MIXED-LAYER ILLITE/SMECTITE DIAGENESIS IN
UPPER JURASSIC CLAYSTONES FROM THE NORTH
SEA AND ONSHORE DENMARK

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ABSTRACT: Claystones of Kimmeridgian-Volgian(-Ryazanian) age are the main source
rocks for oil in the Central Trough, North Sea, but they are too immature for oil generation in the
Norwegian-Danish Basin. This paper describes the diagenesis of mixed-layer illite/smectite
(I/S) in these source rocks as investigated by X-ray diffraction and high-resolution electron
microscopy, interpretation for both methods being supported by computer simulations.
Randomly-ordered I/S with ~70% illite layers, together with discrete illite, kaolinite, and traces
of chlorite and vermiculite, are detrital. The clay mineralogical data suggest the existence of an
island at Gassum during part of the Upper Jurassic. Mixed-layer illite/smectite originating from
volcanic ash is found in two wells in the Central Trough. The volcanism may be related to a
period of stretching in the North Sea graben system during Jurassic-Lower Cretaceous time.
Diagenesis has not altered the I/S in the Norwegian-Danish Basin. In the Central Trough,
however, the amount of illite layers in I/S increases from ~70% to ~90% and the I/S becomes
ordered. The volcanic clays have been changed to randomly ordered I/S with ~50% illite layers.
These changes occurred in the Central Trough simultaneously with oil generation.

Drilling for oil and gas has taken place in the Danish onshore area for fifty years and offshore
in the North Sea since 1968. Commercial hydrocarbon accumulations have not been found
onshore but, in the Central Trough, several wells sourced from Kimmeridgian-Volgian
(–Ryazanian) claystones are producing from Chalk reservoirs. The claystones in the Central
Trough are rich in organic carbon of algal origin, especially in the northern part of the area.
They are pre-mature to mature in the southern and mature in the central and northern
Central Trough (Thomsen et al., 1983). In the Norwegian-Danish Basin the equivalent
claystones are immature, have a lower content of organic carbon, and the organic matter is
more terrestrial in origin than in the Central Trough (Thomsen et al., 1983).

The sedimentology and diagenesis of the Upper Jurassic mixed-layer clay minerals have
been investigated previously by X-ray diffraction (XRD) and chemical analysis. Pearson et
al. (1982) attributed ordering of mixed-layer illite/smectite (I/S) in the Viking Graben to
diagenesis. However, Hurst (1982) and Scotchman (1987) reported the occurrence of ordered
I/S in shallow buried samples from the Moray Firth and from the onshore UK Kimmeridge
Clay Formation, respectively. Dypvik (1983) found an increase in percentage illite layers in
I/S with increasing temperature of the Mesozoic rocks from the Viking Graben and the
Central Trough, but he did not investigate the ordering of the I/S.

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Transmission electron microscopy (TEM) of I/S has given rise to a continuing discussion about the structure and the formation mechanism of I/S. Nadeau et al. (1984) introduced the concept of fundamental X-ray-diffracting particles which could be observed by TEM of fully dispersed clay samples. The I/S XRD spectra were explained as a result of interparticle diffraction. This led Nadeau et al. (1984) to propose a neoformation mechanism for the smectite-to-illite transformation. Ahn & Peacor (1986a) did not find mixed-layers of illite and smectite in a high-resolution TEM (HRTEM) study of the smectite-to-illite transformation in bulk rock claystone samples from the US Gulf Coast; neither did they find fundamental particles. This is in contrast to the XRD findings of increasing ordering with depth in I/S from the same clay formation (Perry & Hower, 1970) and to the neoformation theory of Nadeau et al. (1984).

This paper describes the results of an investigation into I/S diagenesis in time-equivalent Upper Jurassic claystone formations in the Central Trough of the North Sea and the Norwegian–Danish Basin. XRD and HRTEM were used for analysis and computer simulations were applied to support the interpretations. The purpose of the investigation was to assess the variability of I/S parent material and also the relationship between maturity of the organic matter and oil formation on the one hand and I/S diagenesis on the other.

