TECTONIC IMPLICATIONS OF ILLITE/SMECTITE DIAGENESIS, BARBADOS ACCRETIONARY PRISM

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Abstract-The depth distribution of illite/smectite (I/S) compositions was investigated for a well drilled to a depth of 3462 m on Barbados Island, the only subaerial exposure of the Barbados accretionary complex. The classical pattern of increasing percentage of illite interlayers in the mixed-layer clay with increasing burial depth was not observed. Rather, the data describe an irregular, zig-zag trend with depth. This trend is probably the result of reverse faulting in the section. I/S data were also used to infer several kilometers of uplift and subsequent erosion of the section. This study shows that irregular patterns of clay diagenesis with depth can be anticipated for sequences that have undergone complicated tectonism and deformation. Combined with other geologic information, these patterns can help to determine the tectonic history of the sedimentary sequence.

Key Words-Barbados, Diagenesis, Faulting, Illite/smectite, Tectonics.

INTRODUCTION

Diagenesis of clay minerals during burial has been the focus of many investigations (e.g., Burst, 1959; Dunoyer de Segonzac, 1970; Perry and Hower, 1970; Heling, 1974; Hower *et aI.,* 1976; and Boles and Franks, 1979). Most of these studies have dealt with the conversion of dioctahedral smectite to illite via intermediary mixed-layer illite/smectite (I/S) minerals. These investigations have shown that the transition is dependent primarily on temperature, with fluid and solid phase compositions and time playing lesser, but significant, roles. Pressure does not appear to be an important variable. Because of the dominant influence of temperature (i.e., geothermal gradient), sedimentary basins with relatively simple histories of subsidence generally show regular trends of increasing percentage of illite layers in I/S with depth. The extensive Gulf Coast studies, as well as other investigations, have shown that reaction time and composition (particularly the availability of K) influence the extent of diagenesis *ofI/S* at a given temperature, but a fairly regular pattern of increasing percentage of illite layers with burial depth (i.e., temperature) has generally been found.

In contrast, relatively little work has been reported on clay diagenesis in sedimentary sequences which have experienced post-burial uplift or deformation that might have altered an original depth trend related to continuously increasing temperature. Hoffman and Hower (1979) showed that alteration of clays in the Montana disturbed belt was related to thermal insulation resulting from emplacement of thrust sheets, not to original depth of burial in a sedimentary basin. Aoyagi and Kazama (1980) used the temperature dependencies of porosity change and various mineral transformations (including the conversion of smectite to illite) to estimate the amount of uplift and overburden removal of some sedimentary sequences in Japan.

The present paper reports results of a study of clay mineral diagenesis of mudstones in an area undergoing active tectonism. The rocks investigated are from the Barbados accretionary prism and have undergone complicated burial, uplift, and deformation. The study was undertaken: (1) to evaluate the influence of tectonism on clay burial diagenesis and (2) to estimate the magnitude and sequence of tectonic events affecting the sediments from the degree of clay mineral diagenesis.

GEOLOGY AND LITHOLOGY

The following discussion is based on the work of Speed (1981,1983) and Speed and Larue (1982). The island of Barbados exposes the structural high of the accretionary prism of the Lesser Antilles forearc (Figure 1). The island is underlain by a 3-tiered tectonostratigraphy: (1) an upper cap of Pleistocene limestone $(\leq 150 \text{ m thick})$, (2) an intermediate complex of thrust slices of forearc basin strata and autochthonous upperslope basin deposits (\leq 1500 m thick), and (3) a basal complex composed of accretionary fault packets of terrigenous muddy and sandy turbidites and hemipelagic rocks. Rocks of the basal complex exposed at the surface and sampled in wells were deposited in a trench

Figure 1. A. Map showing the location of Barbados Island on the structural high of the Barbados accretionary prism. B. Location of the Turner Hall well on Barbados.

in Paleocene and Eocene time and accreted mainly in late Eocene time. Mudstones of the basal complex are the subject of this study.

The probable geologic history of the basal complex in the upper 5 km of Barbados is as follows: (l) deposition in a trench wedge via longitudinal flow from sources in the Precambrian Guyana shield, (2) offscraping as wedge-shaped packets with concurrent tight folding and attachment to the toe of the accretionary prism, and (3) progressive deformation and uplift during growth of the accretionary prism since the Eocene. The burial history of the basal complex is uncertain. Initial burial by sedimentation before offscraping may have been small, perhaps no more than a few hundred meters. Depths of a given unit of rock in the accretionary prism probably increased progressively as the prism developed through continued thrust imbrication and horizontal shortening. The emplacement of the intermediate complex was probably in Miocene time. Erosion of sediment from the basal complex between Eocene and Miocene times is indicated by the existence of quartzose turbidites in allochthonous forearc basin strata of that age range.

