ABSTRACT: The nature of clay-mineral diagenesis in shales has received much attention because of its possible role in providing material to interbedded sandstones. In particular, the smectite to illite transformation has been cited by many authors as a source of silica during mudrock diagenesis. We suggest that illite is not an end-member itself but a transition between smectite and muscovite. This is supported by chemical and physical differences between the two minerals from the literature.

Mudrocks from the Stanley Formation (Mississippian) of the Ouachita Mountains of Oklahoma and Arkansas were investigated both chemically and petrographically to test the hypothesis that illite undergoes a continuous transformation to muscovite. Results were related to thermal maturity as determined from vitrinite reflectance data of Houseknecht and Matthews (1985) on the same samples. Within the phyllosilicate fraction, the concentration of silica was found to decrease with increasing thermal maturity. Loss of water from interlayer positions also showed a linear decrease across the same interval. This is consistent with the change of illite into muscovite mica.

Coincident with the observed changes in the phyllosilicate fraction, a corresponding increase in the amount, grain-size and percentage of composite grains was found in the non-phyllosilicate fraction. This is interpreted as the result of authigenic growth of solid silica released during clay-mineral transformations. This is consistent with reported increases in the amount of polycrystalline quartz in metapelites compared to their precursors. Because of the increase in the percentage of quartz in metapelites, they are a likely source of abundant silt-size quartz. The approximate mass-balance of silica between the phyllosilicate and non-phyllosilicate fractions also suggests that the Stanley mudrocks behaved as closed systems during diagenesis.

INTRODUCTION

The smectite to illite clay-mineral transformation is well known in the literature of diagenesis. The transition appears to be a continuous one, varying from 100% smectite layers through a mixed-layer sequence until 100% illite layers are found within the clay mineral. After the loss of expandable clay layers (smectite layers), illite becomes more ordered and therefore more crystalline with increasing burial. Consequently, illite is often considered an end-member of this transition (Srodon and others, 1986).

A precise definition for illite has not been agreed upon. It is generally agreed that illite is similar in structure to muscovite, sharing similar polytypes based on the type of layer stacking. Most illite tends to be of the 1Md polytype (Eslinger and Pevleaf, 1988), while most muscovite is of the M2, polytype (Deer et al., 1967). Because of their similar structure, illite and muscovite share a basal spacing of 10 Å and are not discriminated by conventional XRD methods. Consequently, the terms mica and illite have been used interchangeably in many instances for fine-grained, potassium-rich phyllosilicates.

There are several distinctions between illite and muscovite. Both minerals have a distinct chemistry. Illite has more silicon and therefore a smaller negative layer charge than muscovite. Illite therefore requires less potassium to balance this charge. Chemical data compiled from the literature indicate that there is almost as much difference between the chemistry of illite and muscovite as exists between illite and smectite (Table 1). Muscovite is generally coarser-grained than illite, which is itself coarser-grained than smectite. There are also very slight differences in the cell edges, which are extremely difficult to detect except with pure phases. Another important distinction between the two minerals is their thermal stability. Illite generally begins dehydroxylation near 550°C, while muscovite is thermally stable to near 700°C (Todor, 1976). The variation in the temperature of dehydroxylation correlates with illite crystalinity, suggesting a gradational recrystallization of illite into muscovite (Hunziker et al., 1986). These gradational variations between illite and muscovite strongly imply that illite is not itself an end-member but is only a phase halfway between smectite and muscovite. It has only rarely been recognized as such because of the similar positions of peaks on X-ray diffraction diagrams.

The consequences of the continued recrystallization of illite into mica with increasing thermal maturity are important to sedimentary petrologists. The same elements released during illitization of smectite would continue to be released. This includes silicon, iron, magnesium and water. The reaction would also consume aluminum and potassium.

The fate of these elements is not fully agreed upon even during the smectite-illite transformation. As early as 1962 it was suggested that clay-mineral transformations could provide silica to sandstones (Towe, 1962). Many authors feel this is also
the case for other released cations as well as silica (Land and Macpherson, 1992; Curtis, 1978). Other work suggests that
shales behave as closed systems (Totten and Blatt, 1993; Sul-
villian and McBride, 1991; Bloch and Hutcheon, 1992). In either
case, the continued release of silica during recrystallization of
illite almost doubles the amount available for authigenic quartz
growth. If the silica indeed remains within the shale, it could
be a major contributor to the silt-size quartz population.

It is the aim of this research to assess the possibility that
phylllosilicate recrystallization during diagenesis continues after
the smectite to illite transition is complete, with muscovite mica
as the final product. The fate of the released silica is also in-
vestigated, and the contribution to the silt-sized quartz popu-
lation estimated. A method to determine the stage of illite re-
crystallization is proposed.

