ABSTRACT: Replacement of kaolinite by dickite has been observed to occur with increasing depth of burial in sandstones from three different basins on the Norwegian continental shelf. In the Garn Formation (Middle Jurassic) of Haltenbanken, samples from 1.4-2.7 km below the sea floor (110°C) contain kaolinite, whereas deeper than 3.2 km (130°C), mainly dickite is present. In the Statfjord Formation (Late Triassic-Early Jurassic) from Gullfaks and Gullfaks Sør Fields, transformation of kaolinite to dickite occurs at ~3-1 km below the sea floor (120°C). From the Stø and Nordmela Formations (Lower to Middle Jurassic) to the Troms Area, kaolin polytypes have been identified in only two shallow and two deep samples, but the results are consistent with the transformation depth determined in two other areas studied. These occurrences are significant because they allow the temperature of the kaolinite/dickite transformation to be established with greater confidence than had been possible previously. Also the observation of this transformation in all three areas so far examined indicates that it may be a general and predictable feature of kaolin-bearin g sandstones worldwide and therefore a potentially reliable paleo-geothermometer. In most cases, the kaolinite occurs as relatively large vermicular crystals, whereas dickite forms more euhedral, blockier crystals. This morphological difference, together with the nature of the structural difference in octahedral occupancy between the kaolinite and dickite, suggests that the transformation occurs by dissolution and reprecipitation, rather than in the solid state.

KAOLIN POLYTYPES

Kaolinite and dickite are two polytypes of the kaolinite sub-group of clay minerals (Bailey, 1980a) which also includes the polytype nacrite. The AIPEA nomenclature thus recommended the name “kaolinite” for both the polytype and the sub-group, so that when one says that the clay present in a rock is “kaolinite”, it remains unclear whether the polytype present is in fact dickite, kaolinite, both, or has not actually been determined. The alternative sub-group name “kandite” has been specifically disallowed (Bailey, 1980a), so probably the best solution is informal use of the term “kaolin” for the sub-group, as used by Bailey (1980a,b) and in the definition given in the current edition of the Glossary of Geology (Bates & Jackson, 1987). Although this is not the formal sub-group name, the advantage for concise and specific scientific writing is considerable, and there appears to be relatively minor potential for confusion with the alternative use of the term “kaolin” for the rock or industrial material consisting dominantly of kaolinite sub-group clays. In this paper, therefore, “kaolin” will be used as an informal designation for the kaolinite sub-group.
(a general term whereby polytype identity is not specified), while the name “kaolinite” will be used to refer specifically to the polytype.

As many readers may not be familiar with the exact nature of the difference between kaolinite and dickite, a brief review follows. Both kaolinite and dickite have the formula $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$. Their structure consists of a plane of Al atoms octahedrally coordinated to hydroxyl ions, which is bonded to a plane of Si atoms tetrahedrally coordinated to oxygen (Fig. 1). These tetrahedral-octahedral sheet pairs (1:1 layers) have a spacing of $\sim 7.2$ Å perpendicular to the plane of the layering. Successive 1:1 layers are held together by hydrogen bonds between the OH$^-\,$ at the top of the octahedral sheet in one 1:1 layer and the oxygens at the base of the tetrahedral sheet in the next 1:1 layer. The structural energy is minimized when successive layers are stacked so that the distances of these hydrogen bonds are as short as possible. This occurs when the unit-cell of each succeeding layer is offset by either $a/3$ (1/3 of the unit-cell dimension in the $a$ crystallographic direction), as in kaolinite and dickite, or $b/3$, as in nacrite.

The kaolinite sub-group minerals are dioctahedral because the Al cations occupy only two of every three octahedral sites. In kaolinite, the empty octahedral site occurs in the same relative position in each succeeding layer. In both dickite and nacrite, the empty site alternates between positions from one layer to the next (Fig. 1). Kaolinite thus has one 1:1 layer per unit-cell, while two 1:1 layers are necessary to define the unit-cell of dickite. Kaolinite has triclinic symmetry; the $a/3$ displacement of each succeeding 1:1 layer results in $\beta = 104.8^\circ$, and minor displacement along $b$ results in a slight departure from 90° in $\alpha$. Dickite is monoclinic; $\beta$ is the same as in kaolinite, but there is no displacement along $b$.

![Crystal structures of kaolinite (A) and dickite (B). Cross-section of 1:1 layer structure looking parallel to the $a$ axis. In kaolinite the empty octahedral site (X) occupies the same position in successive layers, whereas in dickite it alternates between two positions. Modified (by permission) from Drever (1988, p. 79).](image-url)
Kaolinite and dickite are said to be well crystallized when the stacking of their layers approaches the ideal scheme described above. However, both polytypes can display varying degrees of disorder resulting from random displacement of successive layers by $1/3b$. In kaolinite the degree of such disorder can be extreme, equivalent to a random distribution of the empty octahedral site, but in dickite the degree of disorder seems to be limited.

PREVIOUS WORK ON THE OCCURRENCE AND FORMATION OF DICKITE

Dickite was first recognized by Dick (1888, 1908) and was named by Ross & Kerr (1931). Subsequently, the literature accumulated many reports of individual occurrences of dickite and measurements of its physical properties (references given in Bayliss et al., 1965). Although dickite was first perceived to have a hydrothermal origin, it was later found as a widespread authigenic component of sandstones and other sedimentary rocks having no apparent association with hydrothermal activity (Hemingway & Brindley, 1948; Smithson, 1957; Ferraro & Kubler, 1964; Bayliss et al., 1965). Nevertheless, its common association with hydrothermal activity provided early evidence that dickite might be stable at higher temperatures relative to kaolinite, which was the polytype known to be characteristic of weathering profiles and other very low-temperature environments. An example was described by Schroeder & Hayes (1968) from limestone strata in Kansas, USA where dickite occurs preferentially in porous algal mounds, within an elliptical area 201 km long and extending 97 km eastward from a group of ultramafic igneous intrusions. Kaolinite occurs in less porous lithologies within this area and also in the surrounding territory, where dickite is not observed. The authors interpreted the dickite to have formed from heated groundwaters, possibly mixed with magmatic waters, which moved outward from the intrusions through the conduit-like algal mounds. “Where water movement was restricted or where water had travelled tens of miles from the intrusions, water temperature fell below the temperature for dickite crystallization, and kaolinite precipitated instead”. An alternative possibility is that the widespread kaolinite partly or entirely predates the hydrothermal activity and was recrystallized to dickite and partly remobilized during the hydrothermal event.

