Early Diagenetic Calcareous Coal Balls and Roof Shale Concretions from the Pennsylvanian (Allegheny Series)

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EARLY DIAGENETIC CALCAREOUS COAL BALLS AND ROOF SHALE CONCRETIONS FROM THE PENNSYLVANIAN (ALLEGHENY SERIES)

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Abstract. The calcareous concretions studied formed in different depositional environments associated with the Pennsylvanian cyclothem of east central Ohio. The late syngenetic to early diagenetic coal balls from Lower Freeport Coal in Jefferson County were formed by cellular permineralization of decaying terrestrial plants in a peat swamp. These coal balls, composed of 89.8% calcite, 7.6% pyrite, and 2.6% organic material, preserve six identifiable plant genera, Sphenophyllum, Stigmaria, Medulosae, Taxospermum, Psaronius, and Myeloxylon, as well as fusain and spores. The early diagnostic nodules from roof shale overlying Middle Kittanning Coal in Tuscarawas County, Ohio were formed by authigenic cementation of marine muds that contained brachiopods, including Mesolobus, Lingula, and several productid species, bryozoan fronds, ostracodes, gastropods, crinoids, cephalopods, as well as coprolites, terrestrially-derived fusain, root casts, and megaspore coats. These nodules, composed of 84.5% calcite, 11.3% illite, 1.5% quartz, 0.9% pyrite, and 0.5% organic material, have calcite-filled septarian fissures.

Coal balls, or concretions that formed in peat, and roof shale nodules preserve a portion of sediment that was not severely compressed before lithification. As such, they provide a key for deciphering the diagenetic history of the strata in which they formed and also for recognizing original depositional features. Although roof shale nodules, such as those found in the blue shale overlying the Middle Kittanning Coal (No. 6) in Tuscarawas County, are common in near shore shales of Pennsylvanian age rocks of Ohio, coal balls have been described from coals only in the Monongahela (Good and Taylor 1974) and Conemaugh Series (Rothwell 1976) in Ohio. My study compares the diagenetic concretions noted above and coal balls found near Wellsville, Ohio in the Lower Freeport Coal (No. 6a) of the Pennsylvanian Allegheny Series.

Some coal balls show textural variations that indicate formation under a wide range of energy conditions (Perkins 1976) including large-scale marine inundations that carried marine organisms into a peat swamp environment (Mamay and Yochelson 1962). Most coal balls, however, including those so far described from Ohio, were formed in situ under relatively quiescent conditions in peat swamps. Roof shale nodules, such as those described in my study, were formed in marine muds that often transgressed over peat deposits (Stopes and Watson 1909). At the coal ball locality in Jefferson County, Ohio there is no known example of a marine limestone directly above the Lower Freeport Coal (Lamborn 1930). Instead, the coal commonly occurs below a sandstone that shows no indication of marine origin. Weller (1930) observed in Illinois that such sandstone deposits above coals signify a period of erosion in which sediments originally covering the coal were removed before the sandstone was deposited. The coal probably was represented at that time by compressed peat that resisted erosion. Marine deposits that may have occurred above the Lower Freeport Coal in the Wellsville area probably were removed...
during the diastemic interval prior to the deposition of the overlying sandstone.

**DESCRIPTION OF THE WELLSVILLE EXPOSURE**

The coal ball collecting locality near Wellsville, Ohio is a deep sidehill roadcut on the west side of Ohio Route 7 in Saline Township, Jefferson County within T. 9 N. and R. 2 W. in the north central part of Section 7, Wellsville Quadrangle, 0.74 km south of the Columbiana and Jefferson County line. The locality was recognized in 1961 by Horace Collins, Chief of the Ohio Geological Survey. Several specimens were collected at that time and a few preliminary peels were prepared. A brief mention of the deposit appeared in Denton et al. (1961).

At the collecting locality, the concretion-bearing coal rests upon 0.5 m of light gray, non-bedded underclay. The coal varies in thickness from 1.4 m in areas where coal balls are more prominent to 0.6 m where they are more lenticular, flattened, or absent. The coal is overlain by a massive sandstone, 3 to 3.5 m thick, which, in the outcrop, extends beyond the coal.

