# ORIGIN OF AN UNDERCLAY AS REVEALED BY VERTICAL VARIATIONS IN MINERALOGY AND CHEMISTRY

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**Abstract**—Regular vertical variations in mineralogy and chemistry indicate that underclay beneath the Herrin (No. 6) coal in southwestern Illinois has undergone *in situ* alteration. Alteration resulted from the downward movement of hydrogen ions, as indicated by the progressive leaching of acid-sensitive minerals adjacent to the coal. Mineralogical trends observed in the underclay with increasing depth below the coal include: (1) a decrease in the expandability of mixed-layer illite/smectite (I/S); (2) an increase in the amount of ordered I/S with respect to randomly interstratified I/S; (3) an increase in the amount of discrete illite with respect to expandable clays; and (4) an increase in chlorite and calcite. Ordered I/S is the dominant mixed-layer clay where calcite is present, but randomly interstratified I/S dominates where calcite is absent. The pH of the underclay also increases with depth. These trends are consistent with an origin by acid leaching of a preexisting mineral assemblage that included illite, chlorite, and calcite. Other acid-alteration trends may be expected for different precursor minerals and for different leaching intensities and durations.

**Key Words**—Acid leaching, Coal, Expandability, Illite/smectite, Mixed layer, Underclay.

## INTRODUCTION

Underclay is an argillaceous rock that is commonly found beneath beds of coal. Like other seat rocks, it is generally non-bedded, light colored, contains plant roots (*Stigmaria*) and randomly oriented slickensides, and may be calcitic near its base (Huddle and Patterson, 1961). A sharp contact exists between underclay and the overlying coal, but the lower contact commonly is gradational (Gresley, 1887). Underclay is generally several centimeters to a meter thick, but may be as much as 6 m or more (Grim and Allen, 1938). It occurs in cyclothems that represent depositional sequences repeated throughout the Pennsylvanian (Weller, 1930, 1931, 1956; Wanless, 1931, 1947).

Theories on the origin of underclay differ as to whether underclay has undergone post-depositional alteration. On the basis of a lack of strong vertical trends in mineralogy and the presence of unweathered feldspars throughout the profiles, Grim and Allen (1938) and Schultz (1958) proposed that underclay accumulated with its present mineralogical composition. Other authors indicated that *in situ* alteration is an important process in underclay formation, but disagreed as to when and how this alteration took place. Logan (1842) and Lyell (1841), for example, implied that underclay is a fossil soil upon which coal-forming plants grew. McMillan (1956) suggested that underclay is a fossil gley, a bleached layer of soft clay that forms beneath peat by the leaching action of organic compounds, and Stout (1931) suggested that underclay formed from sed-iments that were altered as they were laid down in a coal swamp.

The fossil-soil theory came under attack as early as 1887, when Gresley pointed out the existence of Stigmarian clays without an overlying coal bed. He also noted the sharp contact between the underclay and the coal, and the fact that *Stigmaria* never pass into the coal. Weller (1930, 1931) also noted that both the underclay and the coal may occur alone and that the thickness of coal is not necessarily related to the thickness of its underclay. Thus, he concluded that the two are unrelated, and that underclay is a fossil soil that formed prior to peat accumulation. In support of this idea, Wanless (1931) found that underclay has the characteristics of a poorly drained soil, and Huddle and Patterson (1961) suggested that *in situ* leaching of substratum may have occurred at any time before peat accumulation, during peat accumulation, or during coalification.

Because mixed-layer clays can be sensitive indicators of environmental conditions (e.g., Eberl and Hower, 1976), the present research was undertaken to determine if a detailed study of mixed-layering in underclay could help establish an acceptable theory on the origin of underclay.

## SAMPLING AND EXPERIMENTAL TECHNIQUES

The Herrin (No. 6) coal member of the Carbondale Formation (Kewanee Group) is the principal economic
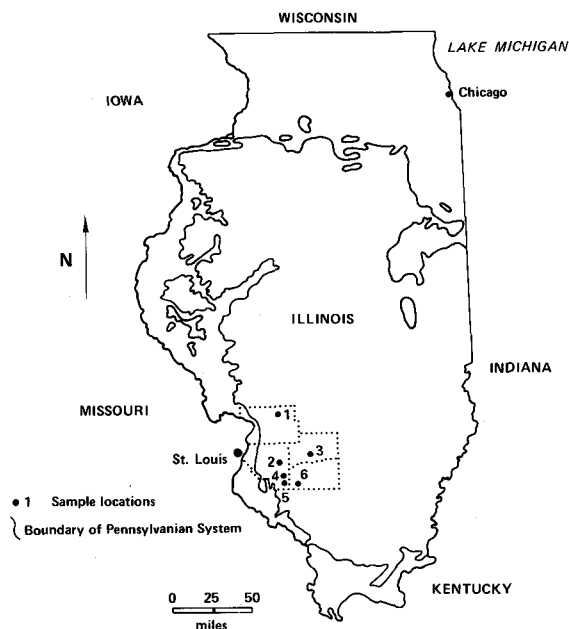
Figure 1.　Core locations. The dotted lines are county boundaries.

coal in Illinois. This seam is present throughout most of the Pennsylvanian rocks in the Illinois Basin; it averages about 2 m thick, but may be more than 4 m thick locally (Hopkins and Simon, 1975). In the study area, the No. 6 coal rests on an underclay that is 1.5 to 2 m thick. Detailed vertical sequences were collected from six core sections from Clinton, Madison, St. Clair, and Washington Counties (Figure 1). Detailed core locations and descriptions are given in Stepusin (1978). A sampling interval of 1–6 cm was used in Core 1, 10–15 cm in the remaining cores. Each core was sampled from directly below the coal/underclay contact throughout the entire clay sequence, and into underlying nodular limestone.