**MATERIALS**

Material from the Mandal, Farsund, and Haugesund Formations in the Central Trough, North Sea, was available as cuttings from the Danish wells Adda 1, E1, G1, II, Lulu 1, M8, U1, and W1, and from the Norwegian wells 2/7-3 and 2/11-1, and as core material from E1 and 2/11-1. Material from the Børglum member of the Bream Formation was available as cuttings from the Danish wells Aars 1, F1, Hyllebjerg 1, J1, Mors 1, and Rønde 1, and as core material from Børglum 1, Frederikshavn City 1, Gassum 1, Haldager 1, and Uglev 1. Material was available as cuttings from the Saude and Tau formations in the Norwegian well 9/4-3. Locations of the wells are given in Fig. 1. The cuttings were washed in distilled water and dried at room temperature. Rock fragments of dark-grey colour, typical of the formations studied (Vollset & Doré, 1984), were selected for analysis.

**METHODS**

*X-ray diffraction*

All samples were gently crushed to pass a 0.125 mm sieve. Calcite was removed by treatment with sodium acetate/acetic acid at pH 5.5 and 100°C, and organic matter by a subsequent treatment using sodium hypochlorite at pH 9.0 and 100°C (Anderson, 1963). Fe- and Al-oxyhydroxides were then removed by Na-dithionite/bicarbonate/citrate at pH 7 (Roth et al., 1969); about 100 mg of dithionite was used per g of sample. Sand and silt fractions were recovered by conventional centrifugation or by elutriation, and the fine clay (<0.2 μm) and coarse clay (0.2–2 μm) fractions were separated in a continuous-flow centrifuge.

Even in the fine-clay fraction, however, discrete illite was present in appreciable amounts, as demonstrated by the d(002) spacing at 5.0 Å in Fig. 2a. Kaolinite was also present in this fraction. The presence of discrete illite in shales compromises the interpretation of mixed-layer minerals in such rocks (Nadeau & Reynolds, 1981). Therefore, the I/S was isolated by a modification of the procedure of Buzagh & Szepesi (1955) (as used by Gibbs (1967)) for the
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FIG. 1. Location of wells examined. (U) indicates wells with a large kaolinite/illite peak area ratio (> 4). Dotted line shows the Gassum Island as drawn by Larsen (1966).

separation of montmorillonite: 1 g of the air-dry, Na⁺-saturated clay was added to 2 ml of 96% ethanol. The paste was left to stand for about one hour and then dispersed in 100 ml 15% ethanol by sonic vibration (20 kHz, 5 s). After 20 min centrifugation at 1600 g the suspension was removed and the precipitate re-dispersed by sonic vibration in 80 ml 15% ethanol. The centrifugation and removal of suspensions were repeated twice. The combined suspensions were made 1 M in NaCl and the clay flocculated. The water–ethanol solution was then removed by centrifugation. From the clay isolated in this way—the I/S fraction—excess NaCl was removed by washing with 96% ethanol. The washed clay was treated by this suspension-flocculation procedure two further times. Less than 10% of the fine-clay fraction was removed during the last separation treatment. The I/S fraction obtained from the last suspension constituted about two-thirds of the fine-clay fraction. The remaining part of the fine-clay fraction was collected by combining and air-drying the sedimented clay from the different ethanol centrifugations.

