CLAY MINERALOGY AND ILLITE CRYSTALLINITY OF THE ATOKA FORMATION, ARKOMA BASIN, AND FRONTAL OUACHITA MOUNTAINS

Christoph Spötl,[1] David W. Houseknecht,[2] and Robert Jaques

Department of Geological Sciences, University of Missouri, Columbia, Missouri 65211

Abstract—Clay mineralogy (including illite crystallinity) was studied in Pennsylvanian synorogenic sediments (Atoka Formation) in the subsurface of the Arkoma Basin and the adjacent Ouachita thrust belt. Vitrinite reflectance values range from $\geq 0.8\%$ at the surface up to as high as 4.7% R_o at the base of the Atoka Formation. The mineralogy of the $<2~\mu m$ fraction of the mudrocks is fairly monotonous and composed of illite ($<10\%$ interstratified smectite), Fe-chlorite, kaolinite, quartz, and traces of feldspars. Kaolinite is common at shallow levels and "disappears" in most wells at a thermal maturity of 1.9–2.1% R_o, suggesting its possible use as an independent paleothermal indicator in this basin. Illite crystallinity (IC) values are fairly high (0.3–0.5° 2θ) and show little variation throughout the entire maturity range. In addition, no relation was observed between vitrinite reflectance and illite crystallinity, indicating that IC is not a useful paleothermal indicator in these rocks. Illite is almost exclusively of the $2M_1$ polytype, suggesting a predominantly detrital origin. Incipient metamorphic and low-grade metamorphic mudrocks in the Ouachita thrust belt to the east of the Arkoma Basin are regarded as the source rocks for the clays of the Atoka Formation. Rapid transportation and deposition by turbidity currents probably played a key role in protecting these unweathered micas from pervasive alteration in the terrestrial environment.

Key Words—Anchimetamorphism, Diagenesis, Illite, Illite crystallinity, Polytype, Provenance, Vitrinite reflectance, X-ray powder diffraction.

INTRODUCTION

Thermal history and modeling of sedimentary basins rely fundamentally on the availability, accuracy, and precision of several paleotemperature indicators. Even studies based on abundant and diverse geologic information (e.g., chronostratigraphy, present-day heat flow, depth-R_o profiles, etc.) commonly do not permit unequivocal interpretations of the burial and thermal history of a given area or well (e.g., Waples *et al.,* 1992). A variety of organic and inorganic, semiquantitative, and quantitative indicators of thermal maturity are commonly used, most notably vitrinite reflectance, T_{max} by pyrolysis, diagenetic mineral assemblages, fluid inclusions, and apatite fission tracks (Frey, 1987; Naeser and McCulloh, 1989; Deming *et al.,* 1990).

Several clay minerals behave sensitively to temperature changes. Probably the most widely applied inorganic paleothermal indicator is the crystallinity of illitic clays, measured by the width of the illite 001 basal X-ray powder diffraction (XRD) reflection on oriented clay mounts ($<2~\mu m$ size fraction). IC studies published since the pioneering work by Weaver (1960) and Kübler (1966) suggest that the IC index (also known as the Kübler or Scherrer index) decreases almost linearly with increasing diagenetic grade and temperature. Because this procedure is rapid, inexpensive and

straightforward, numerous workers have applied it to a variety of argillaceous rocks with highly variable success. There is little doubt that the rate at which the crystallinity index decreases is to a large extent temperature dependent. Many internal and external variables other than temperature, including grain size, provenance, heating rate, effective stress, and total organic carbon (TOC) content of the host rock (e.g., Frey, 1987), also affect IC and place serious limitations on this method. Most workers now agree that this technique can provide useful information about the diagenetic/metamorphic grade of a shale or slate, only if as many of these influencing parameters as possible can be constrained and/or eliminated.

This paper describes the clay mineralogy of Pennsylvanian mudrocks from the central Arkoma Basin of Oklahoma and Arkansas, an area that long has been known for its high level of thermal maturity. Thermal history information of the Arkoma Basin is primarily based on vitrinite reflectance data of coal beds and dispersed organic matter. Because the resolution of vitrinite reflectance analysis decreases with increasing rank, it was initially anticipated that the IC method, in which sensitivity commonly increases with diagenetic grade, would be an appropriate tool to complement vitrinite reflectance measurements.

GEOLOGIC SETTING

The Arkoma Basin is a Paleozoic foreland basin that extends from southeastern Oklahoma to east-central Arkansas and is partially overthrust by the Ouachita mountain belt in the south. Because of its abundant

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[1] Present address: Münzergasse 2, 6060 Hall in Tirol, Austria.