GEOLoGIC SETTING

The pelites studied are from the Stanley Formation (Missis-
sippian), which crops out in the Ouachita Mountains of south-
western Arkansas and southeastern Oklahoma. The Stanley
Formation has a maximum thickness of 3,350 m (Morris, 1989)
and is part of a much thicker sequence of deep-water flysch
deposited off the southern margin of the North American con-
tinent during late Paleozoic time. Many previous studies of bur-
ial diagenesis have focused on the Tertiary sequence of the U.S.
Gulf Coast and rely upon the assumption of relatively constant
provenance. The sedimentation rate during deposition of the
Stanley Formation was very similar to the sedimentation rates
of the Cenozoic Gulf Coast. The much shorter interval of Stan-
ley deposition (10 Ma) minimizes the effect of variations in
source area.

The Stanley Formation is composed of over 70% shale, with
interbedded sandstones and a few tuff beds (Johnson, 1968).
The shale varies in color from olive green to black, and is gen-
erally blocky, although in some localities it is finely fissile.
Stanley shales are very fine-grained, massive, and were depos-
ited as distal turbidites in an abyssal setting (Houseknecht and
Matthews, 1985). The deep-marine depositional environment
of the Stanley shales limits the effect of sea-level changes on
the detrital mineralogy.

In the core area of the Ouachita Mountains in Arkansas, the
Stanley shales have been metamorphosed to phyllite and per-
haps lower greenschist facies. Slate and unmetamorphosed
shale occur outward from the core area. Houseknecht and Mat-
thews (1985) reported that vitrinite reflectance values range
from 0.24 in unmetamorphosed shales to a maximum of 4.28
in phyllites from the core area. Using the relation of Barker
(1988), this represents a maximum temperature variation ap-
proaching 300°C. Guthrie et al. (1986) reported a relation be-
tween illite crystallinity (Kubler Index) and vitrinite reflectance
with a correlation coefficient of 0.82. Based upon XRD patterns
of glycolated samples, mixed layer clays are still present in
shales of low thermal maturity (Ri < 1.5%), although illite is
the dominant clay mineral throughout. A large proportion of
this illite may be detrital because a major source of the sediment
supplied to the Ouachita basin was the stable craton. The
detrital illite would not be involved in the smectite to illite tran-
sition during diagenesis of the Stanley shales but could be in-
volved in thermally driven recrystallization toward micas. The
excellent correlation between illite crystallinity and thermal ma-
turity found by Guthrie et al. (1986) supports this concept.

METHODS

Sample Collection

Samples used for this study are the same as those used in
Houseknecht and Matthews (1985), Guthrie et al. (1986), and
Totten and Blatt (1993). Grab bag samples were collected by
Matthews at least one foot below the surface of the outcrop to
avoid weathering influences. Sample locations are shown in
Figure 1.

Analytical Procedures

Whole-rock major-element composition was determined by
XRF analysis of fused discs. The discs were prepared using a
La,Na-doped lithium tetraborate/lithium carbonate flux. Ana-
lyses were performed at the University of Oklahoma’s Texaco
X-ray Laboratory on a Rigaku SMAX XRF using a rhodium
anode X-ray tube operated at 40 kV and 65 Ma.

Quartz and feldspar were separated from the bulk rock using a
modified sodium pyrosulfate fusion method of Blatt et al.
(1982). Total quartz and feldspar were calculated by weight
remaining after the fusion. Two separate fusions were per-
formed for each sample. The fused residue from one run was
fashioned into a fused disc and analyzed for major element
concentrations by XRF. Quartz percentage and modal feldspar
abundance were calculated from the resulting XRF data. The
chemical composition of the clay-mineral fraction was calcu-
lated by subtracting the non-clay composition from the whole
rock data. The fused residue of the second run was separated
into size fractions. An aliquot of each sized separate was re-
served for petrographic investigation.

Oriented, sedimented mounts were made of the less than 2-
µm fraction of disaggregated whole-rock samples and analyzed
by XRD. The minerals present confirmed the results of Guthrie
et al. (1986) on the same samples. The samples were then
placed in an oven at 500°C for two hours and reanalyzed by
XRD. Peak areas for the 10-Å peak were calculated from both
runs and the difference in peak area calculated.