Diffactometer mounts were prepared by the pipette method with 2-5 mg sample per cm². XRD was carried out with Cu-Kα radiation using a Philips goniometer PW 1771/00 with a 2θ variable divergence slit and monochromator. The specimens analysed were: Mg²⁺- and K⁺-saturated air-dried specimens; Mg²⁺- and Na⁺-saturated glycolated specimens; and K⁺-saturated specimens heated for 1 h at 250°C. The illite percentages and degrees of ordering in I/S were obtained by visual comparison with computer-simulated diffraction traces. Traces of the Mg²⁺-saturated specimens were computer-simulated with the NEWMOD program provided by R. C. Reynolds for two-component mixed-layering. Traces of the Mg²⁺-saturated, glycolated specimens were also computer-simulated with the program of P. D. Cradwick (Cradwick & Wilson, 1978) for three-component mixed-layering.
High-resolution electron microscopy

Six samples were selected from six different wells at different depths: Børglum 1 (986 m), M8 (2387 m), E1 (3444 m), 2/11-1 (3877 m), I1 (3907 m), and 2/7-3 (4178 m). The preparation of electron-transparent thin foils included the following steps: (i) embedding of small pieces of rock in resin, (ii) cutting thin slices perpendicular to the bedding plane, (iii) grinding to ~100 μm in thickness, (iv) argon beam sputtering, (v) coating with carbon to prevent charging in the microscope.

This procedure was chosen for two reasons. First, the thickness of the individual crystallites as well as their distribution in the original rock is maintained; secondly the main part of the crystallites is oriented with the crystallographic c-axis perpendicular to the electron beam. This geometry is required for imaging (001) lattice fringes. (For details see Iijima & Buseck (1978) and Veblen (1983a,b)). In this study a Philips 430 TEM with a 300 kV acceleration voltage was used.

RESULTS AND DISCUSSION

X-ray diffraction

The result of applying the ethanol procedure to the fine-clay fraction is demonstrated in Fig. 2. The diffraction trace of the total fine-clay fraction is shown in Fig. 2a. I/S peaks are seen at 16, 9-8, 8-8, 5-5 and 3-34 Å. In addition, minor amounts of kaolinite (peaks at 7-1 and 3-55 Å) and illite (peaks at 9-8 and 5-0 Å) are present.

![Fig. 2. XRD of Mg²⁺-saturated and glycolated oriented specimens from U1, 2716 m. (a) Total, fine-clay fraction (<0-2 μm). (b) Sedimented fraction. (c) I/S fraction. Cu-Kα radiation.](image)
Peaks due to illite, kaolinite and quartz are enhanced on the trace of the sedimented fraction (Fig. 2b) and significantly depressed on the trace of the I/S fraction. The peaks at 9.8 Å and the shoulder at 5 Å are due to illite segregation (Fig. 2c)—although an exception to this behaviour was shown by material from wells Gassum 1 and Rønde 1, where the I/S fractions were dominated by kaolinite. The ethanol procedure therefore seems to have been effective in removing most of the illite and kaolinite, although poorly crystalline kaolinite may remain in the I/S fraction and may dominate it when only little I/S is present in the sample. Some of the I/S is, however, removed from the I/S fraction by the procedure.

The I/S fractions from Rønde 1 and Gassum 1 were not computer-simulated because they were dominated by kaolinite. Satisfactory NEWMOD computer simulations were obtained for I/S fractions from all other samples except G1, 3441 m and Aars 1, 2425 m and 2465 m. Except for these three samples, the results obtained from computer simulations from glycolated specimens and from air-dry specimens agreed for each sample within 5%. For the Mg$^{2+}$-saturated, glycolated specimens of the samples G1, 3441 m, and Aars 1, 2425 m and 2465 m, the $d$-value of the (002)$_{\text{illite}}/(003)_{\text{smectite}}$ composite peak maximum was significantly lower than would have been expected from the two-component mixed-layer simulation fitted to the 4–10°2θ region. Therefore the three-component program was applied, and this produced a peak pattern in better agreement with the experimental data both in the low-angle region (4–10°2θ) and in the composite peak region (15–20°2θ). The third mixed-layer component of this sample is not chlorite, as the diffraction traces of the K$^+$-saturated specimens showed only a broad 10 Å peak in the 4–10°2θ region. As the third component was modelled with a $d$(001) of 14 Å in glycolated specimens, it was assumed to be vermiculite or a smectite with only one glycol layer between the 2:1 units.