ANALYTICAL METHODS AND MATERIALS

Samples of basal complex rocks were obtained from conventional and sidewall cores from the Turner Hall $#1$ well of Gulf Oil Company. The well penetrated 3462 m of mudstones, silty mudstones, and quartzose sandstones. Twenty-five mudstone samples, covering a

depth range of 383 to 3454 m, were analyzed in this study.

Clay mineralogy of the ≤ 63 - and ≤ 2 - μ m size fractions was determined by X-ray powder diffraction (XRD). Samples were disaggregated by soaking in distilled water, gently crushing in a mortar if necessary, and ultrasonication. Organic matter was removed by treatment of the samples with sodium hypochlorite according to Ristvet (1978); Fe-oxyhydroxides were removed using the method of Mehra and Jackson (1960). Dispersed samples were passed through a 63- μ m mesh sieve and centrifuged to isolate the $\langle 2-\mu m \rangle$ fraction. Oriented mounts were made by pipeting the sample onto a glass slide. Samples were examined on a Philips Norelco X-ray diffractometer using Ni-filtered CuK α radiation. The slides were X-rayed after air-drying, after solvation with ethylene glycol, and for some samples, after heat treatment at 500°C. Scans were made from 2° to $32^{\circ}2\theta$ at $1^{\circ}/\text{min}$ and from 14° to 18°2 θ at $\frac{1}{4}$ °/min.

I/S composition was determined using a variety of methods depending on the nature of the sample. For samples containing I/S with less than about 55-60% illite layers, the valley to peak ratio of the $(001)_{17}$ reflection was used to estimate I/S composition (Hoffman, 1976). Whenever possible, the positions of the combination peaks $(001)_{10}/(002)_{17}$ and $(002)_{10}/(003)_{17}$ were also used to estimate I/S composition (Reynolds and Hower, 1970). Determination of the composition ofI/S with percentage of illite layers equal to or greater than about 55-60 was more difficult because those samples commonly have smaller amounts of I/S and

significant amounts of discrete illite. When possible, positions of the combination peaks were used as described above; otherwise, an estimate of percentage of illite layers was obtained by visual comparison of the profiles to those *ofI/S* in Reynolds and Hower (1970) and Perry and Hower (1970).

Semiquantitative estimates of relative percentages of various clay minerals, quartz, and feldspar in the samples were made by measuring peak area by the triangle method (Mann and Fischer, 1982) and using the weighting factors of Biscaye (1965) and Mann and Müller (1979). This method uses the area under the $(001)_{17}$ peak and a graduated weighting factor dependent on composition to calculate the percentage of I/S. For samples from deeper than 2000 m in the well, small amounts *ofI/S* and high percentage of illite layers made the $(001)_{17}$ peak of the I/S small and indistinct. For these samples, the presence of I/S was noted but not quantified and the quantities of remaining minerals were normalized to 100%.

Thirty-four samples were analyzed for percentage of random reflectance of vitrinite by Robertson Research, Inc. All reflectance values were counted and plotted on a histogram. The lowest mode of reflectance was considered to represent primary vitrinite for the sample.

RESULTS

Mineralogy of Barbados mudstones

The Barbados mudstones are composed chiefly of I/S, kaolinite, discrete illite, chlorite, quartz, and minor plagioclase feldspar. Pyrite and mica were also identified under a binocular microscope in the $>63-\mu m$ fraction of most samples. Estimates of the relative abundances of the minerals in the $\lt 63$ - μ m size fractions are given in Table 1. The major downhole variation of mineralogy is a decrease in I/S content with increasing depth. Samples from the upper half of the well contain $30-75\%$ I/S, whereas deeper mudstones contain < 30% I/S. Several samples appeared from XRD patterns of the $\lt 63$ - μ m fraction to contain little or no I/S, although this phase was identified in the $\langle 2-\mu m \rangle$ fractions of all mudstones. The mudstones from the lower half of the well were found to be richer in kaolinite and chlorite than those from shallower depths. Although these two minerals were not differentiated in the semiquantitative XRD analysis, heat treatments of some samples indicated that kaolinite was significantly more abundant than chlorite.