The coal contains coal balls for a distance of about 18 m in a north-south trend (fig. 1). The coal balls appear either as aggregated masses or as isolated units in the coal with ratio of height to width on the order of 1.5. Bandings in the coal, which are prominently warped around individual coal balls, show disturbance vertically in either direction up to 0.25 m from isolated concretions.

**DESCRIPTION OF THE GNADENHUTTEN LOCALITY**

Roof shale nodules occurred in 2 strip pits operated by the Empire Coal Company of Gnadenhutten, Ohio. They are located in Clay Township, Tuscarawas County within T. 6 N. and R. 2 W. in the northeastern part of Section 3, 2.12 km southwest of Gnadenhutten and in the western part of Section 1, 0.74 km southwest of the village (Gnadenhutten Quadrangle). Both localities are just south of Tuscarawas County road 16.

Concretionary nodules occur in the shales above the Middle Kittanning Coal (No. 6) and to a lesser extent above the Lower Kittanning Coal (No. 5) in this area. At the two localities nodules for

![Figure 1](image_url)

**Figure 1.** Schematic drawing to scale of the Wellsville coal ball outcrop showing the three main concretionary pockets, A, B, and C, in the Lower Freeport Coal (No. 6a). The scale shown applies to both horizontal and vertical profiles.
my study were found along the highwalls in place in the shale above the Middle Kittanning Coal. The shale is about 20 m thick at the first locality and 2.5 m thick at the second. The concretions, exposed irregularly on the highwalls, are seldom aggregated but are roughly oriented in horizontal lines separated vertically by 15 to 30 cm intervals. Where concretion is superficially exposed (fig. 2), the major axis is oriented horizontally and the shale is visibly arched around it.

METHODS AND MATERIALS

Individual concretions were cleaned of mud and fragments of coalified material and then numbered to assure proper identity in all subsequent procedures. Each concretion was sawed (with a diamond-bladed slab saw) normal to its major axis, into roughly parallel 3 cm slices. The sawed surfaces were smoothed of imperfections with carborundum powder (#600) and etched in 10% HCl for 20 to 45 seconds. The wet etched surfaces were studied with a low-power binocular microscope. Dilute safranin Y dye (2%) was used to stain the plant material exposed on the etched surfaces of the coal balls (Kosanke 1945).

The rapid peel technique of Joy et al (1956) was employed to remove material from the etched surface. Before the peels were removed, the mineralogy and organic content was recorded. Some peel sections were removed and mounted on slides with Permount for high power microscopic examination (Stewart and Taylor 1965).

Vertically and horizontally oriented thin sections were prepared for mineralogical study of the concretions to show morphological structures, septaria, and other types of secondary mineralization that were not distinct in the acetate peels. Detailed analysis of the microscopic organic constituents of the concretions was accomplished by using solution residues (Schopf 1965). Rock fragments were dissolved in dilute HCl and the carbonate-free residue was washed and decanted several times, resulting in a sludge which was examined with a microscope. Fossil material was removed and placed in water for more detailed study. This method was especially effective for removal of isolated pyritized microfossils, fusain-like material and megaspore coats.

The minerals present in coal balls and their average composition by weight were determined by chemical analyses. X-ray diffraction was used to identify mineral content of the clays and carbonates. Unweathered sections of approximately equal weight were removed from each concretion crushed, well mixed, and divided into two subsamples. Half a sample was weighed and placed in 10% HCl to remove the carbonate fraction. Half of this residue was placed in dilute H2O2 to oxidize the organic material and the other half sample was analyzed by x-ray diffraction (G. E. Model X RD-6) to confirm the presence of calcite and pyrite in the coal balls.

Six roof shale nodules were sawn and the slices crushed and thoroughly mixed before separating into 2 portions. Each sample was weighed and placed in 10% HCl to remove the carbonate fraction, then dried and reweighed. One subsample was mixed into 1,1,2,2-Tetrabromoethane (Sp. Gr. 2.89) to separate the heavy and light mineral fractions. The lighter portion was isolated, dried, weighed and the organic material was completely oxidized with dilute H2O2. The resultant residue, after drying and reweighing, was washed until all the clay was removed and the final residue was again dried and weighed. The other subsample was analyzed chemically for average composition and by x-ray diffraction to determine carbonate-free mineral constituents.