Random ordering of I/S (with $d$(001) smectite peaks at about 17 Å) was found in the samples from the Norwegian–Danish Basin (Fig. 6) and in the samples from shallow depth in the Central Trough (Fig. 7, E1 (2983 m)). Ordered I/S was found in the more deeply buried samples in the Central Trough. R1 ordering (seen from a $d$(001) at ~27 Å and a $d$(002) at ~14 Å) was predominant in ordered samples (Fig. 7, E1 (3444 m and 3938 m)), whereas R2 ordering (seen from a $d$(003) at about 12 Å) could be present in minor amounts in some R1 ordered samples (Fig. 8, 2/7-3 (4178 m)). R3 ordering (with a shoulder at ~11 Å) was probably present in one sample (Fig. 8, 2/7-3 (3789 m)). Three deeply buried samples (W1, 3816 m; 2/7-3, 3365 m and 3502 m; see Fig. 8) contained randomly ordered I/S. This is discussed later.

XRD patterns from a mixture of thin illite particles (eventually mixed with very thin smectite particles) resemble patterns from mixed-layer particles with both illite and smectite layers (MacEwan crystallites) (Nadeau et al., 1984). Therefore, XRD cannot be used without ambiguity to determine whether mixed-layer particles are present in the XRD specimens. Moreover, the dispersion and pretreatment prior to XRD can disperse mixed-layer particles along smectite layers and thereby result in a mixture of very thin illite and smectite layers. Therefore, mixed-layering in the intact bulk rock must be investigated. In the present investigation this was carried out by HRTEM.

High-resolution electron microscopy

Mineral identifications were partly based on the criteria of Ahn & Peacor (1986a). Packets 5–10 nm thick showing straight and relatively defect-free 10 Å lattice fringes and clear and
well-defined 10 Å reflections in selected-area diffraction (SAD) were described as illite. Such illite packets dominated in the six samples studied in the present investigation (Fig. 3a,b and c). Collapse of smectite layers to a (001) of 10–14 Å in the vacuum of the electron microscope make their identification difficult (Ahn & Peacor, 1986a). However, Ahn & Peacor (1986b) observed extended regions of 20 Å lattice fringes in a K-rectorite. They interpreted this as a regular sequence of illite and collapsed smectite and the 20 Å contrast as due to alternating chemical composition in the interlayers of the mineral (i.e. Na in the smectite and K in the illite interlayer). Lattice fringes of enhanced contrast are observed in Fig. 3d and e. In Fig. 3d, a regular series of enhanced lattice fringes, with a 20 Å periodicity, is observed. Similar images were obtained from all samples except Børglum 1, 986 m. A random distribution of strong lattice fringes is observed in Fig. 3e. We have, to guide the interpretation of the lattice fringe images, made a calculation of the image contrast expected from a model structure of the I/S mineral (Fig. 4). We have chosen an ideal 1M mica structure as the model and have substituted Na for K in every second interlayer. A standard computer program (O'Keefe, 1973) based on the multi-slice method (Cowley & Moodie, 1957) has been used to calculate the lattice fringe pattern for different crystal thicknesses. The results show (Fig. 4) that alternating interlayers of K and Na can produce a 20 Å period in lattice fringe contrast from I/S (collapsed) minerals.

It is outside the scope of this work to give a full discussion of the interpretation problems in lattice imaging of interstratified minerals. We will mention, however, that the 2M mica polytype under special imaging conditions will show a 20 Å periodicity also (Iijima & Buseck, 1978).