The $\lt 2$ - μ m size fractions of the mudstone samples are composed of I/S, kaolinite, illite, chlorite, and minor quartz and plagioclase feldspar. Compositional trends with depth are similar to those observed for the $<$ 63- μ m size fraction; i.e., I/S decreases in abundance downhole whereas kaolinite and chlorite increase. Representative XRD patterns are shown in Figure 2.

Table I. Percentages of clay minerals, quartz, and feldspar in the ≤ 63 - μ m size fraction of Barbados mudstones.¹

¹ I/S = illite/smectite; I = illite; K + C = kaolinite + chlorite, undifferentiated; $Q =$ quartz; $F =$ feldspar; $tr =$ trace quantity; $pr = present$. In samples containing small amounts of I/S which has relatively high numbers of illite layers ($> 55 60\%$ illite), estimates of the percentage of I/S from the peak area method were considered unreliable. The presence of I/S is noted for these samples; the percentages of all other minerals are normalized to 100%.

Composition of the illite/smectite

The composition of the I/S in the $\lt 2$ - μ m size fraction of each sample is plotted against temperature and depth in Figure 3. The present-day geothermal gradient was estimated to be *15°C/km* from the bottom-hole temperature of the well and the range of 11-20°C/km for other wells on the island (Steve Lawrence, Exploration Consultants, Limited, Barbados, personal communication, 1981). The I/S composition range plotted at 20°C is that of mudstone outcrop samples as reported by Larue *et a/.* (1985). The composition-temperature curves for several other representative studies of I/S diagenesis in sediments with simple burial histories are also plotted in Figure 3. Note that the depth axis applies only to Barbados.

Two features of the Barbados I/S vs. temperature curve differentiate it from the other curves: (1) the percentage of illite layers does not increase in a regular fashion with temperature, and (2) samples of a given I/S composition are present at lower temperatures in Barbados than in other areas. Several explanations of the apparently anomalous features of the Barbados curve are possible. The depth-temperature profile of

Figure 2. Representative X-ray powder diffractograms of the $<$ 2- μ m size fraction of mudstone samples from the Turner Hall well (CuK α radiation).

I/S compositions could reflect inherited detrital compositions rather than a diagenetic trend. Alternatively, the I/S composition profile could be a result of tectonic disturbance of the section, caused by folding, faulting, and uplift, and ofthe altered thermal gradient produced by these disturbances. These possibilities will be considered in the discussion section.

Figure 3. Plot of I/S compositions of the $\lt 2$ - μ m fraction of Barbados mudstones vs. temperature and depth. General trends from other investigations of burial diagenesis ofI/S are plotted vs. temperature at indicated geothermal gradients. Curves are from Dunoyer de Segonzac (1970, Douala Basin); Hower *et al.* (1976, Gulf Coast); Boles and Franks (1979, southwest Texas); Freed (1979, Frio Formation, Texas); and Chang *et al.* (1986, Potiguar basin). Note that depth scale applies only to the Barbados data.

Depth profile of vitrinite reflectance

Vitrinite reflectance values vary linearly with depth (Figure 4). A least squares fit to all the data gave a correlation coefficient of. 79. In Figure 4, however, the data are fit with two nearly parallel, offset lines. Correlation coefficients are .83 and .69 for the upper and lower lines, respectively. Offset, linear depth trends of vitrinite reflectance are commonly interpreted to be a result of faulting (Hunt, 1979). Geologic evidence, cited below, for faulting of this section corroborates the interpretation of the data as two separate trends.

DISCUSSION

Evidence for reverse faulting in the section

The abundance of faults with near-vertical dips in outcrop on Barbados indicates the likelihood of encountering faults at depth. Evidence for faulting comes from borehole logs recorded at the time of drilling (unpublished data, Gulf Oil Company). The lithologic log recorded a zone of intercalated units of sand, silt,

and claystones between 1740 and 1900 m. This zone was labeled as a thrust zone in the log, and shows of gas and oil were noted. The dip log indicated a zone of anomalously low dips between 1830 and 1952 m and a 180° reversal in dip direction between 2012 and 2044 m. Although the dip log data did not specifically indicate reverse faulting, the depth correlation of the anomalous dip zones with other data supported faulting in the section. Finally, the mud log, which records the mud weight used in drilling the well, showed several sharp variations in the zone of interest. Mud weight can be used as a crude indicator of formation pressure and is generally kept about 10% over the formation pressure (Gretener, 1977). During drilling of the Turner Hall #1 well, mud weight sharply increased at 1700 m and then sharply decreased at 1825 m, possibly indicating a zone of high pore pressure. Below 1825 m, mud weight continuously increased until mud circulation was lost at 2450 m. The zones of high mud weight and the lost circulation suggest high porosity zones with abnormal formation pressure that might be related to faulting in the section.