It has long been recognized that transformation of kaolinite to dickite occurs as a regional diagenetic process related to increasing depth of burial in sedimentary basins. Apparently the first evidence for this was reported by Smithson (1954), who mapped a geographical boundary between kaolinite- and dickite-bearing Middle Jurassic sandstones in Yorkshire, UK. The dickite-bearing sandstones have been more highly altered by diagenesis in response to greater burial depth, as indicated by greater alteration of feldspar to kaolin, greater quartz cementation, greater corrosion of garnet and staurolite, and increased abundance of authigenic anatase and overgrowths on zircon grains. Additional data on this occurrence were given by Kantorowicz (1984).

Kossovskaya & Shutov (1963) outlined a general model for diagenetic to low-grade metamorphic facies, which include reference to earlier work (in Russian) in which dickite was reported to have replaced kaolinite with increasing grade. Their “quartz-dickite facies” was said to appear at depths of 2.5 km in horizontal Mesozoic strata of the Siberian platform and 1.0–1.5 km in Riphean (late Precambrian) and Lower Paleozoic strata of the Russian platform. Shutov et al. (1970) added the further information that the kaolinite-dickite transition was also observed in the Kitzil, Karaganda, and Pechora basins, but
provided no details on these localities. However, they did show a schematic lithologic column for the Russian platform occurrence with a series of seven differential thermal analysis curves indicating the progressive replacement of kaolinite by dickite in successively older sandy deposits of continental facies. A further stress-dependent dickite-to-nacrite transformation is also suggested, but without details or data.

Ferrero & Kubler (1964) found that Cambrian sandstones in Algeria contain authigenic dickites of relatively large crystal size (20–60 μm), whereas interlayered claystones and siltstones contain kaolinite of probable detrital origin and much finer crystal size (~2 μm). Cassan & Lucas (1966) concluded that the following sequence of diagenetic events had affected these sandstones: (1) alteration of feldspar and mica to form kaolinite plus quartz overgrowths; (2) cementation with dolomite and anhydrite; (3) dissolution of quartz cement and kaolinite and precipitation of dickite; (4) emplacement of oil and subsequent illitization of dickite in rocks that were not oil-filled. Both this occurrence and the Kansas limestones described by Schroeder & Hayes (1968) provide evidence that the transformation of kaolinite to dickite occurs more readily in rocks with higher permeability.

In several subsequent compilations listing reactions indicative of increasing diagenetic grade, the transition of kaolinite to dickite has been reported with reference to the above publications (Dunoyer de Segonzac, 1970, p. 287–290; Hoffman & Hower, 1979, p. 58; Kisch, 1983, p. 335–337; Frey, 1987, p. 31–32). These authors showed considerable diversity in their interpretations. For example, Hoffman & Hower (1979, their Fig. 15) plotted the temperature range of kaolinite-to-dickite/nacrite transformation as 110–140°C, although the only reference cited, Shutov et al. (1970), actually contains no temperature data. On the other hand, Frey (1987) concluded that “the rather limited and divergent results do not favour the use of dickite as an index mineral”.

**ANALYTICAL TECHNIQUES**

In the present study, kaolinite and dickite were distinguished by the combined use of infrared spectroscopy (IRS) and X-ray diffraction (XRD). In practice, however, it was found that there are considerable advantages to using IRS as the primary monitoring technique. There is no difficulty in identifying the kaolin polytype by XRD when it occurs as a pure mineral, but in multicomponent mixtures including non-clay minerals, careful scrutiny and evaluation of the complete XRD pattern is required. In contrast, there are few interfering effects from other minerals in the diagnostic region of the IR spectra, as shown below.

The <4 μm size range was chosen for analysis rather than the standard <2 μm fraction because of the relatively large size of the kaolin crystals as observed in thin-section and by scanning electron microscopy (SEM). Although a still larger size cut-off would no doubt contain more of the total clay present in the rock, the attendant inclusion of substantial amounts of non-clay minerals results in dilution and masking of the kaolin signal. The <4 μm fraction is therefore a compromise intended to maximize the kaolin content of the analysed material.

The degree to which the kaolin content of the <4 μm fraction is indeed representative of the bulk kaolin content of the sandstones analysed is a question of valid concern. Unfortunately, we have not performed the experiments necessary to provide a quantitative answer to this question. The problem is analogous to the uncertainties involved in the conventional practice of XRD analysis of the <2 μm fraction to determine the nature of the
clay content in a sandstone. Although the relative proportions of clay minerals in the fine fraction will be expected to differ from those in the bulk sample, the major species present in the bulk sample will also be substantially represented in the fine fraction. Thus we would expect that if, for example, the kaolin content of the <4 \mu m separate consists overwhelmingly of dickite with only traces of kaolinite, then the bulk kaolin content of the rock will be predominantly, if not also overwhelmingly, dickite. At the same time, we would not expect that a kaolinite:dickite ratio of, for example, 5:2 in the <4 \mu m fraction would necessarily indicate a similar ratio in the bulk sample. Nevertheless, we feel that both kaolinite and dickite may reasonably be inferred to comprise substantial proportions of the bulk kaolin content of that sample.

**IRS**

The <4 \mu m fraction was separated from the sandstone samples after gentle crushing followed by ultrasonic dispersion and sedimentation in deionized water. It was necessary to treat some samples with petroleum ether to remove hydrocarbons. Two mg of the <4 \mu m material were then incorporated into a 13 mm diameter KBr pressed disk and the spectra recorded over the range 4000–250 cm\(^{-1}\) on a Perkin Elmer 580B spectrometer both at room temperature and after heating the disk to 150°C for 16 h and cooling in a dessicator. The disks were then diluted by a factor of 1 to 8 (equivalent to 0.25 mg sample in the disk), to show the spectral detail of intense silicate absorption near 1000 cm\(^{-1}\).