[2] Present address: U.S. Geological Survey, 915 National Center, Reston, Virginia 22092.
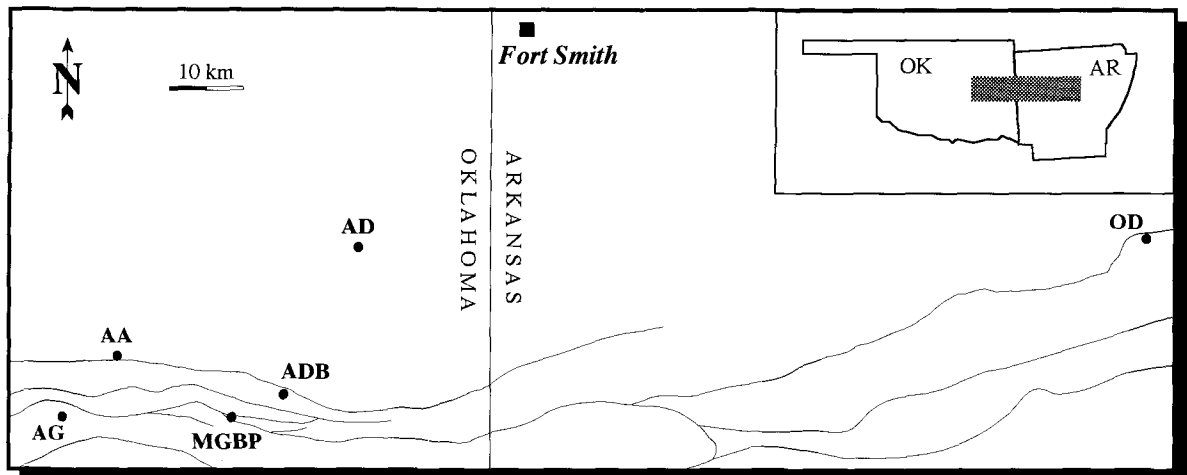
Figure 1. Map showing location of the study area in the Arkoma Basin and northernmost portion of the Ouachita thrust belt showing well locations. The boundary between the Arkoma Basin and Ouachita orogenic belt historically has been defined as the northmost major thrust fault. Surface traces of major thrust faults are indicated by continuous lines (all southward dipping). Key to well names: AA = Anadarko Petroleum Corp., Alford 'A' #1–15; AG = Anson Golden; ADB = Amoco Devil's Backbone #1; AD = Arkla Downes #1–20; MGBP = Mobil Green Bay Packaging #1; OD = Oxy Danville USA A-1.

hydrocarbon reserves at high thermal maturity levels, the basin is a frontier area for natural gas exploration and production in high-grade diagenetic and anchizonal reservoirs (Houseknecht and Spötl, 1993). The basin-fill can be divided vertically into a thin, shallow-marine shelf sequence (Cambrian through lowermost Pennsylvanian) and a thick, overlying wedge-like pile of flysch-to-molasse-type sediments (Pennsylvanian). The clastic wedge, the Atoka Formation, is synorogenic and up to 6.5 km thick in the southern part of the basin and was deposited mainly by E–W-trending submarine fans in a neritic to bathyal environment (Houseknecht, 1986, 1987). Although the Atoka sediments comprise more than three fourths of the entire Pennsylvanian section, they represent only about 5 million years of deposition. Strata overlying the Atoka Formation indicate a gradual shoaling of the basin with widespread deposition of peat interfingering with deltaic and lacustrine sediments. A significant but unknown amount of post-Pennsylvanian section has been eroded, as indicated by the high near-surface coal rank of 1–2% R_o (Houseknecht *et al.*, 1992).

Most samples examined in this study are from the Atoka Formation, a rather homogeneous sequence of dark-gray mudrocks, siltstones, and, locally, turbiditic argillaceous sandstones (e.g., Red Oak sandstone: Houseknecht and Ross, 1992). The siliciclastic detritus was derived predominantly from the evolving Ouachita orogenic belt to the east, and sand dispersal was mostly westward (Houseknecht, 1987; Sutherland, 1988). Minor dispersal systems from the Ozark Dome to the north and from the Illinois Basin to the northeast also delivered sediment into the rapidly subsiding basin. Sediment accumulation rates during deposition of Atoka sediments were up to 1300 m/Ma.

Atoka sandstones and siltstones in the study area contain abundant detrital phyllosilicates, lithic fragments (mainly slates and phyllites), and some feldspars (mostly Na-rich plagioclase), indicating derivation mostly from incipient and low-grade metamorphic rocks. In contrast, siliciclastic sediments underlying the Atoka flysch sequence (e.g., Spiro sandstone) are typically quartz arenites derived from the eroding North American platform to the north (Houseknecht, 1986). Atoka mudrocks and siltstones commonly contain 1–2% TOC, and petrographic analyses indicate that most organic matter is humic (type III kerogen).

Shale cuttings from several wells were studied for clay mineralogy, IC, and vitrinite reflectance. Wells AA and AD are located in the Oklahoma part of the basin, and wells ADB, AG, and OD are located in the frontal part of the Ouachita thrusts just south of the Arkoma Basin (Figure 1). In addition, samples from well MGBP were analyzed in a special study on the IC-TOC relationship.