These borehole logs suggested a fault zone between about 1700 and 2100 m. The mud log also recorded lost circulation at 2450 m, which possibly could be related to this or another fault zone. The offset in the vitrinite reflectance depth profile (Figure 4) was also consistent with a reverse fault at about 2000 m.

Influence of faulting on the I/S composition trend

The gradual increase of percentage of illite layers in l/S with depth (Figure 3) is disrupted by a reversal in trend between 2200 and 3000 m depth. Below 3000 m, the percentage of illite layers again increases with depth. This zigzag trend of I/S composition does not correlate with changes in lithology or mineral abundances (Table 1) which would be expected if the trend were an inherited detrital pattern. Furthermore, the geologic data discussed above indicate the presence of a major fault zone near 2000 m subsurface. Figure 5 is a schematic diagram illustrating how faulting of the section could disrupt a "normal" composition-depth profile and produce a zigzag pattern similar to that observed. Figure 5 shows that by time T_2 , a local maximum in percentage of illite layers develops along the fault plane. The increase in percentage of illite layers as the fault plane is approached from both sides possibly could be caused by (1) preferential heating along the fault due to friction or upward migration of warm fluids through the fault zone, or (2) increased water/ rock ratios or altered fluid compositions in this zone (Heling, 1974).

The maximum in percentage of illite layers in the I/S at 2174 m (Figure 3) is just below the approximate depth range of the reverse fault zone inferred from the borehole logs. The gradual decline of percentage of illite

Figure 4. Plot of vitrinite reflectance vs. depth. Data are interpreted as depicting two linear trends on the semilogarithmic plot. The offset of these two trends may be a result of thrust faulting.

layers in both directions away from the peak is similar to that depicted in Figure 5. A plot of vitrinite reflectance vs. percentage of illite layers (Figure 6) shows that, with the exception of a few samples, a linear relation exists between the two variables. This result supports the concept that the I/S composition trend is diagenetic, not detrital. The three samples that fall significantly off the linear trend are those corresponding to the maximum in percentage of illite layers between

Figure 5. Schematic diagram illustrating possible alteration of a burial diagenetic profile by reverse faulting. The top block shows the increase of percentage of illite layers in *liS* vs. depth at time T_0 . At time T_1 (second diagram), a reverse fault disrupts the sequence causing an offset in the I/S composition profile. The third block (time T_2) illustrates the effect of increased diagenesis along fault zones as a result of fluid migration, altered water/rock ratios, or heating in these zones.

2000 and 2500 m (see Figure 3). The upper two samples are from depths near the bottom of the inferred fault zone. The third sample is from the approximate depth at which mud circulation was lost (2450 m). The relatively low vitrinite reflectance values for these samples suggest that altered fluid compositions or water/ rock ratios rather than preferential heating, may have led to the maximum in percentage of illite layers.

Alteration of clay minerals in fault zones has been noted in another region of the Barbados accretionary prism. The toe of the Barbados prism was investigated during Deep Sea Drilling Project (DSDP) leg 78A (Figure 1). Drilling at DSDP site 541 penetrated several thrust faults and the basal décollement (Moore, Biju-Duval *et aI.,* 1982). Alteration of mixed-layer clays to saponite was noted in sediments from one fault zone and from the decollement (Schoonmaker, 1986). Fault zones and the basal décollement are thought to be major avenues for fluid migration through the accretionary prism (Westbrook and Smith, 1983; Cloos, 1984; Moore and Biju-Duval, 1984). Fluid compositions and water-rock ratios in these zones are probably different from those in the surrounding sediments and are prob-

Figure 6. Plot of random reflectance of vitrinite (%) vs. percentage of illite layers in I/S. The dashed line indicates the general trend of increasing reflectance and increasing percentage of illite layers. The three open symbols denote samples from the inferred fault zone. Their high percentage of illite layers in I/S relative to reflectance suggest that fluid migration, rather than heating along the fault, is responsible for the increased degree of diagenesis in that zone.

ably the reason for altered clay mineralogy (Schoonmaker, 1986).

The existence of a reverse fault, inferred from several lines of evidence, probably explains the irregular trend ofI/S composition with depth in Barbados. In a similar fashion, the following discussion makes use of independent geologic evidence for uplift of the Barbados region to explain the anomalous low-temperature occurrences of I/S rich in illite layers. If this explanation is accepted, the I/S data can then be used to estimate the magnitude of uplift.