Distinction between kaolinite and dickite could be made most easily by assessing the position and relative intensity of the OH-stretching bands in the 3700–3600 cm\(^{-1}\) region (Russell, 1987; Fig. 2A). For well-crystallized kaolinite, there is strong absorption at 3697 cm\(^{-1}\) and a band of medium-strong intensity at 3620 cm\(^{-1}\). Between these bands there is relatively weak absorption at 3669 and 3652 cm\(^{-1}\). In contrast, the strongest absorption in dickite occurs at 3621 cm\(^{-1}\) with medium-strong bands at 3704 and 3654 cm\(^{-1}\). These differences, along with other subtle changes in the Si–O stretching and O–H deformation regions between 1000 and 3000 cm\(^{-1}\), enabled at least a preliminary identification of the dominant kaolin polytype present. In some instances, it was concluded that a mixture of the two polytypes was present, although it was not possible to say whether this was a true mixture or a more intimate dickite/kaolinite stacking within individual crystallites.

Ratios of the relative amounts of the two polytypes were calculated by assessing the spectrum over the range 3800–3500 cm\(^{-1}\) with the wave number scale expanded by a factor of five to provide higher resolution (Fig. 2B). It is over this limited range, the OH-stretching region, that the assessment of the kaolinite/dickite ratio is carried out by comparing the pattern of absorption bands from each sample with a series of computer-generated kaolinite/dickite mixtures derived from the IR spectra of pure, well crystallized kaolinite and dickite. Some allowance may have to be made if OH-containing components like illite and smectite are present, since their presence can influence the final absorption envelope in the hydroxyl stretching region. In any case, the ratios estimated should be regarded as only approximate.

**XRD**

As distinction between kaolinite and dickite depends upon recording non-basal reflections, random crystal orientation is desirable (Wilson, 1987). Where sufficient material was available, samples were prepared by packing into a standard Al holder for a
Philips diffractometer. This does not produce completely random orientation, but is generally adequate to enable the diagnostic non-basal reflections of the kaolin polytypes to be recorded. When only small amounts of material were available, the samples were dried down onto a glass slide from suspension in acetone, providing a relatively random orientation to the platy clay minerals (Paterson et al., 1986). Samples were then run from 3–60° 2θ in a Philips 1130 2 kW diffractometer at 2° 2θ/min using Fe-filtered Co-K radiation. Divergence, receiving and anti-scatter slits of 1°, 0·2 mm and 1°, respectively, were used throughout the recording range.

Initially, assessments were made without prior knowledge of the IRS results. In some samples containing smaller proportions of kaolin minerals, difficulties were encountered due to overlap between potentially diagnostic kaolin reflections and reflections of non-clay minerals. For example, strong kaolinite reflections at 3·84 and 2·52 Å, which have no counterparts in the dickite pattern, also coincide with reflections from feldspar. Similarly, the 2·28 and 2·24 Å reflections for kaolinite also coincide with reflections at these spacings from quartz. Several samples were contaminated with barite, which has reflections approximately coincident with the 2·33 and 2·50 Å reflections of kaolinite and the 3·79 Å reflection of dickite. There are, in fact, remarkably few X-ray reflections which can be used

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Fig. 2. (A) Computer-generated IR spectra for kaolinite (100K), dickite (100D), and various mixtures. (B) IR spectra of <4 µm fraction separated from sandstones from well 34/10-30, showing kaolinite:dickite ratios determined. Sample depths are given in metres on the right.
Fig. 3. XRD patterns of the <4 μm fraction separated from sandstones from well 34/10-30, showing diagnostic peaks (values in Å). (A) 3228 m. Only kaolinite reflections can be observed despite 30% dickite stacking according to IRS. (B) 3245 m. Diagnostic dickite peak at 4.13 Å is weakly apparent despite 40% dickite stacking according to IRS. (C) 3313 m. Diagnostic dickite peaks are indicated. K: kaolinite. D: dickite.

Kaolinite to dickite transformation


unequivocally to distinguish the kaolin polytypes in these circumstances, but the following non-basal reflections were found to be most useful (Fig. 3). For well-crystallized kaolinite, there is a strong (50) reflection at 4.18 Å which is not found in the dickite pattern. Similarly, in dickite there are reflections at 3.79 (55), 3.42 (20), and 2.21 (15) Å which are not found in
the kaolinite pattern. In addition, the 4·12 Å reflection of dickite is twice the intensity of the same reflection for kaolinite (65 vs. 30, respectively). Using the above criteria, it was found that the XRD results were completely consistent with those obtained by IRS. However, mixtures of the two polytypes were detected by XRD in only a few samples, and generally it was a case of identifying only one polytype (Fig. 3).

KAOLINITE-DICKITE TRANSITION IN CORES FROM THE NORWEGIAN CONTINENTAL SHELF

The apparent replacement of kaolinite by dickite related to increasing depth of burial in sandstones has been observed from three different basins on the Norwegian continental shelf (Fig. 4; Table 1). These occurrences are significant because they allow the temperature of kaolinite/dickite transformation to be established with greater confidence than has been possible previously. In the first two examples, present reservoir temperatures are known from drill-stem tests and are believed to be maximum temperatures experienced. Also, the observation of this transformation in all three cases examined indicates that it may be a general and predictable feature of kaolin-bearing sandstones worldwide and therefore a potentially reliable paleogeothermometer.

In the following discussion, it should be remembered that two different data are used for depth values. Individual samples are identified by their “core depth”, where the datum is RKB (relative to kelly bushing). However, because rock temperature and consequently diagenetic reactions are insensitive to water depth, it is more appropriate to discuss and plot analytical results with respect to the datum of RSF (relative to sea floor). All of the samples analysed are sandstones.

Garn Formation, Haltenbanken

This Middle Jurassic (Bajocian-Bathonian), blanket-like sand deposit is an important reservoir unit in several oil and gas fields offshore from mid-Norway (Fig. 5). It averages

![Fig. 4. Locations of area studied. Rectangles A, B, and C show areas enlarged in Fig. 5.](image-url)
### Table 1. Analytical results

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Abbreviations:
- Well sym = symbol used in figs.
- AT = analytical technique
- IRS = infrared spectroscopy
- XRD = X-ray diffraction
- KAO = kaolinite
- DIC = dickite
- K:D = ratio of kaolinite:dickite (interpreted from IRS data)
- ILL = illite
- SMT = smectite
- CHL = chlorite
- QTZ = quartz
- FSP = feldspar
- CAR = carbonate
- BAR = barite

Mineral abundances:
- IRS: Relative peak intensities are shown on a scale from null (--) to a maximum of five (+++++).
- XRD: Abundances are estimated as null (--), low (+), moderate (++), or >50% of sample (+++).
A kaolinite to dickite transformation

haltenbanken

7° 00' - 8° 00'

65° 00' - 65° 78'

N.