METHODS

Splits of approximately 120 samples of borehole cuttings were mixed to obtain average samples for a given depth interval (~30 m). Because drilling mud may potentially contaminate the cuttings, samples were washed repeatedly in deionized water until the supernatant solution remained clear and only >2 mm rock chips remained. The samples were then crushed gently with a mortar and pestle, and the suspensions were transferred in plastic beakers. After 3–4 min of ultrasonic treatment, the <2 µm size fraction was separated by centrifuging. No cation saturation was performed. Two different methods were employed to make oriented clay mounts for XRD analysis: 1) Samples from

wells OD, AA, and AG were prepared by using the filter membrane peel technique method (Drever, 1973; hereafter referred to as the "peel technique"). Slide thickness was not controlled, but weighing of several slides showed that densities are ≥ 2 mg/cm^2, i.e., thin slides were avoided. 2) Samples from wells ADB and AD were prepared by pipetting a suspension onto a glass slide sitting on a ~60°C hot plate (hereafter referred to as the "sedimentation method"). Despite fairly rapid evaporation (≤ 1 hr) grain size segregation occurs (Moore and Reynolds, 1989). Two slides per sample were prepared for all samples. All slides were placed in an ethylene glycol (EG) atmosphere at 60°C overnight. For comparison, samples from well AG were prepared by using both methods.

XRD was performed at 40 kV and 30 mA with CuKα radiation. For IC measurements, slides were scanned in rotating mode from 7.5 to 10.5° 2θ at a step increment of 0.03° 2θ and a scan rate of 1° 2θ/min. The resulting digital data were processed for background correction and Kα_2 stripping. The following slit combination was used: 2 mm divergence slit, 4 and 0.5 mm anti-scatter slits, and 0.3 mm receiver slit. The width of the illite 001 reflection at one-half peak height (IC index) is expressed in $\Delta°\ 2\theta$. In addition, glycolated slides were also scanned from 2°–33° at 2°/min, and the percentage of expandables was determined from the difference between the positions of the 002 and 001 reflections (Moore and Reynolds, 1989).

The illite 001 reflection of a single glass slide was scanned ten times and the precision of the diffractometer was determined to be ±0.02° (2σ). Intra-sample variation, as determined by measuring two glass slides per sample (n = 16), is also less than ±0.02° (2σ). As a consequence, total precision is better than ±0.04° 2θ.

Ten samples prepared by using the sedimentation method (air-dried and glycolated) were analyzed at Kübler's lab in Neuchâtel for comparison with the IC values measured in our laboratory. The IC values determined in this study are consistently higher than Kübler's by 0.07° 2θ. However, we did not correct our IC data accordingly because we were primarily interested in relative IC changes. Note, however, that many IC workers (including Kübler) report IC data for air-dried samples only because the IC values for the anchizone and epizone limits are defined only for air-dried samples (Kübler, 1990; Kisch, 1991). We chose to glycolate our samples because of humidity fluctuations in the lab and IC values change with relative humidity. In addition, ethylene glycol solvation causes illite-smectite (I/S) peaks to shift toward lower 2θ angles, thus reducing the interference of I/S peaks with the discrete illite 001 reflection.

The relative abundance of clay-sized constituents was established by using a simple peak-intensity method. The following XRD reflections were utilized for this approach (normalized to 100%): illite 001, I/S 001/ 002, kaolinite 002, chlorite 004, and quartz 100. Values are relative indicators of changes in mineral abundance but are not accurate quantitative estimates.

Illite polytype abundance was studied in samples from well AG. Clay-size fractions were boiled in 10% hydrochloric acid for 1 hr to dissolve chlorite. Poorly oriented slides were prepared by using acetone and scanned air-dried at 0.25°/min from 20 to 37° 2θ. Quantification of mica polytypes was difficult because published methods using the intensity ratio 2.79 Å/2.58 Å (Maxwell and Hower, 1967; Mukhamet-Galeyev et al., 1985; Wilcoxon et al., 1990) produced $2M_1$ values in excess of 100%. More realistic results were obtained by calibrating against a pure $2M_1$ (pegmatitic muscovite) mica as an external standard, assuming a linear calibration line (cf. Mukhamet-Galeyev et al., 1985).

Vitrinite reflectance measurements were performed on dispersed vitrinite particles concentrated from Atoka Formation mudrocks by using a newly developed technique that allows collection of reflectance data while the microscope polarizer is rotated 360° (Houseknecht et al., 1993). For each sample, this method yields several variables, including mean maximum reflectance, mean rotational reflectance, and mean minimum reflectance. The thermal maturity parameter used in this study is the mean rotational reflectance, which is essentially equivalent to the reflectance parameter commonly used in many dispersed organic studies (R_o or R_{ran}; Houseknecht et al., 1993).

RESULTS

Vitrinite reflectance

Because the coalification and the metamorphism of the organic matter in the Arkoma Basin sedimentary rocks are part of a larger study (e.g., Houseknecht et al., 1992; Houseknecht and Spötl, 1993), we present here only the depth-R_o data relevant to clay mineral diagenesis. In all wells studied, vitrinite reflectance values increase progressively with depth. Modeling of the thermal history indicates that many geologic factors (including geopressure-suppression of thermal conductivity and advective fluid flow) contributed to the three-dimensional thermal maturity patterns within the basin, and these factors will be discussed elsewhere. Near-surface R_o values range from 0.8 to 1.9%, consistent with an increase in thermal maturity of the near-surface Hartshorne coal bed in a W–E direction (Houseknecht et al., 1992). Reflectance values recorded from the base of the Atoka Formation range from 2% in the west to more than 4% in the east (Houseknecht et al., 1992).