The calculations of Fig. 4 and lack of intensity of XRD reflections corresponding to the 2M mica polytype support the interpretation of Fig. 3d that the 20 Å periods can be attributed to a regular series of alternating illite and collapsed smectite layers. We take this as an indication of the existence of ordered I/S in the Central Trough claystones. In extension of this argument we interpret the structure in Fig. 3e, where 10, 20, 30 and 40 Å spacings occur together, as a random distribution of smectite layers in a large crystal. It has not been possible to assign an ordering parameter to the samples based on HRTEM alone because of difficulties in identifying all smectite layers and the time-consuming HRTEM imaging technique. Therefore, a quantitative comparison between XRD and HRTEM has not been possible.

An alternative interpretation of the lattice-fringe images is possible, however. Fig. 3d can be seen as a stacking of fundamental 20 Å illite particles and Fig. 3e as a stacking of 20, 30 and 40 Å thick fundamental particles.

Finally, lattice fringes images of chlorite and mixed-layer chlorite and illite are observed in M8 (2387 m) (Fig. 3f and g).

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**Fig. 3.** (a) M8, 2387 m. Packets of illite showing straight and defect-free 10 Å lattice fringes. The electron diffraction pattern (b) shows a systematic row of clear, well-defined 10 Å reflections used for the one-dimensional lattice image. (c) Børglum 1, 986 m. Detrital illite packet from a shallow depth in the on-shore well. (d) 2/11-1, 3877 m. 20 Å periodicity of enhanced contrast in the lattice fringes is interpreted as an IS-ordered mineral. (e) M8, 2387 m. Enhanced 20, 30 and 40 Å lattice-fringe contrast in randomly-ordered I/S. (f) M8, 2387 m. 14 Å lattice fringes from chlorite. (g) M8, 2387 m. Randomly mixed 14 and 10 Å fringes from a mixed-layer illite-chlorite mineral.
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\[ \text{Na K Na K Na K K K K} \]

\[ t \]

\[ 51A \]

\[ 123A \]

\[ 185A \]

\[ 247A \]

\[ 20A \quad 10A \]
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Discrete clay minerals

Apart from the I/S fraction, traces of chlorite were found by XRD in all fractions of the samples from the Norwegian–Danish Basin except in the wells Gassum 1 and Rønde 1. In the Central Trough, chlorite was detected in the wells M8, G1, U1, and W1, but not by XRD in the deepest samples with the highest vitrinite reflectances and degrees of ordering in I/S, such as in wells 2/11-1, 2/7-3, and E1. Therefore, significant diagenetic chlorite formation has not taken place in the area.

Illite was found in all coarse clay fractions and may largely be considered detrital, because of its grain size and occurrence in all samples irrespective of degree of diagenesis. It was probably derived by erosion of lightly weathered source rocks.

Kaolinite was found in all fractions and may be considered detrital, resulting from erosion of land surfaces which have undergone weathering in a semi-humid tropical climate. Sellwood (1972) found that the dominant clay minerals in Lias shales from the UK are illite and kaolinite. He concluded that these minerals probably reflect the nature of the source areas in the region and that decreasing kaolinite contents in these sediments probably reflect greater distance from source areas. Bjørlykke et al. (1975) found in Well 2/11-1 an average increase in the kaolinite/illite peak area ratio of about 3 from 4100 to 3500 m (the kaolinite/illite peak ratio being the ratio between the areas of the (001) peaks of the two minerals). They suggested that this increase reflected a higher degree of proximity to deltaic or shoreline facies in a regressional sequence. Alternatively, they proposed that the increase could have been due to a climatic change towards more humid conditions at the end of the Upper Jurassic.

In the Central Trough samples, kaolinite/illite peak area ratios were found in general to be low. Therefore, according to the above arguments, the sediments here are offshore facies. In the samples from the Norwegian–Danish Basin, the kaolinite/illite peak area ratios are low in the wells 9/4-3, Aars 1, Hylebjer 1, Haldager 1, and Børglum 1 (<2), high in the wells J1, F1, and Frederikshavn City 1 (~4), and highest in Gassum 1 and Rønde 1 (>5). Apart from the highest values in Gassum 1 and Rønde 1, the variation in the kaolinite/illite peak area ratio in the Norwegian–Danish Basin can be explained by an influx of detrital material from the Fennoscandian Shield, as the wells closest to the Shield have the highest kaolinite/illite peak area ratios. A climatic change is a less likely explanation for this variation, as the sampled formations are time equivalent.