I. B.

O.

Trestakk

Tyrhans

Mikkels

6406

6407

6506

6507

K.

A.

Heidrun

N.

E.

I.

J.

Smorbukk

Middag

P.

Sør

C.

D.

N.

G.

B.

Troms

20° 00' - 21° 00'

7° 00' - 8° 00'

64° 15'

6° 00' - 7° 00'

km

~60 m in thickness and was deposited under both near-shore marine and fluvial conditions in a regressive, fan delta setting. It consists mainly of medium-grained subarkosic arenite with K-feldspar several times as abundant as plagioclase (Ehrenberg, 1990, 1991). The burial depth of the unit increases from east to west across Haltenbanken, mainly due to greater thicknesses of Cretaceous strata toward the west. Burial depth throughout Haltenbanken was dramatically increased relatively recently by deposition of a uniform 1 km thickness of poorly sorted, glacially derived detritus since Mid-Pliocene time (Fig. 6). The reservoir is believed to be presently at its maximum depth and temperature. As described in Ehrenberg (1990) and Ehrenberg & Nadeau (1989), the Garn Formation is fairly uniform in primary sand quality throughout the area studied, but major changes in diagenesis and reservoir quality are associated with increasing burial depth and temperature.

Fig. 5. Locations of wells studied (solid circles) within areas shown in Fig. 4. Open circles show locations of additional wells not sampled. Wells sampled in Haltenbanken are labelled with single-letter well symbols, identified in Table 1. Wells sampled in block 34/10 and the Troms area are labelled with the final characters of the well number (Table 1).
Kaolin in the Garn sandstones occurs mainly as microporous clusters of authigenic crystals filling both volumes between grains and molds of dissolved feldspar grains. Subordinate amounts of kaolin occur as an alteration of mica grains. The amount of kaolin present ranges from <1–8 volume % (including microporosity between booklets) in different samples (Fig. 8 in Ehrenberg, 1991).

Figure 7 shows that there is a clear division between shallower and deeper core samples in terms of the kaolin polytype identified by XRD and IRS. Unfortunately, no cores have been taken in the depth interval 2.7–3.2 km below the sea floor, so that exact depth of the kaolinite-dickite transition and its sharpness as a function of depth are unknown. All of the samples analysed contain either predominantly kaolinite or predominantly dickite, with either minor amounts or none of the complementary polytype. Based on maximum temperature measurements from drill-stem tests, the depth of the kaolinite-dickite transition in the Garn Formation corresponds to a temperature range of 110–130°C (Fig. 8).

**Statfjord Formation, Block 34/10**

The Triassic-Lower Jurassic (Rhaetian-Sinumurian) Statfjord Formation is an important oil reservoir in Gullfaks and Gullfaks Sør Fields and a number of other major oil fields of the northern North Sea area. It is 170–260 m thick in the 34/10 area. In the 34/10 area the subdivisions are, from the base upwards (Petterson et al., 1990; Olaussen et al., 1993):

1. Raude Member—(50–120 m thick) shale, siltstone, and subordinate sheet flood sands deposited in a dry climate with formation of abundant pedoturbated and caliche horizons;
2. Eiriksson Member—(40–100 m thick) stacked channel sands and shaly overbank deposits representing a more poorly drained, humid environment;
3. Nansen Member—(15–70 m thick) a laterally extensive transgressive marine sandstone overlain by heterogeneous shoreline and embayment sediments. The sandstones vary from akosic to subarkosic arenite, with widely variable grain size, sorting, and detrital clay matrix content. The burial history resembles that of the Garn Formation, except for less prominent subsidence in post-Mid-Pliocene time (Fig. 6).

Sandstone samples were analysed from well 34/10-C-2 in Gullfaks Field and from five wells in Gullfaks Sør Field (Fig. 9). A transition from kaolinite-bearing to dickite-bearing samples is observed at ~3.1 km below the sea floor, with the exception of a zone of
anomalous results in well 34/10-2. Drill-stem test data from Gullfaks Sør Field indicate a temperature of ~122°C at 3.1 km below the sea floor (Fig. 8).

In well 34/10-30, the cored interval appears to span the depth of transition from kaolinite to dickite. The two shallowest samples from this well contain exclusively kaolinite; the two deepest samples contain exclusively dickite; and the three intermediate-depth samples contain subequal amounts of both polytypes.

A zone having the opposite pattern is observed in well 34/10-2, where the two shallowest samples from the cored interval contain exclusively dickite, consistent with the depth pattern of the samples from the other wells. However, the two deepest samples contain exclusively kaolinite, and two intermediate depth samples contain subequal amounts of both kaolinite and dickite. An explanation for the “upside-down” trend in this core is not apparent. The two deepest samples are from the Raude Member and are in the water zone, while the four overlying samples are from the Eiriksson Member and are in the oil zone. The two exclusively kaolinite-bearing Raude samples are unusual in containing abundant Fe-rich chlorite coating grain surfaces, but the overlying Eiriksson samples have a more typical petrographic appearance.

The kaolinite-bearing samples from well 34/10-2 are regarded as representing an anomaly to the overall depth pattern. We feel the existence of this inconsistency does not necessarily invalidate the model of temperature being the main factor controlling the transformation of kaolinite to dickite, but rather indicates that local departures can be anticipated due to additional factors not presently understood. The present sample set covers the existing cored intervals from the Statfjord Formation between 2.9 and 3.4 km RSF from block 34/10, but additional cores from Brent Group sandstones in this interval have yet to be examined.
**Stø and Nordmela Formations, Troms Area**

These Lower to Middle Jurassic sandstones form the main reservoirs for several giant gas fields off north Norway. The Nordmela Formation (Sinumurian-Toarcian) is dominantly fine-grained sandstone and shale deposited in a low- to medium-energy, coastal alluvial plain/tidal flat environment (Olaussen et al., 1984). Channel sandstones become more abundant toward the top of the unit. The Stø Formation (Toarcian-Bajocian) consists of fine- to medium-grained sandstone deposited in a high-energy, shoreface/foreshore environment. The sandstones of both units are mainly quartz arenites (Riches et al., 1986). In the two wells sampled, the Stø and Nordmela Formations have thicknesses of ~150 and 200 m, respectively. These wells are located in the Ringvassøy-Loppa fault complex, an extensionally faulted transition zone between the Hammerfest and Tromsø basins (Olaussen et al., 1984).