The evaluation of organic material in the coal balls was based on peel studies from 50 randomly selected coal balls from all areas of the crop. Each coal ball regardless of size was considered as one unit. From many of the coal balls numerous peels were obtained but only 2peels oriented normally to each other were evaluated from each concretion. Each identifiable plant structure or other component was tabulated only once regardless of the number of occurrences within that peel. Percentage of occurrence of plant material in the coal balls was calculated on the basis of all 50 units in the study.

RESULTS AND DISCUSSION

COAL BALLS

The initial mineralization, which formed the coal balls, was the result of cellular permineralization of the plant material present (Schopf 1975). The yellow-brown primary calcite, which comprises the bulk of the matrix, was characteristically bladed or fibrous in crystal form. In thin section the calcite crystals appear to radiate outward from various loci within the concretion, passing undisturbed through interstitial spaces and cell walls. As was found also by Darrah (1941), cell walls in several cases served as terminating boundaries for crystals of fibrous calcite radiating from different loci. The cell wall material, however, was not replaced by the calcite, as organic plant tissues were not affected by etching. In one gymnospermous seed (probably Taxospermum undulatum) fibrous calcite had penetrated the megaspore membrane and two layers of sclerenchyma without disturbing the continuity of its crystal form. Schopf (1971) noted that calcite can interpenetrate cell walls, lumens, and intracellular
spaces without disturbing the histological structures due to the large amount of inherent moisture, which the permineralizing mineral displaces.

Cracks representing the second generation of mineralization have been attributed to compaction after lithification (Mamay and Yochelson 1962) or contraction during the final stages of lithification (Evans and Amos 1961). These were evident in every coal ball examined. Sometimes they were rather isolated pockets having a vuggy appearance, but usually they formed calcite-filled veins, which cut across plant tissue often separating morphologically continuous structures by as much as 12 mm.

Calcite in coal ball veins was present in three forms: fibrous calcite, prismatic calcite or both varieties together in a given vein. When both morphological forms of calcite were present, the fibrous variety grew inward from the walls of the vein indicating earlier deposition, while the interior portion was filled with prismatic calcite. Lindholm (1974) also noted this relationship in septarian veins. Crystals of pyrite commonly occur in small isolated concentrations along the line where the two varieties of calcite were in contact. Very little coloration was apparent in either of the calcitic forms but, of the two, the prismatic calcite had less coloration and probably less impurities.

### Table 1

<table>
<thead>
<tr>
<th>Floral Component</th>
<th>% Occurrence in 100 Peels</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lycopod periderm</td>
<td>84</td>
</tr>
<tr>
<td>Stigmaria sp.</td>
<td>48</td>
</tr>
<tr>
<td>Myeloxylon sp.</td>
<td>44</td>
</tr>
<tr>
<td>Psaronius sp.</td>
<td>24</td>
</tr>
<tr>
<td>Medulosae sp.</td>
<td>4</td>
</tr>
<tr>
<td>Sphenophyllum sp.</td>
<td>4</td>
</tr>
<tr>
<td>Taxospermum undulatum</td>
<td>1</td>
</tr>
<tr>
<td>Sporangia</td>
<td>1</td>
</tr>
</tbody>
</table>

The coal balls in my study were “normal” coal balls (Mamay and Yochelson 1962) that is, they contained only fossil plant material (table 1). Preservation of morphological plant structures in the coal balls was far from perfect. The most abundant component of the coal balls, in fact, was unidentifiable plant debris consisting of leaf litter and smaller plant parts. This material was disintegrated and often heavily pyritized. Prior to permineralization, it had filtered into recesses between larger pieces of lycopod periderm (fig. 3) and other resistant plant organs. Lycopod periderm was the only identifiable floral constituent in 36% of the concretions. Twenty percent of the coal balls contained some amount of fusain-like material, which was typically blocky and fibrous, but had fairly well-preserved cell lumens.