Petrography

The size-separated quartz and feldspar were examined using a
polarizing petrographic microscope. Grain mounts were made
using Petrosopy 154 (index of refraction = 1.540 ± 0.001)
and examined with a Leitz microscope under both plane and
polarized light. For each size fraction, 200 grains were point
counted to determine the percentage of polycrystalline grains.
Chert is commonly defined as polycrystalline quartz with each
individual crystal being less than 20–30 µm in diameter. For
the purpose of this study the term chert includes all polycrys-
talline quartz. Because of the small size of the grains in our
samples, very few quartz aggregates had individual crystals
coarser than 30 µm.

Because sodic plagioclase has an index of refraction similar
to quartz and because twinning and cleavage are usually not
visible in silt-sized feldspar grains, petrographic identification
of feldspar was not considered reliable. The quantity of plagio-
clase in each size fraction was determined by XRD, by com-
SILICA FROM THE ILLITE TO MUSCOVITE TRANSFORMATION DURING LATE-STAGE DIAGENESIS OF SHALES

Fig. 1.—Sample locations in relation to vitrinite reflectance contours (after Houseknecht and Matthews, 1985).

Parison to diffractograms of prepared standards with known proportions of quartz and albite. The most sensitive peak (highest intensity) for plagioclase determinations is the reflection of the 002 plane, with a d-spacing of 3.197 Å. The technique is sensitive to approximately 1% albite, below which the peak is not statistically detectable above background. Additional information on analytical procedures is found in Totten (1992).

RESULTS

The major element chemical analyses are presented in Table 2. Major-element oxides are reported on a 100% anhydrous basis and are in weight percent. Several analyzed samples were omitted from this study based on their anomalous trace-element characteristics (Totten and Blatt, 1993). The anomalous signature is interpreted as indicating a different provenance, hence obscuring diagenetic alteration. Grain-size parameters and major-element concentrations of the non-phyllosilicate fraction are given in Table 3.

The results of this study were evaluated with respect to maximum temperature as calculated from the vitrinite reflectance data of Houseknecht and Matthews (1985). These values were obtained using the relation reported by Barker (1988) assuming a minimum time period at maximum burial temperatures.

DISCUSSION

Phyllosilicate Fraction

A decrease in the amount of silica present in the clay-mineral fraction with increasing thermal maturity is apparent in Figure 2. This decrease appears linear, and continues to maximum temperatures near 300°C, a temperature beyond any reported for complete illitization of smectite. The linear decrease of silica in this fraction suggests that recrystallization of illite into mica with lower silica concentrations continues after illitization. Ideal illite contains 55% SiO₂, while muscovite contains 45% SiO₂. The lower SiO₂ values from the clay-mineral fraction of

| Table 2.—W I L L E R  R O C K  C O N C E N T R A T I O N  D A T A |
|-----------------|------|------|------|------|------|------|------|------|------|
| Spl | SiO₂ | TiO₂ | Al₂O₃ | FeO | MnO | MgO | CaO | Na₂O | K₂O |
| 15  | 63.46 | 0.88 | 20.27 | 7.92 | 0.03 | 2.26 | 0.22 | 1.20 | 3.56 |
| 16  | 65.30 | 0.84 | 20.13 | 6.24 | 0.03 | 1.99 | 0.32 | 1.46 | 3.59 |
| 24  | 61.27 | 0.82 | 21.36 | 8.30 | 0.05 | 2.39 | 0.36 | 1.29 | 3.93 |
| 25  | 64.82 | 0.86 | 20.04 | 7.11 | 0.04 | 1.76 | 0.26 | 1.04 | 3.93 |
| 27  | 65.83 | 0.81 | 17.99 | 7.13 | 0.07 | 2.39 | 0.31 | 1.39 | 4.10 |
| 31  | 67.35 | 0.90 | 21.33 | 3.76 | 0.01 | 1.28 | 0.03 | 0.21 | 5.06 |
| 32  | 65.79 | 0.86 | 18.80 | 6.56 | 0.07 | 2.17 | 0.47 | 1.14 | 3.99 |
| 35  | 60.49 | 0.94 | 21.93 | 7.72 | 0.05 | 2.55 | 0.30 | 1.41 | 4.42 |
| 36  | 61.52 | 1.04 | 23.41 | 7.32 | 0.03 | 1.85 | 0.08 | 0.27 | 4.33 |
| 40  | 67.26 | 0.78 | 17.56 | 6.59 | 0.05 | 1.74 | 0.19 | 2.32 | 3.38 |
| 41  | 59.03 | 0.84 | 20.10 | 6.95 | 0.04 | 1.86 | 0.20 | 1.14 | 4.77 |
| 42  | 63.34 | 0.87 | 22.01 | 4.77 | 0.02 | 1.57 | 0.11 | 1.72 | 5.46 |
| 51  | 65.54 | 0.86 | 19.81 | 6.53 | 0.06 | 1.89 | 0.24 | 0.82 | 4.09 |
| 52  | 59.77 | 0.90 | 22.09 | 8.02 | 0.09 | 1.80 | 0.15 | 2.28 | 4.74 |
| 56  | 62.60 | 0.87 | 20.06 | 7.33 | 0.07 | 2.05 | 0.26 | 2.70 | 3.89 |
| 57  | 63.22 | 0.88 | 20.09 | 7.94 | 0.04 | 2.22 | 0.23 | 1.56 | 3.62 |
| 65  | 70.52 | 0.72 | 17.54 | 3.14 | 0.07 | 2.17 | 0.59 | 2.08 | 3.01 |
| 113 | 63.65 | 0.85 | 21.19 | 6.15 | 0.02 | 2.13 | 0.50 | 1.13 | 4.22 |
| 120 | 64.41 | 0.89 | 19.36 | 6.44 | 0.02 | 2.52 | 0.07 | 0.73 | 4.69 |
| 127 | 65.71 | 0.79 | 18.52 | 6.48 | 0.08 | 2.02 | 0.57 | 2.27 | 3.40 |
| 128 | 61.33 | 0.88 | 21.47 | 7.11 | 0.03 | 2.61 | 0.76 | 1.34 | 4.28 |