Oriented clay samples were prepared for X-ray powder diffraction (XRD) analysis by allowing suspensions of the $<2$-$\mu$m size fraction to dry at room temperature on glass slides. Calcareous samples were treated with 5% acetic acid prior to dispersion to remove excess carbonate. A comparison between acidified and non-acidified samples showed that this treatment did not affect the clay minerals and resulted in an easier dispersion.

Quantitative and semiquantitative XRD analysis of clays in the $<2$-$\mu$m fraction and qualitative XRD analysis of whole-rock samples were made with a General Electric XRD-5 diffraction unit[1] using Ni-filtered CuK$\alpha$ radiation and a log scale, and a Philips Norelco diffraction unit using CuK$\alpha$ radiation with a graphite monochromator and a linear scale. After solvation for several days with ethylene glycol, all $<2$-$\mu$m samples were first

scanned on a log scale at $2°2\theta$/min between $2°$ and $35°2\theta$ to obtain an overall picture of the clay mineral assemblage, and then at the same speed on a linear scale between $14°$ and $21°2\theta$ to determine the exact position of the $(002)_{10}/(003)_{17}$ and $(002)_{10}/(005)_{27}$[2] reflections of mixed-layer illite/smectite (I/S). The latter scans were made two or three times to check reproducibility. Slides were heat-treated for 1 hr at 375°C and examined while hot. This temperature produced complete collapse of expandable layers (Austin and Leininger, 1976), without affecting the intensity of chlorite reflections. Eight $<2$-$\mu$m samples from Core 1 were saturated with $K^+$, air-dried, and then treated with ethylene glycol vapor for 2 days prior to XRD analysis to test for the presence of highly charged expanding layers. Each $K^+$-saturated sample was scanned at $2°2\theta$/min on a linear scale between $2°$ and $35°2\theta$. Randomly oriented powders of 12 whole rock samples from Cores 1, 2, and 6 were X-rayed from $4°$ to $60°2\theta$ at $2°2\theta$/min to identify nonclay minerals.

Clay minerals in the $<2$-$\mu$m fraction were identified from $00\ell$ reflections determined from XRD patterns of ethylene glycol-treated samples (Brown, 1961). Mixed-layer I/S was identified according to the method of Reynolds and Hower (1970). The percentages of kaolinite + chlorite, illite, and expandable phases in the $<2$-$\mu$m fraction were determined semiquantitatively by a procedure modified from Griffin (1970) by Ward (1977), using peak heights rather than peak areas, because peak heights gave better reproducibility. Expandable phases include all clay layers that expand to more than 10 Å following glycolation and that collapse to 10 Å upon heating to 375°C. These phases include smectite, mixed-layer I/S, and vermiculite.

Chemical analyses for major and minor elements were obtained for 44 whole-rock samples from Cores 1, 2, and 6, and for eight $<2$-$\mu$m samples from Core 1. Samples were analyzed by X-ray fluorescence according to methods described in Shimp et al. (1970). Analytical values are reported in detail in Stepusin (1978). Measurements of underclay pH were made with an Orion model 407A specific-ion meter, by adding 1 g of ground clay taken from various depths in Core 1 to 10 ml of distilled water, stirring, and measuring pH after 1 min.

## RESULTS

### Clay mineralogy of Core 1

Variations in expandability and type of interlayering found with depth for Core 1 are shown in Table 1 and

[1] Trade-names are used for descriptive purposes only and do not imply endorsement by the U.S. Geological Survey.