Compaction, presumably due to overburden pressure, was evident in all coal balls examined. Plant organs, such as lycopod periderm, that were more resistant to decay usually resisted compaction. The result was that differential compaction of material occurred in many of the concretions. Most of the root material (which is nearly circular in cross section) shows some indication of compaction before permineralization. The root cells in direct line with overburden pressure show some elimination of air spaces but little alteration otherwise.

The peat deposit in which the coal balls formed was probably the result of an accumulation of fresh plant material and its subsequent degradation to form decomposed leaf litter, disintegrated cells, fusain-like fragments, and small resistant plant parts. The juxtaposition of fragile plant organs with similar organs in various stages of decomposition suggest that there was a continuous addition of new material into the peat swamp environment. Sections of fairly well-preserved *Myeloxylon* petioles showed secretory canals and sclerenchyma around the periphery, and collateral vascular bundles with well-preserved xylem strands. Parenchymatous ground tissue, which usually decays first, was not evident. Other *Myeloxylon* petioles studied were partially compressed and their internal tissues were more or less disorganized.

Plant roots present in the coal balls probably represent the last growing, freshest plant parts introduced prior to
permineralization. Although they consist chiefly of thin-walled parenchymatous tissues, the roots showed the least effect of compaction and commonly retained their shape in the coal balls. Sections of *Stigmaria* showed an elimination of air spaces but their vascular bundles and thin-walled cortical cells were seldom compressed. In many cases, roots penetrated pieces of lycopod peri-

**Figures 2-5**

**Figure 2.** A roof shale concretion in place along the highwall above the Middle Kittanning Coal (No. 6). Note the shale appears to be deflected around the concretion. ×3.5.

**Figure 3.** Two portions of lycopod periderm from a coal ball peel showing the disintegrated cell material and leaf litter that had filtered around them before permineralization. ×0.4.

**Figure 4.** A roof shale aggregate showing an etched surface. The smaller nodule at the top has a crack through the center that is due to overburden pressure, while the rest of the concretion shows typical septarian development. ×1.3.

**Figure 5.** A septarian vein from an etched roof shale nodule. Fibrous calcite lines the walls and prismatic calcite fills the interior of the vein. A brachiopod shell in section, shown by arrow, was disrupted by the venation. ×0.3.

*When multiplied by factor shown will give normal dimensions.*
derm indicating that the peat actually formed a substrate that supported growing vegetation before permineralization. Barghoorn (1952) suggested that plant material, such as roots and rhizomes, that entered a peat deposit by growth, may not have been exposed subsequently to aerobic processes that cause decomposition.

**ROOF SHALE NODULES**

The mode of preservation of roof shale nodules appears to be by "authigenic cementation" (Schopf 1975). Durable fossil organisms were probably preserved in the soft sediment prior to the carbonate cementation. There was no evidence of any permineralization or distortion of fossil components after their encasement in the nodules.

In many nodules the primary cementation was disrupted by radiating septarian fissures (fig. 4). In some concretions the central portions were devoid of cracks, while in others the septaria occurred throughout the matrix. The fissures were usually distinctly polygonal in section toward the center of the nodule but decreased in diameter as they radiated outward as small wiry cracks. Occasionally, the smaller peripheral cracks appeared to be diverted by brachiopod shells in the matrix, however, the larger fissuring commonly displaced fossil material (fig. 5). In some of the concretions the calcite-filled cracks were exposed superficially as lattice-like structures that extend up to 6 mm above the surface.

The amber calcite, which fills the septaria, occurred primarily as the coarse prismatic form of the mineral and fibrous calcite was found only in some of the larger fissures. Where both forms of the mineral occurred in the same crack-filling, the walls of the filling were lined with fibrous calcite and prismatic calcite filled the interior. Pyrite was confined to the periphery of the nodules, especially in the area bordering separate concretionary units in an aggregate.