Major element oxides reported on a 100% anhydrous basis.
<table>
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<th>Sample Size</th>
<th>N</th>
<th>Flavor</th>
<th>OAT</th>
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<th>SWEET</th>
<th>OIL</th>
<th>FRIE</th>
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<th>TASTE</th>
<th>SALT</th>
<th>SUGAR</th>
<th>GRAIN SIZE</th>
<th>VITAMIN D</th>
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TABLE 3: MAJOR-FINETECH CONCENTRATIONS AND GRAIN-SIZE PARAMETERS OF THE NON-PROTIOSTOLIC FACTION
the Stanley shales are due to dilution by lower SiO₂-containing phases such as chlorite.

Because conventional X-ray techniques do not discriminate between illite and muscovite, a means to detect this transition was needed. As mentioned previously, illite contains more water in the interlayer than does mica. To drive off this interlayer water, the samples were heated to 500°C and X-rayed. The patterns were then compared to diffractograms of unheated samples. Loss of interlayer water will result in a decrease in the area of the 10-Å peak representing the basal spacing of the clay. The larger the difference in peak area, the more water that was lost during heating.

Figure 3 illustrates the good correlation between the loss of peak area and increasing thermal maturity. The relationship is linear, suggesting that interlayer water is continually expelled across this entire range of thermal maturity. To validate this interpretation, samples of muscovite from the University of New Orleans mineralogy collection and illite from the clay minerals repository were treated in a similar manner. The results are also shown on Figure 3 and are consistent with the results from the Stanley shale samples. To ensure that adsorbed water was not causing this phenomenon, weight loss after heating to 100°C overnight was compared to thermal maturity, and no correlation was found. Because a major difference between illite and muscovite is the amount of interlayer water present, the expulsion of this water during late diagenesis reflects recrystallization of illite into muscovite.

Differences in the character of the phyllosilicate fraction with increasing temperature are also seen in SEM as shown in Figures 4A and 4B. Samples from areas of lower thermal maturity are of much finer grain size than those of higher maturity. This result is similar to the findings of Guthrie (1985).

Non-phyllosilicate Mineral Fraction

Corresponding changes in the non-phyllosilicate fraction relative to the phyllosilicate fraction were observed. A possible increase in the percentage of the non-phyllosilicate fraction with increasing temperature is shown on Figure 5. This has been reported to be a consequence of quartz growth during clay-
Table 4—Correlation Coefficients for Grain-Size Parameters versus Tmax.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>corr. coef.</th>
<th>sig. level</th>
</tr>
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<tbody>
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<td>Mean</td>
<td>-0.21</td>
<td>&gt;95%</td>
</tr>
<tr>
<td>phi(95%)</td>
<td>-0.49</td>
<td>&gt;95%</td>
</tr>
<tr>
<td>phi(89%)</td>
<td>-0.52</td>
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<td>phi(75%)</td>
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<td>phi(50%)</td>
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<td>phi(16%)</td>
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<tr>
<td>phi(5%)</td>
<td>-0.27</td>
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</table>

Fig. 6.—Grain-size parameters versus correlation coefficients with maximum temperature. The finest sizes correlate the strongest, suggesting authigenic growth in the finest size fractions with increasing temperature.