Unusually rapid decline in porosity with depth in these sandstones (due mainly to quartz cementation) is attributed to post-Late Miocene uplift and erosion of ~1-0-1.2 km of section (in the area of the well sampled in the present study) following attainment of maximum burial depths in Early Tertiary time (Nyland et al., 1992).

In order to test the possibility of a transition from kaolinite to dickite with increasing depth in the Troms area, samples were selected from two nearby wells where substantially different present burial depths were observed to be correlated with major differences in diagenetic grade and reservoir quality (Olaussen et al., 1984). The tops of the Stø and Nordmela Formations are encountered at 1372 and 1518 m RKB, respectively, in well 7119/12-2 vs. 2658 and 2811 m RKB, respectively, in well 7119/12-1. In each well, one sample was analysed from each formation. Both of the samples from well 7119/12-1 were found to contain well-crystallized kaolinite. However, in well 7119/12-1, the two samples analysed contained illite, but virtually no kaolin, apparently reflecting complete illitization of earlier kaolin.

Fortunately, however, the results of a more comprehensive sampling and analysis program for the sandstones in well 7119/12-1 were recently reported by Ramm (1991). Bulk XRD analyses consistently show zero % kaolin content in 22 Stø sandstones analysed and 10-56% kaolin in eight Nordmela sandstones. Two of the Nordmela samples (2820-0 and 2829-9 m RKB) were further examined by unoriented powder XRD analysis of <16 μm separates, revealing that the kaolin clay present consists entirely of dickite. Ramm (1991) presented diffractograms of both samples in his Fig. 9.

The results of the present study, indicating predominantly kaolinite in the two samples from well 7119/12-2, together with Ramm’s determination of dickite in the two Nordmela samples from well 7119/12-1, suggest the presence of a kaolinite-dickite transition in the Troms area at depths somewhere between 1621 and 2820 m RKB, equivalent to 1415 and 2595 m, respectively, below the sea floor. Assuming 1-0-1-2 km of erosion, this gives a paleo-depth range for the transition of 2-4-3-8 km below the sea floor, which is consistent with the results from the other two areas studied (Fig. 10).

**PETROGRAPHIC OBSERVATIONS**

Kaolinite and dickite in the Garn and Statfjord Formations display certain morphological differences that are readily recognizable in thin-section and by SEM. In most samples, these differences could be used to make a preliminary identification of the dominant polytype...
Kaolinite to dickite transformation

Fig. 9. Statfjord Formation, Gullfaks and Gullfaks Sør Fields: plot of sample depths, showing the dominant kaolin polytype present in the <4 μm fraction. Samples are grouped by well, in order of increasing burial depth. Well symbols are identified in Table 1. The central part of the plot is expanded to show detail. Open diamonds = kaolinite; solid diamonds = dickite; half-filled diamonds = mixed kaolinite and dickite.

Fig. 10. Stø and Nordmela Formations, Troms area: plot of sample depths + 1.0 km (to correct for erosion of overlying section), showing the dominant kaolin polytype present in the <4 μm fraction. Samples are grouped by well, in order of increasing burial depth. Well symbols are identified in Table 1. Open diamonds = kaolinite; solid diamonds = dickite.

present, but several Statfjord Formation samples appear to provide exceptions to these generalities, as noted below. Despite differences in the optical properties of kaolinite (optically negative; maximum extinction angle 1–4°) and dickite (optically positive; maximum extinction angle 15–20°), small crystal size makes it impractical to use these differences to distinguish the two polytypes in thin-section. Therefore, the following descriptions necessarily refer to the dominant morphology observed in samples where bulk polytype identity was determined by IRS or XRD in the <4 μm fraction.

If it may indeed be assumed that the polytype identification from the <4 μm fraction is representative of the bulk sample, then the observation of consistent differences in crystal morphology between the kaolinite-dominated and dickite-dominated samples implies that the transformation from kaolinite to dickite occurs by dissolution and reprecipitation. The alternative mechanism of transformation in the solid state would not be expected to produce euhedral dickite crystals having a different morphology from the precursor kaolinite.

Kaolinite

A mixture of widely varying crystal sizes and morphologies is characteristic. The most conspicuous morphology is relatively large (20–40 μm wide by several hundred μm long) vermicular crystals, which typically form curving, semicircular forms (Figs. 11–13). By SEM, these vermicular crystals are seen to consist of disorderly-appearing stacks of thin euhedral plates (Fig. 13A).

The vermicular crystals occur both as isolated crystals and in clusters surrounded by smaller crystals with more blocky morphology (Figs. 11B and 12D). The blocky crystals typically appear similar to the vermicular crystals, except that the stacks are shorter in the c direction.

