicrograph showing calcite (C) engulfing kaolinite (K) pore fill. Eddystone #1, 886.01 m.

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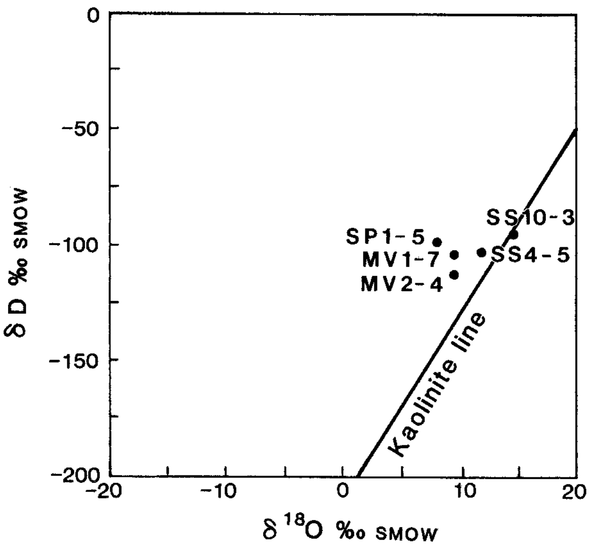

Figure 5. δD *vs* $\delta^{18}O$ for authigenic kaolinite in the Aldebaran Sandstone.

samples from the Springvale-Myrtleville and Merivale Fields has $\delta^{18}O$ values lying in the narrow range of +7.8 to +8.9‰, with a mean of +8.4 ± 0.6‰. δD values, measured for three of these samples, range from −115 to −99‰, and have a mean of −106 ± 8‰. Kaolinite in the three samples from the Permian outcrop belt has contrasting $\delta^{18}O$ values of +11.7 to +14.6‰, with a mean of +12.7 ± 1.6‰. δD values, measured for two of these samples, are −104‰ and −93‰, which have a mean of −98.5 ± 7.8‰.

DISCUSSION

Over the Denison Trough, the Aldebaran Sandstone forms a major aquifer with intake areas lying along the western side of the trough in the Springsure, Serocold, and Consuelo Anticlines. Formation water down to depths of at least 450 m in the vicinity of this outcrop area is fresh. In contrast, to the south and east of the outcrop area, formation water in places is highly enriched in sodium bicarbonate, indicating that here the Aldebaran Sandstone does not function as an active aquifer. Samples from the Springvale-Myrtleville and Merivale Fields (referred to hereafter as the southern samples) are from such an area, whereas samples from the Springsure, Serocold, and Consuelo Anticlines (referred to hereafter as the northern samples) are from the area where the Aldebaran Sandstone is presently undergoing active meteoric flushing near the intake zone.

Southern samples

Kaolinite in the southern samples is clearly an intermediate-stage diagenetic phase. It postdates ankerite, a phase which formed at elevated temperatures (>100°C) during the Late Triassic when maximum burial depths and temperatures were attained in the Aldebaran Sandstone. It predates calcite and probably some quartz overgrowths, phases which in turn are postdated by dawsonite, a late-stage hydrated sodium aluminium carbonate which precipitated in the present hydrologic regime. All of these minerals occur in the gas-bearing sandstones, indicating that they predate the entrapment of gaseous hydrocarbons (Baker, 1991). The occurrence of kaolinite within areas where the strongly alkaline soda brines presently exist further shows that the mineral is not a recent phase in the Aldebaran Sandstone, given that kaolinite precipitation is normally associated with acid, cation-depleted pore waters (Curtis, 1983).