The kaolinite/illite peak area ratios are very high in the coarse clay fractions of the wells Rønde 1 and Gassum 1. The kaolinite peaks at 7-4 Å are broad in the diffraction traces of these wells and the samples have a high kaolinite content and a low I/S content in all fractions (Fig. 5). This high kaolinite content in all fractions corresponds to the soil-type clay XRD pattern of Hughes et al. (1985) for clays from Pennsylvanian strata of the Illinois Basin.

![Fig. 4. Multi-slice calculations of lattice-fringe intensity for a model of I/S. The top images show crystal structure models in [100] projection. To the right a 1M mica model with K in all the interlayer positions. To the left a model of the I/S mineral with alternating interlayers occupied by Na and K. Lattice-fringe images have been calculated for the two structure models for different choice of crystal thickness, t, along [100]. The thickness is given to the left. The column to the left represents the results for the I/S model. It is seen that a 20 Å periodicity is produced for crystals 50–250 Å thick. The ideal mica is represented to the right. Calculations for this model give 10 Å periodicity exclusively. (All the images presented are calculated for a defocus value of -720 Å, corresponding to Scherzer focus for the microscope used.](image)
Hughes et al. found that this clay assemblage resulted from soil processes during deposition. Moreover, one of the samples from Gassum 1 was mottled red and XRD showed significant amounts of hematite. Therefore, the clays from Gassum 1 and Rønde 1 must have originated from soil weathering in emergent sediments. During the Upper Jurassic, sediments were supplied to the Danish sub-Basin from Precambrian rocks and Mesozoic sediments in the Fennoscandian Shield to the north and the Ringkøbing–Fyn High to the south. The red kaolinitic soil clays in Gassum 1 must have formed in situ or have been transported over a short distance. They may have originated from weathered Lower Jurassic or Triassic sediments on the Ringkøbing–Fyn High. However, these soil clays were only found in the wells Gassum 1 and Rønde 1. Therefore, the clay mineralogical results support the finding of Larsen (1966) that a ‘Gassum Island’ (see Fig. 1) existed during part of the Upper Jurassic.

**Diagenesis of I/S**

The compositions of I/S in onshore wells are illustrated in Fig. 6. The I/S from Frederikshavn City 1, 649 m, has 70% illite layers and, from the presence of the 16 and 10 Å peaks and the absence of peaks from ordered phases, is randomly ordered. Identical results were obtained from Børglum 1, 986 m, and Hylebjerg 1, 1850 m, and from the other wells in the Norwegian–Danish Basin, except Gassum 1 and Rønde 1. Comparison between formation temperature and % illite component in Central Trough I/S shows that randomly ordered I/S with 70% illite is found at temperatures of ~100°C. This is in agreement with the
results from the US Gulf Coast (Perry & Hower, 1970; Hower et al., 1976) but it is far in excess of the values of ~70°C attained in the deepest sections of the formations in the Norwegian–Danish Basin and 25°C of the Frederikshavn City 1 well. Therefore, the randomly ordered I/S in the Norwegian–Danish Basin must be detrital, having been weathered and transported from topographic highs in the area.

For most wells in the Central Trough, the I/S depth pattern is illustrated by well E1, Fig. 7. Randomly-ordered I/S with ~70% illite layers is probably of detrital origin, in agreement with the results from the Norwegian–Danish Basin. With increasing depth the illite content increases up to ~90% illite in I/S and this becomes IS-ordered, as seen from the d(001) at ~27 Å and the d(002) at ~13 Å. Minor amounts of discrete illite are assumed to be present in the I/S fraction from E1, 2983 m, from the presence of the d(002) peak at 5-0 Å.