Very fine-grained masses of tiny (1–5 μm) crystals are another morphologic type
Fig. 11. Photomicrographs of sample 2451-65 from well 6507/7-4, showing typical morphologies of kaolinite as seen in thin-section. Analyses of the <4 μm fraction of these samples (Table 1) indicate that the kaolin present is either entirely or very dominantly the kaolinite polytype. Scale bars in A and B are 0.1 mm. C and D have the same magnification as B. A—A kaolinite cluster filling volume between quartz grains. The centre of the cluster, at left, illustrates the kaolinized mica morphology, consisting of tightly packed, radiating sheaves of coarse kaolinite (KM). Surrounding this core are microporous clusters of large vermicular crystals (V) and blocky kaolinite (B). B—Close-up of A, showing vermicular morphology. C—Close-up of A with polarizing filters crossed, showing kaolinized mica morphology. D—A kaolinite cluster consisting of a mixture of blocky to vermicular morphology (upper right) and extremely fine-grained kaolinite (centre). Such mixtures of varying kaolinite crystal sizes are common and possibly are inherited from earlier detrital material that has been altered to kaolinite. For example, the fine-grained material in this view could be derived from a dissolved lithic fragment, as suggested by the distribution of tiny opaque crystals (pyrite).
FIG. 12. SEM photomicrographs showing typical kaolinite morphologies. Analyses of the <4 μm fraction (Table 1) indicate that the kaolin present is either entirely or very dominantly the kaolinite polytype. A & B—Sample 2451-65, well 6507/7-4. The large vermicular crystals are most conspicuous (close-up in B), but areas of much smaller crystal size are also present (lower part of the cluster in A). C & D—Sample 2504-30, well 34/10-C-2. A cluster of relatively finely crystalline kaolinite seen at two magnifications.
Fig. 13. SEM photomicrographs showing typical kaolinite morphologies. Analyses of the <4 μm fraction (Table 1) indicate that the kaolin present is either entirely or very dominantly the kaolinite polytype. A—Sample 2886-60, well 6507/10-1. Coarse vermicules appear as disorderly stacks of thin, flat plates. B—Sample 2450-30, well 34/10-C-2. Masses of widely varying crystal size are characteristic. C—Sample 2476-70, well 6407/6-3. A cluster of large vermicules. D—Sample 3152-40, well 34/10-30. Vermicular crystals with rounded, ragged edges.
Kaolinite to dickite transformation

In some samples, notably those from well 34/10-C-2 in Gullfaks Field, much of the kaolinite occurs in altered (highly kaolinized), very fine-grained rock fragments having diffuse, disintegrating grain boundaries.

Kaolinized mica grains are yet another morphology, displaying all gradations between fresh mica grains with expanded edges and masses of tightly packed kaolinite sheaves reflecting complete kaolinization of the previous mica (Fig. 11C). Commonly the kaolinized mica grains are surrounded by clusters of vermicular and blocky crystals (Fig. 11A), giving the impression of short-range transport of reactant components outward from the mica into the immediately adjacent pore spaces.

It appears certain that the vermicular and blocky kaolinite morphologies all formed authigenically after burial of the sand; textures such as shown in Fig. 11 could not result from detrital sedimentation or infiltration. However, it is possible that some kaolinization of mica grains and rock fragments could have taken place before sand deposition.

Exceptions to the above generalities about the kaolinite-bearing samples are seen in several of the samples from Gullfaks Sør Field. Samples 3379.95, 3388.90, and 3389.70 from well 34/10-2 and samples 3244-65 and 3284-75 from well 34/10-30 contain dominantly kaolinite as determined by IRS, but most of the kaolin observed in thin-section has the morphology of well-formed, uniform booklets which is elsewhere characteristic of the kaolin in dickite-dominated samples, as described below.

Dickite

Perhaps the most striking characteristic of the kaolin clay in thin-sections of the dickite-bearing samples is the relative uniformity of crystal size within individual samples. In most cases, the maximum dimension of the kaolin crystals is $\sim 20-30 \mu m$ (Fig. 14A), but some samples have finer (10 $\mu m$) or coarser (maximum dimension 80 $\mu m$) crystals (Fig. 14B-D).

By SEM, the dickite crystals are seen to be more orderly appearing along the c axis direction than is typical of the kaolin in the kaolinite-dominated samples: in other words, the edges of the individual cleavage plates tend to be aligned, and there tend not to be large gaps between successive plates (Fig. 15). Typically, the crystals are elongated along the b axis direction, but the degree of elongation is variable (Fig. 15). The elongated crystal habit is commonly also visible in thin-section (Fig. 14B).

As noted above, thin-sections of several kaolinite-dominated samples from wells 34/10-2 and 34/10-30 contain mainly kaolin clay having the typical dickite morphology. It is thus uncertain that the morphology shown in Figs. 14 and 15 is exclusively associated with dickite. Therefore, care should be taken not to use morphology alone as a criteria for polytype identification.

Petrographic evidence for differences in reactivity between different morphological types

In thin-section, it is observed that virtually all of the dickite-dominated sandstones from the Garn Formation and some of the dickite-bearing Statfjord sandstones (wells 34/10-2 and 34/10-30) contain both (1) microporous clusters of booklets having uniform dickite morphology, and (2) kaolinized mica grains (Fig. 16A–B). The latter are suspected to consist of kaolinite which has been sluggish in reacting to form dickite, possibly because of larger crystal size and low internal microporosity. The vermicular texture so common in the kaolinite-dominated samples is rare in dickite-dominated samples, having been observed in
Fig. 14. Photomicrographs showing morphologies of kaolin in thin-sections of dickite-dominated samples. Analyses of the <4 μm fraction (Table 1) indicate that the kaolin present is either entirely or very dominantly the dickite polytype. Scale bar in A is 0.1 mm. A—Garn Formation, sample 3889-85, well 6407/4-1. This illustrates the most common crystal size and appearance of dickite. B—Garn Formation, sample 3997-60, well 6506/12-4, dickite with unusually coarse crystal size. C—Statfjord Formation, sample 3809-45, well 34/10-33, dickite with relatively fine crystal size. To the lower left are a rock fragment (R) and a highly expanded mica grain (M). The mica contains much open space between its expanded cleavage plates, but no kaolin is apparent. Expanded mica grains are abundant in this thin-section, but none seem to contain interleaved sheaves of kaolin. D—Statfjord Formation, sample 3313-05, well 34/10-30; another example of what appears to be very finely crystalline dickite (top) adjacent to an expanded mica grain lacking interlayered kaolin.
Analyses of the <4 μm fraction (Table 1) indicate that the kaolin present is either entirely or very dominantly the dickite polytype. A—Sample 3710-70, well 6407/1-2. Crystals are elongated in the a or b direction. B—Sample 3338-40, well 34/10-2. These crystals are somewhat smaller than in the other examples shown here. C—Sample 3784-30, well 6406/3-1. The samples from this well are unusual in containing abundant Fe-rich chlorite (top). D—Sample 3833-50, well 34/10-33.
Fig. 16. Photomicrographs showing details of kaolinized mica grains in thin-sections of dickite-dominated Garn sandstones. Analyses of the <4 \mu m fraction of these samples (Table 1) indicate that the kaolin present is either entirely or very dominantly the dickite polytype. Scale bar in A is 0.1 mm. A—Sample 3898-50, well 6506/12-3. A cluster of dickite (lower right) adjacent to a mica grain, the edges of which are highly expanded and altered to kaolin, possibly kaolinite that has been slow in reacting to form dickite. B—Same area as in A with polarizing filters crossed. C—Sample 3976-60, well 6506/12-4. A cluster of dickite (top) adjacent to a mass of highly birefringent, illitic clay, having a morphology indicating that it formed by illitization of kaolinized mica. Throughout this thin-section, the kaolin with the "dickite morphology" is non-illitized, whereas kaolinized mica exhibits varying degrees of illitization. D—Same sample as in C, showing an expanded, previously kaolinized mica grain, where the kaolin has been completely replaced by illite. Polarizing filters crossed.
Kaolinite to dickite transformation