There are two possibilities that may explain the cause of the acid flushing reflected by kaolinite in the southern samples. Since underlying humic, organic-rich mudrocks of the Cattle Creek Formation entered the oil window during the Triassic Period, the flushing could have been generated as a result of the abiotic thermomaturation of organic matter (e.g., Schmidt and McDonald, 1979; Surdam *et al.*, 1989) and the illitization of smectitic clays (Bjørlykke, 1983) in this unit. Alternatively, the source of the acid flushing could have been fresh water descended from the surface (e.g., Giles and Marshall, 1986; Bjørlykke and Brendsdal, 1986), a plausible mechanism given the fact that the unit underwent major deformation and uplift during the Late Triassic and, as a result, was exposed over the present-day outcrop area during the Early Jurassic. The kaolinite isotope data, together with geological evidence, indicate unequivocally that this latter mechanism is indeed the one which produced the acid flushing.

Since maximum burial temperatures in the Aldebaran Sandstone over the southern Denison Trough were no higher than about 120°C, as estimated on the basis of vitrinite reflectance data (Ro < 0.8%) (Baker, 1989), the mean kaolinite $\delta^{18}O$ value of +8.4‰ indicates that the $\delta^{18}O$ value of the involved water could not have been greater than about −5‰ (Figure 6). On this basis alone, the water must have been at least partly meteoric in origin, ruling out the possibility that the source of the acidic flushing was the adjacent marine, well-illitized (Baker and Caritat, 1992) mudrocks. Waters released by clay mineral reactions from such mudrocks would, in contrast, be enriched in ^{18}O, with $\delta^{18}O$ values possibly as high as +6 to +8‰ based on Gulf Coast data (Fisher and Land, 1986; Taylor, 1990).

It is contended that structural inversion of the basin during the Late Triassic (Ziolkowski and Taylor, 1985) created high topographic relief which permitted meteoric waters to invade the Aldebaran Sandstone at depth under a high hydrostatic head. Penetration would not have taken place until after some degree of uplift and deformation, with the intake zone of the unit probably not actually being exposed over the present-day Permian outcrop area until the earliest Jurassic. Accordingly, kaolinite precipitation temperatures were likely to have been well below the maximum burial temperature of around 120°C. From an average ground surface temperature of 15°C (the Denison Trough was at about 45°S during the Early Jurassic, Smith *et al.*, 1981) and an assumed palaeogeothermal gradient of 45°C/km, a minimum kaolinite precipitation temperature of 45°C can be calculated. This is on the basis that immediately prior to accumulation of the overlying Jurassic-Cretaceous Surat Basin sequence, the Aldebaran Sandstone in this southern part of the Denison Trough was about 700 m below the peneplained Jurassic land surface (Baker, 1989). Given this situation, 70°C is considered to be a realistic upper temperature limit for kaolinite precipitation. Hence, if kaolinite precipitation occurred at temperatures of between 45° and 70°C, water involved must have had a $\delta^{18}O$ value of between about −13.9 and −10.1‰ (Figure 6), indicating, on the basis of these geologically more realistic precipitation temperatures, that it was indeed surface derived. Such a highly ^{18}O-depleted water composition is entirely consistent with the mid to high latitude setting of the Denison Trough during the latest Triassic and Early Jurassic (cf. Bird and Chivas, 1989).

Studies by O'Neil and Kharaka (1976) and Yeh and Savin (1977) suggest that the analysed kaolinite, not having experienced temperatures greater than 90°C subsequent to precipitation (see Baker and Caritat, 1992) and typified by its coarse grain size (crystals commonly 20 to 40 μm wide), would have undergone negligible post-formational oxygen isotopic exchange with pore fluids. Conclusions presented herein based on its $\delta^{18}O$ values are thus considered to be well founded, with little chance of the depleted values reflecting oxygen isotopic re-equilibration with more recent meteoric pore waters. Departure of the kaolinite compositions from the kaolinite line (which represents the locus of isotopic compositions for kaolinite formed at the surface in equilibrium with unmodified meteoric water, see Figure 5) would be due to (1) precipitation at temperatures above those at the surface (Hoefs, 1987), and (2) post-formational partial hydrogen isotopic exchange with slightly heavier meteoric water, a process which is significant at temperatures below 80° to 100°C (O'Neil and Kharaka, 1976; Bird and Chivas, 1988; Longstaffe and Ayalon, 1990).