Clay mineralogy

Illite, Fe-rich chlorite, kaolinite, and quartz are the most abundant minerals in the <2 μm size fraction. Expandable clays (I/S) are only of local significance,

and K-feldspar and Ab-rich plagioclase are present in trace amounts. Illite shows low expandabilities ranging from 4% to 10% S (mostly 6% to 9%). The composition of this illite changes little with depth (see below). Minerals diagnostic of very low-grade metamorphism such as pyrophyllite, mixed-layer paragonite/muscovite, and paragonite were not detected. A significant amount of X-ray amorphous organic matter is indicated by the common presence of an elevated background centering around 3.4 Å. XRD analysis of kerogen concentrates from some of these samples indicates that the kerogen is highly disordered graphite-d_2 (Landis, 1971).

Expandable clay minerals are present as minor trace components in many samples. Typically, a low-intensity plateau is present between the chlorite 001 and the illite 001 reflection, centering broadly at 11.9 to 12.6 Å (glycolated samples). In some samples, a well-defined, intense peak occurs at 12.5 to 12.9 Å and is accompanied by a low-angle superstructure reflection at about 26.5 Å and a reflection at about 9.1 Å. The latter causes a shoulder on the high-angle side of the illite 001 peak. The structure collapses upon air-drying and the resultant reflection strongly interferes with the illite 001 reflection, causing a pronounced low angle-asymmetry. Modeling using software developed by R. C. Reynolds Jr. (Reynolds, 1985) suggests that this component may be a short-range ordered I/S mixed-layer mineral with about 50% S.

Figure 2 shows depth-related abundance of clay minerals in the wells studied. The most obvious trend is the "disappearance" of kaolinite, which occurs consistently at 1.9 to 2.1% R_o. The only exception is well AD, where kaolinite disappears at slightly higher R_o values (between 2.4% and 2.7% R_o). Illite increases at the expense of kaolinite in wells AA, AG, and AD (Figure 2). Chlorite, I/S, and quartz display either no major depth-related trends (e.g., well OD) or show local trends that cannot be correlated between individual wells (e.g., quartz increases down-section in well AA; Figure 2).

Illite crystallinity

The IC measurements yielded unexpected results. Data from wells OD and AA (Figure 2) show virtually no change in IC with depth, samples from well ADB reveal a trend of decreasing IC values with depth (Figure 2), and data from wells AG and AD show a slight increase in IC values downhole. The IC values of samples prepared using the peel technique (wells AA, AG, OD) are mostly 0.3 to 0.4 $\Delta°2\theta$ with slightly higher values for samples from well ADB and AD (Figure 2; for the Atoka section), which were processed according to the sedimentation method. Taking into account the 0.07° difference between values in this study and that determined in Kübler's laboratory, most values suggest anchizonal metamorphism (0.42 to 0.25 $\Delta°2\theta$; Kübler, 1990; Kisch, 1991).

The amount of smectite interstratified within the illite appears to follow the weak trends shown by the IC data, in particular, the increase in IC values with depths in wells AG and ADB (Figure 2). No data on the expandability of the I/S with depth are given because these poorly defined reflections could not be measured precisely.

In order to study the possible preparation-induced errors that could result from using either one of the two methods (peel *vs.* sedimentation), we processed samples from one well (AG) by using both techniques. The results show that the sedimentation method gives systematically higher IC values (i.e., broader peaks) than the peel method, but the trends are the same (Figure 3).

Mica polytypism

Mica polytype abundance was studied in samples from well AG that span almost the entire thermal maturity range in the wells studied (0.8 to 3.5%). After HCl-treatment, most samples still contained minor amounts of chlorite. Comparison of untreated and HCl-treated samples, however, shows that no severe interference problems are caused by the presence of small quantities of chlorite in these samples. All samples show well defined non-basal lines at 4.09 Å, 3.88 Å, 3.72 Å, 3.20 Å, 2.98 Å, 2.86 Å, and 2.79 Å, diagnostic of the $2M_1$ polytype (Bailey, 1984). The 114 reflection at 3.49 Å interferes with the chlorite 005 line at about 3.53 Å. Weak reflections at about 3.07 Å and 3.66 Å suggest the presence of small amounts of polytype $1M$ in three samples. Estimated percentages of $2M_1$ polytype in the Atoka samples from well AG range from 47% to 112% (mean = 74%). Figure 2 shows a slight decrease in the relative abundance of $2M_1$ polytype with depth in the well, which is opposite to the IC trend.