Only a small amount of organic material was preserved in the roof shale nodules. This consisted of a heterogeneous assemblage of both marine animals and terrestrial plant parts (table 2). The most abundant faunal component in

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Roof Shale Nodules</th>
<th>Coal Balls</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surrounding Material</td>
<td>Gray shale overlying Middle Kittanning Coal (No. 6).</td>
<td>Upper part of the Lower Freeport Coal (No. 6a).</td>
</tr>
<tr>
<td>Form</td>
<td>Mostly isolated concretions; also aggregates from 30–37 cm along major axis. Units within aggregate may be separated by a dense concentration of pyrite.</td>
<td>Mostly aggregated masses up to 1 m thick and 3 m long; seldom as individual concretions. Units within aggregate may be separated by 2–3 mm streaks of coaly material.</td>
</tr>
<tr>
<td>Relation to Sediment</td>
<td>Stratum is deflected around the concretions (fig. 2).</td>
<td>Stratum is deflected around the concretions.</td>
</tr>
<tr>
<td>Color</td>
<td>Light to medium gray, very light gray to yellowish on weathered surfaces.</td>
<td>Dark gray, brown to black or reddish brown on weathered surfaces.</td>
</tr>
<tr>
<td>Size</td>
<td>2.5–3.7 cm for major axis of isolated specimens.</td>
<td>5–50 cm for major axis of isolated specimens.</td>
</tr>
<tr>
<td>Shape</td>
<td>Lenticular, discoidal, spheroidal, or lobate.</td>
<td>Lenticular, spheroidal, or subangular.</td>
</tr>
<tr>
<td>Surface Texture</td>
<td>Relatively smooth to pitted with exposed fossils and septarian veins.</td>
<td>Rough with projecting edges, coaly veneer, and exposed plant material.</td>
</tr>
<tr>
<td>Specific Gravity</td>
<td>2.45 g/cm³ average.</td>
<td>2.71 g/cm³ average.</td>
</tr>
<tr>
<td>Fossil Fauna</td>
<td>Abundant brachiopods, including <em>Mesolobus, Lingula</em>, several productid species, some gastropods, bryozoan and crinoid parts, ostracodes, and cephalopods.</td>
<td>None</td>
</tr>
<tr>
<td>Fossil Flora</td>
<td>Megasporic coats.</td>
<td>Lycopods, pteridosperms, spores (table 1).</td>
</tr>
</tbody>
</table>
**Parameter** | **Roof Shale Nodules** | **Coal Balls**
--- | --- | ---
Miscellaneous material | Coprolites, fusain-like material, and root casts. | Fusain-like material.
Mode of Preservation | Authigenic preservation. | Cellular permineralization.
Quality of Preservation | Only the durable fossil components resistant to decay and transport abrasion are represented. | Most of the plant material is disintegrated, only some leaf and root material is delicately preserved.
Minerals Present | Calcite (84.5%), with illite (11.3%), quartz (1.5%), and pyrite (0.9%). | Translucent fibrous calcite radiating from various loci.
Initial Mineralization | Calcareous, but cloudy due to finely disseminated clay. | Non-septarian veins, which often disrupt or displace fossil material.
Secondary Mineralization | Radiate septarian cracks (fig. 3), polygonal near the center and wiry toward the periphery. Some are diverted by fossil material (fig. 4). | Veins filled with brown fibrous calcite oriented normal to the walls, clear prismatic calcite, or both forms with some pyrite.
Fabric of Veins | Mostly filled with amber prismatic calcite. Fibrous calcite line the walls in only a few septaria. Pyrite is mostly absent. | Resulted from cracking due to settling of substrate and overburden pressure.
Origin of Venation | Mostly septarian development primarily due to shrinkage of clay minerals. Some cracks were due to overburden pressure and settling. | Formed in a freshwater peat swamp under static conditions.
Environment of Deposition | Formed in off-shore marine muds. Some movement of sediment indicated but quiet conditions generally prevailed. | Probably derived from a marine transgression over unconsolidated peat. (Direct evidence was removed by erosion).
Origin of Carbonates | Probably derived from the surrounding sea water. | Probably derived from a marine transgression over unconsolidated peat. (Direct evidence was removed by erosion).
Time of Formation | Early diagenetic (fig. 6). | Late syngenetic to early diagenetic (fig. 6).
Chemical Conditions | An oxidizing environment is indicated. | Only limited oxidation occurred. The environment was mainly anoxicogenic.
Depth of Formation | Probably less than 0.5 m from the sediment-water interface. | Probably less than 1 m from the sediment-water interface.