Geological evidence that the acid water was not derived from adjacent marine mudrocks is provided by the ankerite-kaolinite textural relations. Ankerite formed before the kaolinite at model temperatures greater than 100°C (Baker and Caritat, 1992). Accordingly, on the basis of this temperature, the precipitation of kaolinite as well as ankerite must have followed peak acid generation and water liberation from the mudrocks, an event which occurs at temperatures of only around 80° to 90°C (Surdam *et al.*, 1989). The abundance of carbonate fossils in the adjacent marine mud-

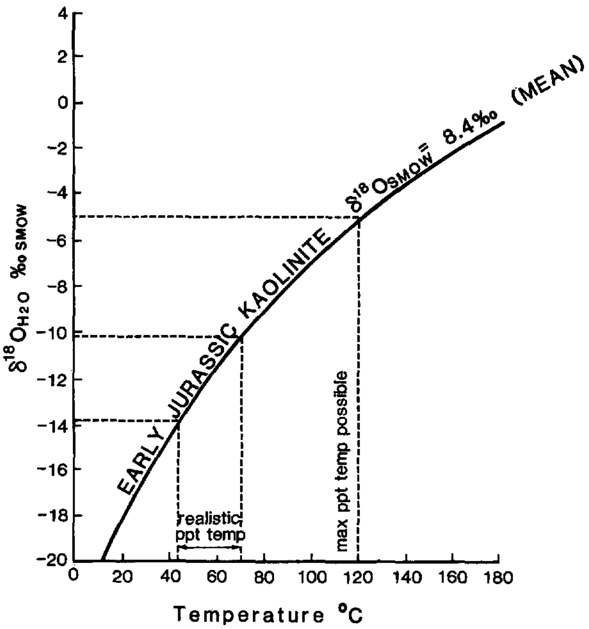

Figure 6. Formation water $\delta^{18}O$ value versus temperature for intermediate-stage authigenic kaolinite in the Aldebaran Sandstone. Curve is for the mean kaolinite $\delta^{18}O$ value, and was calculated using the kaolinite-water fractionation equation given by Land and Dutton (1978).

rock units further argues that mudrock-derived acids were not involved in kaolinite precipitation. Any such acids produced must have been neutralized by these carbonates before entering the Aldebaran Sandstone (e.g., Giles and Marshall, 1986). It is worth noting that the meteoric origin of deep kaolinite in the Aldebaran Sandstone, as implied by the isotope and geological data, contradicts the contention of Bjørkum *et al.* (1990) that meteoric water infiltration is unlikely to bring about kaolinite formation at depth in sandstone units exposed at unconformities.

The Aldebaran Sandstone south and east of the outcrop area ceased being an active aquifer during the Tertiary (Baker, 1991). As a result, entrapped meteoric waters became enriched in sodium bicarbonate, creating a chemical environment conducive to the precipitation of late-stage sodium aluminium carbonate (dawsonite) and incompatible with kaolinite formation.

Northern samples

Many subsurface samples from depths down to at least 520 m in the vicinity of the Permian outcrop area are strongly kaolinitized. Given the present hydrodynamic regime in this area of fresh water flushing, the abundant authigenic kaolinite in these samples is likely to be at least partly a late-stage phase, formed within this regime. Thin section examination of these samples revealed no textural evidence which could be used to determine the relative timing of the kaolinite.