DISCUSSION

Thermal maturity

Vitrinite reflectance data indicate that the studied sections of the Atoka Formation cover the transition from high-grade diagenesis to anchimetamorphism, which is commonly defined by R_o values between 2.5 and 3.1% (Héroux *et al.,* 1979; Kisch, 1987; Robert, 1988). R_o values for the deepest parts of some wells approach the anchizone-epizone boundary. Although such high reflectance values indicate that the rocks of the Arkoma Basin were once exposed to much higher temperatures than at present, the thermal history of the basin remains uncertain. The timing of the regional thermal maximum appears to be confined to the late Paleozoic (Denison *et al.,* 1977; Denison, 1982; Desborough *et al.,* 1985; Arne, 1992), coincident with the Ouachita orogeny. However, the duration and cause of anomalously high thermal events are unresolved. It
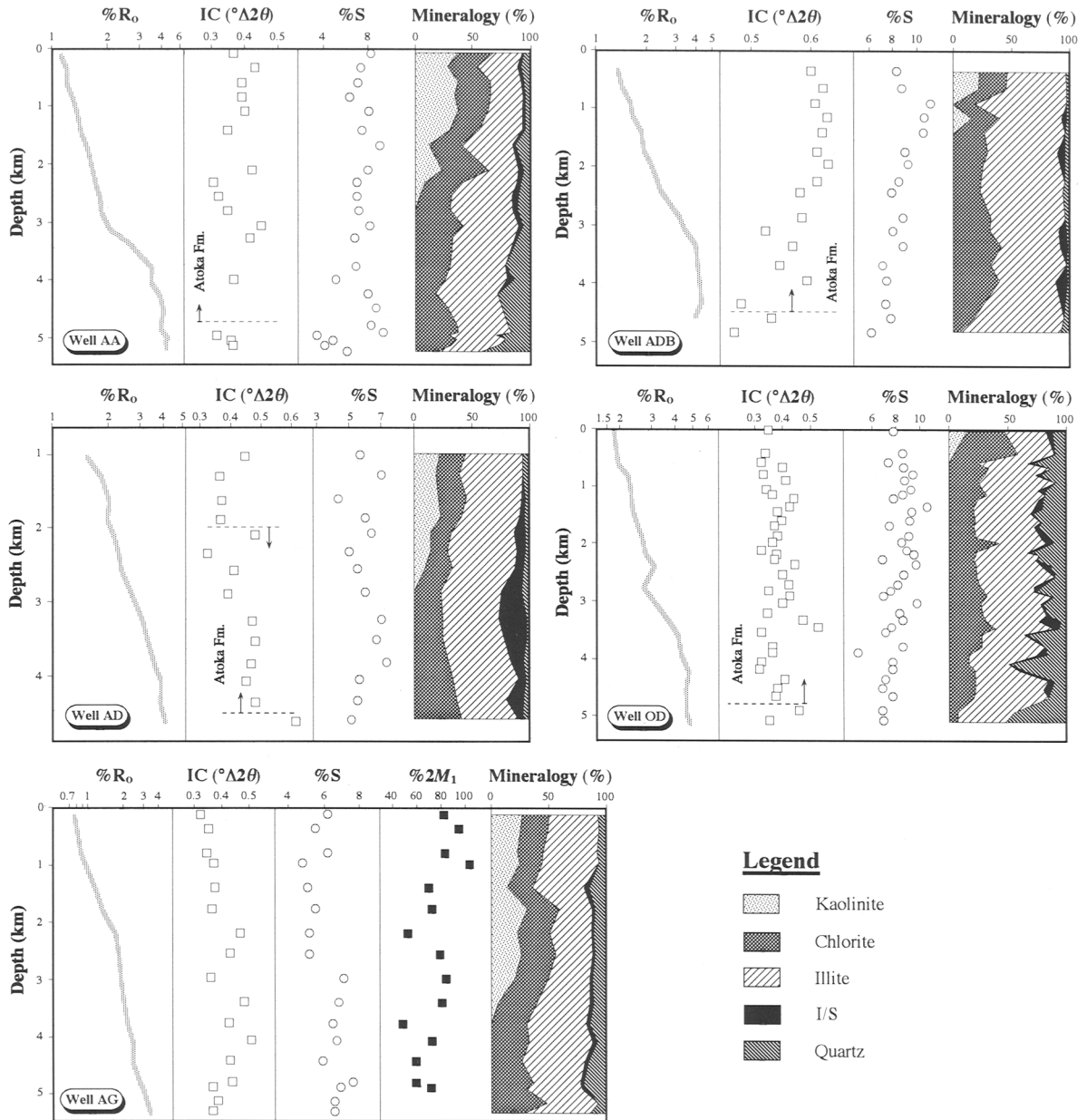
Figure 2.   Illite crystallinity (IC), <2 μm mineralogy, and vitrinite reflectance (R$_o$) from five deep wells in the Arkoma Basin and frontal Ouachita Mountains. The diagram for well AG also includes data on the abundance of illite polytypes. Unless otherwise indicated, samples are from the Atoka Formation. Note logarithmic vitrinite reflectance scale. %S is the expandability of the illite. For well identifications, see Figure 1.

has been suggested that a hydrological regime operated in the basin, producing large-scale fluid migration and subsequent ore mineralizations in adjacent areas (e.g., Leach and Rowan, 1986; Ge and Garven, 1992).