An anomalous I/S depth pattern for the Central Trough is found in the wells 2/7-3 (Fig. 8) and W1. Here, randomly-ordered I/S with 40–50% illite is found at depths (3365 and 3502 m in 2/7-3 and 3816 m in W1) and at temperatures where ordered I/S with ~90% illite layers is
found in other wells. Ordered I/S is also found in these wells just a few hundreds of meters deeper. The samples with 40–50% illite layers are dominated by these mixed-layers. A bentonitic origin for these samples could explain the high content of I/S. Microscopic investigations of the silt fractions has indicated a volcanic origin for at least some of the material in these samples (Lindgreen et al., in prep.). A volcanic origin conforms with the lower illite percentages and degrees of ordering attained, as discussed for bentonites by Nadeau & Reynolds (1981). A Kimmeridgean–Volgian(-Ryazanian) eruption of volcanic ash conforms to the Middle to Upper Jurassic volcanic event previously reported from the Auk Field, just 100 km to the west of 2/7-3 (Howitt et al., 1975). Knox & Fletcher (1978) investigated samples from the Ryazanian part of the Yorkshire Speeton Clay and found four beds of volcanic clay in this formation. Jeans et al. (1982) described Ryazanian to Albian volcanic clays from southern England and found indications for an easterly source probably in the southern North Sea area. Dixon et al. (1981) found two sequences of igneous rocks of earliest Cretaceous age in well 29/25-1 at the edge of the Mid North Sea High, very close to the Auk Field. They summarized data for igneous activity in the North Sea Basin and
concluded that the Jurassic phase of stretching activity in the North Sea Graben System resembles early Miocene activity in the Kenyan segment of the East African Rift, except that the erupted volumes in the North Sea are negligible by comparison with those in Kenya. Volcanic clays in wells 2/7-3 and W1 may originate partly from volcanic ash deposited on adjacent highs (the Mid North Sea High and the Mads High, respectively), and then washed out into the basins by a transgressing sea. The probably volcanic clays are not apparent on the Schlumberger logs and may be present between the sample intervals in the other wells investigated.

Proportions of illite layers in I/S are plotted against published vitrinite reflectance measurements (Thomsen et al., 1983; Thomsen, 1984; Schmidt, 1985) in Fig. 9. The immature onshore well sections from the Danish sub-Basin all contain detrital, randomly ordered I/S. In the Central Trough, this detrital I/S becomes ordered at vitrinite reflectances of ~0-65%, i.e. at oil generation, and the illite content increases during peak oil generation up to 95% illite layers. Unfortunately, vitrinite reflectances have not been published from the wells 2/7-3 and W1, where the volcanic clays are found. However, by inference from
formation temperatures, the volcanic I/S is still randomly ordered with 40–50% illite layers at peak oil generation. These results resemble in part those of Pearson et al. (1982) who found ordered I/S with ~80% illite layers in the Upper Jurassic of Viking Graben associated with vitrinite reflectances of ~0.60% at ordering. Furthermore, in the Moray Firth Basin they found randomly-ordered I/S in the Jurassic sediments associated with much lower organic maturation levels. Hurst (1982) found randomly-ordered I/S with 40–50% illite in Jurassic shales from Brora, NE Scotland, and suggested a detrital origin. He furthermore related IS-ordered I/S found in the same area with I/S of the same composition in Devonian parent rocks. A detrital origin is not acceptable for the ordered I/S of the present investigation because ordered I/S is absent at low formation temperatures in the Central Trough. At variance with our results, Scotchman (1987) reported the occurrence of ordered I/S in the onshore Kimmeridge Clay Formation, UK, in organically immature samples, and proposed it to be authigenic in origin. Discrete illite seems to be present in Scotchman’s diffraction traces of <0.5 μm fractions, and discrete illite may compromise the interpretation of XRD patterns (Nadeau & Reynolds, 1981). However, if present, ordered I/S in the Kimmeridge Clay can be detrital, as ordered I/S may form during weathering (Eberl, 1984).