only a few thin-sections. This relationship implies that the kinetics of the kaolinite-dickite
transformation are dependent upon the nature of the kaolinite starting material, with the
microporous clusters of booklets and vermicules being more reactive than the kaolinized
mica.

In the Statfjord samples, coexistence of dickite morphology and kaolinized mica
morphology is observed only in the thin-sections from wells 34/10-2 and 34/10-30. These
samples all occur near the apparent kaolinite-dickite transition depth. The deeper samples,
from wells 34/10-32A, 34/10-33, and 34/10-16, appear to contain only the dickite
morphology. Expanded mica grains are present in these thin-sections, but interlayered
kaolin appears not to be present between the disrupted mica layers (Fig. 14C–D). It may be
that these expanded mica grains were previously kaolinized, but the kaolinite has since been
dissolved and reprecipitated nearby as microporous clusters of dickite. The presence of
abundant kaolinized mica in the deeper, dickite-dominated samples from the Garn
Formation but not in the Statfjord Formation could reflect the different burial histories of
Haltenbanken and the Northern North Sea (Fig. 6). The latter area has been near its
maximum depth for tens of millions of years, whereas the Garn sandstones attained
maximum depth much more recently, and may not have had time for mineral reactions to
progress as far toward completion.

At a still more advanced stage of burial diagenesis, yet another type of contrast in
apparent reactivity is observed concerning illitization of kaolin in certain samples from the
Garn Formation. In this unit, extensive illitization of kaolin clusters is observed in samples
buried deeper than ~3.7 km below the sea floor (Ehrenberg & Nadeau, 1989). However,
near the top and bottom contacts of the formation, illitization appears to have been
inhibited due to local deficiency of K resulting from earlier extensive kaolinization of

Thin-sections of several samples from these kaolinite-rich leached zones were observed
to contain both microporous clusters of kaolin (shown by IR and XRD analysis to be
dickite) and subordinate amounts of kaolinized mica (inferred to be kaolinite). Throughout
these thin-sections, the kaolinized mica morphology tends to be extensively replaced by
illite, whereas the dickite clusters commonly appear entirely free of illitization
(Fig. 16C–D). This relationship implies that illitization of kaolinite is kinetically favoured
over illitization of dickite.

STABILITY CONSIDERATIONS

The data for kaolinite and dickite shown in Table 2 are found in a number of tabulations
(Robie & Waldbum, 1968; Robie et al., 1979; Haas et al., 1981; Berman, 1988) and are
based on the calorimetric studies of Barany & Kelley (1961) and King & Weller (1961).
Using these data, the stability of the kaolin minerals was recently discussed in a review
paper by Anovitz et al. (1991). They concluded that kaolinite is the stable polytype under all
gеologic P,T conditions, and that the formation of dickite in soils and during diagenesis is
only possible through nonequilibrium processes. However, it is hard to understand the
massive transformation of kaolinite to dickite documented in this paper to have taken place
outside the stability field of dickite.

We believe that this inconsistency probably reflects errors in the thermodynamic data.
The reference free energy difference between the two polytypes is small (Table 2) and
within the uncertainties of the data (Robie et al., 1979). Also, the purity of the materials
TABLE 2. Reference thermodynamic data for kaolinite and dickite from Robbie & Waldbaum (1968).

<table>
<thead>
<tr>
<th></th>
<th>$G_f$ (cal/mol)</th>
<th>$S$ e.u.</th>
<th>$H_f$ (cal/mol)</th>
<th>$V$ (cm$^3$/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kaolinite</td>
<td>-902868</td>
<td>48.53</td>
<td>-979465</td>
<td>99.52</td>
</tr>
<tr>
<td>Dickite</td>
<td>-902287</td>
<td>47.10</td>
<td>-979165</td>
<td>99.30*</td>
</tr>
<tr>
<td>difference</td>
<td>581</td>
<td>1.43</td>
<td>300</td>
<td>0.83*</td>
</tr>
</tbody>
</table>

Volume data marked with one asterisk have been calculated from cell parameters reported in Newham & Brindley (1956) and Newham (1961), while those marked with double asterisk are calculated from the more recent structural determinations of Vish & Von Dreele (1988) and Joswig & Drits (1986).

used in the old calorimetric determinations is questionable. We therefore suggest that new enthalpy and heat capacity determinations are needed for the kaolin polytypes. High P,T phase equilibria involving kaolinite should be reevaluated, both with respect to thermodynamic data retrieval and examination of the kaolin polytypes present.

The molar volumes of the two polytypes are also puzzling. The value of 99.52 cm$^3$/mol for kaolinite has remained unchanged in thermodynamic data bases since reported by Robie & Waldbaum (1968), and it is unclear where this value originated in the first place. We have calculated a molar volume of 98.47 cm$^3$/mol (Table 2), based on lattice parameters reported in Newham & Brindley (1956) and Newham (1961). This value shows better general agreement with the densities given by Deer et al. (1966), which indicated kaolinite to be the more dense of the two polytypes. However, recent structural refinements of both kaolinite (Bish & Von Dreele, 1988) and dickite (Joswig & Drits, 1986) gave molar volumes of 99.35 and 98.58 cm$^3$/mol, respectively (Table 2).