Clay mineralogy trends

Near-surface vitrinite reflectance values in the wells range from 0.8% to 1.8% and correspond to maximum paleotemperatures of 128°–228°C, using the empirical relationship of Barker and Goldstein (1990). Even if these values are used as temperature estimates, they imply, by analogy with other sedimentary basins (e.g., U.S. Gulf Coast), that the major I/S-to-illite transformation reaction has already taken place (Pollastro, 1993), i.e., no randomly ordered I/S is to be expected in the Atoka Formation. This prediction, however, is based on the assumption that the initial Atoka mud had a clay mineralogical composition similar to the

modern Mississippi River sediment (i.e., smectite-rich I/S). Unfortunately, no Atoka samples were studied from thermal maturity levels <0.8% to help determine the initial composition of the Pennsylvanian sediment. In addition, there is no evidence to suggest that the initial Atoka sediment was smectite-rich.

Kaolinite is present in all wells at shallow depths and is absent in the deeper parts where thermal maturities are higher. It "disappears" in most wells at a thermal maturity level of 1.9% to 2.1%. Although some of this kaolinite could possibly be diagenetic in origin, we believe that the bulk of the kaolinite is detrital. This interpretation is based primarily on the work by Weaver (1960, 1961), who showed that clay mineralogical changes in the various foreland basins of the Ouachita orogen, including the Arkoma Basin, are related to changes in provenance. Weaver found that, while kaolinite is rare or absent in Mississippian and older formations, it is a common constituent in Pennsylvanian rocks such as the Atoka Formation. Such regional, basin-wide changes in the clay mineral distribution pattern strongly argue in favor of a detrital source of the kaolinite, caused by major orogenic and/or climatic changes. Weaver also observed a decrease in the abundance of kaolinite towards more distal, basinal facies in the Ouachitas, consistent with a primary sedimentation gradient. We thus feel confident that kaolinite in the Atoka Formation is largely detrital and that its "disappearance" is linked to the thermal history of the area. Studies from other basins concluded that the breakdown of kaolinite at higher temperatures is strongly dependent upon compositional factors (Kisch, 1983, 1987). In contrast, our data imply that for a given lithology and basinal setting the kaolinite disappearance depth may provide a reliable inorganic paleothermal indicator. The absolute thermal maturity level at which kaolinite breaks down, however, obviously varies from basin to basin. Trends of relative abundance of kaolinite and illite in the wells examined (Figure 2) suggest that kaolinite might have reacted to form illite. No such trend exists for chlorite, suggesting that the chloritization of kaolinite was not important.

The occurrence of significant amounts of expandable clay minerals (I/S) in some samples (e.g., well AD) is somewhat unexpected at such high thermal maturity values. Because all samples are from cuttings, it is likely that smectite-rich I/S is from drilling mud (e.g., Pollastro, 1993). The observation that I/S is typically absent at shallow levels and occurs only in certain deeper intervals of some wells (and cannot be correlated between wells) supports this interpretation.

Evaluation of IC measurements

IC workers are aware of the limitations of the method (e.g., Frey, 1987) and considerable effort has been undertaken to standardize IC measurements (Blenkinsop, 1988; Robinson *et al.,* 1990; Kisch, 1990, 1991). The
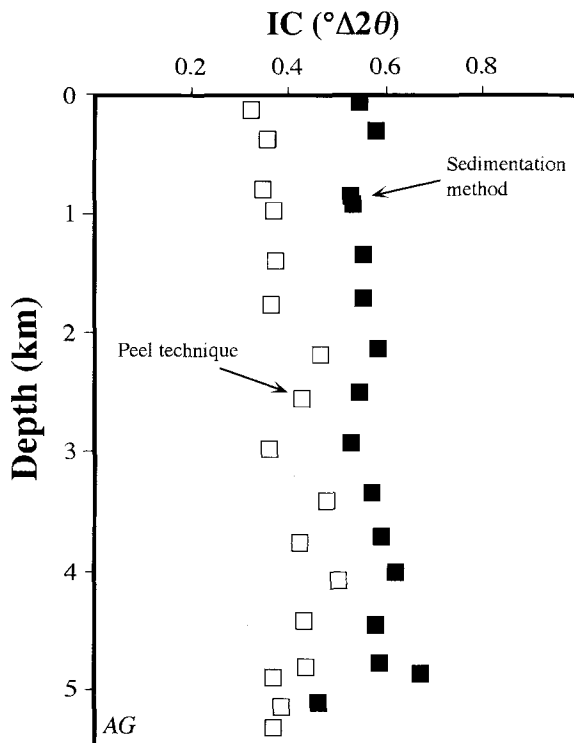

Figure 3. Comparison of IC measured on samples prepared by the peel technique and the sedimentation method (both ethylene glycolated). Note that the samples used for the sedimentation method are not exactly from the same stratigraphic levels as those processed by using the peel method because all the sample material had been used and new samples were taken as close to the initial sampling intervals as possible.

decision to glycolate our slides and the use of two different slide preparation techniques introduce a systematic error in the measured IC values. Because we are primarily concerned with relative changes of IC values within individual wells, these preparation-induced errors will not affect our interpretations. Several geological factors (in addition to temperature) are also known to affect crystallinity measurements of micas, including high TOC contents, presence of I/S, expandability of the illite and heating rate of the rocks (cf. Frey, 1987). In addition, lithology may also be an important controlling factor. However, based on visual examination of the cuttings, we are confident that all samples are from the same lithology, i.e., fine-grained siltstones and shales.