The nodules were *Mesolobus s.p.*, a small articulated brachiopod, which occurred in great abundance in certain units of the Allegheny Series of Ohio (Sturgeon 1936). Rust-colored megaspore coats were the most easily identified terrestrial plant component. These were largely uncompressed and distorted, although a few were broken. Size variation was minimal with an average diameter of 0.25 cm. Unidentified fusinized fibrous-tissue fragments from 0.10 to 0.30 cm in diameter were also commonly distributed throughout the nodules. Coalified plant stems were present on the surface of 2 nodules and a small number of pyritized root casts appeared in the solution residues.

Only the more resistant fossil parts were evident in the roof shale nodules. The soft parts of organisms were not preserved presumably because they were oxidized, consumed, or removed by mechanical transport prior to lithification. The fossil shells were either calcareous or pyritized and many shells of the same species exhibited both types of mineral composition within the same nodule. The formation of roof shale nodules in marine mud probably proceeded relatively rapidly once initiated. This is indicated by megaspores in the...
Figure 6. Sequence of paragenetic events that led to the formation of coal balls and roof shale concretions.

<table>
<thead>
<tr>
<th>EUGENESIS</th>
<th>SYNGENESIS</th>
<th>DIAGENESIS</th>
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<tr>
<td>BIO-DEVELOPMENT</td>
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<td>ACCUMULATION</td>
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<td>CONSOLIDATION</td>
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<td>LITHIFICATION</td>
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<td>TRANSPORT</td>
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<tr>
<td>BACTERIAL ACTIVITY &amp; DECAY</td>
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<tr>
<td>AUTHIGENIC SULFIDE PRECIPITATION</td>
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<td>PEAT</td>
<td>CONSOLIDATED MUD</td>
<td>SHALE</td>
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<td>SOFT MUD</td>
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<td>COAL BALLS</td>
<td>ROOF SHALE CONCRETIONS</td>
<td>TIME</td>
<td></td>
</tr>
</tbody>
</table>
matrix of the nodules being quite rounded and complete and showing little evidence of compression prior to the authigenic cementation of the surrounding matrix. The calcareous and pyritic shell components were naturally much more resistant to compression, whereas the fragile megaspores could not have resisted much overburden pressure and still retain their spheroidal form.

**Comparative Analyses of the Calcareous Concretions**

Physical and chemical differences in coal balls and roof shale nodules arose primarily as a result of their formation in different environments. Twenty-three different parameters (listed in table 2) were used to compare the 2 types of concretions. Four separate categories: eugensis, syngenesis, diagenesis, and episgenesis were used to represent increasing lithification of the enclosing sediment in which the concretions formed (fig. 6). Various overlapping genetic classifications have been used to indicate the origin of concretions (Tarr 1921; Norwood 1958; Pantin 1958). My classification, with the addition of eugensis to indicate the period of development and death of the organic material found in the concretions, allows a more temporal comparison of the genesis of the concretions with changes in sedimentation and lithification.

Although both types of concretions were decidedly diagenetic in origin, it appears that the coal balls began to form during late syngenesis, while the roof shale nodules began to form later during burial, or in early diagenesis. The organic material in the coal balls was permineralized early enough so that bacterial activity, decay, and overburden did not destroy cellular features. The less durable organic matter in the roof shale nodules was destroyed either by transport erosion or by decay prior to the formation of concretions by authigenic cementation during burial and consolidation.

**Acknowledgments.** The author acknowledges the guidance, suggestions, and interest of Dr. James M. Schopf, who directed my attention to the Ohio coal ball locality and supervised the project. Technical assistance was provided by Robert Wilkinson, Jay Spielman, Karen Tyler, and John Ghist. Kenneth Beamer and Michele McCullogh assisted in the field. Dr. Stig Bergström, Dr. K. O. Stanley, and Stephen Jacobson read drafts of this paper in various stages of completion. The employees of The Empire Coal Company of Gnadenhutten, Ohio provided valuable information and free access to operating strip pits. The Friends of Orton Hall fund contributed a grant towards publication.

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