Subsidence took place in both the Norwegian–Danish Basin and the Central Trough during the Upper Jurassic. The Børglum Member claystone of the Norwegian–Danish Basin has a maximum thickness of 120 m in the centre of the Danish sub-Basin (Michelsen, 1978).
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The equivalent Farsund Formation in the Central Trough attains in wells a thickness of more than 1400 m (Jensen et al., 1986). In the Central Trough, large thickness differences may be due to differential subsidence.

Subsidence continued in both the Central Trough and the Norwegian–Danish Basin during the Cretaceous. The subsidence ceased in the Danish sub-Basin after the Paleocene, but increased in the Central Trough where more than 2000 m of marine clays were deposited. According to Madsen (1974) the approximate geothermal gradients for Danish onshore wells and Central Trough wells are 23°C/km and 29°C/km, respectively. However, a high gradient of 42°C/km is calculated for the Lower Cretaceous and Jurassic shales in the Central Trough (Jensen, 1982). The present-day temperatures of the Upper Jurassic claystones are 25°–70°C in the onshore Danish wells and 75°–150°C in the Central Trough. The larger depths of burial and the higher geothermal gradients in the Central Trough have resulted in higher maturities of the organic matter, higher illite percentages, and the I/S ordering in the Central Trough.

Hower et al. (1976) proposed that Fe, Mg and Si released from the illitization of smectite layers formed chlorite. In the present investigation, diagenetic chlorite formation was not found to be significant (based on XRD observations). As the dominant detrital I/S material in most samples had ~ 70% illite layers, little Fe, Mg and Si should be released as a result of the increase in illite content up to 85–95%. Consequently, only minor amounts of chlorite should form, which is in agreement with the observed results. This is clearly different from the Gulf Coast Tertiary, where the detrital I/S with only 20% illite layers is transformed into an I/S with 80% illite at greater depth. This increase is three times the increase in the Central Trough, and the resulting larger release of Fe, Mg, and Si in the Gulf Coast samples may explain the diagenetic chlorite formation there.

CONCLUSIONS

Randomly-ordered and ordered I/S were detected both by XRD on pretreated and isolated I/S and by HREM on bulk rock specimens. Computer simulations proved that ordered I/S will produce a 20 Å periodicity as seen in HREM.

Kaolinite, illite, randomly-ordered I/S with ~ 70% illite layers and traces of chlorite and vermiculite were supplied to the Norwegian–Danish Basin and the Central Trough from the highs in the area. Kaolinite was deposited close to the shore in relatively large amounts together with traces of chlorite and locally vermiculite. Illite and I/S become more dominant with increasing distance from the shore. The main source area was probably the Fennoscandian Shield, periodically together with the Ringkøbing–Fyn High, as judged from the high kaolinite content in J1, F1, Frederikshavn City 1, and in one interval in M8 and I1. The recognition of soil clay assemblages in Gassum 1 and Rønde 1 suggests that ‘Gassum Island’ emerged above sea-level during part of the Upper Jurassic.

Volcanic ash contributed to sedimentation, as observed in the Central Trough wells 2/7-3 and W1, and the volcanism was possibly related to a period of stretching in the North Sea Graben System in Jurassic–Lower Cretaceous times. The volcanic material in the Central Trough was transformed during burial diagenesis to a randomly ordered I/S with ~ 50% illite layers.

The detrital, randomly ordered I/S with an illite content of 70% has not been diagenetically altered over the burial depths encountered in the Norwegian–Danish Basin. In the Central Trough it becomes ordered and the illite content increases up to 95%. In the samples investigated the I/S ordering occurs simultaneously with oil generation.
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REFERENCES


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