Several authors have reported that rocks rich in organic matter show systematically higher IC values than intercalated TOC-poor lithologies (e.g., Ogunyomi *et al.,* 1980; Yang and Hesse, 1991). On the other hand, Frey (1988) did not observe a significant difference in IC values between red and black slates from adjacent formations in the Swiss Alps. To quantitatively evaluate the influence of organic matter on the IC values of the Atoka Formation, we selected a dozen samples
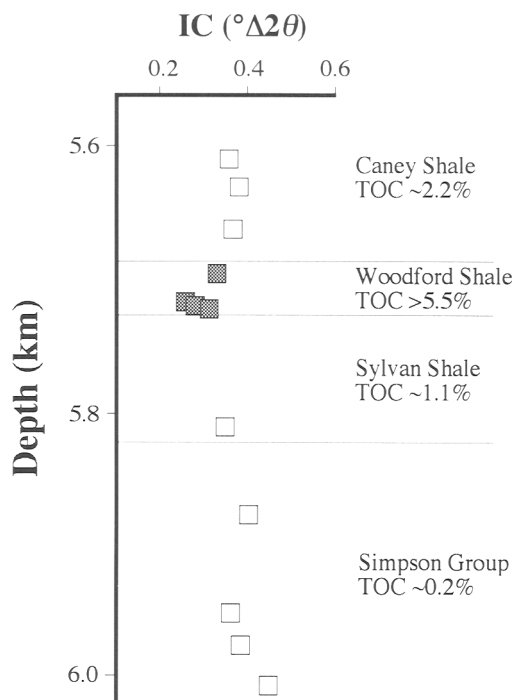
IC (°Δ2θ)

Figure 4. Variation of IC values relative to total organic content (TOC) in well MGBP. Note that TOC-rich samples have similar IC values as TOC-poor samples. Samples from the Woodford Shale are shaded. The R_o values for the studied depth interval range from 3.7 to 4.3% (n = 4).

from the well MGBP (for location see Figure 1) that penetrates lithologies of vastly different TOC contents within a fairly short depth interval (Figure 4). Cuttings were analyzed from the Caney and Sylvan shales having similar TOC contents as the Atoka Formation, from the Woodford shale (black shale, oil-prone source rock), and the TOC-poor Simpson Group. XRD specimens were prepared using the peel technique followed by treatment with EG. The results show little or no change in IC values among lithologies (Figure 4). The IC values for the TOC-rich Woodford Shale are slightly lower than the others, which is the opposite relation from what is commonly reported elsewhere. Therefore, we believe that variable TOC content is not affecting depth-IC trends in the Atoka Formation.

The presence of I/S may cause a more serious problem in the IC measurements. IC measurements of air-dried samples are of limited value, unless more sophisticated manipulations such as peak deconvolution are employed (e.g., Lanson and Champion, 1991). We, therefore, minimized interfering basal reflections of I/S by glycolating the samples. The partially hidden I/S reflection at about 9.1 Å (occurring only in a few samples richer in I/S) poses no problem for IC measurements, because the height of this I/S reflection is always less than a third of the height of the first illite peak. As

a consequence, the peak-width at half-height of the illite 001 peak is not influenced by the interfering I/S line. This is also supported by the absence of a statistically significant correlation between IC and I/S abundance.

Studies in areas that experienced geologically short heating events show that IC lags behind organic maturation/metamorphism (e.g., Teichmüller et al., 1979; Robert, 1988) and may, therefore, show no correlation with vitrinite reflectance values. This possibility can be ruled out for the Arkoma Basin, because even a retarded illitization process should produce a trend of decreasing IC values with depth (with IC values being larger than expected from the vitrinite data).

Interpretation of the IC data

IC measurements from the Atoka Formation of this study do not fit into the ideal conception of increasing illite crystallinity (decreasing IC values) with increasing diagenetic grade. As shown in the previous section, a number of variables known to affect IC values (Frey, 1987) can be ruled out in the case of the Atoka Formation. We, therefore, favor the interpretation that the measured IC values are largely controlled by detrital input. We suggest that the initial Atoka sediment was dominated by highly illitic/sericitic assemblages. Such material does not undergo smectite-to-illite transformation reactions in the subsurface, as is the case in systems dominated by smectite-rich clays (e.g., modern Mississippi River mud). Two lines of evidence support the interpretation that the clays of the Atoka flysch deposits are largely detrital, illite polytype abundance and sandstone petrology.