[2] $(002)_{10}/(003)_{17}$ refers to the XRD peak that results from constructive interference between the 002 reflection of 10-Å layers and the 003 reflection of 17-Å layers in the randomly interstratified mixed-layer clay. This peak migrates with changing proportions of the layers. Similarly, the 002 reflection of 10-Å layers interfere with the 005 reflection of 27-Å layers in IM ordered I/S (Reynolds and Hower, 1970).
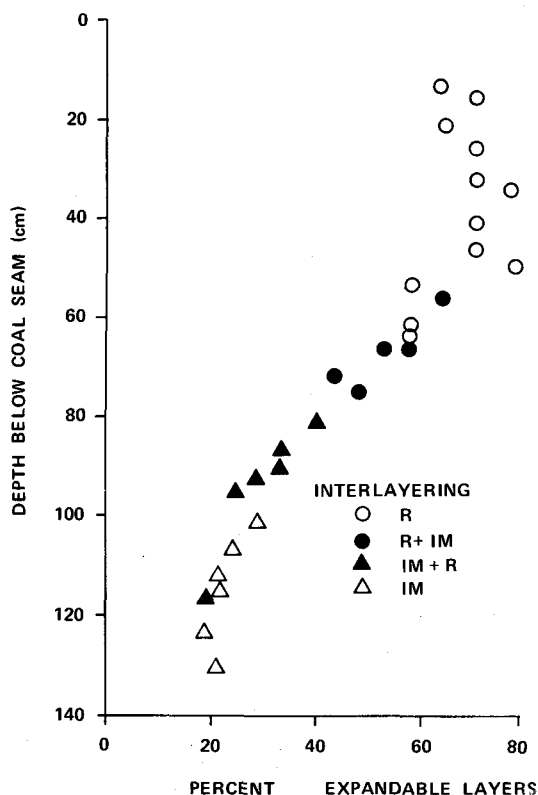
Figure 2.   Relationship between depth below coal, percentage of expandable layers, and type of interlayering for illite/smectite from the <2-$\mu$m size fraction of Core 1. See Table 1 footnotes for an explanation of interlayering symbols.
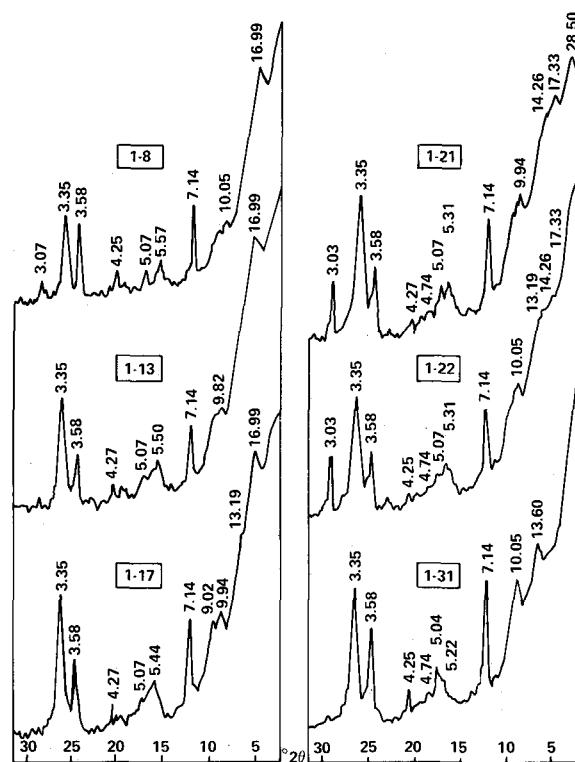


Figure 3.   X-ray powder diffractograms showing trends in clay mineral structure in Core 1. Samples were the oriented, ethylene glycol-treated, <2-$\mu$m size fraction, and were run on a log scale using CuK$\alpha$ radiation.

Figure 2. Throughout an interval of less than 1.5 m, expandability decreases from generally >70% for samples near the coal to about 20% for those at the base of the section. Accompanying this decrease is a change in the type of interlayering. Where the expandability exceeds 40%, randomly interstratified I/S (R) dominates, as is shown by an intense XRD peak at about 17 Å, with non-integral higher order peaks (Figure 3, runs 1-8, 1-13, and 1-17). With increasing depth, however, the intensity of the 17-Å peak decreases, and diffraction peaks at about 13.5 Å, and, in some samples at about 27 Å, intensify (Figure 3, runs 1-21, 1-22, and 1-31). These two peaks are characteristic of nearest-neighbor (IM) ordered I/S (Reynolds and Hower, 1970).

As this change in the type of interstratification takes place, IM and R phases coexist, as is evident in log run 1-21 (Figure 3). Identification of the IM phase commonly is difficult, because the $(002)_{27}/(001)_{10}$ peak at about 13.5 Å is seen only as a break in slope on the shoulder of the 17-Å peak (Figure 3, run 1-17). Further evidence that IM and R phases coexist, however, is given in linear runs made between 14° and 21°2$\theta$ (Figure 4). Here, three peaks are common: the $(002)_{10}$ of discrete illite at 5.07 Å and two peaks between 5.07 Å and

5.61 Å, the $(002)_{10}/(003)_{17}$ of a random phase, and the $(005)_{27}/(002)_{10}$ of an ordered phase (Figure 4, runs 1-13 and 6-13).

Semiquantitative XRD analysis of the <2-$\mu$m fraction (Table 1) shows an inverse relationship between discrete illite and expandable phases. Discrete illite increases from about 20% beneath the coal to more than 40% at the base of the core, whereas expandable phases decrease from a high of nearly 80% to about 40%. In addition, the illite/(illite + expandable phases) ratio is inversely related to the percentage of expandable layers.

Semiquantitative data show a slight decrease in the percentage of kaolinite + chlorite with depth in samples 1-1 through 1-23 (Table 1). Chlorite is absent close to the coal and occurs only in samples 1-21 through 1-31: the $(003)_{14}$ peak of chlorite at 4.74 Å is seen in Figure 3, runs 1-21, 1-22, and 1-31, but not in samples closer to the coal.

Upon K$^{+}$-saturation and treatment with ethylene glycol (Figure 5), the 17-Å peak of randomly interstratified material (run 1-18) migrates to between 14 and 14.7 Å, and the 13.6-Å peak of ordered I/S (run 1-26) migrates to 13 Å. Both migrations indicate a decrease in expandability and are accompanied by a sharpening of the

Table 1.　Clay mineralogy as a function of depth for cores 1–6.