In view of the apparently small free energy difference between kaolinite and dickite at 25°C and 1 bar, it is appropriate to enquire whether surface energy contributions might play a role in helping to stabilize dickite relative to kaolinite. The SEM photomicrographs clearly demonstrate that the dickite crystals, with their blocky morphology, have smaller surface to volume ratios than the thin plate-like crystals comprising the vermicular kaolinite booklets (Figs. 12, 13 and 15). Approximate dimensions of such crystals are given in Table 3. Generally the molar surface energy is given by (symbols explained in Table 3):

$$G_s = 2/3\gamma S$$

Assuming equal sized particles, the surface energy contribution to the local Gibbs free energy is expressed as:

$$G_s = 2/3\gamma Ns$$

The values for this contribution (Table 3) are indeed small compared with the reported Gibbs free energy difference (581 cal/mol), so it may be concluded that surface energy is not a significant factor in the transformation of kaolinite to dickite.

Given the uncertainty in the thermodynamic data, the natural occurrences of kaolinite and dickite can provide an indication of their stability regimes in pressure/temperature space. The diagram in Fig. 17 is consistent with (1) the assumption that kaolinite is the
Kaolinite to dickite transformation

Table 3. Approximate particle dimensions based on SEM photomicrographs and calculated values for the surface energy contribution to the total Gibbs free energy.

<table>
<thead>
<tr>
<th></th>
<th>s</th>
<th>v</th>
<th>N</th>
<th>ΔG_s (erg)</th>
<th>(J)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kaolinite</td>
<td>3280</td>
<td>800</td>
<td>1.25 x 10^{11}</td>
<td>5.12 x 10^7</td>
<td>5.1</td>
</tr>
<tr>
<td>Dickite</td>
<td>522</td>
<td>720</td>
<td>1.39 x 10^{11}</td>
<td>27.3 x 10^7</td>
<td>27.3</td>
</tr>
</tbody>
</table>

Symbols:

γ = surface energy (a value of 100 erg/cm^2 is assumed for the surface tension of solid-water interfaces.)
S = surface area per mole
s = surface area per particle (μm^2)
v = volume per particle (μm^3)
N = number of particles per mole
ΔG_s = surface energy contribution to the total Gibbs free energy

Fig. 17 A summary of the available constraints on the conditions of stability of kaolinite and dickite. Ruled fields represent dickite occurrences. The slope of the univariant boundary between kaolinite and dickite fields is calculated from current enthalpy and volume data. The position of the boundary is constrained by the lowest-temperature of Norwegian shelf dickite occurrence, but the actual position may be at lower temperature, depending on the degree of kinetic overstep needed to initiate kaolinite-to-dickite reaction in Norwegian shelf sandstones.

stable polytype at Earth-surface conditions as indicated by its occurrence as a residual weathering product and in low-temperature diagenetic settings, and (2) the exclusive occurrence of dickite either in high-temperature hydrothermal systems or under medium-to high-grade diagenetic conditions. The appearance of pyrophyllite will define an upper temperature limit to the dickite field at ~300°C. The position of the univariant boundary is limited on the high-temperature side by the first occurrence of dickite at ~120°C in Gullfaks Sør Field and at ~130°C in Haltenbanken. However, the boundary could be located at a somewhat lower temperature, depending on the degree of kinetic overstep (Ridley & Thompson, 1986) needed to form dickite from kaolinite. The slope of the boundary is uncertain, but has been drawn as 78 bar/K in Fig. 17, according to the current entropy (Robie & Waldbaum, 1968) and volume (Bish & Von Dreele, 1988; Joswig & Drits, 1986) data.
Transformation of kaolinite to dickite with increasing burial depth has been observed in sandstone cores from three areas on the Norwegian continental shelf. In the Gullfaks Sør Field, the transition corresponds to a present rock temperature of ~120°C, which is consistent with the more loosely constrained data from the Haltenbanken and Troms areas. Observation of this transformation in all three cases so far examined indicates that it may be a general and predictable feature of kaolin-bearing sandstones worldwide and therefore a potentially reliable paleogeothermometer.

Infrared spectroscopy is recommended as the most sensitive technique for differentiating kaolinite and dickite in sandstones, where large amounts of non-clay minerals are present, and for estimating relative abundances of the polytypes under these circumstances.

Although exceptions to the rule are noted, the kaolinite typically occurs as relatively large vermicular crystals intermixed with clusters of very fine-grained crystals and kaolinized mica, whereas the dickite forms more euhedral, blockier crystals having much more uniform texture throughout individual samples. This morphological difference, together with the nature of the structural difference in octahedral occupancy between kaolinite and dickite, suggests that the transformation occurs by dissolution and reprecipitation, rather than in the solid state.

A very similar morphological difference between early vermicular and later blocky “kaolinite” has been described repeatedly in diagenetic studies of North Sea sandstones, but without identification of the kaolin polytypes involved (Hurst & Irwin, 1982; Kantorowicz, 1984; Thomas, 1986; Glasmann et al., 1989; Haszeldine et al., 1992; Giles et al., 1992). Polytype identification in such studies is needed to determine whether these two commonly reported morphologies in fact reflect kaolinite and dickite occurrence, respectively.

Patterns of coexistence of different morphological types of kaolin observed in thin-section can be used to infer contrasting degrees of reactivity between various kaolin morphologies. Specifically, microporous clusters of kaolinite crystals may react more readily to form dickite than sheaves of kaolinite in kaolinized mica, and, at a more advanced stage of diagenesis, the kaolinized mica may react more readily to form illite than coexisting dickite.

In many previous mineralogical studies, the term kaolinite has been used to refer loosely to all kaolin-group clays, where the specific polytypes present have not been identified. It is recommended that this longstanding convention should now be abandoned and that the term “kaolin” or “kaolin-group clay” be used for this purpose. The polytype names, kaolinite and dickite, should be used only where identification has been confirmed by a reliable analytical technique. We note further that neither the general morphological differences between kaolinite and dickite described in this paper nor the correlation of polytype occurrence with depth should be regarded as a suitable basis for reliable polytype identification without confirmation by IRS, XRD, or differential thermal analysis.

ACKNOWLEDGMENTS

Håkon Rueslåtten of Statoil’s Trondheim Research Lab is thanked for his advice and help during the early stages of this project. Eli Mydland Carlsen contributed the burial history curve for Gullfaks Sør Field used in Fig. 6. We thank Statoil and the partners of the numerous production licenses involved for permission to release this study.
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