Fig. 7.—Percent polycrystalline quartz in the 10- to 20-µm fraction versus temperature. There is a continuous increase in the percentage of polycrystalline grains from about 10% to 60% across the range in maximum temperature. The coefficient of causation ($r^2 = 0.61$) suggests that over 60% of the percentage of polycrystalline grains can be explained by thermal maturity. The remaining 40% is due to other factors, such as the initial percentage when deposited.

Mass Balance Between Grain-size Fractions

From the regression line of Figure 2 the amount of silica released from the clay-mineral fraction across the level of diagenesis encountered in the Stanley is approximately 10%. This amount coincides with the quantity of silica expected to be released during recrystallization of illite to muscovite (Table 1). This compares very well with the total amount of material gained in the non-phylllosilicate fraction (Fig. 9). Because over 90% of this fraction is composed of quartz and chert, it seems apparent that the gain in silica in this fraction is directly related to the loss of silica in the phyllosilicate fraction.

Mineral Diagenesis (Totten and Blatt, 1993). Strong evidence for this conclusion is the increased percentage of fine grained quartz in the samples of high thermal maturity. Table 4 lists the correlation coefficients for different grain-size parameters with maximum temperature. Each grain-size parameter reflects a specific position on a cumulative grain-size frequency curve. For example, the phi (95%) corresponds to 95% on the cumulative curve and represents the finest 5% of the sediment. The parameters representing the finest grain-sizes correlate with maximum temperature much better than those representative of coarser size. These correlations decrease in a systematic fashion with increasing grain-size; in fact, the correlation coefficients themselves correlate extremely well with the grain-size parameters (Fig. 6).

In addition to the quartz fraction increasing in both size and amount during shale diagenesis, this fraction also exhibits an increase in polycrystallinity. This is best seen in the 10- to 20-µm grain-size fraction (Fig. 7). Extension of the regression line to 300°C results in a value of 60% polycrystalline quartz grains. This is consistent with the character of quartz released from schists (Blatt, 1967). The composite nature of quartz grains from samples of high thermal maturity as opposed to the monocrystalline quartz of low thermal maturity is seen in Figure 8.
SILICA FROM THE ILLITE TO MUSCOVITE TRANSFORMATION DURING LATE-STAGE DIAGENESIS OF SHALES

The implication of the mass balance within these rocks suggests that silica is not exported from shales during diagenesis. This is in agreement with whole-rock chemical data from the same rocks (Totten and Blatt, 1993). Even though the continued recrystallization of illite to muscovite releases more silica than previously expected during shale diagenesis, it appears that the silica remains within the shale system and is not exported. While this does not solve the problem of sources of silica for sandstone cementation, it does contribute to our understanding of the origin of silt-sized quartz. A considerable amount of silica precipitates within mudrocks during diagenesis, all within the silt-sized range. This supports the suggestion by Blatt (1987) that metapelites are the source of much of the silt-sized quartz population.

CONCLUSIONS

Results suggest that illite is not the end-member mineral that it is often considered to be, but is a transition phase as clay recrystallizes into mica. Because of the similarity between illite and muscovite diffraction patterns, this is often overlooked. To erase some of the confusion regarding a precise definition for illite, a chemical definition should be applied, and illite could be defined as a high silica, low potassium, more hydrous precursor to muscovite.

In the absence of chemical analyses of the phyllosilicate fraction, a method to determine the relative proportions of illite and muscovite is to measure the difference in peak area of basal plane reflections both before and after heating the sample to 500°C. The varying effect of this treatment probably reflects dehydroxylation of interlayer water. Because illite holds more water than muscovite, the amount of peak area change is greater after heating an illite. Our results show a continual decrease in the area of this peak with increasing thermal maturity, which we interpret as varying degrees of recrystallization of illite to muscovite. One can imagine a mixed-layer illite/muscovite analogous to mixed-layer illite/smectite.

Coincident with the continual expulsion of water after expandable clays are gone, the phyllosilicate fraction exhibits a linear decrease in silica. This is also consistent with recrystallization of illite to muscovite.

During clay diagenesis, silica released precipitates as authigenic solid silica. Both the percentage of quartz and the grain-size of this fraction increase with increasing thermal maturity. An approximate mass-balance for silica exists between the two fractions, supporting previous work that interpreted these shales as closed systems during diagenesis. In addition to the solid-silica fraction increasing in size and amount, the degree of polycrystallinity dramatically increases during shale diagenesis. The percentage of composite grains approaches the percentage reported from schists. The growth of solid-silica in this fraction is consistent with metapelites being a major source of silt-size quartz.

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