| Sample | Depth below coal (cm) | Expandability of dominant illite/smectite (%) | Type of interlayering | Kaolinite + chlorite (%) | Illite (%) | Expandable phases (%) |
|---|---|---|---|---|---|---|
| | | | Core 1 | | | |
| 1-1 | 0–2 | nd[1] | nd | 18 | 24 | 58 |
| 1-2 | 6–7 | nd | nd | 18 | 24 | 58 |
| 1-3 | 13–14 | 64 | R[2] | 10 | 15 | 75 |
| 1-4 | 15–16 | 71 | R | 12 | 17 | 71 |
| 1-5 | 20–21 | 64 | R | 8 | 12 | 80 |
| 1-6 | 25–26 | 71 | R | 16 | 16 | 68 |
| 1-7 | 31–32.5 | 71 | R | 14 | 18 | 68 |
| 1-8 | 33–34 | 78 | R | 15 | 17 | 68 |
| 1-9 | 40–41 | 71 | R | 14 | 16 | 70 |
| 1-10 | 45–46 | 71 | R | 12 | 14 | 74 |
| 1-11 | 49–50 | 78 | R | 17 | 15 | 68 |
| 1-12 | 52–53 | 58 | R | 10 | 22 | 68 |
| 1-13 | 55–56 | 64 | R + IM[3] | 9 | 19 | 72 |
| 1-14 | 60–61 | 58 | R | 9 | 15 | 76 |
| 1-15 | 63–63.5 | 58 | R | 9 | 20 | 71 |
| 1-16 | 64–65 | 58 | R + IM | 12 | 17 | 71 |
| 1-17 | 65–65.5 | 53 | R + IM | 12 | 17 | 71 |
| 1-18 | 70–71 | 44 | R + IM | 11 | 19 | 70 |
| 1-19 | 75–76 | 48 | R | 12 | 22 | 66 |
| 1-20 | 80–81 | 40 | IM + R[4] | 12 | 23 | 65 |
| 1-21 | 85–86 | 33 | IM + R | 10 | 25 | 65 |
| 1-22 | 88–89 | 33 | IM + R | 11 | 25 | 64 |
| 1-23 | 90–91 | 28 | IM + R | 12 | 25 | 63 |
| 1-24 | 94–96 | 24 | IM + R | 15 | 27 | 58 |
| 1-25 | 100–101 | 28 | IM[5] | nd | nd | nd |
| 1-26 | 105–108 | 24 | IM | 16 | 31 | 53 |
| 1-27 | 110–113 | 21 | IM | 15 | 33 | 52 |
| 1-28 | 114–115 | 21 | IM | 17 | 32 | 51 |
| 1-29 | 115–118.5 | 19 | IM + R | 16 | 35 | 49 |
| 1-30 | 122–125 | 19 | IM | 15 | 44 | 41 |
| 1-31 | 129–132.5 | 21 | IM | 19 | 41 | 40 |
| | | | Core 2 | | | |
| 2-1 | 0–3 | 58 | R | 8 | 23 | 69 |
| 2-2 | 10–12 | 58 | R | 6 | 27 | 67 |
| 2-3 | 16–17 | 53 | R + IM | 7 | 26 | 67 |
| 2-4 | 24–25 | 53 | R | 5 | 31 | 64 |
| 2-5 | 29–30 | 44 | R + IM | 9 | 29 | 62 |
| 2-6 | 40–41 | 44 | R + IM | 6 | 33 | 61 |
| 2-7 | 50–51 | 44 | R + IM | 6 | 34 | 60 |
| 2-8 | 60–61 | 40 | IM + R | 5 | 35 | 60 |
| 2-9 | 70–71 | 28 | IM + R | 5 | 39 | 56 |
| 2-10 | 80–81 | 24 | IM + R | 7 | 31 | 62 |
| 2-11 | 90–91 | 24 | IM | 5 | 50 | 45 |
| | | | Core 3 | | | |
| 3-1 | 0–1 | 33 | IM | 5 | 30 | 65 |
| 3-2 | 10–11 | 44 | R + IM | 6 | 32 | 62 |
| 3-3 | 20–21 | 44 | R + IM | 5 | 33 | 62 |
| 3-4 | 30–31 | 40 | IM + R | 6 | 36 | 58 |
| 3-5 | 40–41 | 33 | IM + R | 10 | 34 | 56 |
| 3-6 | 50–51 | 28 | IM | 7 | 36 | 57 |
| 3-7 | 61–62 | 24 | IM | 8 | 38 | 54 |
| 3-8 | 70–71 | 21 | IM | 9 | 43 | 48 |
| 3-9 | 80–81 | 24 | IM | 9 | 44 | 47 |
| 3-10 | 90–91 | 21 | IM | 6 | 44 | 50 |
| 3-11 | 110–112 | 21 | IM | 7 | 46 | 47 |
| 3-12 | 119–120 | 21 | IM | 9 | 44 | 47 |
| 3-13 | 140–141 | 19 | IM | 10 | 48 | 42 |

Table 1. Continued.