Uplift oj the Barbados ridge

The Barbados accretionary prism has been uplifted since the Eocene. Besides uplift of the Barbados ridge as a whole, there is evidence for differential uplift of the Barbados Island area. Based on the depth of deposition of forearc basin rocks now exposed on the island, the rate of the late Neogene uplift of Barbados has been estimated to be 0.07-0.125 *m/l000* yr, averagedover the last 12-15 m.y. (Speed and Larue, 1982). Arching of upraised Pleistocene reef tracts has been used to estimate uplift rates of at least 0.2 m/1000 yr over the last 0.6-0.7 m.y. (Bender *et aI.,* 1979; Speed and Larue, 1982). On the basis of I/S compositions, vitrinite reflectance, thermal alteration indices, and Rock Eval pyrolysis, Larue *et al.* (1985) estimated that basal complex rocks exposed in outcrops on Barbados had been buried to depths of $1-5$ km.

Uplift was probably accompanied by erosion of the basal complex. The presence of resedimented basal complex rocks in some of the forearc basin strata (Larue and Speed, 1984) and the implied basal complex source for a sediment unit lying between the basal complex and the forearc rocks (the T -unit) in the southern part of the island imply that at least some erosion took place prior to emplacement of the forearc thrust sheets.

Estimates of uplift from I/S composition data

The relatively shallow occurrence *ofI/S* rich in illite layers (Figure 3) suggests that the mudstones were exposed to higher temperatures in the past, as would be the case if the rocks are not now at their maximum depths of burial (having experienced uplift and removal of overburden) or if they had been exposed to a higher geothermal gradient in the past. Deposition of the basal complex rocks as turbidites in a trench wedge followed by incorporation in the accretionary prism probably prevented equilibration of the sediments with a normal oceanic thermal gradient. Similarly, the low thermal gradients obtained from Barbados Island wells reflect the rapid rate of tectonic progradation and thickening of the prism which has prevented achievement of a standard thermal gradient (Speed and Larue, 1982).

If the depth trend of I/S composition is diagenetic rather than detrital, and if the current thermal gradient is representative of the past, the I/S data can be used to estimate the maximum burial depth of the basal complex rocks. Mudstone samples from the upper 1500 m of the well contain I/S having 40-50% illite layers (Figure 3). The depth range of these samples corresponds to a temperature range of about 25-40°C. The curves in Figure 3 from areas with simple burial histories cross the 40-50% illite layers composition range over a wide range of temperatures. The curves from southwest Texas (Boles and Franks, 1979), Brazil (Chang *et al.,* 1986), and the Cameroon (Dunoyer de Segonzac, 1970) are similar and yield a temperature range of 60-75°C. A comparison of Barbados with these three areas gives a temperature difference of 20-50°C. This temperature range corresponds to an additional 1.3-3.3 km of burial for the Barbados mudstones. A maximum temperature difference of 85°C (which translates to 5.7 km depth) is obtained using the curve for the Frio Formation (Freed, 1979).

Rough estimates of 1-3 km (and perhaps as much as 6 km) uplift and removal of overburden are similar to those obtained by Larue *et at.* (1985) (1-5 km). In addition, extrapolation of the vitrinite reflectance data for the upper half of the Turner Hall well (Figure 4) to an estimated initial value of 0.2% (Dow, 1982) yields a value of about 3.0 km uplift.

SUMMARY AND CONCLUSIONS

We have interpreted the depth trend of I/S compositions from the Turner Hall well on Barbados Island as a diagenetic trend that was altered by later reverse faulting and uplift. Circulation of fluids in the fault zone may have locally accelerated the clay mineral diagenetic reactions. Estimates of uplift and overburden removal obtained from the I/S and vitrinite data range from 1 to 3 km; one estimate is as great as 6 km. These values are in accord with other estimates obtained independently from geological considerations.

In this study, anomalies in the clay mineralogy of Barbados rocks have been correlated to specific geologic features or events. Thus, clay mineral data may be used to confirm structure models, uplift history, and the timing of various geologic events. For example, according to the fault model (Figure 5), we would conclude that movement along the fault occurred (or continued?) after significant burial and time for development of an original diagenetic curve; that is, movement along the fault was not limited to the original accretion process. If the diagenetic responses of sediments to tectonic events can be identified, clay diagenesis may provide a useful tool in helping to unravel the histories of tectonically complex areas.

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