Diagenetically formed illite is mostly of the $1M$ (or $1M_d$) polytype, whereas $2M_1$ is commonly from igneous/metamorphic environments. The relative amount of $2M_1$ vs. $1M$ systematically increases from high-grade diagenesis to very low-grade (anchizonal) metamorphism (e.g., Hunziker et al., 1986; Yang and Hesse, 1991). The high proportions of $2M_1$ mica polytype in the <2 μm size fraction in samples at $<1\%$ R_o in well AG argue that the bulk of the illite (and hence measured IC values) is detrital. This is consistent with earlier observations by Weaver (1961) that illite in foreland sediments of the Ouachita system has a $2M$ structure. The relative abundance of $2M_1/1/M$ mica polytypes in this well appears to parallel the IC trend, i.e., a decrease in illite crystallinity downhole is accompanied by a decrease in the relative amount of $2M_1$. This can best be explained in terms of a slight increase down-section in the amount of neoformed vs. detrital illite.

Sandstone petrological studies indicate that detrital micas, slaty, and phyllitic rock fragments are abundant in the Atoka Formation (Mack et al., 1983; Zachry, 1983; Houseknecht, 1986; Morris, 1989; Houseknecht

and Ross, 1992). This material was predominantly derived from the eastern Ouachitas, which today are largely concealed beneath Mesozoic and Tertiary sediments of the Gulf Coast Plain. Based on the distribution of unmetamorphosed and very low-grade metamorphic sedimentary rocks in the Ouachita Mountains (see review in Viele and Thomas, 1989) and available subsurface data to the east (e.g., Weaver, 1960, 1961; Graham et al., 1976; Nicholas and Waddell, 1989; Thomas, 1989; Viele and Thomas, 1989), a similar mudrock-dominated, weakly metamorphosed lithology can be anticipated in the eastern Ouachitas. Metamorphic rocks of the southern Appalachians (e.g., Talladega slate belt) could have been an additional source area. We speculate that rapid erosion and sediment transportation into the flysch basin prevented pervasive weathering of the micas in the terrestrial-pedogenic environment. Such an interpretation is supported by the observed high sedimentation rate during deposition of the Atoka Formation.

A previous study by Guthrie et al. (1986) also showed no correlation between R_o and IC for Atoka samples from outcrops in the northern Ouachita Mountains. These authors, however, observed a statistically significant relationship between these two parameters in samples from the older Jackfork Sandstone and Stanley Shale from the same area. These units had source areas different from that of the synorogenic Atoka Formation (Houseknecht, 1987), thus suggesting that provenance may indeed control mica composition. Preliminary results by the authors from the Woodford Shale in wells from the Arkoma Basin also suggest that the IC of shelf-rock micas correlates with the thermal maturity.

Mica composition and "crystallinity" controlled by provenance have only rarely been documented in burial diagenetic sequences. Monnier's work (1982) in the Swiss Molasse Basin, based on XRD analysis of some 2000 samples from exploration wells, is probably one of the most detailed IC studies. His profiles show sections with consistently low IC values (dominated by detrital illite) and others with intervals of increasing IC values overlain by a section of constant IC values (reflecting increasing abundance of diagenetic I/S). Dorsey et al. (1988) examined Pliocene and Pleistocene mudrocks from eastern Taiwan and observed an up-section increase in the degree of illite crystallinity with IC values typical of the epizone in very shallow samples. This IC trend is paralleled by an up-section increase in the abundance of metamorphic rock fragments. The authors attribute these trends to progressive unroofing of metamorphic terranes in the source area during collision-related uplift and deformation. These studies suggest that detrital clay mineral assemblages characterized by anchizonal and/or epizonal IC values do not undergo further diagenetic alteration, thus seriously limiting the use of IC as a sensitive paleothermal index.

SUMMARY AND CONCLUSIONS

Mudrocks of the Pennsylvanian Atoka Formation in the Arkoma Basin and the northern part of the Ouachita thrust belt experienced deep-burial diagenesis, as indicated by R_o values ranging from 0.8 to 4.7%. Despite this large difference in thermal maturity, the clay mineralogy (<2 μm fraction) and in particular IC values and illite polytype abundance do not change significantly within five deep wells studied. These results are surprising, because numerous workers reported systematically smaller IC values (i.e., higher "crystallinities") in progressively deeper buried rocks. Examination of mica polytypism and the detrital composition of interbedded turbiditic sandstones suggests that the bulk of the clay-sized mica is detrital. The initial Atoka mud was derived from the advancing Ouachita orogenic belt in the south that was dominated by very low-grade and low-grade argillaceous sediments.

Detrital kaolinite is common in the <2 μm fraction of shallow samples and "disappears" in most wells at 1.9 to 2.1% R_o, probably related to illitization reactions. Our data suggest that the "disappearance" of kaolinite may provide a useful inorganic paleothermal marker within the Atoka flysch sediments of the study area.

The results of this study underline the difficulties in applying the IC concept to clay mineral assemblages that are strongly provenance-controlled. Sedimentary sequences derived largely from nearby micaceous source rocks (meta-pelites, phyllites, meta-psammites) are likely to be poor candidates for thermal history studies using IC indices. Utilizing a size fraction smaller than 2 μm (although not recommended for IC studies; see Kisch, 1991) as well as more sophisticated XRD peak deconvolution procedures possibly may help to reduce the "detrital noise" in clay mineral populations such as those from the Atoka Formation.

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