| Sample | Depth below coal (cm) | Expandability of dominant illite/smectite (%) | Type of interlayering | Kaolinite + chlorite (%) | Illite (%) | Expandable phases (%) |
|--------|------------------------|----------------------------------------------|------------------------|---------------------------|-------------|------------------------|
| | | | Core 4 | | | |
| 4-1 | 0–5 | 33 | IM + R | 9 | 34 | 57 |
| 4-2 | 15–16 | 44 | R | 3 | 35 | 62 |
| 4-3 | 30–31 | 44 | R + IM | 3 | 36 | 61 |
| 4-4 | 45–46 | 44 | R + IM | 3 | 33 | 64 |
| 4-5 | 65–67 | 40 | IM + R | 2 | 45 | 53 |
| 4-6 | 75–76 | 28 | IM | 1 | 32 | 67 |
| 4-7 | 89–90 | 28 | IM | 1 | 34 | 65 |
| 4-8 | 105–106 | 28 | IM | 3 | 43 | 54 |
| 4-9 | 114–115 | 28 | IM | 4 | 38 | 58 |
| | | | Core 5 | | | |
| 5-1 | 0–2 | 21 | IM + R | 4 | 43 | 53 |
| 5-2 | 15–16 | 28 | IM + R | 3 | 36 | 61 |
| 5-3 | 29–30 | 33 | IM + R | 2 | 33 | 65 |
| 5-4 | 45–46 | 33 | IM + R | 2 | 31 | 67 |
| 5-5 | 59–60 | 21 | IM | 1 | 42 | 57 |
| 5-6 | 75–76 | 28 | IM + R | 1 | 46 | 53 |
| 5-7 | 91–92 | 21 | IM | 2 | 52 | 46 |
| 5-8 | 105–107 | 24 | IM | 1 | 47 | 52 |
| 5-9 | 115–116 | 24 | IM | 1 | 43 | 56 |
| 5-10 | 135–137 | 19 | IM | 2 | 77 | 21 |
| | | | Core 6 | | | |
| 6-1 | 0–2 | 53 | R + IM | 2 | 34 | 64 |
| 6-2 | 7–8 | 53 | R + IM | 2 | 28 | 70 |
| 6-3 | 14–15 | 53 | R | 1 | 27 | 72 |
| 6-4 | 29–30 | 48 | R + IM | 1 | 21 | 78 |
| 6-5 | 45–46 | 33 | IM + R | 5 | 33 | 62 |
| 6-6 | 59–60 | 24 | IM + R | 2 | 28 | 70 |
| 6-7 | 75–76 | 21 | IM + R | 3 | 31 | 66 |
| 6-8 | 89–90 | 24 | IM | 1 | 43 | 56 |
| 6-9 | 105–107 | 21 | IM | 1 | 57 | 42 |
| 6-10 | 121–122 | 21 | IM | 1 | 71 | 28 |
| 6-11 | 140–142 | 21 | IM | 1 | 54 | 45 |
| 6-12 | 155–156 | 24 | IM | 1 | 62 | 37 |
| 6-13 | 170–171 | 24 | IM + R | 1 | 58 | 41 |
| 6-14 | 180–181 | 24 | IM | 1 | 48 | 51 |

[1] nd = no data available.
[2] R = randomly interstratified illite/smectite.
[3] R + IM = random illite/smectite is dominant.
[4] IM + R = ordered illite/smectite is dominant.
[5] IM = ordered illite/smectite.

002 reflection of illite at about 5 Å, indicating that the mixed-layer clays contain highly charged (vermiculite-like) expandable interlayers.

## Clay mineralogy of Cores 2–6

XRD data on the five other cores generally support the observations made on Core 1. Similar trends in expandability, in type of interlayering, and in mineral abundance are seen in data for Cores 2 and 6 (Table 1). In both cores, the percentage of expandable layers decreases from more than 50% below the coal to 24% at the base of the core. At greater expandabilities, randomly interstratified I/S is the dominant phase, with IM

ordering appearing as the percentage of expandable layers decreases. During this transition, R and IM phases coexist. Semiquantitative XRD data for Core 2 show an increase in discrete illite and a decrease in expandable phases with depth below the coal. A similar, but more erratic trend, is seen in data for Core 6.

The data for Core 3 have less of a trend, with expandability decreasing from greater than 40% (excluding sample 3-1) to about 20%, with a corresponding increase in discrete illite and decrease in expandable phases. Trends in data for Cores 4 and 5 are less distinct. In Core 4, the percentage of expandable layers decreases from 44% to 28% (excluding sample 4-1), and
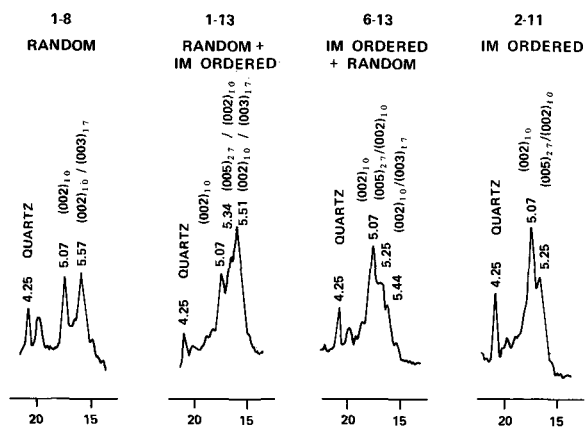
Figure 4. X-ray powder diffractograms showing mixed-layer peaks used to determine the percentage of expandable layers in illite/smectite. Samples were the oriented, ethylene glycol-treated, <2-$\mu$m size fraction, and were run on a linear scale using CuK$\alpha$ radiation. The bottom axis is in °2$\theta$.
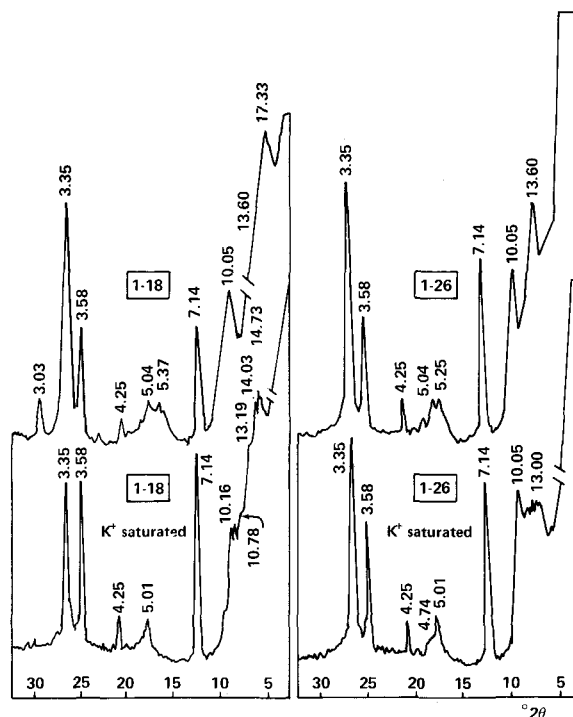