Kaolinite in the northern samples, with a mean $\delta^{18}O$ value of +12.7‰, is considerably enriched in ^{18}O compared to the southern samples. This is consistent with its precipitation from post-Mesozoic, low-latitude meteoric waters. These waters would have been isotopically heavier than the mid to high-latitude Mesozoic waters involved in the precipitation of the intermediate-stage kaolinite on account of the northward drift of the Australian continent to lower latitudes since the Mesozoic Era (see Bird and Chivas, 1989). Using present-day surface water $\delta^{18}O$ values of −6 to −4‰ (based on data in Bird and Chivas, 1988), the isotopic temperature of kaolinite formation in the three samples is calculated to be about 20° to 60°C higher than present day temperature at the sample depths, probably indicating that the $\delta^{18}O$ values have been depressed somewhat by the presence of visually masked, isotopically lighter Mesozoic kaolinite in the same samples. This evidence, together with the proven existence of early, isotopically light diagenetic kaolinite in the Aldebaran Sandstone, supports Bird and Chivas' (1989) contention that true $\delta^{18}O$ values of weathering kaolinites analysed by them in outcropping sandstones of the Aldebaran Sandstone and Cattle Creek Formation are actually higher than the measured because of the presence of co-existing, masked, early diagenetic kaolinite with a low $\delta^{18}O$ value.

The two northern samples for which δD was measured have δD and $\delta^{18}O$ values that plot on and very close to the kaolinite line (Figure 5), indicating that the kaolinite in these samples precipitated at near surface temperatures and has undergone little or no post-formational hydrogen isotopic exchange. This is entirely consistent with the interpreted late-stage origin of the kaolinite in this area.

Origin of secondary porosity

Secondary porosity in the Aldebaran Sandstone has formed as a result of feldspar and rock fragment dissolution and ankerite decementation. It accounts for over half of the total visual porosity in some sandstones, and is largely responsible for the good reservoir quality existing in the unit over the economically important southwestern part of the Denison Trough.

Kaolinite in the southern samples tightly infills areas which were clearly occupied by ankerite (see Figure 2), yet, in other parts of the same thin section where ankerite was never present, kaolinite may be absent despite the presence of open, well connected primary pores. This intimate relationship between authigenic "dissolution" kaolinite and carbonate decementation porosity has been noted by others (in Curtis, 1983; Franks and Forester, 1984; Burley *et al.*, 1985), and may be explained by the fact that, (1) over the acid range, kaolinite solubility rapidly decreases as pH rises, and (2) ankerite dissolution would have caused a local rise in pH of the pore fluid. Simultaneous precipitation of kaolinite with ankerite dissolution would thus be expected (see Curtis, 1983), implying that freshwater flushing was the mechanism by which ankerite decementation porosity was created in the Aldebaran Sandstone.

Volumetrically important grain dissolution pores in the southern samples are clearly post-compaction, and predate dawsonite as shown by the occurrence of this mineral within some of the pores. In view of this timing relationship, it is suggested that the same freshwater flush which created the kaolinite-infilled ankerite decementation porosity also created most grain dissolution porosity within the unit. Dissolved feldspars are likely to have supplied the necessary silica and aluminium for kaolinite precipitation.

SUMMARY AND CONCLUSIONS

Stable isotope analysis was carried out on four intermediate-stage and three late-stage authigenic kaolinites extracted from subsurface sandstones of the Aldebaran Sandstone. Results show that the intermediate-stage kaolinite is highly depleted in deuterium and ^{18}O, indicating that precipitation was from meteoric water rather than fluids generated at depth in the adjacent marine mudrock units. Ingress of this water is linked to a Late Triassic phase of deformation and uplift in the Denison Trough, which led to exposure of the Aldebaran Sandstone by the Early Jurassic prior to further regional sediment accumulation. The intimate association between kaolinite and ankerite dissolution porosity in the unit suggests that the same water was responsible for the creation of the ankerite dissolution porosity. Leaching of labile grains probably also occurred during this time, giving rise to existing volumetrically significant grain dissolution porosity and the silica and aluminium necessary for kaolinite precipitation.

Late-stage kaolinite in the Aldebaran Sandstone is relatively enriched in deuterium and ^{18}O, reflecting precipitation largely from post-Mesozoic meteoric water which was isotopically heavier than the Mesozoic water involved in intermediate-stage kaolinite precipitation. The temporal change in the isotopic composition of these waters is related to the latitude dependence of the isotopic composition of meteoric water, and to the northward drift of the Australian continent to lower latitudes since the Mesozoic Era.

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