Figure 5. X-ray powder diffractograms of oriented, <2-$\mu$m sample prior to K-saturation (above) and after K-saturation (below). All samples were treated with ethylene glycol and run on a linear scale using CuK$\alpha$ radiation.

ordering type changes from R to IM, with the two phases coexisting in between. The amounts of discrete illite and expandable phases show no regular variation with depth. In Core 5, no systematic variations in either expandability or in amounts of discrete illite and expandable phases are seen, but the type of interlayering changes from IM + R to IM with increasing depth.

*Non-clay mineralogy*

XRD patterns of randomly oriented powders indicate that quartz, calcite, dolomite, plagioclase feldspar, gypsum, and pyrite are present in the underclays. Quartz and calcite dominate these assemblages.

*Chemical analyses*

From chemical data presented in Stepusin (1978), CaO increases markedly with depth below the coal. Because nearly all of the CaO is present in the mineral calcite, a plot of the percentage of CaO vs. depth for Core 1 (Figure 6) indicates the change in calcite content of the core. The CaO content is minimal to a depth of about 70 cm; below this depth calcite is more abundant. At about this depth, a change in ordering type occurs, from R-dominant to IM-dominant (see Figure 2 and Table 1). Similar trends are seen in Cores 2 and 6.

Chemical trends can be seen for other elements after attributing all of the CaO to calcite and then normalizing the analyses to 100%. An inverse correlation exists between K$_2$O and the percentage of expandable layers in Core 1, particularly in the <2-$\mu$m samples (Figure 7), as is expected from illite chemistry. Whole rock samples from Cores 2 and 6, however, do not show distinct trends in K$_2$O. Also, no correlation exists between the SiO$_2$/Al$_2$O$_3$ ratio and the percentage of expandable layers for the whole rock or <2-$\mu$m fractions of any cores.

*Underclay pH*

The general increase in pH with depth for the underclay from Core 1 (Figure 8) indicates a source for hydrogen ions from above. The sudden increase in pH from 5.7 to 8.4 at a depth of about 70 cm approximately marks the appearance of calcite and a change in dominant I/S ordering type, as discussed above.

## DISCUSSION

Several depth-dependent mineralogic trends exist in the underclay of the Herrin (No. 6) Coal. With increasing depth below the coal: (1) expandability of I/S decreases; (2) the amount of ordered I/S increases with respect to the random phase; (3) discrete illite increases while the amount of expandable phases decreases; (4) chlorite increases; and (5) calcite increases. These trends, especially the regular decrease in the percentage of expandable layers with depth (Figure 2), are not consistent with normal sedimentation, but are characteristic of *in situ* alteration.

It is unlikely that this alteration is related to either a temperature or a pressure gradient, because the trends occur within an interval of only 1.5 m. More likely, they result from a chemical gradient, such as changing pH. Such a gradient is shown in Figure 8, although minimal pHs observed in the laboratory may partly result from
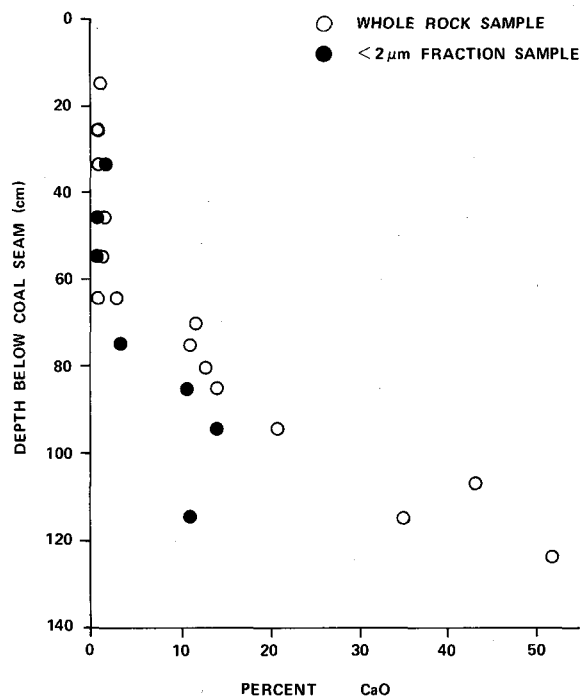
Figure 6.   Relationship between the percentage of CaO and depth below coal in Core 1.



Figure 7.   Relationship between percent $K_2O$, percentage of expandable layers, and interlayering type for the whole rock and the $<2$-$\mu m$ size fraction in Core 1. See Table 1 footnotes for an explanation of interlayering symbols.

oxidation of pyrite in the core during storage. Huddle and Patterson (1961), however, reported underclay pHs as low as 3.9. Indeed, all the mineralogical trends observed in the underclay are consistent with an origin by acid leaching of sediment that initially contained illite, (or I/S of low expandability), chlorite, and calcite. The initial sediment probably also contained other minerals, such as quartz, kaolinite, and feldspar, that were not necessarily involved in the acid reactions. Absence of the acid-sensitive minerals calcite and chlorite from the region adjacent to the coal is thus explained. Acids would also leach interlayer $K^+$ from illite, thereby forming mixed-layer clay.

Leaching $K^+$ from illite would initially occur under gentle conditions, with calcite present to buffer the pH, and would lead to the formation of some IM-ordered clay. In this near-neutral pH zone, IM-ordered I/S and discrete illite would coexist. It might be argued that illite would not be leached in the presence of calcite, but Grim and Allen (1938) reported that illite in weathered Illinois glacial deposits was appreciably altered before all of the calcite had been dissolved. In support of this mechanism for the formation of IM-ordered I/S, Norrish (1973) reported that $K^+$-depletion of micas leads to regular interstratification, and Churchman (1980) found IM-ordered material forming from mica in a New Zealand soil. In addition, illite leached by acid produces an IM-ordered structure (oral communication, J. Hower,
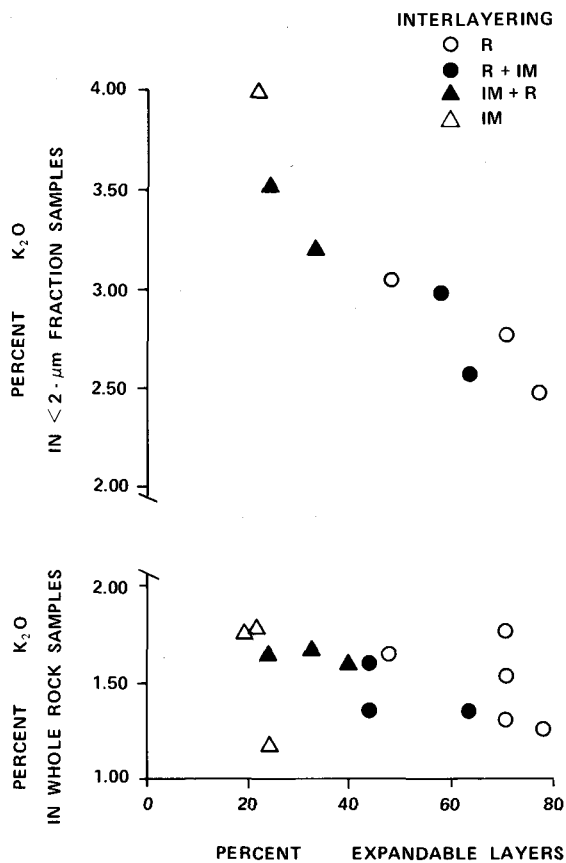
Dept. of Geology, University of Illinois, Urbana, Illinois).

Where most of the calcite had been destroyed, a condition that would exist at the top of the underclay, more intense leaching conditions would prevail, leading to further expansion of the ordered phase and to its eventual transformation into a random phase. The small reversal in the mixed-layering trend adjacent to the coal (samples 1-3, 3-1, 4-1, and 5-1, Table 1), if it is not simply experimental error, may represent a local increase in $K^+$ derived from vegetal matter in the adjacent coal. Such a $K^+$ increase also was suggested by Huddle and Patterson (1961) based on the refractory properties of underclay, and by McMillan (1956) based on underclay chemistry.

The tendency of some interlayers to collapse on K-saturation reveals that some of the expanding layers in the mixed-layer clays have a high layer charge, a result in harmony with an origin involving the leaching of $K^+$ from illite. All of the expanded layers are not expected to be reconstituted to illite on $K^+$-saturation,
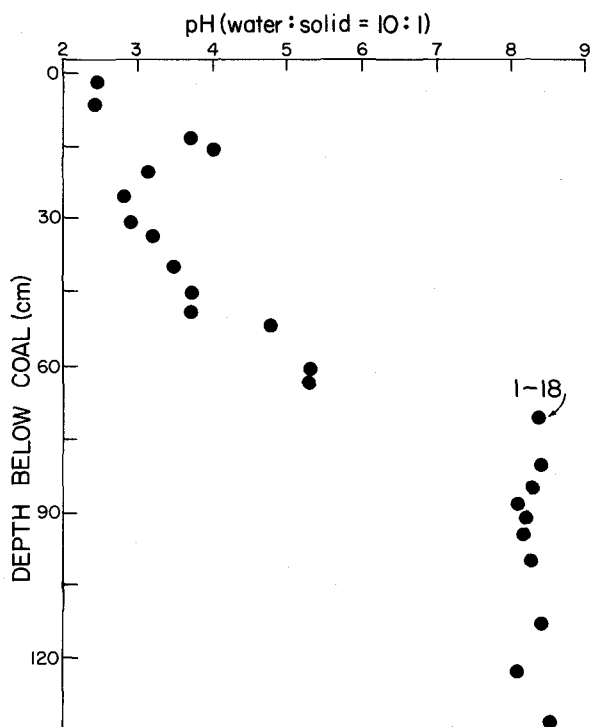
Figure 8. Relationship between depth below coal and underclay pH for the whole rock, Core 1.

include kaolinite, boehmite, and diaspore-rich underclays.

Plastic underclays may be the first stage in a leaching sequence in which boehmite and diaspore-bearing underclays are the last stage, as has been suggested by Huddle and Patterson (1961). These authors also emphasized the importance of plants to the leaching process, in that plants can exchange hydrogen ions for $K^+$, and then move $K^+$ upward to where it can be flushed away by swamp water and floods.

The time of alteration has not been distinguished. Did leaching occur during soil formation prior to swamp development, or did it occur under the coal swamp, or after burial, or during all three periods? If the measured underclay pHs (Figure 8) are indicative of present field conditions, some *in situ* acid alteration is still taking place today.

## ACKNOWLEDGMENTS

because various irreversible changes take place in the 2:1 layer as $K^+$ is leached, such as a loss of octahedral cations and structural distortions (Norrish, 1973). Inverse relationships between the abundance of illite and expandable phases and between the illite/(illite + expandable phases) ratio and expandability also support this mechanism for mixed-layer formation.

## CONCLUSION

Underclay beneath the No. 6 coal has undergone significant post-depositional alteration by acid leaching. This leaching overprints the original detrital mineralogy; hence, in addition to indicating *in situ* conditions, the composition of underclays also may reflect conditions in the sediment source area, as suggested by Schultz (1958) and may have responded to lateral variations in the basin of deposition, as suggested by Parham (1964) and Williams *et al.* (1968). The effect of thermal diagenesis, if any, on an underclay during its long burial history is unknown.

Different parent mineralogies will lead to alteration trends that differ from those described here, as will different leaching durations and intensities. With regard to the latter, the mixed-layer sequence found in this study is: illite → I/S ordered → I/S random, with increasing intensity. This series encompasses the plastic underclays. It may be correct to extend this series to

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**Резюме**—Регулярные вертикальные изменения минералогии и химии указывают на то, что подстилающая глина под Герринским (№ 6) углем в южно-западном районе Иллиноя подвергалась видоизменению на своем месте. Видоизменение было следствием движения ионов водорода вниз, на что указывает прогрессивное вымывание кислочувствительных минералов, прилегающих к углю. Минералогические тенденции, наблюдаемые в подстилающих глинах при увеличении глубины под углем, включают: (1) уменьшение расширяемости смешаннослойного иллита/смектита (И/С); (2) увеличение количества упорядоченного И/С по сравнению со случайно внутринапластованным И/С; (3) увеличение количества неявного иллита по сравнению с расширяющейся глиной и (4) увеличение содержания хлорита и кальцита. Упорядоченный И/С является преобладающей смешанно-слойной глиной, если присутствует кальцит, тогда как случайно внутринапластованный И/С преобладает, если кальцит отсутствует. С глубиной также увеличивается pH подстилающей глины. Эти тенденции находятся в согласии с происхождением путем кислого вымывания ранее существующих минеральных скоплений, включающих иллит, хлорит, и кальцит. Другие кислоизменяющие тенденции могут ожидаться для различных предвестников минералов и для различных интенсивностей и продолжительностей. [E.C.]

**Resümee**—Regelmäßige vertikale Änderungen in der Mineralogie und chemischen Zusammensetzung zeigen, daß der Basalton unter der Herrin-Kohle (No. 6) im südwestlichen Illinois eine *in situ* Umwandlung mitgemacht hat. Die Umwandlung war die Folge von sich abwärts bewegenden Wasserstoffionen, wie aus der zunehmenden Auslaugung der säureempfindlichen Minerale im an der Kohle angrenzenden Gestein hervorgeht. Mineralogische Trends, die in dem Basalton mit zunehmender Tiefe unter der Kohle beobachtet werden, sind: (1) eine Abnahme der Quellbarkeit der Illit/Smektit-Wechsellagerung (I/S); (2) eine mengenmäßige Zunahme einer regelmäßigen I/S in Bezug auf eine unregelmäßige I/S-Wechsellagerung; (3) eine mengenmäßige Zunahme von Illit an sich in Bezug auf quellbare Tone; und (4) eine Zunahme von Chlorit und Calcit. Eine regelmäßige I/S ist die vorherrschende Wechsellagerung, wenn Calcit vorhanden ist, eine unregelmäßige Wechsellagerung überwiegt jedoch dann, wenn Calcit fehlt. Der pH des Basaltones nimmt mit zunehmender Tiefe zu. Diese Trends stehen in Zusammenhang mit einer Entstehung, die durch eine saure Auslaugung der zuvor existierenden Mineralvergesellschaftung, die aus Illit, Chlorit, und Calcit bestand, bewirkt wurde. Andere Trends einer Säureumwandlung kann man bei anderen Ausgangsmineralen annehmen sowie bei anderen Auslaugungsintensitäten und anderer Auslaugungsdauer. [U.W.]

**Résumé**—Des variations verticales regulières de minéralogie et de chimie indiquent que la sous-argile sous le charbon Herrin (no. 6) en Illinois du sud-ouest a subi une altération *in situ*. L'altération a resulté du mouvement vers le bas d'ions hydrogène comme l'indique le lavement progressif de minéraux sensibles à l'acide adjacents au charbon. Les tendances minéralogiques observées dans la sous-argile proportionnellement à la profondeur sous le charbon comptent: (1) un décroissement de la capacité d'expansion d'illite/smectite (I/S) à couches melangées; (2) une augmentation de la quantité d'I/S ordonnée respectivement à l'I/S interstratifiée au hasard; (3) un accroissement de la quantité d'illite discrète par rapport aux argiles expansibles; et (4) une augmentation de chlorite et de calcite. L'I/S ordonnée et l'argile à couche melangée est dominante où la calcite est présente, tandis que l'I/S interstratifiée au hasard domine où la calcite est absente. Le pH de la sous-argile augmente aussi proportionnellement à la profondeur. Ces tendances sont compatibles avec une origine de lavement par un acide d'un assemblage minéral pré-existant qui comprenait de l'illite, de la chlorite, et de la calcite. D'autres tendances d'altération par un acide peuvent être attendues pour d'autres minéraux précurseurs et pour des intensités et des durées de lavement différentes. [D.J.]